365003

IC 8650

Bureau of Mines IC 8650

PB-238 666

Bureau of Mines Information Circular/1974

Technology and Use of Lignite

Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 9-10, 1973

UNITED STATES DEPARTMENT OF THE INTERIOR

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Information Circular **8650**

Technology and Use of Lignite

Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 9-10, 1973

Compiled by Gordon H. Gronhovd and Wayne R. Kube
Grand Forks Energy Research Laboratory, Grand Forks, N. Dak.

UNITED STATES DEPARTMENT OF THE INTERIOR
Rogers C. B. Morton, Secretary

BUREAU OF MINES
Thomas V. Falkie, Director

As the Nation's principal conservation agency, the Department of the Interior has responsibility for most of our nationally owned public lands and natural resources. This includes fostering the wisest use of our land and water resources, protecting our fish and wildlife, preserving the environmental and cultural values of our national parks and historical places, and providing for the enjoyment of life through outdoor recreation. The Department assesses our energy and mineral resources and works to assure that their development is in the best interests of all our people. The Department also has a major responsibility for American Indian reservation communities and for people who live in Island Territories under U.S. administration.

This publication has been cataloged as follows:

Gronhovd, Gordon H comp.

Technology and use of lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 9-10, 1973. Compiled by Gordon H. Gronhovd and Wayne R. Kube. [Washington] U.S. Bureau of Mines [1974]

262 p. illus., tables. (U.S. Bureau of Mines. Information circular 8650)

1. Lignite—Congresses. 2. Coal liquefaction—Congresses. 3. strip mining—Congresses. 4. Fly ash—Congresses. 5. Reclamation of land—Congresses. 6. Powerplants—Congresses. I. Kube, Wayne R., jt. comp. II. U.S. Bureau of Mines. III. Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., 1973. IV. Title. (Series)

TN23.U71 no. 8650 622.06173

U.S. Dept. of the Int. Library

For sale by t rinting Office

Stock No. 2404-01684

BIBLIOGRAPHIC DATA SHEET

1. Report No. BuMines IC 8650

2.

3. Recipient's Accession No.

4. Title and Subtitle
Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 9-10, 1973

5. Report Date
November 1974

6. Performing Organization Code

7. Author(s)
Gordon H. Gronhovd and Wayne R. Kube

8. Performing Organization Rept. No.

9. Performing Organization Name and Address
Grand Forks Energy Research Laboratory
Bureau of Mines, USDI
P.O. Box 8213 University Station
Grand Forks, ND 58201

10. Project/Task/Work Unit No.

11. Contract/Grant No.

12. Sponsoring Agency Name and Address
U.S. Department of the Interior, Bureau of Mines and the University of North Dakota, University Station, Grand Forks, ND 58201

13. Type of Report & Period Covered
Symposium, 1973

14. Sponsoring Agency Code

15. Supplementary Notes

Seventh symposia in a series of meetings at two-year intervals.

16. Abstracts The proceedings of the 1973 Lignite Symposium held at Grand Forks, N. Dak., May 9-10, includes 16 papers: Operations of Western Company; New Developments in Open Cast Mining Technology at Rheinische Braunkohlenwerke, AG, Koeln; Big Stone Plant: Design Features and Fuel Handling; Shipment, Storage, and Handling Characteristics of Dried Low-Rank Coals; Gas From Coal: Fuel of the Future; Some Studies on Stack Emissions From Lignite-Fired Power Plants; Pilot Plant Scrubber Tests to Remove SO_2 Using the Soluble Alkali in Western Coal Fly Ash; Experience in Use of Brown Coals From the Latrobe Valley, Australia; Design Considerations for 575 MW Units at Big Brown Steam Electric Station; Design and Operating Experiences of Steam Generators for Low-Grade Brown Coal; New Day Dawns for Great Plains Coals; Utilization of Lignite and Subbituminous Ash; Low-Btu Fuel Gas for Power Generation; CO_2 Acceptor Process: Status of Development; Laboratory Studies on Solution-Hydrogenation of Lignite; and The Liquefaction of Subbituminous Coal.

17. Key Words and Document Analysis. 17a. Descriptors

- Energy
  - Fossil fuels
  - Lignite
  - Subbituminous coal
  - Brown coal
  - Oil
  - Gas
- Environment
  - Air quality control
- Mining
- Processing
- Combustion
- Scrubbers
- Liquefaction
- Hydrogenation
- Thermal electric plants
- Fly ash
- Reclamation
- Drying
- Storage
- Transportation

17b. Identifiers/Open-Ended Terms

- Wet scrubbing stack emission
- Open pit mining
- Reclamation of mined land
- Fly ash utilization
- Energy supply options
- Stack emissions
- Power plant design
- Power plant operation
- Synthetic natural gas
- Low-Btu gas
- Liquefaction of lignite and subbituminous coal
- Drying of lignite and subbituminous coal
- Storage of dried low-rank coal
- Utilization of Australian brown coal
- Mining practice in Germany

17c. COSATI Field/Group 07A; 07C; 07D; 10A; 10B; 14B; 21B; 21D;

18. Distribution Statement Sold by Sup. Docs., GPO, [redacted], Catalog No. I28.27:8650, Stock No. 2404-01684; available from NTIS in microfiche only for $2.25.

19. Security Class (This Report) UNCLASSIFIED

20. Security Class (This Page) UNCLASSIFIED

21. No. of Pages 272

22. Price $

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11. **Contract/Grant Number.** Insert contract or grant number under which report was prepared.

12. **Sponsoring Agency Name and Address.** Include zip code.

13. **Type of Report and Period Covered.** Indicate interim, final, etc., and, if applicable, dates covered.

14. **Sponsoring Agency Code.** Leave blank.

15. **Supplementary Notes.** Enter information not included elsewhere but useful, such as: Prepared in cooperation with . . . Translation of . . . Presented at conference of . . . To be published in . . . Supersedes . . . Supplements . . .

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    (b). Identifiers and Open-Ended Terms. Use identifiers for project names, code names, equipment designators, etc. Use open-ended terms written in descriptor form for those subjects for which no descriptor exists.
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19 & 20. **Security Classification.** Do not submit classified reports to the National Technical Information Service.

21. **Number of Pages.** Insert the total number of pages, including this one and unnumbered pages, but excluding distribution list, if any.

22. **Price.** Insert the price set by the National Technical Information Service or the Government Printing Office, if known.

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TECHNOLOGY AND USE OF LIGNITE

Proceedings: Bureau of Mines-University of North Dakota Symposium,
Grand Forks, N. Dak., May 9-10, 1973

Compiled by

Gordon H. Gronhovd[1] and Wayne R. Kube[2]

ABSTRACT

Sixteen papers concerned with the technology and utilization of low-rank fossil fuels are presented as the proceedings of the 1973 lignite symposium. This symposium, the seventh in a series of biennial meetings, was cosponsored by the Bureau of Mines and the University of North Dakota.

INTRODUCTION

The Bureau of Mines of the U.S. Department of the Interior and the University of North Dakota have since 1961 cosponsored biennial lignite symposia to bring to the attention of interested parties developments in the technology and utilization of low-rank fossil fuel. Usually the meetings are held on the campus of the University of North Dakota, but in 1965 and 1971 they were held in Bismarck, N. Dak., to facilitate field trips to lignite mines and lignite-fired electrical generating stations. The present proceedings compile the edited papers presented at the seventh symposium, which was held at Grand Forks, N. Dak., on May 9-10, 1973.

The 1973 lignite symposium attracted nearly 400 registrants from many States, the District of Columbia, seven Canadian Provinces, and four European countries. Generally, the registrants represented middle to upper level management and technical personnel of energy-oriented organizations from Governments, universities, and industries. Much interest was expressed concerning the potential of the low-rank coals from the Northern Great Plains

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[1] Chief, Grand Forks Energy Research Laboratory, Bureau of Mines, Grand Forks, N. Dak.

[2] Professor of Chemical Engineering, University of North Dakota, Grand Forks, N. Dak.; chemical engineer, Grand Forks Energy Research Laboratory, Bureau of Mines, Grand Forks, N. Dak.

Province. Proceedings of previous symposia including the 1958 lignite forum have been published to allow further dissemination of information.[3]

Presiding at the various sessions were Wm. L. Crentz, Assistant Director--Energy, Bureau of Mines, Washington, D.C.; Alan G. Fletcher, Dean, College of Engineering, University of North Dakota, Grand Forks, N. Dak.; P. J. Reiten, Professor of Mechanical Engineering, University of North Dakota, Grand Forks, N. Dak.; A. W. Johnson, Dean, Graduate School, University of North Dakota, Grand Forks, N. Dak.; and A. M. Cooley, Professor of Chemical Engineering, University of North Dakota, Grand Forks, N. Dak. Those in attendance were welcomed by W. E. Koenker, Vice President, Academic Affairs, representing the University of North Dakota, and T. A. Henrie, Deputy Director--Mineral Resources and Environmental Development, representing the U.S. Department of the Interior.

The many contributions of J. L. Elder, former chief of the Grand Forks Energy Research Laboratory, warrant special notice. Mr. Elder was cochairman for the first six symposia and was instrumental in developing the program for the 1973 lignite symposium prior to his transfer to the Bureau's underground gasification project as a research supervisor. Subsequently, he retired in June 1973.

The symposium consisted of three half-day sessions, a luncheon, and a banquet meeting, during which 16 papers were presented.[4] Subject matter covered mining and the environmental aspects associated with open pit mining, powerplant design and operation, emissions from powerplants and their control,

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[3] Kube, W. R., and J. L. Elder (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Bismarck, N. Dak., May 12-13, 1971. BuMines IC 8543, 1972, 145 pp.
Elder, J. L., and W. R. Kube (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 1-2, 1969. BuMines IC 8471, 1970, 174 pp.
Kube, W. R., and J. L. Elder (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April 27-28, 1967. BuMines IC 8376, 1968, 201 pp.
Elder, J. L., and W. R. Kube (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Bismarck, N. Dak., April 29-30, 1965. BuMines IC 8304, 1966, 124 pp.
Kube, W. R., and J. L. Elder (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April-May 1963. BuMines IC 8234, 1964, 128 pp.
Elder, J. L., and W. R. Kube (comp.). Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April 1961. BuMines IC 8164, 1963, 113 pp.
North Dakota Economic Development Commission. North Dakota Lignite Forum, Speech Summaries. Bismarck, N. Dak., 1958, 49 pp.

[4] Company and trade names are used throughout these proceedings for clarity and identification purposes only, and their use does not imply endorsement or recommendation by either the Bureau of Mines or the University of North Dakota.

gasification and liquefaction, and the potential for utilization of western coals.

Abstracts of the papers presented at the 1973 lignite symposium follow in order of presentation.

ABSTRACTS OF PAPERS

Operations of Western Energy Co.

By W. P. Schmechel, Vice President and General Manager,
Western Energy Co., Butte, Mont., and Richard L. Hodder,
Research Associate, Agriculture Experiment Station,
Montana State University, Bozeman, Mont.

The Colstrip mine of Western Energy Co. in 1973 was the third largest coal mine in the United States with over 5.5 million tons produced. It is estimated that 12 to 13 million tons/year will be mined in 1980. Strip mining uses 60-cu-yd draglines and 17-cu-yd loading shovels. An extensive reclamation program is underway coordinated with the mining plan. The major problem in reclamation of the mined land is lack of moisture. Studies are underway on methods for effectively retaining moisture to promote revegetation. Plant nutrients and plant adaptibility are also critical and are being evaluated.

New Developments in Opencast Mining Technology at Rheinische Braunkohlenwerke AG, Koeln

By Hans J. Leuschner, Head, Planning Department,
Rheinische Braunkohlenwerke AG, Koeln,
West Germany

Advanced techniques will allow profitable opencast mining at depths of 1,650 feet with a volume stripping ratio of 7.6:1. Presently, mining is at 660 feet for 160-foot seams. Bucket-wheel excavators of 260,000 cu yd/day are planned. Estimated production rate will be 100 to 130 tons per man-day. Mining patterns are detailed. In past mining operations, 44 villages with 18,500 inhabitants have been moved along with railroads, highways, and even rivers. Reclamation efforts have been highly successful with restoration to farm land or woods and lake areas depending on the political-economic conditions.

Big Stone Plant: Design Features and Fuel Handling

By Orville B. Johnson, Superintendent, Production,
Engineering and Construction, Otter Tail Power Co.,
Fergus Falls, Minn., and Robert F. Middleton,
Assistant Superintendent, Production Operation,
Otter Tail Power Co., Fergus Falls, Minn.

To be completed by May 1975, the Big Stone plant is a 440-MW lignite-fired electric generating station using cyclone burners. Larger than any unit presently burning North Dakota lignite, the operation is a joint venture of

Montana-Dakota Utilities Co., Northwestern Public Service Co., and Otter Tail Power Co. Otter Tail Power Co. is to build, operate, and maintain the plant. Located near Big Stone Lake in northeastern South Dakota, the plant will use cooling ponds with 7,000 acre-ft/yr being withdrawn from Big Stone Lake. Two to three million tons/yr of lignite will be delivered by unit train from an open pit mine of the Knife River Coal Mining Co. near Gascoyne in southwestern North Dakota. Over 200 flat-bottomed gondola cars of 100 tons capacity will form two unit trains of 100 cars each. A round trip including loading and unloading will require 48 hours. An unique feature of the rail cars is that they will be covered with a hinged roof system that closes and opens automatically for loading and unloading.

Shipment, Storage, and Handling Characteristics of Dried Low-Rank Coals

By L. E. Paulson, chemical engineer, S. A. Cooley,
mechanical engineer, and R. C. Ellman, Research
Supervisor, Grand Forks Energy Research Laboratory,
Bureau of Mines, Grand Forks, N. Dak.

Rail car quantities of lignite and of subbituminous coal were dried in a modified entrained dryer and shipped to the laboratory where the dried coal was stockpiled. Despite relatively high coal temperature when removed from the cars, successful storage was achieved for 2 years. A conical pile formed from a stable stockpile of lignite self-ignited, indicating that the dried low-rank coal was quite reactive even after an extended aging period. Forming a stockpile was more difficult and dust problems were greater than with undried coal. In pilot plant tests, the reactivity of dried coal toward oxygen in the air was found not to vary significantly with moisture content. However, temperature of the dried particles was critical. Heat loss through a bed of dried coal is low. Both dried subbituminous coal and lignite regained moisture when exposed to an atmosphere of high humidity. Tests indicated that the settled bulk density of dried material was not significantly different from that of the original coals.

Gas From Coal: Fuel of the Future

By G. Alex Mills, Chief, Division of Coal,
Bureau of Mines, Washington, D.C.

Synthetic fuels are of increased interest in the United States because of present and projected energy shortages. Coal is the most abundant energy source and can be converted into gaseous and liquid fuels. Applications have been filed with the Federal Power Commision to construct two large gasification plants in the Four Corners area. It is projected that in 15 years, 36 gasification plants will be in operation to produce 3 trillion cu ft of gas annually, about 13 pct of present use. Total cost for plants and mines should be about $14 billion, and over 36,000 people will be required for operation. Approximately 300 million tons of coal will be required or about 50 pct of present total production. Cost for gas would be in excess of $1 per Mcf. First plants will use the Lurgi dry-ash gasification process, and later units will use one of several processes being developed. The Bureau's Synthane gasification process was discussed and other Bureau processes were mentioned.

Some Studies on Stack Emissions From Lignite-Fired Powerplants

By G. H. Gronhovd, Chief, P. H. Tufte, chemical engineer,
and S. J. Selle, mechanical engineer, Grand Forks Energy
Research Laboratory, Bureau of Mines, Grand Forks, N. Dak.

Tests were made on three pulverized-coal-fired (pc-fired) plants, a cyclone-fired unit, and a spreader-stoker plant. Less than 1 ppm of SO_3 was found in stack gases. Usually, SO_2 concentrations were less than 2.0 lb/MMBtu fired and in some instances were less than the 1.2 lb/MMBtu set by national standards. A 40-pct reduction in emission would allow meeting standards. The SO_2 discharge is strongly influenced by the sodium content of the lignite. Indications are that sulfur retention in the ash is less with cyclone firing than with pc firing and that NO_x concentrations are higher. Particle size of fly ash when burning low-sodium lignite is larger using cyclone firing than with pc firing. Sulfur retained in ash was from 10 to 40 pct. Higher sodium contents in fly ash were found to reduce resistivity of the ash.

Pilot Plant Scrubber Tests To Remove SO_2 Using Soluble Alkali in Western Coal Fly Ash

By P. H. Tufte, chemical engineer, Grand Forks Energy Research
Laboratory, Bureau of Mines, Grand Forks, N. Dak.,
E. A. Sondreal, Acting Engineer in Charge, Gasification and
Combustion, Grand Forks Energy Research Laboratory, Bureau of
Mines, Grand Forks, N. Dak., K. W. Korpi, mechanical engineer,
Abe W. Mathews Engineering Co., Hibbing, Minn., and
G. H. Gronhovd, Chief, Grand Forks Energy Research Laboratory,
Bureau of Mines, Grand Forks, N. Dak.

Tests were run on two pilot wet scrubbers. One, at the Grand Forks Energy Research Laboratory, was a 110-cfm flooded-disk unit, and the other, at the Hibbard plant in Duluth, Minn., was a 3,000-cfm spray scrubber. A high-alkali lignite-fly ash from the Glenharold mine in North Dakota and a low-alkali subbituminous coal fly ash from the Colstrip mine in Montana were tested. Using Glenharold fly ash in quantities typically found in flue gases from a pulverized-coal-fired boiler and the small scrubber, SO_2 removal was as high as 40 pct of an initial 1,100 ppm. With the small unit and subbituminous coal fly ash in the normal quantities from a cyclone-fired boiler, SO_2 removal was about 20 pct of an initial 1,100 ppm. Approximately 35 pct of an initial SO_2 concentration of 1,100 ppm was removed using the subbituminous coal fly ash from pulverized-coal firing during tests at the Hibbard plant. The Duluth operation, nonclosed loop, used 9 pct makeup in proportion to the liquid feed to scrubber, and no scaling problems were encountered. In tests at Grand Forks, closed loop, no significant change in SO_2 removal was noted during approach to steady-state operation. Some problems with scaling did occur.

Experience in the Use of Brown Coals From the Latrobe Valley, Australia

By R. S. Higgins, Research Manager, Scientific Division,
Planning and Investigations Department, G. Bonafede,
Head, Material Technology Section, Scientific Division,
Planning and Investigations Department, and L. Hoch,
engineer, Hazelwood power station, Latrobe Valley
Department, State Electricity Commission of
Victoria, Richmond, Victoria, Australia

On a geological basis, coal reserves of the Latrobe Valley have been estimated at 47,500 million tons proved and 37,500 million tons inferred. On proved resources, some 29,000 million tons occur with less than 100 feet of overburden, and 10,000 million tons are considered recoverable. The Yallourn power station has a total capability of 546 MW. The Hazelwood station consists of eight 200-MW units with the first of four planned 350-MW units being commissioned. The larger units use pulverized-coal firing. Usually the brown coal has about 67 pct moisture, but many contain as much as 73 pct. Some difficulties were encountered in maintaining stable combustion, especially in the larger units. Design changes have essentially solved the problem except for very high moisture and ash content. Laboratory tests indicate that flame stability is also dependent of coal source (inorganic constituents and petrology) and particle size. Ash deposition from different coals are of sulfate-bonded type but exhibit marked variations in quantities and characteristics of fireside deposits. Water blowers are more effective than steam soot blowers in controlling ash deposition depending on location of deposit. Off-load cleaning is usually required after 3,000 to 4,000 hours of operation. High water solubility of coal ashes cause some difficulties in operation of ash sluice system and in disposal of waste water.

Design Considerations for 575-MW Units at Big Brown Steam Electric Station

By W. H. McAlpin, Project Manager, Fossil Fuels, and
B. B. Tyus, Senior Mechanical Engineer, Texas
Utilities Services, Inc., Dallas, Tex.

Dallas Power and Light Co., Texas Electric Service Co., and Texas Power and Light Co. jointly own the Big Brown plant consisting of two 575-MW units. The first unit was initially operated in December 1971 and the second in November 1972. These units featuring "outdoor" construction and a supercritical pressure design are the largest in the country to use lignite. Pulverized-coal-fired boilers deliver 4,025,000 lb/hr of steam at 3,825 psig, 1,005° F/ 1,005° F and are designed for dry bottom operation. Eight bowl mills, four on a side, pulverize the lignite with an individual mill feeding a level of burners. Furnace bottom ash is collected in a bottom ash hopper and transported hydraulically to a storage pond. Fly ash is collected in the precipitator hoppers and transported pneumatically to a silo for subsequent disposal to a landfill area. Operation has been at or near design capacity for extended periods. Initially, ash deposition was considered excessive, and 46 additional wall blowers were installed to provide satisfactory cleaning.

Design and Operating Experiences of Steam Generators for Low-Grade Brown Coal

By Thomas Buck, Head, Sales Department, and Udo Schuler,
Head, Mill Construction Department, EVT Energie und
Verfahrenstechnik GmbH, West Germany

Bown coal has been used extensively in boilers for raising steam for generation of electric power. Size of the pulverized-coal-fired units has increased over the years with 600-MW stations now being commissioned. Fuel characteristics are important in determining plant design because brown coal may have heating values from 1,450 to 9,000 Btu/lb with moisture contents as high as 73 pct and ash as high as 50 pct. It may be necessary to provide a blending plant at the mine to insure uniform quality of the coal. Various designs and layout of burners are discussed. Designs of mills, steam generators, and evaporator systems are reviewed, as were general methods for ash removal and ash fouling control.

New Day Dawns for Great Plains Coals

By Carl E. Bagge, President, National Coal Association,
Washington, D.C.

The large reserves of western coals are a key to the Nation's continued prosperity and comfort as well as security. Because of the present energy crisis, a quarter of our petroleum requirements are imported. Imports of petroleum are increasing and together with planned large imports of natural gas will contribute to an increase in our international trade deficit. The largest part of imports will be from countries that may place considerable political as well as economic pressure on the United States. Lignite and subbituminous coal represent a large, relatively unused source of low-cost energy. Restrictions on use of high-sulfur fuels has increased interest in these coals. Long-term contracts have been signed that extend the market area of subbituminous coal to Texas, Minnesota, Iowa, and Oklahoma. Such movement by unit train displaces other coals and will be necessary until adequate SO_2 removal technology is provided. Major use of western coals will be in production of gaseous and liquid products. Research and development need to be increased drastically to provide the required technology. Announced plans for gasification projects indicate that dry-ash Lurgi gasification, followed by methanization, will be used initially rather than more modern methods being investigated. Impact of large installations such as required for gasification will affect economic, environmental, and population considerations.

Utilization of Lignite and Subbituminous Ash

By Oscar E. Manz, Professor of Civil Engineering,
University of North Dakota, Grand Forks, N. Dak.

With increased use of lignite and subbituminous coal for electrical generation, the quantities of fly ash produced also increases. Although extensive application of cyclone-fired boilers will decrease the production of fly ash and increase production of slag per quantity of electrical power, total available fly ash should be higher. From 1971 to 1972, production of lignite fly ash increased by 70 pct, but percentage of utilization remained nearly constant. Fly ash from western low-rank coals is more basic than bituminous coal fly ash, which presents difficulties in meeting present ASTM standards. Research is underway to establish performance of basic fly ashes and to have standards modified so as to permit their acceptance. Physical and chemical data on western fly ash are given. Roadbuilding applications for soil stabilization and poz-o-pac base mixtures have used the largest tonnage of lignite fly ash in North Dakota with satisfactory results. The North Dakota State Highway Department has specifications for lignite fly ash to be used as a mineral filler. Research on use of fly ash in bricks, road base material, and other applications is continuing at the University of North Dakota. Work by other organizations is reviewed.

Low-Btu Fuel Gas for Power Generation

By P. S. Lewis, Research Supervisor, R. J. Belt,
chemical research engineer, and A. J. Liberatore,
mechanical engineer, Morgantown Energy Research Center,
Bureau of Mines, Morgantown, W. Va.

Fuel gas for power generation can be used to help meet near-term energy requirements arising from shortages of natural gas and low-sulfur oil and coal. Coal gasified with air and steam at moderate pressure gives a low-Btu fuel gas that can be desulfurized at temperature and pressure, thus meeting clean air standards. Benefits from stirring the bed of a pressurized gas producer were described, and results were discussed for three coals having widely different properties: Lignite, a moderately caking coal from western Kentucky, and a strongly caking coal from West Virginia. The aforementioned coals from western Kentucky and West Virginia are high in ash and sulfur and have large reserves that could be utilized for gasification purposes. Development of a process for removing hydrogen sulfide from hot producer gas was described in which a regenerable solid sorbent composed of iron oxide and fly ash is used. Elemental sulfur can be recovered during regeneration, and activity of sorbent is maintained over 175 regeneration cycles.

CO_2-Acceptor Process: Status of Development

By Carl E. Fink, Project Manager, Consolidation Coal Co.,
and Marion H. Vardaman, Manager, Technical Operations,
Stearns-Roger Corp., Rapid City, S. Dak.

The CO_2-Acceptor Process is being developed by Consolidation Coal Co. in a pilot plant operation at Rapid City, S. Dak., under a contract with the Office of Coal Research. Personnel from Stearns-Roger Corp. constructed and are operating the plant. Designed specifically for low-rank, noncaking coals, the process uses a dolomite acceptor to provide the thermal requirements for gasification by sensible heat and by the exothermic reaction of CO_2 in the gas with the calcined dolomite. Gasification reactions between steam and coal carbon occur at 11 atm and 1,500° F to produce a synthesis gas for methanization without use of a shift converter. Resulting pipeline gas would have a heating value of 900 to 950 Btu/scf. The pilot plant has been in operation for about a year to provide data for design of a 250-MMscfd commercial unit. Eight runs have been made using char as feedstock. Sustained gasification has not been achieved but information from each test has made the succeeding runs more successful. The process was explained and mechanical/operational difficulties and solutions described. When operation has been established using char, two lignites and a subbituminous coal will be gasified. Dolomites and limestone from Montana, Wyoming, North Dakota, and South Dakota will be evaluated as acceptor.

Laboratory Studies on Solution-Hydrogenation of Lignite

By Donald E. Severson, Principal Investigator, Project Lignite;
Professor and Chairman, Department of Chemical Engineering,
A. Max Souby, Project Manager, Project Lignite, and
Wayne R. Kube, Professor, Chemical Engineering,
the University of North Dakota, and chemical engineer,
Grand Forks Energy Research Laboratory,
Grand Forks, N. Dak.

Batch autoclave tests on solution-hydrogenation of lignite were made to provide information for development and design of a small continuous unit under contract with the Office of Coal Research. Reaction time and solution temperatures have been investigated. Conversions in excess of 90 pct (moisture- and ash-free basis) have been obtained using anthracene oil as a solvent in atmospheres of carbon monoxide, carbon monoxide plus hydrogen, or hydrogen. The effect of concentrations of cations in lignite on conversion was studied. Results indicate that shift catalyst cations, such as sodium, calcium, and potassium, increase formation of lighter products. Sufficient concentrations of cations seem to be present in natural lignites to maximize yields of solvent refined coal.

The Liquefaction of Subbituminous Coal

By Dat Nguyen, Graduate Assistant (presently with
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Bench-scale tests showed that in 5 minutes, 70 to 80 pct (moisture and ash free) of a subbituminous coal could be converted into benzene-soluble products by hydrogenation using carbon monoxide and water. Phenanthrene at a 2:1 solvent-to-coal ratio appears to be the best solvent. For highest conversions, a final temperature of 475° C at 5,500 psi is required in the presence of 1 to 3 pct of a sodium salt as catalyst. An incomplete factorial experimental design was used to indicate significance of variables in tests using massive quantities of molten $KCl:ZnCl_2$ catalyst to convert subbituminous coal to benzene-soluble material. It was found that conversions were significantly higher at 2,500 psig than at 500 or 1,500 psig; that conversion of minus 140-mesh coal was higher than minus 100- plus 140-mesh fraction; and that at high pressure there was significant interaction effect between coal size and pressure. Salt:coal ratio and $KCl:ZnCl_2$ mole ratio had no significant effect on conversion. Parameters used for tests have essentially the same effect on subbituminous coal or lignite. The molten salt mixture used as a catalyst became poisoned, probably by coal nitrogen. Use of high-temperature gas removal procedures reduced nitrogen content of the salt-coal mixture.

OPERATIONS OF WESTERN ENERGY CO.

by

W. P. Schmechel[5] and Richard L. Hodder[6]

Mining at Colstrip, Mont.

The Western Energy Co. surface mine at Colstrip, Mont., has been the third largest coal mine in the United States for the past 2 years. Western Energy began mining in 1968, when 150,000 tons of coal were extracted. Production in subsequent years was as follows: 1969--521,000 tons, 1970--1,658,000 tons, 1971--5,161,000 tons, and 1972--5,501,000 tons.

The Montana Power Co., parent company of Western Energy Co., acquired the coal properties from the Northern Pacific Railway Co. in 1959. Before that, the railroad had operated a surface mine at Colstrip for 35 years. Actual mining began in 1924 and terminated in 1958. Peak production during those years occurred in 1943 when 2,577,000 tons of coal were mined for use in steam locomotives. Complete dieselization of the rail line in 1958 obviated coal as locomotive boiler fuel. During the years of Northern Pacific mining, 44 million tons of coal were extracted in mining approximately 1,000 acres of land.

The Colstrip mine was the first all-electric surface coal mine in the Nation's history. Electricity drove the dragline and loading shovel and charged the batteries to power locomotives that delivered coal to the main Northern Pacific rail line 35 miles north of Colstrip. Montana Power Co. extended a 100-mile, 55,000-volt, electrical transmission line from Billings, Mont., to Colstrip to supply the energy needed in mining. Western Energy has continued to use all of the mining equipment acquired from the Northern Pacific, supplemented by additional bulldozers and scrapers.

Since the initial acquisition of the Colstrip property, Western Energy has acquired reserves in the vicinity totaling 850 million tons of coal.

The subbituminous coal at Colstrip comes from the Rosebud coal vein. The seam averages 25 feet in thickness with overburden as great as 150 feet. The average heating value of the coal is 8,700 Btu/lb. Ash content is 8 pct, and the sulfur content averages 0.7 pct. The low sulfur content of the coal has been a prime reason for reviving the Colstrip mine, just as it has been in increasing interest in mining activity throughout the Fort Union coal region.

Western Energy is supplying coal for 25 pct of the generating capacity of the Montana Power Co. and also substantial volumes to midwestern utilities.

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Today, Western Energy is preparing for expansion of mining operations to accommodate, by 1980, a production level of between 12 and 13 million tons annually. The planned production increase reflects enlarged Midwest utility markets and the need to supply two 350-MW electrical generating units under construction at Colstrip by the Montana Power Co. and Puget Sound Power and Light Co. of Bellevue, Wash.

During the next 2 years, a substantial construction program will be in progress at Colstrip. Two new mining areas will be readied for production. Grading and bridge construction for a new railroad spur and loop track have been completed. The mile-long railroad spur track will be laid before September. The loop track will be installed in 1974. The 2-mile-long loop track will pass beneath a new loading tipple to allow in-motion loading of unit trains. New machinery to remove overburden and dig coal includes two 60-cu-yd draglines and two 17-cu-yd loading shovels. The total investment in machinery, coal loading equipment, and railroad facilities will be about $30 million.

As important as construction in the expanded mining areas is evaluation of the environment of the region and preplanning for reclamation. Baseline wildlife evaluation and hydrologic studies are underway in the projected mining areas and for the surrounding 10-mile by 20-miles region. Additional studies of rangeland, soil chemistry, vegetation, and small mammal populations are being conducted, as is an analysis of the coal for the presence of 14 trace elements.

In 1968, before the first acre of ground had been disturbed at Colstrip, Western Energy realized that a comprehensive reclamation program would have to accompany the mining of coal. The Agricultural Experiment Station at Montana State University in Bozeman, Mont., was engaged to conduct research that would lead to procedures for restoration of mined land to its previous use or a better use. Close coordination between the reclamation program and mining efforts has been beneficial to both, and reclamation at Colstrip currently is keeping pace with mining.

Concepts and Implementation of Reclamation Research

Is there a good definition of reclamation? - for restoration? - for rehabilitation? Everyone has his own impression of what reclamation means. The work reclamation should be eliminated because it connotes that the land has already been abused. A way must be found to take the wreck out of reclamation.

One must do better than merely restore mined lands, for the creative potential for accomplishing great things is very real. Only several short years ago, reclamation in the semiarid west commonly meant irrigating dry lands to make them productive. Presently, reclamation emphasis is focused toward surface mine spoils, and irrigating the desert in a reclamation sense seems far from the present problem. Or is it?

Irrigating is a means of supplying a critical limiting factor to a specific agricultural problem, usually in an area where other requirements are adequate. Supplying the limiting factors is what must be done to accomplish effective reclamation.

Critical Factors in Reclamation

What are the common critical limiting factors pertaining to reclamation of surface mined lands in the semiarid west? Obviously, water is the major factor; availability of plant nutrients is another prerequisite for spoils are notoriously sterile; alkalinity, drought, extremely high temperatures, evaporation and excessive winds cannot be overlooked. These three requirements--water, plant nutrients, and plant adaptability--are the major limiting factors, and they have been stated in the order of importance for most of eastern Montana.

But supplying the three key requirements for successful revegetation is not enough, and this has become very obvious from previous research. It is the right combination of essential factors at the critical time that results in successful revegetation. The lack of any one crucial factor or the timing of its availability will detrimentally influence results and in most cases insure complete failure. This essential combination of factors occurs naturally at such infrequent intervals that natural and unaided reclamation is an extremely slow process in the semiarid west.

In the coalfields of eastern Montana, the mean annual precipitation is about 12.5 inches. At Colstrip, which is higher in elevation than much of the surrounding country, mean annual precipitation is about 15 inches. In 1971, there was more than 19 inches of rainfall. This sounds like a great growing season, but in this year of unusual annual precipitation, the growing season was the driest in many decades. Flood rains occurring in October inflated the accumulated rainfall figures. This, of course, did not help the spring growth of cool season grasses that complete a seasonal cycle between April and July.

The normal precipitation pattern presents problems for a semiarid situation. Conservatively, one-fourth to one-third of the annual precipitation comes in the form of snow. Most of this sublimates on the site or blows away to the nearest draw where it is lost as runoff when melted. Another fourth may come as low-intensity storms that hardly dampen the surface of the soil. With excessive winds, high temperatures and exposed spoils, evaporation is extreme, often five times as great as precipitation during the growing season, so this portion of the annual precipitation is not effective. High-intensity storms are common in summer, and a good share of this moisture quickly runs off the typically steep mine spoils. Drizzle-type storms are most effective, but these gently lingering-type storms are rare except in springtime. Actually, the effective precipitation utilized does not average 12.5 inches annually but is far less.

Approach to Reclamation

The picture sounds bleak and perhaps hopeless, but what can be done about this predicament and what are the chances of success?

The water problem has been attacked, not with irrigation using pit water that is potable and must be removed from the pit and can be utilized on spoils, but with water conservation plans and devices to trap and store most if not all of the natural precipitation. In doing this, the following are accomplished: (1) Control of excessive erosion typical of spoils; (2) establishment of a soil mosture reserve capable of sustaining a desirable vegetative cover; and (3) relief from the severe compaction of spoils that inhibits the rapid penetration of roots and moisture into the spoils.

Water is being conserved by--

1. Spoils drainage design and by reducing slope gradients.

2. Placing a porous absorbent layer of segregated materials on the surface of spoils.

3. Manipulating the surface to trap and store precipitation and increase infiltration.

4. Mulching with organic mulches to reduce evaporation and erosion and to increase infiltration.

Essential plant nutrients are being supplied by--

1. Addition of chemical fertilizers.

2. Planting of legumes and other nitrogen-fixing species.

3. Incorporating soils crops into the spoils, thus increasing organic material and plant nutrients.

Plant selection used to designate tolerant adaptable forage species includes--

1. Native perennial trials.

2. Introduced perennial selection.

3. Annual species testing.

4. Combinations of the aforementioned three methods.

This combination of interdependent approaches to reclamation has been effective and will become more so as experience is gained in coordinating the essential requirements for successful and rapid growth.

How practical is this system approach and how expensive? The answers to these questions depend strictly on the efficiency of the mining including simultaneous and coordinated processes of extraction and reclamation because these must become mutually beneficial. In practice, reclamation preplanning must always precede anticipated mining activity.

The concepts of reclamation are changing just as the rules of the mining game are changing. Mining as an extraction process by itself is not longer acceptable. Extraction simultaneously coupled with an efficient reclamation system is the new challenge. Reclamation in a mining sense must be composed of a system of coordinated processes used to achieve a preconceived result necessitated by excavation. Reclamation must be an aid to the extraction process or ways will be found to circumvent the responsibilities involved. Efficient coordinated systems of reclamation and extraction are basic because business is dependent on a reasonable margin of profit and common inefficiencies in reclamation are rapidly becoming intolerable. One important way that the extraction process can benefit from the reclamation investment is to apply this knowledge to the extraction process in predicting mining problems, in estimating both reclamation and extraction costs, and in developing more effective mining techniques and more efficient handling of materials. But to make this efficiency possible, sound basic preplanning must replace the happenstance of the past.

Planning for Reclamation

Five phases of planning are essential. Adapted to agriculturally oriented reclamation applicable to Montana, these are--

1. Preplanning studies.
2. Purpose, design, and reclamation plan.
3. Development plan, schedule, engineering, and shaping.
4. Surface manipulation, topsoiling, and revegetation.
5. Management.

Preplanning studies help predict the problems and costs of extraction as well as reclamation. Analyses of overburden cores provide pertinent information on toxicity, stratigraphic ratios, weathering rates, water infiltration, percolation rates, water holding capacity, and influences on seed germination and plant growth. Thus, spoils selection, efficient spoils handling, and reclamation techniques can be determined far in advance of extraction. Other essential preplanning studies include ground-water characteristics, surface-water-flow patterns, soils surveys, forage inventories, wildlife inventories, domestic grazing patterns, and archeological evaluation. These together form the irrefutable basis upon which to judge the success of reclamation; many are basically pertinent to predicting an efficient extraction technique or in purchasing the proper equipment to do the total job.

In Montana, a reclamation plan with the intended designated purpose must be stated, submitted, and accepted before a company can start a mine. This requirement is necessarily based on predictions, and the most sound predictions are provided by preplanning studies.

A development plan must evolve from a reclamation plan after it is modified to reflect actually encountered mining problems. It should contain (1) an adjusted mining technique to effectively segregate overburden and place selected spoils on the surface and (2) a mining schedule so that reclamation processes can efficiently be kept concurrent with extraction. Effective shaping of spoils to conform to a preconceived design is essential to promote total efficiency, to eliminate rehandling, and to minimize expensive drainage problems.

Surface manipulation is essential to trap scant moisture and concentrate it as a soils moisture reserve in quantities sufficient to insure seed germination and rapid establishment of seedlings, control erosion, and relieve compaction. Current regulations require that soil be salvaged and used when quantity and quality specifications are met.

Mangement is the key to sustained vegetative cover and effective land use. It applies to all land forms whether used intensively or extensively. The prolonged success of reclamation efforts in semiarid areas will ultimately be determined by effective management.

Systems for Water Retention

Several systems of water retention on spoils by surface manipulation are being tested including deep chiseling, gouging, offset-listering, and dozer basins. These systems are all being compared to determine relative efficiency.

Deep chiseling is a surface treatment that loosens compacted soils for a depth of 6 to 8 inches. The process creates a series of parallel slots in the surface on the contour of the spoils that effectively impede waterflow and markedly increase the infiltration rate. Chiseling forms a cloddy seedbed that is ideal to receive broadcast native seed mixtures. Some benefits of chiseling are relatively temporary because erosion from low-intensity storms may be sufficient to fill the slots and thus the initial high absorption rate diminishes. This treatment is effective on relatively flat slopes during the first spring to help establish a vegetative cover, which when adequate will be a lasting erosion control system.

Offset listering (fig. 1) is a surface configuration consisting of alternative elongated pits approximately 6 inches deep and 4 feet long. The design is especially effective in reducing saltation and in impeding runoff.

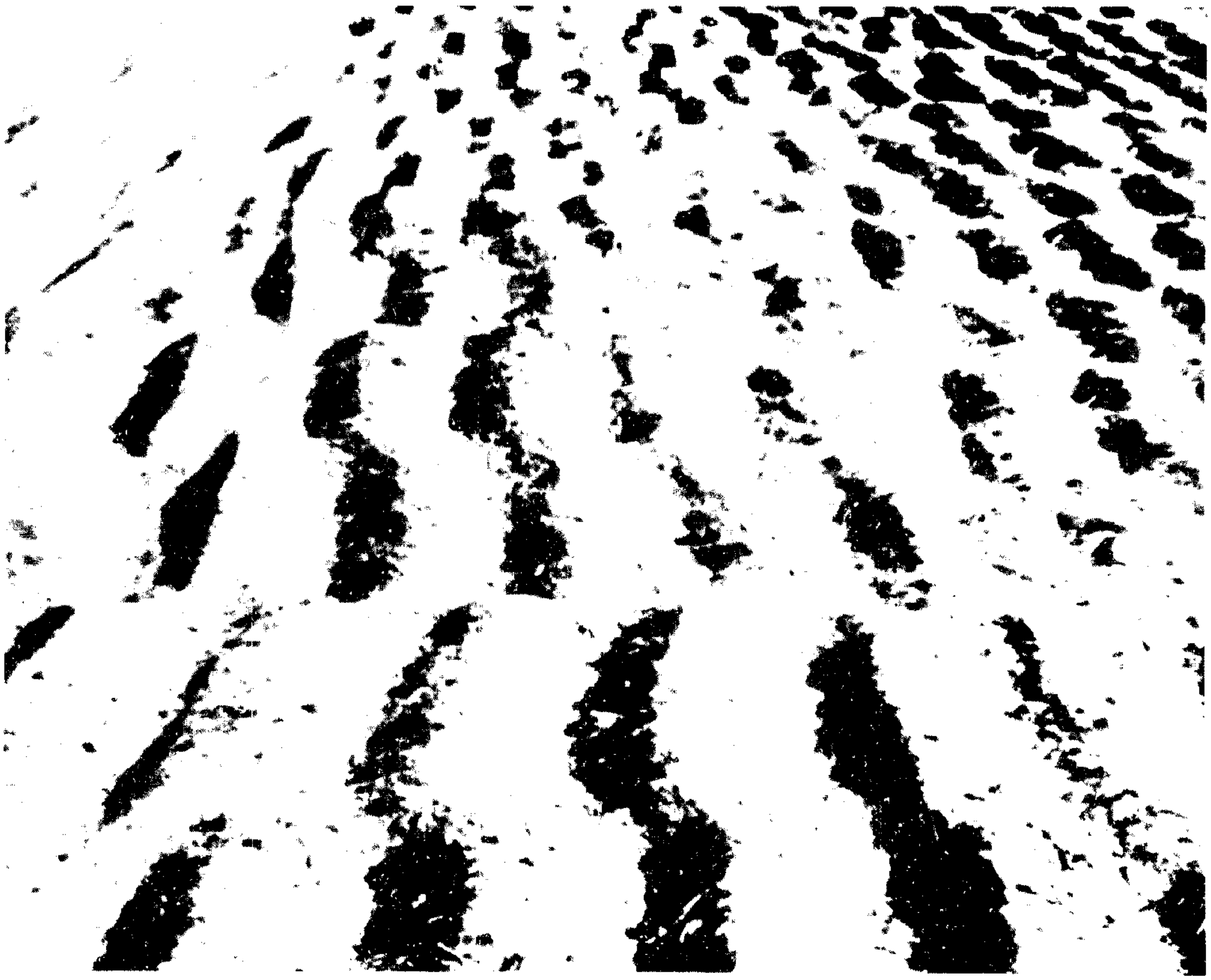

FIGURE 1. - Offset listering showing the shallow pits.

Gouging (figs. 2-3) is a surface configuration composed of many depressions approximately 10 inches deep, 18 inches across, and 25 inches long. This pattern is amenable to gradual slopes and flat areas. Gouging is surprisingly effective in conserving runoff from moderate-intensity storms and in causing differential melting of snow in the winter to retain snow moisture that would otherwise be lost. Gouging, too, creates a cloddy seedbed ideal for broadcast seeding.

Dozer basins (fig. 4) are large depressions designed to accomplish what terracing is intended to do but without the characteristic precision, hazards, and expense. Dozer basins are usually about 3 feet deep, 25 feet long, and placed on the rough contour at an interval of about 30 feet. This spacing is adequate to allow for combinations of treatments that will improve water retention capacity and overall efficiency. Precipitation intercepted within each minidrainage accumulates in the basin bottom in quantities sufficient to thoroughly saturate the basin limits. The increased soil moisture

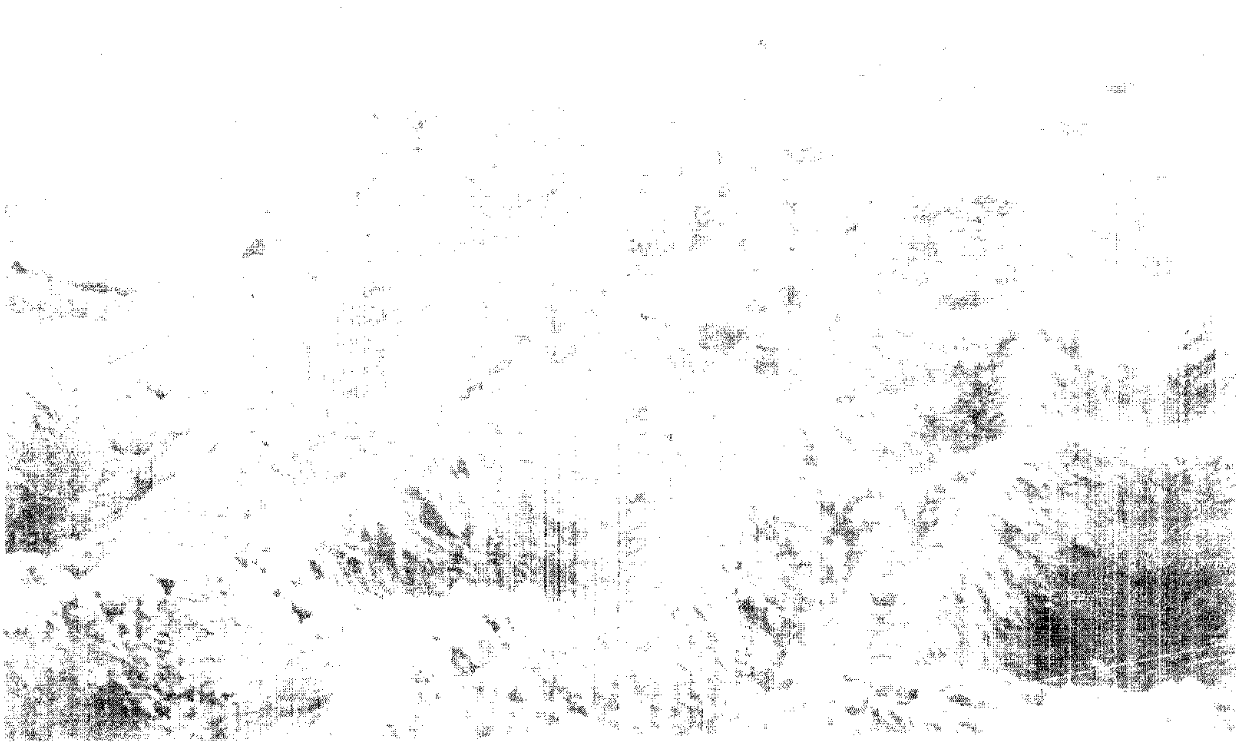

FIGURE 2. - Gouging to conserve runoff.

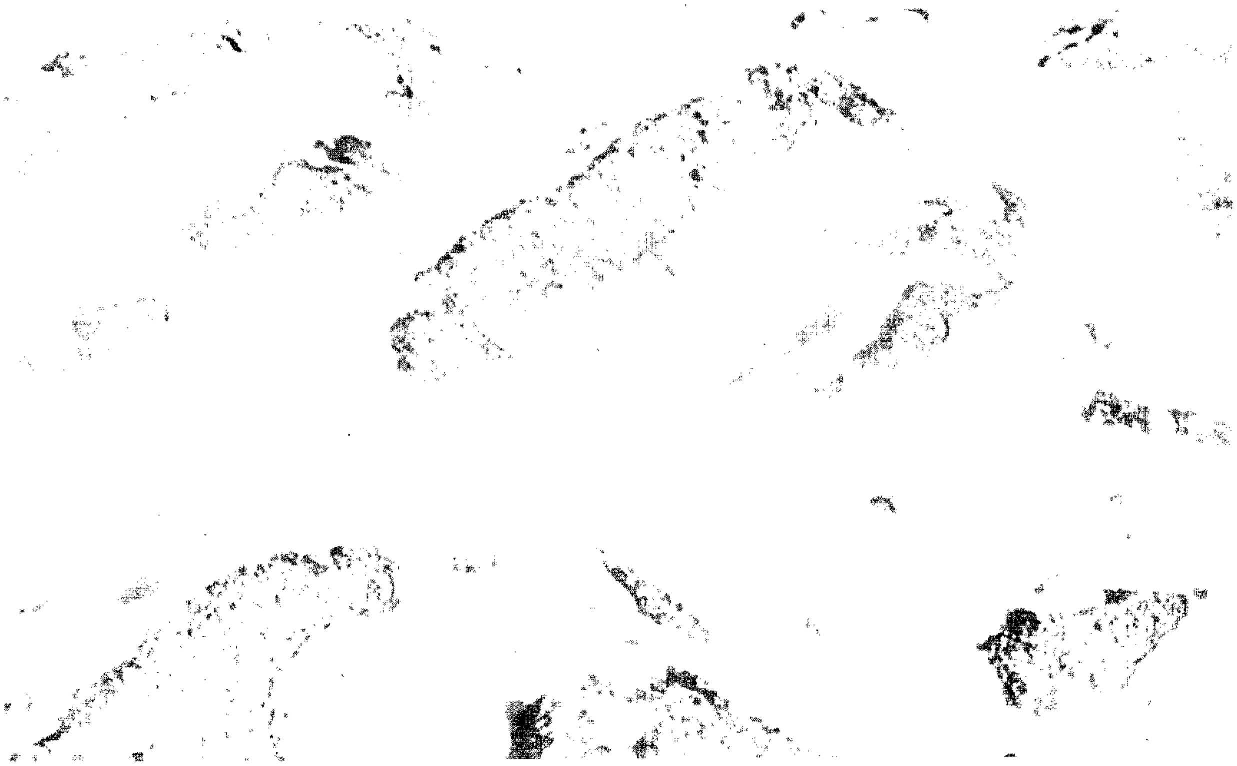

FIGURE 3. - Effect of gouging on snow retention.

FIGURE 4. - Dozer basins for collection of excessive runoff.

availability assures the establishment of a nucleus stand of vegetation the first growing season from which it can spread between basins to make a complete cover.

NEW DEVELOPMENTS IN OPENCAST MINING TECHNOLOGY AT REHEINISCHE BRAUNKOHLENWERKE AG, KOELN

by

Hans J. Leuschner[7]

Introduction

The largest lignite deposit of Western Europe, which covers 970 square miles and contains 60 billion short tons of brown coal, is located in West Germany. The area forms a triangle with points at Cologne, Duesseldorf, and Aachen, as shown in figure 5.

With modern technology about 13 billion short tons of lignite can be exploited to a depth of 1,650 feet in already developed and planned open mines. The seam averages 160 feet in thickness but occasionally branches out into as many as five separate seams.

During the last 2 decades mining technology was characterized by two developments: Growing annual production rates--from 66 million to 105 million short tons during the period between 1950 and 1972, and the shifting from the conventional flat-open mine to modern deep-open mines. Production data since 1950 are given in figure 6.

The profitableness of the Rhenish lignite industry is attained by use of large operating units that can exploit the benefit of cost reduction most efficiently. Consequently, production has been concentrated in a few large mines. By 1960, all lignite corporations in the area merged into only one corporation--the Rheinische Braunkohlenwerke AG. During 1950 to 1972, the number of operating opencast mines was reduced from 23 to 6.

Mining Practices and Utilization

The increasing depth of the mines is reflected by the growth of overburden excavation--59 million cu yd in 1950 compared with 280 million cu yd in 1972. The ratio "overburden volume to lignite volume," which indicates the profitableness of opencast lignite mines, soared from 0.8 to 1 in 1950 to 2.5 to 1 in 1972.

Realistic projects were developed for the opening of the Hambach mine. Five billion short tons of lignite reserves will be mined at a depth of 1,650 feet. The ratio of "overburden volume to lignite volume" will exceed 7.6 to 1.

For the time being, 80 pct of the lignite production, eventually reaching 125 million short tons annually, will be for thermal electric powerplants. The remaining 20 pct is for production of lignite briquettes. The district's

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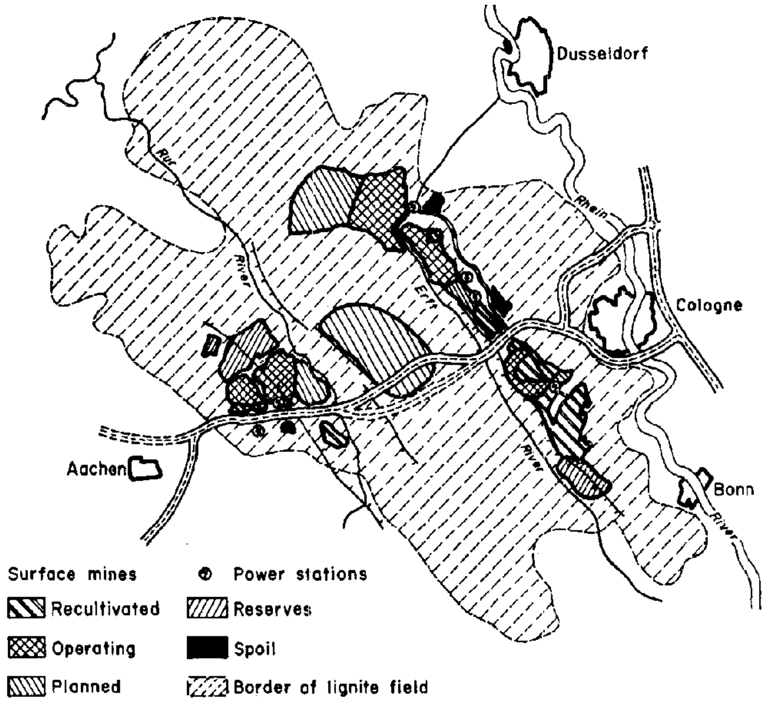

FIGURE 5. - Map of the Rhenish lignite district.

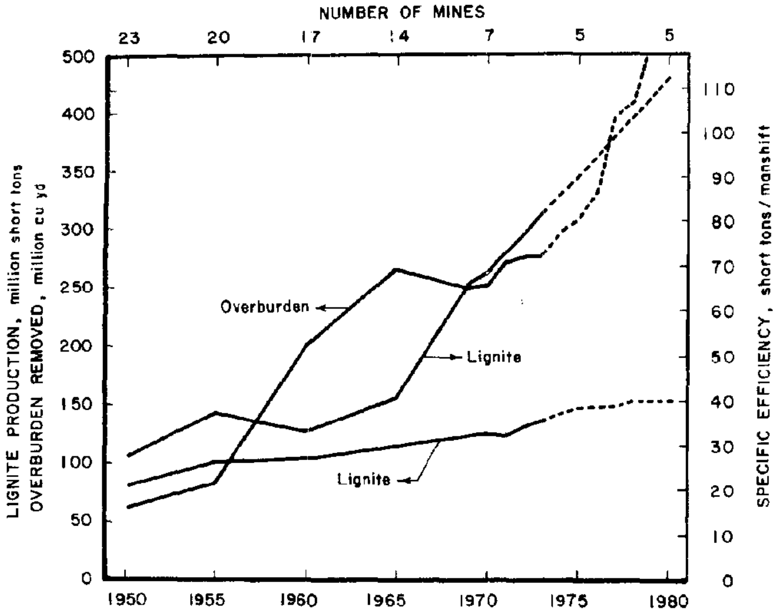

FIGURE 6. - Production data for the Rhineland lignite district, 1950 to 1980.

presently installed electric power capacity is 7,800 MW; by the end of 1975, the capacity will total 11,500 MW, at which time 85 pct of the lignite production will be for power station fuel. In 1972, the district contributed 22 pct toward West Germany's total electric power production from thermal powerplants.

The large volume of excavated material to be moved, and the fact that mining depths are now about 660 feet and in the future will be as much as 1,000 feet, indicates that bulk transportation of material will be the crucial problem to be solved by the Rhenish lignite industry.

Under these circumstances, strip mining is not feasible, nor could belt bridges--spanning the distance between the excavation and the spoil bank levels--be used. The deposit requires opencast mines of large dimensions where the overburden is excavated on several levels by continuously operating devices and carried by high-capacity transport equipment along the mine border to the spoil bank levels in the mined-out sections of the open pit. A plan map of the arrangement for mining is presented in figure 7.

In 1972, 42 million short tons of lignite and 97 million cu yd of overburden was excavated in the Fortuna-Garsdorf mine, which is the largest open mine of the district. This equals 206 million short tons of material, which is the largest recorded annual output ever reached by any mine.

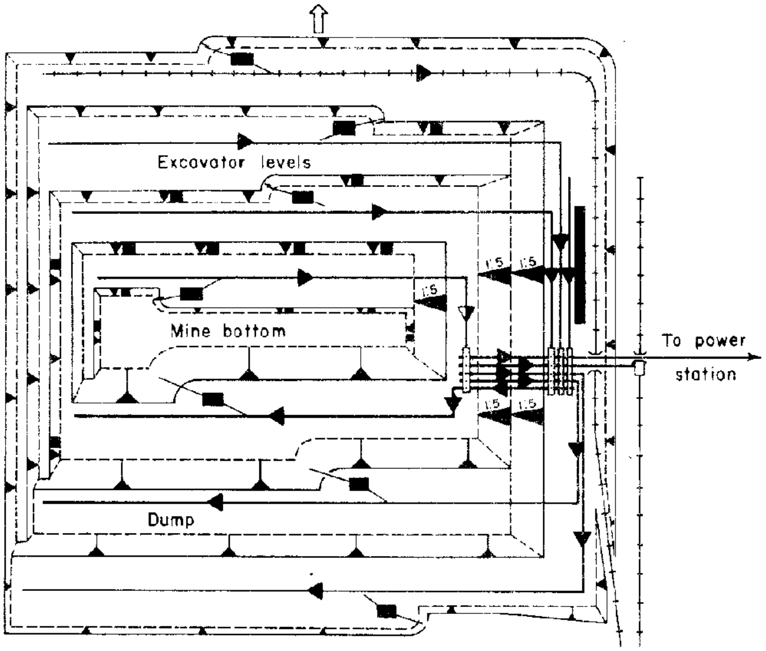

FIGURE 7. - Proposed mining and transportation arrangement.

Bucket-wheel excavators with a daily output of 130,000 cu yd of unconsolidated sediments are being used (fig. 8). Because the annual capacity of the Fortuna mine must be increased to produce 55 million short tons of lignite and 130 million cu yd of overburden in the coming years, additional machines with doubled capacity are envisaged. These 260,000-cu-yd-per-day bucket-wheel excavators are expected to streamline operations substantially and increase the specific efficiency per man per shift from 100 to 130 short tons of lignite.

Design and dimensions of bucket-wheel excavators

FIGURE 8. - Bucket-wheel excavator of 130,000-cu-yd-per-day capacity.

are decisively influenced by the desired excavation depth and characteristics of the soil. Stability of the slope depends on geologic strata. With increasing excavation height, the angle of slope must be reduced. This can only be attained through lengthening the bucket-wheel boom, which, however, tends to increase overproportionately both the total weight and the investment costs for the machine. An optimum relationship between capacity of the excavator and the height of excavation is necessary to keep the total excavation cost at a minimum. Another significant dimension is the bucket-wheel diameter, which is primarily gaged to the desired production capacity. Other important aspects are the ground stability of the various cut fractions and the demand for minimum specific transportation movements as related to the production volume.

It was deemed practical to adhere to the basic dimensions of the 130,000-cu-yd-per-day machine in development of the new bucket-wheel excavator. To guarantee unencumbered excavation and loading operations, the excavating step was isolated from the loading portion by means of a connecting telescoping belt bridge. The upward excavation height is 165 feet, and the maximum possible downward cut is 59 feet. The excavation range can be widened by lifting or lowering the excavator level above or below the level of the loading device, so that a total excavation height of 320 feet can be reached.

The machine measures 650 feet in length. A 70-foot diameter was selected for the bucket wheel, which is equipped with 18 buckets holding 8.2 cu yd each. With the 230-foot boom of the bucket wheel, a block width of 330 feet can be reached. The excavator weighs 29 million pounds. The installed electric power will total 14,000 kW of which 2,500 kW is for the drive of the bucket wheel (fig. 9).

For the development of the new machines, 2 decades' experience with the 130,000-cu-yd unit was available. Particular emphasis was given to maximum reliability of operation because shutdowns caused by operating disturbances are bound to affect the daily output of the mine proportionate to the increased capacity of individual machines. Consequently, the transportation system of the excavator was simplified, and the caterpillar drive was reinforced.

The doubled capacity of the excavator must be matched by corresponding developments at the stackers that dump the overburden. Capacity of the stackers will be increased by use of wider conveyor belts. The installed daily capacity of the stackers--310,000 cu yd per day--slightly exceeds capacity of the excavators so that temporary load peaks of the bucket wheel can still be dumped by the stacker without necessitating shutdowns. A three-belt stacker with a receiving belt, supported on a separate driving unit, was chosen to keep the construction weight as low as possible. Accelerated speed of the unloading belt of the 330-foot boom also permits a further decrease of the construction weight of the stacker and widens the range of discharge (fig. 10).

The working range of a stacker is determined by boom length and gradient of the discharge belt; for faultless operation, the gradient should not exceed 12°. The new machine is able to stack up to a height of 115 feet. The depth in downward stacking is influenced by the fact that the discharged material must be built up on the bottom of the spoil banks to solidify the slopes. Assuming a 35° dumping slope, the resulting height will be about 200 feet. To widen this range, the machine is designed to enable the stacker to be placed up to 65 feet below the level of the dumpside belt. The stacker's unloading range thus totals about 380 feet in height.

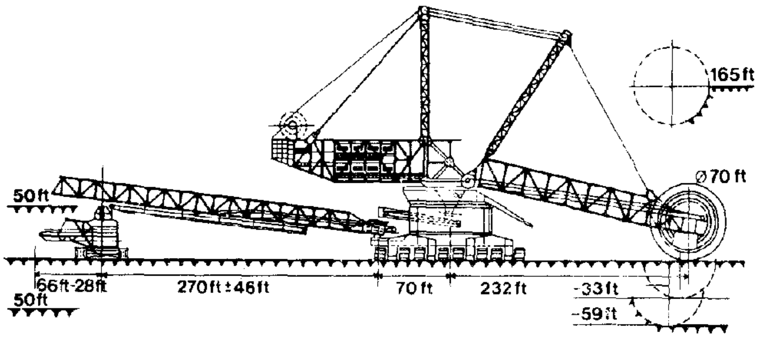

FIGURE 9. - Bucket-wheel excavator of 260,000-cu-yd-per-day capacity.

With a view to the enormous capacity of the new machines, load concentrations in one spot on the ground must be avoided to enable the building of stable spoil banks. Therefore, the unloading techniques tend to prefer dynamic unloading operations that keep utilizing the full swinging of the unloading boom.

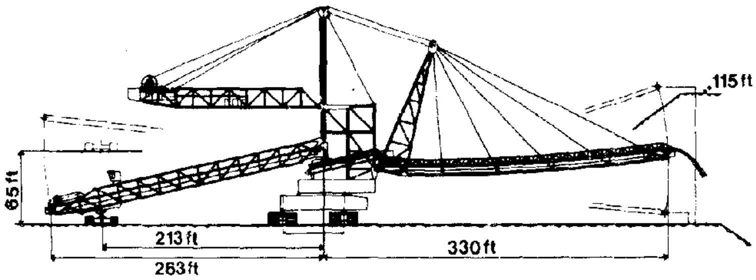

FIGURE 10. - Three-belt stacker having a daily capacity of 310,000 cu yd.

Transportation System

The immense transportation operations necessitated by the particular deposit characteristics absorb about 40 pct of the total operating cost. In 1972, 580 million short tons had to be transported. This transportation work equals 3 billion short ton miles. It is therefore essential that the transportation device be both most effective technically and most economical.

In the Rhenish district, railroad trains and conveyor belts are used almost exclusively for transportation. Over long distances and for small ascents, electric locomotives will continue to be used to haul the 125-cu-yd wagons containing overburden and the 149-cu-yd wagons containing lignite to the various coal consumers. A train moving overburden is shown in figure 11.

Within opencast mines, trains are usually not as applicable as conveyor belts because of their poor ability to climb. Also, in terms of streamlining operations, conveyor belts are superior to trains. All conveyor belts are

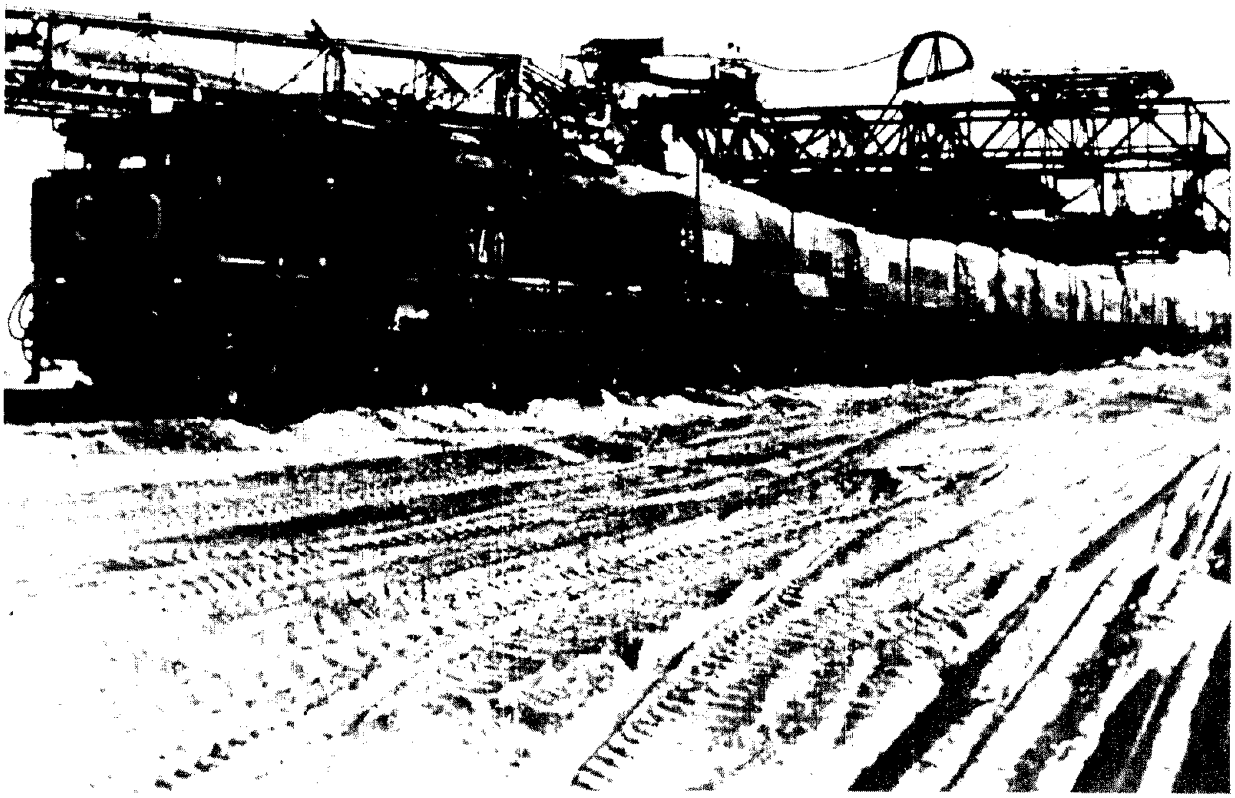

FIGURE 11. - Train moving overburden.

operated and controlled at only one dispatching desk. Automatic devices control the unmanned driving and transfer stations and will stop the operations in case of emergency. The most important device is installed in the loading funnels. A probe--based on electric capacity-monitoring--safeguards from overloading (fig. 12). Other essential control devices are being tried to check slipping of the belts on the driving drums, to monitor belt tensions, and to detect longitudinal cracks in the belts.

Automation keeps the man-hours for conveyor belt operations at a minimum. Similar reductions in railroad operations are limited because of the necessity for trackshifting.

For belt operation, the most economical system must be devised. Mine conditions are decisive. Size and shape of the minefield influence the length of the levels on which the volume transported depends. The dimension of the system also depends on the selected exploitation pattern. For so-called parallel operation, level belts lead to front-end belts, which connect the excavation levels with the spoil bank levels at the border of the opencast mine. The material must be distributed to the different spoil bank levels to build up stability, and lignite must be kept separate from overburden; therefore, each level must be equipped at the junction points with transfer devices having back-and-forth sliding heads to permit loading on the different belts

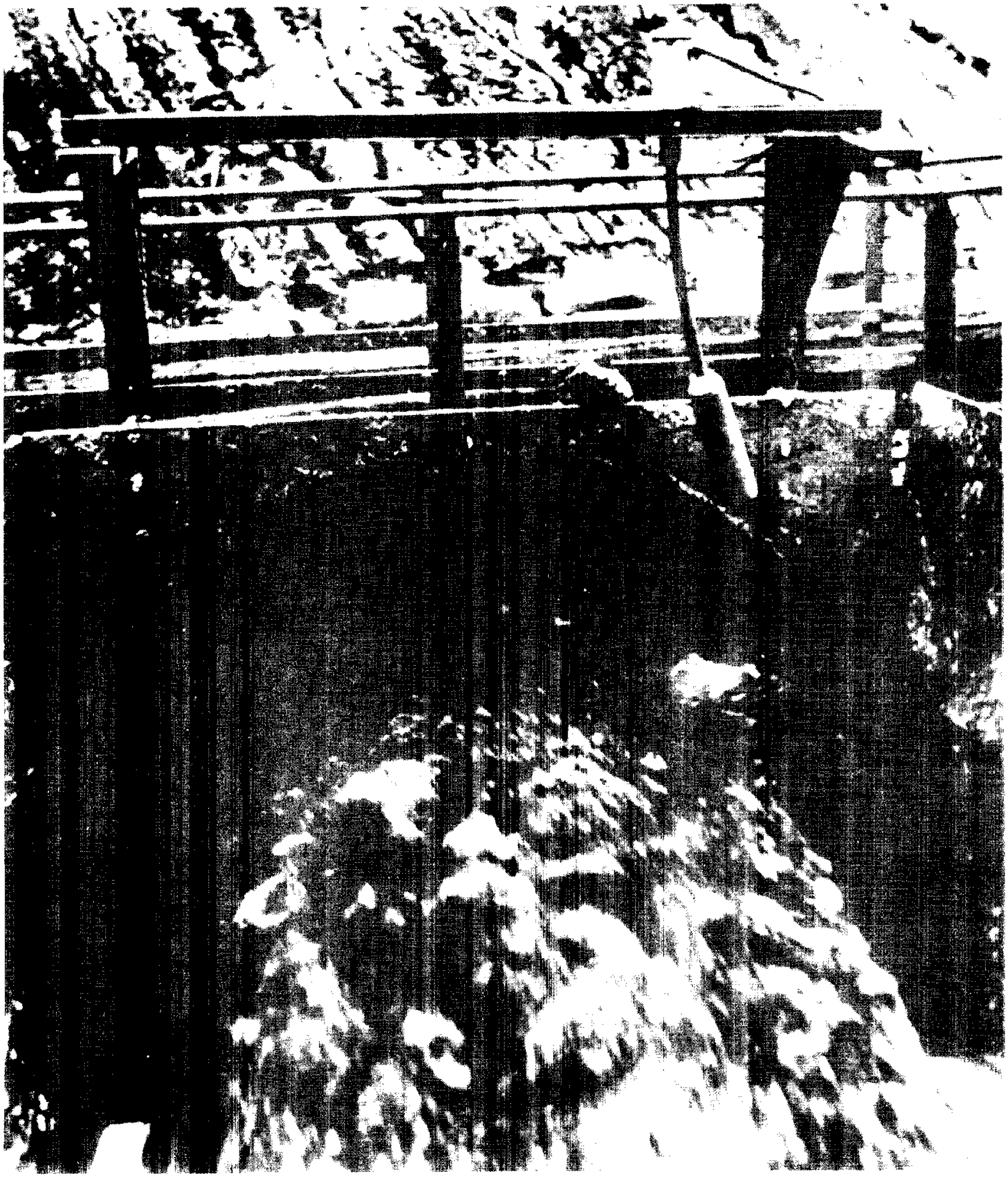

FIGURE 12. - Monitoring device to indicate overloading at a belt station.

that run across to the excavator level belt. This arrangement is shown in figure 13.

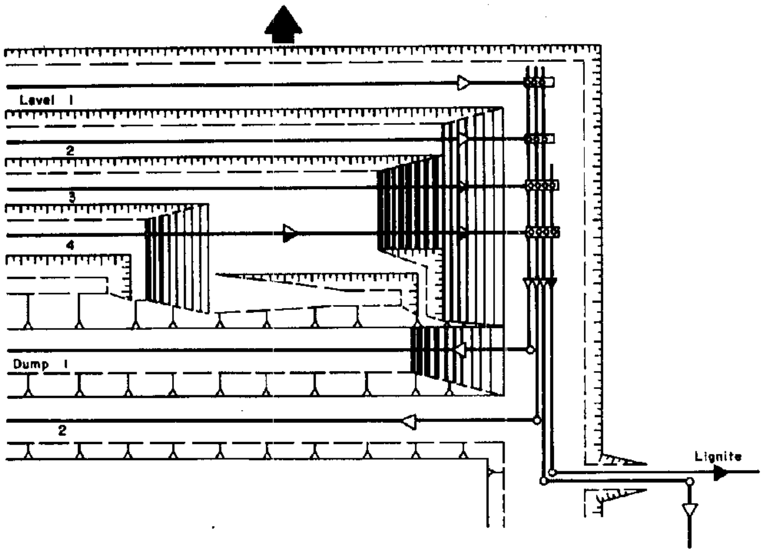

FIGURE 13. - Parallel operation guiding of conveyor belts.

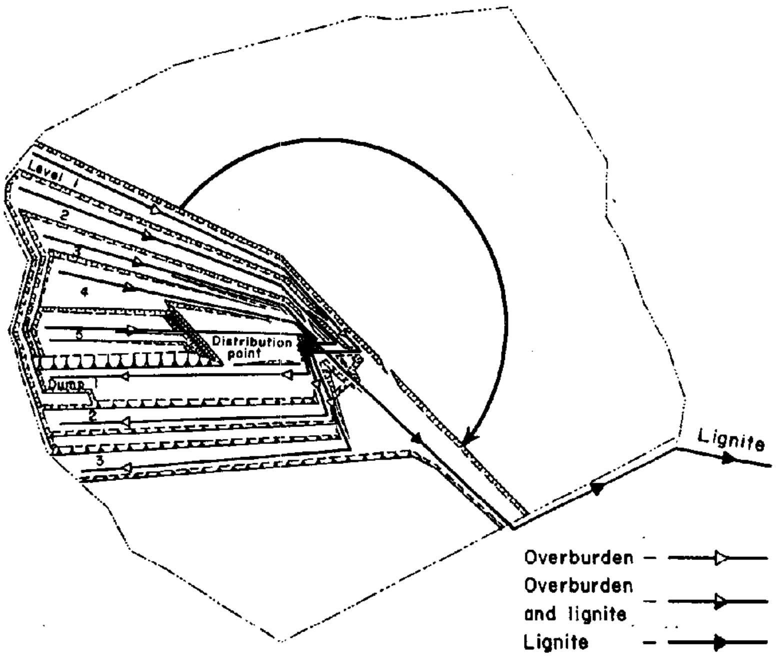

FIGURE 14. - Swinging operation guiding of conveyor belts.

For the swinging operation pattern, a distribution point is established--if possible in the center of masses--around which the excavating and spoil bank levels are swung. The belt ways of the different levels meet at this point. Figure 14 shows this configuration.

Both exploitation patterns are, however, not quite satisfactory in that the material is transported from the excavation point along the mine border to the spoil bank site.

A third possibility, the so-called diagonal belt conveyor pattern, was recently used for the first time in the Rhenish district. With this method, the excavation and spoil bank sides are connected diagonally, substantially shortening the distance for transport. The disadvantage is that all material to be transported passes to the bottom of the mine and is hoisted from there to the individual spoil banks or to the lignite hopper. The savings involved in reduced transport work will, however, offset this defect. The diagonal method is illustrated in figure 15.

Conveyor Belts

The operation of the new types of excavators and stackers requires modern belt conveyor systems having twice the capacity of previous models. The belts are designed for an hourly capacity of 33,000 short tons. Driving stations can be equipped with up to six electromotors, each one with a capacity of 1,500 kW. The planned width of the wire-rope armed belts (manufacturing type St 5,400) is 118 inches. The belt speed will be 21 fps.

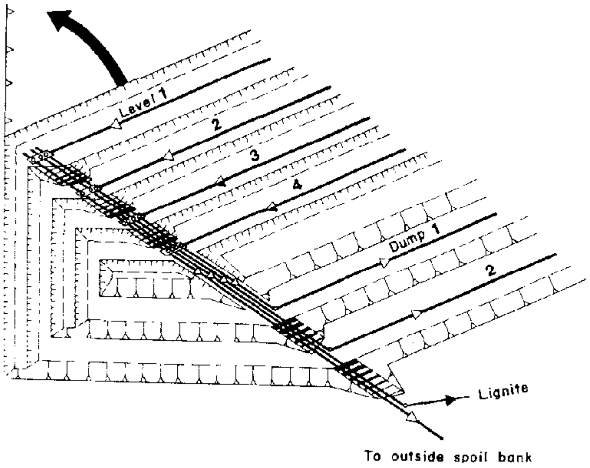

FIGURE 15. - Diagonal belt conveyor between mining site and dump site.

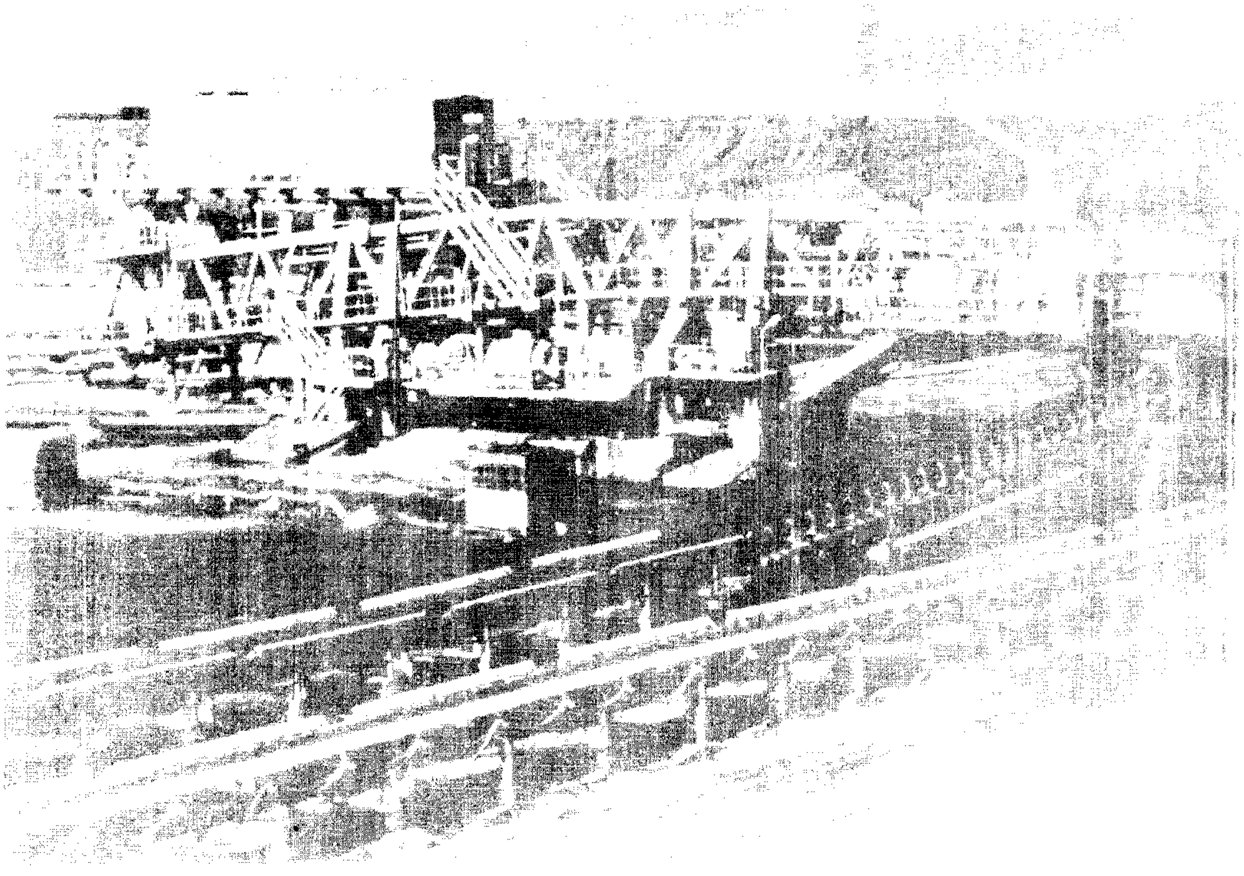

FIGURE 16. - Conveyor head station with 1,500-kW driving units.

One of the modern 1,500-kW driving stations is already operating at the Fortuna mine. Permanently installed compound-walking devices enable

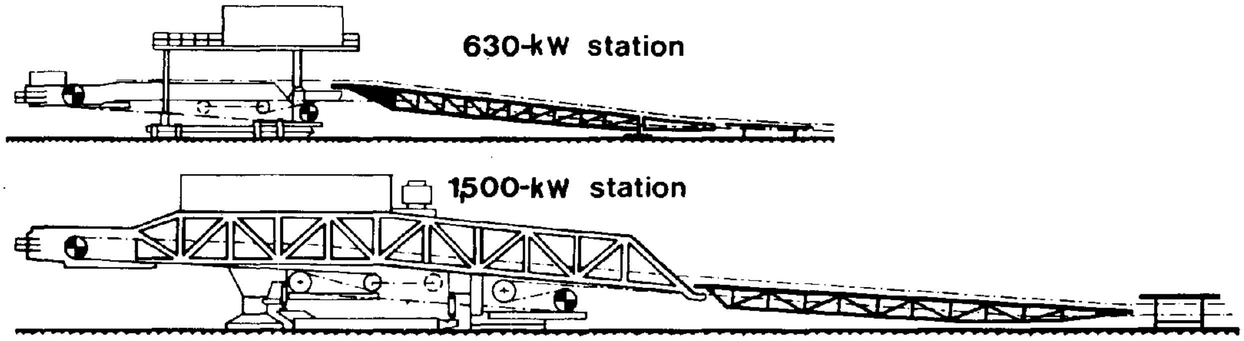

FIGURE 17. - Comparison of 630-kW and 1,500-kW head stations.

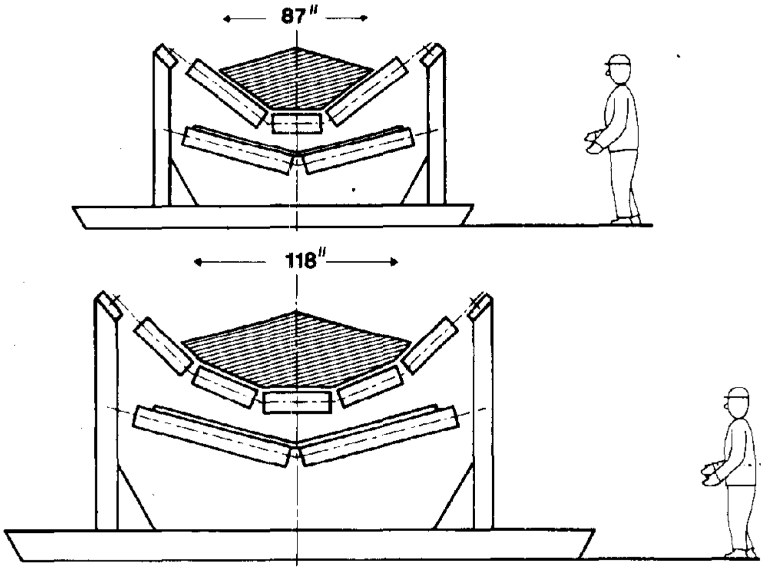

FIGURE 18. - Cross-sectional view of conveyor belts.

self-propelled motion of the 1.8-million-pound station when the belt system is shifted. The conveyor head station of this installation is shown in figure 16. The size of the new 1,500-kW station is compared with that of the 630-kW station in figure 17.

Corresponding to the driving stations, the individual belt frames will likewise be substantially enlarged. Figure 18 shows the differences in dimensions for the two units. Repair work at these belt systems such as replacement of rollers will not be feasible without mechanical assistance.

Development of new machines and belt conveyors indicates that the Rhenish lignite mining industry has decided on capacity increases of opencast mining machinery to attain streamlining and cost reductions. Nevertheless, another increase in efficiency by raising the hourly capacity substantially beyond the 33,000 short tons mark will not be feasible unless transport technology takes a new course. In particular, new belt driving devices are necessary. To meet this challenge, the Rhenish mining industry has joined forces with various scientific institutions and numerous companies of the component industries to find solutions to the problems.

Spoil Reclamation

In addition to the problems of opencast mining technology, several problems must be solved that stem from deep open pit mining covering large areas.

These could be summarized under the heading "environmental impact." Especially two environmental factors influence the costs involved. First, the Rhenish lignite district contains a population of 1 million at a density of 1,070 inhabitants per sq mi, making it one of the most densely populated areas in Europe. The United States has about 47 inhabitants per sq mi. Second, the lignite is covered by a layer of loess, which is one of the most fertile soils in the entire European community and is intensively cultivated. The major crops are grain and sugar beets, with some vegetables being grown in the immediate vicinity of major cities.

Large operating units with equally large production capacities are necessary to find and to finance technically feasible and economically reasonable solutions to the environmental problems that develop during and particularly after mining exploitation.

The mining areas include public railroads, highways, and even rivers. As mining operations expand, these have to make way, which means that houses, farms, and whole villages have to be moved. Since the beginning of the present mining operations, 44 villages with 18,500 inhabitants were resettled. An additional 9,000 people will be moved in the next 2 decades. In connection with the resettlement of villages, the necessary technical and organizational measures are combined with efforts toward improved social conditions. Thus, the structure of the new villages and communities is considerably enhanced, as shown in figure 19. Upon completion of the mining operations, the surface of the exhausted mines is restored so that the entire area can be used.

The major portion of the mining districts in the north was once mostly cultivated land. Accordingly, efforts are made to reclaim a matching portion of the spoil banks for agricultural purposes. To attain this goal, the loess, which may be as thick as 65 feet, is useful. The loess is excavated separately and a 3- to 7-foot layer is spread over the spoil bank surface either by dry spreading or by sluicing.

The south mining district was not cultivated when lignite mining began. Here, the high slopes of the spoil banks are reclaimed by forestation. The surface of the spoil banks is covered by a mixture of gravel and loess, and then planted with some 36 species of trees and 18 species of shrubbery. Lakes are created in the remaining unfilled hollows as shown in figure 20. The new landscape developed in the wake of lignite mining--lakes and woods in the southern district and farm lands interspersed with wooded slopes in the northern district--add valuable recreational areas benefiting the inhabitants of this densely populated country.

Only proper reclaiming will insure that further demands for land to be used for open lignite mining will be granted. Governmental authorities and autonomous corporations including representatives of the local population are watching the operations, and all must be satisfied. Moreover, it is imperative to familiarize the public of the exploitation measures and their economic significance. An information center has been established in an old moated castle, and annually about 50,000 visitors are informed about lignite mining. The center is shown in figure 21.

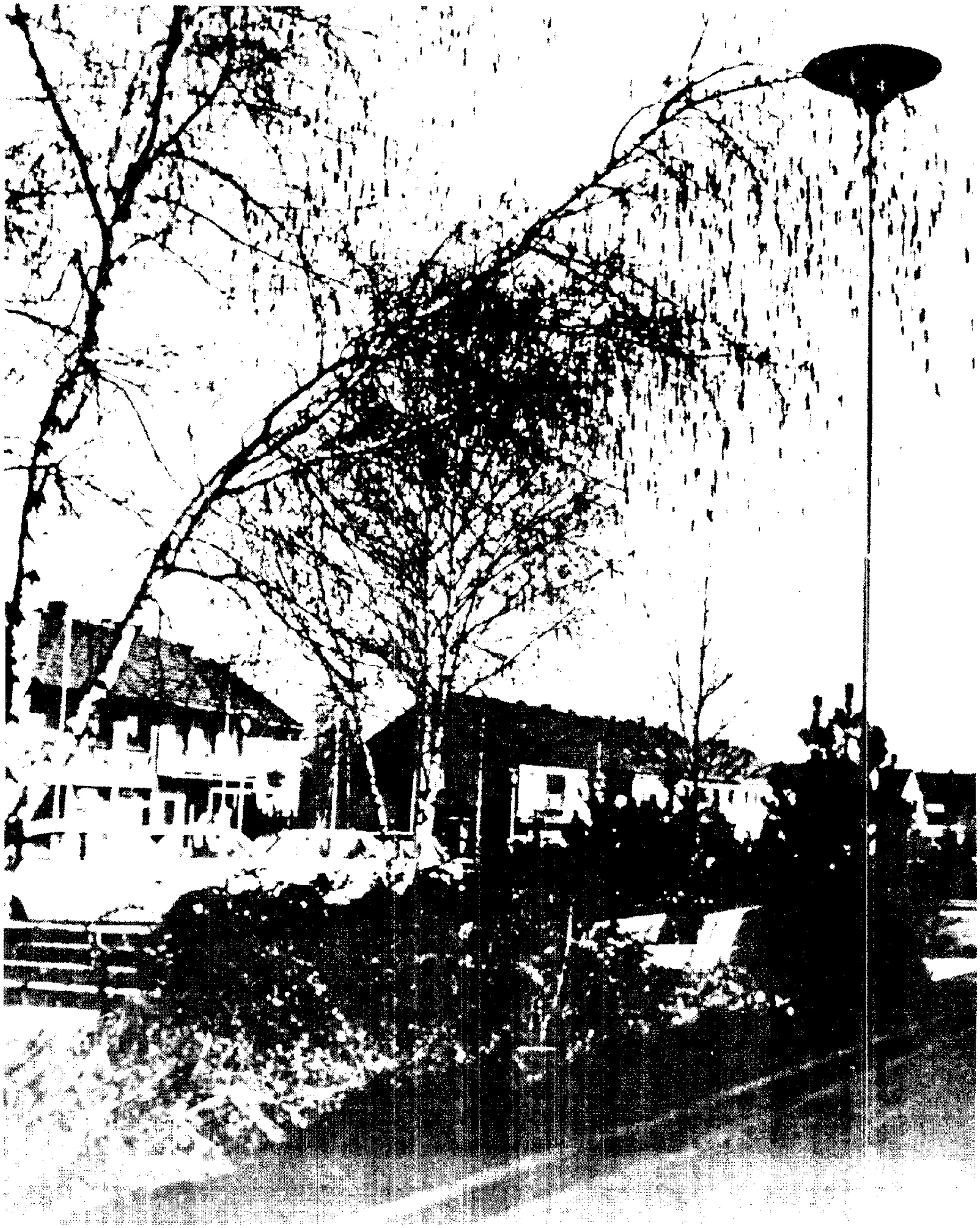

FIGURE 19. - Neu-Moedrath, a resettled village.

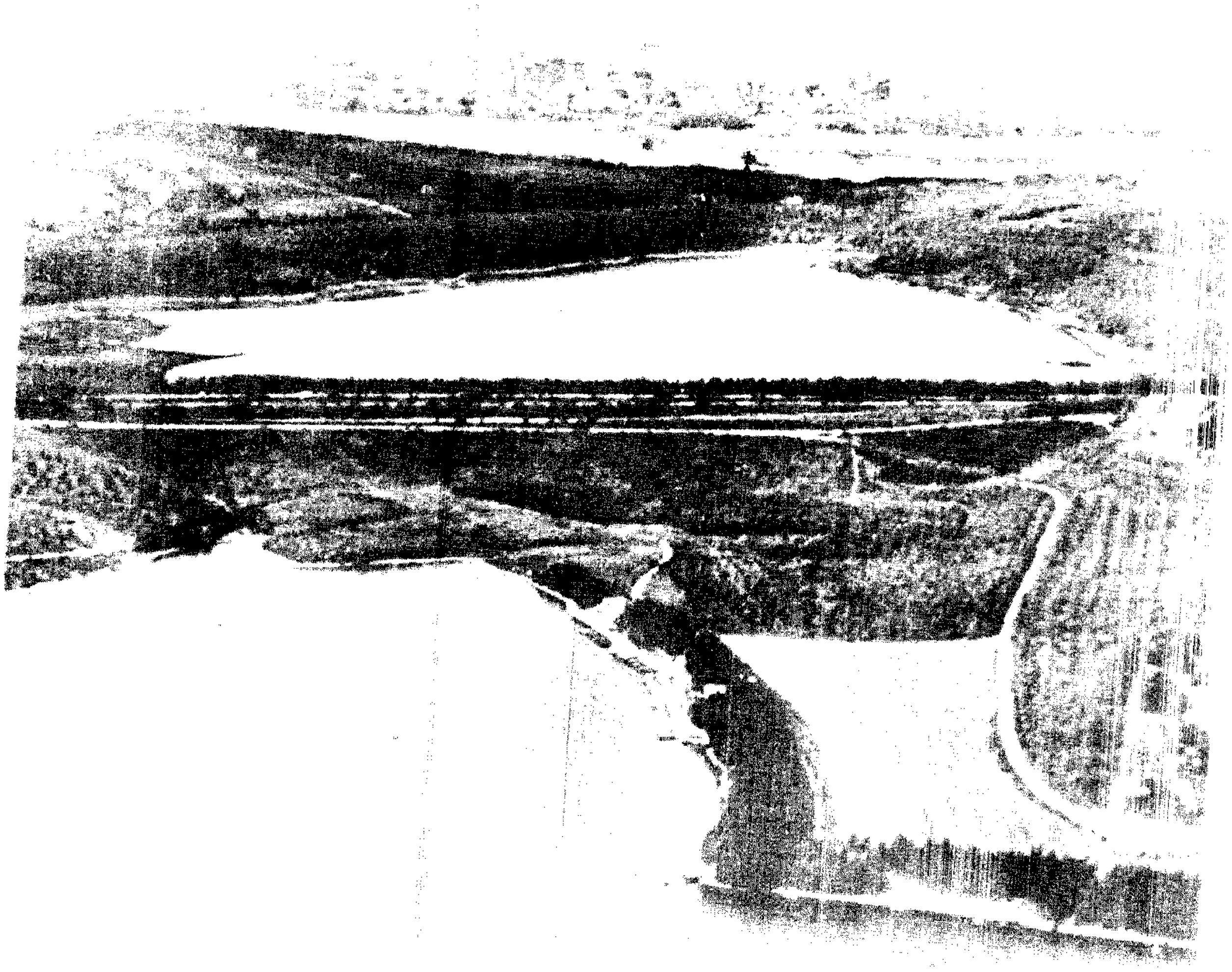

FIGURE 20. - Woods and lakes in a reclaimed district.

FIGURE 21. - Information center at Castle Paffendorf.

BIG STONE PLANT: DESIGN FEATURES AND FUEL HANDLING

by

Orville B. Johnson[8] and Robert F. Middleton[9]

Introduction

The Big Stone plant will be a 440-MW lignite-fired electric generating plant located in Grant County near Milbank, S. Dak. A joint venture of Montana-Dakota Utilities Co., Northwestern Public Service Co., and Otter Tail Power Co., the plant is scheduled for commercial operation by May 1, 1975. Otter Tail Power Co. has been appointed agent for the owners with the responsibility and authority to build, operate, and maintain the plant. Engineering design and construction was awarded to Bechtel Power Corp. of San Francisco, Calif. Areas served by the three companies are shown in figure 22.

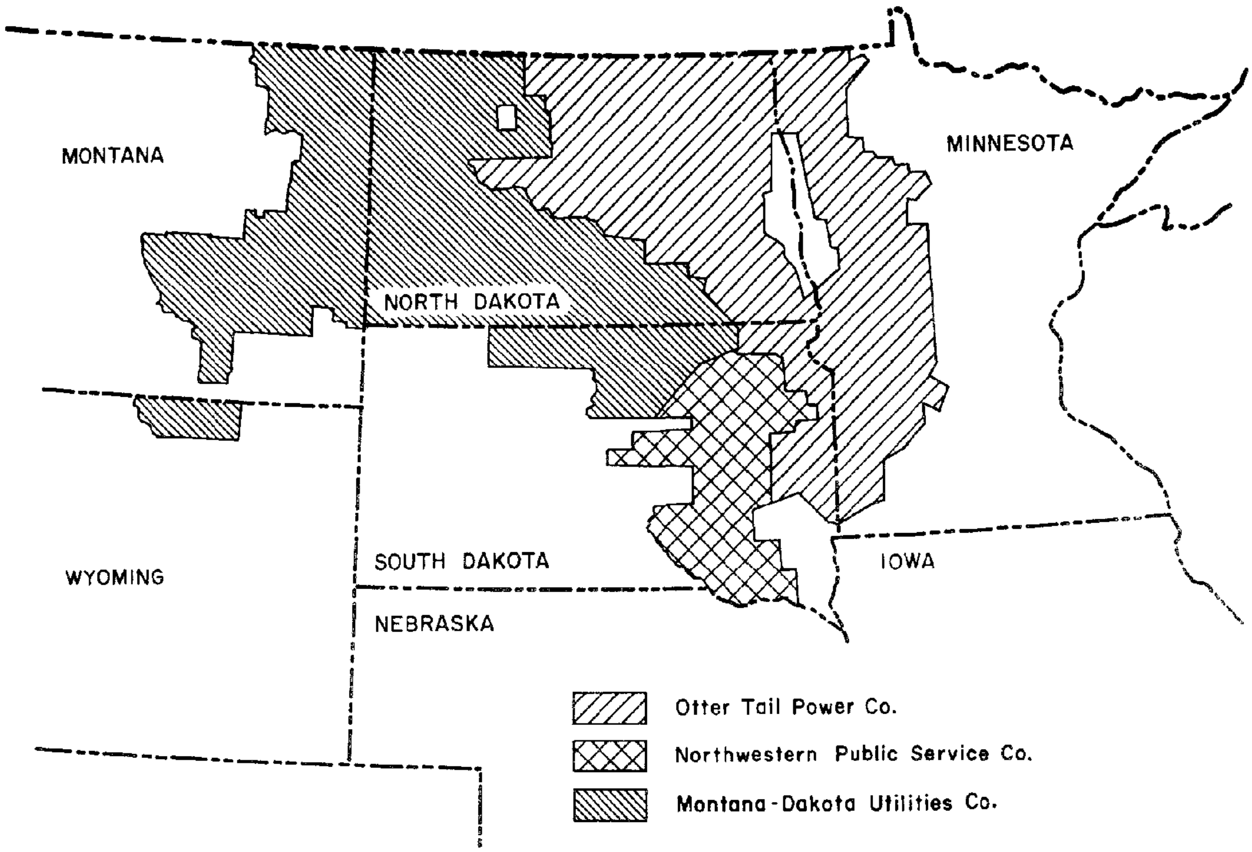

FIGURE 22. - Service areas of cooperating companies.

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Both authors are with Otter Tail Power Co., Fergus Falls, Minn.

Various committees, comprised of members of the three companies, are delegated the responsibilities of overseeing the details of construction and operation. The Coordinating Committee, consisting of A. V. Hartl, President of Otter Tail Power Co., Chairman, A. D. Schmidt, President of Northwestern Public Service Co., and David Heskett, President of Montana-Dakota Utilities Co., has the responsibility to provide liaison between the owners and to supervise and coordinate the functions of the other committees. Accounting, budgeting, auditing, and financial matters are the primary responsibilities of the Auditing Committee. Design, construction, operation, maintenance, and general supervision of the agent are the function of the Engineering and Operating Committee. Some significant dates relative to the joint venture and plant design are as follows:

1. September 9, 1969--The three companies signed a letter of intent for the Big Stone plant.
2. October 1969--Bechtel Power Corp. was hired as design engineers and constructors.
3. November 1969--Plant site selected by Bechtel and approved by the three companies.
4. February 1970--Commitment with Westinghouse Corp. was made for a major equipment package, including turbine-generator, condenser, isolated phase bus, transformers, switchgear, feedwater heaters, large motors, auxiliary turbines, and forced-draft fans.
5. July through September 1970--Permit applications to regulatory agencies of South Dakota and Minnesota.
6. December 1970--Commitment to Babcock & Wilcox for the steam generator.
7. May 1971--Contract for earthmoving, site preparation, and construction of the cooling pond.
8. June 17, 1971--Earthwork was started at the site.
9. December 22, 1972--Contract for the emission control system awarded to Wheelabrator-Frye, Inc.
10. March 1, 1975--Construction completed.
11. April 30, 1975--Preliminary operation and testing completed.
12. May 1, 1975--Commercial operation.

Site Evaluation

Development of the Big Stone plant came about through several engineering studies. Site evaluations were conducted by Bechtel Power Corp. Of several sites considered, a location utilizing water from Big Stone Lake was determined to be the most feasible for a 200- or 400-MW unit. Because Otter Tail Power Co. already had a generating station at Ortonville, Minn., it would seem logical to add the new unit to the existing station. However, physical limitations were such that the new unit would have to be separated from the existing plant. Because of the space limitation of the site, the economics of using the existing plant personnel to operate both units could not be realized and it was decided to develop a new site. Figure 23 is a schematic plan showing locations of major items at the Big Stone site.

Stone & Webster Management Consultants, Inc., of New York conducted a joint generation study for the three utilities. The conclusion from this

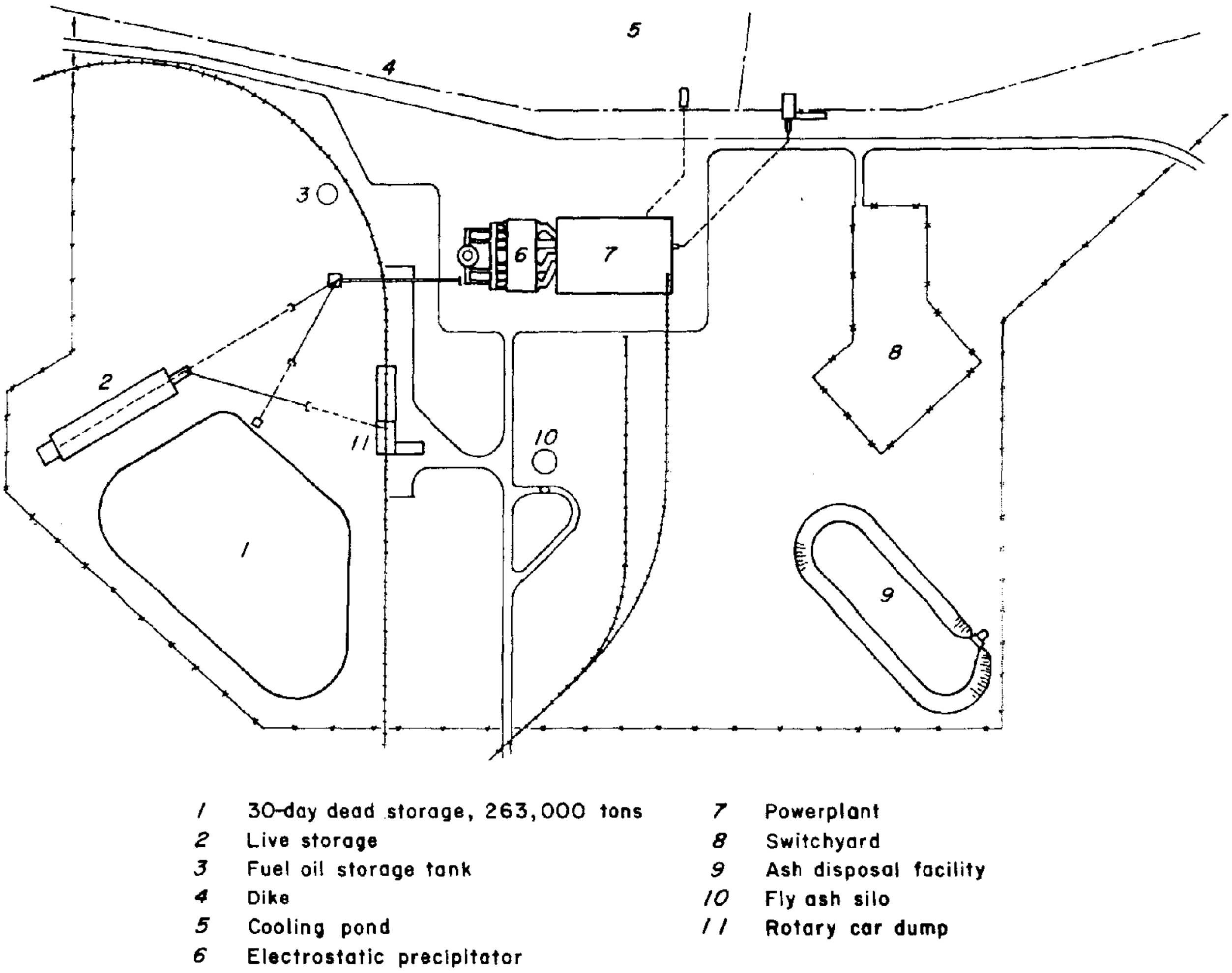

FIGURE 23. - Location of major items at Big Stone site.

study and the one by Bechtel was that a 400-MW station would be constructed in South Dakota adjacent to Big Stone Lake to utilize makeup water from Big Stone Lake. The basic design feature of the site is a cooling pond with provision for zero discharge of waste water. The zero discharge concept was conceived and developed in conjunction with Bechtel Power Corp. by H. R. Cowles. Until his retirement in December 1972, Cowles was Vice President, Planning, for Otter Tail Power Co. and Project Manager for the Big Stone plant.

Water Requirements

Applications were made to and permits granted by the following agencies to enable appropriation of water from Big Stone Lake for plant use:

South Dakota Water Resources Commission
Minnesota Department of Natural Resources
Department of the Army, Corps of Engineers
South Dakota Department of Health, Division of Sanitary Engineering and Environmental Protection

Minnesota Pollution Control Agency
South Dakota-Minnesota Boundary Waters Commission
South Dakota Department of School and Public Lands
Upper Minnesota River Watershed District
East Dakota Conservancy Sub-District
United States Environmental Protection Agency.

Under the terms of the various agreements, 7,000 acre-ft/yr from the Big Stone Lake may be appropriated for plant use. Because of restrictions based on lake levels, a pumping station was constructed on Big Stone Lake, as shown in figure 24, including one 10-cfs pump and two 50-cfs pumps. This arrangement allows flexibility of high pumping rates during spring runoff and high lake level conditions and a reduced pumping rate during normal lake level conditions. Because no pumping is allowed below a minimum lake level, sufficient storage is provided in the cooling pond to take care of evaporation losses during those periods.

The cooling pond and eventual evaporation ponds are shown on figure 25. Water from Big Stone Lake is pumped into a 5-acre boiler makeup water pond and overflows from this pond to the 340-acre cooling pond. Computer operation studies of the pond with 320-cfs circulating waterflow at maximum load conditions indicate the average maximum condenser-inlet-temperature will be 85° F or less with a 30° F temperature rise through the condenser. Maximum temperature conditions with high ambient temperatures and low pond levels are predicted to be 95° F at condenser inlet and 125° F at the condenser discharge.

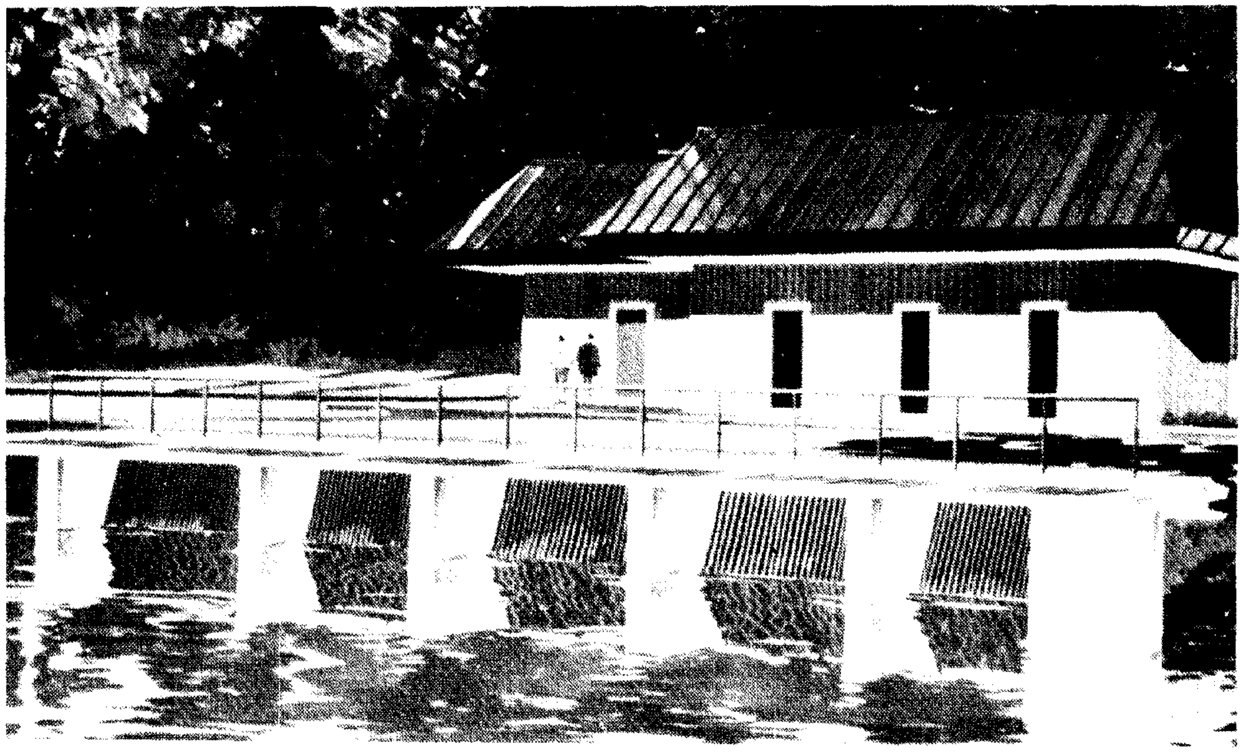

FIGURE 24. - Pump intake structure on Big Stone Lake.

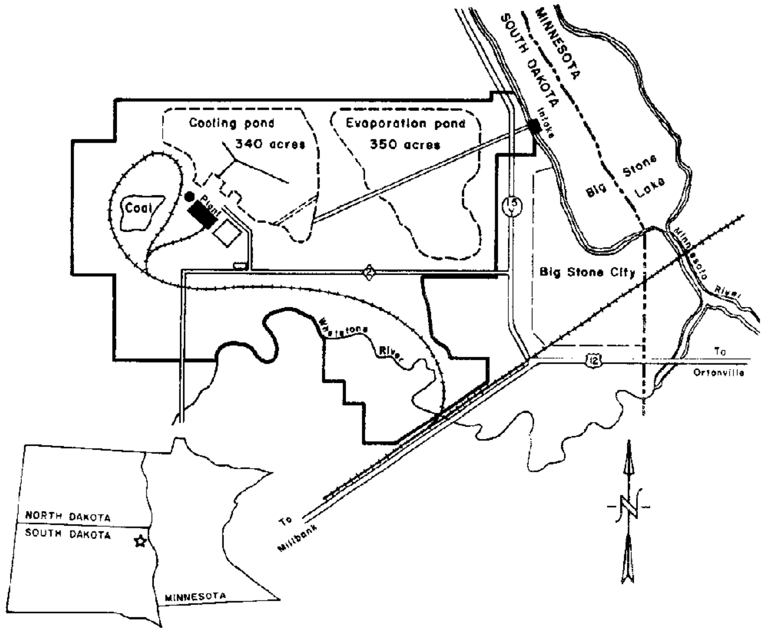

FIGURE 25. - Cooling and evaporative ponds for Big Stone Lake plant.

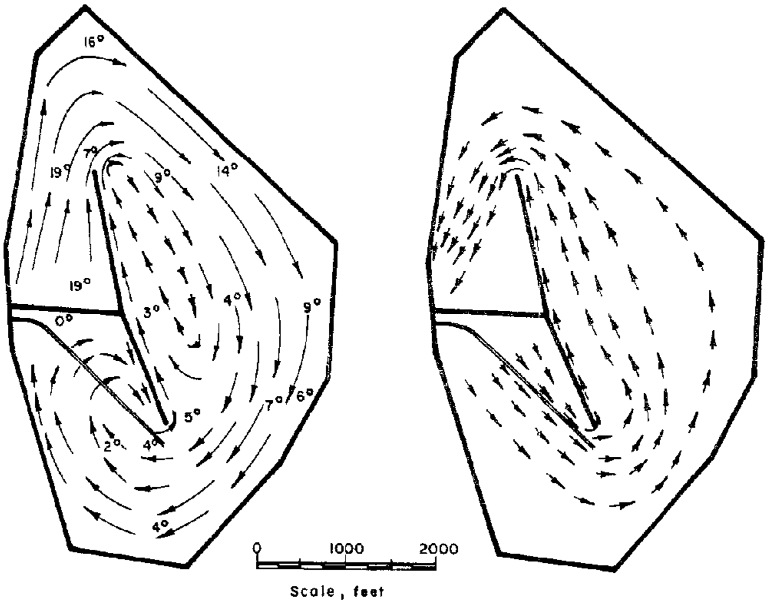

FIGURE 26. - Model and predicted flow patterns for cooling pond.

To determine the optimum location of baffle dikes for the cooling pond, a hydraulic model study was conducted by J. F. Ruff of Colorado State University, Engineering Research Center, Civil Engineering Department, at Fort Collings, Colo. Figure 26 shows a picture of the model with surface flow patterns and temperatures of the final baffle dike arrangement. It is interesting to note that subsurface flow recirculates and immediately reduces the discharge water from 31° F above inlet temperature to only 19° F above.

Blowdown water from the cooling pond will ultimately be pumped to evaporation ponds for disposal. It is expected that the plant will be able to operate on the cooling pond for 6 years before concentrations are sufficiently high that blowdown will be required.

The pumping station, pipeline, and cooling pond were completed in 1972. Pumping operations were begun in November, and the pond was filled to normal operating level by early January 1973.

Turbine-Generator

Although the turbine-generator contract was awarded to Westinghouse in February 1970, a comprehensive study was conducted by Bechtel prior to actual selection of a specific unit. Two basic units were considered, both 400 MW, 2,400 psig/1,000° F/ 1,000° F tandem/compound, one unit to have two-flow 31-inch last row blading (LRB) and the other to have four-flow 23-inch LRB. The economic comparison

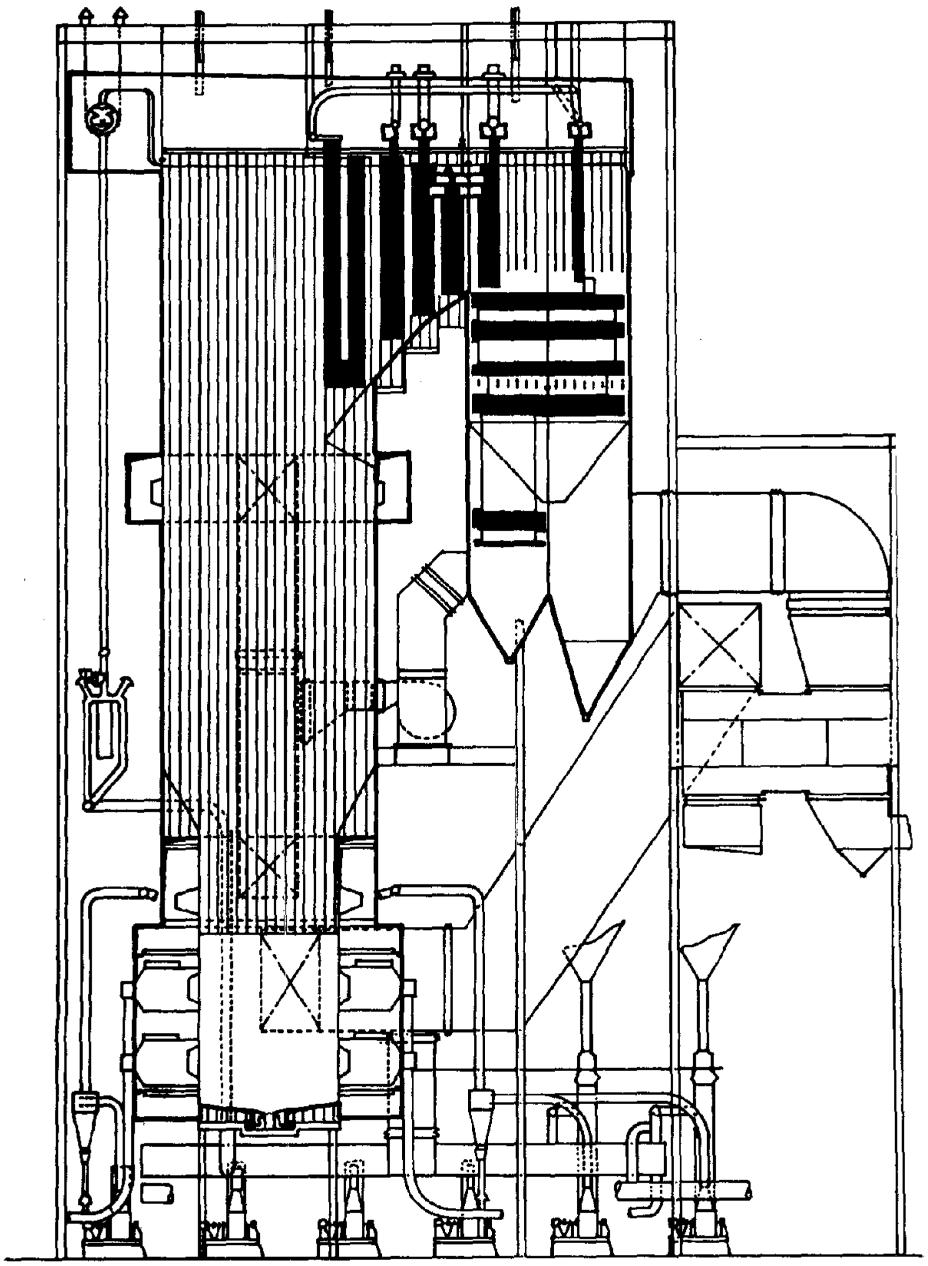

FIGURE 27. - Cross section of boiler for Big Stone plant.

favored the two-flow 31-inch LRB. However, there was considerable reluctance to accept this unit because of many failures the industry had experienced with 31-inch blading and the reduced capability because of steam flow restrictions in the low-pressure turbine. Westinghouse presented conclusive evidence to allay any fears of blade failure. Boiler negotiations that were proceeding during this same period helped influence the choice of the turbine. By utilizing exhaust steam from the intermediate pressure turbine to drive auxiliary turbines for the boiler feed pumps and forced draft fans, it was possible to increase the net capability of the station. The choice was made to purchase the tandem-compound, single-reheat, double-flow unit with 31-inch LRB. The nameplate rating of the unit is 414.6 MW at 2,500 psig/1,000° F/1,000° F, 3.25 inch Hg. The net output of the unit with 5 pct overpressure and maximum steam flow of 3,250,000 lb/hr will be 440.0 MW.

Boiler Selection

Choosing a boiler developed into a technical and economic comparison of pulverized versus cyclone firing. The higher horsepower requirement of the cyclone unit was compensated by greater net plant capability because of auxiliary turbine drives on the forced draft fans and boiler feed pumps. In addition a pulverized coal-fired unit would have required additional building size. With gas recirculation, the cyclone unit adequately met the temperature limitation of 1,950° F leaving the furnace and entering the first convection pass. One of the factors that had a considerable bearing on choosing the Babcock & Wilcox cyclone-fired boiler was the successful operation of the cyclone unit at Minnkota Power Cooperative's Milton R. Young plant at Center, N. Dak. A cross-sectional view of the 48-foot-wide by 45-foot-deep boiler is shown in figure 27. At 3,250,000-lb/hr steam flow, the fuel-firing rate will be approximately 365 tons of lignite per hour.

The boiler building is designed so that it can be washed down after overhauls and during operation if desirable.

Electrostatic Precipitator and Ash Removal

The Wheelabrator-Lurgi precipitator and some of the design parameters are shown on figure 28. The precipitator is divided into two sections. Each section contains two chambers with guillotine dampers in the inlet and outlet so that any chamber can be removed from service for maintenance with the other three remaining in service.

Precipitator and accessories are designed for maximum reliability and maximum flexibility in servicing equipment with the plant operating at its maximum continuous rating corresponding to a flue gas flow rate of 2,032,000 acfm. Arrangement and sizing of the gas chambers is such that with one chamber isolated for service, the remaining three chambers can accommodate the total gas flow while maintaining at the outlet a specified particulate emission rate of not more than 0.1 lb/10^6 Btu. During periods of single-chamber isolation, it will be necessary to proportion gas flow through the remaining three chambers by adjusting louver dampers on the outlet ducts. In addition, eight smoke density monitors are located at the outlets of the precipitator. These monitors, together with the outlet louver control, will be used to balance the gas flow in the system for maximum precipitator efficiency.

Expected precipitator performance is as follows:

Gas flow rate................acfm..	2,032,000	2,032,000	2,032,000
Number of chambers in operation....	4	4	4
Inlet loading.............gr/acf..	1.1	2.2	3.3
Outlet loading............gr/acf..	0.006	0.009	0.012
Precipitator efficiency.......pct..	99.46	99.59	99.66

The maximum acceptable particulate emission rate of 0.1 lb/10^6 Btu is equivalent to 0.026 gr/acf. With 6.46 pct ash in the coal and 40 pct of the total ash going to the precipitator, inlet loading would be 1.1 gr/acf.

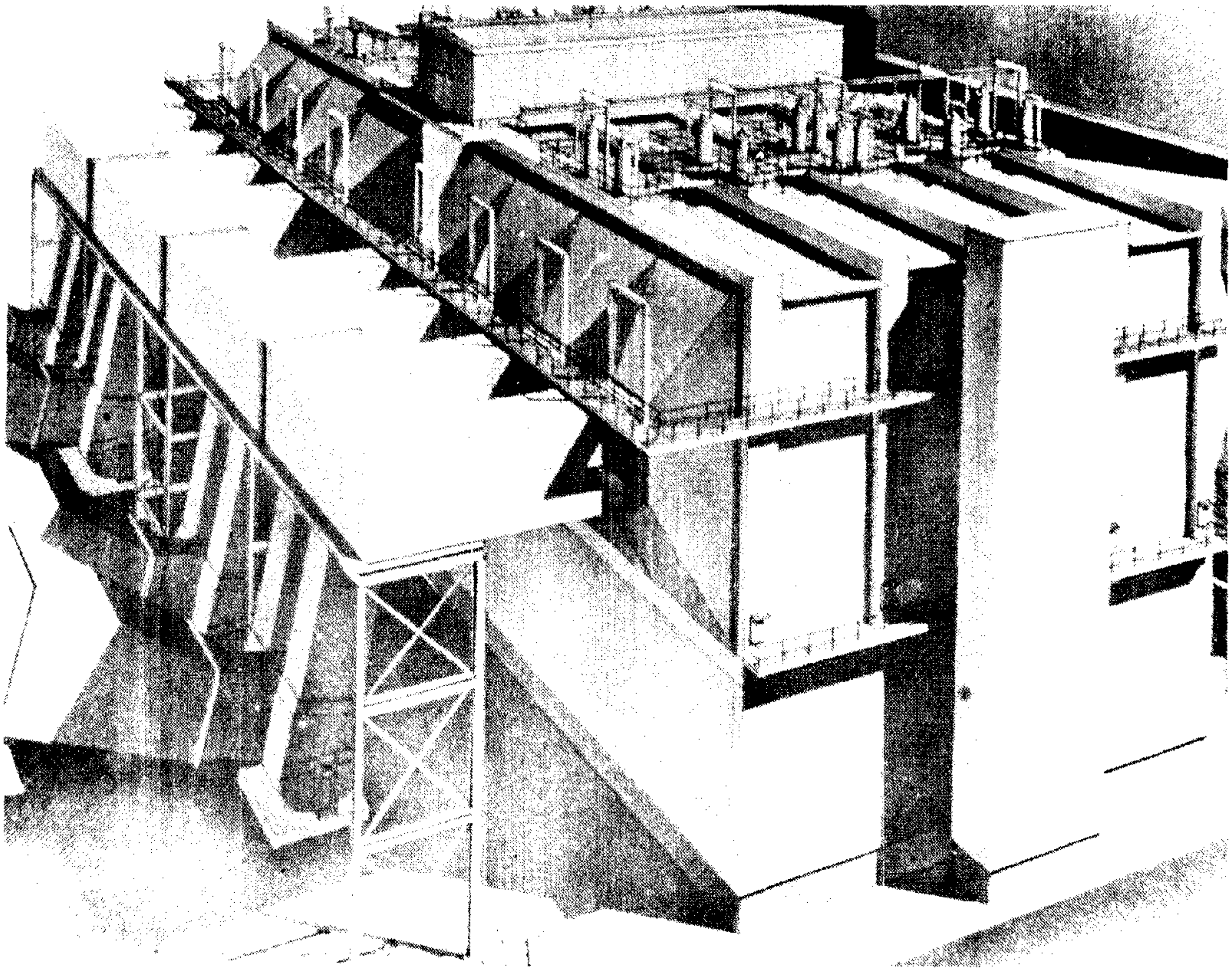

FIGURE 28. - Electrostatic precipitator for Big Stone plant.

Because of industry's experience with large induced draft fans, it was decided to install four fans with sirocco blading. An economic evaluation of two-speed versus single-speed motors revealed a substantial saving in favor of the two-speed motors. The induced draft fans will be equipped with two-speed phase amplitude modulated (PAM) motors.

Ash collected in the air heater hoppers, economizer hoppers, and slag tanks will be sluiced to an ash settling basin. Precipitator ash will be conveyed pneumatically to an elevated ash silo. Ash from the silo can be unloaded through a dustless unloader for disposal on the site or unloaded dry for transport to a commercial market. Any ash not disposed of commercially will be hauled to an area on the plant property and disposed of by a sanitary landfill method.

Auxiliaries and Transmissions

The fuel-oil-fired auxiliary boiler is sized to supply 150,000 lb of steam per hour at 165 psig and 500° F. Capacity is based on building heating requirements plus startup steam for the auxiliary turbines driving the boiler feed pumps and forced draft fans.

Both the 10,738-hp boiler-feed pump auxiliary turbines and the 10,205-hp forced draft fan auxiliary turbines exhaust to the main condenser.

Transmission line facilities to the plant include two 115-kV and two 230-kV lines. Two 115-kV lines or either of the 230-kV lines will provide sufficient power to start the plant. The 13.8-kV startup power will be supplied from the tertiary winding of the 115-kV/230-kV interchange transformer.

The Big Stone plant will burn between 2.5 to 3.0 million tons of lignite each year, all mined by Knife River Coal Mining Co. at Gascoyne in southwestern North Dakota.

Unit Train Concept

Early in the design conferences leading to construction of Big Stone, it was decided that the most economical and satisfactory method of transporting these quantities of lignites would be by unit trains. Consequently, the Bechtel Power Corp. was directed by the owners to begin investigating how this decision could be implemented through a practical lignite loading, transportation, and unloading system.

Out of these investigations grew the Big Stone Unit Train, a unique train of identical cars carrying a specific commodity from a single source to a single destination on a regular schedule. The Big Stone partners will purchase over 200 flat-bottomed gondola cars of 100-ton capacity to be split into two trains of 100 cars each that will operate on a closed-loop system, with one train at the mine being loaded while the other is being unloaded at the plant. The tracks at both terminals are designed as loops to reverse train direction at each end without switching.

The trains will operate over the Milwaukee Road main track through Hettinger, N. Dak., and Mobridge, Aberdeen, and Milbank, S. Dak., as shown in figure 29, with locomotive power provided by the Milwaukee.

A typical schedule would show one train leaving Gascoyne at 1200 hours and arriving at the Big Stone plant at 0800 hours the next morning. This in-transit time of 20 hours includes, however, crew changes at Hettinger, Mobridge, Aberdeen, and Milbank, with additional inspection service and light mechanical maintenance at Aberdeen. Four hours are allotted for the actual unloading operation, and another 20 hours for return time to the mine, where 4 hours are again scheduled for loading time.

Mine facilities being constructed for the unit train operation include a loop spur capable of holding the entire train while it is being loaded,

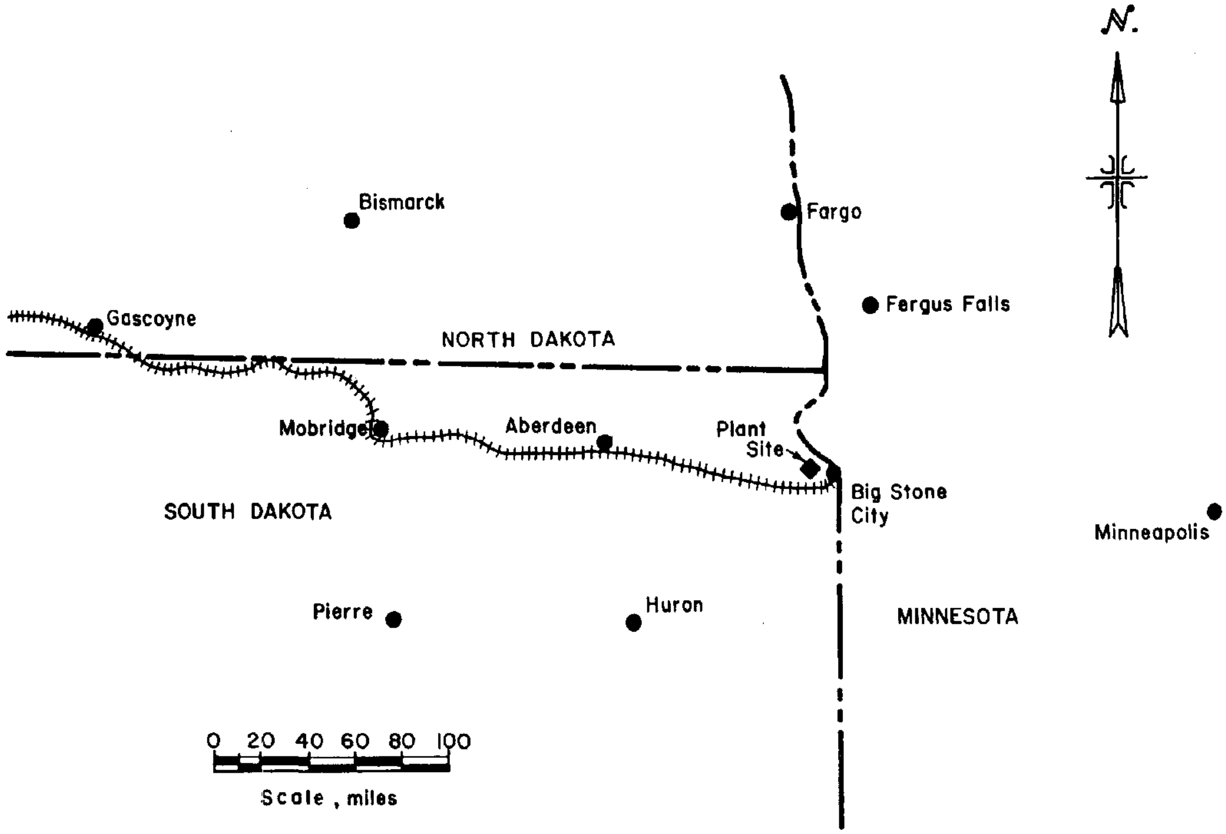

FIGURE 29. - Route for unit train, Gascoyne to Big Stone City.

lignite handling and storage systems, and an over-track tipple designed to allow flood-loading of the cars without stopping the train. A pant-leg chute will be constructed under the tipple to transfer lignite flow from one car to the next with a minimum of spillage between cars.

Unloading the cars at the plant will be accomplished using a rotary dumping mechanism that will clamp each car in turn to a track platform and then rotate it to an almost upside-down position. After returning the empty car to its normal position, an automatic car positioner will move the entire train forward the length of one car and the dumping process will continue. Cycle time on the rotary dumper will approximate 90 sec per car if no difficulty is experienced in removing lignite adhering to the inside of the car body, but conveyor capacity will effectively limit the cycle time to 2 min per car. Swivel couplings will eliminate the necessity of uncoupling each car as it is dumped, as the axis of car rotation will be at the coupling.

Unit Train Coal Cars

The most novel feature of the Big Stone Unit Train, however, will be the specially designed coal car developed by Bechtel in cooperation with the Milwaukee Railroad. This coal car will be a nominal 100-ton-capacity flat-bottom gondola unit constructed of steel and furnished with a system of hinged roof covers specifically designed to allow the rapid loading and unloading required in true unit train service. The roofed car design has three important functions:

1. It prevents snow and rain from adding moisture to the lignite being transported while helping to retain heat, thus reducing freezing problems.

2. It prevents falling or windblown snow from accumulating in the empty car, thus helping to prevent a buildup of frozen lignite on the sides, ends, and floor of the car during winter operation.

3. It prevents windblown loss of lignite, which could amount to as much as 1 pct of the transported amount, and eliminates environmental problems associated with that loss because of dusting along the track right-of-way.

Several details of the unit train car are shown in figure 30. The car has roller masts on each of the roof segments that are engaged by an operating "scroll" rail at the mine facility and raised to an open position as each car reaches the loading area under the tipple. It is this feature of the hinged roof covers that allows flood-loading of the train without stopping. The

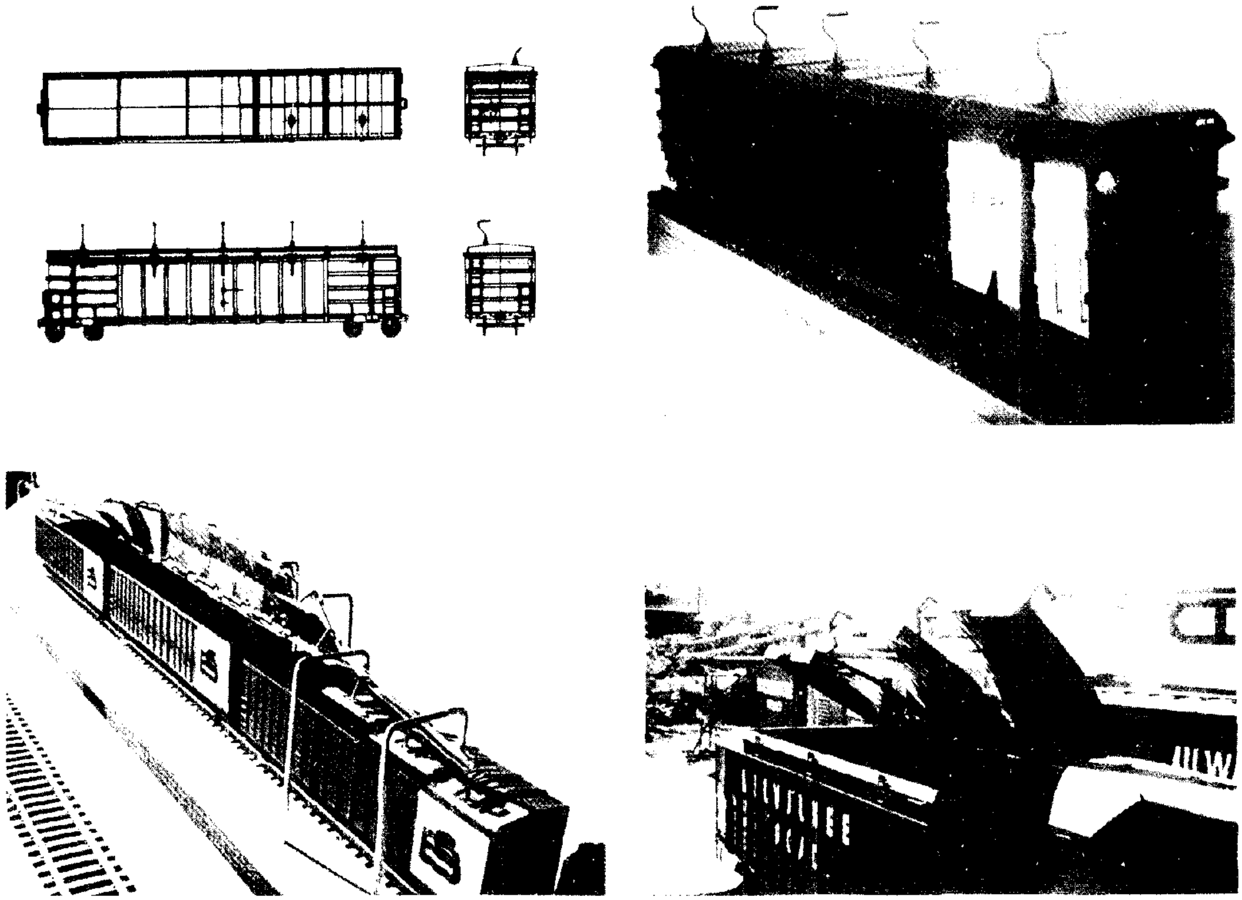

FIGURE 30. - Details of unit train coal car.

photograph of the model train shows masts engaged in the scroll rail and all lids in open position. Following the loading operation, the roller mast and lid are returned to a closed position by the "scroll" rail.

Since the lids are latched down during transit, a method of unlatching has been developed for use both at the loading point and the plant rotary dumper. This system consists of a pressure bar that will be rigidly constructed beside the track just ahead of the "scroll" rail at the mine, which will apply pressure to the unlatching mechanism from the side. The spring-loaded lid will then pop up between 2 to 3 inches allowing the tipple operator to determine whether or not each lid is actually unlatched prior to entry into the "scroll" rail. At the end of the "scroll" rail downward pressure will be applied to the roller masts to relatch each lid.

Similar arrangements are to be provided at the rotary dumper, except that in this case no "scroll" rail is required to lift the lids. As each car is rotated in the dumping process, the lids will fall open of their own accord, much like the tailgate on a dump truck as the front of the box is raised. A device exerting downward pressure on the covers will be used here, as at the mine, to insure positive latching.

To test this entire concept, the Milwaukee Railroad made available a standard 70-ton open-top gondola for test purposes.

The car was fitted with segmented lids of the same design as proposed for the Big Stone cars and a roller mast was attached to each lid. The "scroll" or lifting rail was designed by Bechtel and built to full scale in the Milwaukee yards alongside a portion of trackage.

A series of tests was then begun, including a great number of passes through the lifting rail to ascertain the durability of the rail, lids, and mast. At the same time, a latch for each lid was designed and installed for durability testing, along with a device for automatically unlatching the lid prior to entering the rail.

After this extensive testing, the car was taken out on the road and moved with normal traffic. A dynamometer car complete with instrumentation accompanied the test car to record all data acquired from strain gages and acceleration measuring devices attached at critical locations, and no unusual results were obtained. The scroll rail will be removed and reassembled at the mine facility when testing is completed.

Lignite Handling

Lignite handling equipment at the plant will be divided into two general areas, the unloading system and the reclaim system. Schematic views of loading, unloading, and in-plant handling are depicted in figure 31. The actual unloading of the lignite cars will be accomplished on a rotary dumping device along with an automatic car positioner. The dumper has been so located on the track loop that space is available on the entrance side for the possible later construction of an infrared car-thawing shed.

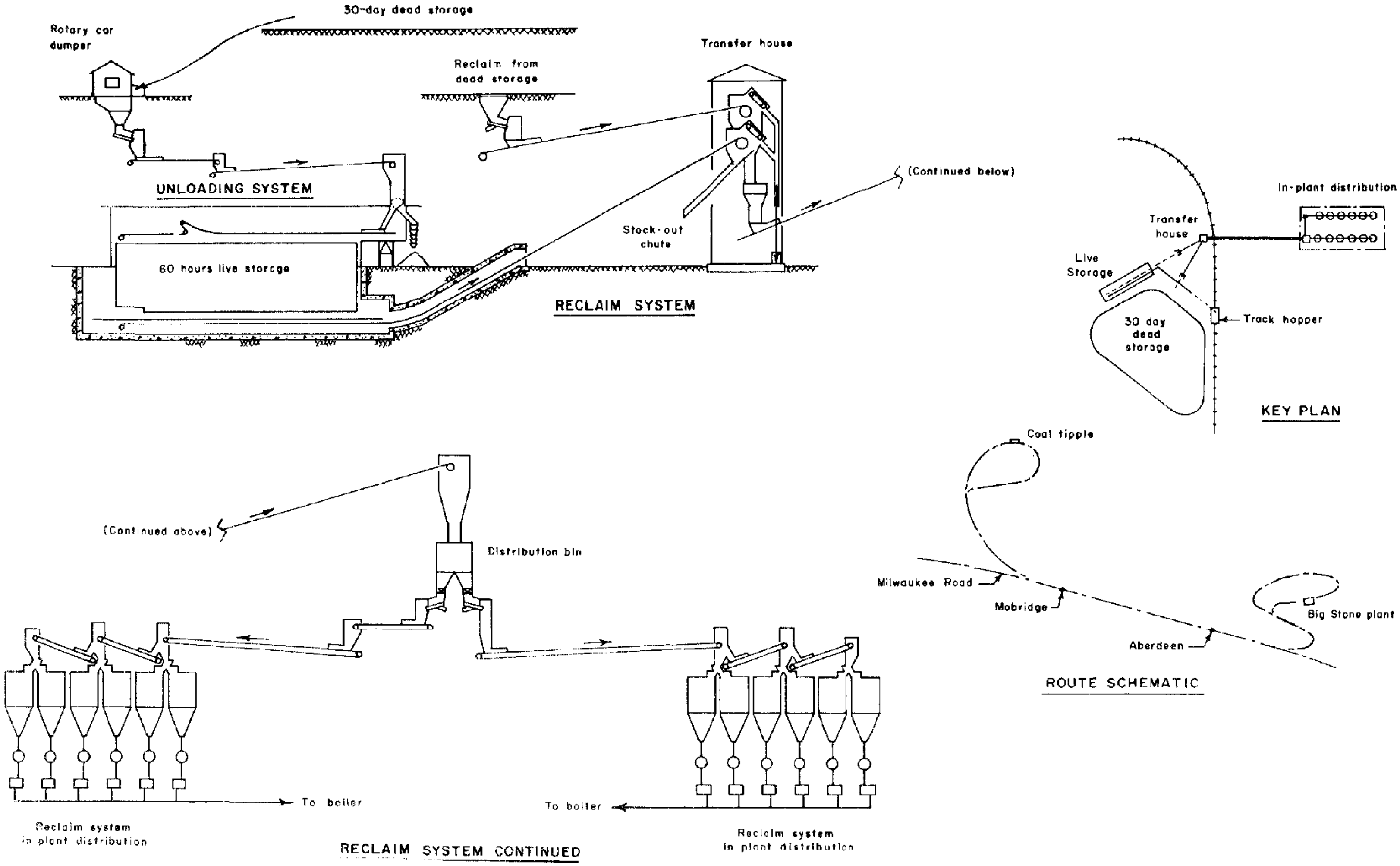

FIGURE 31. - Loading, unloading, and in-plant handling of lignite.

Present plans are to begin dead storage stockpiling late in 1974 so that much of the storage activities will be accomplished during cold weather. By exercising the lignite unloading system under the worst conditions, it is expected that most problems associated with low-temperature handling will be identified during a period when unloading times are not as critical as during later plant operation. At this time it will also be determined whether or not a thaw shed for the rail cars will be required.

The lignite will fall a minimum of 12 feet from the open car to the 18- by 18-inch grizzly below the dumper, hopefully breaking up any larger lumps of lignite that may have frozen in the car. Provisions are being made, however, for the possible later installation of a hydraulic spade if required. Below the grizzly, a hopper with enough capacity to hold three carloads of lignite will feed a 3,000-ton/hour conveyor belt through four vibratory feeders.

Flow on this 72-inch conveyor belt will pass over a recording belt scale to a transfer point at the end of a covered live storage pit with 60-hour storage capacity. From this transfer point lignite will either flow to the ground or onto another 72-inch, 3,000-ton/hour conveyor belt with a tripper system providing distribution along the entire length of the live storage pit. Lignite that drops on the ground will either be moved to dead storage or to the emergency reclaim yard hopper. In this latter case, the capability will exist of transferring fuel directly from the train to the plant in case of difficulties in the live storage area.

Sonic devices suspended below the tripper conveyor will sense pile height in the pit and adjust tripper speed and/or positioning accordingly.

From the bottom of the live storage pit, a rotary plow feeder with a capacity of 550 tons/hour will load a conveyor belt of the same capacity for movement of fuel to the transfer-crusher house in the yard.

Fuel entering the transfer house on this conveyor first will pass under a magnetic separator. After the separator, a motorized flop-gate will provide the alternative of either dropping the lignite to the ground through a stock-out chute or passing it to the crusher system. The stock-out chute will allow complete removal of all fuel from the live storage pit if necessary.

The crusher system consists of two 550-ton/hour crushers designed to reduce the 3-inch as-received lignite to 3/4 inch or less. A bypass around the dual crusher arrangement will allow conveying of 3-inch lignite directly to the plant.

From the transfer house, twin 36-inch belts, each with a capacity of 550 tons/hour, lead to the in-plant fuel distribution system. In the plant, the lignite will first enter a distribution bin from which two conveyor systems will be fed, one on each side of the boiler. Each of the six cyclones on the back and the six on the front of the boiler will have individual bunkers of 8 hours storage capacity. A 550-ton/hour conveyor will transport fuel to a hopper above and between the first two bunkers. When both of these bunkers are full, fuel will back up into the distribution hopper and cascade over onto the next conveyor, which feeds bunkers three and four. Finally, as three and four become filled, fuel cascades onto the last conveyor feeding bunkers five and six.

Level sensors in all bunkers control shutoff of fuel at proper levels, and reestablishment of fuel flow will be controlled by a pulse timer in the gravimetric feeder systems below the bunkers.

From the feeders, lignite will be introduced into individual crushers for each cyclone where it will be reduced in size to 1/4 inch or less. Transported by hot air from the crusher, the fuel enters a cyclone separator where lignite is separated from the water-vapor-saturated air and lifted into the cyclone burner with a new blast of drying air. Saturated air from the cyclone separator will be vented into the boiler above the upper burner level.

A 263,000-ton dead storage pile will provide 30 days of emergency fuel at the plant maximum burn rate of 365 tons/hour. Reclaim from dead storage will be through an opening in the side of the rotary dumper structure into the hopper below. Since this is the normal unloading point for unit train coal also, no changes in control mode will be necessary during dead storage reclaim other than to reset the belt scale for inventory control.

In case of difficulty with the rotary dumper, the conveyors between the dumper and transfer-crusher house, or live storage reclaim equipment, a separate yard hopper will be used for emergency reclaim. A conveyor of 550-ton/hour capacity will transport fuel from this hopper directly to the transfer-crusher house if necessary.

In addition to the lignite handling and storage systems, capacity is being provided for storage of 1,000,000 gal of fuel oil in two 500,000-gal storage tanks complete with truck and rail unloading facilities. Normal plant usage of fuel oil would not ordinarily warrant such large storage capacity, but in the winter of 1974-75, oil will be used for plant heating as well as for startup, and the total of these two quantities may well approach 1,000,000 gal.

In view of the recent difficulty in acquiring fuel oil and the poor prospects for improvement in that situation, it was felt that prudent design would incorporate enough storage to allow purchase of oil requirements in early summer for use during the following winter.

SHIPMENT, STORAGE, AND HANDLING CHARACTERISTICS OF DRIED LOW-RANK COALS

by

L. E. Paulson,[10] S. A. Cooley,[11] and R. C. Ellman[12]

Introduction

The U.S. Geological Survey estimates that 56 pct of the reserves of coal in the United States, based on mapping and exploration data, are lignite and subbituminous coals.[13] The majority of these low-rank coals are in the Western States of North Dakota, Montana, and Wyoming and are characterized by relatively low sulfur and ash and have high moisture contents. If moisture was removed by drying, these coals would be upgraded in heat content per unit weight, and their physical characteristics for a variety of processing techniques and utilization systems would be improved. These coals have been used without treatment in powerplants located within a relatively limited consuming area; thus possible benefits of full-scale drying have not been justified. The new expanded market for low-sulfur fuels has renewed interest in upgrading processes for low-rank coals and particularly in the problems involved in transporting, handling, and storing the reactive dried material.

Industrially dried lignite is produced during the winter months by Baukol-Noonan, Inc., at Noonan, N. Dak., for the purpose of freezeproofing as-mined lignite.[14] About 15 pct of a carload of lignite is dried to a moisture content of approximately 20 pct and mixed at car walls and bottoms with untreated lignite to eliminate winter handling problems. A few carload shipments of dried lignite have also been made for specific test purposes. No particular difficulties were encountered, but no effort was made to record and evaluate the difference between handling and shipping dried and as-mined lignite.

Subbituminous coals may have moisture contents that range from 20 to 30 pct; lignites have moisture contents ranging from 35 to 40 pct. The major portion of moisture present exists as free moisture in pores and capillaries within the physical structure of the coal. When a particle surface is exposed to an atmosphere unsaturated with water vapor, moisture is released until the vapor pressure of moisture at the particle surface and that of the surrounding

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All authors are with the Grand Forks Energy Research Laboratory, Bureau of Mines, Grand Forks, N. Dak.

[13] Averitt, P. Coal Reserves of the United States, January 1, 1967. U.S. Geol. Survey Bull. 1275, 1969, 116 pp.

[14] Ellman, R. C., J. W. Belter, and L. Dockter. Freezeproofing Lignite. BuMines RI 6677, 1965, 28 pp.

atmosphere are in equilibrium. With an increase in temperature, the rate of water release increases. When a particle loses moisture, the surface shrinks and forms fissures, producing new surfaces that are reactive towards oxygen. The consequence of drying low-rank coals is the production of a mixture of sizes of varying moisture content that is hot, dusty, and highly reactive toward oxygen.

Much research has been done in the past on drying of low-rank coals,[15] and a variety of techniques are suitable and technically feasible. The characteristics of dried material in respect to transportation, handling, storage, heat transfer properties, amount of moisture the dried particles will regain, and bulk density have not been evaluated. The present work was initiated to determine and evaluate these properties and to develop procedures that would reduce or eliminate the problems associated with the movement and storage of a dried low-rank coal from producing plant to point of use.

This paper includes observations made on two carloads of dried lignite and one carload of dried subbituminous coal and of the status of research on the physical characteristics of dried coal done at the Bureau of Mines Grand Forks Energy Research Laboratory. The dried lignite was obtained and shipped with the cooperation of Baukol-Noonan, Inc., and the Burlington Northern Railroad. One carload of subbituminous coal was similarly provided by Western Energy Co. of Billings, Mont., dried at Baukol-Noonan, Inc., and shipped to the laboratory. Pilot plant facilities were developed for drying bulk quantities for test purposes, and suitable procedures were devised for evaluating and studying the characteristics of these samples.

Handling, Shipping, and Storing Rail Car Quantities of Dried Lignite and Subbituminous Coal

Lignite

The first test carload, containing 49 tons of 1/4-inch by 0 lignite dried from 35 to 22.5 pct moisture, was loaded into a standard open-top, bottom-dump car on July 22, 1970, at the Baukol-Noonan mine. The shipment arrived 5 days later at the Grand Forks Energy Research Laboratory. The temperature of the coal in the car was measured at five depths at each of eight locations in a grid pattern within the car both at the mine and after completion of shipment. The average temperature at departure from the mine was 116° F. During the 5-day transit the average temperature increased to 154° F, but no localized

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[15] Cooley, A. M. Pneumatic Drying of Lignite. Paper in Technology and Use of Lignite. Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April 1961, comp. by J. L. Elder and W. R. Kube. BuMines IC 8164, 1963, pp. 30-35.

Harrington, L. C., V. F. Parry, and A. Koth. Technical and Economic Study of Drying Lignite and Subbituminous Coal by the Fleissner Process. BuMines Tech. Paper 633, 1942, 84 pp.

Parry, V. F., W. S. Landers, E. O. Wagner, J. B. Goodman, and G. C. Lammers. Drying and Carbonizing Fine Coal in Entrained and Fluidized State. BuMines RI 4954, 1953, 43 pp.

hot spots or fires could be observed. Maximum temperature recorded at arrival was 186° F. The weight of the car was recorded at the mine and after arrival in an attempt to measure wind loss. Unfortunately, the car was exposed to a severe rainstorm during transit and wind loss could not be determined.

The dried lignite was unloaded by an under-car conveyor and carried to the stockpile site with a front-end loader. Each bucketful was placed and then leveled by the bucket of the loader as it operated at the perimeter of the pile. It was planned to construct the pile in 6-inch lifts placed by a dump truck as it unloaded. This procedure could not be carried out, partly because of equipment limitations but also because the dried lignite could not be compacted in layers of sufficient stability to provide the traction required by the truck. The finished stockpile had a 26- by 35-foot base, a height of 5 feet, relatively steep side slopes, and a flat top of 8 by 16 feet. The completed test stockpile is shown in figure 32.

The dried lignite cooled during unloading and stockpiling operations. The average temperature of the pile was 123° F, measured at 41 points at stations installed immediately after stockpiling was completed. On two separate occasions during the fall season of 1970, one small area on the west side of the pile ignited after a period of unusually high winds. Temperatures in the pile remained at 100° F or above until October 1970 but slowly decreased to an average temperature of 57° by January 1971. From that time until the pile was dismantled in the summer of 1972, the average temperature reflected seasonable temperature variations.

On September 3, 1970, a second carload, 43 tons of 1/4-inch by 0 lignite dried from 35- to 21.3-pct-moisture content, was similarly produced at the mine, shipped to the laboratory, and stockpiled. After loading, the average temperature, measured at 18 locations in a grid pattern, was 143° F. The car was in transit for 7 days, during which the average temperature increased to 168° F. Maximum temperature recorded on arrival was 229° F. A second attempt to determine wind loss was also unsuccessful.

The second car was unloaded more slowly than the first car to provide greater cooling during construction of the test stockpile. In addition, greater effort was made to construct the pile in leveled layers by the use of a tractor and drag. The completed pile had a depth of 3 feet and a base of 18 by 33 feet with gently sloped sides. Completed stockpile 2 is shown in figure 33. The average of initial temperatures measured at installed stations was 115° F. The temperature of pile 2 rapidly and steadily decreased to near freezing during the winter and thereafter varied with seasonal ambient temperature changes. No hot spots or heating occurred in this pile. It remained stable until it was dismantled in the summer of 1972.

During the last week of July 1972, a conical pile of approximately 21-foot base diameter and 10 to 12 feet in height was constructed from one-half of each of stockpiles 1 and 2. The temperatures at both piles when moved were in equilibrium with ambient temperature. The moisture content of a representative sample of pile 1 was 22.5 pct, but the moisture content of pile 2 was much higher--27.2 pct--and free water could be observed on the

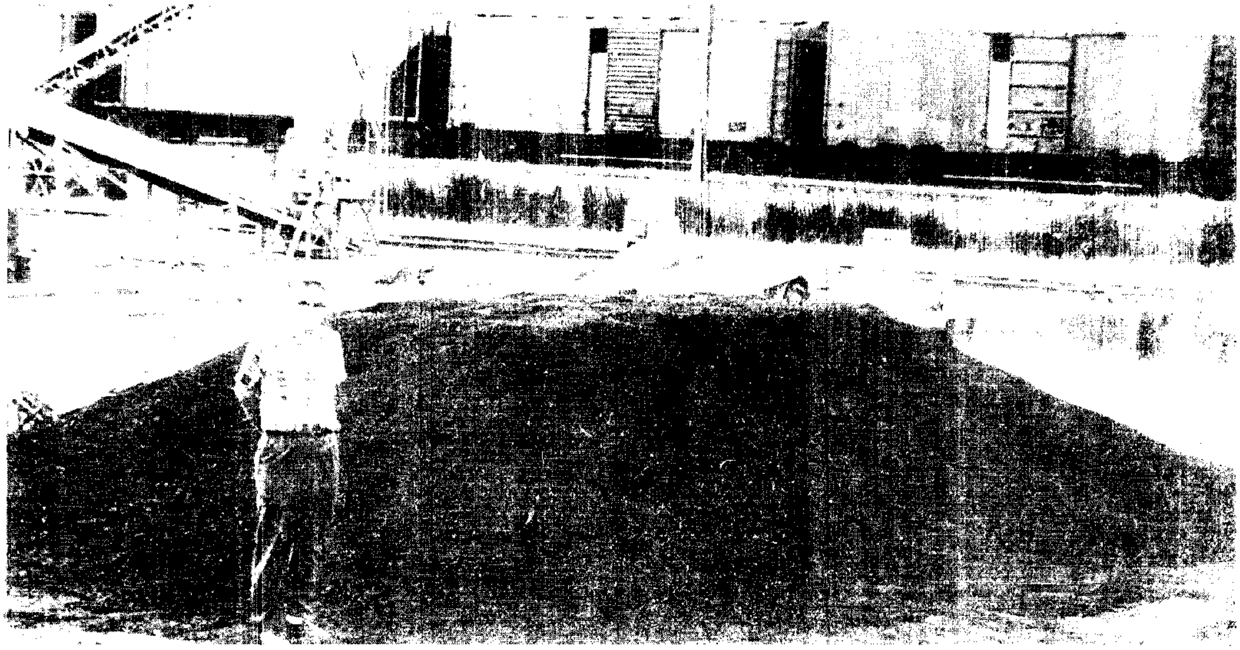

FIGURE 32. - Completed test stockpile 1, July 1970.

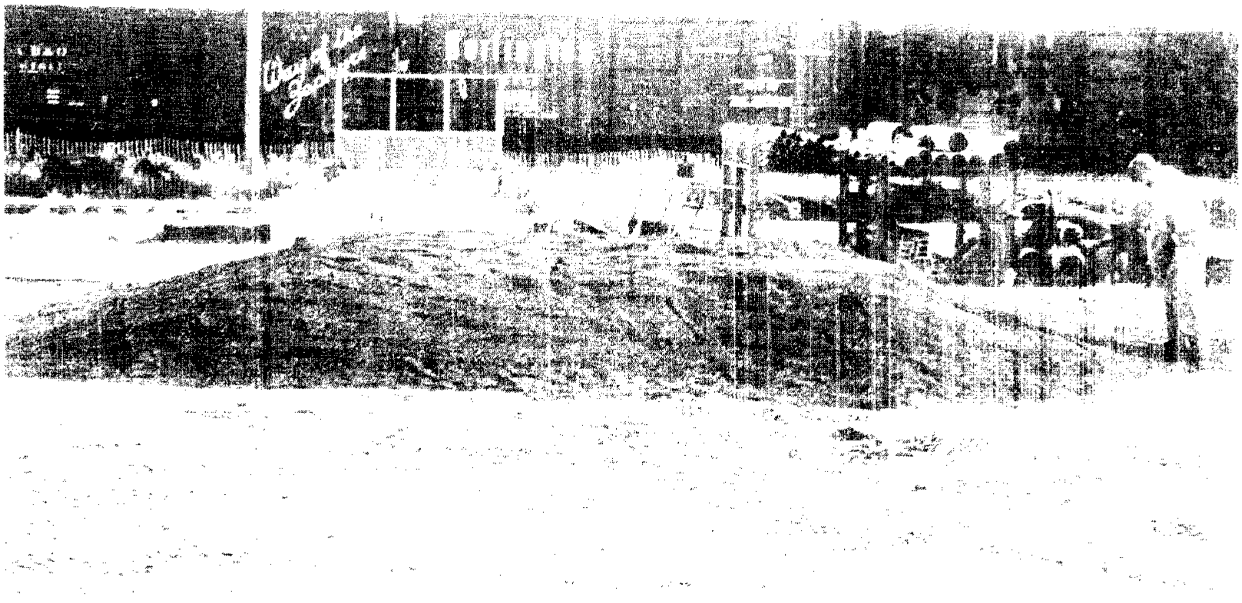

FIGURE 33. - Completed test stockpile 2, September 1970.

surface of particles. Apparently rainwater had pentrated all portions of pile 2. The center portion of pile 1 had remained dry. The conical pile was constructed using a raised conveyor fed by a front-end loader. A bucketful was alternately removed from each of the two stockpiles. For a period of

4 weeks, temperatures measured at eight stations in the conical pile showed no change. The remaining portions of the two original stockpiles were also stable. The second half of both piles were added to the conical pile the week of September 3, 1972. The completed conical pile containing 92 tons of dried lignite was approximately 35 feet in diameter and 15 feet high.

Temperatures in the pile remained stable until October, when the top portion of the pile began to heat. Temperatures at many locations soon exceeded 150° F. The snow on the top melted to form a black crown as shown in figure 34. During November and until the end of December 1973, the heating spread throughout the pile until almost all points were over 125° F and temperatures ranged up to 170° F. At the end of December, numerous small areas ignited at the base of the pile, and the coal was spread and cooled.

Subbituminous Coal

In March 1971, two carloads of 1-1/4-inch by 0 subbituminous coal were shipped from the Colstrip mine, Colstrip, Mont., to the Baukol-Noonan mine for drying. The as-mined moisture content was 26.6 pct. The particle size of the subbituminous coal was much larger than the 1/4-inch by 0 lignite normally fed to the dryer. As a consequence, the subbituminous coal of 26.6 pct moisture content could be dried to only 21.4 pct moisture. One carload, containing 42 tons, was shipped to the Grand Forks Energy Research Laboratory for storage studies. After loading, the average temperature of the dried subbituminous

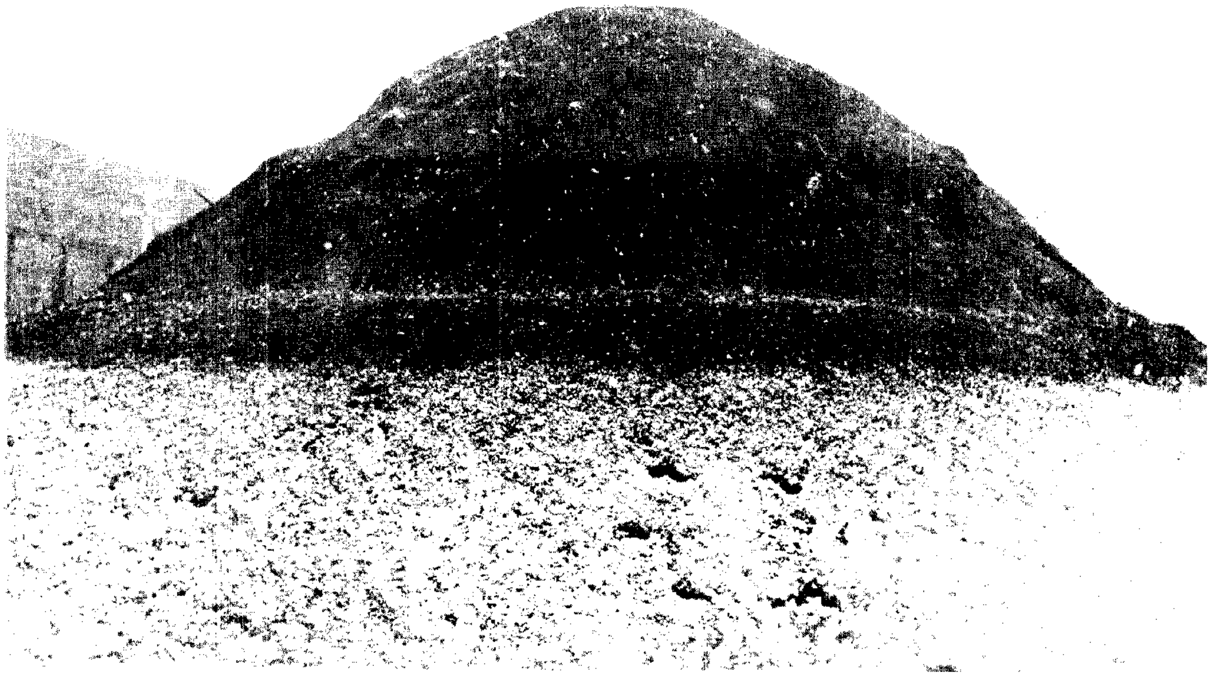

FIGURE 34. - Conical test pile constructed from test piles 1 and 2, December 1972.

coal was 108° F. Five days later, on arrival at the laboratory, the average temperature was 123° F. Maximum temperature recorded was 189° F.

Test stockpile 3 was constructed with a base of 18 by 33 feet, a depth of 3 feet, and with gentle side slopes. Cooling, mixing, and leveling of the dried coal was promoted during the construction portion of stockpile 3 by traversing the pile with an agricultural-type harrow after each portion was placed on the pile. Completed test stockpile 3 is shown in figure 35. The highest temperature in the completed pile was 93° F and the average temperature rapidly decreased to 48° F during the remainder of the winter. To date, there has been no evidence of heating in this stockpile.

Size Distribution and Progressive Change in Fuel Composition

Size distribution of dried test coal is given in table 1. Size consist of piles 1 and 2 are very similar; however, the size consist of dried subbituminous coal in pile 3 is significantly larger. Proximate analysis and heating value of samples collected at intervals from each of the piles is given in table 2. The variation in moisture content of periodic samples of each of the three stockpiles demonstrates the changes that occur because of seasonal or weather experiences. Pile 1 initially lost moisture during the extended

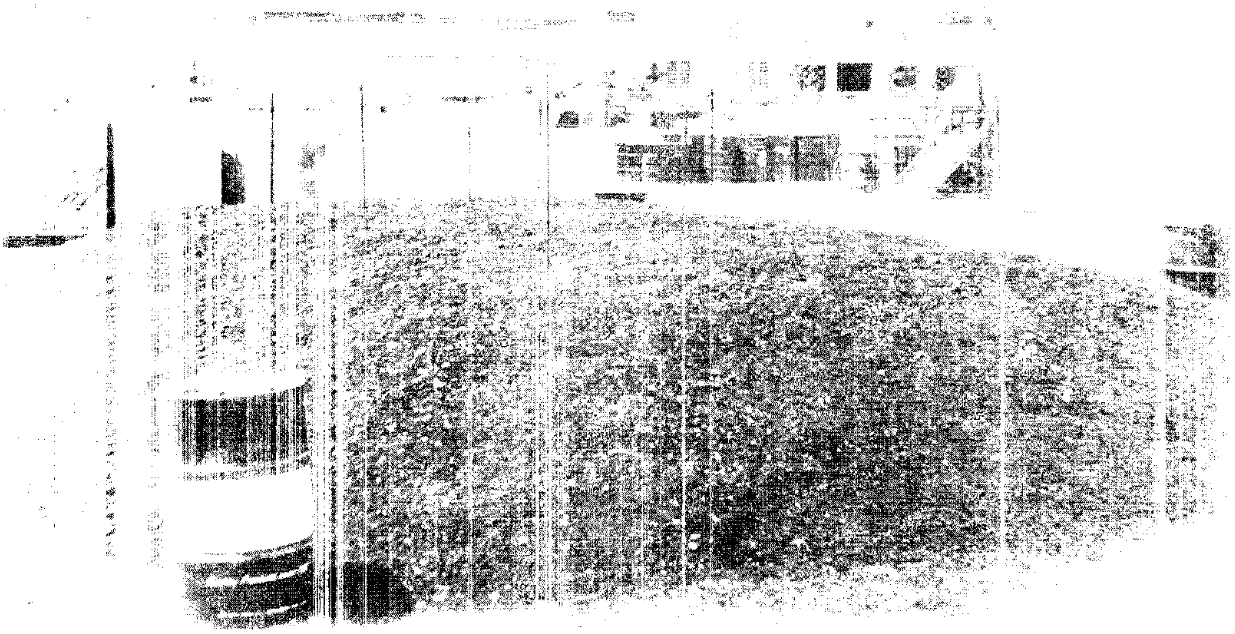

FIGURE 35. - Completed test stockpile 3, March 1971.

period of cooling. All stockpiles demonstrate the effect of moisture penetration. Changes in heating value are not significant, considering the range of values that could be caused by variation in samples and tolerances in analytical determinations.

TABLE 1. - Size distribution of stockpiled dried low-rank coals, pct

Size fraction	Pile 1, dried lignite	Pile 2, dried lignite	Pile 3, dried subbituminous
Plus 3/4 inch..........	-	-	9.0
3/4 by 1/2 inch........	-	-	12.0
1/2 by 1/4 inch........	-	-	25.0
1/4 inch by 4 mesh.....	24.5	17.5	10.0
4 by 8 mesh............	32.5	33.5	18.0
8 by 16 mesh...........	20.5	21.5	12.0
16 by 30 mesh..........	12.5	14.0	6.5
30 by 50 mesh..........	6.0	8.0	3.5
50 by 100 mesh.........	2.0	3.5	2.0
100 by 200 mesh........	1.0	1.0	1.0
Minus 200 mesh.........	1.0	1.0	1.0

TABLE 2. - Proximate analysis and heating value of periodic samples collected from stockpiles

Proximate, as received	Stockpile 1, dried lignite				Stockpile 2, dried lignite			
	Initial	April 1971	Sept. 1971	Oct. 1972	Initial	April 1971	Sept. 1971	Oct. 1972
Moisture.......pct..	22.5	13.4	13.4	22.5	21.3	19.9	16.8	27.2
Volatile.......pct..	31.2	-	34.2	30.8	28.9	-	33.7	28.9
Carbon.........pct..	37.1	-	39.1	35.7	34.4	-	39.7	34.4
Ash............pct..	9.2	11.9	13.3	11.0	9.5	11.0	9.8	9.5
Total.....pct..	100.0	25.3	100.0	100.0	94.1	30.9	100.0	100.0
Btu/lb (maf)........	12,210	11,680	11,510	11,690	12,165	12,105	11,850	11,960

	Stockpile 3, dried subbituminous		
	Initial	Sept. 1971	April 1972
Moisture.......pct..	20.6	18.3	23.6
Volatile.......pct..	30.1	31.5	29.6
Carbon.........pct..	39.3	38.3	38.4
Ash............pct..	10.0	11.9	8.4
Total.....pct..	100.0	100.0	100.0
Btu/lb (maf)........	12,950	12,680	12,300

maf = moisture and ash free.

Summary of Observations on Carload Shipments of Dried Low-Rank Coals

Shipment

When loaded directly from the dryer, the dried coal is at an elevated temperature and in a highly reactive condition. The temperature throughout the car may be expected to increase after loading and during shipment because of reaction with oxygen contained in the air present in void spaces. Considering the elevated temperature, any additional air circulation through the dried coal could readily produce rapid increases in temperature.

Temperature increase during shipment periods of 5 to 7 days in open-top cars was, however, not sufficient to produce ignition. Wind losses could not be determined; however, the physical characteristics of particles suggest that such losses would undoubtedly occur.

Unloading

Dried low-rank coals will readily flow from bottom-dump cars. Compared with as-mined coals, the dust nuisance was higher. Dried subbituminous coals created a much greater dust problem than did dried lignites. Temperature of dried coals readily decreased during handling operations. A period in which such cooling may take place, either during handling or after placement in a stockpile, should be provided. The lower the initial temperature of the stockpile, the better.

Stockpiling

Compaction of stockpiles of dried low-rank coals is more difficult than for as-mined coals because dried particles tend to slide and flow. The permeability coincidental with a low degree of compaction aids in initial cooling in small stockpiles, but it is likely that low degree of compaction, by allowing air infiltration, would promote heating in piles of the size required in commercial operation. With little compaction, rainwater and melted snow penetrate the interior of stockpiles. Gentle side slopes on stockpiles reduce air penetration by wind pressures but also reduce the amount of water runoff. Small test stockpiles of low compaction and gentle pile slopes have been found to be stable in respect to spontaneous heating but subject to variations in moisture content due to weather experiences.

Satisfactory large-scale stockpiling of dried coal requires that a method or technique be developed by which greater compaction can be obtained than is possible with procedures used for as-mined low-rank coals.

A dried low-rank coal remains reactive even after extended storage periods. Development of spontaneous heating in the conical pile demonstrated that despite an extended aging process, dried lignite is still capable of comparatively rapid spontaneous increases in temperature. The manner of stockpiling utilized in both piles 1 and 2 was essentially satisfactory in respect to spontaneous heating tendencies for the quantities stored and highly reactive initial condition of the test samples. The techniques utilized provide a basis for development of procedures by which large-scale, stable stockpiles can be constructed.

Drying and Testing of Pilot Plant Samples of Lignite and Subbituminous Coal in the Laboratory

Carload-scale tests of drying and stockpiling could not be as flexible as required in respect to the control of drying or treatment conditions. Pilot plant-scale studies were therefore initiated in which test samples of coal were dried at controlled test conditions. Oxidation tendencies, cooling rate, moisture uptake, and density were determined for dried coal samples as a function of variables such as moisture content of product, source of coal, and treatment conditions. To date, studies have been made on lignite from the Glenharold mine, Stanton, N. Dak., and on a subbituminous coal from Gillette, Wyo., furnished by Atlantic Richfield Corp.

Tests were conducted primarily on 1/2-inch by 0 coal because it and the dried product produced are of a practical commercial size. Moisture content of lignite tested was 36.3 pct. Moisture content of subbituminous as-received sample was very high--34.3 pct--and obviously mined under very wet conditions because of the presence of surface moisture. The principal variables studied to date are the influence of moisture content on the characteristics of the dried product and the effect of temperature and aging on the oxidation tendencies of dried low-rank coals.

Dryer

A Link Belt Roto-Louvre type dryer, on loan from the Chemical Engineering Department of the University of North Dakota, was used to produce samples of dried coals for testing. The auger is 31 inches in diameter and 10 feet long. A blower with capacity of 1,500 cu ft of air per min pulls the drying gases into the dryer and exhausts them to the exterior through a cyclone to remove particulate matter. The total weight of particulate matter collected by the cyclone and carried out by gases was measured for the range of conditions used to prepare test samples. The maximum weight of particulate matter at any operating condition was found to be 2 pct or less of product weight. Most particulate matter was collected in the cyclone. Dryer exhaust gases to the atmosphere were well below the limits permitted by industrial pollution standards. Particulate matter collected in the cyclone collector of the exhaust system was not included as product.

Heat for drying was provided by a natural gas burner installed in the inlet gas duct where combustion gas was tempered by addition of air. Drying gases were oxidizing owing to the excess air. Natural gas flow for all tests was adjusted to produce a drying gas temperature at the control point within the duct of 1,200° F. Drying gases were admitted between the outer shell and the louvres under the bed and passed upward through the coal being dried. The coal was gently tumbled as the dryer rotated.

Product was discharged to a hopper and carried to a drum receiver by an auger. The moisture content of the product was governed and controlled by the residence time in the dryer, which was varied by changing the rate of feed. The product temperature was a function of the degree of drying. For tests reported, the product temperature ranged from 130° to 135° F for lignite dried to midmoisture content range and to 165° F when dried to lower than 10 pct moisture. The pilot plant dryer is shown in figure 36.

Oxidation Tests

The rate and amount of oxygen reacting with dried low-rank coals is a measure of reactivity and the tendency to heat spontaneously.

The parameters associated with the oxidation reaction for low-rank coals at specific and controlled conditions have been studied by a number of

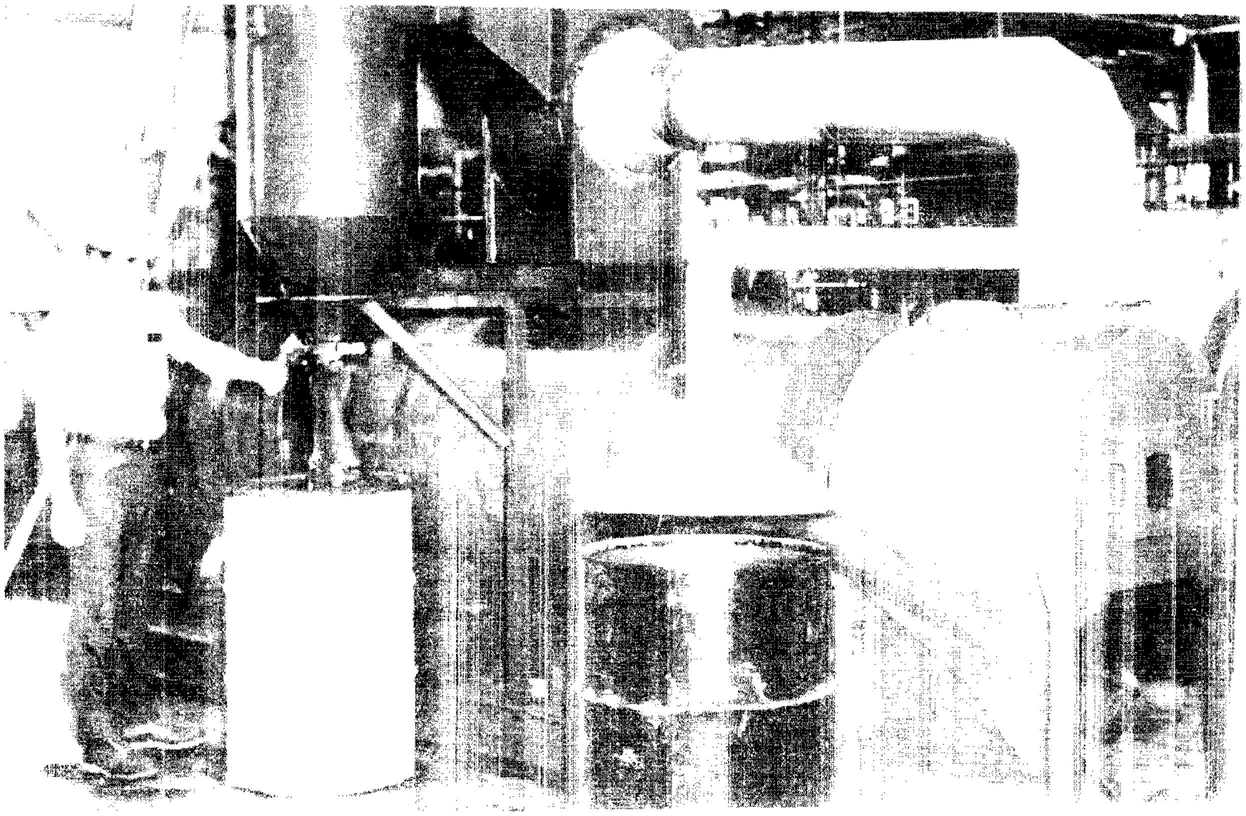

FIGURE 36. - Pilot plant coal dryer.

investigators.[16] The objective of the present investigation was to establish a comparative but still practical measure of the oxidation potential as encountered in a commercial process.

The quantity of oxygen reacted and reaction rate was measured by periodically sampling the atmosphere in a water-jacketed, insulated, sealed chamber 26 inches in diameter and 5 feet long. Test samples were charged to the chamber through an 11-inch-diameter port in one end. One-inch-square bars welded at intervals on the inside circumference provided a mixing action to the coal as the chamber was slowly rotated. The temperature of the coal and of the chamber interior was measured using thermocouples. The temperature in the chamber was controlled using external natural gas burners to heat the water in the jacket. A 50-pound sample of coal was found to provide a measurable and significant absorption of oxygen from the 17.6 cu ft of air available. The oxidation chamber is shown in figure 37.

The rate at which a sample of dried coal reacted with the given supply of oxygen was determined for five conditions. Two tests, designated "cycles 1 and 2," measured the reactivity of coal (1) directly from the dryer and (2) after a single cooling in a limited quantity of air. The remaining three conditions measured the effect of temperature on samples of dried product that had been cooled in a closed container.

For a test, the chamber was preheated to the desired temperature, the 50-pound sample of coal charged, and the chamber rotated at 3½ rev/hr. Samples of the chamber gases were collected each hour for 4 hours, and the carbon dioxide and oxygen content were determined. At the end of each hour, the temperature of the air and coal in the chamber and the atmospheric pressure were recorded. As oxygen in the chamber became depleted, a slight vacuum was created that caused additional air to enter the chamber. The amount of air leakage was determined by calculating the nitrogen balance each hour and then calculating the oxygen added.

First-Cycle Test

The objective of the test designated "first cycle" was to determine the rate at which the dried coal absorbed oxygen immediately after it was produced. The oxidation chamber was preheated to the temperature of the dried coal at discharge from dryer. A 50-pound sample, taken directly from product receiver of the dryer was charged to the oxidizer. The total moles of oxygen absorbed by the 50-pound test sample progressively as a function of time for four individual tests on dried lignite of 18.2- to 19.3-pct-moisture content is shown

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[16] Elder, J. L., L. D. Schmidt, W. A. Steiner, and J. D. Davis. Relative Spontaneous Heating Tendencies of Coal. BuMines Tech. Paper 681, 1945, 24 pp.
Sondreal, E. A. Studies on Low-Temperature Oxidation of Lignite as Related to Storage. Paper in Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April-May 1963, comp. by W. R. Kube and J. L. Elder. BuMines IC 8234, 1964, pp. 57-93.

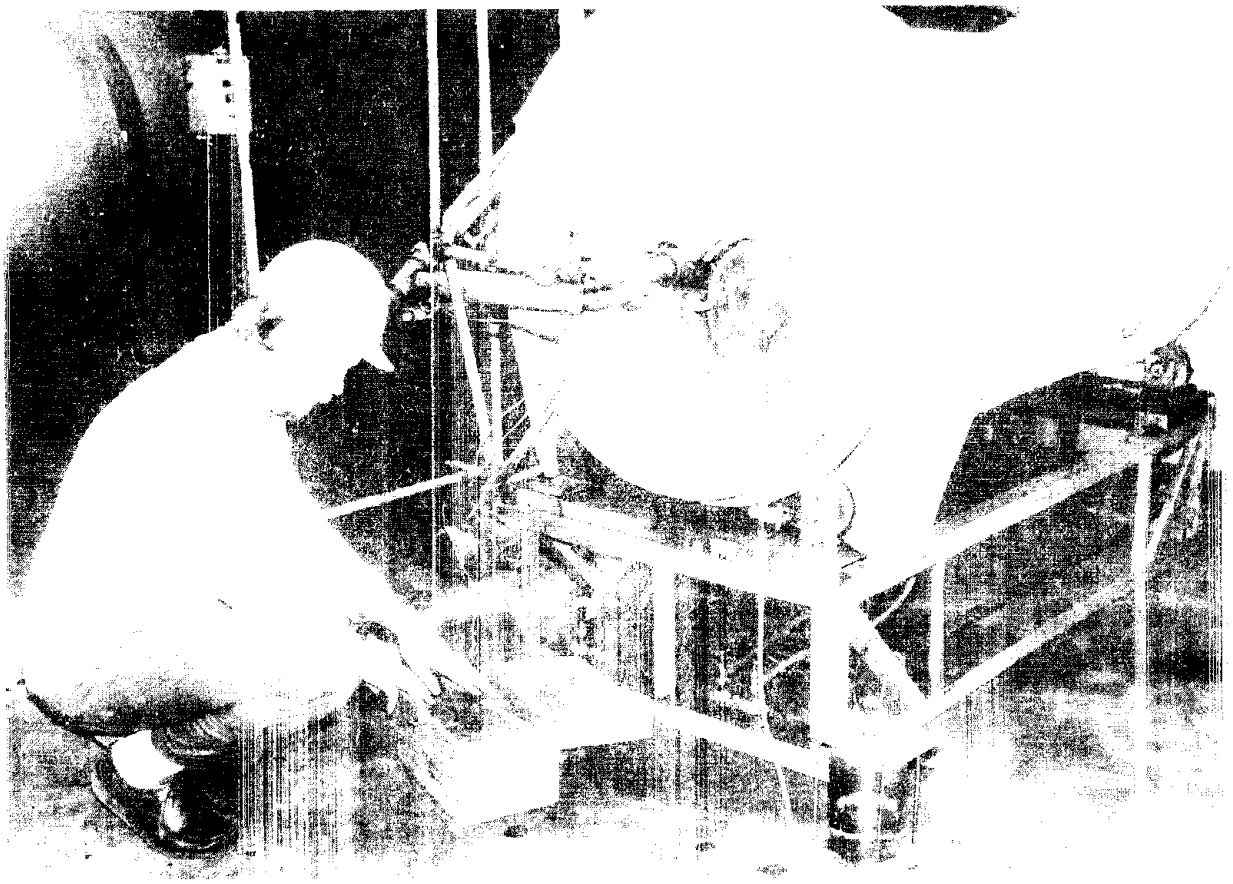

FIGURE 37. - Test chamber to determine reactivity of dried coal toward oxygen.

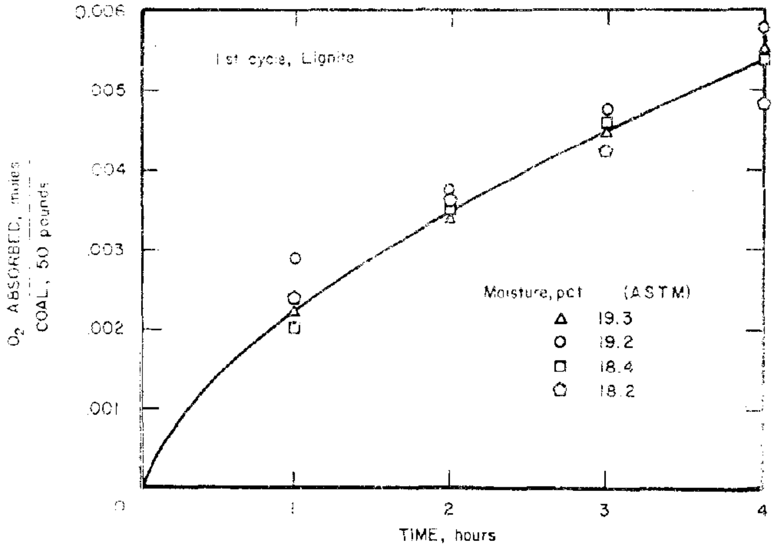

FIGURE 38. - Reproducibility of test method—first-cycle tests.

in figure 38. In this paper, oxygen absorption is expressed as pound moles of oxygen absorbed per 50 pounds of sample. All variations in the drying, sample preparation and analysis, and testing procedure contribute to differences in the amount of oxygen absorbed. The values plotted indicate reproducibility.

Nine first-cycle tests were made using individual samples of dried Glenharold lignite, and four first-cycle tests were performed on samples of dried Wyoming subbituminous coal. For the lignite, the moisture content of test samples ranged from 8 to 27 pct, and for the subbituminous

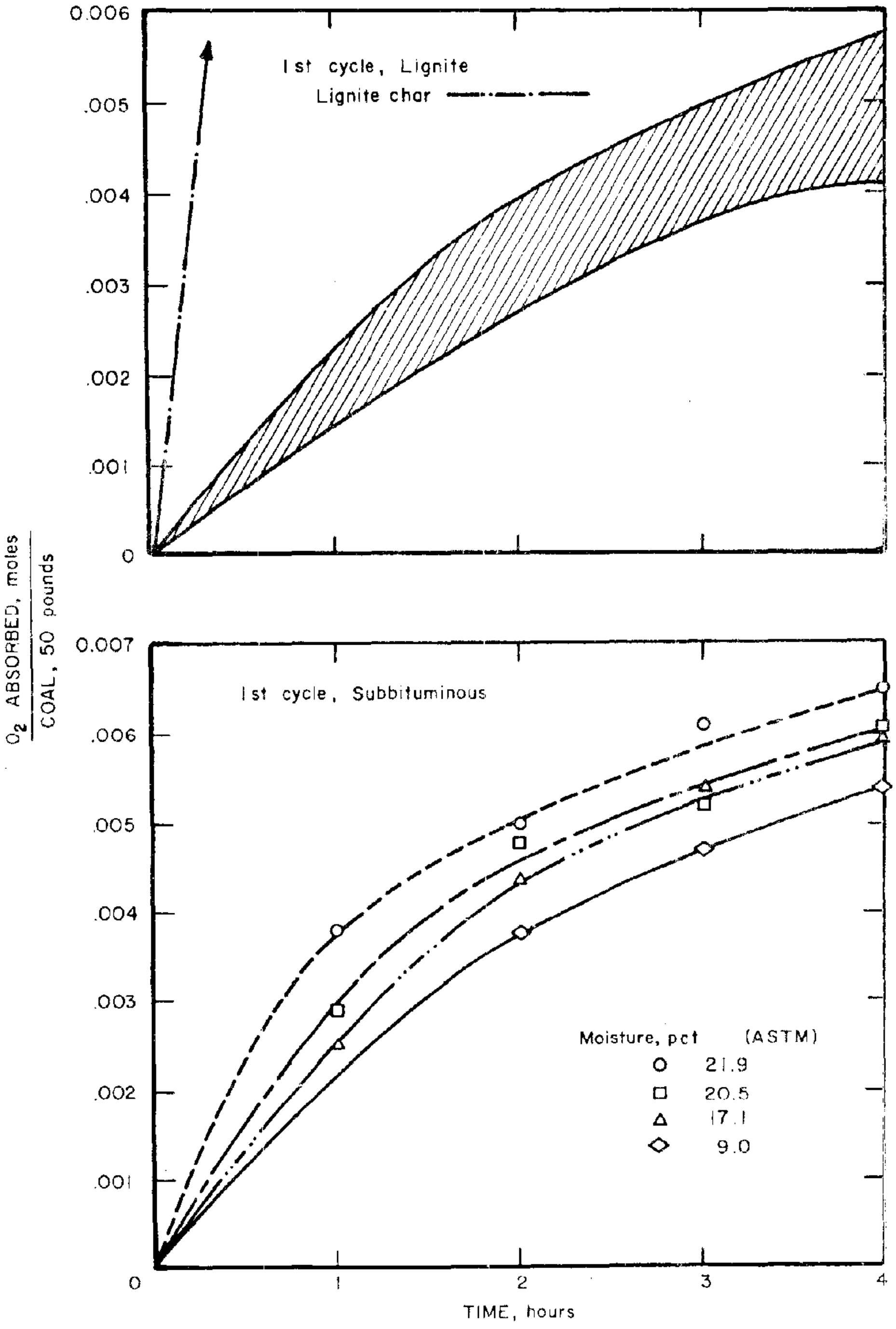

FIGURE 39. - Oxygen absorbed by dried lignite, lignite char, and subbituminous coal—first-cycle tests.

coal, from 9 to 21.9 pct. The moles of oxygen absorbed by the 50-pound samples of lignite (plotted as a range) and of subbituminous coal as a function of time is shown in figure 39. Previous studies[17] have shown a relationship between reactivity to oxygen and moisture content of sample when drying was done in an inert atmosphere. Drying for these tests, however, was done in an oxidizing atmosphere so that residence time in dryer and exposure to oxidation varied with product moisture content. Size consist was also subject to variation. Therefore, a definite relationship between moisture content and reactivity to oxygen was not established. Test results may also demonstrate that compensating factors that occur during drying, as would be carried out in commercial practice, tend to decrease differences in reactivity. Individual test data is

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[17] Second work cited in footnote 16.

plotted for subbituminous coal. The apparent relationship between moisture content and reactivity in these cases may not be valid because the number of tests and the range of moisture content is limited. However, subbituminous coal does appear to be slightly more reactive than lignite.

The product temperature of the dried lignite ranged from 165° F for samples with 8-pct-moisture content to 135° F for samples of 27-pct-moisture content; the temperature of dried subbituminous coal ranged from 150° F for the sample with 9-pct-moisture content to 135° F for the sample with 21.9-pct-moisture content. The most reactive dried coal samples depleted the oxygen content in the chamber to 9.5 pct at the end of 4 hours. Also shown in figure 39 for comparison is the reactivity of freshly produced 15 pct volatile matter lignite char and freshly dried lignite. The char sample was tested in the oxidation drum at 200° F, the temperature at which the sample was collected from the pilot plant carbonizer. The extreme reactivity of freshly produced lignite char toward oxygen at this temperature is indicated by the data.

It would be expected that when oxygen reacts with a carbonaceous material, carbon dioxide is produced. The rate at which CO_2 is given off during the first cycle per 50 pounds of lignite and subbituminous coal is shown in figure 40. For both coals, the amount of carbon dioxide released is very limited. Either the carbon dioxide remains on the particle or other oxygen compounds are produced.

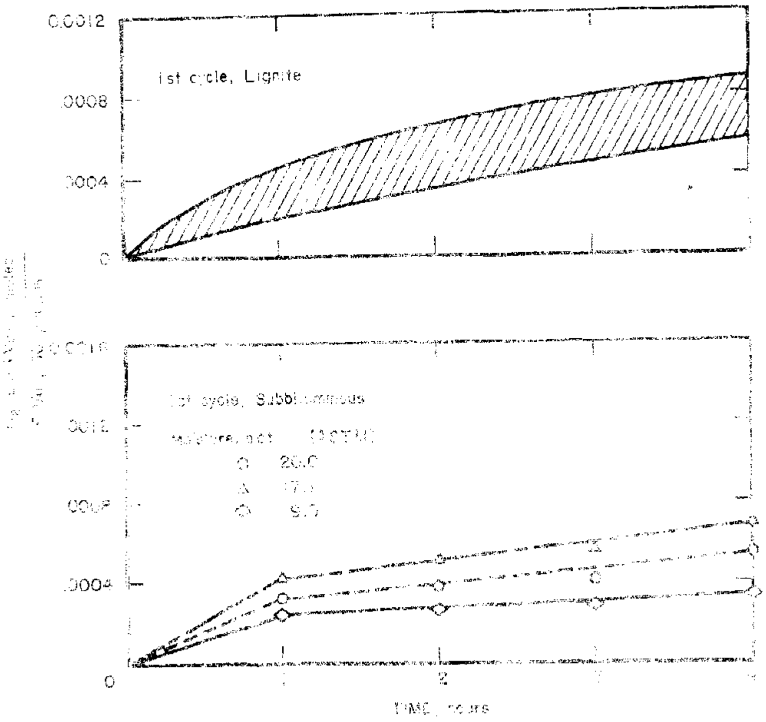

FIGURE 40. - Carbon dioxide evolved in first-cycle tests of dried lignite and subbituminous coals.

Second-Cycle Test

The influence of a cooling and aging process on the oxygen absorption characteristics of dried coal was determined in second-cycle tests. A similar process would possibly be feasible in commercial practice. For this test, the samples used in the first-cycle tests were permitted to cool to room temperature in the oxidation chamber then removed and placed in a plastic bag. The oxidation chamber was then reheated to 130° F, the coal sample replaced in chamber, and the test procedure repeated. Test data obtained in the second-cycle tests from three lignites of moisture contents from 18.4 to 19.2 pct is shown in

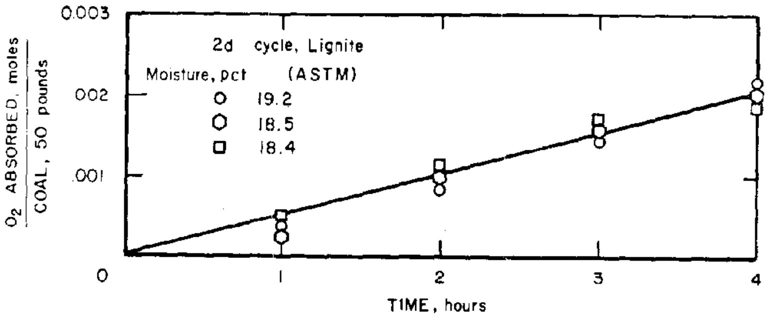

FIGURE 41. - Reproducibility of test method, second-cycle tests.

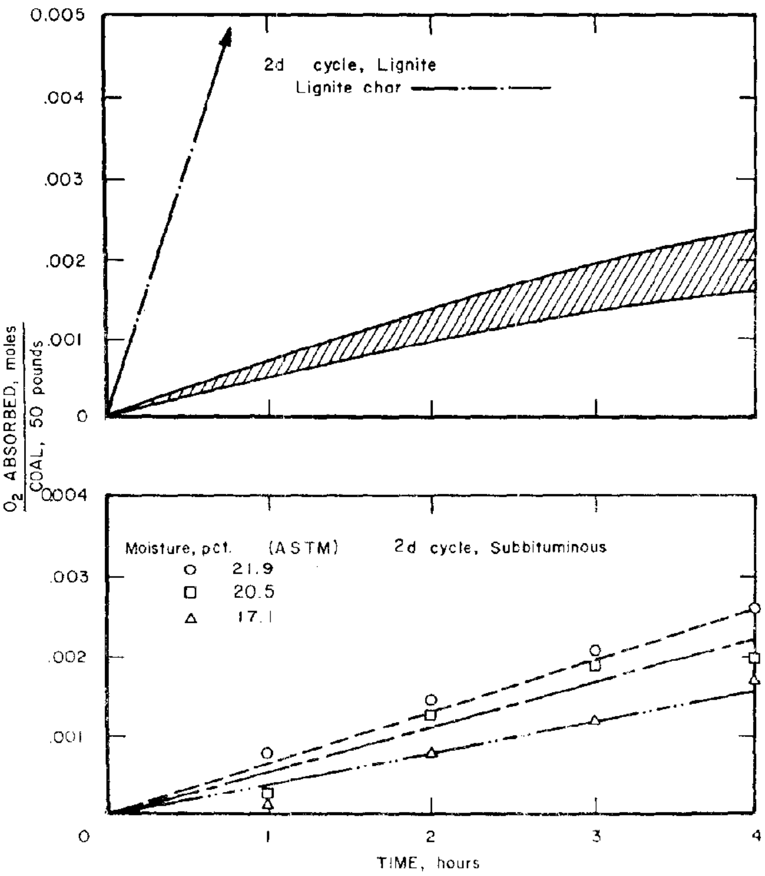

FIGURE 42. - Oxygen absorbed by dried lignite, subbituminous coals, and char—second-cycle tests.

figure 41 to indicate the reproducibility of test procedure. The range of values obtained in second-cycle tests is somewhat less than that obtained for first-cycle tests.

Test results from second-cycle tests for samples having moisture content ranging from 8.0 to 27 pct for lignite and 17.1 to 22 pct for subbituminous coal are given in figure 42.

Compared with results obtained in the first-cycle tests, the amount of oxygen reacted by both lignite and subbituminous dried coals is significantly reduced by the cooling and aging treatment. The range of values for lignites of different moisture contents was also reduced as a result of the treatment. Again a definite relationship could not be detected between the degree of oxygen absorption and moisture content for lignites, and values are consequently plotted as a range.

Results obtained from a second-cycle test performed on the sample of lignite char are also included. The temperature at which the char was tested was the same as for the dried coal. Although there appears to be some reduction in reactivity compared with first-cycle tests, char is still much more reactive than dried lignite or subbituminous coals.

The cumulative moles of carbon dioxide evolved per 50 pounds of dried product for the Glenharold lignite and for the subbituminous coal during the second-cycle 4-hour test period are shown in figure 43. As for the first cycle, the amount of carbon dioxide released is very low.

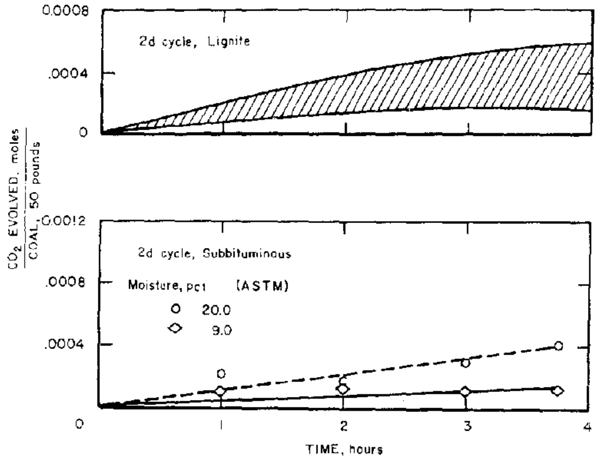

FIGURE 43. - Carbon dioxide evolved in second-cycle tests of dried lignite and subbituminous coals.

Temperature Tests 1, 2, and 3

The rate at which coal oxidizes is a function of temperature. Tests were performed to evaluate the difference between oxygen absorption rates of dried coals at 75°, 130°, and 190° F. The samples of dried coal for these tests were prepared by cooling the hot product from the dryer in a filled, closed container. The oxidizer was heated to the desired temperature level, and the tests was performed as described previously. Samples of as-received lignite and subbituminous coal were also tested for comparative purposes.

At a temperature of 75° F (room temperature), there was no detectable oxygen absorption for any sample. At a temperature of 130° F the amount of oxygen absorbed was quite similar to that reported in the second-cycle tests. At 190° F the rate of oxygen uptake is markedly increased for both lignite and subbituminous coal.

The amount of oxygen absorbed by the 50-pound samples of lignite as a function of time at 130° and 190° F are given in figure 44. Within the range indicated for lignite, values were obtained in seven tests on samples with moisture contents ranging from 18.4 to 36.5 pct. Results obtained from four samples of dried subbituminous coal ranging in moisture content from 9.0 to 34.3 pct are given in figure 45. For both subbituminous and lignite coals, highest moisture content represents the as-received samples. As indicated previously, the surface of the as-received subbituminous coal was noticeably wet with corresponding unusually high moisture content.

The oxygen absorption characteristics of samples of as-received and undried lignite when tested at temperatures of 130° and 190° F showed oxygen absorption properties very similar to the samples of dried lignites utilized in these tests. After one cycle of cooling experienced by these samples, the rate of oxygen absorption is determined primarily by temperature. Based on the size studied, test lignites after drying and cooling were as reactive as raw coal at the same temperature. Data suggests, however, that the dried subbituminous coal is more reactive than the undried. For both lignites and

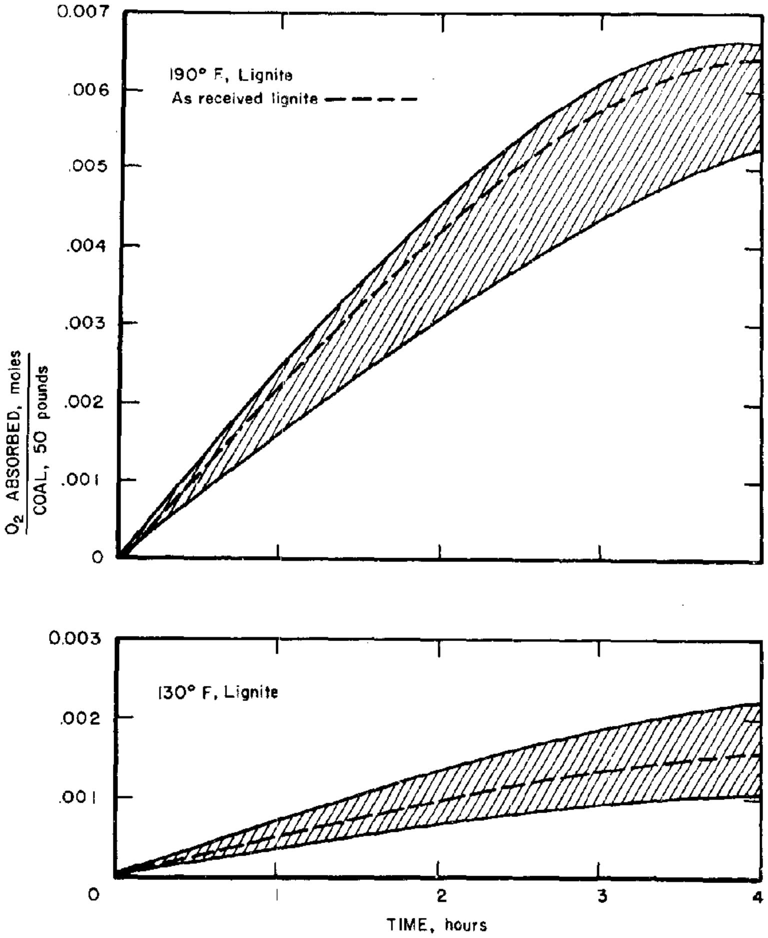

FIGURE 44. - Oxygen absorbed by dried lignite as a function of time at 130° and 190° F.

subbituminous coal, a cooling procedure after it was produced would greatly decrease the rate at which the dried material absorbs or reacts with oxygen.

Figures 46-47 show the evolution of carbon dioxide from test samples of lignite and of subbituminous coal, respectively, as a function of time and temperature. A range of values is plotted for lignite of different moisture contents. For both coals, the carbon dioxide given off is significantly influenced by the temperature of the coal and is significantly greater at 190° F than at 130° F. The amount of carbon dioxide released was stoichiometrically less than the oxygen absorbed in all tests.

Tests were also performed to compare the reactivity of the dried lignite from test stockpiles 1 and 2 with a sample of similar moisture content prepared in the pilot plant dryer and also to a sample of as-received coal from the same mine. The rate of oxidation was measured at 75°, 130°, and 190° F. Results are shown in figure 48.

In contrast to results obtained on lignite from the Glenharold mine, the reactivity of Baukol-Noonan lignite, freshly dried to 23 pct moisture, was greater than that of the as-received coal. The dried coal from the stockpiles was much less reactive than either of the other samples, which demonstrates that extensive aging does reduce reactivity. Despite a low tendency to absorb oxygen, the fact that this aged dried lignite could heat when stored improperly was demonstrated by the heating that occurred in the conical stockpile made from previously stored lignite.

Cooling Rate Tests

An effective and obvious procedure to reduce the rate of oxidation and potential for spontaneous heating is to reduce the temperature of dried coal before shipment. Such cooling could be incorporated in the production process

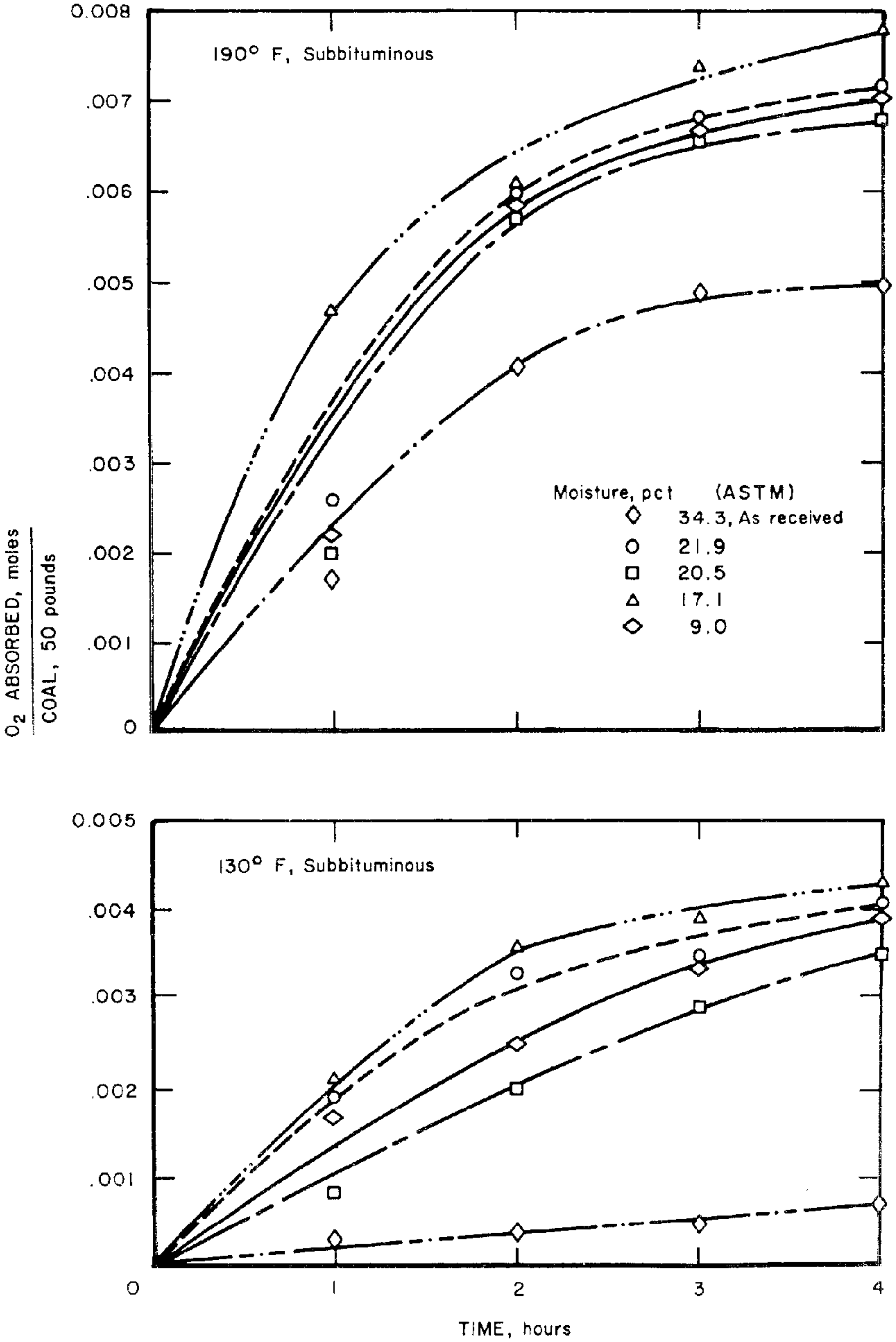

FIGURE 45. - Oxygen absorbed by dried subbituminous coal as a function of time at 130° and 190° F.

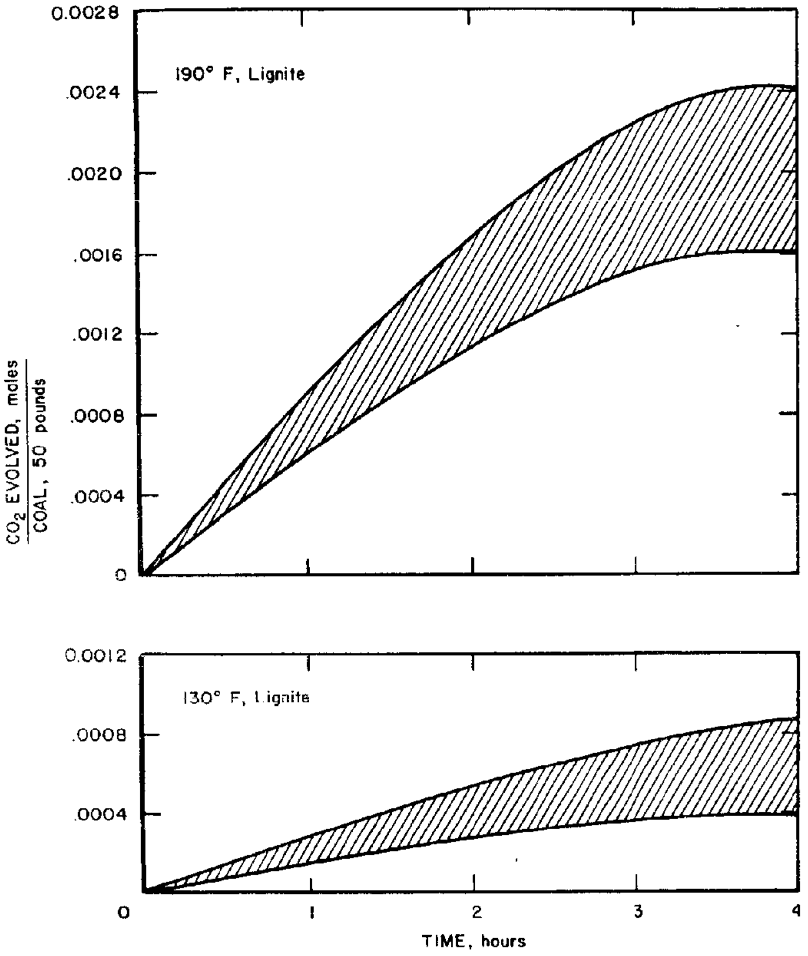

FIGURE 46. - Carbon dioxide evolved from dried lignite at 130° and 190° F.

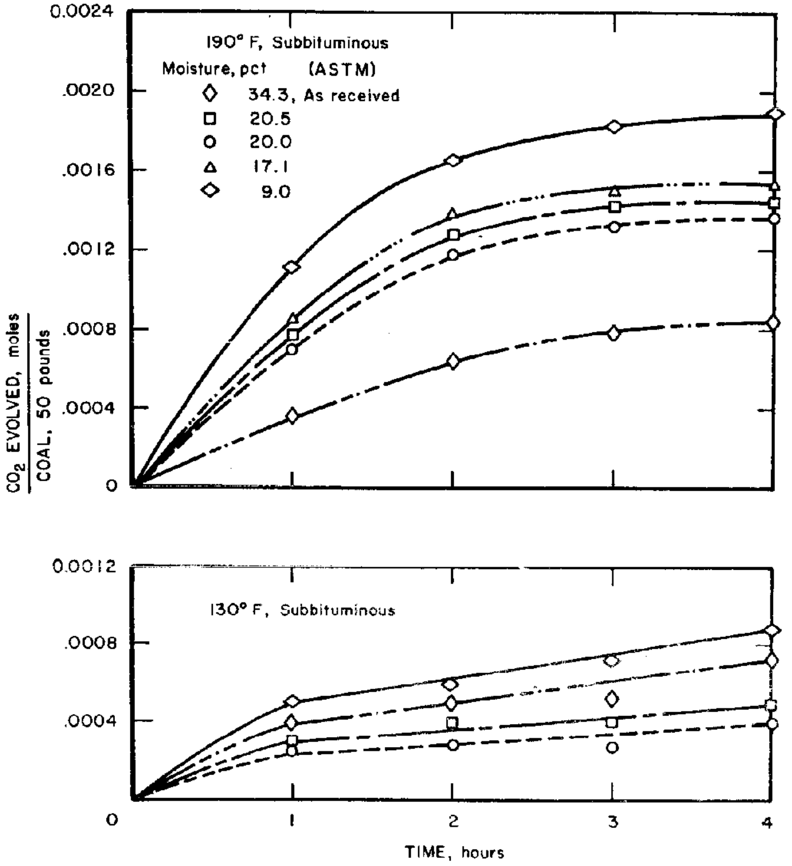

FIGURE 47. - Carbon dioxide evolved from dried subbituminous coal at 130° and 190° F.

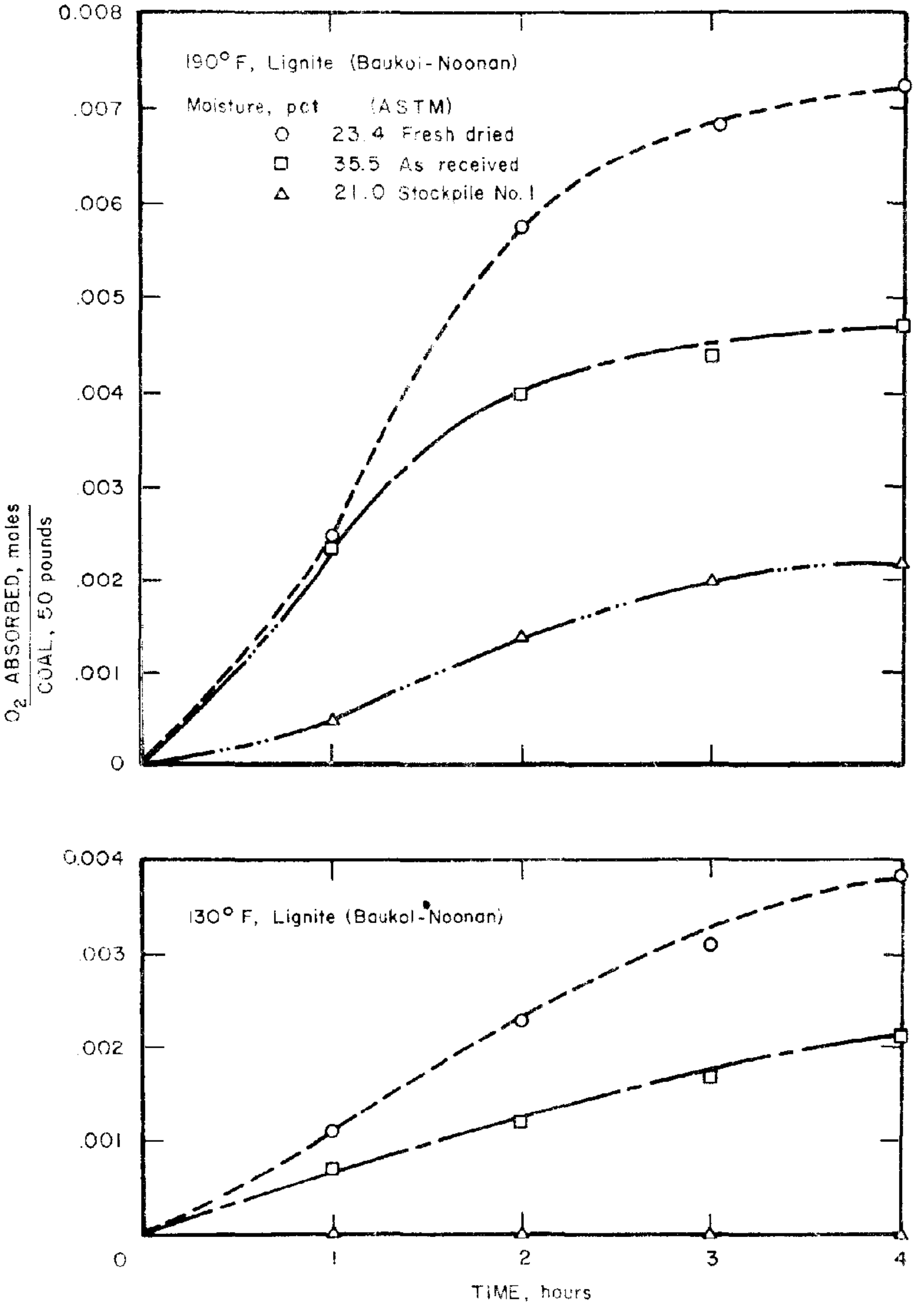

FIGURE 48. - Comparison of oxygen absorption of as-received, freshly dried, and dried and aged Baukol-Noonan lignite at 130° and 190° F.

by holding bins or during shipment. Accordingly, data was obtained on the heat transfer characteristics of test samples in a simulated hopper or bin exposed to ambient room temperatures. Approximately 1,400 pounds of dried product was placed in a covered 38-inch-diameter by 43-inch-high steel container. Temperatures were measured at specific locations in the container by a system of thermocouples and continuously recorded. During an initial period that ranged from 30 to 60 hours, the temperature of the dried coal increased for each test because of the reaction and absorption of oxygen present in the void spaces. During this period in each test, the exterior surface of the sample container was insulated so that a maximum temperature value could be determined corresponding to conditions in which heat loss to surroundings was minimum. When the maximum temperature was established, the insulation was removed, and the coal was allowed to cool to determine rate of heat transfer to the surroundings.

The average value of the four individual measurements located at the center of the container as a function of time is plotted for lignite samples of 8 and 19.2 pct moisture in figure 49. The initial temperature of the lignite dried to 8-pct-moisture content was 165° F. A maximum temperature of 175° F was reached after 30 hours, at which time the insulation was removed. The sample of lignite of 19.2-pct-moisture content had an initial temperature of 127° F and reached a maximum of 139° F also after 30 hours. The temperature of test samples decreases very slowly, indicating that heat loss in shipment or storage is a slow process for dried coals.

Figure 50 shows similar data for subbituminous coal. The initial temperature of subbituminous coal dried to 9-pct-moisture content was 150° F and a maximum of 190° F was reached after 45 hours. The sample dried to 17.1 pct moisture was 135° F initially, but also reached a maximum of 190° F after 60 hours. The temperature at the container center for the sample of 17.1-pct-moisture content did not decrease for the period shown. Temperatures at the outside of the cooling drum did show, however, a decrease in temperature over the time period. Compared with those of dried lignite, heat transfer rates in dried subbituminous coal were significantly lower. Results indicate that the temperature rise in the carload shipments of dried low-rank coals previously described is due to reaction with oxygen of the air within void spaces.

Moisture Reabsorption Tests

Dried coal will release or absorb moisture until equilibrium is reached corresponding to the vapor pressure of water present in the surrounding atmosphere. The rate at which the moisture content of samples of dried lignite and subbituminous coal increased if subjected to saturated conditions was evaluated. The samples for these tests were prepared by cooling the dried coals in an inert atmosphere and subsequently exposing "as-dried" 5-gram lots to a 100-pct-water-saturated atmosphere at room temperature. The 5-gram samples in watchglasses were simply floated on the surface of a water-filled pan that was covered with plastic sheeting to maintain 100 pct humidity. Each day for a total of 10 days, three of these samples were removed and the moisture content determined.

The rate of moisture pickup and equilibrium moisture content for dried lignite and subbituminous coals produced in this investigation is shown in figure 51. Data for dried lignite samples were obtained in 11 tests on samples having moisture contents from 8 to 27 pct. Data on subbituminous coal was obtained in four tests of samples of 9 to 21 pct moisture. The major part of the moisture gain occurred in the first 2 to 3 days. Before drying, moisture content of the lignite was about 36 pct and about 30 pct for the unwetted subbituminous coal.

The rate of moisture pickup by the dried material is approximately the same for both lignite and subbituminous coals. Lignite and subbituminous coal, when dried to the 8-pct range, reabsorbed moisture to the 22- to 25-pct level. For samples of both coals dried to the 21- to 24-pct moisture level, a 27- to 30-pct level was reached at equilibrium.

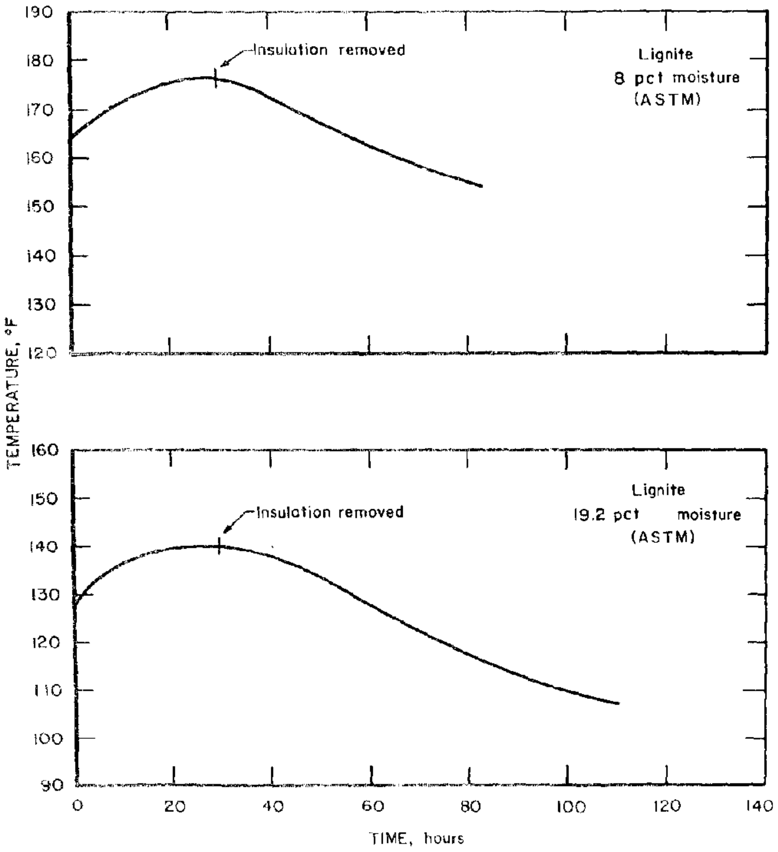

FIGURE 49. - Temperature history of dried lignite in a simulated bin.

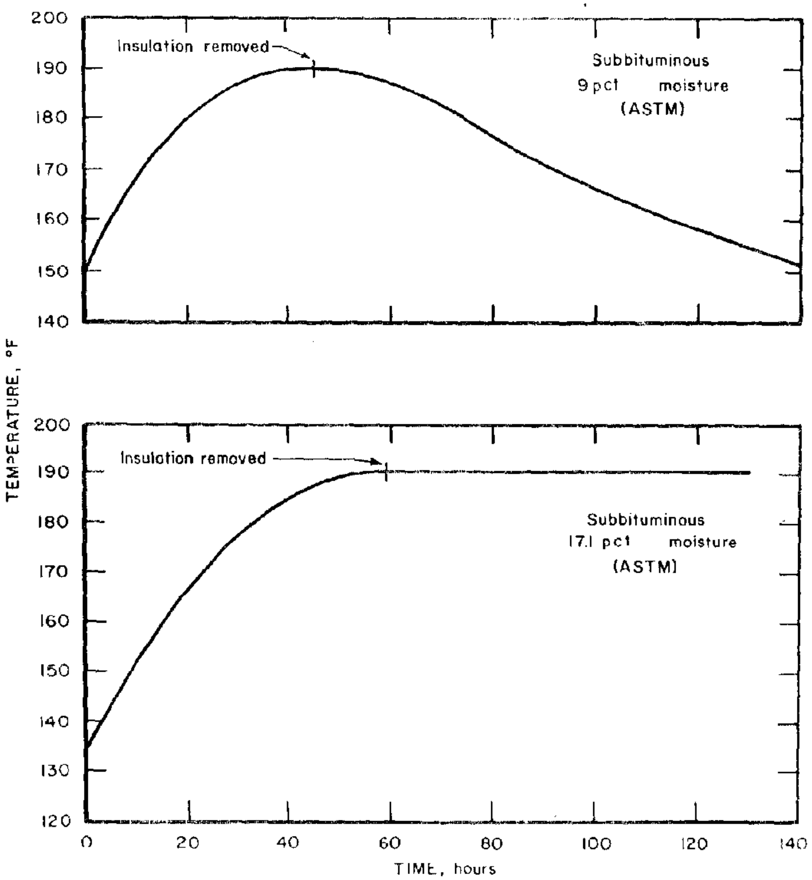

FIGURE 50. - Temperature history of dried subbituminous coal in a simulated bin.

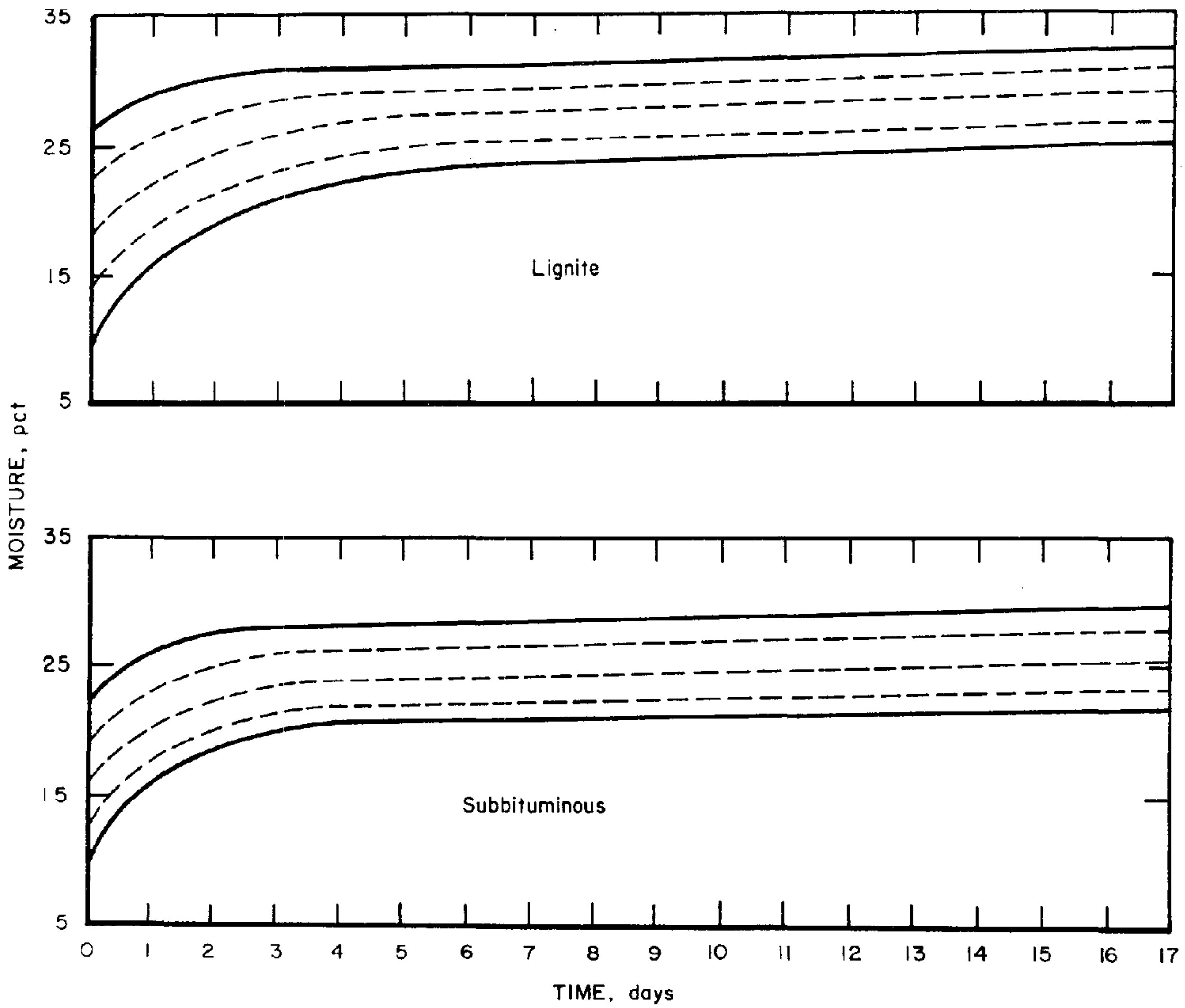

FIGURE 51. - Moisture gain and equilibrium moisture content of dried low-rank coals.

Samples of the stockpiled Baukol-Noonan coal were also tested. Figure 52 shows the moisture change of the stockpile sample compared with that of freshly dried Baukol-Noonan lignite. In each case the starting moisture content was similar, and the moisture reabsorption rate was also similar.

Density and Settling Tests

The bulk density of dried low-rank coals determines the size of handling and transportation equipment required to transport given quantities of material in a given time. The container for heat transfer tests was also utilized to provide information on settling characteristics and bulk density. After the container was filled for the heat transfer tests, it was weighed and the heat transfer test was performed. When the temperature studies were completed, the container was opened, and the walls of the drum were vibrated with a jackhammer until no further settling of the coal occurred to obtain an estimate of

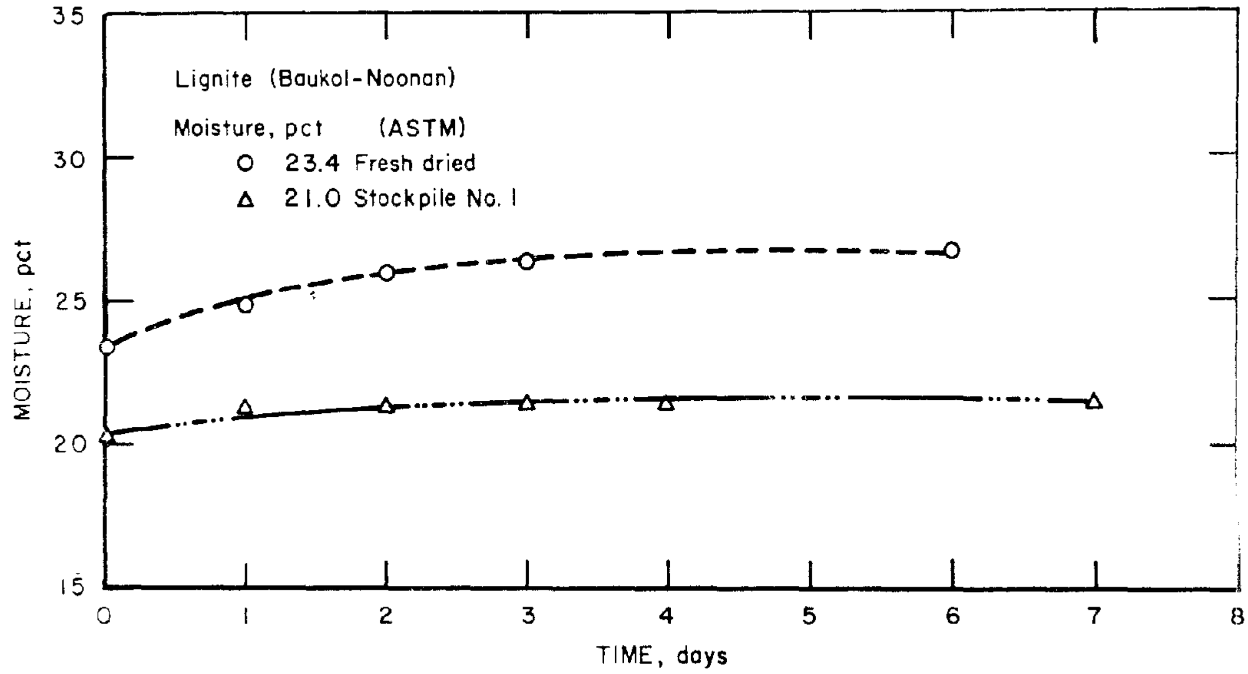

FIGURE 52. - Moisture readsorption of dried Baukol-Noonan lignite.

its "settled" density. The bulk density computed from the change in volume occupied by the coal before and after the vibration procedure increased by about 10 pct for dried subbituminous coals and up to 13 pct for dried lignites.

Values determined are shown in figure 53. Moisture content of lignite samples ranged from 8 to 36.2 pct for the as-received lignite; for the subbituminous coal, the moisture contents ranged from 9 to 34.3 pct for the as-received subbituminous coal.

Bulk density of a sample of Baukol-Noonan lignite collected from the test stockpiles was also measured. Loose packed density was 50.5 lb/cu ft and the vibrated density was 53.2 lb/cu ft. The moisture content of the sample was 22 pct.

Size Consist of Dried Coal Samples

A size analysis was made of a sample of dried product as tested. The initial size analysis of the lignite and of dryer product at four levels of moisture content is given in table 3. Similar data for the subbituminous coal and dried product are given in table 4.

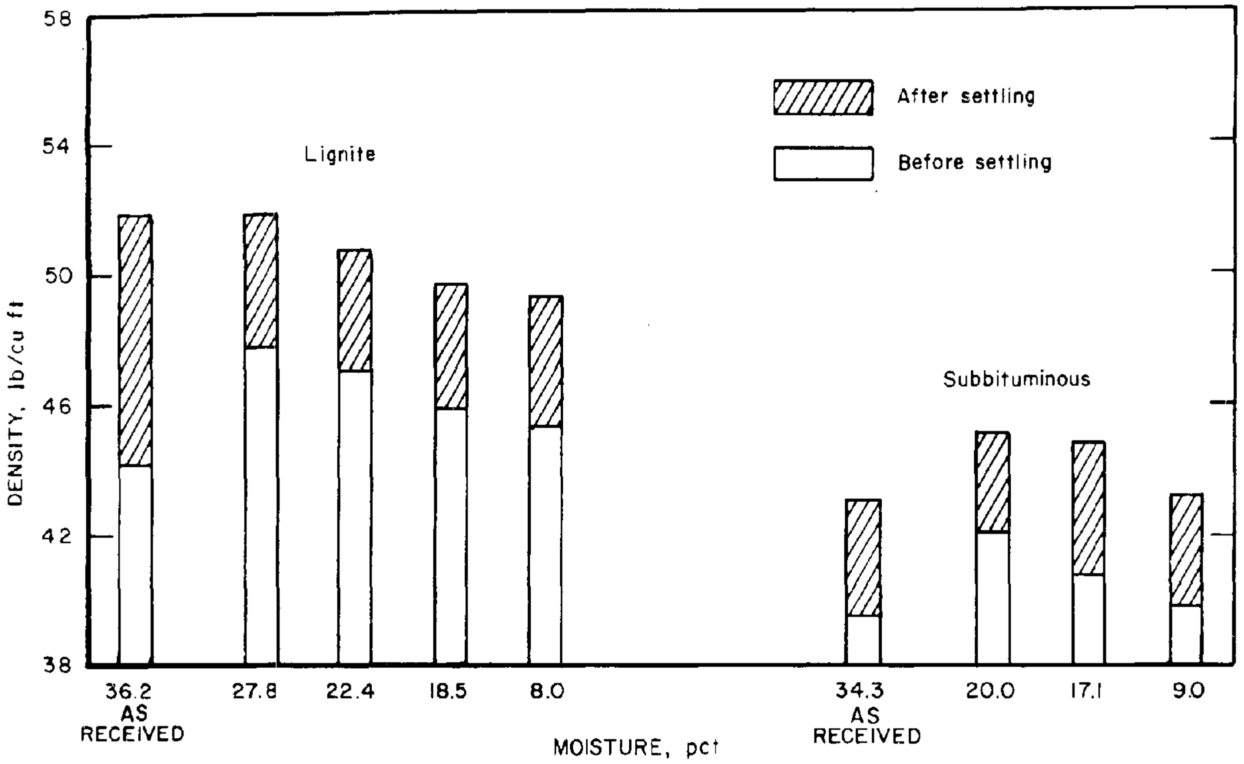

FIGURE 53. - Bulk density of dried low-rank coals.

TABLE 3. - Size distribution of dried lignite

	Size distribution, pct				
	Initial	Product			
Moisture content, pct........	36.3	27	22.3	18.4	8
Size fraction, wt-pct:[1]					
Plus 4 mesh...............	17	4	7	7	10
4 by 8 mesh...............	23	26	18	18	22
8 by 16 mesh..............	26	28	27	27	30
16 by 28 mesh.............	11	14	14	14	14
28 by 48 mesh.............	11	11	14	14	11
48 by 100 mesh............	5	8	8	8	6
100 by 200 mesh...........	4	4	5	5	3
Minus 200 mesh............	3	5	7	7	3

[1] Tyler sieve series.

TABLE 4. - Size distribution of dried subbituminous coal

	Size distribution				
	Initial	Product			
Moisture content, pct	34.3	21.9	20.5	17.1	9.0
Size fraction, wt-pct:[1]					
Plus 4 mesh	30	7	12	25	12
4 by 8 mesh	27	15	16	30	25
8 by 16 mesh	20	25	23	25	29
16 by 28 mesh	11	16	12	9	14
28 by 48 mesh	7	13	13	5	9
48 by 100 mesh	3	10	8	3	4
100 by 200 mesh	1	5	6	1	4
Minus 200 mesh	1	9	10	2	3

[1] Tyler sieve series.

Summary of Pilot Plant Test Results

Test samples of dried low-rank coals have been studied and compared as a function of moisture content. A procedure was developed by which the relative reactivity of test samples can be compared and evaluated. To date, a number of the physical properties of test samples have been evaluated and compared as a function of moisture content.

The reactivity of the freshly dried test samples of lignite to oxygen was not found to vary significantly with the moisture content. This suggests that the degree of moisture reduction is not a major consideration in respect to product reactivity. A major factor in respect to reactivity of lignite under any given sample condition is temperature. A process in which the dryer product is cooled and exposed to air was found to be very effective in reducing reactivity towards oxygen. In general, dried subbituminous coal was somewhat more reactive than dried lignite. At elevated temperatures, reactivity of dried subbituminous coal was found to be greater than that of the as-received coal tested at the same temperature.

Reaction with oxygen of void space air produces an initial temperature rise in dried coals during transport or in stockpiles. The rate of heat transfer in dried coals is slow. Little cooling can be accomplished in an enclosed bin or stockpile within reasonable time periods without benefit of a suitable conductant mechanism or procedure. Dried low-rank coals will regain a portion of the moisture removed if exposed to saturated conditions. The lower the initial moisture content of dried coal, the lower will be the final moisture content after exposure. In no case does the moisture content return to original moisture content. A period of 3 to 4 days of continuous exposure is required for the reabsorption process to be completed.

The bulk density of dried low-rank coals without a vibration or settling treatment is greater than that of as-mined coals. A vibration or settling procedure was found to increase bulk density of dried coals but to a lesser degree than that which occurred for the as-mined coal. Results suggest that volume requirements for handling and storage of a given weight of dried low-rank coals are not greatly different than those required for as-mined coals.

Status of Investigation

The experience and information developed provide an improved basis upon which the problems involved in shipment, handling, and storage of dried low-rank coals can be projected. Undoubtedly other as-yet-untested variables will also be important, but progress has been made in evaluating a number of important factors. It is particularly significant that dried lignites and subbituminous coals have been produced, shipped in open-top cars with normal handling, unloaded, and placed in stockpiles that were stable for several years.

The relative tendency of dried low-rank coals to react with oxygen of the air has been compared with that of as-mined coals. The value of cooling and aging to reduce this tendency has been demonstrated. Data regarding the physical characteristics of dried low-rank coals has been compiled including bulk density, heat transfer properties, and moisture regain and loss. This information has direct application to consideration of the practical aspects of commercial-scale processing.

Future efforts will be to develop a simple and practical method by which the reactivity and dust will be reduced and the compaction properties improved to permit safe and trouble-free transportation, handling, and storage of dried low-rank coals.

GAS FROM COAL: FUEL OF THE FUTURE[18]

by

G. Alex Mills[19]

The Age of Synthetic Fuels

The lignite symposium is a special opportunity to discuss energy problems and opportunities. The Red River Valley is famous for its farming capabilities, and farming is basically an energy conversion process. But North Dakota is destined to become increasingly famous as the site of enormous lignite deposits, themselves an agricultural product, and what is more, as the site of plants for converting lignite into electricity and into synthetic fuels.

Synthetic fuels are of sharply increasing interest in the United States. Commercial plans for the manufacture of gas from coal are progressing rapidly. The age of synthetic fuels from coal for heating homes, for generating electricity, for running factories, and for powering cars has arrived, but paradoxically, it is not here. It is not here because it will be a decade before synthetic fuels will make any appreciable impact on the Nation's energy supply. However, it is with western coals that the first commitments are being made in the United States for the manufacture of synthetic fuels from coal. The energy business is so large that a lead time of about 10 years is required to make any appreciable change.

Applications have been made to the Federal Power Commission to construct two large coal gasification plants in the Four Corners area. These will use German technology--essentially the so-called Lurgi Process--which has been in commercial operation in Europe. For future installations, new improved processes now in an advanced pilot plant state in the United States are believed to offer economic advantages and to have a wide application to American coals.

The spotlight was turned on coal gasification in the President's June 1971 energy message in which he said, "As we carry on our search for cleaner fuels, we think immediately of the cleanest fossil fuel--natural gas. But our reserves of natural gas are quite limited in comparison with our reserves of coal. Fortunately, however, it is technically feasible to convert coal into clean gas that can be transported through pipelines. The Department of the Interior has been working with natural gas and coal industries on research to advance our coal gasification effort....We are determined to bring greater focus and urgency to this effort."

The tempo has been further accelerated by recent recommendations of Congressional energy committees and by the President's second energy message

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[18] Luncheon address.

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delivered less than a month ago. Special emphasis is proposed for development of low-cost, clean-burning forms of fuel from coal. The proposed fiscal year 1974 budget included a 27-pct increase to $120 million for coal research and development.

Coal Gasification in 1985-90

The first gasification plant will not be completed for at least 3 years. However, a dozen years later, projections indicate that 36 plants will be in operation, each manufacturing 250 million scfd of pipeline gas and each consuming 6 to 10 million tons of coal annually, depending on the type of coal. This would mean the following:

Coal.--A total of about 300 million tons of coal would be utilized each year just for making gas, an increase of 50 pct from present total coal production in the United States. A commitment of 9 billion tons of coal would be required for a 30-year reserve.

Water.--Each plant will use a total of 6,000 or more gpm of water. Since the average person uses 300 gpd, water consumption for these 36 plants is equivalent to water usage of a million persons.

Dollars.--Each plant will cost over $400 million including the mine, for a total cost of about $14 billion. For comparison, the net assets in the United States of the 30 largest petroleum companies were $37 billion in 1971. The value of the gas at the plant at $1 per Mcf will be $3 billion annually. This amounts to approximately the total annual sales value at the mine of all U.S. coal today. In the recent past, the price of gas at the wellhead has been about 20 cents per million Btu (the cheapest energy fuel except for lignite). Certainly the most phenomenal change will be the quantum jump in the price of "new" gas. A further economic factor is the potential savings by improved gasification processes. For example, a savings of 20 cents per million Btu, thought to be possible with the newer processes being developed, would correspond to a savings of $600 million every year.

People.--It has been estimated that over 1,000 persons per plant would be employed, about 500 in the mine and 500 in the gasification plant.

Gas Produced.--The 36 plants would produce an aggregate of 3 trillion cu ft of gas annually. The present yearly gas usage is 23 trilling cu ft. The 3 trillion cu ft is sufficient to supply the total energy needs (coal, oil, and gas) of 9 million persons in the United States at the present energy-use level. Finally, it may be noted that this 3 trillion cu ft will fulfill only a fraction of the demand since the gas gap--the shortfall between supply and demand in 1985-95--has been estimated to be as much as 17 trillion cu ft. So the potential market for gas would be six times larger than the amount produced by 36 plants.

The estimates for the 1985-95 period are not an over-enthusiastic dream but are projections made by expert committees officially reporting to the

Federal Power Commission and the National Petroleum Council. Neither group has a reputation for radical pronouncements.

It is estimated that it will take 15 years to construct the first 36 plants. The projection is that the next 36 will require only 5 years.

Energy Projections and Choices

To indicate what coal gasification can do for our future energy needs, first consider projections made by the Bureau of Mines and others. Our energy needs are expected to grow 3.5 pct annually for the balance of this century. This means that by 1985 nearly twice as much energy must be provided compared with the present, and nearly three times as much will be needed by the year 2000.

Natural gas now supplies one-third of the Nation's total energy requirements, including approximately half the demand for residential, commercial, and industrial consumers. One-fourth of the needs of steam electric powerplants is also supplied by gas. At present, natural gas represents the largest energy fuel in terms of Btu's both produced and used in the United States. Moreover, the rate of gas consumption has far exceeded the growth rate of other fuels and continues at an annual rate of increase of nearly 6 pct. During the last 5 years, more gas has been used than found, and gas reserves are inadequate to satisfy future needs.

What alternate choices do we have to meet our energy needs? Nuclear energy cannot be a major short- nor medium-term answer since nuclear energy is expected to supply only one-fourth of our electrical energy needs by 2000. Electrical energy represents less than 10 pct of our total energy use.

The vast importation of foreign oil offers a choice with many problems. We are importing about 4 million barrels of oil per day. If we depend on foreign oil to supply our increased energy needs, it will require importation of 15 million barrels per day by 1985. This would require a line of supertankers from here to the Near East spaced only about 7 miles apart. It will be difficult, if not impossible, to support a cost escalating from the present $4 billion to $25 billion by 1985. The potential of tens and even hundreds of billions of U.S. dollars in foreign hands has also become a recognized hazard.

All the foregoing factors, together with the great increases in the cost of clean energy and current technical advances, provide the opportunity for "gas from coal" to be the "fuel of the future."

What Is Coal Gasification?

Coal gasification consists of the chemical transformation of solid coal into gas. Pipeline gas consists essentially of methane. It has a heating value of about 1,000 Btu per cu ft and is virtually free of sulfur.

Synthetic gas having the specifications of natural gas can be manufactured from coal by first producing a synthesis gas, followed by purification and catalytic methanation. The Bureau of Mines Synthane Process begins with coal preparation, in which coal is ground to a minus 20-mesh powder. It is then pretreated with oxygen to destroy caking properties because heating causes some coals to swell and plug the reactor. Then follows the gasification step in which synthesis gas is formed by reacting steam and oxygen with coal. The synthesis gas contains carbon monoxide, hydrogen, and methane as valuable components, and carbon dioxide, sulfur compounds, tars, and dust as impurities that must be removed before further processing.

After tars and dust are removed, the gas is conducted over a "shift" catalyst where the reaction between carbon monoxide and steam occurs to form hydrogen and carbon dioxide. The objective is to bring the hydrogen-to-carbon monoxide ratio to 3 to 1, which is required for the subsequent methanation step that uses 1 volume of carbon monoxide plus 3 volumes of hydrogen to form 1 volume of methane plus 1 volume of water.

An intensive purification step precedes methanation since strict sulfur removal is required to prevent inactivation of the nickel catalyst. The product from the methanation step has a heating value of about 950 Btu/cu ft and is entirely suitable for use as a pipeline gas.

The Bureau of Mines has a long productive history in coal gasification research. In fact, it is fair to say that all of the processes under development in the United States depend to a significant degree upon research carried out over many years by the Bureau. It is to the Bureau's credit that much of its gasification research was done in anticipation of a national need for synthetic gas.

The Bureau Synthane Process

At present, four large-scale coal gasification processes are at the pilot plant stage in the United States.

The Synthane Process, signifying the manufacture of synthetic methane, takes advantage of advances in modern fluid-bed design technology in contacting coal with steam and oxygen in the gasification step.

There are several process features to the Synthane Process that recommend it. First, the process is simple in that the gasifier consists of a single fluid bed. This is believed to provide for reliable and trouble-free operation and may be the most important feature of the Synthane Process.

Second, raw coal of any kind, including lignite, is fed to the unit in such a manner that all the gases produced including devolatilization products are made available as product gas to maximize production of methane.

Third, by using the concept of both a high-temperature zone and a low-temperature zone in the gasifier, the methane yield within the gasifier is increased. In fact, more than half the ultimate methane is made directly in the gasifier, which production minimizes both oxygen consumption and the volume of gas that must be purified and methanated.

Finally, a unique methanator is employed. This unit employs a nickel catalyst flame that is sprayed directly onto heat exchange tubes. Heat removal through the tube wall controls reaction temperature very well.

Status of Synthane Pilot Plant

A large pilot plant is under construction of a 5-acre site at the Bureau of Mines station at Bruceton, near Pittsburgh, Pa. Design was based on extensive work carried out in a laboratory pilot plant processing 10 to 30 pounds of coal per hour. Additionally, an intermediate-scale pilot plant has been operated by Hydrocarbon Research, Inc., at 400 psi and charging 600 pounds of coal an hour. This test demonstrated several of the key features of the Synthane Process on a larger scale including the fluid-bed integral pretreater, an 18-inch-diameter gasifier, and a steam-oxygen inlet cone. The pilot plant under construction will process 3 tons of coal per hour and is capable of producing about 1.25 million cu ft of pipeline gas per day. The pilot plant design was completed by the Lummus Co. in mid-1972. The Lummus Co. was also authorized to purchase three long-delivery items, including the gasifier vessel. The Rust Engineering Co. has contracted to build the Synthane plant with the completion date scheduled for August 1974. Overall cost of plant construction will be about $12 million. Additional important engineering assistance is being received from the C. F. Braun Co., which is under contract with Office of Coal Research (OCR), U.S. Department of the Interior, and the American Gas Association to study and make recommendations in regard to all gasification processes being funded by the Department of the Interior.

Lignite Gasification

Lignite has been tested extensively in the laboratory pilot plant. Because of high chemical reactivity, lignite can be processed at more than twice the rate of subbituminous coal and still obtain a high degree of conversion. For example, at the 1970 American Gas Association Synthetic Pipeline Gas Symposium, data were presented indicating that lignite at 870° C showed a 77-pct conversion in the Synthane Process, compared with 74-pct conversion for Illinois No. 6 at 950° C. Not only was the conversion higher, the temperature was 80° lower.

The high reactivity of lignite probably depends partly on its chemical structure and partly on the alkaline ash content, which is known to accelerate the slow steam-carbon reaction. Here is a case where the ash constituents of lignite, including the sodium content, can be a benefit.

Is gasification of lignite of commercial interest? The first two gasification plants using subbituminous coal are planned for New Mexico, about 500 miles from Los Angeles. The extensive North Dakota lignite deposits are only a little farther from high-density population centers, about 800 miles from Chicago. Moreover, gas transmission lines to which the substitute natural gas (SNG) can be added now go through North Dakota or are nearby. Additionally, the low cost of lignite is important since the cost of coal can amount to 40 pct of the cost of the gas. Cost estimates for synthetic gas made by the Lurgi process are $1.10 per million Btu using western coal compared with

$1.45 using bituminous coal. It is expected that the new processes being developed will decrease costs by $0.15. These costs are based on a plant producing 250 million cu ft daily, utility type financing, and a 10-pct return on the rate base. The rate base comprises 60 to 75 pct borrowed capital and the rest equity capital. If 100-pct equity and 12-pct discounted cash flow are used instead of utility-type financing, gas costs could increase $0.25. For reference purposes, it costs about 18 cents to transport 1,000 cu ft of gas 1,000 miles.

In a recent application for water permits, the Michigan Wisconsin Pipeline Co. requested a total of 375,000 acre-ft per year of water for a future enormous lignite-gasification project to be located in North Dakota. This water rate corresponds to 120 billion gal of water annually. Gasification plants would be located in the vicinity of Garrison Reservoir. Adequate lignite is available; a total of 4,384 million tons of lignite reserves are present in five fields. Twenty-two plants would be built, and each plant would require 17,000 acre-ft of water annually. It was estimated that investment in developing the necessary mines for this program would exceed $1 billion and the investment in coal gasification would exceed $5 billion.

Other Bureau Coal Gasification Programs

In addition to the Synthane project, the Bureau of Mines is carrying on several other important coal gasification research projects. Interesting work is being carried out with the stirred fixed-bed pilot plant in Morgantown, W. Va. Bureau of Mines Report of Investigations 7408 describes work completed at Grand Forks using a slagging fixed-bed unit. In addition, work is progressing on the hydrogasification of coal at Pittsburgh, Pa., using the Hydrane Process.

Perhaps the most unusual current work is the in situ or underground gasification underway in Wyoming. Combustion was initiated late in March of this year with promising results. New ideas to be tested include directional drilling, hydrofracturing, and use of chemical explosives to produce porosity in underground coalbeds to make them suitable for gasification in place. Many of these techniques have been developed in petroleum technology.

Until a few years ago, energy projections for the United States did not include a synthetic gas having a lower heating value than pipeline gas. Now, there are compelling fuel efficiency incentives to develop lower-Btu gas processes mainly for the electric power industry especially utilizing combined power cycles. Any oxygen-steam process for making SNG, such as the Synthane Process, can make a low-Btu gas by using air instead of oxygen for gasification and by eliminating shift conversion, CO_2 removal, and methanation. In addition, special opportunities are also offered by underground gasification and the Bureau's stirred fixed-bed process.

Finally, there are two areas of lignite gasification that I would like to mention. Both involve gasification but result in final production of a liquid. In the first, lignite is converted to a gaseous mixture of carbon monoxide and hydrogen that then is catalytically converted to methanol. Methanol is a liquid with value both as a boiler fuel and for use in automobiles. Its use in automobiles is made favorable because it has a research octane number of 106 (it is used in racing cars) and it produces much less pollution than does gasoline.

The second is concerned with the gasification of part of the lignite to carbon monoxide, followed by treatment of lignite with steam and the CO to form a liquid useful particularly as a boiler fuel. This process, discovered by the Bureau in Pittsburgh, is of interest as an improvement to the solvent refining of coal. Further work has been carried out by Berg and his students at the University of Montana, and the process should also be applicable in the pilot plant experiments on solvent-hydrogenation of coal to be carried out by the University of North Dakota.

As indicated, progress is slow on coal-to-gas processing technology. Our energy economy is so vast and so complex that about 10 years of lead time is required for any appreciable change to be made. Clearly, there is a need to accelerate our efforts toward commercial production of clean synthetic fluid-fuels--both gas and oil--from coal. Development of our domestic energy resources (and coal is our most abundant one) is the most sensible move that we can make toward strengthening our negotiating position in deals for oil from the Middle East and elsewhere.

SOME STUDIES ON STACK EMISSIONS FROM LIGNITE-FIRED POWERPLANTS

by

G. H. Gronhovd,[20], P. H. Tufte,[21] and S. J. Selle[22]

Introduction

Stack emission limits for new stationary sources of greater than 250-MMBtu/hr input were promulgated on a national basis by the Environmental Protection Agency (EPA) in 1971.[23] These standards include limits for SO_2, NO_x, and particulate matter and are applicable to all plants constructed or significantly modified after August 17, 19 1. In addition, there are State, and sometimes local, regulations pertaining to emissions from new and existing powerplants.

Little information is available in the literature on the quantity and quality of stack emissions from boilers burning North Dakota lignite. For this reason NO_x limits for lignite-fired plants were not specified in the National Emission Standards. Lignite from the Northern Great Plains Province is currently used in plants totaling about 1,200-MW capacity, and plants now under construction will increase the total to 2,000 MW by 1975. During the past 8 years but most actively since 1970, the Bureau of Mines has conducted field tests in which data on furnace exit and stack emissions from lignite-fired plants have been obtained. This report presents data on SO_2, SO_3, NO_x, and solid particulate emissions obtained in these field tests. Such data should be of value to those responsible for the design, procurement, or operation of air pollution control devices for lignite-burning plants.

Acknowledgments

The cooperation of Otter Tail Co., Basin Electric Coop., Central Power Electric Coop., and Minnkota Power Coop in permitting tests at their plants is gratefully acknowledged. Without their support the tests would not have been possible.

Powerplants Tested

Tests were conducted at three pulverized-coal-fired (pc-fired) plants, one cyclone-fired plant, and one spreader-stoker-fired plant, each burning lignite from a different mine in North Dakota. Information on each of the plants tested and the source of lignite burned are given in table 5. This report includes data from a total of 46 test days, 39 of which were with pc firing.

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[23] Environmental Protection Agency. Standards of Performance for New Stationary Sources. Federal Register, v. 36, No. 247, Dec. 23, 1971, pp. 24876-24895.

TABLE 5. - Powerplants included in emission studies

Plant name............	Hoot Lake	Leland Olds	Wm. J. Neal	Milton R. Young	F. P. Wood
Plant location........	Fergus Falls, Minn.	Stanton, N. Dak.	Voltaire, N. Dak.	Center, N. Dak...	Grand Forks, N. Dak.
Company..............	Otter Tail Power Co.	Basin Electric Power Coop.	Central Power Electric Coop.	Minnkota Power Coop.	Minnkota Power Coop.
Size of installations tested, MW.	50	215	20	235	12
Type of firing........	Pulverized coal.	Pulverized coal.	Pulverized coal.	Cyclone burner...	Spreader stoker.
Burner location.......	Tangential......	Front and rear walls.	Front wall......	Front wall.......	
Source of lignite.....	Beulah mine and Gascoyne mine of Knife River Coal Mining Co.	Glenharold mine of Consolidation Coal Co.	Velva mine of Consolidation Coal Co.	Center mine of Baukol-Noonan, Inc.	Larson mine of Baukol-Noonan, Inc.
Dates of tests........	1965, 1966, 1970, 1972	1970, 1971	1971	1970, 1971, 1972	1971, 1972
Type fly ash collector during tests.	Cyclone, pilot ESP,[1] Commercial ESP.[1]	Cyclone, pilot ESP.[1]	Cyclone.........	Cyclone..........	Cyclone.

[1] ESP = electrostatic precipitator.

Calculation Methods and Test Procedures

General

Principal emphasis was on the measurement of SO_2 emissions and their relationship to sulfur level in the coal burned. For such a study a complete ash and sulfur balance on the boilers tested would have been desirable. However, the difficulty and expense involved in collecting, weighting, and sampling large quantities of ash (15 to 20 tons per hour for a 200-MW unit) makes a complete sulfur balance quite impractical within the framework of a limited program. Data on SO_2 are based only on sampling of the coal feed and analysis of SO_2 in the flue gas.

Calculation Method

The SO_2 emission per million Btu input and the percentage of input sulfur in the lignite that is emitted as SO_2 can be calculated from the coal and the flue gas analyses without knowing the weights of coal burned or volume of flue gas produced. Given the carbon, sulfur, and heating value of the coal and the Orsat analysis of the flue gas, a balance on the carbon, assuming that all of the carbon in the coal appears as CO_2 and CO in the flue gas, yields the following equations:

$$\text{lb } SO_2/\text{MMBtu} = 5.33 \times \frac{C}{(CO_2 + CO)} \left(\frac{\text{ppm}}{\text{HHV}} \right), \qquad (1)$$

and

$$\text{S.E.} = 2.67 \times 10^{-2} \times \frac{C}{(CO_2 + CO)} \left(\frac{\text{ppm}}{S} \right), \qquad (2)$$

where ppm = parts per million of SO_2 in the flue gas, volume per volume on a dry basis;

CO_2 + CO = vol-pct in flue gas from Orsat analysis;

C = wt-pct carbon from the ultimate analysis of fuel;

S = wt-pct sulfur from ultimate analysis of fuel;

HHV = higher heating value of the coal, Btu/lb;

and S.E. = pct of input sulfur in the coal emitted as SO_2.

Data from ultimate analysis and heating value should be on the same basis--as received or moisture free.

A typical North Dakota lignite[24] has 65 pct carbon, 10 pct ash, and 10,800 Btu/lb on a dry basis. Using these values the pounds of SO_2 per million

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[24] Sondreal, E. A., W. R. Kube, and J. L. Elder. Analysis of the Northern Great Plains Province Lignites and Their Ash: A Study of Variability. BuMines RI 7158, 1968, 94 pp.

Btu can be calculated for various sulfur levels. These data along with corresponding parts per million SO_2 in the flue gas, based on 30 pct excess air, have been plotted in figure 54. For each sulfur level the percentage of SO_2

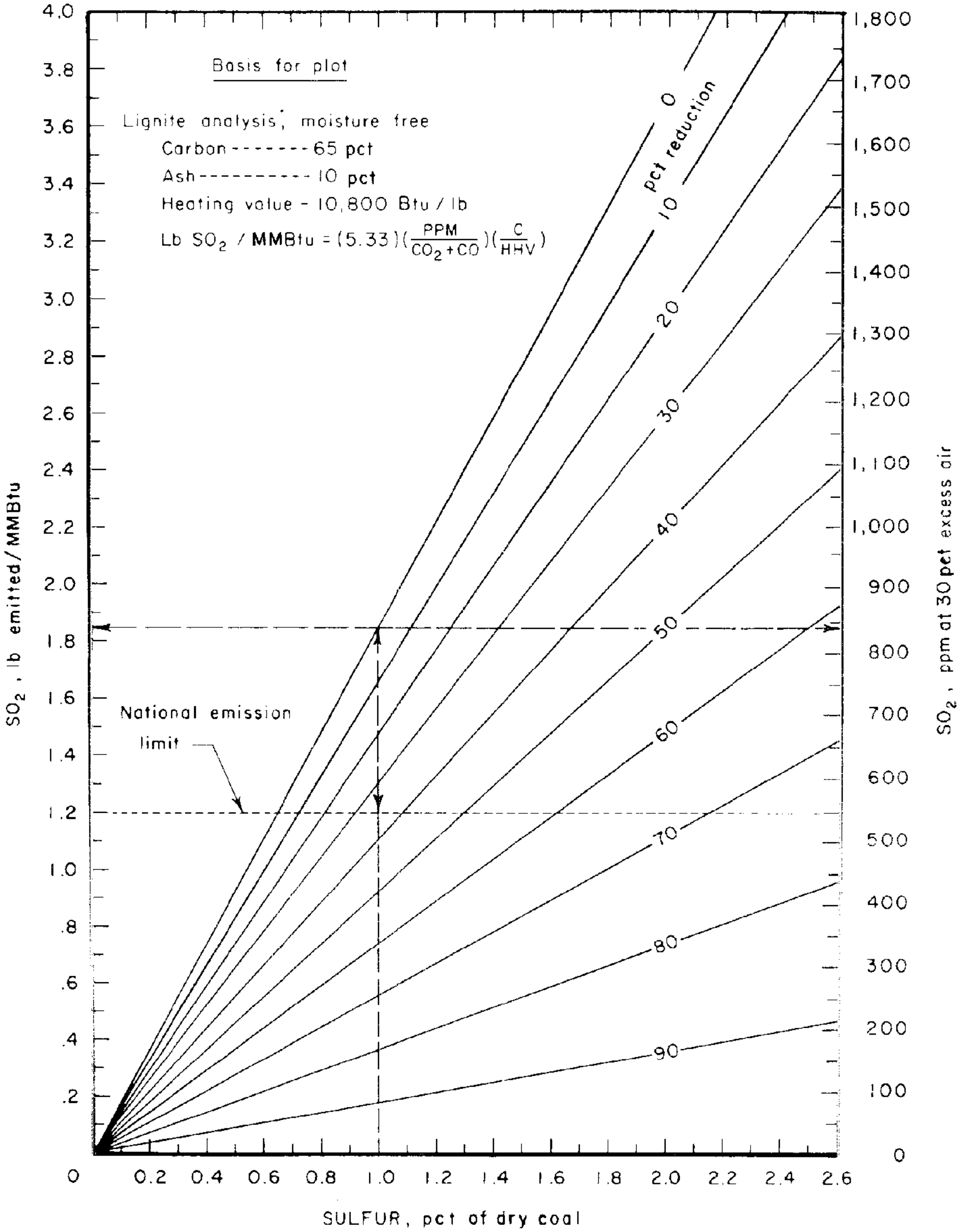

FIGURE 54. - Chart for determination of sulfur emissions and percentage removal required to meet national emission limit.

removal required to meet the 1.2 lb/MMBtu national emission limit is given by the lines of percent removal. For example (illustrated by the dashed lines on the chart), a lignite having 1-pct-sulfur content on a dry basis would have an equivalent SO_2 in the flue gas of 1.85 lb/MMBtu or 837 ppm. A 37-pct reduction in SO_2 would be required to meet the national emission limit.

The NO_x emission rate reported as NO_2 can be calculated by a similar method as for SO_2, using the equation

$$\text{lb } NO_x/\text{MMBtu} = 3.83 \times \frac{C}{(CO_2 + CO)} \left(\frac{\text{ppm}}{\text{HHV}} \right), \qquad (3)$$

where ppm = parts per million of NO_x in the flue gas, volume per volume on a dry basis.

Coal Sampling and Analysis

In sampling the lignite feed to the boilers, the objective was to collect a representative sample that would give an accurate value of the sulfur input during the SO_2 sampling period. The coal handling equipment and the facilities for sampling varied from plant to plant. The usual sampling procedure was to collect grab samples from each coal feeder at 1/2-hour intervals and combine these samples for determination of average sulfur content.

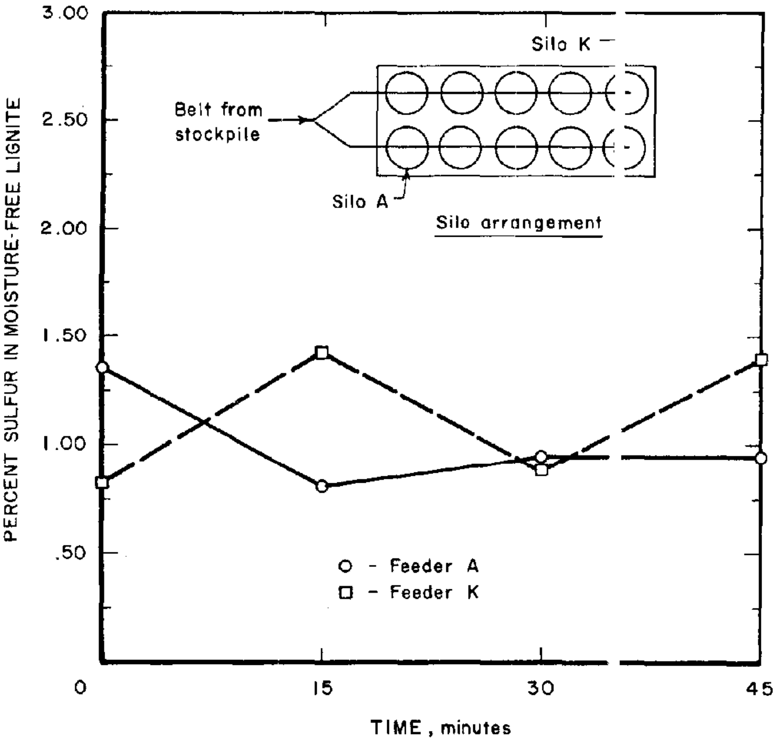

FIGURE 55. - Variation in sulfur content of lignite with time and point of sampling.

Special tests were run at both the Leland Olds and Milton R. Young plants to determine the variability of sulfur content in the lignite. At the Leland Olds plant, grab samples were collected at four 15-min intervals simultaneously from the two storage silos that were physically farthest apart. Each sample was split into four aliquots, two of which were analyzed by one laboratory and two by another. The results of the sulfur determinations at the two laboratories are shown in table 6, and the average of the four determinations for each sample is plotted in figure 55. The relative differences in sulfur contents of lignite between feeders at any given time or from a given feeder in a 15-min interval approached

100 pct. Within- and between-laboratory results were usually within the ASTM tolerances of 0.05 and 0.10 percentage points, respectively. Similar results were found for data from the Milton R. Young plant.

TABLE 6. - Variation in sulfur content of lignite at the Leland Olds plant

Time, min	Silo	Sulfur concentration, pct moisture free				
		Lab. G		Lab. P		Average
		Sample 1	Sample 2	Sample 3	Sample 4	
0...........	A	1.49	1.34	1.33	1.41	1.39
	K	.76	.82	.93	.82	.83
15...........	A	.79	.77	.82	.85	.81
	K	1.42	1.25	1.55	1.56	1.45
30...........	A	.96	.94	.96	.96	.95
	K	.91	.91	.95	.94	.93
45...........	A	.90	.94	.97	1.01	.95
	K	1.37	1.37	1.46	1.46	1.42

The data indicate the difficulty in obtaining a representative lignite sample for sulfur balances. Sampling is a factor that is too often overlooked in powerplant testing, and some of the variation in data for the SO_2 tests could be attributed to nonrepresentative coal samples despite the emphasis placed on coal sampling.

SO_3 and SO_2 Sampling Procedure

Sulfur trioxide and SO_2 determinations were made by single point sampling in the flue gas ducts in each of the boilers tested. Usually the samples were collected in the duct between the boiler exit and the air heater, but some samples were collected after the air heater. In one case, samples were collected from the stack.

The apparatus used for SO_3 and SO_2 determination was similar to that described by Lisle and Sensenbaugh.[25] The flue gas is drawn through a condenser maintained at 140° to 194° F where the SO_3 is selectively condensed and collected. The flue gas then passes through a bubbler containing a 3-pct H_2O_2 solution. The resulting sulfuric acid is titrated to a methyl purple endpoint using a standard NaOH solution.

Orsat analyses were usually taken before and after each 30-min test for SO_2. All of the SO_2, SO_3, and NO_x values have been normalized to a 30-pct excess air basis for ease of comparison.

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[25] Lisle, E. S., and J. D. Sensenbaugh. The Determination of Sulfur Trioxide and Acid Dew Point in Flue Gas. Combustion, v. 36, No. 7, January 1965, pp. 12-15.

NO_x Sampling Procedure

The NO_x sampling was usually done at or near the SO_2 sampling location. The procedure used was the Phenoldisulfonic acid method, specified as "Method 7" by the EPA in the Federal Register.[26] Because the test is more difficult to perform than the SO_2 test, fewer determinations were made.

Particulate Sampling and Study

During most of the tests, samples of fly ash were collected for determination of chemical analysis, particle size distribution, and, in later tests, for electrical resistivity. The fly ash samples were obtained from mechanical dust collector hoppers, pilot or commercial electrostatic precipitator hoppers, or from aspirated flue gas samples. Particle size analysis was done by the Bahco method and laboratory electrical resistivity measurements were by a method similar to that given in ASME PTC-28.[27] In the later tests, two "in situ" methods were used for resistivity studies. Details of these test procedures have been published.[28]

Results and Discussion

Summary of SO_3 Results

A total of 51 determinations of SO_3 content were made at the various lignite-burning plants. In most of these tests, a trace of SO_3 was detected but in no case did it exceed 1 ppm, even with SO_2 levels as high as 1,395 ppm. Such low SO_3 concentrations differ considerably from those published for eastern bituminous coals where it is normal for the SO_3 content to be 1 to 2 pct of the SO_2 content. The lack of free SO_3 in the flue gas from lignite-fired boilers is believed to be caused by the presence of reactive alkali in the fly ash. Any SO_3 produced rapidly reacts with the alkali to produce sulfates.

Summary of SO_2 Results

Data on the coal analysis, ash analysis, and SO_2 in the flue gas for the 46 test days are given in table 7. Included are data collected during 39 test days with pc firing, 5 days with cyclone firing, and 2 days with spreader-stoker firing. Figure 56 shows a frequency distribution of pounds of SO_2/MMBtu input for the test days, which include all variations in coal analysis, ash analysis, and firing method. For 17 of the test days, the SO_2 emissions levels were between 1.0 and 1.5 lb/MMBtu and for another 16 they were between 1.5 and 2.0 lb/MMBtu. For 13 test days, the SO_2 emissions were below the 1.2 lb/MMBtu national limit. There were only 7 test days in which the SO_2 was above 2 lb/MMBtu, at which level a 40-pct reduction would be required to meet the 1.2 lb/MMBtu level.

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[26] Work cited in footnote 23.

[27] American Society of Mechanical Engineers. Determining the Properties of Fine Particulate Matter. ASME PTC-28, New York, 1965, pp. 15-17.

[28] Seele, S. J., P. H. Tufte, and G. H. Gronhovd. A Study of the Electrical Resistivity of Fly Ashes From Low-Sulfur Western Coals Using Various Methods. Pres. at Air Pollution Control Assoc. Meeting, Miami Beach, Fla., June 18-20, 1972, APCA Preprint 72-107, 1972, 31 pp.

TABLE 7. - Summary data for SO_2 emissions tests at lignite-fired plants

Test date	Boiler load, pct of rated capacity	Lignite analysis, pct					Coal ash analysis, pct				SO_2 in flue gas			
		Moisture	Moisture-free basis								Number of tests	Average SO_2 ppm at 30 pct excess air	Lb SO_2 per MMBtu	Pct of input sulfur emitted
			Ash	Sulfur	Carbon	Heating value, Btu/lb	SiO_2	Al_2O_3	CaO	Na_2O				
HOOT LAKE PLANT--BEULAH LIGNITE--PC FIRED														
7/25/65	80	35.6	10.3	1.09	65.4	10,830	14.2	9.8	20.1	8.2	4	420	0.93	48
7/28/65	82	35.9	10.1	1.03	65.4	10,840	14.4	10.2	21.1	9.0	4	450	1.00	52
8/16/65	82	36.7	9.9	1.15	65.4	10,900	17.0	11.4	26.0	1.7	2	800	1.76	84
8/18/65	84	36.6	10.2	1.10	65.8	10,970	19.7	12.0	26.3	1.6	2	830	1.83	92
9/7/65	102	36.1	11.9	1.36	NA	10,570	17.6	11.0	23.0	4.8	2	820	1.87	73
5/2/66	84	36.1	11.8	1.52	64.4	10,600	20.8	10.5	23.3	5.3	2	740	1.66	58
5/3/66	84	36.3	10.8	1.31	64.8	10,720	19.7	10.9	23.1	5.6	4	700	1.55	63
5/4/66	84	36.3	10.8	1.25	65.0	10,710	19.5	10.9	21.6	5.8	4	710	1.58	68
5/5/66	84	36.1	11.8	1.54	64.6	10,740	17.4	9.2	22.0	5.4	4	840	1.86	65
5/6/66	84	35.8	11.8	1.33	65.1	10,690	19.4	10.7	22.6	5.5	4	840	1.88	76
7/9/70	104	34.4	10.4	1.32	65.2	10,910	16.7	10.9	25.2	7.8	2	610	1.34	59
7/10/70	104	34.9	9.8	1.10	65.5	10,910	16.5	10.6	24.9	3.8	3	630	1.37	69
8/11/70	112	34.3	9.8	.92	64.1	10,670	14.6	10.6	21.2	7.5	1	515	1.14	66
8/12/70	112	34.5	9.8	.97	64.1	10,670	16.9	9.9	21.5	3.8	8	585	1.29	71
8/13/70	110	34.0	9.8	1.06	64.1	10,670	14.4	10.9	24.2	5.1	3	575	1.27	64
9/12/72	100	35.4	10.1	1.11	65.2	10,780	17.9	12.0	26.5	1.1	5	680	1.51	74
9/13/72	100	35.8	9.9	1.21	64.6	10,740	17.3	12.0	26.2	1.0	13	770	1.70	75
9/14/72	100	35.8	10.0	1.14	64.8	10,780	18.7	11.6	25.1	.9	13	820	1.81	86
9/15/72	100	35.2	9.5	1.03	65.4	10,800	18.9	12.1	28.2	.9	10	780	1.74	91
9/18/72	84	35.7	10.9	1.14	63.9	10,530	20.6	11.5	20.3	6.1	12	600	1.34	61
9/19/72	84	35.7	10.3	1.03	64.5	10,680	19.7	11.2	20.0	6.1	22	570	1.27	66
9/20/72	84	35.2	10.4	1.12	64.4	10,660	20.1	11.5	21.2	6.1	9	550	1.22	59
HOOT LAKE PLANT--GASCOYNE LIGNITE--PC FIRED														
7/27/70	96	40.1	14.8	1.52	61.3	10,300	32.4	11.6	15.9	3.1	1	1,120	2.45	83
7/28/70	106	41.1	14.8	1.33	61.3	10,300	28.1	12.7	18.9	5.8	4	1,170	2.56	99
7/29/70	108	41.2	14.8	1.38	61.3	10,300	37.1	13.1	16.4	3.0	6	1,360	2.97	100
7/30/70	110	41.8	14.8	1.68	61.3	10,300	32.0	13.1	17.0	3.5	3	1,325	2.90	89
7/31/70	108	41.7	14.8	1.68	61.3	10,300	34.3	13.8	16.7	2.1	3	1,395	3.05	94
8/4/70	110	40.0	14.8	1.71	61.3	10,300	32.3	10.3	15.6	2.0	3	1,300	2.84	86
LELAND OLDS PLANT--GLENHAROLD LIGNITE--PC FIRED														
8/25/70	93	36.7	10.4	0.63	63.1	10,580	30.7	11.0	20.2	8.8	8	425	0.93	78
8/26/70	93	37.8	11.5	.65	62.9	10,570	31.4	11.8	17.0	8.0	4	470	1.03	84
8/27/70	84	35.2	11.1	.61	62.7	10,560	37.5	13.6	16.2	7.7	16	470	1.02	89
9/9/70	56	36.8	11.7	.85	62.5	10,580	35.1	13.6	17.7	6.0	16	655	1.42	89
9/10/70	55	36.4	11.7	.84	62.1	10,470	35.7	13.8	17.2	7.0	4	590	1.28	80
10/28/70	92	36.8	12.7	.75	62.3	10,425	35.3	13.4	18.7	6.2	6	520	1.14	79
10/29/70	100	38.1	12.1	.74	63.6	10,700	33.0	12.2	19.2	7.8	12	510	1.11	81
4/14/71	100	34.3	13.2	1.09	62.9	10,550	33.8	11.2	17.0	5.1	5	660	1.44	70
WM. J. NEAL PLANT--VELVA LIGNITE--PC FIRED														
7/27/71	100	NA	9.1	0.57	64.7	10,920	38.1	13.0	22.6	0.9	7	355	0.77	74
7/28/71	100	NA	9.1	.70	64.7	10,920	46.0	13.6	16.4	.8	9	525	1.14	89
7/29/71	100	NA	9.1	.66	64.7	10,920	34.1	14.5	23.4	.9	12	455	.99	82
MILTON R. YOUNG PLANT--BAUKOL-NOONAN (CENTER) LIGNITE--CYCLONE FIRED														
11/24/70	106	36.5	11.9	0.90	63.9	10,490	30.9	12.7	19.4	0.7	6	765	1.71	100
4/16/71	92	37.3	12.5	.93	62.9	10,580	29.1	13.3	21.8	.8	10	735	1.61	91
9/6/72	106	36.5	20.6	1.11	55.9	9,270	45.8	14.0	11.9	.4	5	930	2.02	86
9/7/72	106	37.1	19.4	.97	56.7	9,270	44.8	14.3	13.8	.4	17	785	1.76	82
9/8/72	106	38.2	13.8	1.10	59.2	9,550	29.9	10.9	19.4	.7	17	765	1.68	73
FRANKLIN P. WOOD PLANT--BAUKOL-NOON (LARSON) LIGNITE--SPREADER STOKER FIRED														
11/17/71	100	35.0	10.7	0.59	65.7	10,940	32.5	14.1	15.6	10.9	8	130	0.29	27
12/15/72	100	33.0	12.5	.54	64.7	10,880	32.8	14.4	16.8	8.6	18	194	.42	43

NA Not available.

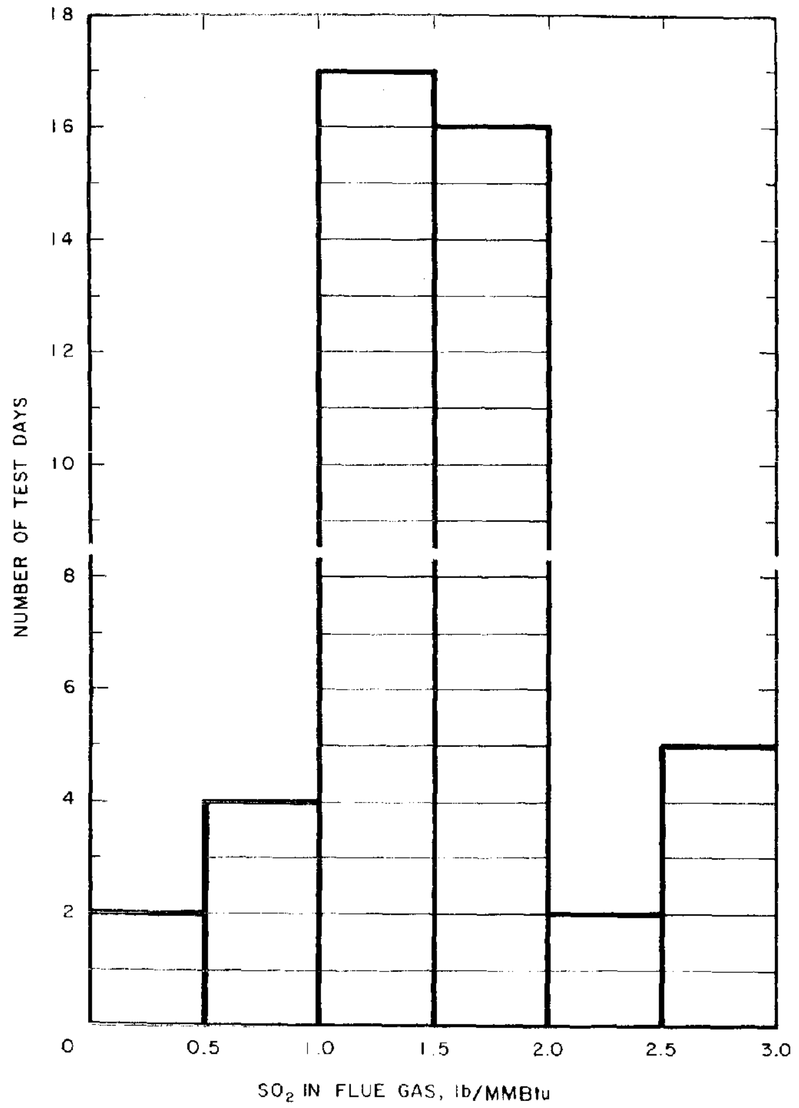

FIGURE 56. - Frequency distribution of SO_2 emission level.

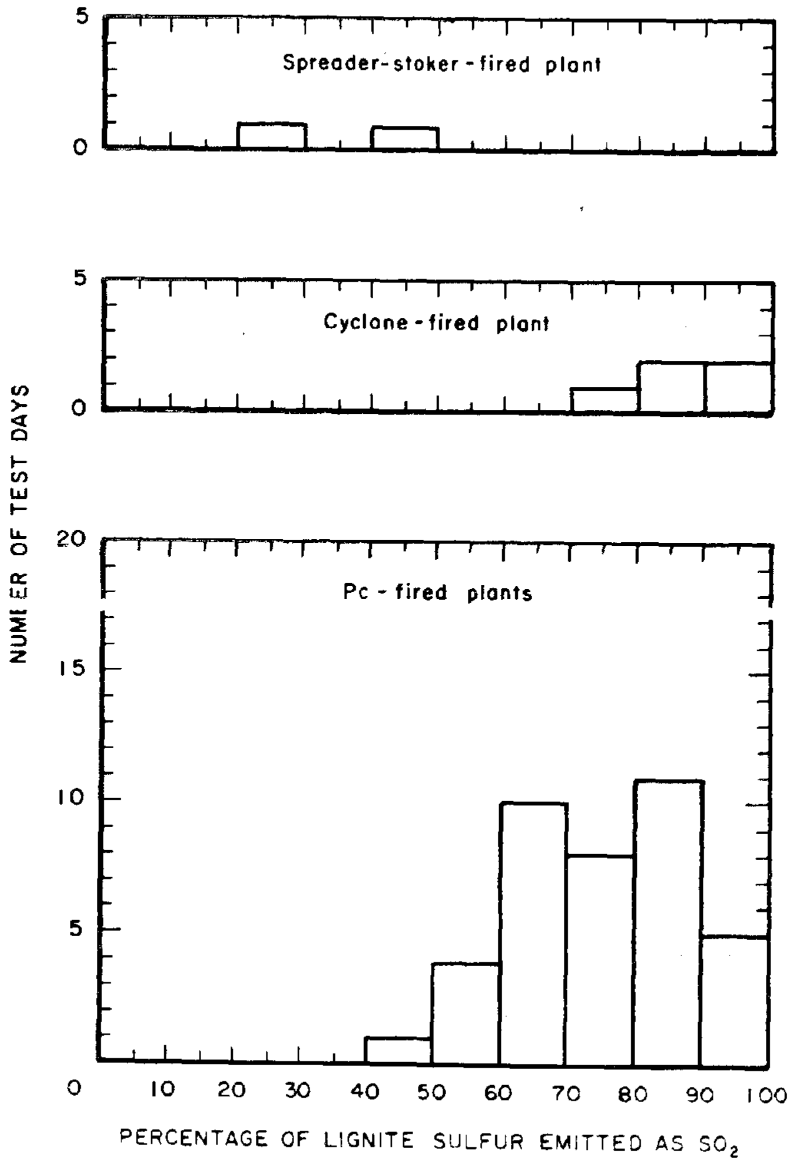

FIGURE 57. - Percentage of lignite sulfur emitted as SO_2.

Figure 57 shows a frequency distribution of the test days that the percentage of sulfur in the lignite emitted as SO_2 (referred to as S.E.) was in a given range. Results are shown separately for each method of firing. In 29 of the 39 test days with pc firing, S.E. was in the range of 60 to 90 pct. The minimum value was 48 pct, and the maximum was 100 pct. Some of the reasons for these variations will be discussed later.

With cyclone firing, S.E. was in the 80- to 100-pct range for 4 days and in the 70- to 80-pct range for 1 day. One would expect less retention of the sulfur with the ash in a cyclone furnace because of the smaller percentage of ash that is carried through the boiler as fly ash. That portion of the ash that is melted into slag contains very little sulfur.

Two test days were on a spreader-stoker-fired plant. In one test, S.E. was 29 pct, and in the other, 42 pct. The lignites burned during these tests were very low in sulfur, 0.59 and 0.56 pct on a dry basis, and the ash had high sodium oxide contents of 10.9 and 8.6 pct. These factors combined to produce the very low emission rates, which cannot be assumed to be typical for spreader-stoker-firing. The grate ash from a spreader-stoker unit should not retain much sulfur and the percentage of ash carried over is less than for pc firing, so less sulfur retention would be expected.

Prediction of SO_2 Emissions

The retention of sulfur in lignite fly ash is due to the presence of alkali constituents such as calcium, magnesium, sodium, and potassium in reactive form. Calcium and sodium sulfates have been identified in significant concentrations in lignite fly ash by X-ray diffraction. Water-solubility stoichiometric data suggests all soluble sodium and potassium in the fly ashes are present as sulfates, but only a portion of the soluble calcium is as sulfate. The water solubility of magnesium in the fly ash is highly pH sensitive, and sulfate in solution does not increase as magnesium dissolution increases with lowered pH, suggesting there is little $MgSO_4$ in a typical lignite fly ash. Since potassium is generally present in lignites in very small amounts, the alkalis most responsible for SO_2 retention in lignite fly ash are calcium and sodium.

Not all calcium and sodium is available for reaction with SO_2. Electron microprobe analyses of individual fly ash particles have indicated a significant CaO and Na_2O content in most glassy silicate and aluminum silicate particles appearing in lignite fly ash. These particles result from intimate contact of very fine silica and clay with organically bound calcium and sodium in burning coal particles. This type of particle accounts for the significant fraction of calcium and sodium, which remains insoluble in water, even in solutions having low pH. It is believed that the alkali appearing in these insoluble glassy particles is not reactive towards SO_2. Thus silica and clay inherent in the ash could be expected to reduce the fraction of the ash that is active in SO_2 pickup.

Field test data from pc-fired units were examined using statistical regression to determine whether correlations existed between ash composition, sulfur content, and the percent sulfur emitted as SO_2. The bivariate relationships between individual constituents sodium, calcium, silicon, aluminum, total ash, and sulfur contents in the lignite, and percent sulfur emitted as SO_2 are shown in figure 58. The most clearly discernible trends are the increased percent emitted with increased concentrations of SiO_2 and Al_2O_3 in the coal on a dry basis. Increased sodium and calcium decrease the percent sulfur emitted; however, the correlations are very poor, especially for calcium. No correlation between initial sulfur content and percent emitted is evident. A slight increase in percent of sulfur emitted with increased ash content is indicated, perhaps relatable to increased silica and clay contents in high-ash coals.

Multiple linear regression was employed with more success using the basic model of the form

$$\text{S.E.} = C_1 \ (\text{Element A})^W \times (\text{Element B})^X + C_2 \ (\text{Element C})^Y \times (\text{Element D})^Z, \qquad (4)$$

where Elements A, B, C, D, --- were SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, and S tested in a multiplicity of combinations. Components were expressed as percent of moisture-free lignite. Exponents W, X, Y. Z, --- ranged from -2 to +2 and were tested in a multiplicity of combinations.

The most useful correlation developed for the pc-fired test data combined good predictability and simplicity:

$$\text{S.E.} = -12.7 \frac{CaO}{Al_2O_3} - 48.1 \frac{Na_2O}{SiO_2} + 110.1,$$

and

$$\text{and } R^2 = 0.711. \qquad (5)$$

A plot of actual versus predicted percent emission is shown in figure 59.

The R^2 indicates 71 pct of the variance in the data is explained by the correlation, a significant improvement over individual predictors. An R^2 of 100 pct would indicate perfect agreement between the data and the equation.

The effect of each variable on S.E. can be illustrated by considering the change in S.E. with a 1-pct increase in each ash component on a dry coal basis. This would be roughly equivalent to a 10-pct increase on a percent-of-ash basis. A 1-pct increase in Na_2O (dry coal basis) or a 10-pct increase in the ash would cause a decrease of 20 pct in S.E. A 1-pct increase in CaO would cause a 10-pct decrease in S.E., a 1-pct increase in SiO_2 would cause a 6-pct increase in S.E., and a 1-pct increase in Al_2O_3 would cause a 10-pct increase in S.E. It should be noted that in other regression relations developed, the effect of sodium relative to calcium may be increased to a 3:1 ratio. The generally greater effect of sodium reflects a higher reactivity toward SO_2.

The prediction formula confirms quantitatively that alkalis are responsible for SO_2 removal, and silicates and clays would retard removals by tying up alkalis. The ratio of CaO to Al_2O_3 suggests that CaO reacts preferentially

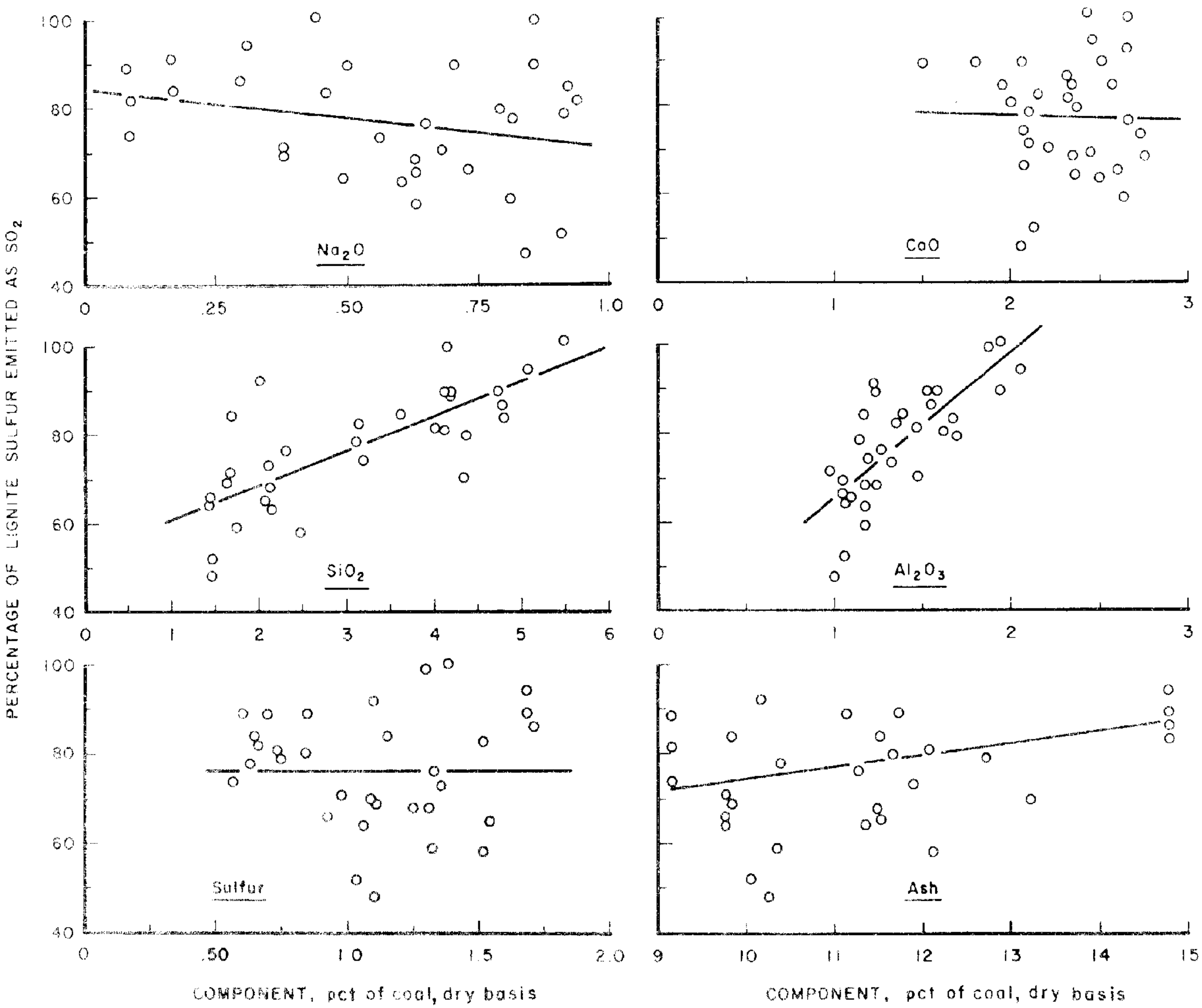

FIGURE 58. - Correlation of lignite and ash analysis with percentage of sulfur emitted for pc-fired units.

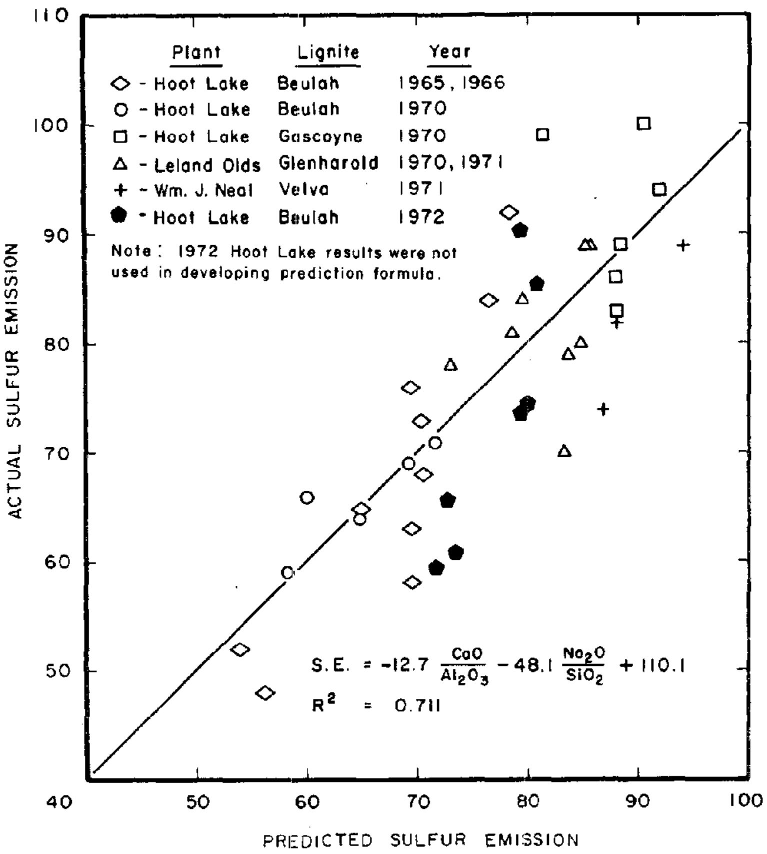

FIGURE 59. - Predicted versus actual sulfur emission (S.E.) for pc-fired plants.

with clay, but Na_2O would react more readily with silica as mechanism for decreasing alkali reactivity.

The fact that the model suggests that S.E. is not a function of sulfur level deserves additional discussion. Sulfur was considered in all possible combinations in the model, but did not increase R^2 more than a trivial amount. In the equation for S.E., coal sulfur is a divisor $\left(\text{that is,}\right.$

$$\left. S.E. \approx \frac{\text{ppm } SO_2 \text{ emitted}}{\text{S in coal}} \right).$$

If both sides of equation 5 are multiplied by percent S in dry coal, the apparent dilemma is resolved. Then S.E. becomes absolute sulfur emitted, expressed as a function of the product of the alkali clay silica ratio with sulfur. It can then be seen that the model suggests sulfur retention is relatable to a first-order alkali sulfur reaction scheme.

In figure 59, some data not included in the regression are plotted. These are data for 1972 Hoot Lake, Beulah lignite tests. All other points on the plot were used to develop the relationship. The correlation explains data from the Hoot Lake tests as well as those originally included in the regression.

Effect of Sodium on SO_2 Emissions

Sodium has been shown to have the greatest effect on sulfur emitted from lignite-fired boilers. In tests at the Hoot Lake plant in 1972, low- and high-sodium lignites from one mine were tested for succeeding weeks. The sodium content was the only substantial variable in the tests (see analysis in table 7), and any variation can be attributed to changed sodium concentration. The results from 3 test days on each coal is shown in figure 60. The parts per million SO_2 decreased from about 800 ppm to about 590 ppm when the sodium was increased from 0.9 to 6.1 pct. A disadvantage of the high sodium content is, of course, an increased ash fouling rate.

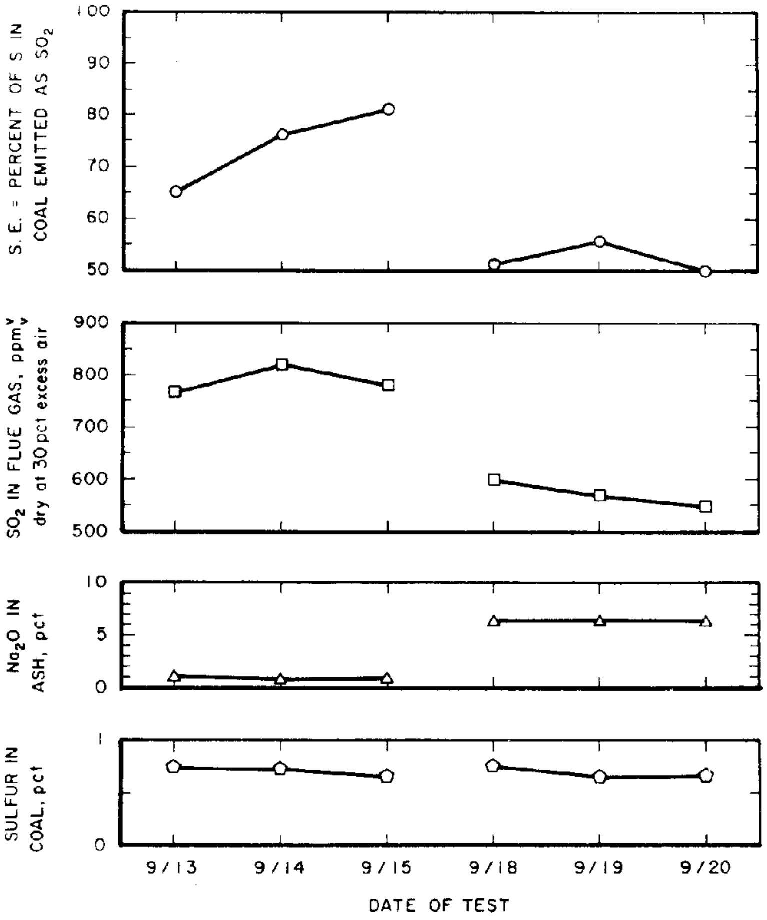

FIGURE 60. - Effect of sodium content in lignite on SO_2 emissions, Hoot Lake tests, 1972.

Effects of Boiler Load on SO_2 Emissions

Sulfur balance studies conducted on plants burning German brown coals have shown that the percent of sulfur retained in the ash is greatly influenced by boiler load and gas residence time.[29] Most tests reported in this study were at 85 to 110 pct of rated load as shown in table 7. The one exception is at the Leland Olds plant where load was reduced to 55 pct for 2 test days. The results show no significant change in SO_2 emission when compared with the higher load tests.

Sulfur Retention in Various Ash Fractions

During most of the field tests, samples of ash were collected from various locations in the boiler for chemical and physical analyses. Results of chemical analysis of selected ashes from the various boilers are shown in table 8. Fly ash samples from both the Leland Olds and Hoot Lake plants were collected during special tests in which a pilot electrostatic precipitator was operating on a side stream of flue gas. In both cases, the electrostatic precipitator dust is considerably enriched in SO_3 as compared with the cyclone dust collector sample. In tests conducted at the Hoot Lake plant in 1965-66, four sodium levels in the lignite were studied, and fly ash samples aspirated from the flue gas ahead of the air heater in each case. The SO_3 contents of aspirated ash ranged from 6.8 pct with the lowest sodium lignite to 28.3 pct with the highest sodium lignite. During the high-sodium tests, considerable ash was being retained in the furnace because of boiler fouling.

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[29] Speich, P. Schwefelbilanzuntersuchungen an Braunkohlenkesselanlagen. (Investigations of Sulfur Balance at Lignite-Fired Boiler Plants.) Braunkohle, v. 17, No. 9, September 1965, pp. 364-371.

TABLE 8. - Partial analysis of selected ash samples from field tests of lignite-fired boilers

	Component in ash, pct			
	CaO	MgO	Na_2O	SO_3
PC-FIRED TESTS				
Hoot Lake plant, 8/12/70:				
Mechanical dust collector.................	28.8	9.1	4.2	4.5
Pilot electrostatic precipitator..........	26.4	8.9	5.3	8.7
Leland Olds plant, 8/28/70:				
Mechanical dust collector.................	19.9	6.0	6.9	2.3
Pilot electrostatic precipitator..........	20.1	5.8	7.9	6.0
Hoot Lake plant, 1965-1966: Samples aspirated from boiler exist dust:				
Test 1....................................	32.9	10.3	2.3	6.8
Test 2....................................	28.5	8.6	7.2	11.6
Test 3....................................	26.2	9.1	9.4	14.1
Test 4....................................	20.1	9.3	16.3	28.3
CYCLONE-FIRED PLANT				
Milton R. Young plant, 1970:				
Mechanical dust collector.................	32.4	8.8	1.4	3.9
Aspirated from gas to dust collector......	31.5	8.4	1.4	6.7
Bottom slag...............................	31.8	9.5	.3	.2
SPREADER-STOKER-FIRED PLANT				
Franklin P. Wood plant, 1971:				
Mechanical dust collector.................	23.5	6.4	11.8	2.9
Aspirated from dust collector exit gas....	3.8	.9	30.5	38.0
Grate ash.................................	19.3	4.8	8.8	1.0

The results for the cyclone-fired plant again show that the aspirated fly ash from the furnace exit contained considerably more SO_3 than the fly ash caught in the mechanical dust collector. The bottom slag had only 0.2 pct SO_3.

Tests conducted on a spreader-stoker-fired boiler during operation on high-sodium lignite showed that the dust collector ash had 2.9 pct SO_3 but that the aspirated dust at the dust collector outlet had 38.0 pct SO_3 and 30.5 pct Na_2O. The grate ash had 1.0 pct SO_3.

To summarize these data, the results show wide variation in the amount of sulfur retained with the ash, depending upon alkali content and temperature history of the ash. The fine fly ash fractions are most enriched in SO_3; however, without ash balance data one cannot determine the quantity of input sulfur retained in the various fractions. Bottom slag from a cyclone and grate ash from a spreader-stoker retain little sulfur.

Summary of NO_x Results

Figure 61 shows the results of NO_x determinations at the various plants. The daily averages shown have been adjusted to 30 pct excess air for comparative purposes. The levels for the pc-fired plants ranged from about 300 to 520 ppm. For the cyclone-fired plant, the levels ranged from 570 to 690 ppm, significantly higher, as would be expected from the higher combustion temperatures employed. The value of 230 ppm for the spreader-stoker plant is surprisingly low. These values can be compared with a limit of 0.7 lb/MMBtu (about 550 ppm) for bituminous coal-fired boilers as specified by the national emission standards. At the present time, lignite-fired boilers are specifically exempted from an NO_x limit in the national emissions standards.

Fly Ash Size Distribution

Two of the most important factors affecting design of fly ash control equipment are the fly ash loading and particle size distribution in the flue gas exiting from the boiler. Determination of dust loading was not a major part of these studies, but size distribution was determined on fly ash samples aspirated from boiler exit ducts during some of the tests. Figure 62 shows the Bahco size analysis of fly ash taken from the boiler exit flue gas at a pc-fired, cyclone-fired, and spreader-stoker-fired boiler. The spreader-stoker fly ash was coarsest, having a mass median diameter of 44 µm with

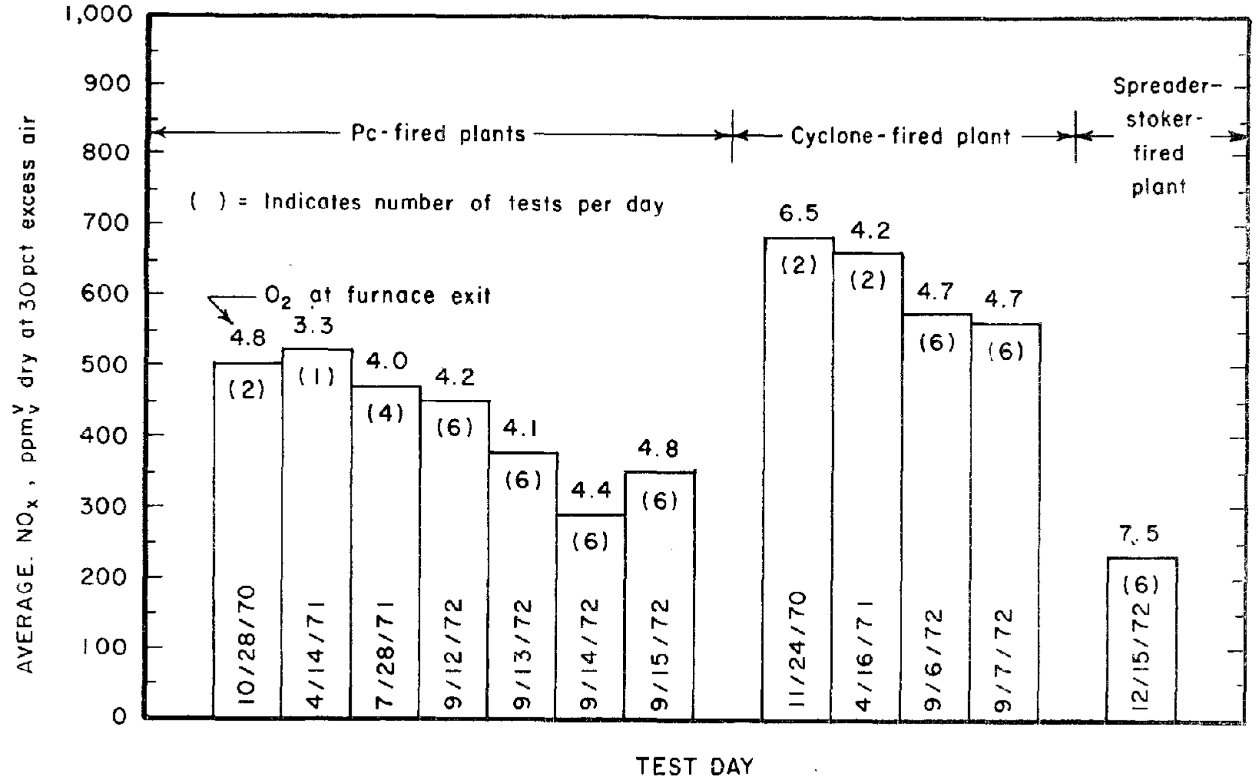


FIGURE 61. - NO_x emissions from various lignite-fired plants. (Boiler loads ranged from 92 to 106 pct of rated capacity.)

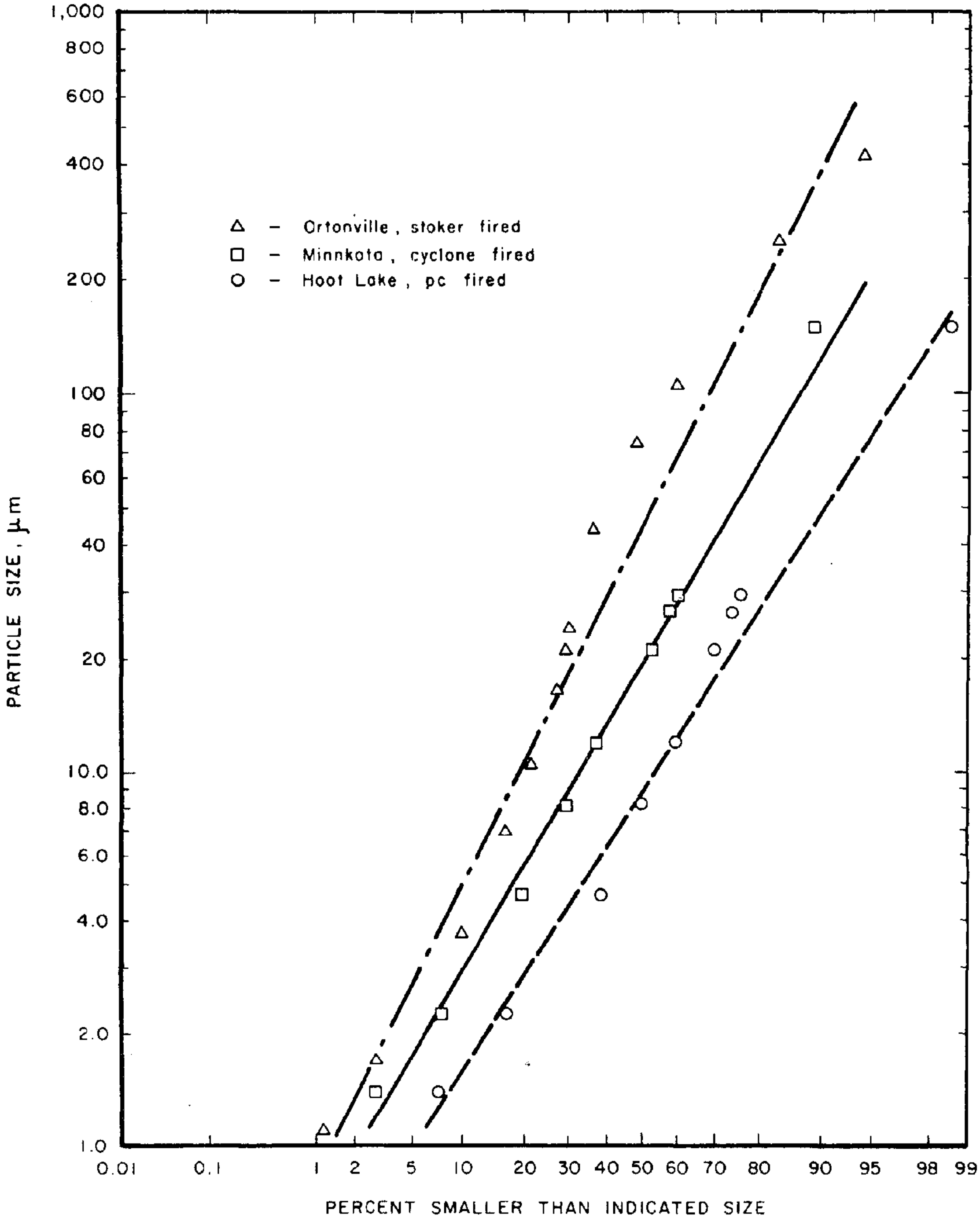
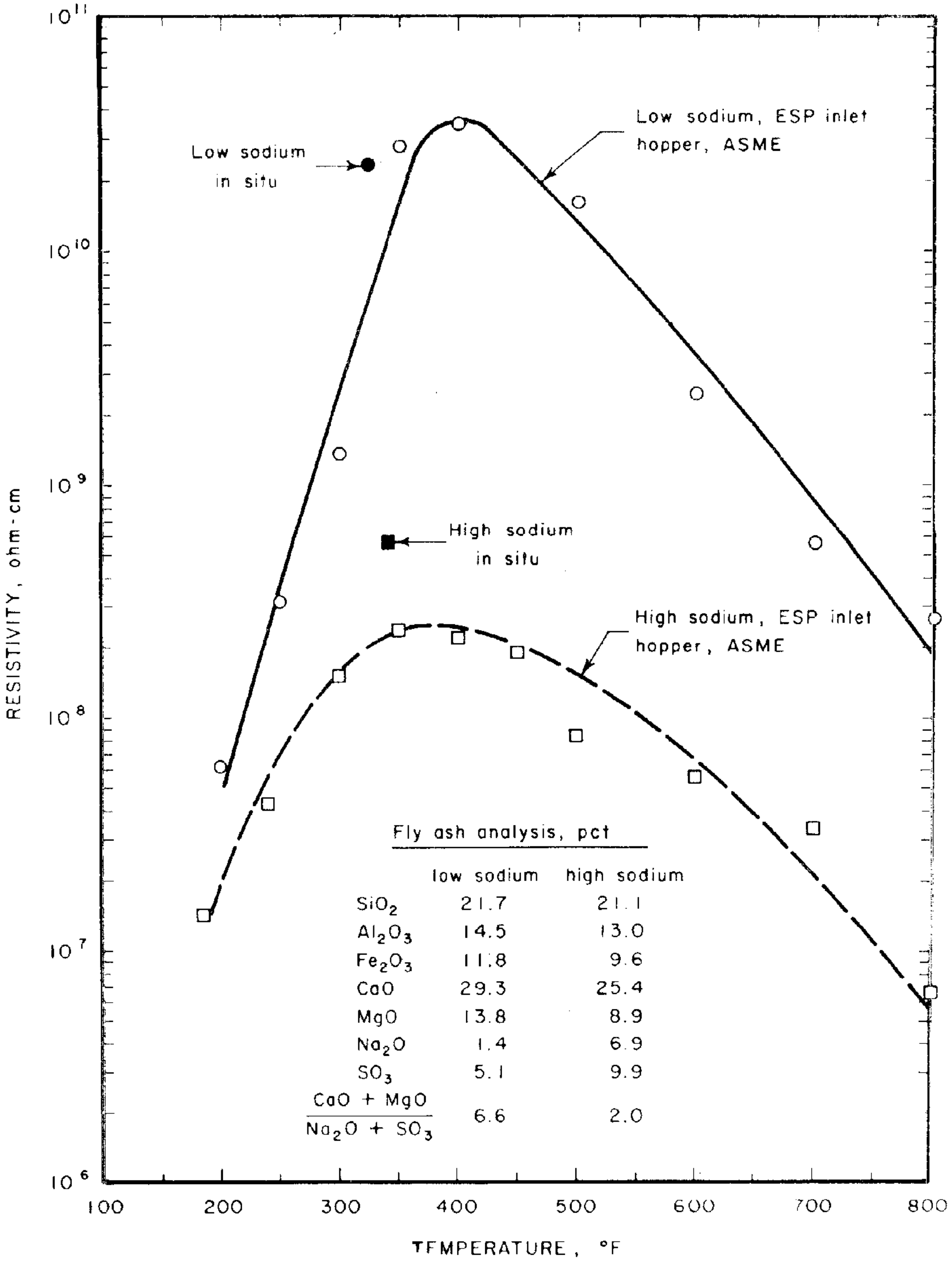


FIGURE 62. - Size analysis (by Bahco method) of fly ash samples aspirated from boiler exit duct.

18 pct below 10 μm. It has been stated in the German literature on brown coal firing that cyclone firing produced a finer fly ash than did pc firing. This was not confirmed by the tests at Milton R. Young and Hoot Lake plants in 1972.



Fly ash analysis, pct

	low sodium	high sodium
SiO_2	21.7	21.1
Al_2O_3	14.5	13.0
Fe_2O_3	11.8	9.6
CaO	29.3	25.4
MgO	13.8	8.9
Na_2O	1.4	6.9
SO_3	5.1	9.9
$\frac{CaO + MgO}{Na_2O + SO_3}$	6.6	2.0

FIGURE 63. - Effect of sodium content on electrical resistivity of lignite fly ash.

The mass median size for the cyclone-fired boiler was 18.5 μm with 33 pct below 10 μm and for the pc-fired boiler the mass median size was 7.8 μm with 54 pct below 10 μm. It should be noted that the cyclone-fired boiler was burning a very low sodium coal during these tests, and it is possible that increased volatilization might increase the fine fraction if high alkali lignites were used.

Electrical Resistivity of Fly Ash

An important factor affecting the electrostatic precipitation of fly ash is the electrical resistivity of the ash. A series of laboratory resistivity tests was run on the fly ash from a number of low-sulfur western coals with the basic data presented previously.[30] A computer study indicated that the ratio $(CaO + MgO)/(Na_2O + SO_3)$ in the fly ash was the best indicator of resistivity for those samples, with resistivity varying directly as the ratio.

In more recent work, the emphasis has been on in situ field resistivity determinations. The effect of sodium in the fly ash is additionally demonstrated by data from the 1972 Hoot Lake tests, shown in figure 63. The low-sodium ash showed a significantly higher (100 X at 350° F) resistivity than did the high-sodium ash for both the laboratory and in situ results. The results also confirm the applicability of the ratio described previously, as the higher resistivity fly ash shows the higher value for $(CaO + MgO)/(Na_2O + SO_3)$. The values determined are within the range for satisfactory precipitator operation despite the low sulfur content of the lignite burned. This is confirmed by the satisfactory performance of the electrostatic precipitator (ESP) units on the Hoot Lake plant since their startup in the spring of 1972.

Summary and Conclusions

For the past few years, the Bureau of Mines has collected data on stack emissions from various lignite-fired powerplants. Included were tests on pc-fired, cyclone-fired, and spreader-stoker-fired boilers operating on lignites. Most emphasis was on the study of SO_2 emissions and how they relate to input sulfur level, firing method, ash analysis, and other factors. Limited data was collected on NO_x emissions from the various plants. Chemical and physical properties of fly ash as a function of firing method were determined, and electrical resistivity studies of the fly ashes were made. A method is given for calculating SO_2 and NO_x emission levels and percent of input sulfur emitted as SO_2 without knowing the weights of coal burned or flue gas produced.

The principal conclusions are the following:

1. The 1.2-lb/MMBtu SO_2 emission limit was exceeded in 33 of the 46 test days. However, with a 40-pct reduction most of the tests would have been under the limit.

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[30] Work cited in footnote 28.

2. Significant amounts of sulfur were retained by the ash in most of the tests. The percentage sulfur retained ranged from 0 to 73 pct with most of the results being between 10 and 40 pct.

3. Insufficient data are available to definitely establish a difference in sulfur retention between firing methods, although there are some indications that the retention is less with cyclone firing than with pc firing.

4. The percentage of sulfur retention in ash was not closely reproducible, and some of the variation is probably a result of variability in the input sulfur. Results show the extreme importance of adequate coal sampling if the sulfur balance data is to be meaningful.

5. The SO_2 emission is strongly influenced by the sodium content of the coal. Increasing the sodium from 0.9 to 6.1 pct in the ash reduced the SO_2 in the combustion gases from 800 to 590 ppm.

6. Sulfur retention in the ash can be predicted from the chemical composition of the ash with some degree of confidence. The computer-derived relationship shows that increasing CaO and Na_2O increases sulfur retention, and clay and silica in the ash tend to decrease sulfur retention.

7. Boiler load does not greatly influence SO_2 emissions on the basis of limited testing of a pc-fired unit.

8. NO_x emissions are greater for cyclone-fired than for pc-fired units.

9. The fly ash particle size at the boiler outlet when burning low-sodium-content lignite is greater using cyclone firing than for pc firing.

10. As predicted by a correlation developed in laboratory tests, increasing the sodium content of the ash from 0.9 to 6.1 pct reduced resistivity by a factor of 100 in field tests.

PILOT PLANT SCRUBBER TESTS TO REMOVE SO_2 USING SOLUBLE ALKALI IN WESTERN COAL FLY ASH

by

P. H. Tufte,[31] E. A. Sondreal,[31] K. W. Korpi,[32]
and G. H. Gronhovd[33]

Introduction

Sulfur dioxide emission from powerplants has become the subject of much recent legislation and extensive engineering study. The most fully developed process for reduction of sulfur emissions is the lime-throwaway wet-scrubbing process. The present study utilizes an adaptation of the basic priniples of lime and limestone scrubbing in that the necessary alkali is the calcium oxide in fly ash derived from western coals. The process under consideration combines the known capability of wet scrubbing to remove a high percentage of fly ash from flue gas with the potential of the alkali in some coals to remove a significant portion of the SO_2 in the flue gas. Fly ash alkali could be augmented with lime (for additional SO_2 removal) if required to meet emission regulations.

The Federal standard for new stationary sources limits SO_2 discharge to 1.2 lb/MMBtu heat input.[34] Local and State standards may in some instances require lower limits based on emissions or ambient air quality. For a typical North Dakota lignite, the Federal standard of 1.2 lb/MMBtu is equivalent to a sulfur content of 0.65 pct dry basis in the coal if all coal sulfur is emitted as SO_2. A survey of Northern Great Plans lignites[35] indicates an average sulfur content of 1.0 pct on a dry basis for 212 samples.

A significant portion of the sulfur may react with the fly ash alkali to effectively reduce emissions, as discussed in a paper by Gronhovd, Tufte, and Selle at this symposium. The quantity of sulfur removed is variable between 0 to 60 pct of the total sulfur, dependent on alkali, clay, and silica contents of the ash.. A nominal retention of 20 pct would leave 1.5 lb/MMBtu in the stack gases from burning a typical lignite having 1 pct sulfur on a dry basis. Some SO_2 control will probably be necessary if a large portion of North Dakota lignites are to meet the Federal emission standards.

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[32] Mechanical engineer, Abe W. Mathews Engineering Co., Hibbing, Minn.

[33] Chief, Grand Forks Energy Research Laboratory, Bureau of Mines, U.S. Department of the Interior, Grand Forks, N. Dak.

[34] Dibelius, N. R., and W. F. Marx. The Status of Environmental Legislation Affecting Fossil Fuel Fired Power Generating Equipment, Combustion, v. 44, No. 2, August 1972, pp. 11-15.

[35] Work cited in footnote 24.

Assuming average values for Northern Great Plains lignites of 24.6 pct CaO in ash, 9.9 pct dry ash, and 1.0 pct dry sulfur,[36] the stoichiometric ratio of total CaO in the coal to total sulfur would be 1.40. The variability for this ratio is large, but a significant portion of SO_2 could be removed with the CaO in the ash, provided that the calcium was available for reaction. These considerations led to the present work using fly ash as an agent for SO_2 scrubbing.

To implement this study, a 110-scfm pilot plant flooded-disk scrubbing system was constructed and tested at the Grand Forks Energy Research Laboratory. To data, two western coal fly ashes, one of high-alkali content and one of low-alkali content have been tested. The effects of the level of ash addition and the level of SO_2 in the gas on SO_2 removal were evaluated. Acetic acid was added in selected tests to promote alkali solubility.

Additional work was done with a 3,000-scfm Krebs Elbair spray-impingement scrubber at the Minnesota Power and Light Co. at Duluth, Minn. Tests on ash alkali scrubbing in this system were performed cooperatively by the Abe W. Mathews Engineering Co. and the Federal Bureau of Mines. The ash tested was from the same source as the low-alkali ash used in the Grand Forks pilot plant tests.

The basic principles of operation are the same for both scrubbing systems. Sulfur dioxide is absorbed to form sulfurous acid, sulfurous acid is oxidized to sulfate, and sulfate is precipitated as gypsum. Under certain conditions, sulfur may be precipitated as sulfite. The limiting factor for closed-loop steady-state operation, which is defined for this report as no further buildup of sulfur species as SO_3 in solution, is the CaO furnished by the Ash. Sufficient CaO must be available to precipitate all SO_2 absorbed. Thus, the amount and solubility of CaO in fly ash becomes important.

The chemistry of flue gas-ash-water systems is partially known from studies on lime wet-scrubbing processes but is complicated because of the large number of chemical species. Present knowledge does not permit design based on first principles. For a summary of the state of the scrubbing art consult Slack,[37] and for equilibrium chemistry, Lowell.[38]

Test Program at Grand Forks

Test Equipment

In the flooded-disk scrubber shown in figure 64, a flow of liquid spills from the edge of the disk and is swept through a surrounding annulus by a

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[36] Work cited in footnote 24.

[37] Slack, A. V. Sulfur Dioxide Removal From Waste Gases. Noyes Data Corp., Park Ridge, N.J., 1971, 200 pp.

[38] Lowell, P. S. A Theoretical Description of the Limestone Injection-Wet Scrubbing Process. National Technical Information Service, U.S. Department of Commerce, Springfield, Va., v. 1-2, Rept. PB 193-029 and PB 193-030, June 9, 1970, 230 pp.

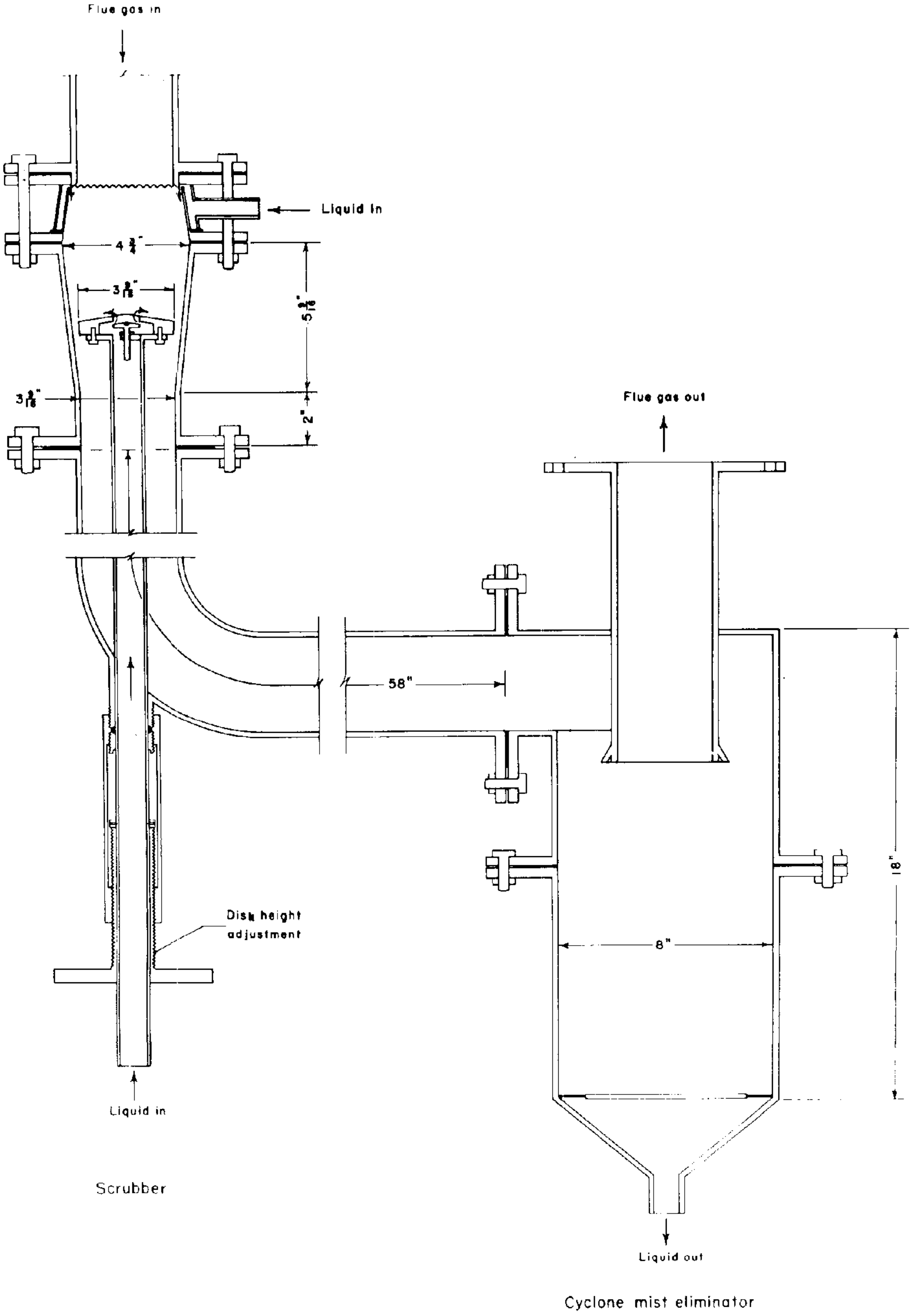

FIGURE 64. - Flooded-disk scrubber and cyclone mist eliminator.

downward flow of flue gas. Turbulence in the two-phase flow through the annulus shatters liquid droplets and promotes scrubbing. Provision for raising and lowering the disk within the tapered section permits changes in pressure drop while maintaining the same gas flow. The pressure drop is normally maintained at 10 inches H_2O. An auxiliary flow of liquid is directed through a serrated weir above the disk to establish a sharp boundary between wet and dry zones above the disk constriction. Flows are nominally 2 gal/min to the disk and 0.5 gal/min to the weir. Liquid is separated from the exiting gas by a cyclone mist eliminator that operates at about 99-pct efficiency at 1.5 inches pressure drop. Gas residence time between the disk and the cyclone exit is 0.3 sec.

The scrubber system shown in figure 65 includes the disk scrubber, cyclone separator, a catch tank, two 150-gal batch mixing tanks, and a 500-gal settling tank. Flue gas is generated by a nominally 75-lb/hr pc-fired pilot plant combustor, which has previously been described.[39] Before the scrubber, the flue gas is directed through an electrostatic precipitator where 99^+ pct of the fly ash is removed and discarded. All test ash used in scrubbing is added to the scrub liquid in the mixing tanks. None is added from the combustion gas.

The materials of construction for liquid and flue gas line were stainless steel. Mix and settling tanks were of mild steel and lined with paint. Liquid feed lines were deliberately oversized to allow for some scale buildup.

The closed-loop liquid flow circuit proceeds from the bottom of the cyclone separator to one of the two ash mixing tanks, then to the settling tank, and finally back to the top of the scrubber.

Operation and Test Procedure

All tests began with a 4-hour warmup period to establish steady operation of the test furnace. During the early phase of the program, tests were completed in an 8-hour period; however, in the later phase of the work, tests were 72 to 96 hours of continuous operation. Test results reported in some instances include the consolidated results of successive test periods conducted at the same conditions.

The lignite burned was selected to produce flue gas containing nominally 400 ppm SO_2. Higher levels were achieved by adding SO_2 from a gas cylinder to the flue gas.

All of the flue gas generated in the furnace was fed to the scrubber. Inlet gas flow at 280° F was nominally 170 acfm or 110 scfm and varied by ±10 pct. Gas outlet temperature was nominally 115° F. The liquid rate to the

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[39] Gronhovd, G. H., A. E. Harak, and P. H. Tufte. Ash Fouling and Air Pollution Studies Using a Pilot Plant Test Furnace. Paper in Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota, Grand Forks, N. Dak., May 1-2, 1969, comp. by J. L. Elder and W. R. Kube, BuMines IC 8471, 1970, pp. 69-88.

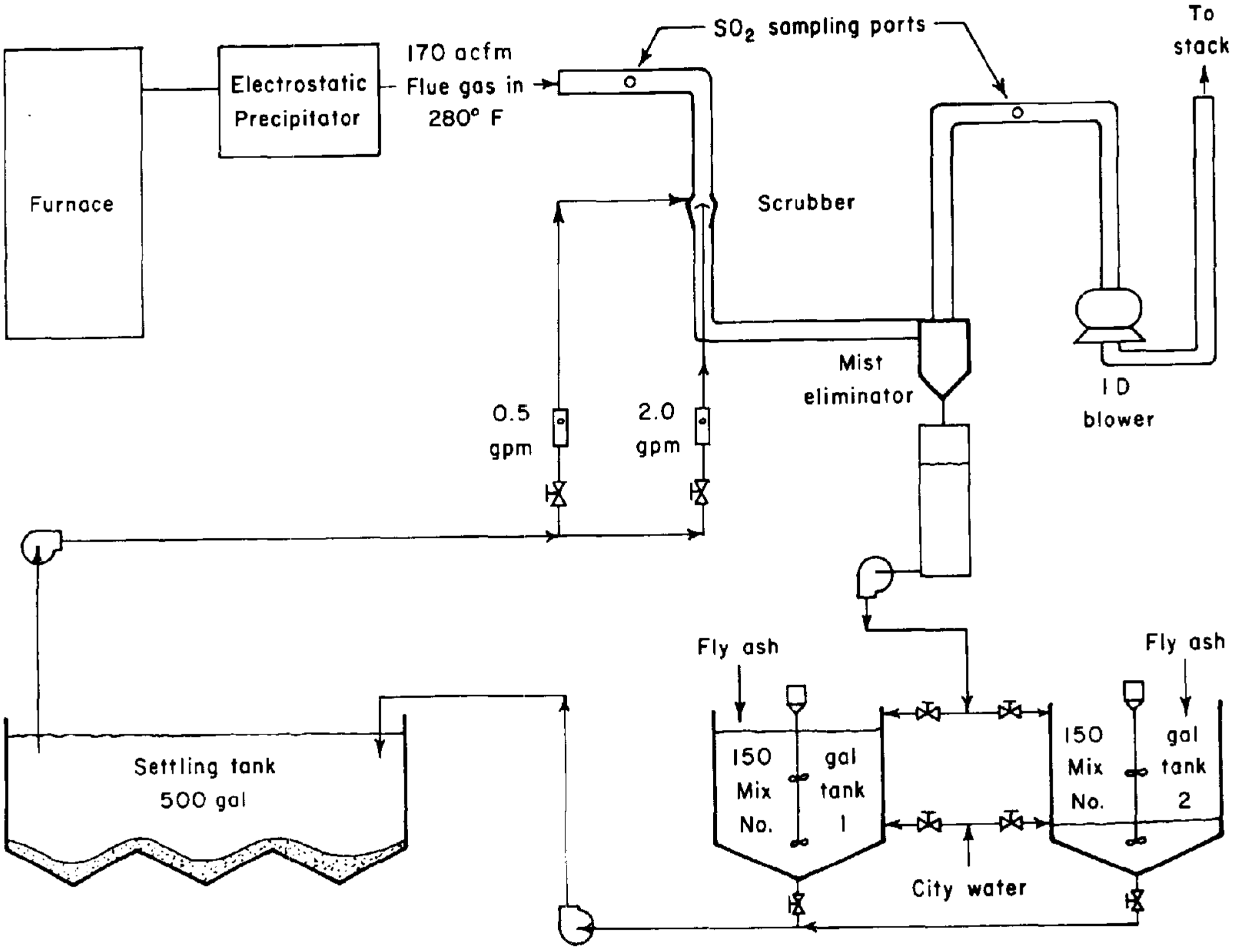

FIGURE 65. - Flowsheet for Grand Forks wet scrubber system.

scrubber averaged 2.5 gal/min and was varied to maintain constant liquid-to-gas ratios of 15 gal/1,000 acf or 22 gal/1,000 scf. The inlet scrubbing liquid temperature averaged 100° F, and the outlet, 110° F.

A weighed amount of fly ash was added to each 150-gal batch in the mixing tanks. This and the maintenance of a constant liquid-to-gas ratio established a fixed proportion of ash to flue gas, which could be interpreted as a dust loading. Addition of ash to the mixing tank permitted precise control of the type and amount of ash added. Such control would not have been possible if the ash had originated in the furnace and entered with the flue gas.

Mixing tanks were operated alternately. One was emptying while the other was filling and being mixed.

Delay time in the mix tanks was nominally 50 min, sufficient to dissolve nearly all of the ash going into solution in a one-pass batch extraction, as shown in laboratory beaker tests. The 160-min residence time in the settling tank reduced the solids content by 80 pct. The time required for complete turnover of liquid was approximately 4 hours.

No liquid was deliberately drained from the system but about 3 gal/hr of makeup was required to compensate for spray loss, evaporation, and pump leakage. Mean residence time based on total flow rate was 220 hours. Solids collecting in the settling tank were allowed to accumulate over the course of several weeks of testing.

Gas measurements were made at the inlet and outlet to determine temperature, pressures, and percentages of SO_2, O_2, and CO_2. Inlet gas flow was measured periodically using a pitot traverse. A Bureau of Mines modification of the Shell Thornton method[40] was used for measuring flue-gas SO_2 each hour. Samples were taken simultaneously at the scrubber inlet and outlet, using a 3-pct hydrogen peroxide solution as SO_2 absorbent. Sampling at the scrubber outlet required a heated sampling line with filter to prevent entrainment of condensate. Oxygen and CO_2 concentrations were measured by Orsat analysis; the oxygen level at the scrubber was typically 7 pct, and CO_2, about 12.5 pct.

Scrubber effluent and feed were sampled each half hour for pH level, which was also determined for each batch from the mix tanks prior to pumping. Every 4 hours, samples of feed and effluent were retained for laboratory analysis. Calcium and magnesium in solution were determined by the EDTA wet chemical method.[41] Calcium was also measured by X-ray fluorescence. The Na and K were obtained by the flame photometer method. Sulfur in solution was determined by the $BaCl_2$ gravimetric method and by X-ray fluorescence. Solution analyses were reported as equivalent CaO, MgO, Na, K, and SO_3. It is recognized these may not be the actual species occurring in solution. Particular note should be made that sodium and potassium are reported on an elemental basis in the solution analyses, and on an oxide basis in the ash analyses.

The suspended solids were filtered, and selected samples were analyzed by X-ray fluorescence. The fly ash fed to the system was analyzed using both wet chemical and X-ray fluorescence methods. Selected ash residues were evaluated by X-ray diffraction. In this paper, concentration in the gas phase expressed as parts per million is a volume/volume ratio, and in liquids or solids, parts per million is a weight ratio.

Results

Laboratory Tests on Dissolution of Fly Ash

Because water-soluble alkali plays a very important part in wet scrubbing, several types of laboratory solubility tests were made on the Colstrip and Glenharold ashes used in the scrubber tester. Analyses of these ashes are

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[40] Goksoyr, H., and K. Ross. The Determination of Sulfur Trioxide in Flue Gases. J. Inst. Fuel, v. 35, No. 255, April 1962, pp. 177-179.

[41] U.S. Bureau of Mines. Methods of Analyzing and Testing Coal and Coke. BuMines Bull. 638, 1967, pp. 78-79.

presented in table 9. These ashes were used in both beaker and pilot plant tests.

TABLE 9. - Analyses of fly ashes used in scrubber tests

Component	Glenharold,[1] pct	Colstrip,[2] pct
Loss on ignition........	0.1	1.3
SiO_2....................	35.9	51.5
Al_2O_3....................	13.0	20.7
Fe_2O_3....................	10.8	9.5
TiO_2....................	.4	.6
P_2O_5....................	.3	.4
CaO.....................	23.9	9.3
MgO.....................	6.2	3.9
Na_2O....................	6.9	.2
K_2O.....................	1.0	.8
SO_3......................	2.4	.8
Total..............	100.9	99.0

[1] Fly ash produced at Basin Electric Co. powerplant near Stanton, N. Dak. Collected from mechanical dust collector. Derived from Glenharold lignite, Mercer County, N. Dak.

[2] Fly ash produced at Minnesota Power and Light Co. Hibbard powerplant at Duluth, Minn. Collected from mechanical dust collector. Derived from Colstrip subbituminous, Rosebud County, Mont.

The first series of laboratory tests used varying concentrations of dilute acetic acid to wash ash at two proportions of ash to water. The concentration of soluble species in the Glenharold ash increased both with the amount of ash added and with the drop in pH associated with increased concentration of acetic acid, as shown in table 10. Table 11 shows the dissolution of Colstrip ash in a 120-ppm-acetic acid solution.

The addition of acetic acid to the extraction liquid increased the dissolution of CaO and MgO, but not of Na, K, and SO_3. The dissolvable Na and K are thought to be present as soluble sulfate. The insoluble Na and K and the insoluble portions of CaO and MgO are believed to be combined in inert glassy-fly-ash structure. Previous work on ash fouling has shown that a fraction of the alkali is present in glassy spheres, precluding dissolution. Dissolution of MgO was increased from 0 to 17 pct by addition of progressively more acetic acid up to 1,960 ppm, owing to the pH sensitivity in solubility of MgOH. Calcium oxide dissolution was increased by a factor of three, perhaps because of complexing by acetate and the influence of pH. Data, now shown, on effect of acetic acid on calcium dissolution in a pilot plant scrubber solution presumed to be saturated with CaO, indicates a 10-pct increase in solubility associated with addition of 900 ppm acetic acid.

TABLE 10. - Laboratory dissolution of Glenharold ash in water and acetic acid solutions[1]

	Quantity in ash added, ppm	Ppm dissolved	Pct	Ppm dissolved	Pct	Ppm dissolved	Pct	Ppm dissolved	Pct
		0 ppm acetic acid (pH 11.15)		98 ppm acetic acid (pH 10.95)		245 ppm acetic acid (pH 8.15)		490 ppm acetic acid (pH 5.45)	
Ash (2,000 ppm):									
CaO.......	478	74	15.5	105	22.0	136	28.5	181	37.9
MgO.......	124	0	.0	2	1.6	6	4.8	16	12.9
Na_2O......	138	9	6.5	11	8.0	12	8.7	14	10.1
K_2O.......	20	1	5.0	8	4.0	1	5.0	1	5.0
SO_3.......	48	28	58.3	31	64.6	32	66.7	33	68.8
		0 ppm acetic acid (pH 11.1)		490 ppm acetic acid (pH 10.3)		980 ppm acetic acid (pH 9.6)		1,960 ppm acetic acid (pH 5.1)	
Ash (4,000 ppm):									
CaO.......	956	140	14.6	272	28.5	312	32.6	362	37.9
MgO.......	248	0	.0	5	2.0	19	7.7	43	17.3
Na_2O......	276	22	8.0	26	9.4	27	9.8	30	10.9
K_2O.......	40	1	2.0	2	5.0	2	5.0	2	5.0
SO_3.......	96	64	66.7	62	64.6	60	62.5	63	65.6

[1] Beaker-scale tests were run by adding ash to the extraction liquids (acetic acid in distilled water) in the proportions indicated and stirring until a constant pH was obtained. Sitrring time was nominally 30 min. Analyses of ash are given in table 9.

TABLE 11. - Laboratory dissolution of Colstrip ash in acetic acid solution[1]

(Acetic acid solution; 120 ppm acetic acid, pH 5.9)

	Quantity in ash added, ppm	Ppm dissolved	Pct
Ash (2,000 ppm):			
CaO..................	208	34	16.3
MgO..................	86	8	9.3
Na_2O..................	4	1	20.0
K_2O..................	6	0	.0
SO_3..................	18	11	61.1

[1] Beaker-scale tests were run by adding ash to the extraction liquid (acetic acid in distilled water) and stirring until a constant pH was obtained. Stirring time was nominally 30 min. Analyses of ash are given in table 9.

In all beaker tests, the dissolution process as evidenced by change in pH was nearly complete in 15 min, with no change after 45 min. This finding, in solutions that were presumed not to be saturated, suggested at first that the maximum dissolution was achieved in a single extraction. However, when multiple extractions were performed in water and in scrubber solutions as shown in table 12, CaO and MgO continued to dissolve in successive extractions. Typically, a total dissolution two to three times that of the first extraction was achieved. Other beaker tests showed that the percent of CaO soluble increases with dilution down to an ash proportion of 1,000 ppm in water. Approximately 40 to 60 pct of the calcium in the Glenharold ash and 40 to 85 pct of the calcium in the Colstrip ash could be dissolved. The mechanism for continuing dissolution in multiple extractions does not appear to be repeated saturation of the batch solutions since varying and generally reduced amounts were dissolved in successive extractions. However, the results do suggest that recycling of ash from the settling tank back to the mix tanks in the pilot plant scrubber might increase utilization of ash.

In comparison of laboratory dissolution of ash species with experience in the pilot plant, material balances were run on transient periods of pilot plant operation prior to CaO saturation. Quantity of ash added and the increase in solute concentrations were noted. Calculations were based on the amount of ash added to one 150-gal batch in a mix tank and the average change in solute concentrations associated with the delay time for the batch. Results in table 13 indicate that species in the Glenharold ash were dissolved to extents greater than indicated by a single laboratory extraction in water but considerably less than by multiple laboratory extraction in scrubber solution. Species in the Colstrip ash were dissolved to a greater extent than for the Glenharold. The dissolved fractions for CaO and MgO were greater than in the single laboratory extractions with dilute acetic acid but not as high as in multiple laboratory extraction.

A pilot plant study on the effect of mixing time was made using Glenharold ash at 20,000-ppm concentration in 150 gal of tapwater agitated by a 1/2-hp mixer. The concentrations of Na and K leveled out within 5 min. No MgO dissolved. The SO_3 concentration increased to within 90 pct of its 2-hour values in 5 min. The CaO, however, took 30 min to reach 90 pct of the 2-hour level.

TABLE 12. - Laboratory dissolution of ash by multiple-extraction method[1]

300-ml extractions	CaO, ppm dissolved (pct)			MgO, ppm dissolved (pct)		
	In scrubber solution	In 1:1 diluted solution	In water	In scrubber solution	In 1:1 diluted solution	In water
	Glenharold ash[2]					
1	125(17.1)	170(23.2)	108(14.7)	0	16(9.5)	0
2	91(12.4)	102(13.9)	36(4.9)	16(9.5)	6(3.6)	0
3	91(12.4)	34(4.6)	34(4.6)	16(9.5)	0	0
4	103(14.1)	17(2.3)	25(3.4)	40(23.7)	65(38.5)	0
5	0	6(0.8)	32(4.4)	40(23.7)	49(30.0)	0
6	0	6(0.8)	20(2.7)	40(23.7)	0	0
7	ND	ND	16(2.2)	ND	ND	ND
Wash	12(1.6)	ND	34(4.6)	ND	ND	ND
Total	422(57.6)	335(45.7)	305(41.6)	152(89.9)	136(80.5)	0
	Colstrip ash[3]					
1	79(27.8)	136(47.9)	63(22.2)	8(7.5)	56(52.3)	0
2	34(12.0)	11(3.9)	18(6.3)	16(15.0)	0	0
3	34(12.0)	34(12.0)	7(2.5)	24(22.4)	0	0
4	22(7.7)	22(7.7)	14(4.9)	0	8(7.5)	0
5	10(3.5)	11(3.9)	9(3.2)	16(15.0)	0	0
6	0	11(3.9)	5(1.8)	0	0	0
7	ND	ND	ND	ND	ND	ND
Wash	12(4.2)	19(6.7)	10(3.5)	ND	ND	ND
Total	191(67.3)	244(85.9)	126(44.4)	64(59.8)	64(59.8)	0

ND Not determined.

[1] 0.915 gram of ash was placed in 300 ml of liquid and stirred for 1 hour. Undissolved ash was separated by centrifuging and saved for subsequent extraction. The liquid was analyzed to determine the quantities of Ca, Mg, NA, K, and SO_3 that dissolved. The extraction liquids used were solution from the pilot plant scrubber, a 1:1 dilution of this solution, and distilled water. Ash residues were ignited to 800° C after the third extraction. "Wash" represents the quantities dissolved in wash and transfer liquids, where measured. Analyses of ash are given in table 9.

[2] Quantities added to extraction liquid:
Total ash - 3,050 ppm
CaO in ash - 733 ppm
MgO in ash - 169 ppm

[3] Quantities added to extraction liquid:
Total ash - 3,050 ppm
CaO in ash - 284 ppm
MgO in ash - 107 ppm

TABLE 13. - Dissolution of ash in pilot plant tests[1]

	Quantity added, ppm	Range of conc in solution, ppm	Ppm dissolved	Pct dissolved
	Glenharold ash[2]			
Ash	3,040			
CaO in ash	727	207- 770	138	18.7
MgO in ash	188	8- 56	20	7.8
Na_2O in ash	210	50- 155	35	16.8
K_2O in ash	30	6- 16	3	10.2
SO_3 in ash	73	509-1,606	-	0
	Colstrip ash[2]			
Ash	900			
CaO in ash	94	358-1,043	42	45.3
MgO in ash	39	43- 268	13	34.3
Na_2O in ash	2	15- 30	1	2.1
K_2O in ash	3	3- 8	0	3.9
SO_3 in ash	8	621-2,561	-	-

[1] Amounts dissolved are computed from the change in concentrations in scrubber liquid with time during transient pilot plant operation.

[2] Analyses of ash are given in table 9.

Preliminary Pilot Plant Testing

Pilot plant tests were made at an SO_2 input of 500 ppm using untreated tapwater as a scrubbing liquid, without recycle. Increasing the water rate from 10 to 25 gal/1,000 acf brought about a linear increase in sulfur dioxide removal efficiency from 20 to 50 pct.

Flue gas velocity at the disk annulus ranged from 100 to 200 ft/sec, at several water-to-gas ratios, without effect on SO_2 removal efficiency. The corresponding ΔP across the disk ranged from 3 to 13 in H_2O. Thus, in this scrubber, power input measured by pressure drop did not effect SO_2 scrubbing efficiency.

Tests using a lime slurry without recycle were made at stoichiometric ratios of 1:1 and 2:1 for CaO to 500 ppm SO_2 in the flue gas. The liquid rate was 20 gal/1,000 acf. Scrubbing efficiencies at the stoichiometric ratios of 1:1 and 2:1 were 69 and 80 pct, respectively. By comparison, Slack[42] cites removals of 73 pct at 600 ppm and 42 pct at 500 ppm using 2:1 to 3:1 stoichiometric $CaCO_3$ in a venturi systems.

Preliminary tests using fly ash as a scrubbing medium were made without recycle to test the effect of concentration of dissolved alkali on scrubbing efficiency. In figure 66, the percent SO_2 removed from a flue gas of 500 ppm is indicated to be correlated with "available" alkali in solution, suggesting

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[42] Work cited in footnote 37.

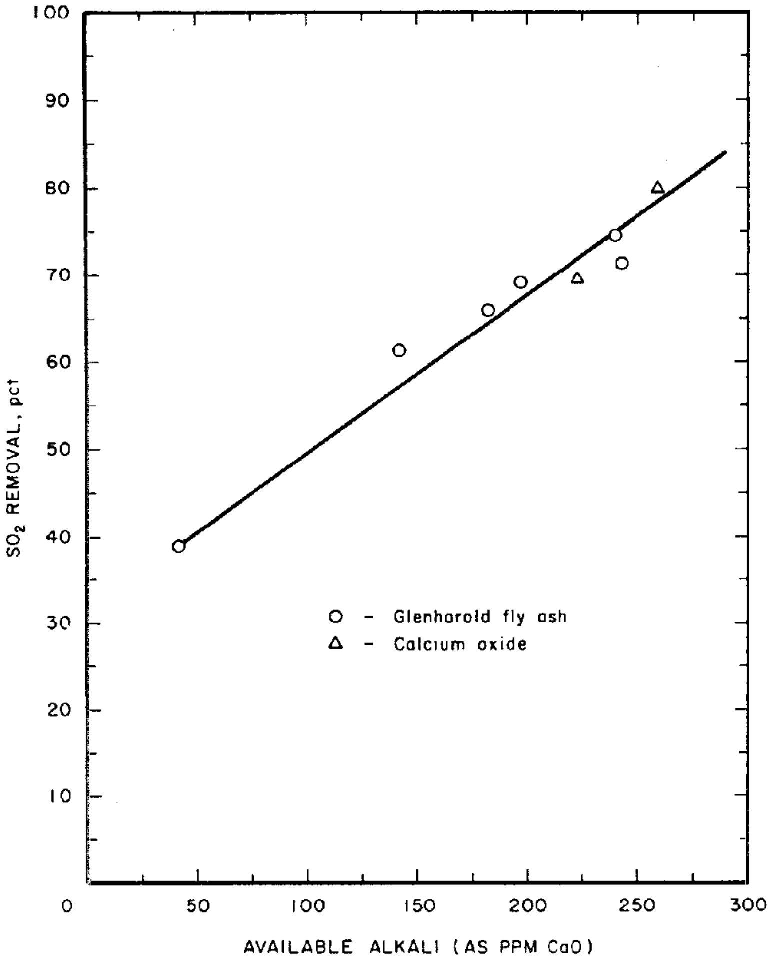


FIGURE 66. - Effect of scrubber solution alkali on SO_2 removal at 500 ppm inlet SO_2, once-through operation without recycle.

that the nonsulfate alkali may play a role in SO_2 removal. (Available alkali is defined as that in the feed liquid in excess of that stoichiometrically required for SO_3 in solution.) The preliminary tests are a simplification of the situations encountered in later recycle tests where total ion concentrations are higher.

Recycle Tests

Variables Studied

The main variables tested were the analysis or source of the ash, the amount of ash added, and the inlet level of SO_2. The effects of acetic acid and sodium addition were also investigated.

Fly ashes were derived from Glenharold lignite mined near Stanton, N. Dak., and Colstrip subbituminous coal mined near Colstrip, Mont. (Analyses are in table 9.) Glenharold ash was high in alkali, and the Colstrip ash tested was low in alkali. The Glenharold fly ash was produced at the Basin Electric powerplant at Stanton; and the Colstrip fly ash at the Minnesota Power and Light Co. plant at Duluth, Minn. All fly ash was obtained from mechanical dust collectors.

The two levels of ash addition chosen represent 80 and 25 pct of the total ash in a typical North Dakota lignite having 10 pct ash on a dry basis. The 80 pct might correspond to the fraction of ash leaving a pc-fired boiler, and the 25 pct, that leaving a cyclone-fired unit. The two levels include most firing conditions for lignite. Expressed as dust loading, the 80 pct is 4.0 grains per standard cu ft (gr/scf), and the 25 pct, 1.3 gr/scf.

Inlet SO_2 levels were approximately 400 ppm and 1,100 ppm SO_2, or 0.9 lb SO_2/MMBtu and 2.4 lb SO_2/MMBtu covering most discharge levels that had been encountered in field test experience. These concentrations represent 0.5 pct and 1.3 pct in terms of total coal sulfur on a dry basis, assuming all sulfur in the coal appears as SO_2 in the flue gas.

Acetic acid was added to both Glenharold and Colstrip ash slurries in selected tests in amounts from 300 to 1,000 ppm on a weight basis.

Sodium was added to the Colstrip ash solution in the form of sodium acetate to raise the sodium level in the solution to that found in the Glenharold solution, as well as to a much higher level.

Glenharold Ash Addition at High and Low Rates

Time histories of variables for scrubbing 500 ppm SO_2 in flue gas with Glenharold fly ash at both high and low add levels are given in figure 67. The first 144 hours are at the high ash-add rate, and the last 48 at the low ash-add rate.

The solution at the start of the test was tapwater. Ash was added in batches as testing progressed. The SO_2 removal at 4 gr/scf of Glenharold ash increased from 40 pct at the start to an average of 53.4 pct over the last 48 hours of the 144-hour test period. The increase in removal coincided with a rise in concentrations of dissolved ions during the approach to a steady state, and the higher percentage removal is considered the better measure of long-term operation using a closed-loop system.

The SO_3 and CaO concentrations in solution increased for the first 24 hours, at which time apparent saturation for CaO was reached at 900 to 1,000 ppm. The rate of SO_3 increase lessened as the CaO concentration peaked, indicating start of $CaSO_4 \cdot 2H_2O$ precipitation. Sulfur trioxide concentration increased throughout the test to 2,600 ppm at conclusion. The gradual increase in SO_3 coincided with a small decrease in CaO, which averaged 777 ppm for the last 56 hours. Sodium concentration increased throughout most of the test, reaching a constant level at 580 ppm. Magnesium ion concentration increased gradually to 210 ppm. The scrubber feed and effluent pH levels at 8.5 and 5.5 did not change appreciably after the first 24 hours.

The starting solution at the low ash-add rate was similar to that at the end of the high ash-add test. After the add rate of Glenharold ash was reduced from 4 to 1.3 gr/scf, SO_2 removal efficiency immediately decreased to the range of 30 to 40 pct; the pH dropped to the range of 3 to 4; and the solution concentrations changed. The SO_3 level in solution increased sharply for the first 24 hours and at a reduced rate the second 24 hours. The calcium in solution leveled off slightly higher than in the high ash tests, 1,000 ppm. The sodium level in solution remained relatively unchanged, averaging 505 ppm. The magnesium ion concentration increased rapidly from 200 to 440 ppm during the low ash test, probably because of the lower pH.

Summarizing the effects of reducing the amount of Glenharold ash added, average SO_2 removal decreased from 53 to 36 pct; pH dropped; Ca^{++} and Mg^{++} in solution were increased; sodium was relatively unchanged; and SO_3 concentration increased greatly.

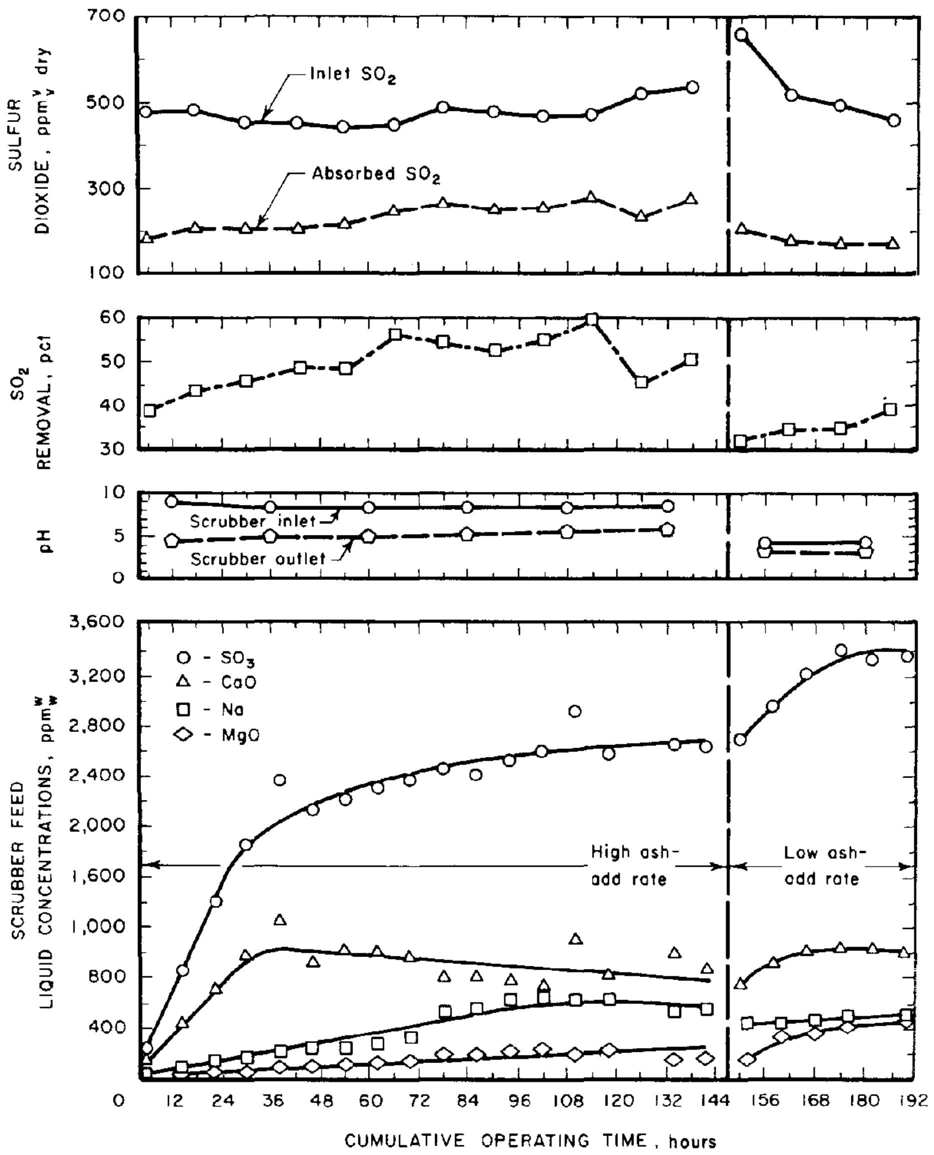


FIGURE 67. - Test history for low and high addition rates for Glenharold ash at low SO_2 concentration in gas.

Glenharold Ash at High SO_2

Glenharold fly ash at the high ash-add rate of 3.9 gr/scf was also tested at a higher inlet concentration of SO_2, 1,100 ppm. The time history for the high SO_2 test is given in figure 68.

The solution and at the start of the 1-day high-SO_2 test was that resulting from the high ash, low-SO_2 test, after acetic acid was added. The presence of acetic acid may have increased removal efficiency.

The SO_2 removal at the 1,100-ppm inlet level decreased from 55 pct to 40 pct over the 24-hour test. The SO_3 in solution increased from 4,500 to 5,400 ppm; and CaO dropped slightly to 1,200 ppm. Other changes in the solution were small increases in MgO and Na, and a small drop in pH.

A larger absolute amount of SO_2 was removed at the higher inlet concentration (445 ppm versus 259 ppm) even though the percentage of removal was lower (40.9 pct versus 53.4 pct).

Colstrip Ash at High and Low Add Rates

Time histories of variables for scrubbing 340 ppm SO_2 with Colstrip fly ash at both low and high add levels are given in figure 69. The first 144

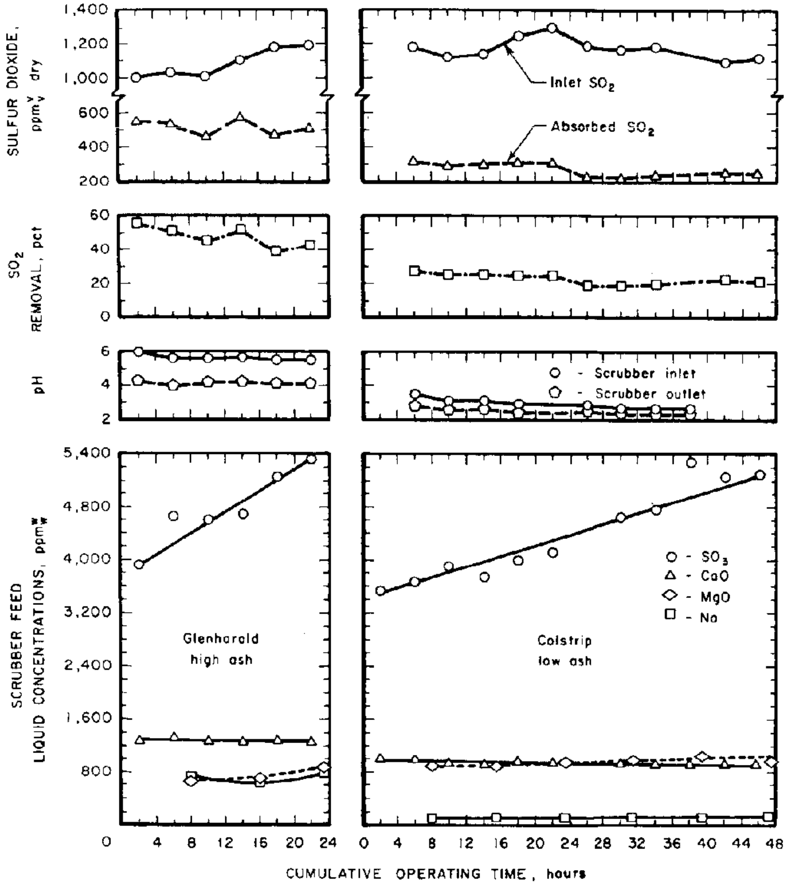


FIGURE 68. - Scrubbing histories at high SO$_2$ concentration in gas using Glenharold and Colstrip coal ashes.

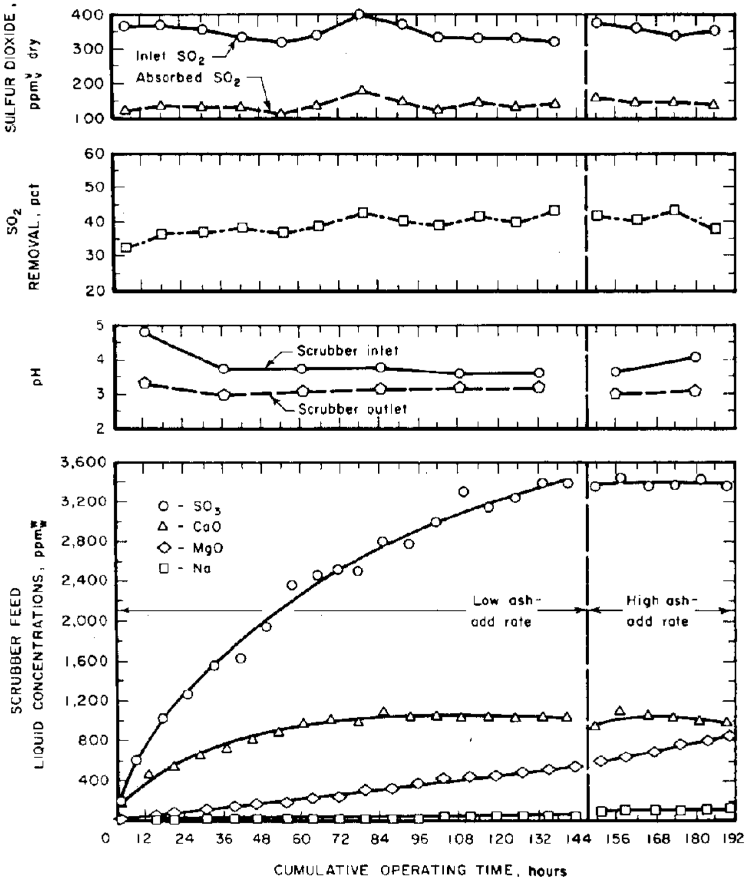


FIGURE 69. - Test history for Colstrip coal ash at low and high addition rates and low SO$_2$ concentration in gas.

hours were at the low ash-add rate, and the last 48 hours, at the high rate. The low-high add sequence was a reversal of that employed in Glenharold tests.

The test was started with tapwater. Ash was added in batches as testing progressed. The SO_2 removal at 1.3 gr/scf of Colstrip ash increased from 35 pct initially to an average of 42 pct over the last 24 hours. The SO_3 and CaO in solution increased for the first 72 hours, when apparent saturation for CaO was reached at 1,000 to 1,100 ppm. Thereafter, CaO remained unchanged for the duration of the low-ash test, averaging 1,054 ppm. The SO_3 continued to increase at a lesser rate for the remainder of low ash test, reaching 3,400 ppm at test end. Sodium concentration remained low, increasing to only 70 ppm, reflecting the low percentage of Na_2O in Colstrip fly ash. Magnesium ion concentration increased throughout the 6-day period, reaching 540 ppm. The average pH at inlet and outlet were 3.54 and 3.14, respectively.

The solution at the start of the high ash-add rate test was that existing at the end of the low addition run. After the increase, to 4 gr/scf of Colstrip ash, the SO_2 removal efficiency remained virtually unchanged at 41 pct. The pH of the feed increased slightly to 4.3, but the outlet pH remained unchanged. The SO_3 concentration in solution remained at 3,400 ppm. Concentration of calcium in solution dropped slightly at the outset but leveled out at 1,015 ppm. The sodium concentration increased to a constant level of about 120 ppm. Continuing the rate of increase noted in the low add test, concentration of magnesium increased from 580 to 880 ppm.

To summarize the effect of increasing the amount of Colstrip ash added from 1.3 to 4 gr/scf, no change occurred in SO_2 removal efficiency; SO_3 and CaO in solution changed only slightly; and Na and MgO increased significantly.

Colstrip Ash at High SO_2

Colstrip fly ash at the low ash-add rate of 1.3 gr/scf was also tested at an average SO_2 input of 1,172 ppm. The time history of the 2-day high-SO_2 test is shown in figure 68. The starting solution was that existing at the end of the high ash, low-SO_2 Colstrip test shown in figure 69.

The SO_2 removal percentage declined from 25 pct at the start to an average of 20.6 pct for the second 24 hours. The SO_3 in solution increased throughout the test from 3,790 to 5,300 ppm. The CaO, MgO, and Na levels remained virtually unchanged throughout the test, averaging 938, 982, and 120 ppm, respectively. The pH of the feed dropped from 3.2 the first 24 hours to 2.7 the second.

Comparing the tests at low- and high-SO_2 levels, the absolute amount of SO_2 removed increased from 144 to 237 ppm, but average removal percentage decreased from 40.8 to 20.6. The most evident changes in the solution were the increase in SO_3 and the drop in pH. The CaO in solution remained unchanged, and MgO and Na continued to increase at about the same rate throughout both tests.

Acetic Acid Addition

Acetic acid was added to Colstrip ash at the end of the low ash, high-SO_2 test. Addition at a concentration of 300 ppm produced no effect. When the concentration was increased to 1,000 ppm, the SO_2 removal for an input of 1,200 ppm SO_2 gradually increased from 22 to 32 pct over 28 hours. From the previous test, the average removal without acetic acid addition was 20.6 pct. The changes in the solution were a continuation of the increase in SO_3 observed in the nonacetic acid test and a gradual increase in MgO. The CaO and Na remained unchanged. The solution pH was gradually increased from 2.6 to 3.5.

Addition of acetic acid to Glenharold fly ash was made at the low SO_2 level and the high ash-add rate. With 500 ppm acetic acid added, the SO_2 removal percentage increased to 64 pct, compared with 53 pct with no acetic acid. Owing to unrelated changes in the scrubber solution in intervening tests, no direct comparison can be made between the concentration of ions with and without acetic acid. The pH in the acetic acid test was lower by about 1 unit.

Sodium Addition

Sodium was added at the end of the Colstrip low ash, high-SO_2 test. Scrubbing efficiency prior to addition was 30 pct at 1,200 ppm. The addition of sodium acetate to raise the Na level from 158 to 440 ppm resulted in an immediate increase of removal efficiency to 45 pct. Thereafter, the removal percentage gradually declined to 28 pct during 16 hours. The levels of CaO, MgO, and Na in solution did not change, but SO_3 continued to increase as during the previous test. The pH of the scrubber feed increased from 3.15 to 3.25 upon addition of NaAc, and then gradually declined to 2.85 during 16 hours when a second addition of sodium acetate was made, bringing the level of Na to 1,100 ppm. Once again a sharp increase in removal to about 45 pct was seen with a decline to 31 pct during the remaining 8 hours. The SO_3 in solution continued to increase while other ion concentrations remained unchanged. A sharp increase in pH of scrubber feed was noted on the second NaAc addition, from 2.85 to 3.65, with a gradual decay to 3.35 at end of test.

Results suggest that sodium addition produced a temporary beneficial effect, possibly due to increase in solution pH. However, the sustained level of SO_2 removal was not greatly affected by addition of up to 1,100 ppm Na.

Tapwater Control Test

Closed-loop tests were run without addition of ash for the purpose of establishing a baseline of absorption. Tests were run both by starting with water in the system and adding no ash and by discontinuing ash addition after ash-add tests on both Glenharold and Colstrip ashes.

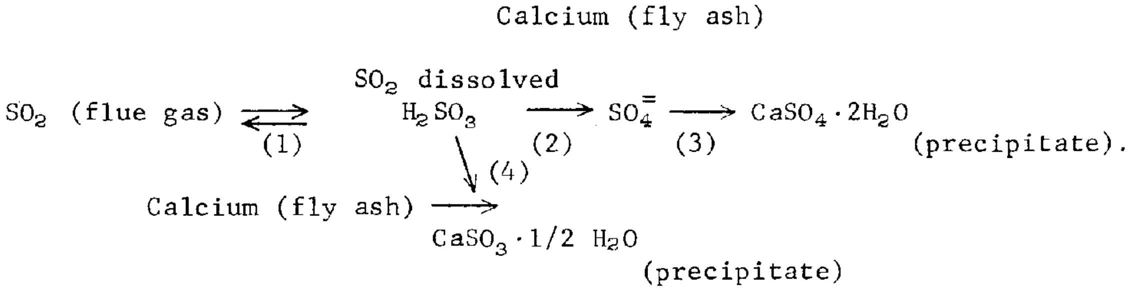
In the test started with water, sulfur dioxide removal efficiency averaged 13.9 pct during the test at an average input level of 338 ppm SO_2. The SO_3 concentration in the scrubbing liquid increased linearly throughout the test to 2,400 ppm. A significant amount of alkali was dissolved by the scrubbing liquid from scale and ash left from previous tests. (The amount of ash reaching the solution from the electrostatic precipitator is considered insignificant.) The constant Ca concentration in solution is evidence that saturation with respect to gypsum may have been reached. The pH reached 3.0 both in and out after the first test day and decreased 0.05 units over the last 2 days. Final levels of CaO, MgO, and Na in the solution were 670, 110, and 50 ppm, respectively, which are not greatly different than those in certain ash-add tests.

Similar SO_2 removal was obtained in tests where ash addition was discontinued after the scrubbing solution had been saturated by prior testing. The percentage of removal decreased to 16.3 pct after discontinuance of Glenharold ash, and to 11.9 pct after Colstrip ash test.

Because of dissolution of CaO from scale and ash, the system may have increased absorption over the level that would have been experienced with a clean system operated on tapwater. However, the tests served as a control since the state of scale in the system was similar to that in tests with ash additions. Thus, even though solution analyses do not appear greatly different, a positive dynamic effect on SO_2 removal due to ash addition is clearly indicated.

Discussion and Interpretation of Results

The model assumed for the scrubbing process is as follows:



(1)

Step 1 is the dissolution of gaseous SO_2 into the scrubbing solution. The driving force is the difference between the partial pressure of SO_2 in the flue gas and the solution SO_2 back pressure, which is a complex function of solution chemistry. The second step is the oxidation of the sulfurous acid or sulfite to sulfate. This may be catalyzed by species introduced with the ash. The third step is precipitation of the sulfate by calcium as gypsum. An alternate mode of sulfur removal, indicated as step 4, is precipitation of calcium sulfite. Steady-state SO_2 removal is achieved when steps 3 and 4 balance step 1, that is, when no sulfur species accumulate in the solution with time.

Effects of Alkali Added and Input SO_2

The ranges of variables studied were 0 to 4 gr/scf ash added and 340 to 1,150 ppm inlet SO_2. Summary data for the Glenharold tests appear in table 14, columns A, B, and C, and that for Colstrip in columns D, E, and F.

TABLE 14. - Data summary for Glenharold and Colstrip fly ashes in closed-loop operation

	Glenharold fly ash			Colstrip fly ash		
Ash-add level	High ash		Low ash,	High ash,	Low ash	
SO_2 level	Low SO_2	High SO_2[1]	low SO_2	low SO_2	Low SO_2	High SO_2
	(A)	(B)	(C)	(D)	(E)	(F)
OPERATING DATA						
Input ash (as dust loading) gr/scf	3.854	3.919	1.224	4.043	1.26	1.255
SO_2, ppm_v^v dry:						
In	485	1,087	472	346	336	1,152
Out	226	642	304	205	196	915
Removed	259	445	168	141	140	237
Removal pct	53.4	[2]40.9	35.6	40.8	41.7	20.6
pH:						
In	8.44	[2]5.45	4.3	4.03	3.54	2.6
Out	5.46	4.10	3.05	3.03	3.14	2.3
SCRUBBING SOLUTION ANALYSIS						
SO_3, ppm_w^w:						
Feed	2,562	[2]5,297	3,390	3,375	3,399	[2]5,140
Effluent	2,602	5,646	3,468	3,482	3,482	[2]5,300
CaO, ppm_w^w:						
Feed	859	[2]1,257	1,033	1,024	1,060	934
Effluent	777	1,176	991	1,015	1,054	938
MgO, ppm_w^w:						
Feed	206	[2]885	443	861	520	993
Effluent	213	909	434	877	544	982
Na, ppm_w^w:						
Feed	580	[2]810	495	116	63	120
Effluent	586	788	505	118	70	120
K, ppm_w^w:						
Feed	33	[2]50	33	17	14	21
Effluent	34	46	33	18	14	21
Total alkali in solution as equivalent SO_3, ppm:						
Feed	2,693	5,071	3,251	3,478	2,706	3,602
Effluent	2,602	4,963	3,219	3,472	2,758	3,584
REACTANT STOICHIOMETRY						
Quantities added in ash, g-mole/MMscf:						
CaO	1,066	1,084	338	435	136	135
MgO	384	390	122	253	79	79
Na_2O	277	282	89	8	3	3
K_2O	25	27	9	22	7	7
Total alkali	1,752	1,783	558	718	225	224
Flue gas:						
SO_2 input g-mole/MMscf	580	1,299	564	414	402	1,377
SO_2 removal g-mole/MMscf	310	531	201	169	168	284
Mole ratio $CaO_{in}/SO_{2_{in}}$	1.84	0.834	0.599	1.052	0.337	0.098
Mole ratio $CaO_{in}/SO_{2_{removal}}$	3.44	2.039	1.683	2.578	0.808	0.476
STEADY-STATE INDICES OF PERFORMANCE						
Indicated percent of scrubbed SO_2 precipitated from solution[3] (an index of transiency)	94.5	65.7	86.5	98.3	82.8	54.1
Hypothetical steady-state removal[4] percentage	50.5	26.9	30.8	40.1	34.5	11.1
Percent of CaO in input ash that must be dissolved to sustain the hypothetical steady-state removal	27.5	32.2	51.4	38.1	102.5	113.7

[1] Acetic acid added to scrub solution.

[2] Final value of transient condition used.

[3] Index defined as the percent of SO_2 removed that is precipitated. Calculation is based on the difference between total removal indicated by gas analyses and the transient increase in dissolved sulfur as SO_3.

[4] Hypothetical removal = percent removal (experimental) X indicated percent precipitated/100.

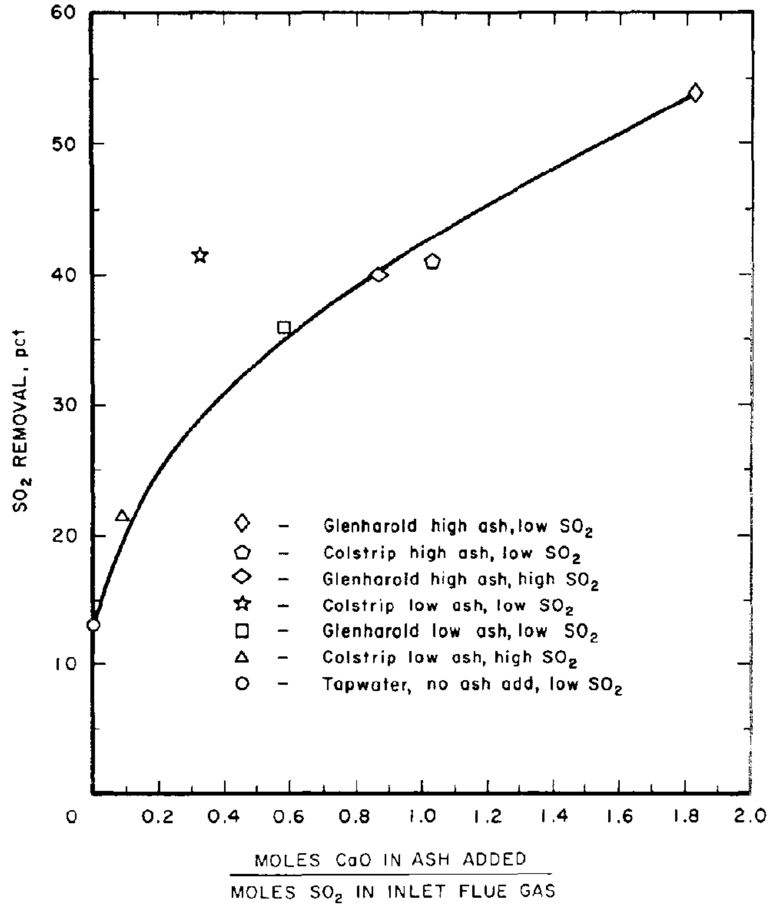


FIGURE 70. - The effect of molar CaO/SO_2 inlet ratio on SO_2 removal.

The parameter selected to represent the effects of alkali and input SO_2 is the molar ratio of CaO in ash added to input SO_2. Since sulfur is removed from the closed loop by precipitation with calcium, the amount of CaO added limits the amount of SO_2 that can be scrubbed at steady state. The inlet molar ratios (CaO/SO_2) studied ranged from 0.098 to 1.84 (table 14). In figure 70, sulfur removals are shown to correlate well with the CaO/SO_2 input ratio for both high- and low-SO_2 tests. The exception is the Colstrip low-ash, low-SO_2 test, for which no explanation is available.

The variables in figure 70 were found to be linearly related when plotted on log-log coordinates, which led to the following equation for representing percent removal of SO_2:

$$SO_2 \text{ removal, pct} = 43.5 \left(\frac{\text{moles CaO in ash added}}{\text{moles } SO_2 \text{ in inlet flue gas}} \right)^{0.34} \qquad (2)$$

By multiplying parts per million inlet SO_2 and converting units of measure, the absolute removal of SO_2 is

$$SO_2 \text{ removal, ppm} = 0.96 \text{ (dust loading, gr/scf} \times \text{pct CaO in ash)}^{0.34}$$
$$\times \text{ (ppm inlet } SO_2)^{0.66}. \qquad (3)$$

In this form, the effects of entering CaO and entering SO_2 on the absolute amount of SO_2 removed are illustrated. Doubling the entering CaO by increasing dust loading or a higher percent in the ash would increase the absolute removal by 26 pct. Doubling the inlet SO_2 concentration would increase absolute removal by 58 pct. The equations predict the experimental points for ash-add tests in figure 70 within 16 ppm, except for the Colstrip low ash, low-SO_2 test.

Comparing data from Glenharold and Colstrip tests given in table 14, the increase in input SO_2 from nominally 400 ppm to 1,100 ppm raised the absolute amount of SO_2 absorbed significantly, from 259 to 445 ppm for the Glenharold and 140 to 237 ppm for the Colstrip. However, removal percentages decreased from 53.4 to 40.9 pct for the Glenharold, and from 41.7 to 20.6 pct for the Colstrip as SO_2 level was increased. Equation 3 illustrates the increase in absolute removal that occurs with increased inlet SO_2, and equation 2 illustrates the corresponding decrease in percent removed.

The positive slope of the curve (fig. 70) indicates that increased addition of CaO definitely does improve the SO_2 absorbing ability of the solution recycled to the absorber. Thus, an increase in CaO added is inferred to reduce solution back pressure, probably by lowering the inventory of SO_2 (dissolved) and of H_2SO_3 in solution. A direct measurement of $SO_3^=$ as distinguished from $SO_4^=$ in solution could be used to substantiate this effect. Unfortunately, this analysis was not routinely performed. A possible mechanism for the effect is the formation of $CaSO_3$ precipitate (step 4 in equation 1), which would tend to occur as the pH was increased because of additional ash.

The increase in absolute removal with increased inlet SO_2 is caused by the greater driving force associated with a higher average partial pressure of SO_2 in the gas. The drop in percentage removal suggests that the solution back pressure is also somewhat increased as a result of the larger absolute amount of SO_2 entering the liquid and remaining to increase the sulfurous acid inventory in solution.

Relationship of Solution Composition to Scrubbing

In the present study to date, scrubbing has been a gas-liquid process, with essentially a clear solution passing through the scrubber. The amount of fly ash not removed in the settling tank and therefore passing through the scrubber in the feed liquid provided no significant amount of dissolvable calcium as compared with that already in solution. Thus, based on the proper analysis, it should be possible to correlate scrubber performance directly with concentrations in solution.

Based on the analytical data obtained, no consistent effect could be established between solute concentrations and SO_2 removal efficiencies (table 13). That the species measured were truly not the important determining factors of removal efficiency is further indicated by the fact that removal percentages remained relatively unchanged during the periods of transient solute buildup shown in figures 67 and 69.

In the absence of sufficient analytical data to fully support interpretation of the results, the following mechanism is postulated as one that is consistent with what is known. The solution leaving the scrubber contains large recirculating concentrations of Ca^{++}, $SO_4^=$, and possibly HSO_4^-, and a much smaller concentration of absorbed SO_2, which is rapidly hydrolyzed to sulfurous acid. The sulfurous acid reduces the pH and thereby lowers the solubility of $CaSO_4$, causing gypsum to be deposited as a scale. The solution of sulfurous acid is sufficiently saturated to be a poor absorbing liquid if immediately recycled to the inlet; however, the process of ash addition furnishing soluble calcium and the hold time in mixing and settling tanks reduces the sulfurous acid content and the resulting SO_2 back pressure so that absorption is again possible.

The reactions postulated for reducing sulfurous acid and solution back pressure are thus oxidation to the sulfate with subsequent precipitation as $CaSO_4$ (steps 2 and 3) and direct precipitation of $CaSO_3$ (step 4). The neutralization reaction between ash alkali and sulfurous acid may also influence back pressure of SO_2 through its direct effect on hydrogen ion concentration. Addition of greater amounts of ash tends to increase pH, which argues in favor of $CaSO_3$ precipitation. However, X-ray diffraction patterns of solid residues indicate $CaSO_4$ hydrates to be the usually precipitated species. Sulfite precipitate was identified in only one diffraction pattern, but the threshold of detection for sulfite leaves some possibility that it was generally present in small amounts.

In the reaction scheme postulated, the Ca^{++} concentration is fixed by one or both of the precipitation equilibria (steps 3 and/or 4). Adding more ash would not greatly effect these equilibria except by influence of pH on solubility, which explains the lack of importance of Ca^{++} in solution as a measure of performance. The addition of ash would, however, determine the amount of $SO_3^=$ that could be removed by either route, which illustrates the importance of the quantity of calcium added. Hydrogen ion concentration is also important. The explanation presented depends on the concentration of sulfurous acid in solution being the controlling factor determining solution absorption potential, and this remains to be substantiated by direct measurement. There is the subjective observation that addition of ash to the mixing tank produces an immediate disappearance of a strong SO_2 odor, indicating a rapid reduction in SO_2 back pressure.

One other consideration in the link between ash addition and removal of sulfurous acid is the role that various ash species, and particularly iron, may play in catalyzing oxidation (step 3). This consideration deserves to be investigated further under the conditions of an ash-based scrubbing system.

Steady-State Considerations

A necessary condition for steady-state operation is the availability of enough calcium to precipitate the sulfur removed. The mole ratio of total CaO input to SO_2 removed appears in table 13. For the Colstrip tests, this ratio is less than 1, indicating that the reported removal rates could not be sustained at steady state. The increasing SO_3 level in solution throughout the Colstrip tests further substantiates this concept.

Transiency of SO_3 in the scrubbing solution occurred throughout all tests to some extent. To evaluate transiency, a number of indices were computed from sulfur balances. The "index of transiency" in table 13 is the fraction of the SO_2 removed from the gas which is apparently precipitated, calculated as the difference between the total SO_2 absorbed and that represented by the increase in solute sulfur. This index, ranging from 54.1 to 94.5 pct (table 13), indicates a wide variation in transiency. An index of 100 pct would represent a complete steady-state operation.

A "steady-state removal percentage" appearing in table 13 was calculated by multiplying the index of transiency by the experimentally obtained percent removal. As this hypothetical SO_2 removal percentage represents the sulfur precipitated, it is the removal level that can be assumed to be sustainable if removal is controlled by precipitation. For the high-SO_2 tests for Glenharold and Colstrip, hypothetical steady-state removals are 26.9 pct and 11.1 pct, respectively, compared with 40.9 and 20.6 pct observed. The hypothetical steady-state removals for high ash, low-SO_2 tests are scarcely altered from reported values, being 50.5 versus 53.4 pct for Glenharold and 40.1 versus 40.8 pct for Colstrip.

Whether the hypothetical steady-state removal percentages are reasonable was further tested by computing a percent of input CaO solubility necessary to sustain such removal (table 13). The required solubilities for the low-ash Colstrip tests are slightly in excess of 100 pct. Otherwise the required solubilities fall in or below the 40- to 80-pct range observed in laboratory dissolution tests.

In evaluating the effects of transiency on test results, it should be emphasized that an increase in total dissolved sulfur ($SO_4^=$, $SO_3^=$, etc.) had little influence on SO_2 removal or pH. Also, in tests of more than 1-day duration, the concentration of sulfur in solution tended to approach a steady value with time. For unchanged removals, such steady-state values imply that precipitation increased as a steady state was approached. If the stoichiometric limit imposed by the calcium available in the ash were exceeded, increased precipitation would not occur.

Scaling

Scale formation due to calcium sulfate precipitation caused problems during extended scrubber operation. Buildup in scrubber liquid effluent lines and at the flooded disk necessitated shutdown for cleanup when severest scaling was encountered. Scale was also noted in the mix tanks, in the duct from the disk to the cyclone, and in the cyclone. Cleaning was required between some tests. Occasional scaling occurred in the settling tank. A typical analysis of the scale indicates about 80 pct $CaSO_4$, the remainder being fly ash. X-ray diffraction analyses indicated the presence of gypsum, $CaSO_4 \cdot 2H_2O$, and plaster of paris, $CaSO_4 \cdot 1/2\ H_2O$. The indication of plaster of paris is thought to be caused by loss of water of hydration from gypsum upon drying at 100° C. Calcium sulfite, $CaSO_3 \cdot 1/2\ H_2O$, was also identified in one test in limited amounts. The solution pH was above 7.0 in this test, a level at which $CaSO_3 \cdot 1/2\ H_2O$ solubility is very low. Magnesium and sodium levels in deposits were normally quite low.

Slurry piping was 1 inch diameter for flows of about 3 gal/min. In one 24-hour period, 3/16 inch of buildup was observed in a scrubber effluent line. Plugging at the disk was caused by wet-dry zone buildup above the weir and

and disk. Considerable enrichment in $CaSO_4$ was found in both type deposits. It is estimated that under the severest conditions encountered, operations would be limited by either scrubber or liquid line plugging to 2 days without cleaning. During the several days of transient operation after startup and prior to saturation of the solution with gypsum, no scaling or deposition was noted.

Some analyses were made of settling tank residue. Typical analyses showed a slight depletion of CaO from the ash feed and a slight enrichment in SO_3. This indicates the settled material was a combination of leached fly ash and precipitated gypsum. The bottom layer of settled ash became quite hard in all tests and was found to be depleted both in CaO and SO_3.

Test Program at Duluth

Tests were conducted jointly by the Abe W. Mathews Engineering Co. and the Federal Bureau of Mines at the M. L. Hibbard Steam Electric Generating Station of the Minnesota Power and Light Co. at Duluth, Minn. The cooperation of the Minnesota Power and Light Co. in providing facilities and assistance with testing is gratefully acknowledged.

The scrubber tested was of the same type as two commercial units that have been operating on 58-MW pc-fired boilers at the Minnesota Power and Light Station at Aurora, Minn., since June 1971.[43]

Test Equipment

The Krebs Elbair wet scrubber (fig. 71) directs spray concurrent with gas flow against a rod deck of closely spaced vertical rods. Scrubbing occurs through the rebound region of spray ahead of the rod deck and as the gas passes between the rods. Energy for scrubbing is introduced in the high-pressure spray. Spray is separated from the exiting gas by a chevron-type mist eliminator. Scrubbers may incorporate a number of stages in series to achieve improved removal efficiencies.

The scrubber tested consisted of a wet elbow and three scrubber stages. Total gas pressure drop was 2.5 inches of water. Spray to the wet elbow and first stage was at 200 lb/sq in; that to the second and third stages was 43 lb/sq in. Drainage was through seal pots.

Stack gas was obtained from pc-fired boiler 3 at the Hibbard Generating Station. No attempt was made to withdraw the test gas fraction isokinetically.

The complete scrubbing system shown in figure 72 included the Elbair scrubber, mix delay tanks, a settling tank, and a sump.

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[43] McGuire, E. F., and W. F. Arndt. Use of Western Coal and Air-Water Quality Control by Minnesota Power and Light Co. Paper in Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Bismarck, N. Dak., May 12-13, 1971, compiled by W. R. Kube and J. L. Elder. BuMines IC 8543, 1972, pp. 40-48.

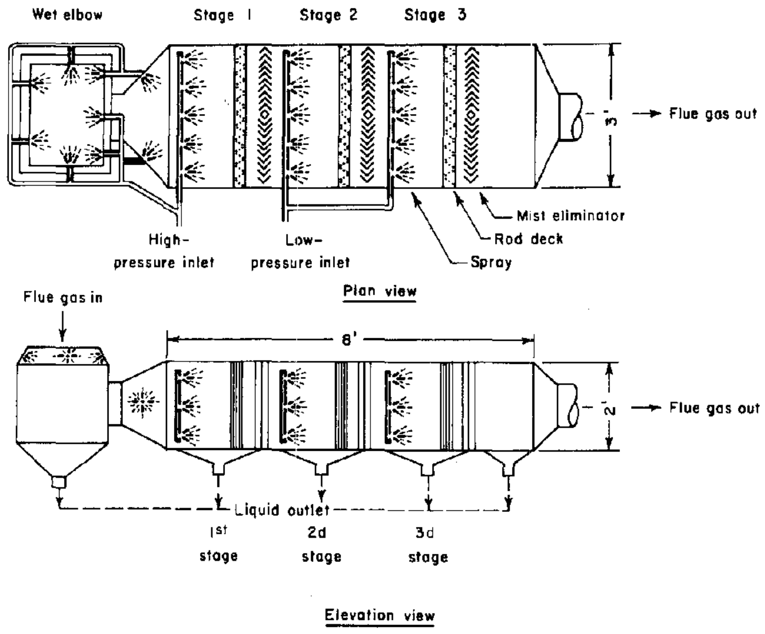

FIGURE 71. - Spray impingement scrubber used in Duluth tests.

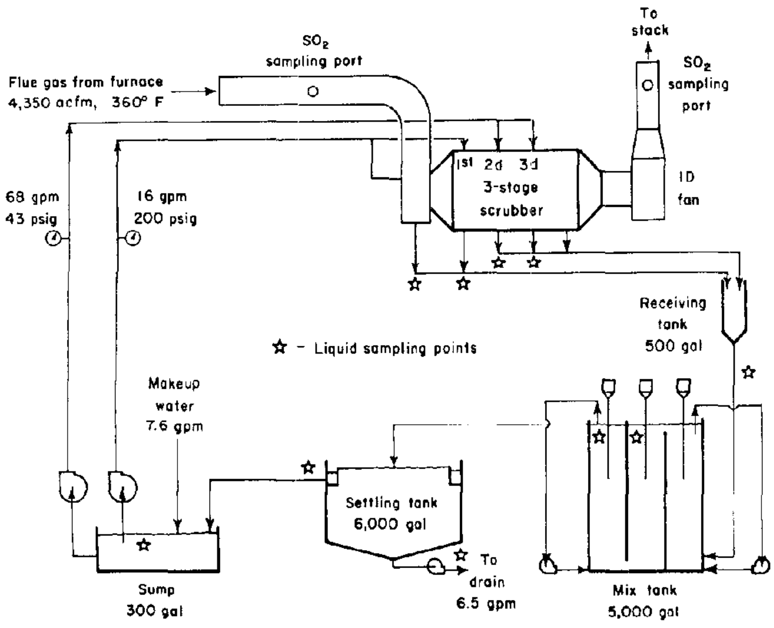

FIGURE 72. - Flowsheet of pilot wet scrubber system tested at Duluth, Minn.

Operation and Test Procedure

Operation was started January 11, 1973, and continued through January 26, with two weekend shutdowns and a 10-hour shutdown on January 18. Total operating time was 221 hours, or 9 days, all at essentially the same operating conditions.

When burned, the subbituminous coal from the Colstrip mine, Colstrip, Mont., averaged 1.44 pct sulfur on a moisture-free basis and produced an average SO_2 concentration of 1,100 ppm at the scrubber inlet. The average sulfur in the coal represents 2.53 lb SO_2/MMBtu; input to the scrubber averaged 2.69 lb SO_2/MMBtu. The higher sulfur rate indicated by the inlet SO_2 analysis results from sampling inconsistency, where gas and coal samples were not taken coincidentally or in sufficient numbers to give precise results.

The stack gas diverted to the scrubber averaged 4,350 acfm (2,800 scfm) or approximately 5 pct of the stack gas produced by the boiler operating at 22.5 MW. Gas temperature at the inlet averaged 360° F.

Ash entered the scrubber with the stack gases and represented 49 pct of the ash in the 9.3-pct-ash coal on an as-received basis. Average dust loading at the inlet to the scrubber was 2.27 gr/scf; dust loading at the exit was 0.015 gr/scf, indicating a fly ash removal efficiency of 99.3 pct.

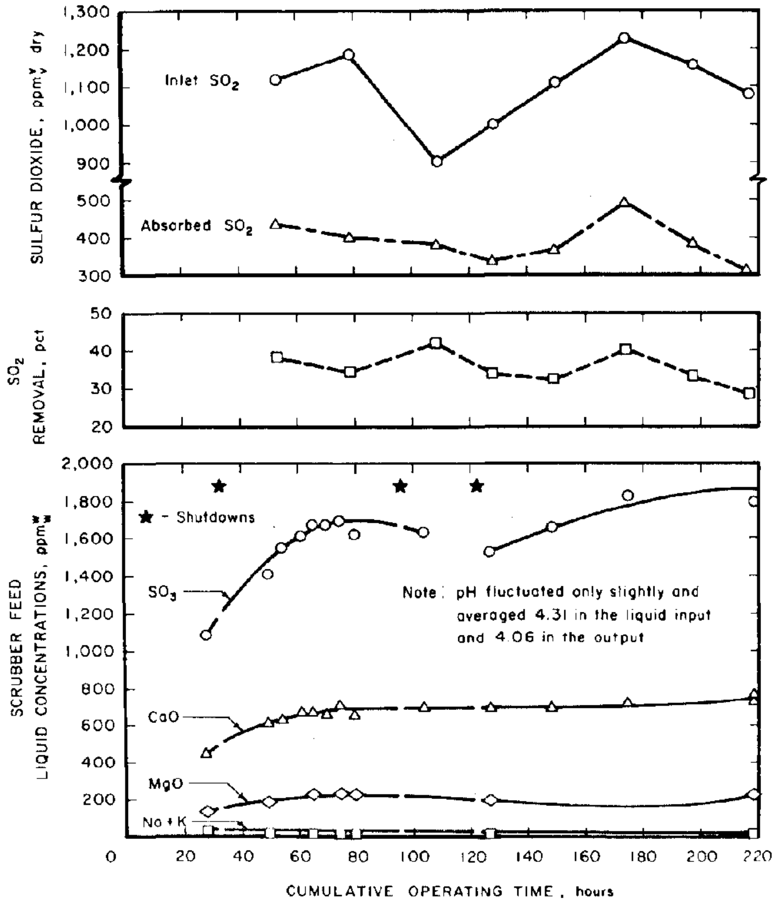

FIGURE 73. - Scrubbing history for Duluth tests using Colstrip ash.

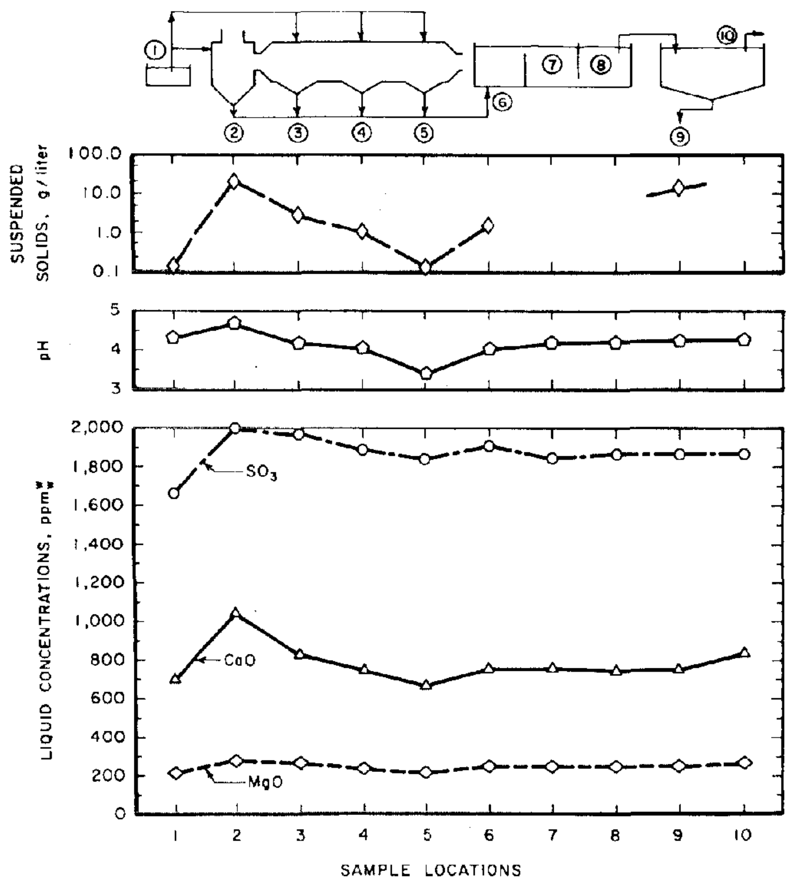

FIGURE 74. - Solution analysis at various locations in the Duluth scrubber based on averages for the last 160 hours of testing.

The total liquid rate to the scrubber was 84 gal/min or 31 gal/Mscf of stack gas. Flows by stages were 8 gal/min to the wet elbow, 8 gal/min to the first stage, 34 gal/min to the second stage, and 34 gal/min to the third stage. Slurry was discharged to the drain at 6.5 gal/min, and 1.1 gal/min was lost as evaporation and pump leakage; makeup was added at 7.6 gal/min. The mean residence time in the stirred mix tanks was 60 min. Baffles prevented short circuiting. Total liquid turnover time based on flow through the scrubber was 2.4 hours. Total residence time in the system based on outlet flow was 29 hours. Liquid temperatures were 128° F at the wet elbow, 126° F at the first stage, 123° F at the second stage, and 120° F at the third stage.

During operation, water samples, temperatures, and pH's were taken daily or more often from each of the 10 points indicated in figures 71-72. Coal from the coal feeders and ash from the mechanical dust collector were collected several times daily and consolidated into daily composites for analysis. Gas sampling for SO_2, O_2, and CO_2 was performed at the inlet and outlets points designated in figure 72. Dust-loading data were obtained at the same points. Analytical procedures used were the same as those described for tests at the Grand Forks Laboratory.

Results

Changes in SO_2 removal and in the specie concentrations in scrubbing feed liquid with time for the 221-hour test are shown in figure 73. Solution pH, not shown, changed little with time and averaged 4.31 in the feed and 4.06 in the combined liquid discharge. Liquid concentrations increased to near-steady values of 1,650 ppm SO_3 and 700 ppm CaO after 60 hours and changed only slightly for the 160-hour remainder of the test. These concentrations are lower than those in tests performed at Grand Forks using Colstrip ash.

The concentration of SO_2 in the inlet gas ranged from 908 to 1,230 ppm and averaged 1,100 ppm. Removal of SO_2 averaged 35.3 pct and was not consistently increased or decreased by any of the operating parameters. The overall trend, discounting intermediate peaks and valleys, was a slight downturn in percent removal from the start to the end of the test.

Average solution concentrations for the last 160 hours of the test are plotted for 10 sampling points in figure 74. The changes in liquid composition with position (fig. 74) and in the material balances (tables (15-16) reflect the distribution of scrubbing, dissolution, and settling occurring through the system. A large part of the fly ash was removed in the wet elbow, over 100 pct based on sampling of the liquid. Fly ash removal was nearly complete after the second stage, so that only 0.6 pct was removed by the third stage out of an overall fly ash removal of 99.3 pct.

Using analyses of liquids, the absolute amount of SO_2 removed was greatest in the second stage because of the higher liquid rate to the second stage compared with the wet elbow and first stage. However, the specific removal in proportion to liquid rate was highest in the wet elbow, where the wash liquid picked up 340 ppm SO_3. Each successive stage absorbed a lower concentration into solution, with the SO_3 in the liquid for the third stage increasing by only 180 ppm.

TABLE 15. - Material balance across scrubber stages for the Duluth test

	Gas stream		Combined liquid streams		Individual liquid streams by stages							
					Wet elbow		1st stage		2d stage		3d stage	
	Inlet	Outlet	Inlet	Outlet	In	Out	In	Out	In	Out	In	Out
Total mass flow..lb/hr[1]..	13,012	-	41,832	-	3,984	-	3,984	-	16,932	-	16,932	-
Ash...............lb/hr..	54.59	0.36	6.18	58.88	0.59	91.72	0.59	13.02	2.50	17.53	2.50	2.83
Ash removal........pct[2]..	-	99.34	-	96.53	-	(166.9)	-	22.77	-	27.53	-	0.59
SO_2...............lb/hr..	31.31	20.27	55.62	63.72	5.30	6.38	5.30	6.29	22.51	25.56	22.51	24.92
SO_2 removal.......pct[2]..	-	35.27	-	25.87	-	3.47	-	3.16	-	9.73	-	7.70

[1] The gas flow through the scrubber is equivalent to that produced from combustion of 1,200 lb of Colstrip coal per hour at 30 pct excess air.

[2] Removal percentages are computed as the amounts removed out of the gas or absorbed into the liquid over the amount entering in the gas stream, times 100. Removals are based on independent analyses on liquid and gas samples, not necessarily obtained at the same time. Incomplete closure on balances is discussed in the text.

TABLE 16. - Material balance across the entire scrubber system for the Duluth test

	Inputs			Outputs				Closure = $\frac{\text{output}}{\text{input}} \times 100$
	Gas[1]	Fly ash	Makeup water	Gas	Unremoved fly ash	Liquid to drain	Solid to drain	
Solids............lb/hr..	-	54.59	-	-	0.360	-	59.23	109.16
Pct of input............	-	-	-	-	0.66	-	108.49	
Sulfur (as SO_2)...lb/hr..	31.31	0.66	-	20.27	-	4.83	0.62	80.45
Pct of input............	97.95	2.05	-	63.40	-	15.12	1.93	
CaO...............lb/hr..	-	5.65	0.095	-	0.037	2.70	3.20	103.37
Pct of input............	-	98.35	1.65	-	0.65	47.05	55.67	
MgO...............lb/hr..	-	2.33	0.216	-	0.015	0.89	1.66	100.55
Pct of input............	-	91.53	8.47	-	0.60	34.83	65.12	
$Na_2O + K_2O$........lb/hr..	-	0.70	0.044	-	0.0046	0.088	0.59	92.14
Pct of input............	-	94.02	5.98	-	0.62	11.83	79.69	

[1] The gas flow through the scrubber is equivalent to that produced from combustion of 1,200 lb of Colstrip coal per hour at 30 pct excess air.

The material balance on the overall system given in table 16 indicates reasonable closure for all species except sulfur as SO_2, for which only 80 pct of the input is in the output. The liquid to the drain accounts for spproximately 43 pct of the SO_2 removed from the gas stream. The sulfur in solids going to the drain was approximately equal to the sulfur in entering fly ash.

The increase in pH for liquid passing through the wet elbow (fig. 74) indicates that an excess of ash compared with SO_2 is removed and dissolved in the wet elbow. In stages 1, 2, and 3, the pH decreases, indicating that more SO_2 was absorbed than ash dissolved. The gradual increase in pH through the mixing and settling tanks indicates a slight dissolution of ash and/or desorption of SO_2 into the atmosphere.

The scrubbing system was observed to be virtually free of scale buildup for the entire 221-hour test.

Discussion and Interpretation of Results

The removal efficiency of 35 pct reduced the SO_2 emission rate to approximately 1.7 lb SO_2/MMBtu. This discharge rate would be less than the 2.0 lb SO_2/MMBtu that becomes effective in Minnesota in June 1973[44] but more than the Federal standard for new plants (1.2 lb SO_2/MMBtu).[45]

Average dust loading of 2.27 gr/scf was equal to 49 pct of the total ash in the coal based on 9.3 pct average ash on an as-received basis. The calcium and magnesium oxides in the ash input to the scrubber were equivalent to only 32.2 pct of the entering SO_2. The removal of slightly more than a stoichiometric amount of SO_2 in relation to ash alkali (35.3 pct) in part was caused by the 8-pct liquid discharge to the drain that permitted sulfur to leave the system without being precipitated. Some SO_2 may also have been desorbed from the scrubbing liquid in the mixing and settling tanks, possibly explaining the poor closure on the system sulfur balance.

The absence of scaling is attributable to operating below the saturation concentration for calcium salts when using 9 pct dilution by makeup water. The rate of makeup and liquid discharge would perhaps be impractical for a large plant because of the amounts of water involved. However, the concept of operating just below saturation by using small amounts of makeup water in the feed is worthy of further investigation as a means of avoiding scaling in a system operating on dissolved alkali.

The two functions of ash alkali in scrubbing are that the soluble alkali should enhance the SO_2 absorbing properties of a scrubbing liquid, and that the absorbed SO_2 be precipitated as a calcium salt to permit solid waste

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[44] Minnesota Pollution Control Agency. Minnesota State Regulations. Air Pollution Control. Rules, Regulations, and Air Quality Standards, April 1972. Available upon request from Department of Administration, Centennial Bldg., St. Paul, Minn. 55155.

[45] Work cited in footnote 34.

disposal. The mechanism for scrubbing proposed in the discussion of Grand Forks results applies also to the Duluth test, except that no calcium salts were precipitated in the Duluth test because of dilution by makeup water.

During all operations at Duluth, ash continually entered the scrubber at an essentially fixed rate; thus, the Duluth test did not establish the extent to which dissolved ash enhanced SO_2 absorption. The closed-loop tests at Grand Forks did permit a comparison of operation with and without addition of ash in varying amounts, with increased addition of alkali in ash significantly increasing SO_2 removal efficiency.

The removal percentage of 35 pct in the Duluth test is somewhat improved over the 20+ pct removal obtained in the Grand Forks test conducted on Colstrip ash at the high-SO_2 level (1,100 ppm). The improved percentage removal can be attributed to one or more of the following differences: A higher liquid-to-gas ratio of 31 gal/1,000 scf at Duluth compared with 22 gal/1,000 scf at Grand Forks, a longer gas residence and contact time in the scrubber of 1 sec compared with 0.3 sec, a higher ash rate of 2.27 gr/scf compared with 1.25 gr/scf, and the discharge of sulfur out of the system in the liquid discharge to the drain. The increase in dissolved sulfur as SO_3 in the wash water passing through the scrubber is approximately 50 pct greater for the Duluth test, at 240 ppm compared with 160 ppm.

Conclusions and Recommendations

Results and conclusions are as follows:

1. Sulfur dioxide was removed from flue gas to extents of 20 to 60 pct using wet scrubbing with ash alkali from two Western U.S. coals. Removals are sufficient to bring stack emissions into compliance with Federal and State standards for some coals. An important advantage of an ash alkali process in future commercial application is that it avoids the need to supply large tonnages of limestone to a powerplant.

2. In some closed-loop tests in which insufficient CaO was introduced to precipitate the SO_2 absorbed into the solution, removals reported cannot be considered to represent removal rates for indefinitely sustained operation. However, where a considerable excess of CaO was introduced, the rates can with some certainty be considered to be representative of long-term operation. Trends during the longest test run identified no downturn in removal efficiency as a steady state was approached.

3. The addition of fly ash to a scrubbing liquid does significantly increase the scrubbing effectiveness of that liquid, and the amount of CaO added in relation to inlet SO_2 is an indicator of scrubbing potential. Thus, the process is specifically suited to those Western U.S. coals that combine the properties of relatively low sulfur content and high alkali content. The other alkali constituents in the ash, Na, K, and Mg, may also be beneficial, although their effect was not clearly established.

4. The absolute amount of SO_2 absorbed increases along with the inlet SO_2 level; however, the percentage of removal declines.

5. The SO_2-removal efficiency is not affected greatly by the total amounts of sulfur or alkali in solution. The forms of the solute species and pH are, therefore, indicated to be the important factors.

6. Removal of sulfur from a closed-loop ash-scrubbing system is principally by precipitation as gypsum, $CaSO_4 \cdot 2H_2O$. This indicates that the absorbed SO_2 must be oxidized. Under some conditions, $CaSO_4 \cdot 1/2\ H_2O$ and $CaSO_3 \cdot 1/2\ H_2O$ were found.

7. Scale buildup caused by precipitation of gypsum in the scrubber is a problem when a system is operated at the saturation level. Dilution of the feed to the scrubber with makeup water prevents scaling but also necessitates disposal of a liquid waste stream.

Recommendations for additional study are--

1. To establish by direct measurement the primary importance of the sulfurous acid content of the scrubbing liquid as the factor determining SO_2 back pressure and resulting scrubbing efficiency, and further to investigate the processes of neutralization, oxidation, and precipitation that govern the sulfurous acid balance.

2. To conduct a critical analysis based on theoretical calculations and experiments to determine the equilibrium relationship between gaseous SO_2, scrubbing liquid, and solid species in a ash scrubbing system; calculations to include use of Lowell's[46] computer program for calculating equilibrium, and pilot plant tests for approaching equilibrium by rapid recycling of a lower total liquid capacity of the system.

3. To perform extended tests to establish that SO_2 removal rates indicated are definitely sustainable beyond all indications of transient operation.

4. To investigate additional ash types to establish the relationship between SO_2 removal and measured properties of the ash; this should include a search for laboratory methods for characterizing ash beside direct chemical analysis and solubility tests.

5. To investigate operating variables that affect optimization of an ash-scrubbing process, including scrubbing temperature, gas residence time, liquid rate at steady-state operation, ash recycle, and feed dilution to prevent scaling.

6. To consider problems of ash disposal that may be caused or intensified by sorption of SO_2.

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[46] Work cited in footnote 38.

EXPERIENCE IN THE USE OF BROWN COALS FROM THE LATROBE VALLEY, AUSTRALIA

by

R. S. Higgins,[47] G. Bonafede,[48] and L. Hoch[49]

Introduction

The State Electricity Commission of Victoria (SECV), when constituted in 1918, was given responsibility for the development of the brown coal resources of Victoria for electricity and solid fuel production. Continuous large-scale development of the brown coalfields of the Latrobe Valley has provided the basis for the power and fuel undertakings of the SECV that now comprise three opencuts, one briquet factory, three operating power stations, and a further power station in which the first unit is being commissioned. Yallourn Power Station has a mixture of range connected plant and units of 120-MW capacity and has a total capability of 546 MW. Hazelwood consists of eight 200-MW units, while the 350-MW unit now being commissioned at Yallourn W Power Station is the first of four 350-MW installations. Total SECV investment in the Latrobe Valley now exceeds $650 million.

Over the history of the SECV to June 1972, a total of 425 million tons of brown coal has been mined, and in 1971-72, production was 22 million tons for the year. When the plant under construction has been commissioned and generation capacity has increased to over 3,000 MW in 1974, coal production will rise progressively to about 33 million tons per year and total coal reserves of 1,100 million tons will have been commited for the remaining life of these plants.

An extensive program of geological investigations carried out over more than 25 years by the SECV has defined the extent of the Latrobe Valley brown coal deposits and the principal areas most suitable for opencut winning. Numerous papers, including those by Thomas and Baragwanath,[50] Edwards,[51] Gloe,[52] and others, have described the stratigraphy and structure and other

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[49] Engineer, Hazelwood Power Station, Latrobe Valley Department.

All authors are with the State Electricity Commission of Victoria, Australia.

[50] Thomas, D. E., and W. Baragwanath. Geology of the Brown Coals of Victoria, Parts 1-4. Min. Geol. J. (Victoria), 1949-1951, v. 3, No. 6, pp. 28-55; v. 4, No. 1, pp. 36-52; v. 4, No. 2, pp. 41-63; v. 4, No. 3, pp. 36-50.

[51] Edwards, A. B. Coal Types in the Yallourn and Latrobe Brown Coal Seams. Proc. Australasian Inst. Min. & Met., 1947, Nos. 146 and 147, pp. 23-69.

[52] Gloe, C. S. The Geology of the Latrobe Valley Coal Field. Proc. Australasian Inst. Min. & Met., 1960, No. 194, pp. 57-125.

features of the coal measures, which underlie an area from 5 to 10 miles wide extending for over 30 miles. Figure 75 shows a plan of the major structures and of the concealed outcrops of the brown coal seams that have been identified in the different areas. On a geological basis, the coal reserves of the Latrobe Valley have been calculated as 47,500 million tons proved and 37,500 million tons inferred.

Of the proved resources of coal, some 29,000 million tons occur with less than 100 feet of overburden over the uppermost seam. However, when provision is made for factors such as present townships, rivers, realistic opencut planning, and a limitation on cost of coal winning to approximately present cost, these resources are reduced to about 10,000 million tons.

Two major difficulties have been encountered in the use of brown coals from the Yallourn Opencut and the Morwell Opencut in power station boilers. The first boilers installed in 1924 to generate power from Yallourn brown coal were grate fired, and some difficulties were experienced in burning the coal, which has a normal moisture content of 67 pct but can be as wet as 73 pct. The problem of unstable combustion was encountered in serious proportions when the mill-fired Yallourn C boilers (fig. 76), producing 200,000 pounds of steam per hour, were commissioned in 1954; despite a number of significant developments in handling and firing, flame-outs can still occur in the 120-MW Yallourn E boilers.

Since the application of pulverized fuel firing, difficulties have been experienced from ash depositing on heating surfaces; at Yallourn, the boilers

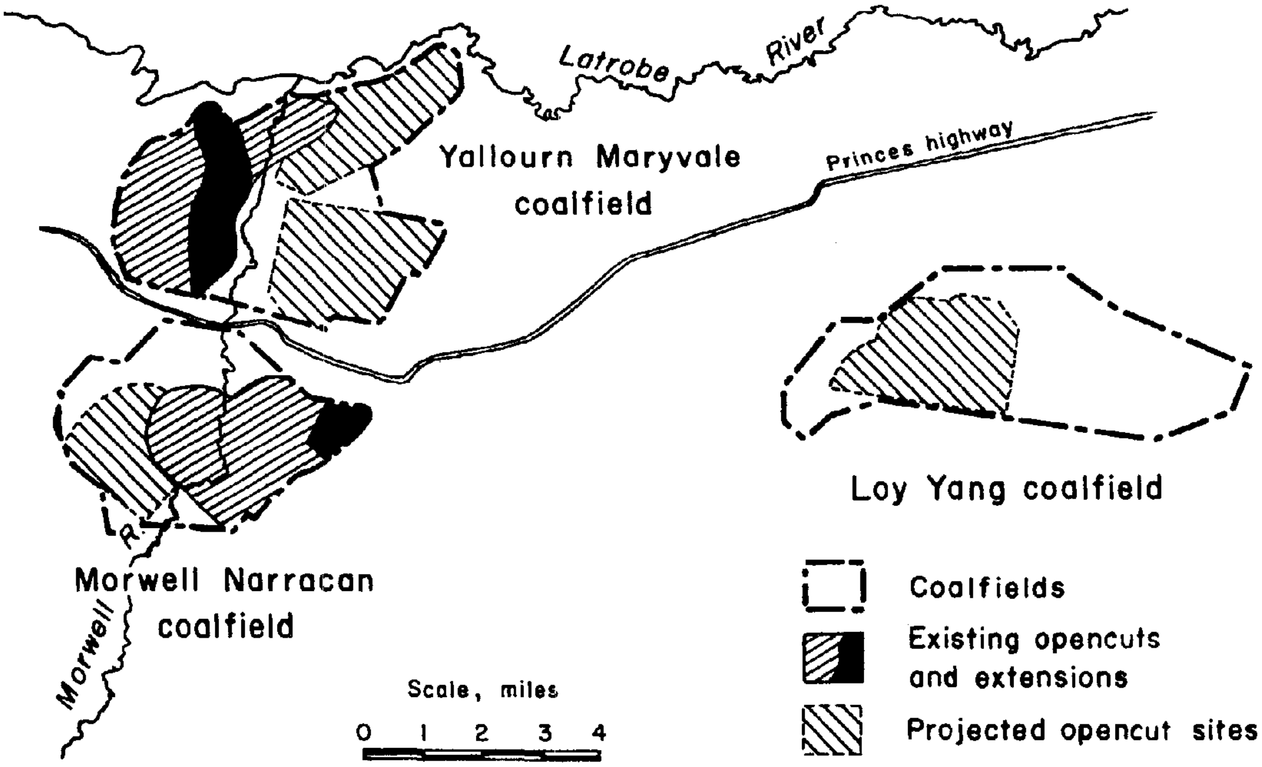

FIGURE 75. - Brown coal deposits of the Latrobe Valley.

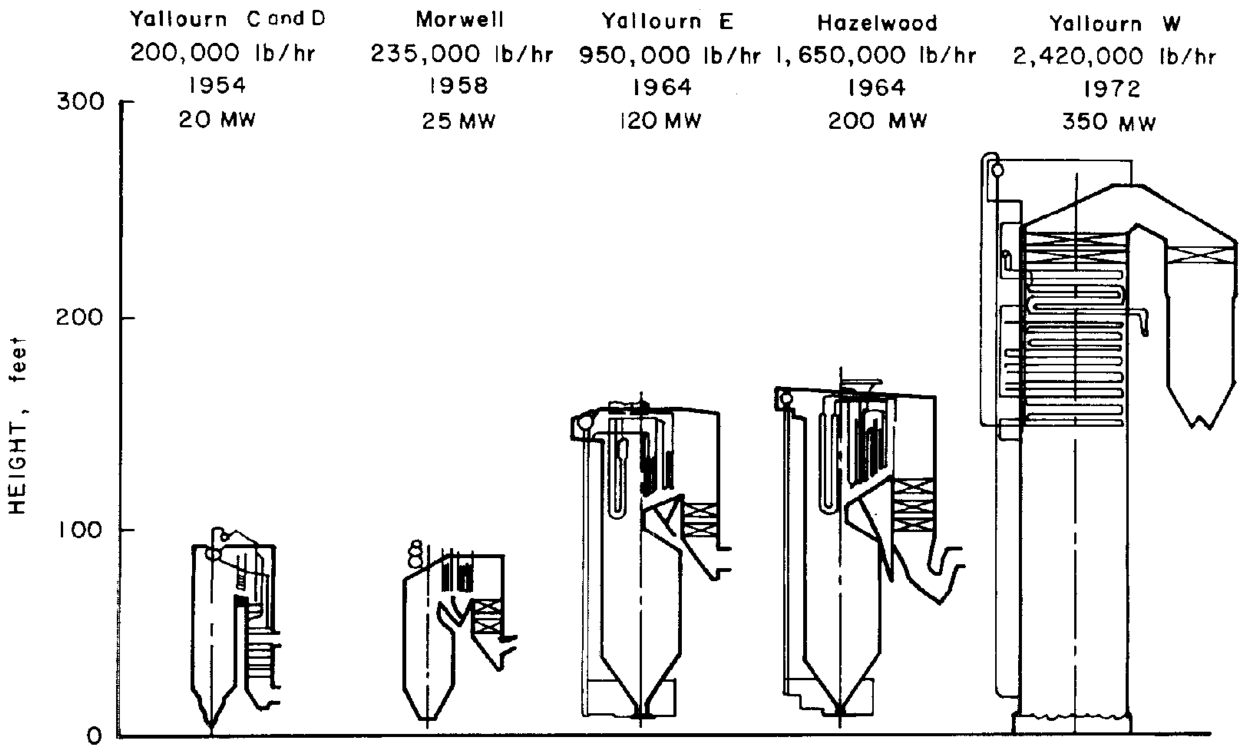

FIGURE 76. - Comparative sizes of Latrobe Valley boiler plants fired with pulverized coal.

were equipped extensively for on-load cleaning with steam soot blowing in the furnace, superheater, and economizer regions. Shot cleaning was used in the economizers of the smaller boilers. Equipment and practices were developed to enable the boilers to operate between annual overhauls without an intermediate off-load cleaning outage, but this availability was achieved at the expense of boiler efficiency. In 1958, very severe fouling was encountered with the first use of coal from the Morwell Opencut. Boiler deposits from Morwell coal formed more rapidly and (particularly in furnaces and superheaters) were much more difficult to remove than those from Yallourn coal. The first 200-MW boiler at Hazelwood Power Station could be operated initially for less than 500 hours before cleaning although 8,000 hours without recourse to off-load cleaning had been specified. A development that has contributed significantly to the alleviation of this problem has been the application of water soot blowing to the furnace walls of the boilers.

Acknowledgment

This paper is published with the permission of the State Electricity Commission of Victoria, Australia, and the authors acknowledge the contributions made by their colleagues in providing information and in preparing the material it contains.

Boiler Operation

Combustion Stability

The problem of unstable combustion in mill-fired boilers was first encountered when the Yallourn C boilers were commissioned. The fuel used has a normal moisture content of about 67 pct, which is considerably higher than that of the German coals for which the firing equipment had originally been designed. Steam raising, running the turbine up to speed, and synchronizing were achieved using auxiliary burner equipment that operated on pulverized coal with approximately 15 pct moisture. The main mills were then brought into service, but ignition was lost when the auxiliary firing was shut down with the furnace alternately blacking out completely and then reigniting with heavy pressure surges. To keep the plant in commercial operation, it was necessary to run auxiliary firing equipment continuously to maintain ignition on the main burners. Even when operating in this manner, excessive furnace pulsations were still experienced.

Development of Combustion Systems

Different burner designs were tried as successive boilers were commissioned, and the first modification to improve conditions was the installation of large areas of refractory around the burners themselves. Refractory "belts" were subsequently fitted at burner level around the furnace of the four boilers then in service, and for the first time no auxiliary firing was necessary, although furnace pulsations were still excessive. Finally, cyclones were fitted in the fuel ducts to enrich the fuel/gas mixture delivered to the lower part of each burner. With an enriched mixture containing about 80 pct of the fuel and one-third of the conveying gas introduced through the lower half of each burner and the remainder admitted through the upper half, a strong stable flame was obtained at the lower burner and satisfactory operating conditions could be maintained. All boilers in C station and in D station were equipped with these separators, and it was found possible to remove the refractory previously installed.

Investigations leading to the solution of the combustion instability problem for Yallourn C and D boilers have been described by Roberts[53] and Kennedy and Warren.[54] This fuel separation system, pioneered at Yallourn, has since been adopted in plants overseas as an effective technique when dealing with brown coals that are difficult to burn.

The 120-MW boilers of the Yallourn E Station were equipped with exhausters capable of varying the gas/fuel ratio of the mixture entering the furnace; cyclones for further enriching the mixture were not provided. However, when

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[53] Roberts, F. H. Pulverized Brown Coal for Power Generation in Victoria. Proc. 2d Conf. on Pulverized Fuel, London, England, November 1957, Inst. Fuel, Paper 15, pp. D1-D10.

[54] Kennedy, W. B., and D. R. Warren. Boiler Furnace Combustion of Raw Brown Coal. Commonwealth Australia, Dept. of Supply, Aeronautical Res. Lab. Tech. Memo. A.R.L./M.E. 153, March 1956, 6 pp.

burning wetter than average coal, fairly wide fluctuations in furnace pressure were experienced with these boilers, and auxiliary firing was used fairly frequently to stabilize combustion. Angled plates fitted in the fuel ducts above the exhausters resulted in an improvement by changing the distribution of fuel between the two openings of each burner group; "concentrators" of this type were eventually fitted to all ducts on both boilers. A test carried out with wetted coal indicated that combustion stability should be satisfactory with coals of moisture contents up to approximately 71 pct.

However, further difficulties were experienced with fluctuating furnace pressures, and loss of ignition occurred on a number of occasions. During one period of operation, it was found necessary to avoid coal from the base of the Yallourn seam that could have relatively high ash contents; ultimately it was decided to install cyclone separators similar to those developed for C and D stations. This modification resulted in significant improvements in boiler operation, but it has not completely eliminated periods of instability; some flame-outs in E station have occurred that cannot be related to changes in moisture or ash contents of the coal. In contrast, combustion instability has not occurred in power stations supplied with the 63-pct-moisture Morwell coal.

Coal Property Investigations

Because of the continuing trouble experienced with Yallourn E and the prospect of the 350-MW Yallourn W boilers operating on the same Yallourn coal, intensive investigations are being conducted on the coal itself to identify the coal properties that contribute to combustion instability. These studies have ranged from the ignition of single particles of brown coal to tests in a 75-lb/hr experimental furnace.

A study of the ignition of single particles of brown coal in a hot gas stream[55] has established, both from a mathematical model and experimentally, that ignition is affected by such physical factors as particle size, gas temperature, oxygen partial pressure, and moisture content. However, with these physical factors controlled at set levels, different Latrobe Valley brown coals still display wide variations in ignition behavior. Morwell coals are found to ignite more readily than Yallourn coals, and some areas of coal in the Yallourn Opencut with particularly poor ignition characteristics have been detected. Samples from bores in an undeveloped field (Loy Yang) were tested for comparison and found to be generally less reactive than the Morwell sample but more reactive than Yallourn samples.

These findings have been duplicated with a second bench-scale test that involves timing the ignition of a 0.5-gram sample of milled coal placed in a muffle furnace at 500° C. This simple test has been used for extensive surveys of the variation in ignition characteristics in the Yallourn Opencut.

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[55] Thomas, G. R. Ignition of Brown Coal Particles. M. Eng. Sc. Thesis, University of Melbourne, Melbourne, Australia, 1970, 138 pp.
Thomas, G. R. Ignition of Latrobe Valley Brown Coals: Single Particle Studies. State Electricity Commission of Victoria, Sci. Div., Rept. No. 268, 1972, 10 pp.

Investigations in the 75-lb/hr pulverized-fuel experimental furnace at the Herman Central Scientific Laboratory (H.C.S.L.)[56] of the SECV have shown that furnace pressure fluctuation and effective flame radiation temperature can be used as measures of flame stability, and correlations obtained with water addition to the flame are consistent with the data obtained in early trials on Yallourn C Station boilers by Roberts[57] (fig. 77). The stability of the flame in the experimental furnace to water addition was dependent on coal source (fig. 78) and also decreased with increasing fuel particle size; Morwell flames were more stable than those of Yallourn with Loy Yang intermediate between the two. These observed relative flame stabilities support the findings of the bench-scale ignition tests and are generally consistent with the operating characteristics of commercial boilers burning Yallourn and Morwell coals.

It has been shown[58] that coal source is a factor that influences the particle size and moisture content of the pulverized fuel resulting from the milling and drying process. Thus the milling and drying characteristics of coal can also influence combustion stability.

Ignition and flame stability investigations have been extended in an attempt to explain the poor ignition phenomenon and identify areas of poorly igniting coal within the Yallourn Opencut. It has been bound that the ignition characteristics are influenced by the inorganic constituents and the petrology of the coal in a particular seam. Experiments with ion-exchanged samples of coal have shown that some exchangeable cations in the coal catalyze ignition reactions, and others appear to have an inhibiting effect. Lithotype identification of the samples studied has established that, within a particular seam, the occurrence of poorly igniting coal is confined to the darker "lignitic" bands of coal; the lighter colored "earthy" brown coals have better ignition characteristics. The darker coal also has the higher moisture content[59] so that combustion instability problems in Yallourn power stations are likely to be aggravated when the boilers are being supplied with a high proportion of the darker coal. Unfortunately, identification of this banding by color is only possible after the coal face has been exposed for some weeks.

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[56] Anderson, B. The Effect on Combustion Stability of Water Additions to Pulverized Brown Coal Flames. State Electricity Commission of Victoria, Sci. Div., Rept. No. 270, 1973, 7 pp.

[57] Work cited in footnote 53.

[58] McIntosh, M. J. The Effect of Drying on the Breakage of Brown Coal. State Electricity Commission of Victoria, Sci. Div., Rept. No. 246, 1971, 21 pp.
McIntosh, M. J. Prediction of Brown Coal Drying in a Mill-Drying System. State Electricity Commission of Victoria, Sci. Div., Rept. No. 261, 1972, 14 pp.

[59] George, A. M. Some Physical Properties of Brown Coal Related to Seam and Lithotype. State Electricity Commission of Victoria, Exploration and Geol. Div., Petrological Rept. No. 14, 1972, 25 pp.

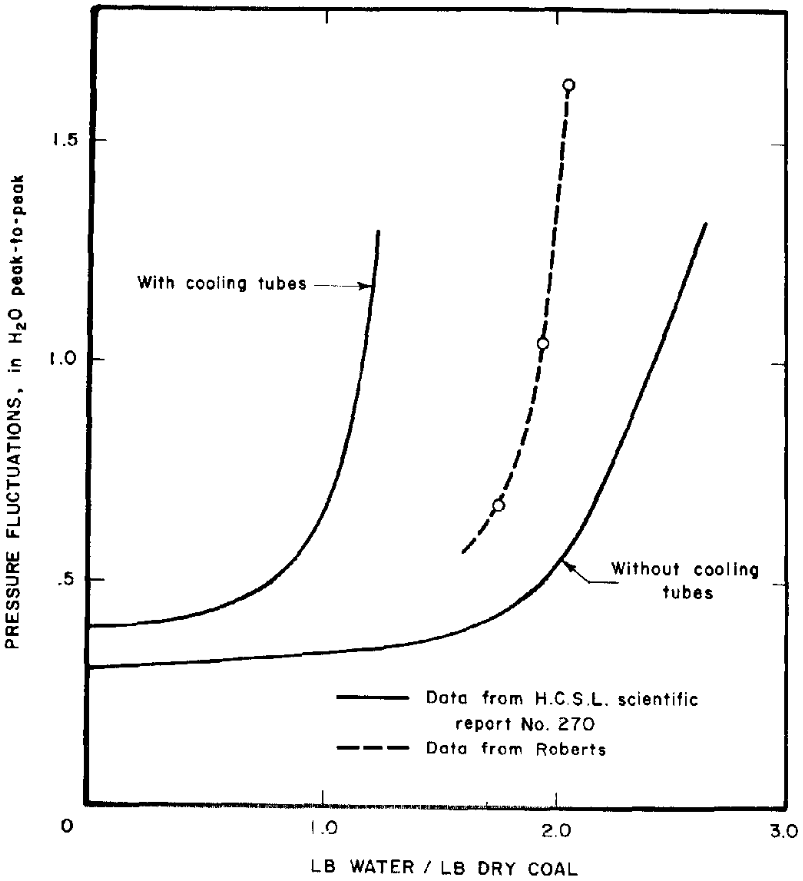

FIGURE 77. - Effect of coal moisture on pressure fluctuations in a combustion chamber.

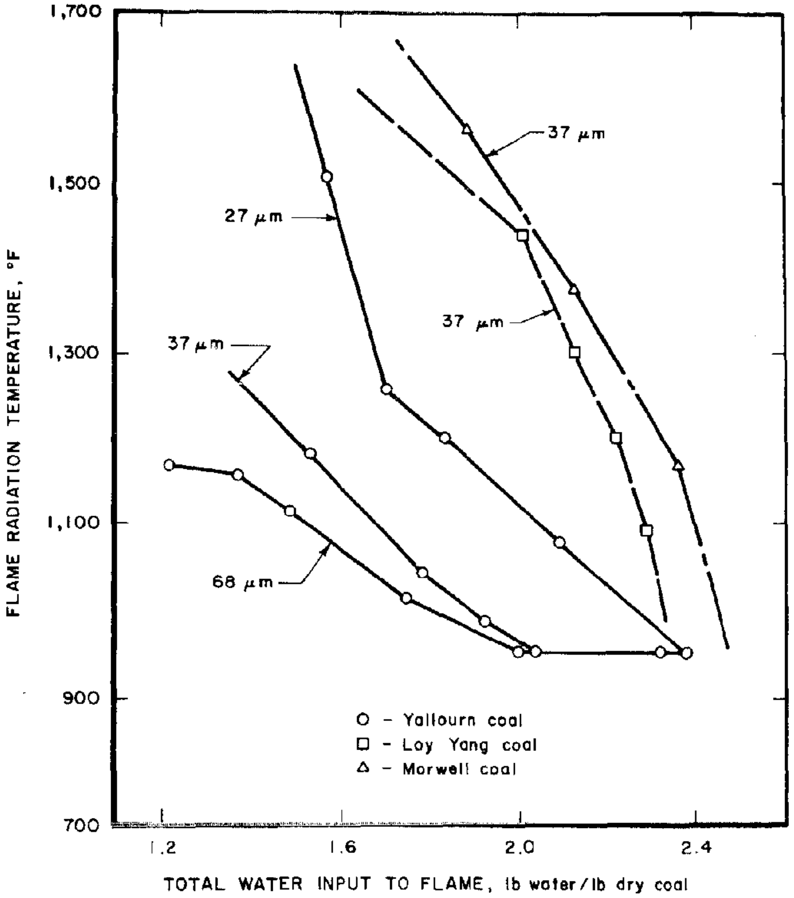

FIGURE 78. - Effect of coal type, particle size, and coal moisture on flame radiation temperature.

Ash Deposition

Although all seams in the Latrobe Valley area show many similarities, there are important differences in moisture content and inorganic constitution. The opencuts at Yallourn and Morwell are in geologically distinct coal seams. In table 17, typical analyses for coal and coal ash are presented.

TABLE 17. - Typical coal and ash analysis[1]

	Yallourn				Morwell			
	High ash		Low ash		High ash		Low ash	
	Coal	Ash	Coal	Ash	Coal	Ash	Coal	Ash
Moisture as received.....pct..	66.0	-	67.0	-	61.0	-	62.0	-
Ash, dry basis...........pct..	2.6	-	1.2	-	4.8	-	2.5	-
Carbon, dry basis........pct..	65.0	-	66.6	-	66.5	-	69.5	-
Hydrogen, dry basis......pct..	4.9	-	4.6	-	4.8	-	4.9	-
Inorganic constituents, pct:								
Iron total..................	.25	13.5	.18	21.6	.50	15.0	.26	14.8
Calcium.....................	.05	2.7	.05	5.8	.54	15.5	.75	42.0
Potassium...................	.008	.4	.005	.5	.02	.5	.003	.3
Chlorine....................	.05	.1	.10	.2	.16	2.1	.05	.1
Sulfur......................	.40	14.1	.26	19.2	.50	26.1	.26	18.7
Silicon.....................	.37	30.7	.06	10.7	.35	15.4	.01	.8
Aluminum....................	.31	22.7	.04	6.5	.05	2.0	.01	.8
Magnesium...................	.20	12.7	.20	27.6	.43	15.0	.28	18.3
Sodium......................	.06	3.1	.07	7.9	.25	6.8	.08	4.4

[1] Coal analysis is expressed as pct element, dry basis; ash analysis, as pct oxide in ash.

Morwell coal contains more ash than does Yallourn coal, and this becomes more significant if the coals are compared in terms of effective heat release because of the difference in moisture contents. However, the more important differences are those related to the proportions of the inorganic constituents.

In general, the Morwell coal contains much higher concentrations of calcium and sodium than does Yallourn coal, and the ratio of calcium to magnesium is dominated by calcium rather than magnesium.[60] In both coals, calcium, magnesium, and sodium are present as carboxylates, but in the Morwell coal some sodium occurs in the form of sodium chloride. Other elements occasionally present as carboxylates include iron, potassium, and aluminum. Iron and aluminum are often uniformly distributed through the coal matter as oxides or hydroxides. Sulfur is mainly present as part of the coal molecule but also

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[60] Durie, R. A. The Inorganic Constituents in Australian Coals--Part 3, Morwell and Yallourn Brown Coal. Fuel, 1961, v. 40, p. 407.

occurs in pyritic form, and chlorine can be attached to the coal molecule or be present as halite.[61]

Extraneous ash consists of quartz, clay minerals, and pyritic inclusions (predominantly marcasite), and the amounts present can fluctuate widely. Although cracks filled with sand create local anomalies in the Yallourn coal, the general level of extraneous ash is lower than that in Morwell coal.[62]

Ash deposits from combustion of the different coals are of the same sulfate-bonded type, but the differing proportions of the inorganic constituents cause marked differences in the quantities and characteristics of the fireside deposits in the boilers.

The average furnace heat flux in a boiler burning Yallourn coal remains practically unaltered at approximately 25,000 Btu/ft^2 hr after 11 to 12 months of steaming.[63] In a boiler fired with Morwell coal, the average furnace heat flux drops from 25,000 to 18,000 Btu/ft^2 hr after 4 to 5 months of operation, and the overall heat transfer coefficient in the secondary superheater decreases from 11 to approximately 5 Btu/ft^2 hr ° F.[64] In a boiler fired with Yallourn coal, the secondary superheater coefficient drops from 11 to approximately 8 Btu/ft^2 hr ° F after 11 months.

Nature of Deposits

The nature of fireside deposits, formed during the combustion of Latrobe Valley coal, has been extensively studied over the last 13 years in programs

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[61] Murray, J. B. The State of Combination of the Major Ash-Forming Constituents in Latrobe Valley Coal. State Electricity Commission of Victoria, Sci Div., Rept. No. 145, 1968, 5 pp.; No. 203, 1970, 8 pp.; No. 224, 1971, 5 pp.; No. 233, 1971, 3 pp.; No. 249, 1972, 4 pp.

[62] Henderson, P. L. (ed.). Brown Coal, Its Mining and Utilization. Melbourne University Press, Melbourne, Australia, 1953, pp. 52-56.

Baragwanath, G. E. Some Aspects of the Formation and Nature of Brown Coal, and of the Behavior of Brown Coal Ash in Water-Tube Boilers, With Special Reference to Victorian Deposits. Australasian Inst. Min. & Met., Proc. (1962) H_2O_2, S. pp. 131-249.

[63] Waring, P. T., and W. J. Morley. The Measurement of Heat Flux in Furnaces and Its Application to Brown Coal-Fired Boilers. Inst. of Eng., Australia, Thermodynamics Conf.--Ind. Applications, Adelaide, Australia, August 1970, 5 pp.

[64] Hall, T. V., and N. F. Bowles. Improvement in Availability of Brown Coal Boilers. Inst. of Eng., Australia, Thermodynamics Conf.--Ind. Applications, Adelaide, Australia, August 1970, 8 pp.

conducted by the Commonwealth Scientific and Industrial Research Organization[65] and by the SECV.[66]

Gas side deposits found in boilers can be divided into three major groups:

1. Deposits exposed to radiant heat,

2. Deposits formed on convection surfaces,

and 3. Loose agglomerates of fly ash.

The first group includes the greatest variety of deposits both in terms of chemical composition and physical properties.[67] Samples range from dense siliceous slags to highly sulfated bonded deposits of various degrees of hardness. Slags and clinker-type deposits are found on the sloping walls of the furnace hoppers and around the burner mouths and gas offtake throats. (Figure 79 is a schematic diagram of a 200-MW boiler from Hazelwood Power Station.) Hard sintered formations have also been observed in the outer layers of deposits collected from the furnace wall tubes at both burner and gas offtake levels although the inner layers are usually softer and more sulfatic. Slags and clinkers have bulk densities between 1.8 and 2.9 (on occasions as high as 3.1), and the density of the particles can vary from 2.3 to 4.3.

Generally, hopper and burner level deposits are richer in iron and silica than those collected at the gas offtake level (both from water wall tubes and gas offtakes), but there are exceptions.

Sulfatic, bonded deposits are particularly noticeable in the higher regions of the furnace and on platen superheaters. In these areas they resemble, in composition and physical appearance, the deposits collected from the secondary and primary superheaters.

The individual particles that form hard sintered deposits have a glassy appearance and are translucent. The particles from sulfatic deposits are

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[65] Grant, K., and J. H. Weymouth. The Influence of the Inorganic Constituents in Victorian Brown Coals When Used in Gas Generators and Industrial Boilers. Inst. of Fuel, Symp. on the Inorganic Constituents of Fuel, Melbourne, Australia, May 1964, pp. 127-138.
Brown, H. R., and D. J. Swaine. Inorganic Constituents in Australian Coals. Inst. of Fuel, Symp. on the Inorganic Constituents of Fuel, Melbourne, Australia, May 1964, pp. 23-44.

[66] Bonafede, G. The Behavior of Inorganic Constituents During the Combustion of Victorian Brown Coals. Internat. Conf. on Fireside Corrosion and Fouling in Thermal Power Stations, Frankfurt/Main, Germany, May 30-31, 1972, 17 pp.

[67] Bonafede, G., and M. Dent. Chemistry of Fireside Deposits Formed During Combustion of Latrobe Valley Coals, Parts 1-4. State Electricity Commission of Victoria, Sci. Div., Rept. No. 154, 1969, 6 pp.; No. 174, 1969, 6 pp.; No. 202, 1970, 5 pp.; No. 209, 1970, 7 pp.

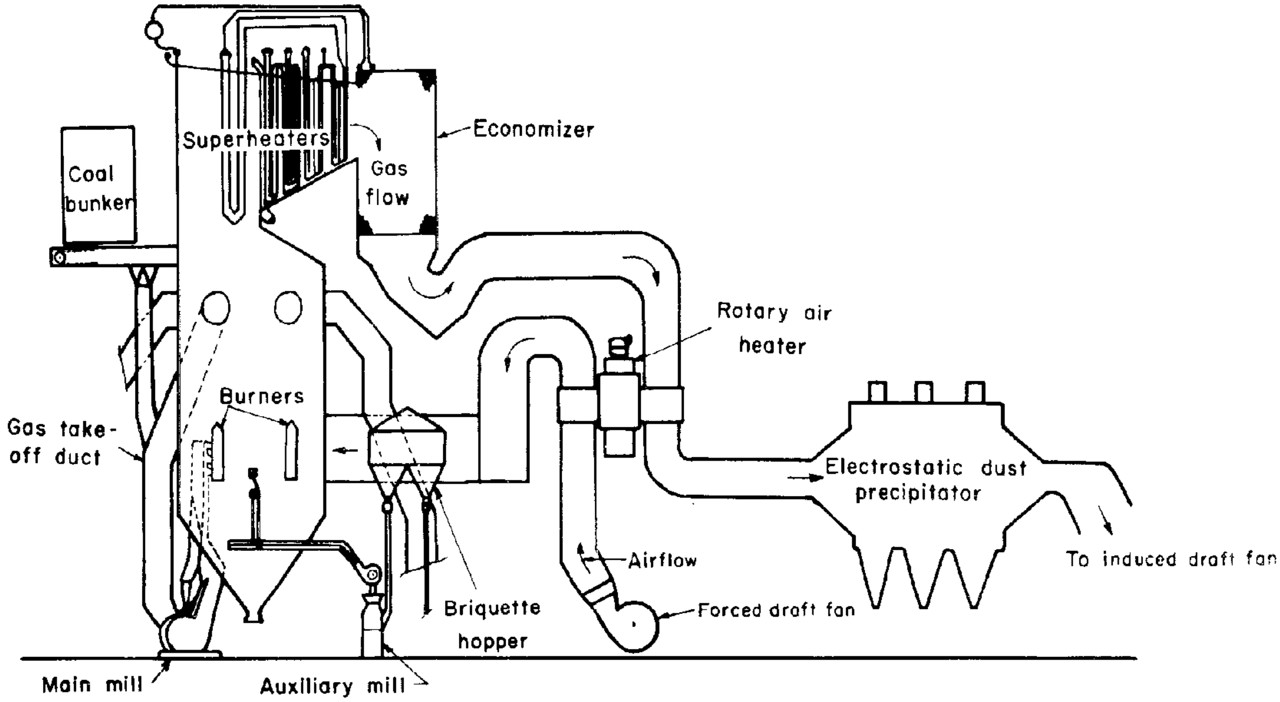

FIGURE 79. - Schematic diagram of a 200-MW boiler, Hazelwood Power Station.

finer, more crystalline and opaque. Chemical analyses of some typical furnace deposits are shown in table 18.

TABLE 18. - Chemical analysis of furnace deposits

	Morwell coal-fired boiler				Yallourn coal-fired boiler			
	Hopper slag	Burner level	Feeder level	Furnace top	Hopper slag	Burner level	Feeder level	Furnace top
Component, wt-pct:								
Sulfate, as SO_3...	1.4	13.1	25.0	34.8	1.2	12.9	18.1	29.4
Silica, as SiO_2...	36.9	10.2	11.4	8.6	58.2	4.2	14.8	4.9
Alumina, as Al_2O_3.	5.8	2.9	2.8	1.5	6.8	4.4	14.2	8.4
Iron, as Fe_2O_3....	31.8	30.2	23.8	9.9	13.0	63.0	31.2	30.7
Calcium, as CaO...	10.4	20.1	16.8	21.5	4.4	1.9	3.2	3.5
Magnesium, as MgO.	10.2	17.3	12.6	15.6	9.9	3.5	14.8	7.7
Sodium, as Na_2O...	3.6	3.8	6.0	6.8	4.2	6.0	5.5	13.8
Potassium, as K_2O.	.2	.2	.2	.2	.3	.3	.3	.6
Chloride, as Cl...	Nil	Trace	.1	.1	Nil	Nil	Nil	Nil

Deposits formed on convection surfaces, such as superheater deposits, vary in bulk density between 1.3 and 2.6 and in particle densities from 2.4 to 3.4. The deposits have a distinct layered structure, and the individual layers themselves are formed by alternating darker and lighter colored bands. The inner layer seldom exceeds 1 to 2 mm (0.040 to 0.080 inch) in thickness. It is soft and consists of submicrometer or 1- to 2-µm crystalline, light-colored particles. Chemically, the inner layer is rich in alkalies and sulfates and can contain considerable amounts of chloride. The outer layers are usually darker and can be hard and strongly bonded. The main bonding agents have been identified as thenardite, anhydrite, and mixed sodium/magnesium sulfates.

Examination of deposits by the scanning electron microscope has shown that individual oxide and siliceous particles are enveloped by sulfate and that the envelopes coalesce into a dense sulfatic matrix (fig. 80).

Economizer deposits are not unlike superheater deposits in chemical composition but are usually more friable and are sometimes present in the form of loosely held agglomerates of fly ash. Air-heater deposits are physically similar to economizer deposits but less sulfated.

Analyses for some of the deposits sampled from convection surfaces are given in table 19. A listing of minerals detected in fireside deposits is given in table 20, together with an indication of their significance in different locations.

During a period of boiler operation, the chemical and physical properties of deposits undergo noticeable changes. The main factors responsible for these changes are (1) continuous sulfation together with some formation of ferrites and silicates and (2) changes in surface and gas temperatures leading to sintering and smelting, and also to evaporation and the diffusional migration of volatile compounds.

The consolidation of furnace deposits is associated with changes in chemical and mineralogical properties, but similar changes do not accompany the hardening and increase in density of superheater deposits.

TABLE 19. - Chemical analysis of superheater, economizer, and air heater deposits

	Morwell coal-fired boiler				Yallourn coal-fired boiler			
	Secondary superheater	Primary superheater	Economizer	Air heater	Secondary superheater	Primary superheater	Economizer	Air heater
Component, wt-pct:								
Sulfate, as SO_3....	38.8	43.7	38.5	25.9	37.6	42.1	36.5	19.2
Silica, as SiO_2....	4.0	4.2	3.6	7.1	8.1	6.5	3.6	19.4
Alumina, as Al_2O_3.	1.0	.9	.5	2.0	9.5	8.3	12.7	10.4
Iron, as Fe_2O_3.....	6.0	5.1	5.1	7.9	16.0	13.7	13.5	21.6
Calcium, as CaO....	23.1	17.7	24.3	29.9	4.6	4.7	3.8	5.8
Magnesium, as MgO..	16.5	12.4	15.4	21.3	10.7	12.7	14.8	20.7
Sodium, as Na_2O....	10.5	16.2	10.1	4.9	12.4	10.7	14.2	3.3
Potassium, as K_2O..	.2	.7	.3	.2	1.1	1.2	1.3	.2
Chloride, as Cl....	.2	.3	.3	1.1	Trace	Trace	Trace	.05

TABLE 20. - Minerals detected in fireside deposits

Mineral	Chemical formula	Main bulk of deposits					Inner layer all locations	Fly ash
		Furnace		Superheater	Economizer	Air heater		
		Bottom	Top					
Oxides:								
α-quartz.............	SiO_2.........	[1] (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	O (M,Y)	F (M,Y)
Cristobalite..........	SiO_2.........	O (M,Y)	O (M,Y)	-	-	-	-	-
Coesite..............	SiO_2.........	O (M,Y)	-	-	-	-	-	-
Periclase............	MgO..........	O (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	O (M,Y)	F (M,Y)
Lime.................	CaO..........	O (M,Y)	F (M)	F (M)	F (M)	F (M)	O (M,Y)	F (M)
Magnetite............	Fe_3O_4........	[1] (M,Y)	F (M,Y)	O (M,Y)	O (M,Y)	O (M,Y)	-	O (M,Y)
Haematite............	Fe_2O_3........	[1] (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)
Sulfates:								
Anhydrite............	$CaSO_4$........	O (M,Y) -	F (M) O (Y)	[1] (M) O (Y)	[1] (M) O (Y)	[1] (M) O (Y)	F (M) O (Y)	[1] (M) O (Y)
Thenardite...........	Na_2SO_4.......	O (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	F (M,Y)	[1] (M,Y)	F (M,Y)
Magnesium sulfate.....	$MgSO_4$........	O (Y)	O (Y)	F (Y)	F (Y)	F (Y)	F (Y)	F (Y)
Ferric sulfate........	$Fe_2(SO_4)_3$....	-	-	-	-	O (M,Y)	-	-
Other salts:								
Calcite..............	$CaCO_3$........	-	-	-	-	O (M)	-	O (M)
Halite...............	NaCl.........	-	-	-	-	O (M)	O (M)	F (M)
Ferrites:								
Calcium ferrite.......	$CaOFe_2O_3$.....	F (M) O (Y)	F (M) O (Y)	O (M,Y) -	O (M,Y) -	O (M,Y) -	- -	O (M,Y) -
Magnesium ferrite.....	$MgOFe_2O_3$.....	F (M,Y)	F (M,Y)	O (M,Y)	O (M,Y)	O (M,Y)	-	O (M,Y)
Silicates:								
(a) Olivines:								
Monticellite........	$CaMg(SiO_4)$...	F (M,Y)	O (M,Y)	-	-	-	-	-
Forsterite..........	$Mg_2(SiO_4)$....	F (M,Y) -	F (M,Y) -	F (Y) O (M)	F (Y) O (M)	F (Y) O (M)	- -	F (Y) O (M)
Fayalite............	$Fe_2(SiO_4)$....	F (M,Y)	O (M,Y)	-	-	-	-	-
Kirschsteinite......	$CaFe(SiO_4)$...	O (M,Y)	-	-	-	-	-	-
(b) Pyroxenes:								
Diopside............	$CaMg(Si_2O_6)$..	F (M,Y)	O (M,Y)	-	-	-	-	O (M,Y)
Enstatite...........	$MgSiO_3$.......	F (M,Y)	O (Y)	O (Y)	O (Y)	-	-	O (Y)
(c) Melilite group:								
Akermanite..........	$Ca_2Mg(Si_2O_7)$.	F (M,Y)	-	-	-	-	-	-
Gehlenite...........	$Ca_2Al_2SiO_7$...	O (M,Y)	-	-	-	-	-	-
Kilchoanite.........	$Ca_3(Si_2O_7)$...	O (M,Y)	-	-	-	-	-	-
(d) Plagioclase group:								
Anorthite...........	$CaAl_2Si_2O_8$...	O (M,Y)	-	-	-	-	-	-

F = Frequently detected in considerable quantities.
O = Occasionally detected as a minor component.
M = Detected in Morwell coal-fired boilers; Y = Detected in Yallourn coal-fired boilers.

[1] Major component.

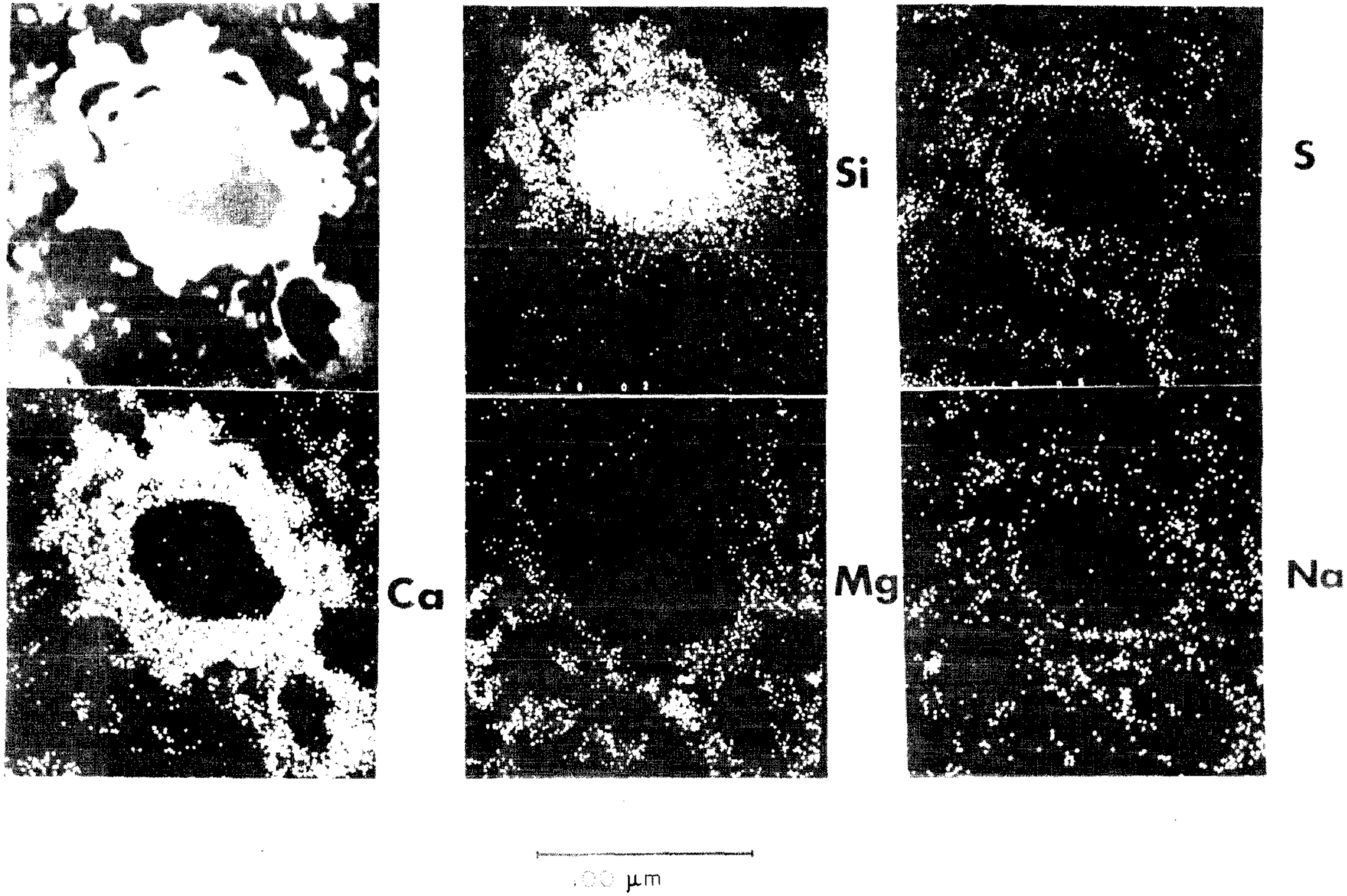

FIGURE 80. - Element distribution in a superheater deposit particle by electron microprobe analysis.

An important factor in the consolidation of superheater deposits is migration of sodium salts.[68] Examinations by scanning electron microscopy and the electron probe microanalysis of soft superheater deposits reheated in a laboratory muffle furnace have shown the existence of dense sodium-rich layers and the consolidation of the structure when this layer is fused (fig. 81). The so-called "outer skin" observed on many freshly sampled superheater deposits resembles closely, in composition and appearance, the inner layer and was probably formed on cooling by the migration of sodium salts from the core of the deposit. If fireside deposits are not removed during a shutdown, this sodium-rich outer layer will soften and melt when the boiler is restarted causing a dense layer within the deposit and providing a sticky surface that will retain impacting fly ash particles.[69]

In 1966-67 the nature and extent of fireside corrosion of superheater tubes in Latrobe Valley boilers was investigated.[70] The observed corrosion rates varied between 0.007 and 0.015 mg/cm^2 hr over an extended period, and generally corrosion of ferritic and austenitic superheater tubes did not appear to be excessive for superheaters conveying steam at temperatures below 1,050° F. (Note that 0.015 mg/cm^2 hr is equivalent to 0.008 in per 10,000 hours.)

Low-temperature corrosion has occurred on a number of occasions in air heaters, but when the metal temperatures are maintained above 300° F, the corrosion rate is negligible. Special materials such as vitreous enamel coatings have also been used to improve corrosion resistance at low temperatures.

Boiler Cleaning Practices

Because steam soot blowing was found to be unsuccessful in cleaning deposits from the water tube walls of the boilers at Hazelwood Power Station, water blowers (water jet deslaggers) were introduced some 6 years ago. Each boiler, generating 1,650,000 pounds steam per hour at 1,600 psig and 1,055° F, is now equipped with 42 water blowers that effectively clean 50 pct of the furnace area. The boilers are fired with Morwell coal and consume some 6,000 tons of raw coal per day.

There are 38 single-nozzle (4-mm diameter) Babcock & Wilcox water blowers and 4 retractable oscillating water blowers of SECV design installed in the furnace walls and operated in an automatic sequence. Figure 82 indicates the locations of all soot blowers on a boiler and gives detailed information on make, model, nozzle size, operating pressures, and speeds. Each step of the sequency starts three to four blowers and an adjustable timer spaces the steps

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[68] Bartlett, D. J., and G. Bonafede. The Migration of Alkali Salts in an Inert Porous Medium. State Electricity Commission of Victoria, Sci. Div., Rept. No. 178, 1969, 6 pp.

[69] Kiss, L. T., L. K. Ingram, and K. A. Herd. Preliminary Study of Off-Load Boiler Cleaning at Hazelwood Power Station. State Electricity Commission of Victoria, Sci. Div., Rept. No. 257, 1972, 12 pp.

[70] Bonafede, G., and A. R. Ellery. Fireside Corrosion of Superheater Tubes in Latrobe Valley Boilers. State Electricity Commission of Victoria, Sci. Div., Rept. No. P.R.-3, 1968, 22 pp.

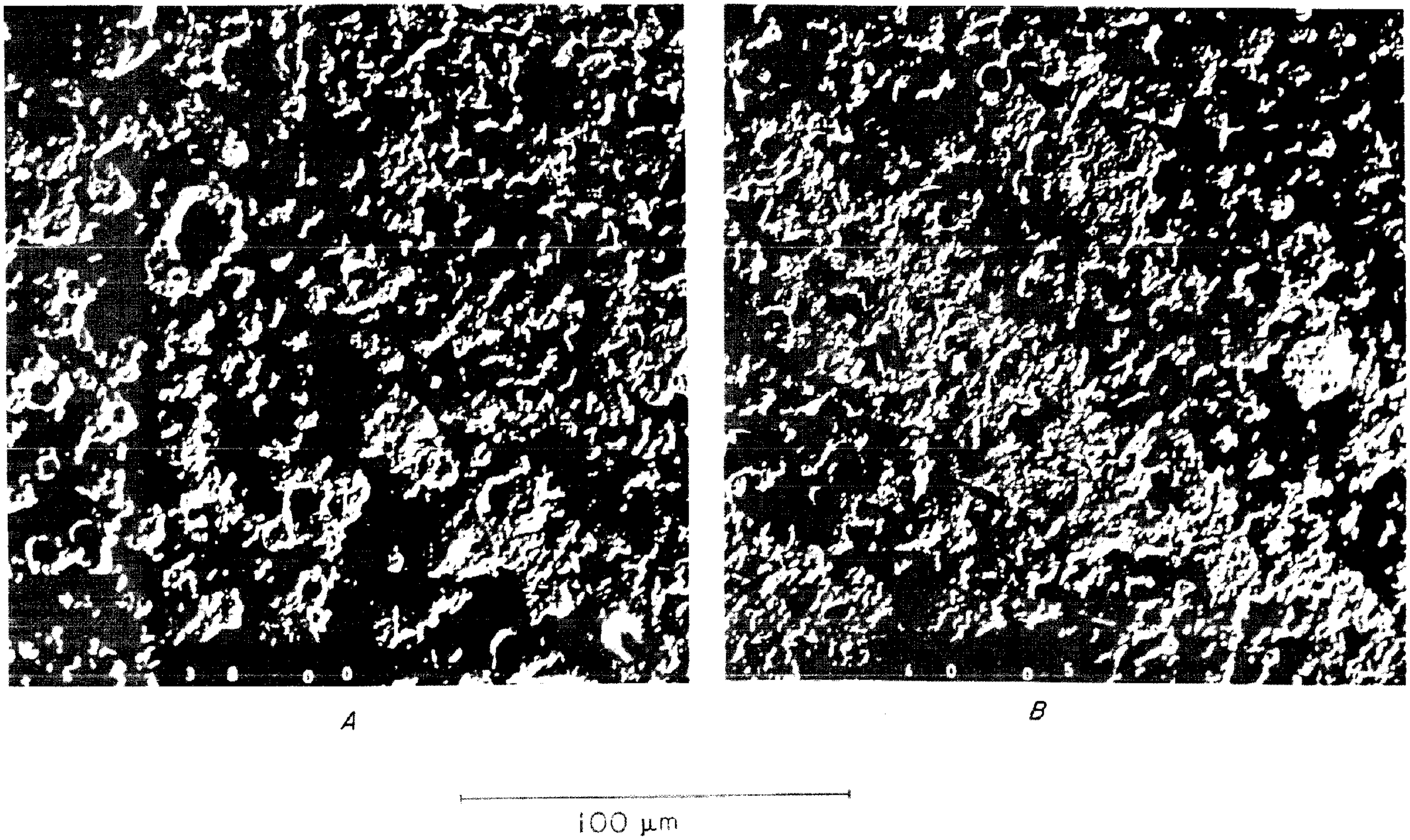
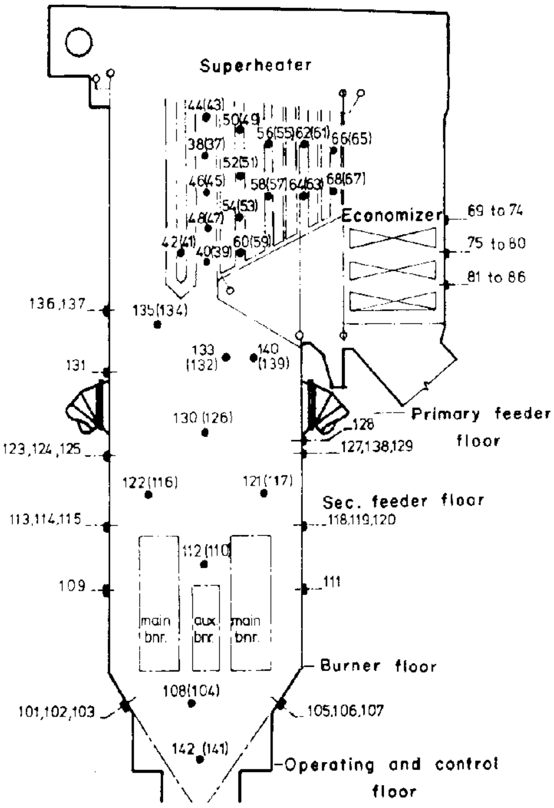


FIGURE 81. - Superheater deposit surface by scanning electron microscope. *A*, Initial conditions; *B*, after reheating.



Sootblower type	Blower number	S/B Operating pressure (psi)	Remarks
Furnace (water)	101 to 142	100-180	Babcock & Wilcox: Type, Mk2 Mk3 Travel rate, 6.5 6.5 in/min Rotation, 1.5 3.5 rpm Water consumption, 5 gpm SEC oscillating type: Travel rate, 3 fpm Oscillation, 6/min Minimum distance from tubes, 6 ft Nozzle size, S/B's 101-112, 4-mm diam 113-142, 3-mm diam
Platen (steam)	41 to 42	200	Diamond rock type IK300 25 ft travel, two nozzles 7/8- or 1-in diam
Secondary superheater (steam)	37 to 60	350-380	Diamond rock type IK300 25 ft travel, two nozzles 7/8- or 1-in diam
Primary superheater (steam)	61 to 68	170-200	Diamond rock type IK300 25 ft travel, two nozzles 7/8-in diam
Economizer (steam)	67 to 74 75 to 86	250-280 180	Diamond rock type IK300 20 ft travel, two nozzles 7/8- or 1-in diam

FIGURE 82. - Soot blower locations and equipment detail as installed on a 200-MW boiler, Hazelwood Power Station.

so that the full sequence is executed during one 8-hour shift; the water blowers situated below the burners are operated twice and the blowers above the burners only once per shift. The delay between the steps of the sequence provides the possibility for manual operation of any blower when deposit buildup in that region requires more frequent blowing. If a consistently stronger cleaning effect is required, the soot blowers can be equipped with heads having two nozzles, each of 4-mm diameter.

Measurements have shown (fig. 83) that the heat flux remains practically constant for the surface cleaned by water blower compared with a 75-pct deterioration in the heat flux after 500 hours for an uncleaned surface.

The tangent tube construction of the boiler furnaces, with no membrane walls and very limited use of finned tubes, was undoubtedly a factor in the successful application of on-load water washing. However, a number of difficulties were experienced during the conversion from steam soot blowing. The long retractable oscillating water blowers, which are installed in the side walls of the furnace hopper and immediately below the "nose" on the rear wall of the furnace (141, 142, and 139, 140, respectively, on fig. 82), have proved to be very successful but have resulted in some water-side circulation problems. The blower oscillates through 90° and four jets are installed on the head, the two forward-facing jets being intended to clean surfaces past which the blower travels and the two backward-facing jets to clean the vertical side wall through which the blower protrudes. The rear jets also clean the sloping walls of the furnace as the blower advances towards the limit of its travel. Some of the wall tubes that are cleaned by the oscillating water blowers form the roof of the furnace and the superheater enclosure, and it was found that a number of conditions could arise that significantly reduced the natural circulation in these tubes.

Tube temperature measurements have been made from the back of tubes in the furnace roof, and circulating flow rates with tubes have been determined by pitot measurements. It has been found that excessive quantities of water from a blower can reduce natural circulation sufficiently to result in a 45° F increase in tube metal temperature. Blower design changes were made to minimize the excessive cooling when the water jets impinged on the walls near a corner. The simultaneous cleaning of tubes by an oscillating water blower in

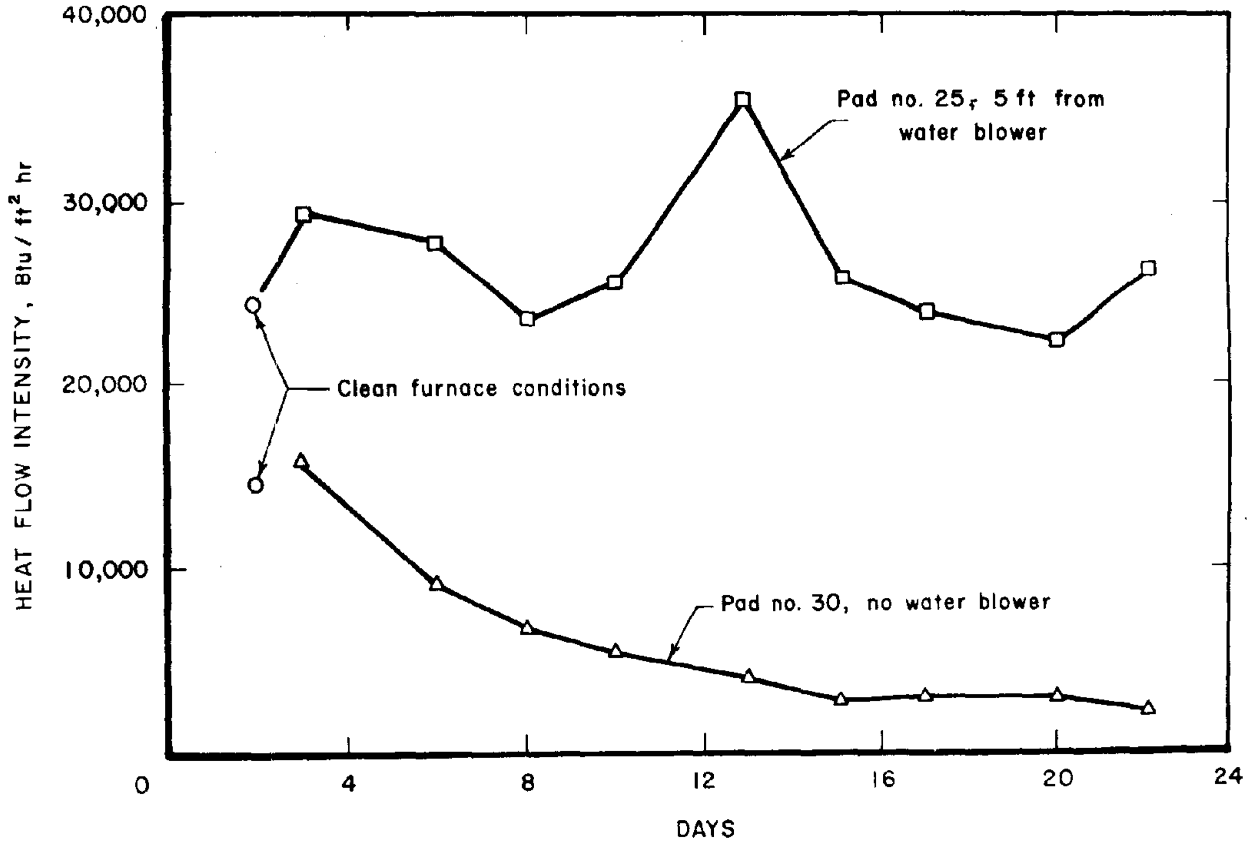


FIGURE 83. - Heat flux variation for washed and uncleaned surfaces.

the superheater area resulted in an increase in tube metal temperature of 135° F and is believed to have caused three tube failures. To overcome this problem, the water blower head was redesigned with a 3-mm-diameter nozzle, and the automatic control sequence was altered to prevent the simultaneous operation of soot blowers affecting the same tube.

The alinement of the two rear nozzles on an oscillating blower so that both jets strike the same tube can stop the natural circulation within that tube, and it is possible that flow could reverse. When monitoring circulation rate on a tube during the operation of an oscillating water blower, a no-flow condition was observed that persisted after the blower completed its operating cycle. The tube metal temperature indication continued to rise steadily under these conditions until, when 135° F above the usual temperature, the operator reduced load by 25 MW. This sudden load reduction permitted the natural circulation in the tube to reestablish and the tube wall temperature returned to its normal level.

Experience at Hazelwood Power Station has shown that the single rotary water blower (equipped with one or two nozzles) is unlikely to influence the natural circulation in a boiler, even in locations where the heat flux is low, but the simultaneous operation of two adjacent water blowers should be avoided.

Some cracking has been observed on the water-washed sections of tubes in the vicinity of tube butt welds or attachment welds. Accelerated thermal fatigue investigations on tube samples have failed to reproduce this cracking after more than 30,000 cycles, and it is considered that restraint effects must be significant in the boiler. Further laboratory-scale investigations are proceeding, and the operation of the water blowers in the power station is being developed toward minimizing the quenching effect but maintaining effective cleaning of the heat transfer surface.

In the superheater regions conventional steam soot blowing is used; rack-type Diamond soot blowers with two 7/8-inch-diameter nozzles are installed in the platen and secondary and primary superheaters, and are operated in pairs. In the platen and the primary superheaters, steam at a pressure of 200 psig is used, and in the superheater the steam pressure is 350 psig. Steam supply for the soot blowers is taken from the inlet to the secondary superheater at 1,650 psig and 800° F. The installation on four boilers of a steam pressure reducing station to permit distribution to the soot blowers at 600 psig has considerably reduced maintenance costs. Soot blowing is controlled by an automatic sequencing unit with provision for any soot blower to be operated out of sequence. Normal operation provides for superheater soot blowing twice per shift.

Under typical operating conditions, the flue gas temperature at the inlet to the economizer would rise by approximately 150° F per shift if the superheater soot blowers were not operated. Because the factors currently limiting the length of boiler operation without off-load cleaning are the maximum permissible gas temperature at the inlet to the economizer (1,300° F) and the lowest acceptable final steam temperature (1,000° F), satisfactory soot blower operation is essential. A comprehensive system of field reporting and maintenance priorities provides for daily attention to problems not covered by the normal programed maintenance. The effectiveness of the soot blowers in maintaining the heat transfer capacity of the superheaters is monitored by calculating heat transfer coefficients for the separate superheating stages to determine trends.

Water washing of superheaters is regarded as an interesting but doubtful prospect.[71] Laboratory investigations are being conducted on low-alloy superheater steels (1 pct Cr, 1/2 pct Mo and 2-1/4 pct Cr, 1 pct Mo) to determine sensitivity to thermal fatigue. Indications to date are that cracking does not occur in the 1 pct Cr, 1/2 pct Mo alloy at metal temperatures up to 500° C (930° F) and up to 16,000 quench cycles, but there is slight cracking after 30,000 cycles; above 500° C, cracking occurs independent of the number of cycles.[72] The two steels investigated exhibit similar tendencies, but the cracking is more severe in the higher alloy steel.

To prevent the overheating of superheater tubes by excessive soot blowing at a time when the furnace heat transfer is restricted, the unit controller is aided by the indication in the control room of metal temperature on 30 selected tubes in the platen and secondary and primary superheaters. During soot blowing, the metal temperature of the tubes cleaned will normally increase by some 45° to 55° F, but this increase will be greater if the steam flow in the tube is low. Consequently, soot blowing is restricted during startup or low-load conditions. The metal temperature monitoring system installed was developed from temperature measurements made on some 120 tubes and early experience with superheater tube failures. The temperature measurement is made in the boiler vestibule and has been found to be convenient and reliable and also useful as a steam temperature indication. The temperature limits applicable to different groups of tubes are indicated on the temperature recorders, and the operating staff take decisions relating to the extent of soot blowing from the control room.

Efforts are being made to limit the peak in the distribution of gas temperature across the boiler in the region of the superheaters. It has been established that higher temperatures increase the rate of deposition and cause deposits to be denser and more difficult to remove from the boiler during off-load cleaning.[73] The temperature distribution in this region is affected by the combination of mill-burner units that are in service at any time (seven out of eight being required to maintain full load) and by the firing

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[71] Ellery, A. R. On-Load Water Washing of Superheater Tubes To Improve Boiler Availability, Part 1. Initial Study at Morwell Power Station. State Electricity Commission of Victoria, Sci. Div., Rept. No. 187, 1969, 8 pp.
Newton, J. D. On-Load Water Washing of Superheater Tubes To Improve Availability, Part 2. Interim Report on the Metallurgical Effects of Two Years Operation in No. 7 Boiler, Morwell Power Station. State Electricity Commission of Victoria, Sci. Div., Rept. No. 230, 1971, 5 pp.

[72] Newton, J. D. Thermal Fatigue Cracking in 1 pct Cr 1/2 pct Mo Superheater Tube Steels. State Electricity Commission of Victoria, Sci. Div., Rept. No. 265, 1972, 9 pp.

[73] Work cited in footnote 69.

conditions within the furnace (since swirling conditions are generated by the burner system) and is accentuated by the fact that there is no crossover (from the center of the gas path to the side) in the steam flow between the platen and primary superheaters. Some progress has been made in leveling out this temperature distribution by balancing coal feed rate to the mills.

Economizers are of a steaming type and when the boiler is nearing an off-load clean, the steaming rate can exceed 30 pct. Six rack-type, Diamond steam-soot blowers with two nozzles of 7/8-inch or 1-inch diameter are provided for each of the three banks of the economizer. The soot blowers are operated in groups of three in sequence from the top bank to the bottom following the superheater soot blowing cycle. The soot blowers mounted above the middle and bottom bank of the economizers are 16 inches or less above the top rows of tubes and erosion has caused tube failures. Soot blowing was first restricted to once per shift for these lower banks, then on the four boilers equipped with a steam reducing station, the pressure to the soot blowers involved was reduced from 250 psig to 180 psig. At this pressure, once-per-shift operation provided insufficient cleaning, and the sequence was restored to twice per shift. On the other four boilers with steam pressure at 250 psig, the nozzle diameters were reduced to 1/2 inch, and the most affected tubes were protected with sleeves. The effectiveness of the different cleaning conditions was monitored by calculating heat transfer coefficients for the economizer from operating conditions and determining the short- and long-term trends.

Shot cleaning was practiced on the tubular air heaters of the first two boilers until vitreous enameled tube ends were installed to reduce acid dew-point corrosion at the cold tube plant; since then, these air heaters have only been cleaned off-load. The other six boilers have rotary air heaters, each equipped with 12 Babcock & Wilcox, fixed lance, multiport soot blowers and the soot blowing of the air heater is included in the boiler sequence, twice per shift, after the superheater and economizer blowing.

Initial difficulties with high differential pressures across the air heater were caused by the combined effects of plate disintegration and ash deposition. Plate disintegration was found to be caused by vibration under the action of soot blowers using insufficiently superheated steam at unnecessarily high pressures. A reduction in pressure from 250 psig to 180 psig significantly increased plate life, but tests carried out with an experimental Diamond IK 110 blower have shown that 90 psig is sufficient for effective cleaning so that further improvement might be possible. Acid corrosion conditions exist for a part of each air heating, heat transfer cycle, but vitreous enamel coating of the plates provides good protection.

Boiler operation has now improved to the state where off-load cleans are expected after 3,000 to 4,000 hours of operation although abnormalities can force earlier outages. The approach to cleaning is to soak the deposits with water for about 48 hours and then dislodge them with jets of water from hand-held fire nozzles. Some hand cleaning may then be required in the superheater areas but the boiler can be returned to service after 5 to 7 days if no major maintenance is required. The maximum water demand for washing is 70,000 gal/hr, and all the water and ash is sluiced to ash ponds for disposal.

Ash Disposal

Although the ash that forms deposits on the heat transfer surfaces of the boilers creates limiting operational problems, it is only 2 to 4 pct of the total ash in the coal consumed. The distribution of ash in a boiler at Hazelwood Power Station has been estimated as 85 pct, collected by electrostatic precipitator; 11 to 13 pct, collected in furnace hoppers after fallout or removal by on-load cleaning from furnace, superheater, economizer and air heater; and 2 to 4 pct, collected on heating surfaces as deposit and removed off-load.

Typically, then, burning 4-pct-ash coal in the 200-MW boiler, there will be some 75 to 80 tons/day of fly ash from the precipitators, 10 to 15 tons/day of coarser ash or agglomerates from the various ash hoppers on the boiler and about 2 to 4 tons/day added to the deposits within the boiler.

Electrostatic Precipitation of Fly Ash

Electrostatic precipitators were first installed with the 200-MW units at Hazelwood and are also being installed with the 350-MW units under construction at Yallourn W. The 120-MW units at Yallourn that have been in service for 12 years have now been equipped with precipitators, and plans are being developed for the older plant.

Fly Ash Characteristics

The chemical compositions of fly ashes formed from Yallourn and Morwell coals reflect the differences in the original inorganic constituents, but size distributions are very similar. Table 21 gives size distributions and chemical compositions for typical fly ashes sampled from the flue-gas stream with conventional dust sampling equipment. The larger size fractions of fly ash, sampled from the boilers burning Morwell coal, usually contain a considerable proportion of agglomerates formed from light-colored skeletal particles.

Microscopically, the Morwell fly ash consists predominantly of very fine yellow and white crystalline particles, which have been identified as anhydrite, thenardite, periclase, and quartz. It has a relatively low silica content and is less sulfated than most of the deposits. Black magnetic cenospheres ranging in size from 1 mm to less than 66 μm in diameter are frequently observed and consist mainly of magnetite. In contrast the Yallourn fly ash is less sulfated, has higher contents of iron and alumina, a lower calcium-to-magnesium ratio, is darker, less agglomerated, and contains fewer black magnetic cenospheres.

TABLE 21. - Size distribution and chemical analysis of fly ash

	Morwell coal						Yallourn coal					
	Coal ash	Fly ash size, µm					Coal ash	Fly ash size, µm				
		>147	104 to 147	74 to 104	66 to 74	<66		>147	104 to 147	74 to 104	66 to 74	<66
Percentage by weight..	-	4.4	2.3	2.4	2.0	88.9	-	3.4	0.9	2.5	1.5	91.8
Pct loss on ignition..	-	20.0	15.0	3.6	3.3	3.4	-	65.0	62.0	19.2	10.6	8.0
Chemical analysis, wt-pct:[1]												
Sulfate, as SO_3.....	16.5	n.d.	n.d.	n.d.	n.d.	21.4	12.8	n.d.	n.d.	n.d.	n.d.	10.9
Silica, as SiO_2.....	14.2	12.0	18.0	23.4	18.5	5.9	8.8	37.0	37.4	14.9	7.1	3.9
Alumina, as Al_2O_3...	.6	2.8	2.1	2.2	2.4	2.4	53.3	26.0	22.4	17.3	19.7	38.6
Iron, as Fe_2O_3......	14.6	11.1	10.6	9.9	9.0	10.2	8.4	18.2	18.0	16.2	17.1	14.8
Calcium, as CaO.....	23.6	24.0	15.4	18.5	18.8	25.4	2.5	4.0	2.0	4.0	5.7	4.2
Magnesium, as MgO...	19.9	18.7	17.5	13.8	14.5	21.9	9.8	6.0	5.2	19.7	22.4	18.4
Sodium, as Na_2O.....	8.2	13.8	14.4	12.8	12.8	10.4	4.0	5.3	2.6	8.8	10.5	6.2
Potassium, as K_2O...	.3	.9	1.2	.6	.6	.5	.3	.8	.8	.2	.2	.3
Chloride, as Cl.....	2.0	n.d.	n.d.	n.d.	n.d.	4.6	n.d.	n.d.	n.d.	n.d.	n.d.	2.2

n.d. = Not determined.

[1] Corrected for loss on ignition.

The essential conclusion from the examination of electron micrographs of freshly formed fly ash particles is that the coal particles burn out to mainly skeletal and cagelike formations, although some inorganic constituents are released as spherical or irregularly shaped particles and the more volatile sodium compounds are released as vapors.[74] The micrographs in figure 84 show typical partly burnt coal particles and ash particles. The volatilized sodium compounds condense readily on cooled metal surfaces and on solid ash particles and are rapidly sulfated. The branches of the skeletal particles consist of oxides of calcium, magnesium, and iron that can react with extraneous ash and form silicates (for example, forsterite) or can be sulfated by the sulfur oxides present in the gas.

Precipitation Efficiency

The physical and chemical properties of Latrobe Valley brown coal ash appear to be favorable for electrostatic precipitation, and in the existing plant, collection efficiencies of well above 95 pct are achieved. Laboratory trials conducted on coals from different seams show differences in precipitation behavior but indicate that efficiencies of 95 pct or higher can be expected for the collection of fly ash at future power stations.[75]

The main problems encountered in the operation of electrostatic precipitators are corrosion of electrodes and of structural parts, and difficulties experienced in handling the fine, light and fluffy fly ash.

Ash Handling and Disposal

Present Practice

Ash disposal systems used with the boilers burning Latrobe Valley coals consist of hydraulic recirculating systems incorporating an ash settling pond. The greatest operating difficulties encountered with these wet disposal systems are associated with the high water-soluble content of the Latrobe Valley coal ashes: Precipitation of salts from solution causing blockages of valves and pipelines; corrosion-erosion of equipment due to the combined effects of the saline solution and entrained solids; disposal of effluent solutions to maintain a stable concentration of dissolved solids in the recirculating system.

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[74] Bonafede, G., and W. M. Drew. A Study by Electron and Optical Microscopy of Fly Ash Particles From Brown Coal-Fired Boilers, Parts 1 and 2. State Electricity Commission of Victoria, Sci. Div., Rept. No. 181, 1970, 5 pp.; No. 215, 1970, 5 pp.

Bonafede, G., and P. Dimitrakakis. A Study by Electron and Optical Microscopy of Char and Ash Particles Produced During the Rapid Heating of Brown Coal Particles in a Laboratory Furnace. State Electricity Commission of Victoria, Sci. Div., Rept. No. 228, 1971, 11 pp.

[75] Paulson, C. A. J. Investigation of the Electrostatic Precipitation of Fly Ashes From Loy Yang Seam Coal, Australia. Commonwealth Sci. and Ind. Res. Organization, Miner. Res. Lab., Investigation Rept. No. 90, 1971, 7 pp.

FIGURE 84. - Electron micrographs of partly burnt particles (*A* and *B*) and fly ash particles (*C* and *D*).

Typical compositions of the two main kinds of ash and their solutions are given in table 22 and table 23, respectively. The ash from the Hazelwood Power Station, operated on Morwell coal, is sluiced into a settling pond from which the supernatant water containing the soluble salts overflows into a recirculating pond and is then pumped back to a return water reservoir for the cycle to be repeated. A proportion of the water from the ash pond has to be discharged and replaced with fresh general service water to control the salt concentrations in the sluicing water, and a maximum of 500,000 gal/day of saline ash pond water has been diverted. After 15 months the disposal of this saline effluent to an outfall sewer had to be discontinued because of blockages caused by precipitated calcium carbonate. The saline water can contain up to 900 mg/l of calcium in solution, and the main cause of precipitation was the dissolved carbon dioxide in raw sewage.

TABLE 22. - Analysis of ashes from Yallourn and Morwell coals

	Yallourn	Morwell
Sulfate, as SO_3.........pct..	11.4	25.6
Silica, as SiO_2.........pct..	28.7	3.5
Alumina, as Al_2O_3.......pct..	11.5	2.2
Iron, as Fe_2O_3..........pct..	22.6	7.1
Calcium, as CaO.........pct..	6.2	27.3
Magnesium, as MgO.......pct..	14.1	20.4
Sodium, as Na_2O.........pct..	5.2	10.0
Potassium, as K_2O.......pct..	.5	.3
Chloride, as Cl.........pct..	.3	2.5

TABLE 23. - Analysis of ash solutions from Yallourn and Morwell coals

	Yallourn			Morwell			Existing deashing system, Hazelwood
	1	2	3	1	2	3	
Conductivity..mmho/cm.	8	18	50	11	18.5	50	13.4
pH....................	7.5	7.8	8.7	12.6	12.9	13.0	12.5
Calcium, Ca.......ppm.	450	630	600	840	890	700	800
Magnesium, Mg.....ppm.	300	1,020	3,000	n.d.	n.d.	n.d.	7
Sodium, Na........ppm.	1,040	3,420	16,800	1,270	2,450	12,300	2,050
Potassium, K......ppm.	60	200	740	n.d.	n.d.	n.d.	80
Sulfate, SO_4......ppm.	4,640	13,120	42,200	2,930	3,370	16,600	2,100
Chloride, Cl......ppm.	142	370	5,900	570	1,070	4,560	1,080

n.d. = Not determined.

Recent investigations indicate that the formation of objectionable calcium carbonate deposits can be overcome by mixing the saline water with the effluent from a local paper mill prior to discharge into the sewer.[76] The

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[76] State Electricity Commission of Victoria, Latrobe Valley Department. Disposal of Saline Water Effluent. Rept. by the Working Group, February 1972, 5 pp.

paper mill effluents contain 1 to 5 g/l of organic matter of a ligninsulfonate type, which apparently prevents formation of hard calcium carbonate scale by acting as a dispersant or chelating agent.

Another approach to the alleviation of this problem is by carbonating the saline water with flue gas, and this is being investigated on a pilot scale. Excessive amounts of calcium can be precipitated as calcium carbonate and settled out before the saline water is conveyed to the sewer.

At Hazelwood Power Station 10 g/l of total dissolved solids is adopted as the concentration limit within the recirculating system itself because extensive deposits of calcium salts are formed in the return water lines at higher concentrations. This limit is equivalent to a conductivity of 14 to 15 mmho/cm.

The fly ash collected by the electrostatic precipitators on boilers fired with Yallourn coal will be sluiced into a large basin that is the worked-out Yallourn North Opencut. At present the basin holds approximately 2×10 gal of relatively pure water (conductivity 400 μmho/cm), which will be recirculated through the ash sluicing system until conductivity rises to 60 mmho/cm.

A solution of Yallourn ash can be concentrated five to six times more than can a Morwell ash solution because it has a lower calcium concentration and a lower pH and is therefore less likely to form deposits that can cause blockages in pipelines or valves. The limiting figure of 60 mmho/cm has been selected to insure that sodium sulfate, the main salt present in solution, will not precipitate under the most adverse temperature conditions anticipated.

The main problems facing this disposal system for Yallourn ash are the possible seepage of saline water from the basin that could contaminate ground and surface waters, and the disposal of the effluent after the conductivity of the water in the pond reaches 60 mmho/cm in about 15 years.

Ash Disposal for Future Developments

Alternative systems of ash disposal are being investigated to determine the most satisfactory approach in relation to future power generating developments in the Latrobe Valley. Broadly the alternatives are the following: Recirculating hydraulic ash handling as discussed previously; dry disposal of conditioned ash within overburden dumps or worked-out opencut areas, a once-through hydraulic system with a pipeline carrying ash slurry to the ocean (approximately 60 miles).

For dry ash disposal it will be necessary to minimize dust generation in the handling system. One approach to this is to moisten the fly ash with water after which the ash tends to solidify because of the formation of hydrated sulfates and carbonates of calcium and magnesium. Solidification creates some handling problems, but it may prove to be a significant benefit by reducing the extent of the subsequent leaching of salts from dumped ash. Current investigations cover the physical and chemical properties of water-conditioned ash, its behavior when mixed with different types of overburden, and the practical extent of leaching from ash dumps. The development of equipment suitable for handling and conveying dry or water-conditioned ash has commenced.

An ocean outfall system is an interesting prospect. The water-soluble components of Latrobe Valley ashes are similar to the salts present in sea-water so that no serious effects would be expected. However, this system raises many problems of resource management that require serious consideration in addition to problems of solution chemistry such as the precipitation of magnesium hydroxide from Yallourn ash effluent by the admixture of high-pH Morwell ash effluent solution. The feasibility of this system is under consideration.

Although investigated broadly there would appear to be little prospect for finding a means of utilizing significant quantities of Latrobe Valley fly ash. The large proportion of soluble salts in the ash is the principal limiting characteristic and any treatment process proposed would be additionally complicated by the day-to-day variability in ash composition. No system of treatment and utilization has been devised that can either simplify the problems of ash disposal or contribute to a reduction in ash disposal costs.

DESIGN CONSIDERATIONS FOR 575-MW UNITS AT BIG BROWN STEAM ELECTRIC STATION

by

W. H. McAlpin[77] and B. B. Tyus[78]

Introduction

Dallas Power & Light Co., Texas Electric Service Co., and Texas Power & Light Co. generate and distribute electric energy in adjoining service areas located entirely within the State of Texas. These areas cover 75,000 square miles in the north central, eastern, and western parts of the State that contain a population estimated in excess of 3.7 million. Electric energy demand is increasing at a rate that necessitates a continuous evaluation of energy sources to provide future requirements.

Historically, natural gas has been utilized as the predominant fuel for electric energy generation. Texas Power & Light Co. used Texas lignite at one of its powerplants until the early 1940's. A powerplant associated with an industrial installation in central Texas has utilized lignite as fuel since about 1953.

In 1967, these three companies decided to install a jointly owned powerplant to utilize locally available lignite for electric energy generation. This jointly owned powerplant is known as Big Brown Steam Electric Station and is located approximately 90 miles south of Dallas. The plant has two 575-MW generating units presently in operation--unit 1 was initially operative in December 1971, and unit 2, in November 1972. Steam cycle for the units was based on supercritical pressure design. Plant design incorporates an outdoor concept. Figure 85 shows a plot plan for the station. Plant cooling water is provided from a lake constructed on Big Brown Creek, a tributary of the Trinity River. Electric power transmission facilities have been provided to interconnect with load centers of the owner companies. Fuel for the plant is obtained from lignite deposits in the immediate vicinity.

Turbine Generators

The station's two main turbine generator units were supplied by Westinghouse Electric Corp. and designed for outdoor installation. Each turbine is tandem-compound, 3,600-rpm, four-flow, and designed for operating steam conditions of 3,500 psig, 1,000° F throttle, and 1,000° F reheat. Each generator is hydrogen-cooled, 3,600-rpm, 18,000-volt, 3-phase, 60-cycle, and includes a rotating-rectifier-excitation system and voltage regulator.

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[78] Senior Mechanical Engineer.
Both authors are with Texas Utilities Services, Inc., Dallas, Tex.

Feedwater and Steam Cycle

Condensate deaeration is provided in each main unit condenser. Mechanical vacuum pumps are provided for air removal from each condenser. Condensate polishing demineralizers are utilized to maintain low solid concentrations in the feedwater. Seven closed-extraction feedwater-heaters are used in each water cycle. Pumping equipment for each water cycle includes two 50-pct-capacity low-pressure condensate pumps, one 100-pct-capacity condensate booster pump, and one 100-pct-capacity boiler feedwater pump in series. A mechanical driven steam turbine, normally supplied with extraction steam from the main turbine, is arranged to drive both the condensate booster pump and boiler feedwater pump.

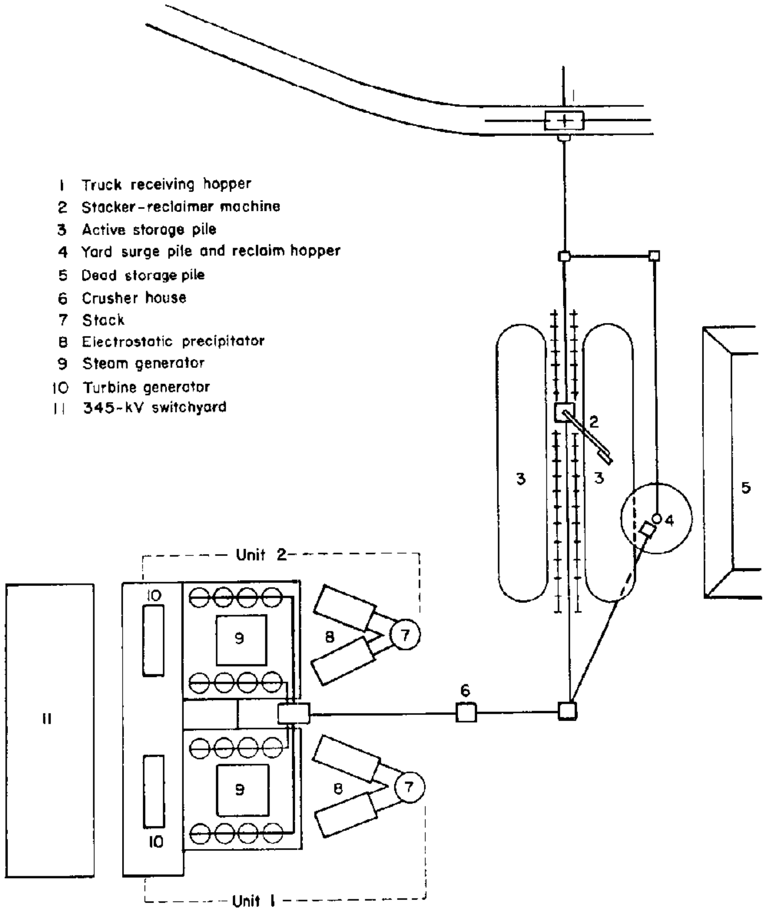

FIGURE 85. - Plot plan for Big Brown Station.

Main Steam Generator Units

The station's main steam generator units (boilers) were supplied by Combustion Engineering, Inc. They are combined circulation type--supercritical, radiant, reheat, divided furnace, balanced draft, and tangentially fired. Each unit is designed to deliver 4,025,000 pounds of steam per hour at 3,825 psig, 1,005° F and reheat to 1,005° F. Figure 86 is a section view of the station's main steam generators. The units were designed to burn lignite having the typical analysis given in table 24.

Each steam generator, designed for dry bottom operation, has a furnace center division wall. Eight No. 943 Combustion Engineering, Inc., bowl mills, located four on each side of the unit, are provided to pulverize the lignite for combustion. Each mill supplies fuel to a level of burners (each level consisting of eight fuel nozzles--one per corner). Gravimetric feeders supplied by Stock Equipment Co. are provided to regulate feed of fuel to the mills.

Main burner ignitors are provided with natural gas fuel. The ignitors are used to bring the unit up "on line."

TABLE 24. - Typical analysis of Texas lignite

(Heating value, as received: 7,600 Btu/lb;
Hardgrove Grindability Index: 48)

Proximate, wt-pct:	
Moisture	30.8
Volatile matter	31.7
Fixed carbon	27.0
Ash	10.4
Ultimate, wt-pct:	
Moisture	30.8
Carbon	43.0
Hydrogen	3.5
Oxygen	11.1
Nitrogen	.6
Sulfur	.6
Chlorine	.2
Ash fusibility, ° F:	
Initial deformation:	
Reducing atmosphere	2,075
Oxidizing atmosphere	2,105
Softening (H = W):	
Reducing atmosphere	2,220
Oxidizing atmosphere	2,255
Softening (H = 1/2 W):	
Reducing atmosphere	2,230
Oxidizing atmosphere	2,275
Fluid:	
Reducing atmosphere	2,310
Oxidizing atmosphere	2,375

H = height of sample. W = width of sample.

The forced draft fans were supplied by Westinghouse and the primary air fans and induced draft fans were supplied by the Air Preheater Company Corp. Two regenerative-trisector type air heaters are provided on each unit to recover heat for primary and secondary combustion air.

Control of ash accumulation in the furnace and gas spaces is accomplished with soot blowers supplied by Copes-Vulcan. Each unit has 34 retracts, 192 wall blowers, and 4 air-heater soot blowers that use compressed air for blowing medium. Three 4,500-hp motor-driven multistage centrifugal-type soot blowing air compressors, supplied by Allis Chalmers, are provided to supply soot blowing air for the two steam generators.

An ash handling system, supplied by United Conveyor Corp., is provided for furnace bottom ash and fly ash removal. Furnace bottom ash is collected in a bottom ash hopper and transported hydraulically to an ash storage pond near the plant. Fly ash is collected in the precipitator hoppers and transported pneumatically to a 2,800-ton-storage-capacity silo for subsequent disposal by truck haul to landfill areas nearby.

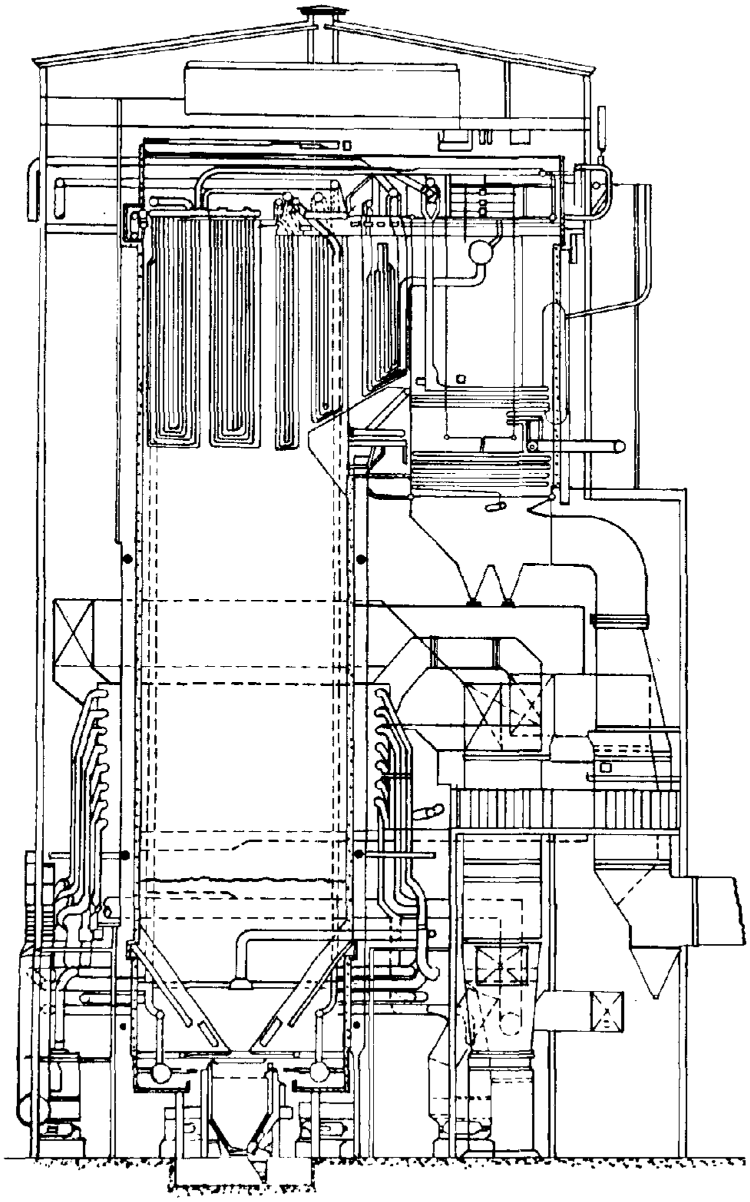

FIGURE 86. - Steam generator unit for Big Brown Steam Electric Station.

Electrostatic precipitators, supplied by Research Cottrell, are installed after the air heater and ahead of the induced draft fans to remove the particulate (fly ash) from the flue gas to the stacks. A consultant performed a "powerplant emission study" using stack effluent dispersion models to predict the required stack height to prevent exceeding the ambient air quality standards. Two (one per unit) 400-foot-high concrete stacks with brick liners were installed. Tests taken indicate that the station is in compliance with Texas Air Control Board emission regulations.

A single startup boiler, supplied by Combustion Engineering, Inc., is provided to supply essential steam requirements during startup of either of the main units. These requirements are for turbine steam seals, for heat required for condensate deaeration during startup, for maintaining cold end temperature of the air heaters, and for the boiler feed pump and condensate booster pump turbine drives. The startup boiler is fired with natural gas and has a steaming rate of 190,000 lb/hr at 185 psig and 550° F. This boiler is only required during a main unit startup and is maintained on hot standby while the main units are providing their own steam requirements.

Fuel Handling System

Lignite from adjacent deposits is delivered to the station in 120-ton truck haulers. The lignite-handling system of the station, supplied by McNally Pittsburgh Manufacturing Corp., consists primarily of a truck receiving hopper, two coal breakers that reduce the received lignite to 6- by 0-inch, a rotary bucket-wheel stacker/reclaimer machine that has a stack rate of 2,200 tph and a reclaim rate of 1,000 tph, a yard reclaim hopper, two ring crushers to reduce the 6- by 0-inch lignite to a minus 3/4-inch size, several belt conveyors, and surge bins. Miscellaneous equipment includes dust suppression systems to wet the lignite for dust control and dust collecting systems to maintain negative pressure on the transfer chutes and bins to minimize emission of dust at these points. The stacker/reclaimer machine stacks out and forms two long active yard-storage piles for use when lignite is not being received. Mill silos and surge bins are sized for approximately 4 hours storage supply; therefore, considerable stress is placed on the lignite handling system to maintain continuous fuel delivery to the steam generators.

Cooling Water Reservoir

Cooling water for the station is provided from a lake of approximately 2,400 acres in surface area. The cooling water system includes pumps and canals to provide water circulation in the lake to accomplish the required heat rejection. Provisions including a closed-loop water cycle for bottom ash handling system and a settling pond for storm water runoff from lignite storage area were included in the design of the project to prevent contamination of water from conditions that may result from the station operations.

Electrical Transmission

A 345-kV switchyard provides for transmission facilities to load centers. Main power transformers from the electrical generators to the switchyard are rated at 19 to 345 kV.

Construction

Placement of orders for the station equipment started in 1967. Erection of the station facilities started in the fall of 1968. The station is located about 21 miles from the closest railroad; therefore, all erection materials and station equipment were hauled by truck to the site. Texas Utilities Services Inc. was designated by the owners to serve as the agent to design, construct, and operate the jointly owned powerplant facilities. Consultants were used for detailed engineering and design. Construction was by contract.

Operating History

The two steam units have operated successfully for a significant period of time. Operation at or near design load for extended periods has been achieved.

There were minor problems relating to equipment and operation during startup and operation of the units. Ash concentration on furnace walls and other adverse conditions that may be associated with lignite combustion have been minimal. Ash concentrations in the burner zone were considered excessive, and the boiler manufacturer has installed 46 additional wall blowers in these areas for cleaning purposes. Ash accumulation in gas passes and on convection surfaces has been minimal.

Operating conditions associated with these units have been considered satisfactory to date.

DESIGN AND OPERATING EXPERIENCES OF STEAM GENERATORS FOR LOW-GRADE BROWN COAL

by

Thomas Buck[79] and Udo Schuler[80]

General Trends

Brown coal is used extensively in European countries for power generation. When Kramer-hammer mills and KSG-beater-wheel mills were developed after 1920, it was possible by combining the grinding and drying processes to inject the pulverized material into the combustion chamber of a boiler, and ignite and burn it in suspension. After 1950, the technique of pulverized firing of brown coal became more refined and rapidly found widespread application. The steam generator capacity of brown coal-fired units in West Germany was increased to about 500 tph of steam during 1954. The first steam generators for 650 tph were put into operation in Poland in 1962. These units were designed and manufactured under license from Kohlenscheidungsgesellschaft mbH. Steam generators producing 1,000 tph with corresponding 300 MW of electric energy were put into operation in Germany after 1966. Boilers of 2,000-tph capacity equivalent to 600 MW will be commissioned in 1973. Before the operating experience gained with these units can be applied in other countries, careful consideration must be given to firing techniques, and the condition and characteristics of the coal must be evaluated.

Types of Brown Coal

It is generally known that the designation "brown coal" includes solid fuels having moisture contents to 75 pct, ash contents to 50 pct, and lower heating values between 800 to 5,000 kcal/kg (1,450 to 9,000 Btu/lb). Such wide ranges in coal quality require varying techniques for preparation and firing.

Some analytical data for solid fuels from different sources are presented in diagram form in figure 87. During transformation of wood to peat, to brown coal, and to various kinds of black coal, the volatile matter decreases while carbon content and calorific value increase. In the same sequence, the oxygen and hydrogen contents decrease. In general, even limited regional coal deposits exhibit these characteristical variations. In the same sequence the density of the coal substance increases. As shown in figure 87, the selected parameters at best indicate a wide band relationship when considering coal originating from different parts of the world and especially for the brown coals.

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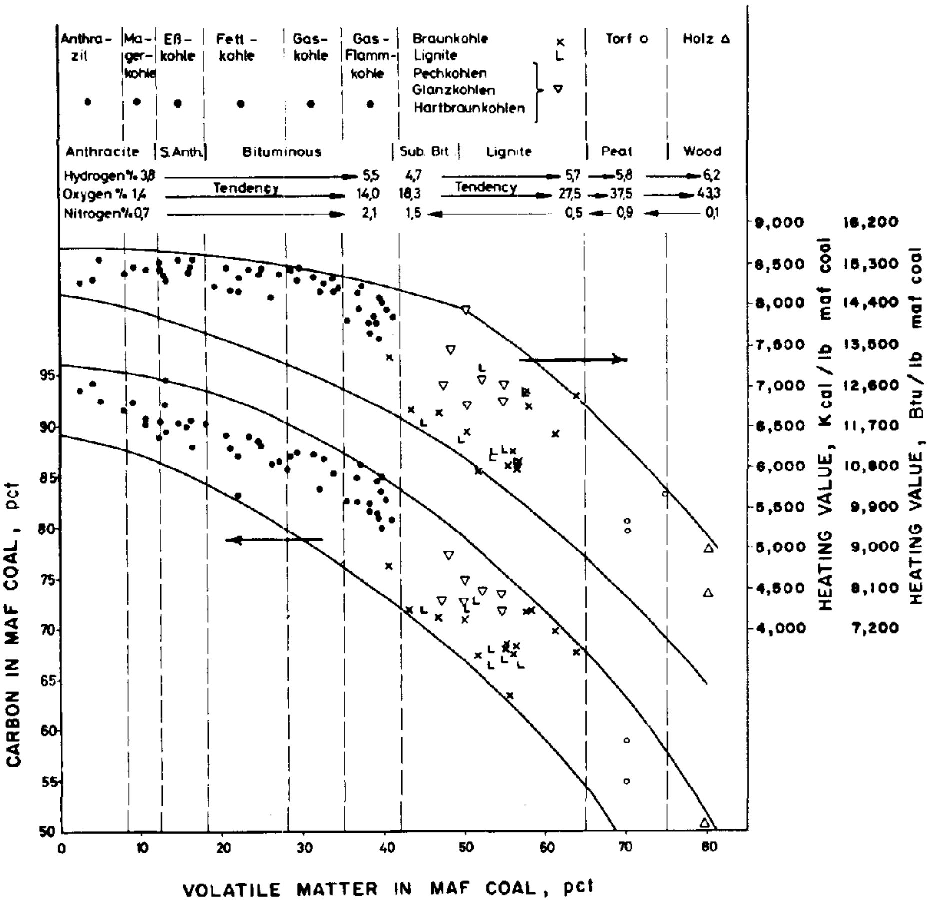


FIGURE 87. - Classification of various solid fuels.

In figure 88, analytical parameters are presented that are important in selection of firing equipment for brown coal and lignites. Included are heating values and moisture and ash contents from some well-known brown coal deposits. Earthy brown coal of the highest known moisture content is fired at the Yallourn power station in Australia. The area indicating medium moisture and ash contents and heating values between 800 and 2,500 kcal/kg (1,450 and 4,500 Btu/lb) includes earthy brown coals from various European sources in Germany, Poland, Greece, Spain, Yugoslavia, and Hungary. The older lignites from the United States have heating values to 4,000 kcal/kg (7,200 Btu/lb), lower moisture contents of 25 to 40 pct, and ash contents between 5 and 15 pct. In contrast, various brown coals from Bulgaria and particularly from the Tuncbilec deposit in Turkey have a relatively high ash content of about 48 pct and are similar to hard brown coal. Consequently, their position on the diagram is quite far removed from the more usual coals.

Yallourn and Tuncbilec coal have similar heating values with widely different moisture and ash contents. Thus, preparation and firing techniques have to take these variations into account to avoid difficulties during plant operation.

Other characteristics than those in figure 88 have to be considered during the design for mechanical preparation of the coal. These included such items as ungrindable long fibrous xylithes, greasy clays or loam, and abrasive pyrites or sand that may be present in varying amounts.

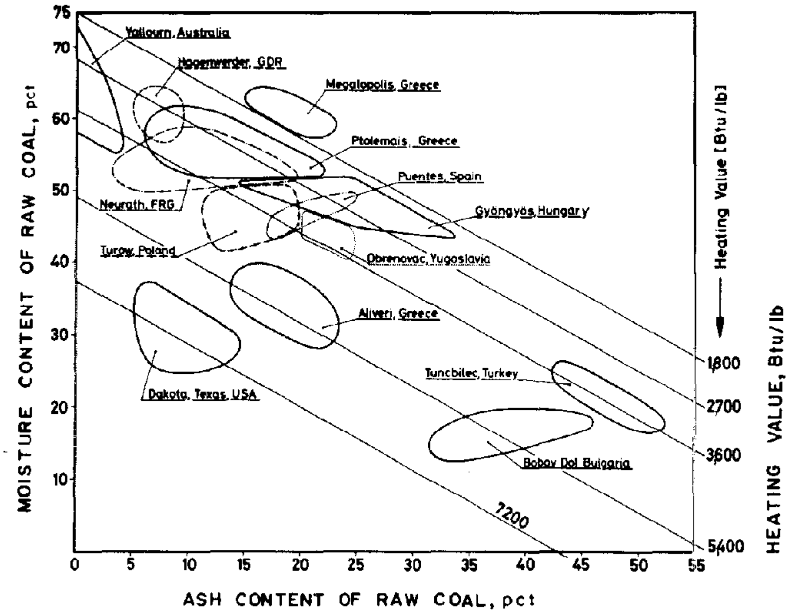


FIGURE 88. - Diagram for raw brown coal and lignites.

Effect of Mining Methods on Quality of Coal

Opencast mining is the principal method used for brown coal. Coal seams, between 2 and 150 m in thickness, have intermediate layers of varying thickness. Figure 89 shows, as a typical example, a cross section through a seam of an opencast mine at Kardia in Greece. Mining is carried out using efficient rotary scoop excavators. Mineral layers less than 0.5 to 1 m thick cannot be mined separately from the coal.

Inclusion of extraneous mineral matter lowers the heating value of the mined coal. In design of the preparation and firing plants, it is important to consider the effects of these extraneous minerals. A mixing plant should be provided prior to pulverization to insure uniform quality of the coal, which will reduce problems such as those associated with ignition.

Firing Equipment

Besides corner firing, front- and roof-firing have been used for brown coal-fired steam generators with capacities to 500 tph. For steam generators with capacities between 1,000 tph and 2,000 tph, tangential firing is used exclusively. With tangential firing, the burners are arranged in the wall of the combustion chamber so as to be directed towards a "ring" in the combustion chamber. The combustion flame fills the combustion chamber. Because of intense mixing of pulverized coal with combustion air, retention time of the coal dust particles in the combustion chamber is increased. When firing hard brown coals of low moisture content, the technique used for bituminous coal is applied in that one mill furnishes several burners with pulverized coal. With earthy brown coal of higher moisture content, it is common to have a mill to serve only one burner.

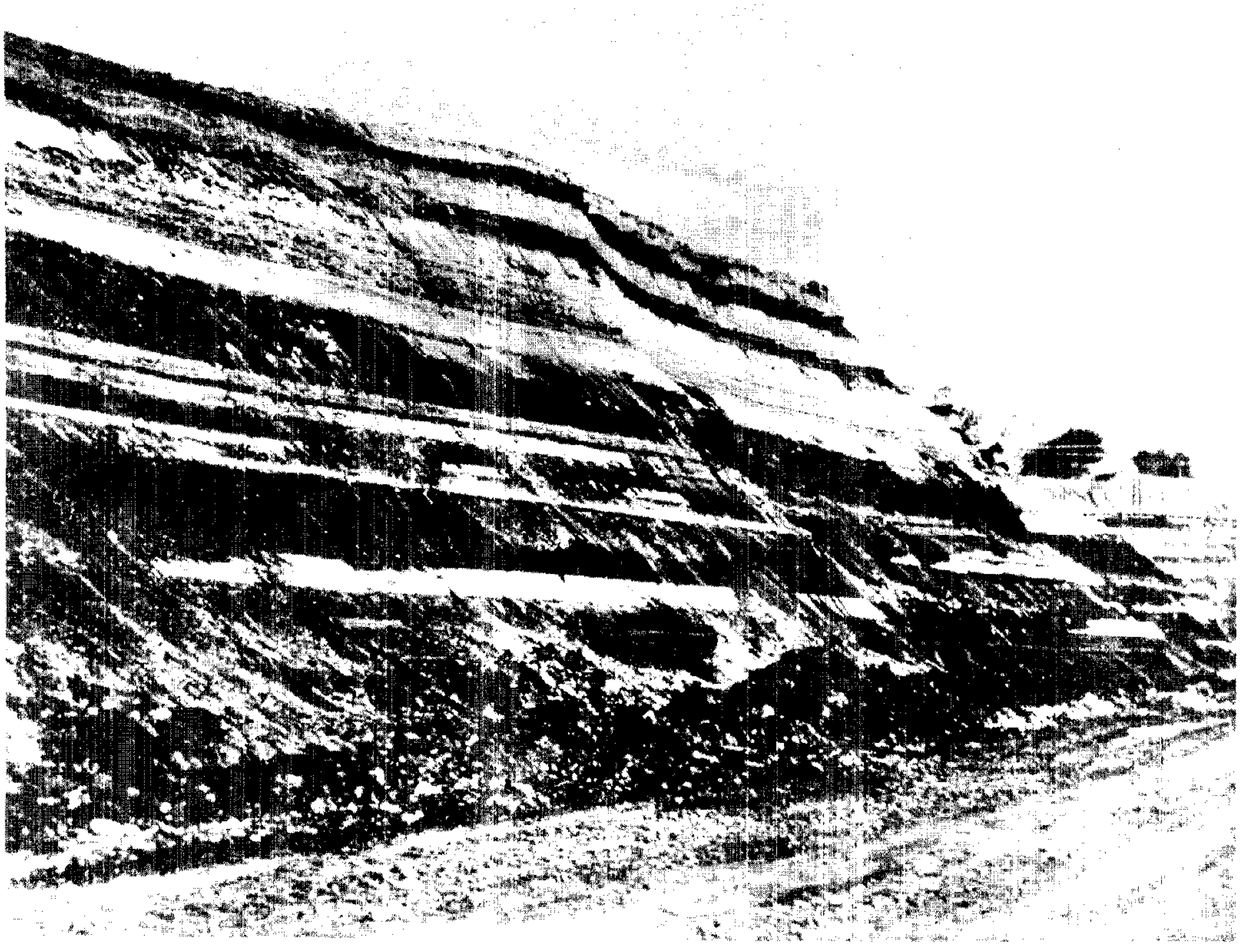
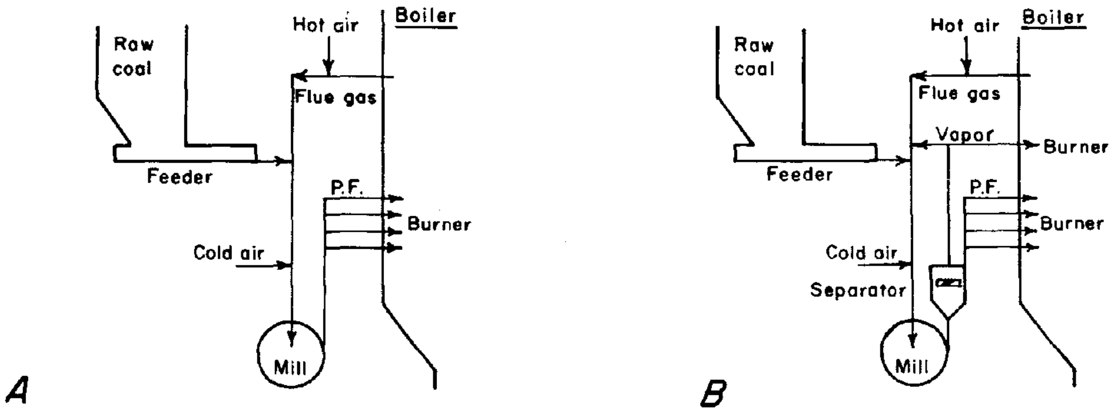


FIGURE 89. - Brown coal in the opencast mine at Kardia, Greece.

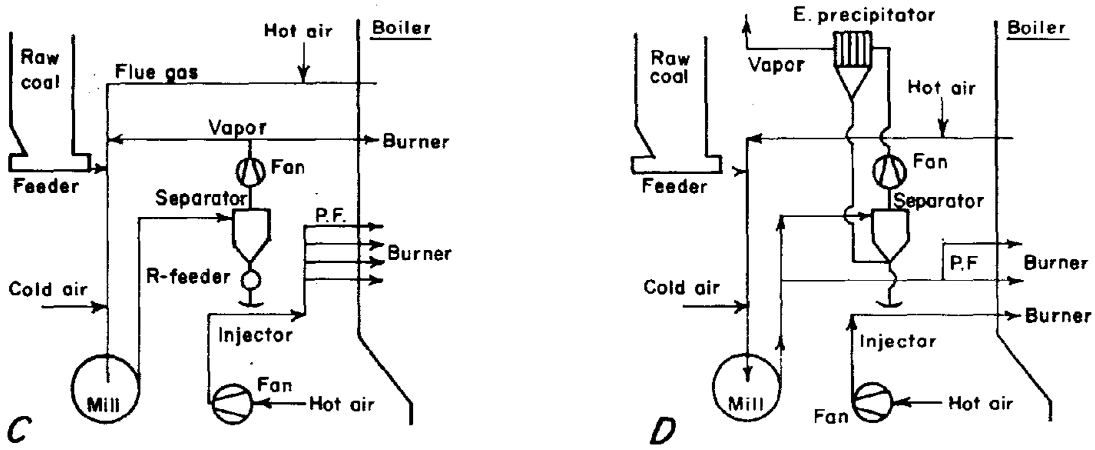
Most of the firing methods which have been used can be arranged within the diagrammatically illustrated group of systems shown in figure 90. Figure 90A shows the direct injection system. Raw coal leaving the bunker travels on a feeder into the mill where it is simultaneously pulverized and dried by hot gases. The combustible pulverized fuel is fed to the burner and into the combustion chamber together with the vaporized moisture and conveying gas. The analysis for coal used at the Niederaubem power station in West Germany are quite typical. This firing system can be used for a large proportion of the West European raw brown coals.

Figure 91 shows the arrangement of the pulverization system and associated burners for the steam generator of a 600-MW unit. Included are the flue gas recirculation duct, the raw coal supply into the flue gas recirculation duct, the mill, and the pulverized fuel ducting to the burner. The burner cross section is subdivided into three groups, each of which contains two pulverized fuel outlets and outlets for bottom air, intermediate air, and upper air, respectively.



Niederaubem, West Germany 600 MW
Direct fired
LHV 2,960 - 4,600 Btu/lb
Moisture 52 - 57 pct
Ash 2 - 13 pct

Yallourn, Australia 350 MW
Direct fired
LHV 1,800 - 3,800 Btu/lb
Moisture 58 - 73 pct
Ash max. 3 pct



Tuncbilec, Turkey 150 MW
Indirect fired
LHV 3,600 Btu/lb
Moisture 22 - 24 pct
Ash 42 pct

Megalopolis, Greece 125 MW
Indirect fired
LHV 1,600 Btu/lb
Moisture 64 pct
Ash 13 pct

FIGURE 90. - Preparation and firing systems for brown coal and lignite.

If the moisture contents increase to more than 50 pct and ash content is low, direct injection of the pulverized fuel may be used as shown in figure 90B. This firing method insures that ignition characteristics of the pulverized fuel conveying gas mixture is increased by removing part of the inert water vapor. There is also an associated increase of temperatures in the combustion chamber that aids in firing of coals that are difficult to ignite.

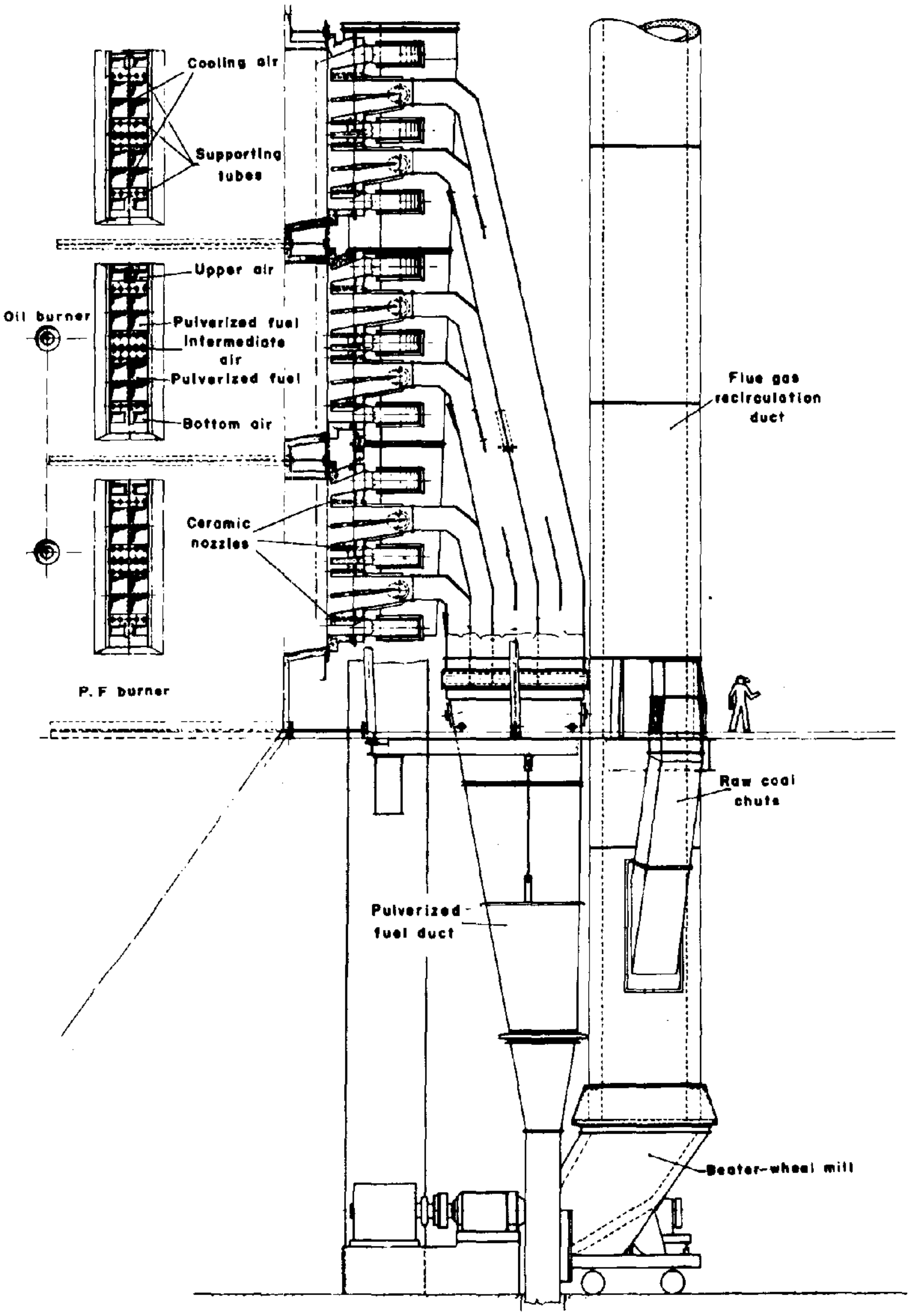


FIGURE 91. - Firing system with beater-wheel mill for 600-MW units.

The coal dust gas mixture is rotated in the centrifugal separator located within the pulverized-fuel ducting so that pulverized-fuel particles are directed against the separator wall and then conveyed as an enriched pulverized-fuel gas mixture to the pulverized-fuel burners. The vapors with a low pulverized-fuel content enter the combustion chamber above the ignition zone where the fine pulverized fuel present is burnt. Applications of this method have been made in the power station at Yallourn in Australia, the Ptolemais plant in Greece, and the Hungarian power station at Gyongyos. Figure 92 shows the Ptolemais plant including the pulverized fuel ducting and the vapor separation device that separates the pulverized-fuel gas mixture into portions rich and low in pulverized fuel. The rich pulverized-fuel gas mixture is burned in both lower burner levels with the addition of bottom and upper air. The less rich pulverized-fuel gas mixture is injected above the ignition zone. However, it can be seen from the diagram shown in figure 90B

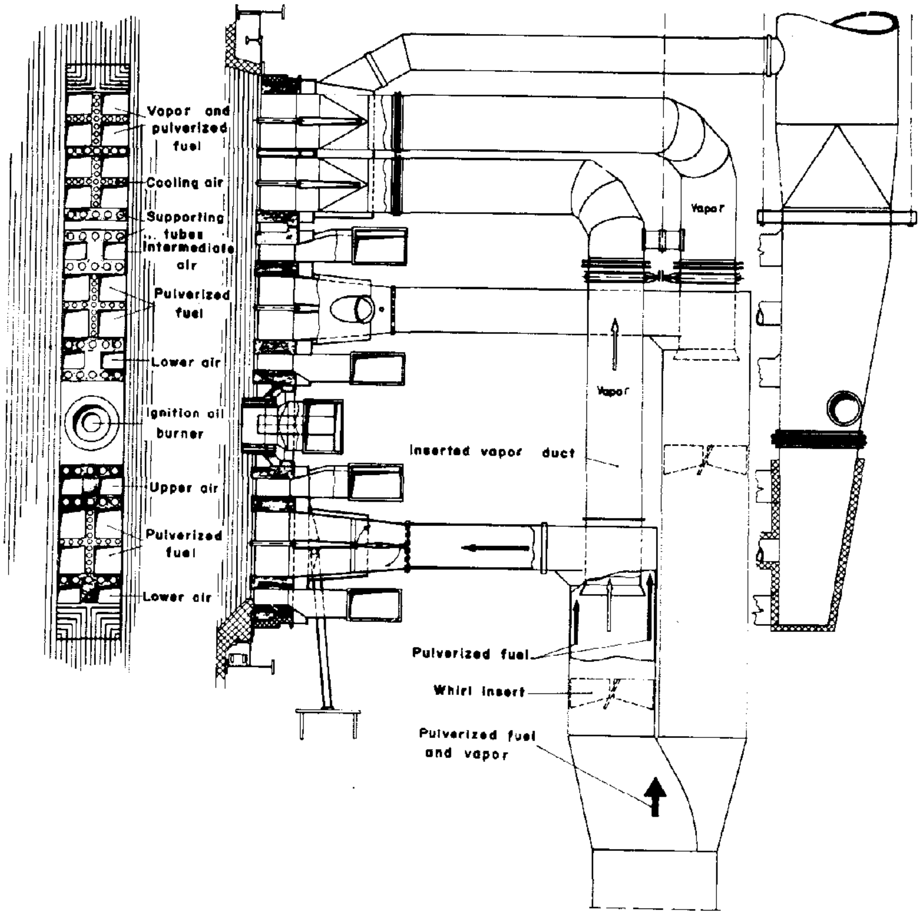


FIGURE 92. - Vapor burner for 300-MW brown coal-fired unit, Ptolemais IV, Greece.

and 90C that vapor separation is not recommended if the readiness to ignite is impaired by a high ash content. In these cases, splitting the mixture into a lean pulverized fuel portion and another portion rich in pulverized fuel will not achieve the same success as with coals of high moisture contents and lower ash contents.

The firing system in figure 90C is recommended for coals with high ash contents. The pulverized fuel is freed from the vapor on leaving the centrifugal separator and collected in a small intermediate bunker. The pulverized fuel is fed to the burners of the boiler plant with the aid of primary air via rotary feeders and injection nozzles. The portion of the vapors with the low pulverized-fuel content are fed to burners above the ignition zone with the aid of a fan. In the case of the Turkish plant (Tuncbilec), this procedure allows a favorable relationship between pulverized fuel and conveying air quantities and improves ignition readiness of the pulverized fuel. Use of hot air as a conveying medium gives favorable ignition and combustion conditions.

The firing system shown in figure 90D illustrates a special design of the vapor firing shown in figure 90B. As with the usual case of vapor firing, the pulverized-fuel gas mixture is separated after the mill outlet into rich and lean pulverized fuel portions, but the lean pulverized-fuel mixture is precleaned in a separator, and the dust is removed in an electrostatic precipitator. This system has advantages particularly in the preparation of brown coals that have very low calorific values and high moisture contents. For hard brown coal that has a low calorific value, a low moisture content and high ash content, the vapor separation shown in figure 90D cannot achieve the same success as it would with moist coals. With proper systems, the preparation and firing of brown coal with calorific values down to 800 kcal/kg (1,450 Btu/lb) is possible.

The influence of the coal quality on the selection of the firing system has been emphasized. It must be recognized that in marginal cases other design conditions can have a decisive influence on the selection of the system. In this connection, instances when the calorific values drop below the average value because of insufficient blending are important. Requirements for a very large capacity reserve in the grinding plant for coal with the lowest calorific value in one case and for a very low-load operation capability of the steam generator on the other, can lead to the selection of the next more expensive firing system.

Combustion Chamber Dimensions

After selection of a suitable firing system, dimensions of the furnace space must be established. Important are the volumetric and cross-sectional heat release rates as well as the burner zone heat release. Figure 93 shows the relationship of the volumetric and cross-sectional heat release in regard to the capacity of steam generators. Volumetric heat release decreases when the steam generator capacity increases while the cross-sectional heat release increases. Experience has established limits for heat release in the burner zone to keep fouling and slag deposits to a minimum near the burners.

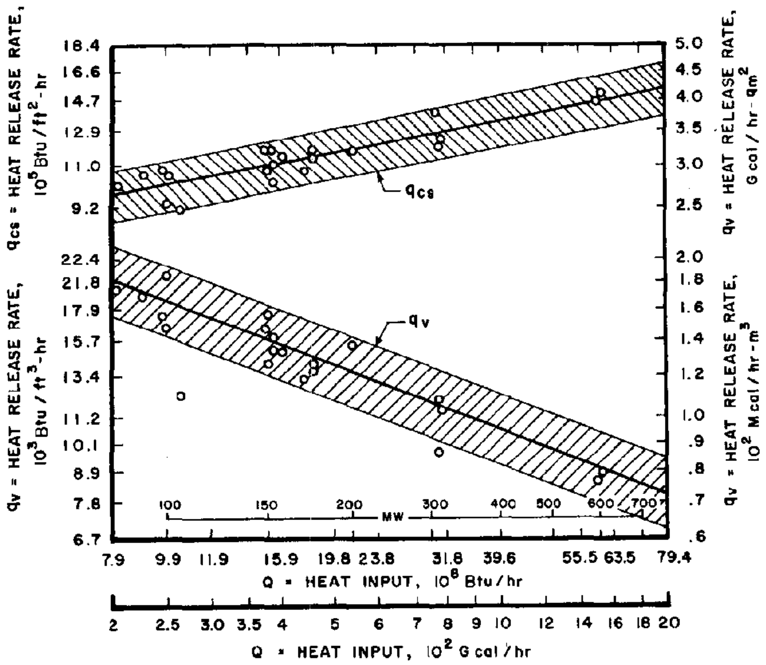


FIGURE 93. - Volumetric and cross-sectional heat release related to the capacity of steam generators.

Volumetric heat release is a measure of retention time of the combustion gas in the combustion chamber. It is usually assumed that pulverized particles follow the same path as the combustion gas. With increasing cross-sectional heat release, higher axial-flue-gas velocities result within the combustion chamber when an evenly distributed flow over the cross-sectional area is obtained. With an increase in the steam generator size, the conditions for complete combustion will be more favorable with the same pulverized fuel fineness because retention time will be increased and the larger particles can be held in suspension.

The combustion chambers of the steam generators for the 600-MW plants Niederaubem and Neurath in West Germany have a cross-sectional dimension of 20 by 20 m and a furnace height of 69 m with the corresponding cross-sectional heat release rate of about 4 Gcal/m^2h and a volumetric heat release rate of about 0.8×10^2 Mcal/h M^3 corresponding to 14.7×10^5 Btu/sq ft hr and 8.9×10^3 Btu/ft^3 hr, respectively.

Layout of Burners

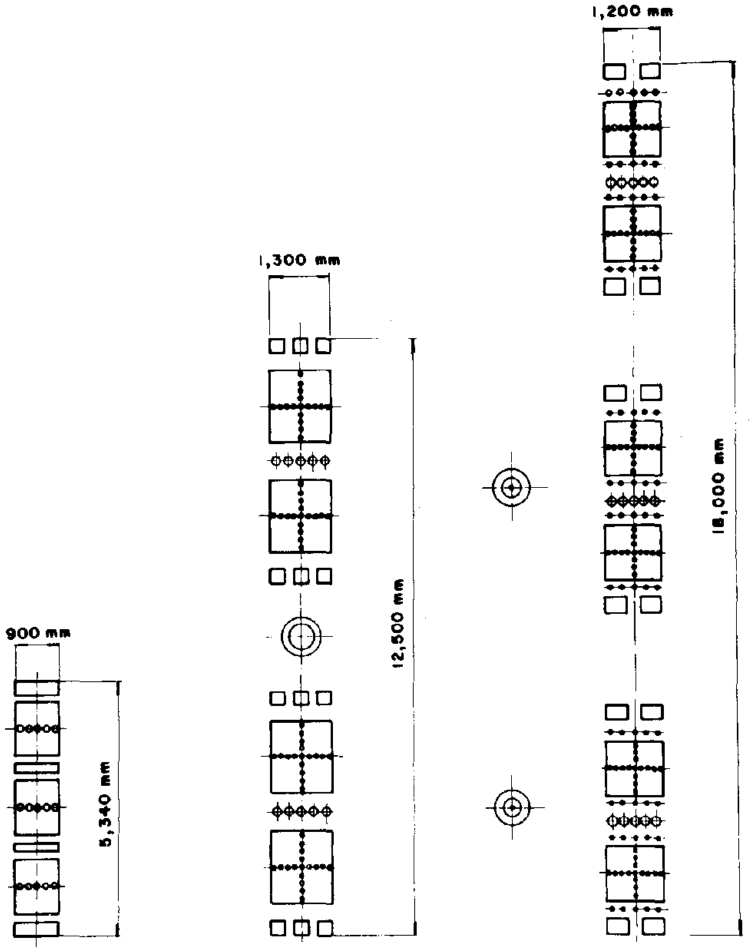
Other technical requirements include observing definite pulverized-fuel mixture exit velocities, secondary air velocity, energy and impulse conditions at the burner, and a velocity reduction of pulverized fuel streams and airstreams at increasing distance from the burner outlet. Limiting damage by excessive temperature on shutdown is done mechanically by correct dimensioning of the exit cross sections of burners, by use of ceramic linings, and by air cooling.

Figure 94 gives the general construction and main dimensions of burners using Rhein brown coal for 150-, 300-, and 600-MW units. With geometrical similarity, it can be seen that the distribution already achieved at the burners for 300-MW plants occur for the 600-MW units. Each burner has three groups comprising of upper air, bottom air, intermediate air, and two pulverized fuel outlets. The following technical data for the burner for 600-MW plants is of interest.

Total height:	18 m (59 feet).
Width:	1.2 m (3.95 feet).
Fuel throughput:	Approximately 120 tph.
Heating value:	1,850 kcal/kg (2,960 Btu/lb).
Hot air temperature:	280° C (536° F).
Pulverized-fuel outlet velocity:	Approximately 12 m/sec (39 ft/sec).
Secondary air outlet velocity:	Approximately 55 m/sec (181 ft/sec).

Pulverizing and Drying Units

For grinding raw brown coal in units with an hourly throughput above 40 tons, only mills can be applied that draw in the hot flue gases for the drying process from the furnace and simultaneously generate the head to compensate for pressure loss in the flue gas resuction ducts and the following pulverized-fuel ducts and burners. It is common to all mills that the major part of the pulverizing and grinding is done by the beater wheel or fan wheel. The quality of the processed material is dependent on the raw coal and to great extent on a uniform coal feed, the selected type of mills, operational methods, and control of the grinding and drying processes according to predetermined results of a heat balance calculation.



Unit capacity (MW)	150	300	600
Mills type	6 KSG-N90.60	6 KSG-N200.45	8 KSG-N270.45
Furnace dimensions (m)	12,5x11,5x18,7	14,928x13,872x33,7	20,0x20,0.69,0
Burner zone heat release			
(Gcal/m²h)	1,83	1,08	1,10
10^3 (Btu/ft²hr)	675	400	406

FIGURE 94. - Cross section of brown coal burners.

Feeding

In addition to reaching average predetermined hourly throughput, the feeder is particularly responsible for avoiding unwanted fluctuations in the coal quantities. Since mills have no storage capacity, they are unable to compensate for such fluctuations in coal flow that might create pulsation in the furnace. The apron plate conveyor as shown in figure 95 proved to be suitable for feeding a difficult raw brown coal. The coal is taken from the bunker and transported to the coal chute by contact with the rough surface of the apron plate conveyor. A roller fitted with arms, installed at the outlet of the feeder, insures that even a sticky coal is uniformly distributed and loose (no agglomerates) before discharge into the flue-gas recirculation duct. There the coal is predried by contact with the hot flue gases.

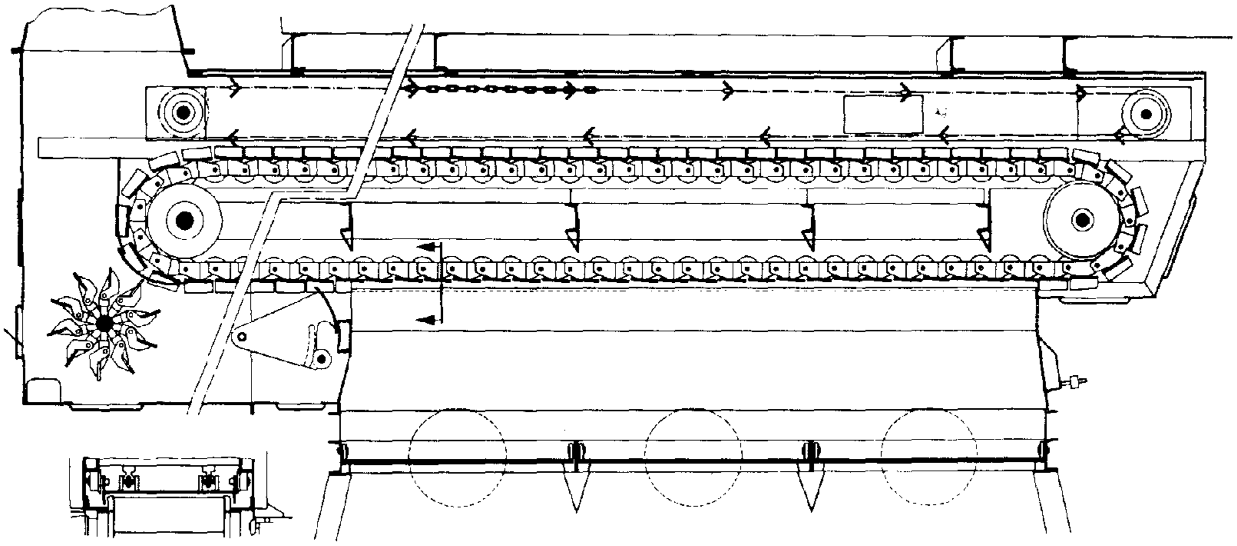


FIGURE 95. - Apron plate conveyor for brown coal.

Mill Types

Only overhung-mounted beater-wheel mills are in competition with fan-type beater mills having bearing on each side for coal throughputs above 40 tph. EVT and its licensees have manufactured or have on order 2,020 beater wheel mills types N and S at present. Two hundred and twenty of these pulverizers are operating with throughputs of 65 to 135 tph of an earthy brown coal. Forty mills were ordered for capacities of 30 to 55 tph for lignites and hard brown coals. For processing earthy brown coals, the mill with classifier shown in figure 96 is widely used. The raw coal is transported together with the flue gases into the rotating beater wheel where impact crushes the coal and the hot gases simultaneously reduce the moisture content. In the classifier, the pulverized fuel is separated from coarse particles and transported together with the gas to the burner. The coarse particles are returned to the mill. If coal with a high content of sandy impurities is to be pulverized, mills without classifiers as shown in figure 97 can be used to avoid increased wear. It is possible to employ hammer groups positioned in the mill door that are driven separately and in opposite direction to the rotating beater wheel. In this manner an increased grinding effect is obtained by use of only a few hammers. Both mill types shown are designed for processing earthy soft-brown coal with relatively high moisture for throughputs to 135 tph. The large spaces inside the mill housing and the classifier indicate the necessity for large throughput-quantities of drying and conveying gases.

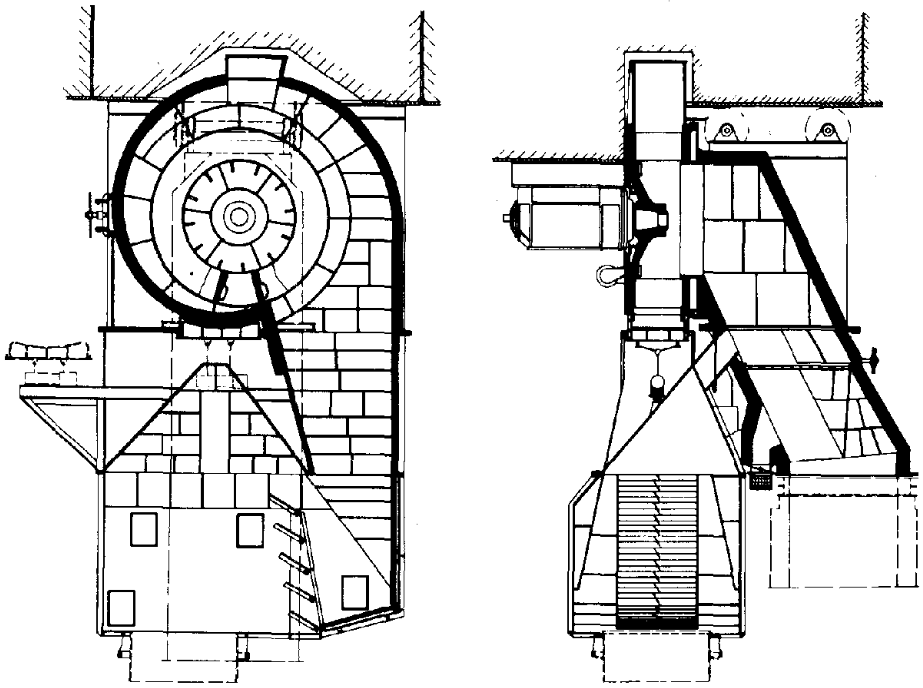


FIGURE 96. - EVT beater-wheel mill, type KSG-N 240.45 (20 tph raw brown coal).

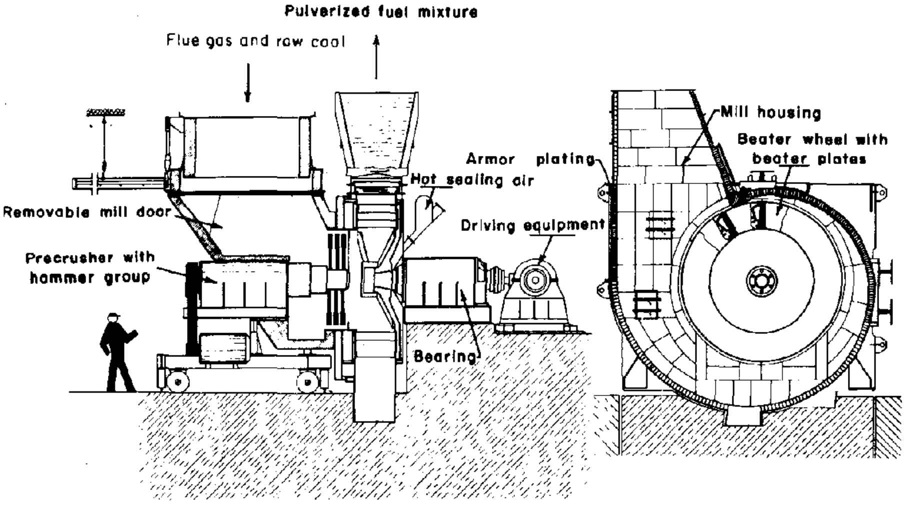


FIGURE 97. - EVT beater-wheel mill, type KSG-N 270.45 (135 tph raw brown coal).

For processing low-moisture lignites and hard brown coals, beater-wheel mills type KSG-S having lower gas throughputs are used. Figure 98 shows a semischematic section of the mill with classifier. The beater wheel has a diameter of 3,400 mm and operates at 500 rpm.

Heat Balance for Pulverizing and Grinding Process

The mechanical resistance of the coal to pulverizing and the required fineness of pulverized fuel as well as the required drying capacity is of importance when determining size of a mill. The required drying capacity is determined by taking into account the demanded load variations in the mill and in firing. The thermal balance provides the basis for the control of the drying and grinding process during future operation as well as in intial sizing. Figure 99 gives the results of drying calculations for various coals in pulverizers of different sizes. For drying raw brown coal, there are available flue gases with temperatures up to 950° C (1,740° F), hot air of 280° C (540° F), cold flue gases of 180° C (356° F)

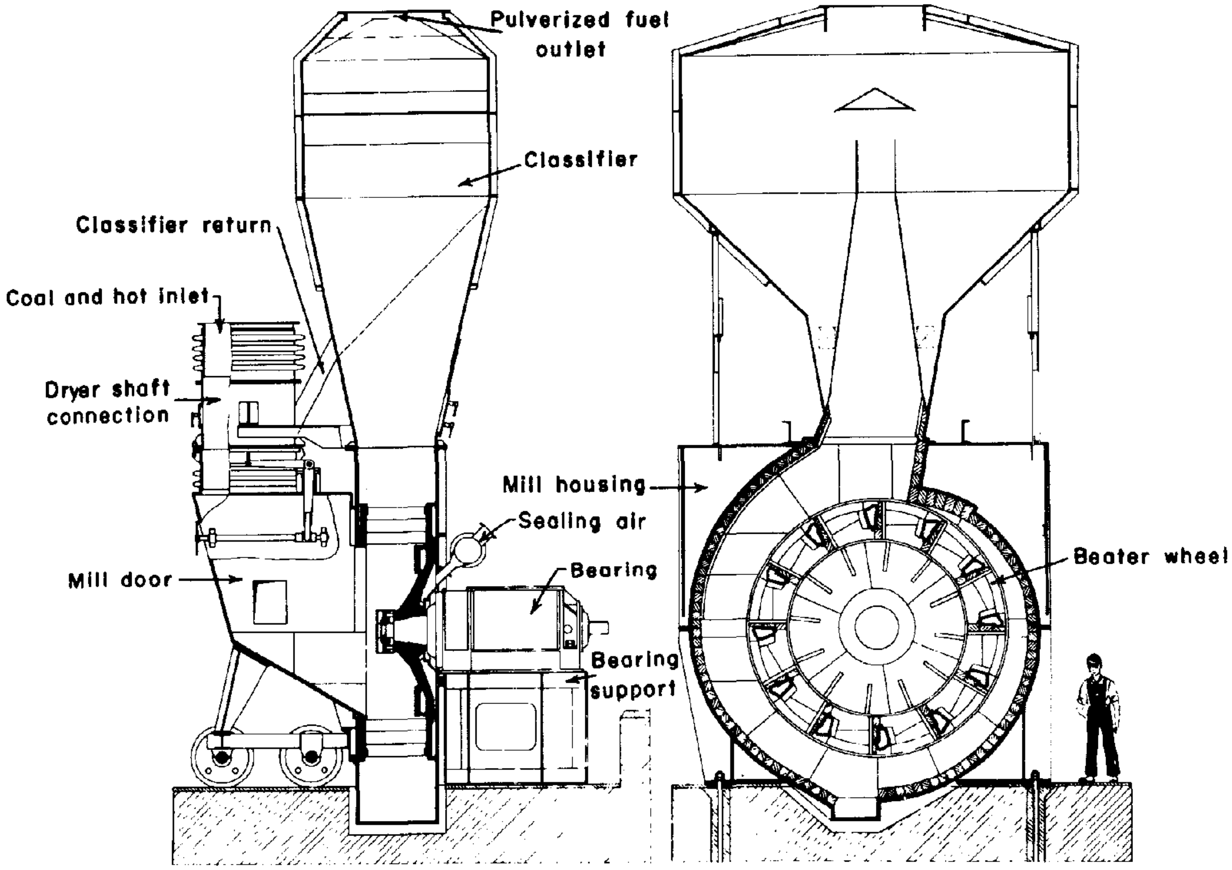


FIGURE 98. - EVT beater-wheel mill, type KSG-S 45.50 (55 tph of lignite).

from the boiler outlet, and cold air for the thermal protection of the mill during planned operational shutdown or in emergency.

The percentage of gas in the conveying volume depending on throughput is shown for different cases. For burning coal of an average moisture content of 50 pct, approximately 9 pct of the conveying gas volume is supplied by hot air at full load. With decreasing coal output and constant mill outlet temperature of 180° C (356° F), the hot air percentage increases at the expense of a reduction in the hot flue gas quantity to compensate for loss of cooling by the coal. At the Australian powerplant, the Yallourn lignite of 65 pct moisture is dried almost exclusively with flue gases during full-load operation. Temperature during part-load conditions is controlled by use of recirculated vapors.

Low-moisture coals are different. When processing 30-pct-moisture lignites in the Aliveri powerplant, 32 pct of hot air enters the pulverizer at full load. With an increase in the hot air volume in the partial load zone of the pulverizers, the secondary air velocity of at least 20 m/sec (66 ft/sec) must be maintained at the burner outlet.

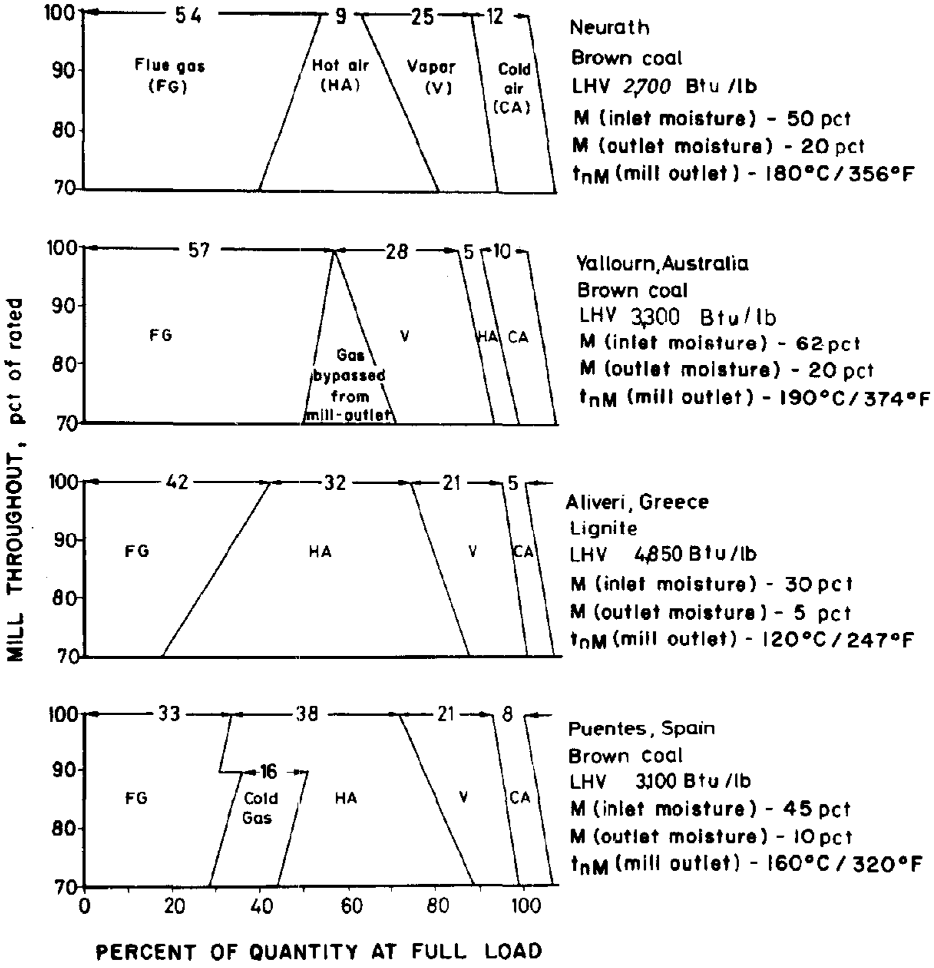


FIGURE 99. - Drying processes for various coals in beater-wheel mills.

In the Spanish powerplant at Puentes, earthy brown coal of 45 pct moisture is pulverized. The load conditions and fuel range necessitate operation of the pulverizers during full-load conditions with 38 pct hot air in the conveying gases when grinding fuel having the designed analysis. It is planned to deliver cold gas for cooling purposes when decreasing the output so that ignition stability is maintained in the partial load range of the mill and an intolerable increase of the hot air portion is avoided.

Another aspect in limiting the hot air flow for temperature control of the pulverizer at reduced output is the problem of maintaining an inert mixture so as to avoid dust ignition in the pulverizer or ducting during all operational phases. By tempering the gases drawn from the furnace with hot air, the CO_2 content in the flue gas drawn from the furnace is reduced below 14 pct. The part load zone of the pulverizer is limited so as to prevent the mill outlet CO_2 content from falling below 4 pct. With sufficiently inert gases, temperatures between 120° and 180° C (247° and 356° F) are admissible using present grinding and drying techniques.

For operation purposes, all results from calculations are summarized in a "mill diagram." An example is given in figure 100. This diagram shows range in fuel consumption to be burned in future operation, the quantity of raw brown coal to be processed at a given boiler load, and the number of mills needed. Usually the method of increasing the control range of the mill is by changing speed of the beater wheel.

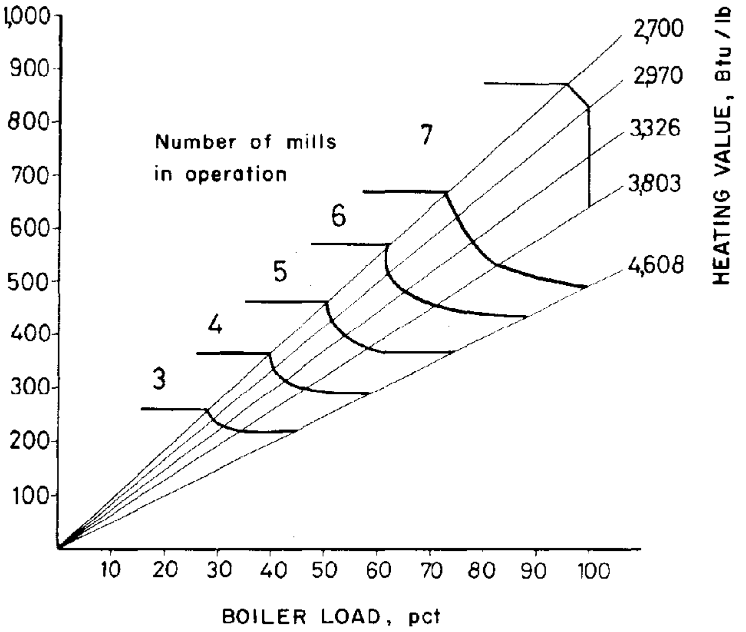


FIGURE 100. - Mill operation diagram (EVT beater-wheel mills, type KSG-N 270.45). Power station Neurath D and E, West Germany.

Arrangement of Pulverizing and Firing Equipment at the Steam Generator

The arrangement of the pulverizers and the burners around the steam generator depends on the firing system that is selected. Figure 101 shows a direct injection system without vapor separation that meets the requirements of direct and shortest pulverized-fuel and flue gas recirculating ducting. With this design selected for four 600-MW units at two West German powerplants, fully symmetrical arrangements could be realized. The identical design of flue gas recirculating ducts and pulverized-fuel ducts facilitates not only design and manufacture but also makes important simplifications in operation. Corrections that may be required for distribution of the pulverized fuel to the individual burner fingers need to be tested on one unit only. The test results can be applied to other installations.

Steam Generators

In the past various designs were used for the arrangement of heating surfaces such as L type, inverted L type, two pass, and single pass. The selection of the L type (fig. 102) was based on the assumption that a high percentage of ash separation could be attained in the bend between the furnace and convection pass. It was also expected that additional ash retention would take place in the horizontal gas pass (L and inverted L).

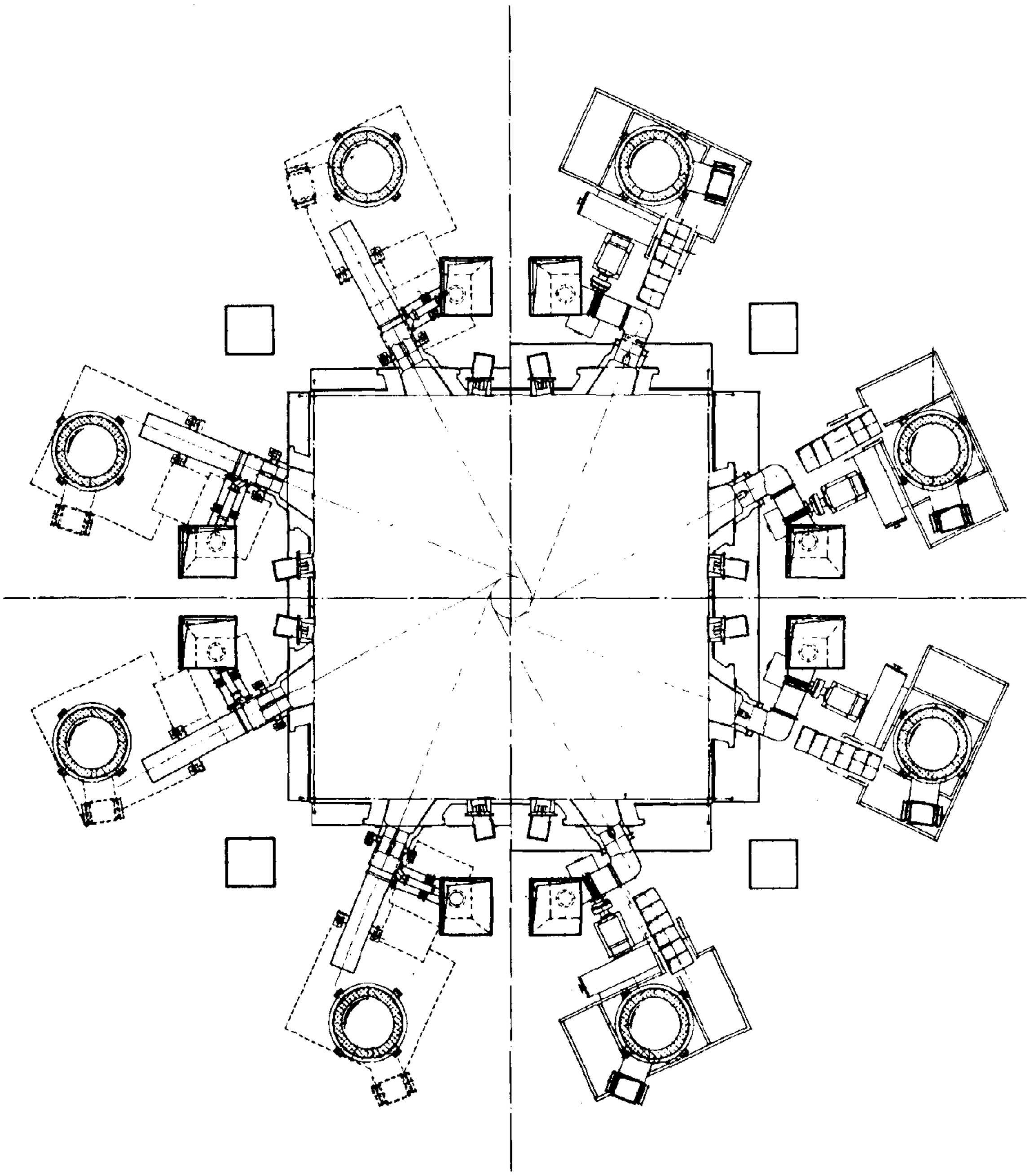
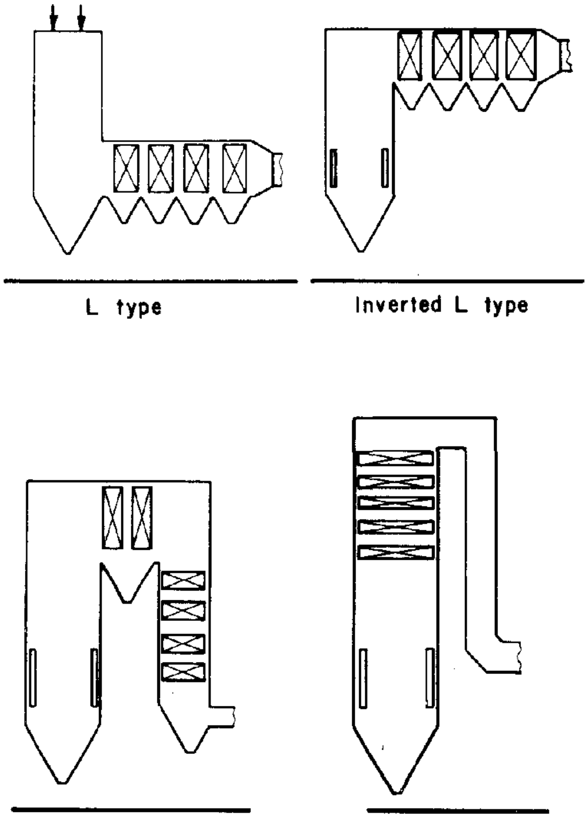


FIGURE 101. - Mill and burner arrangement for 600-MW brown coal-fired units.



L type

Inverted L type

Two pass

Single pass

FIGURE 102. - Design concepts for steam generators.

The latest 300- and 600-MW units are built exclusively as single-pass units (fig. 103). The gases leave through the side of the convection surfaces positioned above the combustion chamber into a downward gas pass and hence to regenerative air heaters that may be arranged vertically or horizontally. The air heaters are followed by electrostatic precipitator and induced draft fans. Use of the single-pass design gives an evenly distributed velocity profile over the path of the flue gas. Erosion is reduced by retardation in velocity of ash particles in the upward flow. The single-pass design facilitates a simpler boiler enclosure that has favored its application even where wear problems are not of primary importance (Australia, Greece). When firing fuel with a lower heating value of 1,500 kcal/kg (2,700 Btu/lb) and an ash content of 20 pct, the total ash throughput is equivalent to the total fuel that would be fired in a bituminous coal-fired unit of same capacity.

The major unknown factor for the prediction of the wear in a powerplant is the raw coal. It has been impossible in the past to predict the specific net wear of the grinding parts on the basis of the physical and chemical characteristics of the raw coal. As shown in figure 104, data from wear tests in a beater-wheel mill gives the specific net wear of beater plates depending on the percentage of particle size over 0.09 mm in the pulverized fuel for various raw brown coals. With the same dust fineness and the same ash content of the raw coal, brown coal fired at the West German powerplant Frimmersdorf

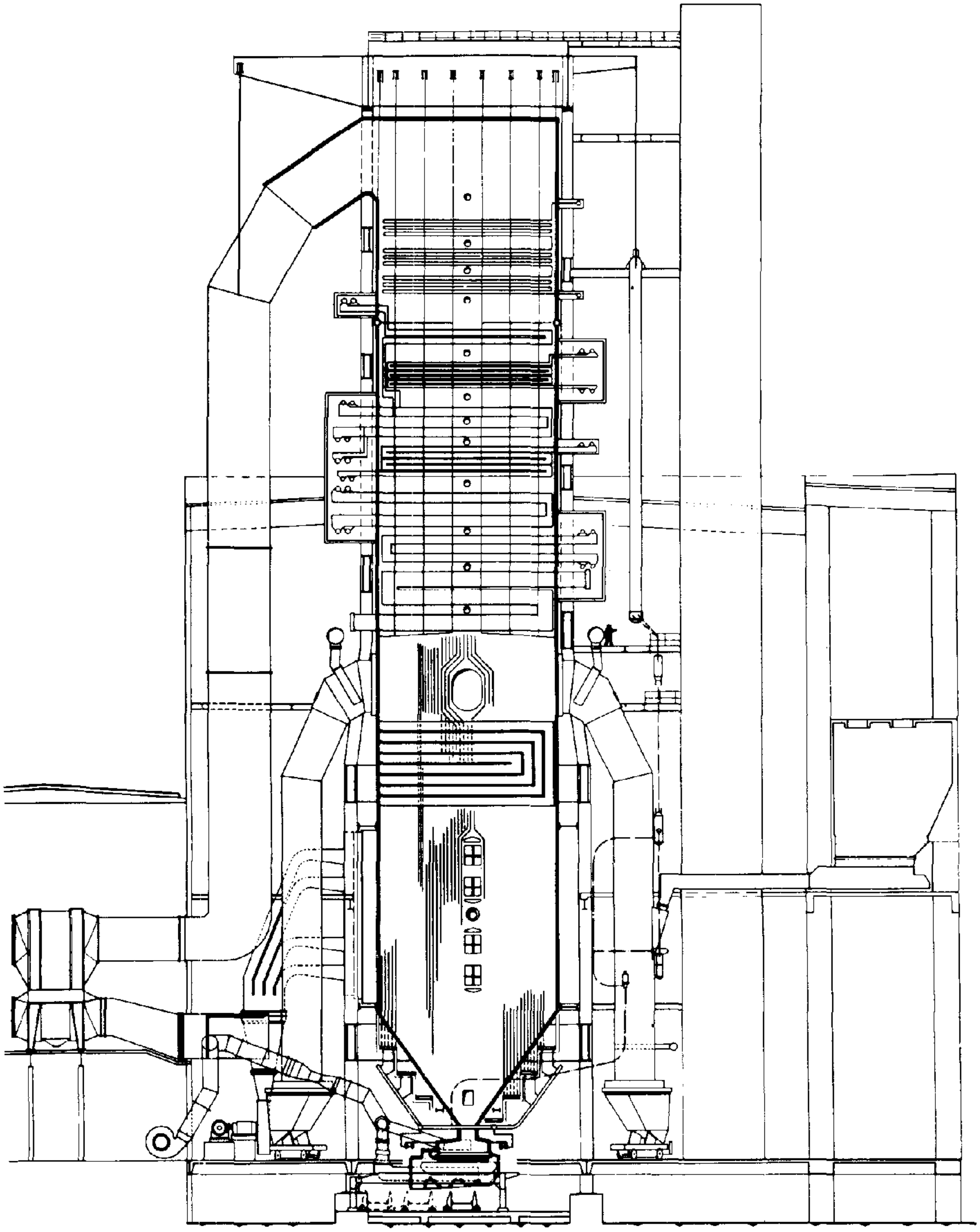


FIGURE 103. - Single-pass design for 300-MW unit.

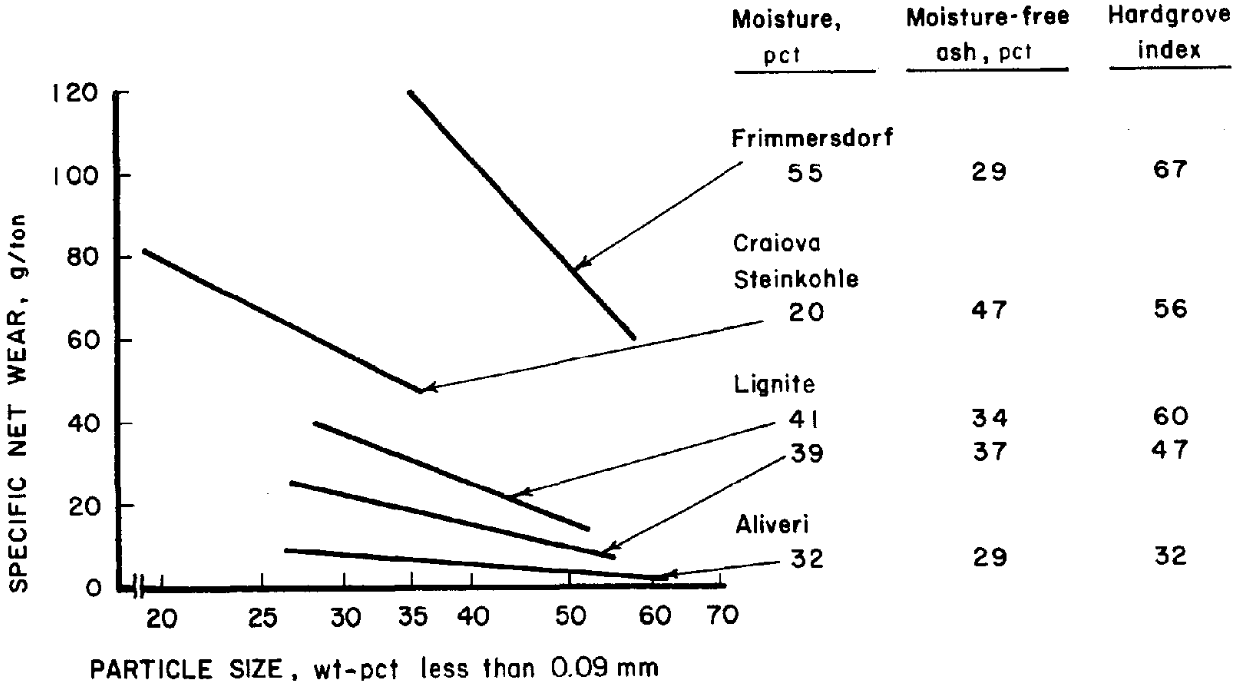


FIGURE 104. - Specific wear for various raw brown coals.

has 20 times the wear as the coal fired in the Greek powerplant Aliveri. Frimmersdorf coal causes wear that is four to eight times that of 35-pct-ash lignite used in the Rumanian powerplant Craiova. Even knowledge of the percentage of SiO_2 in the chemical analysis of the ash would not enable a prediction of actual wear.

Special measures are required at the following points: In the bends of the pulverized-fuel ducts, at the burner, at the flue gas recirculation duct openings, at the tube penetrations, at the bends of the gas ducts, and at the internal stiffeners in the gas ducts. The geometry of bends as well as bifurcations was optimized to minimize wear by model flow tests. As the effect of wear sometimes increases with the cube of the gas velocity, velocities in most cases are limited to about 10 m/sec (33 ft/sec).

Ash Removal

For removal of furnace ash--about 10 to 20 pct of the total ash--wet ash conveyors are used. The quantity of ash discharged is not constant. Under certain operating conditions there may be occasions when three to five times of the normal amount is discharged. To prevent thermal overloading of the ash conveyor, which could result in explosive pressures being developed in the trough, traveling grates are installed below the furnace hopper to guarantee a controlled flow into the ash conveyor. Various arrangements are in use. Figure 105 shows the arrangement with two grates that travel in opposed directions and discharge into a centrally located ash conveyor. The discharge position is protected from direct ash infiltration by means of a roof that is water cooled.

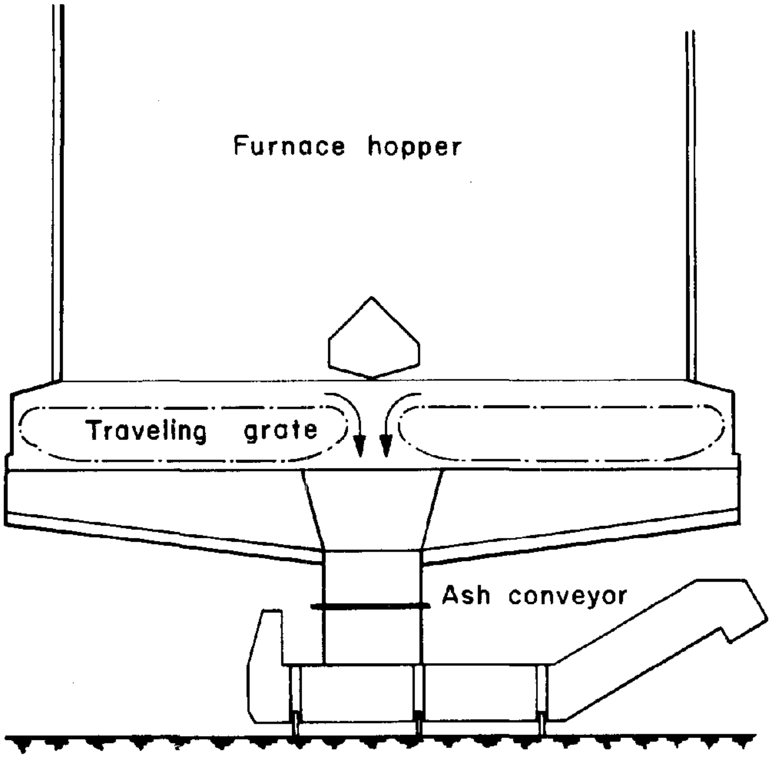


FIGURE 105. - Arrangement of grates and ash conveyor below furnace hopper.

Air-cooled traveling grates enables combustion of unburnt substance to take place. This is of special importance if the coal includes wood or fibers that cannot be ground in the mill and consequently do not completely burn in suspension.

Fouling

Ash analyses and thus ash fusion behavior of coals varies considerably even within the same mine. Coal with high alkali contents (sodium or potassium) in the presence of sulfur are especially prone to fouling.

To counteract fouling of the furnace walls, the following general rules are followed: Low heat release per cross-sectional area; low burner zone heat release; and low hot-air temperature. In most cases cleaning equipment will be required for the furnace. Water blowers have proved to be most efficient. The furnace exit temperature is of main importance for the fouling of the first convection heating surface. For economic reasons the temperature range is limited. To decrease the gas exit temperature of a 600-MW unit from 1,000° C (1,832° F) by 100° C (180° F) the furnace height would have to be increased by more than 50 pct.

To reduce fouling, the first heating surface is constructed of platens with transverse tube pitches of 600 to 1,000 mm (1.96 to 3.28 feet). For the latest units only as in line tube arrangement is used for all convection heating surfaces. Steam soot blowers are used in cleaning.

Evaporator Systems

Natural circulation is used for superheater outlet pressures up to 170 atu (2,400 psi). In West Germany for pressures between 175 and 190 atu (2,500 and 2,700 psi), once-through systems are applied exclusively. A similar tendency can be observed in some East European countries.

Both pure once-through and once-through with superimposed circulation systems are used. Furnace walls are fully welded in both systems. In the case of pure once-through systems (fig. 106), the evaporator tubes are wound spirally as high as the furnace outlet. Beyond this level, tubes continue

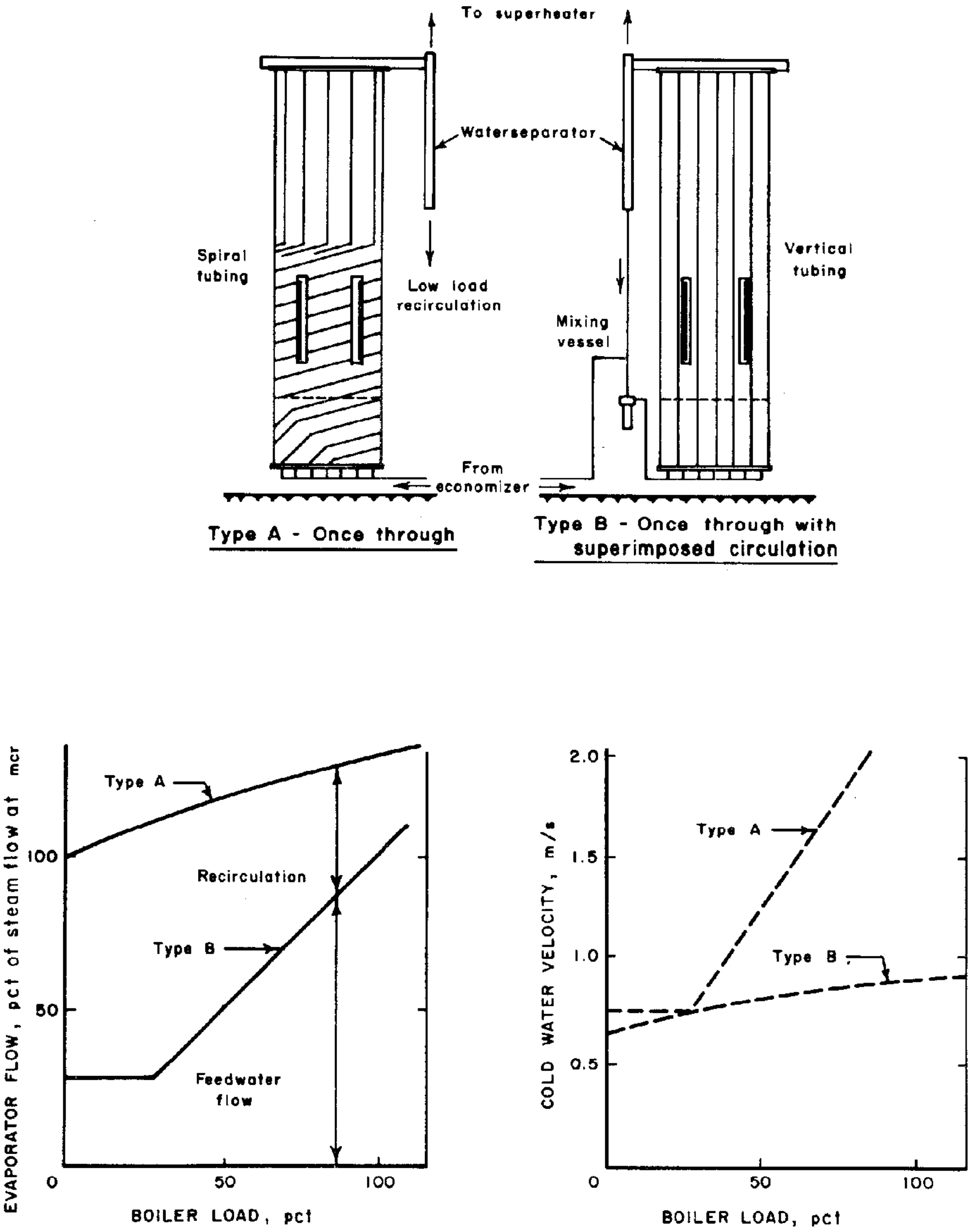


FIGURE 106. - Once-through systems.

vertically to make provision for tube penetrations of the convection heating surface elements. The flow through the evaporator decreases in direct relation to load. Below a 30-pct load, flow has to be maintained by means of circulating pumps or water recirculation to the feedwater vessel. The selection of the mass flow within the tubes has to be made with respect to the minimum load.

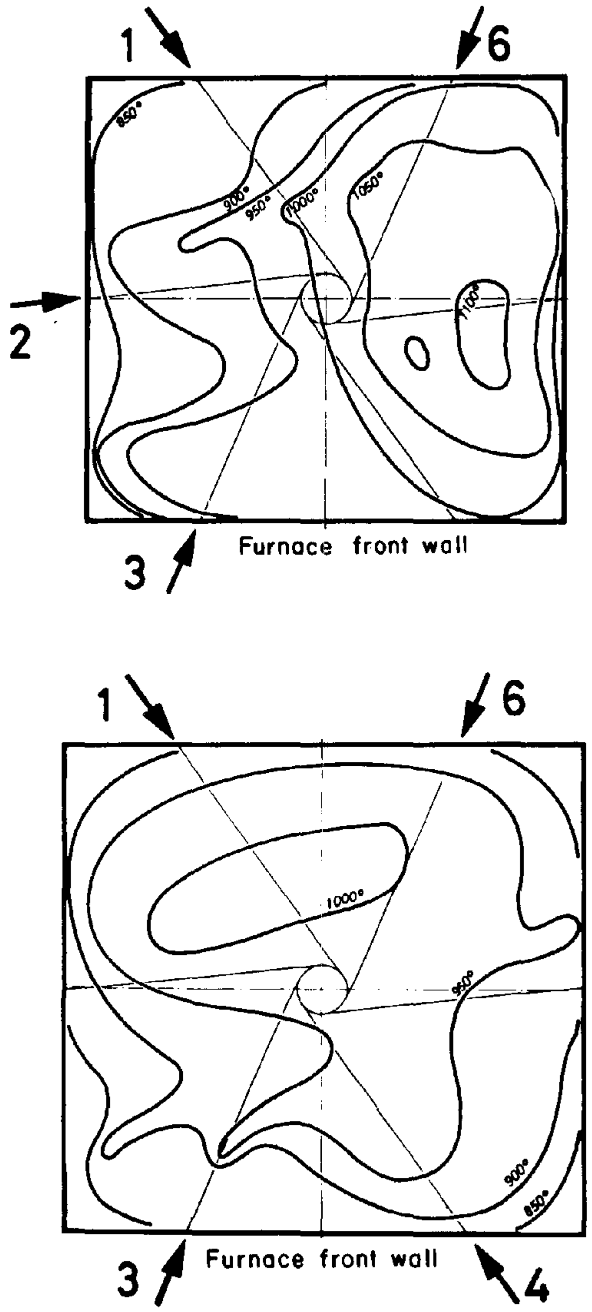


FIGURE 107. - Temperature profiles at furnace exit for different mill combinations.

For a system employing superimposed circulation, the tubes extend vertically over the whole height. The water level of the steam generator in the water separator after the evaporator is maintained by use of a feed pump. A circulating pump positioned in the main stream circulates a mixture of feedwater plus recirculated water from the separator. A nearly constant flow is maintained by the circulation pump over the load range that permits the mass flow to be selected according to the minimum control rate (MCR) point. Shorter tube length and lower mass flow are responsible for low evaporator pressure drop. The vertical tube system facilitates the design of the enclosure of burners and gas recirculation duct openings.

Heating Surfaces

In the arrangement of heating surfaces for brown coal-fired steam generators, the following additional factors have to be considered: Temporary variations in the heating value and different mill combinations.

Figure 107 shows two temperature profiles developed from measurements taken at the furnace outlet for a 300-MW unit equipped with six mills. The mills in operation are marked by arrows. To balance steam temperatures, the surfaces are divided into two or four parallel systems with separate temperature controls. Each of these is further subdivided into several sections. There are three to five sections for the superheater, and two to three for the reheater. The connecting piping between the series connected stages is additionally crossed in such a way that each section passes over the complete furnace width. A typical arrangement for a 600-MW unit is shown on figure 108.

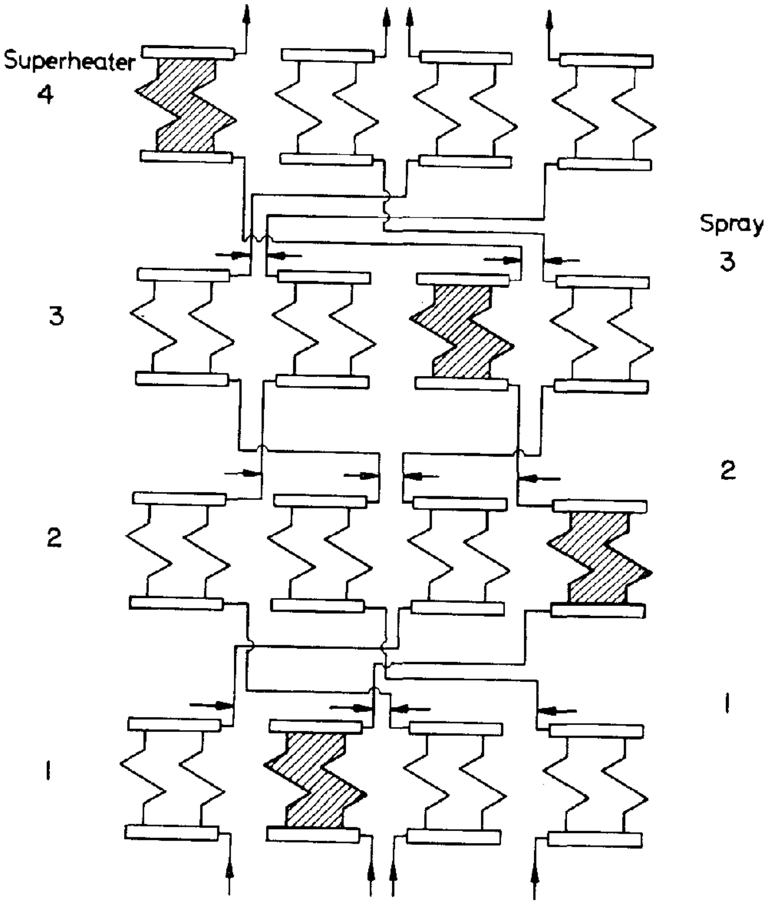


FIGURE 108. - Typical superheater flow scheme for a 600-MW unit.

Reheat Temperature Control

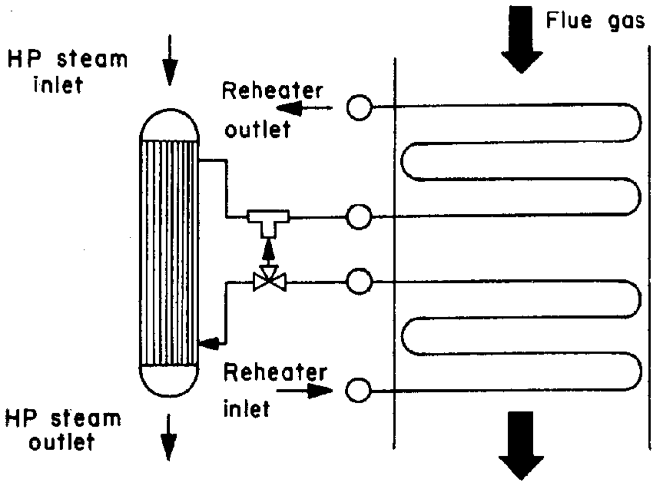
The following methods are used for reheat temperature control: Spray water injection, and heat exchanger (Biflux or Triflux).

The use of tilting burners for brown coal firing is precluded because of design reasons. Heat exchangers are applied for the large ranges in reheater control or if the ratio of water:water ash-free coal is subject to wide variations.

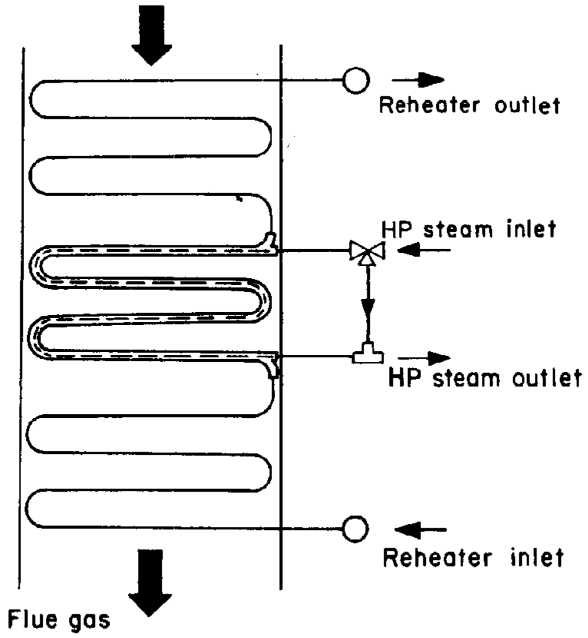
The Biflux system (fig. 109) consists of external heat exchangers where the reheat steam is heated using high-pressure (HP) steam. Temperature is changed by varying the reheat flow via three-way valves. In the case of the Triflux the heat-exchange surfaces consist of reheater tubes heated by flue gas in which concentric high-pressure tubes are positioned. Control of temperature is influenced by variations in quantity of the high-pressure steam.

Startup

Before the first mill can be started, furnace temperatures have to be established that are sufficiently high to warm the mill through the gas recirculation duct. Either light or heavy oils are used for this purpose. The capacity of the ignition firing should be not less than 25 pct of rated load in order to allow an uninterrupted increase in temperature. Figure 110 shows a startup diagram for a 300-MW unit after a 48-hour shutdown period. After about 30 min of operation with oil, the first mill is started, warmed up, and control initiated. After a further period of 60 min for temperature increase in the superheater and reheater, the second mill is started. After synchronizing and loading of the turbine, the third, fourth, and fifth mills are started at short intervals. In the case shown, the oil firing was kept in operation until the end of the startup procedure for safety reasons. Normally oil is only used for low load or if the heating value of the coal falls conderably below the design parameter.



Biflux system



Triflux system

FIGURE 109. - Reheat temperature control by use of heat exchanger.

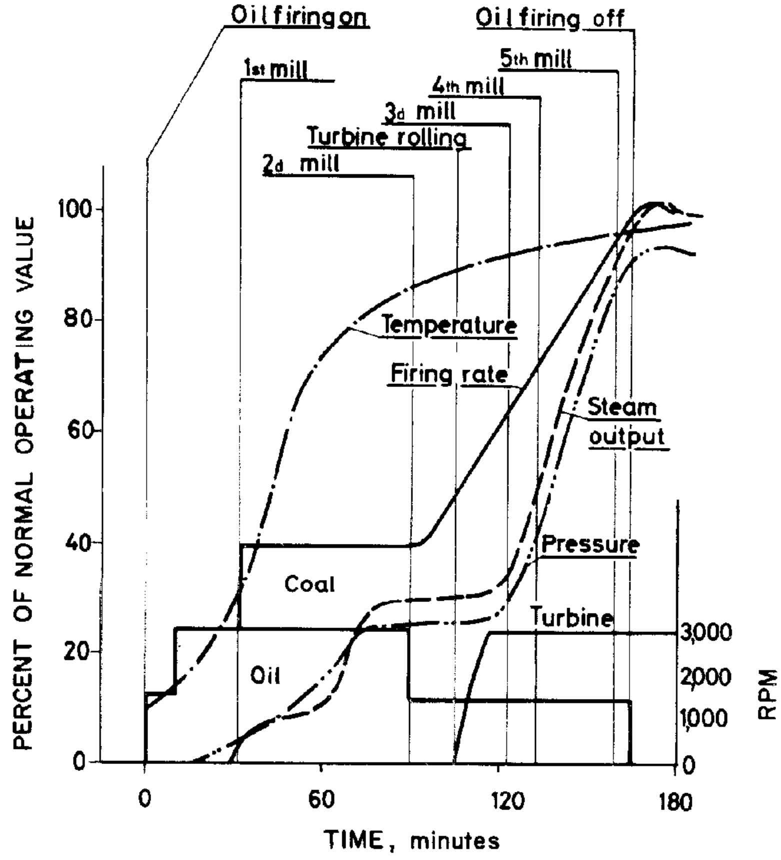


FIGURE 110. - Startup diagram for a 300-MW unit.

Development Trends

The steam generator in figure 111 is part of a 300-MW unit designed for the following parameters:

Steam output, 916 tph (2,020,000 lb/hr);

Pressure superheater outlet, 175 atu (2,490 psi);

Superheater/reheater temperature: 542°/542° C (1,007°/1,007° F).

The steam generator is of single design operating in the once-through principle with superimposed circulation. The cross section of the furnace has a width of 16.7 m (54.6 feet) and a depth of 15 m (49.1 feet). The total height of the unit amounts to 80 m (262 feet). The milling system comprises of eight beater-mills type KSG N240, each designed for a coal throughput of 95 tph. Because of the low heating value, the vapor firing mode was selected. If the calorific value of the coal falls below 1,000 kcal/kg (1,800 Btu/lb), oil support firing has to be introduced. Three additional steam generators of smaller capacity are operating at the same station. The initial unit has more than 100,000 hours of operation with a high degree of availability.

In Germany, several 300-MW units have been in operation over longer periods. Encouraged by the operating results of these boilers, six 600-MW units were ordered by Rheinisch Westfälische Elektrizitätsgesellschaft, Essen (RWE). The first one will be on load by the end of 1973. Figure 112 shows a section through the steam generator.

The parameters are the following--

Steam output: 1,886 tph (4,160,000 lb/hr).

Pressure at superheater outlet: 175 atu (2,490 psi).

Superheater/reheater temperature: 530°/530° C (986°/986° F).

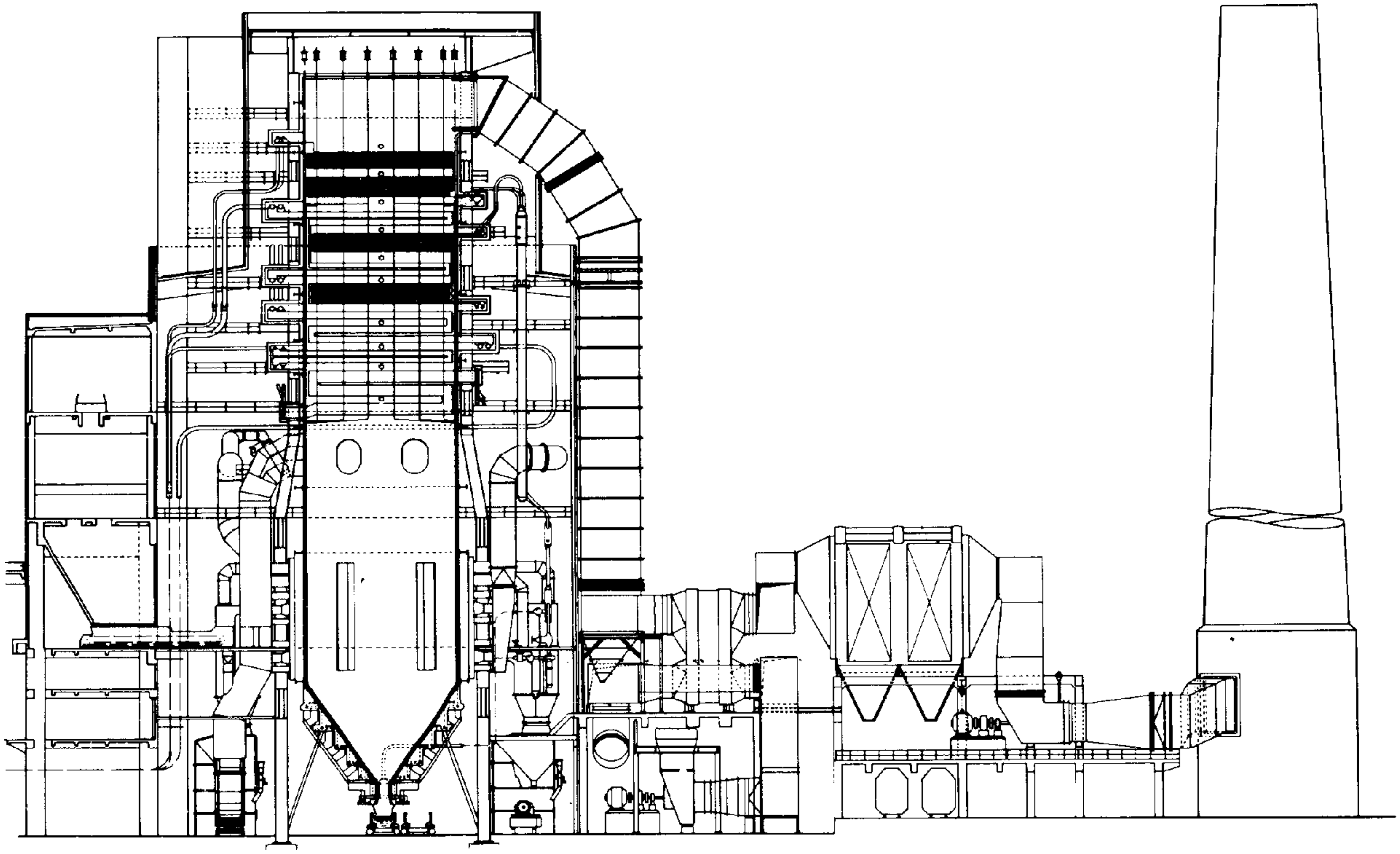


FIGURE 111. - Steam generator for a 300-MW unit (Greece).

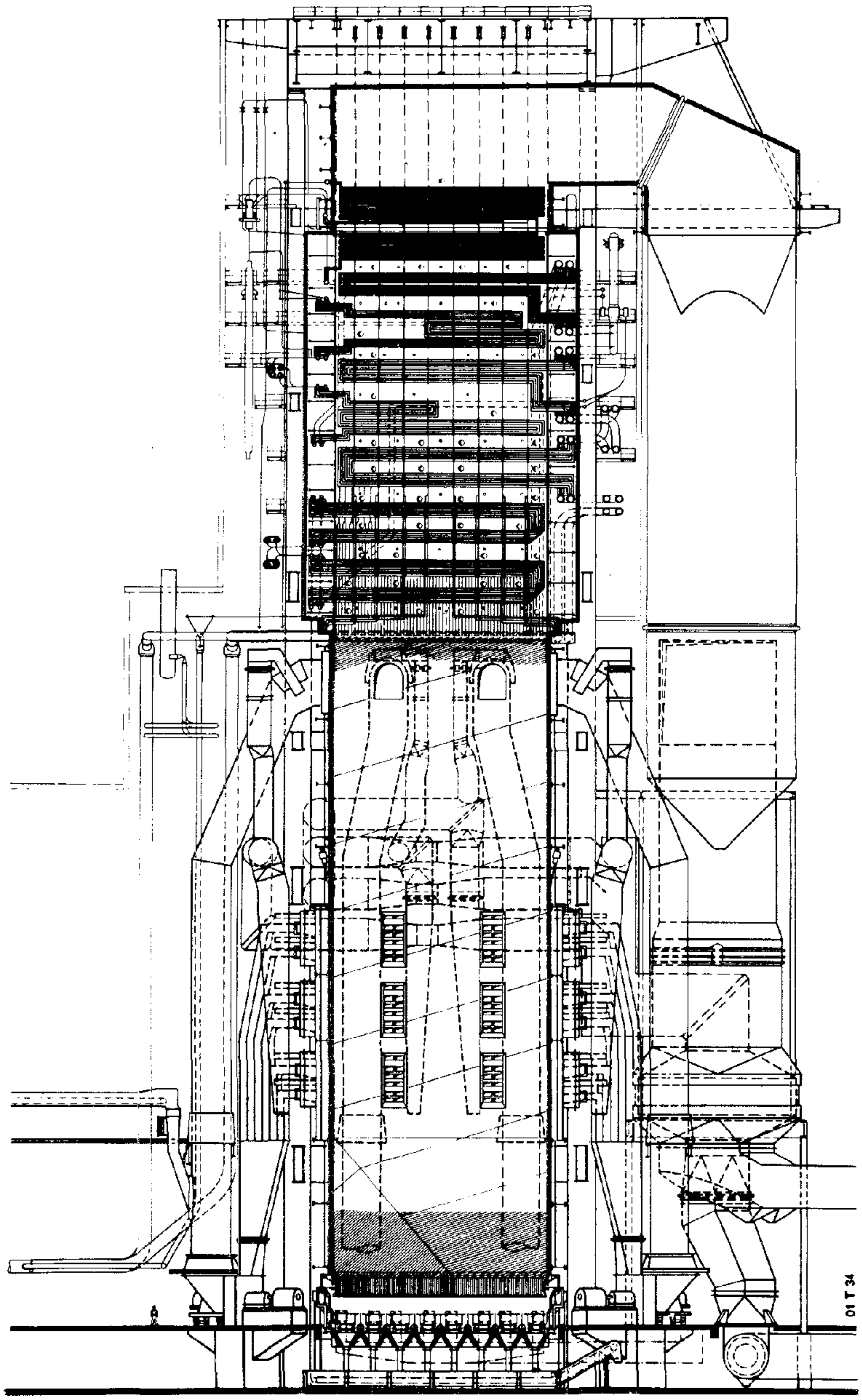


FIGURE 112. - Steam generator for a 600-MW unit (Germany).

The furnace volume is 22,000 m^3 (710,000 cu ft). The total height is 125 m (410 feet). Furnace design is based on pure once through with spirally wound evaporator tubes. The basic heating value of the coal is 1,650 kcal/kg (2,970 Btu/lb) with moisture contents up to 57 pct and ash contents up to 12.5 pct permitting the use of a firing system without vapor separation. For grinding and drying of 900 tph of fuel, eight mills of the type KSG N 270 are required; each mill is designed for an output of nearly 130 tph.

NEW DAY DAWNS FOR GREAT PLAINS COALS[81]

by

Carl E. Bagge[82]

Introduction

Anyone connected with the energy industry in this country today, and anyone concerned with our future supplies of energy, comes to the West with the same sort of respectful awe that Ebenezer Scrooge might feel on a visit to Fort Knox. The coal reserves of the West represent a repository of energy so enormous, and to date so little developed, that we as a nation have only begun to realize their potential.

The vast coal reserves of the West are a key not only to our continued prosperity and comfort but to our national security and perhaps our very survival as a major world power.

A few years ago--entirely too few, as it turns out--that statement would have been dismissed as the ravings of either a zealot or a chamber of commerce secretary. Today it is so widely recognized as to be almost a cliche--but a cliche that loses none of its truth through repetition.

The present energy crisis did not burst upon us without warning. There were warnings aplenty, but we as a nation pressed onward heedlessly toward trouble.

Consequently, at present we are importing a quarter of our petroleum, and increasing that share almost day by day. To pay for that oil, we are throwing our international trade out of balance, and have twice devalued the dollar in moves caused at least in part by our petroleum imports.

Moreover, after 2 decades of short-sighted wellhead price regulation, we are running out of natural gas and are preparing to compound the damage to our trade balance by importing vast amounts of liquefied natural gas and to make synthetic gas in this country from imported petroleum feedstocks.

Most of the additional oil we must import is not available from Canada or Venezuela, our chief suppliers in the past. We must turn to the Middle East, where some governments are openly hostile to the United States and where many of the producing countries are either nationalizing the oil industry outright or demanding larger and larger shares of ownership. Since Western Europe and Japan are even more dependent than we are upon Middle East oil, higher prices may be only the least troublesome results we can anticipate. There are some reports already that Saudi Arabia, which has been a friendly and reliable

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[81] Banquet address.

[82] President, National Coal Association, Washington, D.C.

source of oil for the United States, has suggested certain alterations of our foreign policy in return for their oil.

How much longer will it be before more Middle East countries make such suggestions--and before they become not suggestions but requests and then demands? And if that happens--let me be optimistic and say "if," not "when"--what are our alternatives?

These are the prospects, economic and diplomatic, that President Nixon addressed in slightly more veiled and diplomatic language in his April 18 energy message. The Persian Gulf lies nearly half a world away from Grand Forks, but in this symposium you are discussing one of our prime alternatives.

Coal as Alternative in the Energy Crisis

The thrust of the President's message was a recognition of the need to make America more nearly self-sufficient in energy resources. He said we have the capacity and resources to avert a major crisis if only we take the proper steps and take them now.

The first thing to be done in attacking a complex problem is to list your assets, and the President began his list with these words:

> "More than half of the world's total reserves of coal are located within the United States. This resource alone would be enough to provide for our energy needs for well over a century."

He went on to take notice of the fact that the extraction and use of coal is so beset by environmental problems that less than 20 pct of our energy needs are met by coal and the health of the entire coal industry is seriously threatened.

He spoke of coal's problems, saying that if the use of coal is further reduced, "we would have to increase our oil imports rapidly with all the trade and security problems this would entail."

President Nixon noted that coal production has been limited by competition from natural gas at prices held down by unrealistic Federal price regulation at the wellhead, and by environmental concerns, mine health and safety regulations, and by uncertainty over Federal and State strip mining regulations.

While recognizing all these problems, the President said, "I urge that highest national priority be given to expanded development and utilization of our coal resources."

So far, so good. But I must say in candor that the coal industry does not feel the President went far enough or did enough good. He did urge customers to use coal when they can. He urged governors to make this easier by delaying secondary air quality regulations where health concerns are not involved. He urged State utility commissions to allow utilities to pass along to customers, as part of the fuel charge, the costs of pollution control equipment, including both stack scrubbers to remove sulfur dioxide and coal gasification equipment.

Status of Coal Research

The President made no recommendation for increased research funds for coal, beyond the fairly modest amounts proposed in the 1974 budget. For coal research and development this comes to only $120 million, even including mine health and safety research and a previously uncommitted $21 million fund in the Department of the Interior. The total for coal research is $200 million less than the Atomic Energy Commission is allotted next year for work on the Liquid Metal Fast-Breeder Reactor alone.

I do not suggest taking away a nickel of the funds for the fast-breeder reactor. I think we are going to need that machine eventually. But even its staunch advocates say that it is 20 to 30 years away from commercial application--and when we get it, it will be just another way to make electricity. Except as it might free other fuels for other uses, it will not help our urgent need for hydrocarbon fuels for transportation, home heating, and industrial use. Somehow, the scale of spending proposed for coal research does not jibe with the concept of making increased development of coal our highest national priority.

President Nixon did declare that when additional research is shown to be essential, he will do his best to provide the funds. That is fine as far as it goes--but it does not seem to allow more funds now for investigation of promising new lines of research that may have been pigeonholed because of the cheeseparing financing of coal research in the past. Researchers cannot make a case for a stepped-up program until they have made a beginning--and this approach seems to allow no new beginnings.

The Case for Western Coal

Research has a direct bearing on the future of coal in the West. That future looks good by any rational standard. However, several things are necessary to turn the great promise of western coal into performance, and one of them is expanded and accelerated research. For proof of this, we need look no further than the fact that the need for synthetic gas from western coal is already upon us, and our domestic research on gasification methods is still in the pilot plant stage.

The vast reserves of coal in the West have been known and used locally ever since the demand for fuel outstripped the handy supply provided by the buffalo. However, except for essentially local markets, there was little development of western coal. Ironically, the current increase in the use of western coal is based at least in part not upon research but upon the lack of it--the lack of proved and commercially available devices for removing sulfur oxides from the stack gases of plants burning the higher sulfur coals of the Middle West.

This has opened new markets for low-sulfur coals from Wyoming and Montana--markets that were formerly out of reach because of freight costs. With aggressive use of unit trains and with the economies attainable by surface mining thick seams, western coal producers are serving utilities more than a thousand miles away, in States like Iowa, Minnesota, or Illinois, and are bidding for markets even further east.

This eastward movement may be replacing high-sulfur coal from established mines, and in due course it will have to compete against local coals used with stack scrubbers when that technology is available for control of sulfur oxides.

But meanwhile, western coal is winning new customers in areas that would flabbergast any oldtime coal man. For example, a new powerplant at Avinger, in east Texas--the erstwhile stronghold of natural gas--will be supplied with coal from a mine at Gillette, Wyo., 1,483 miles away. That is a movement southeastward into new territory, and it is a long-term contract. Another company has contracted to supply a powerplant at Muskogee, Okla., from a mine in Campbell County, Wyo.

The west coast, of course, has been reaching out to tap western coal reserves via long-distance power transmission. From the great generating stations of the Four Corners area to the Mohave plant in Nevada and northward to the Pacific Power and Light plant at Centralia, Wash., western coal is being translated into electric power and is moving west.

In these cases a little more research is involved--the studies that made possible the long-distance transmission of power at high voltages, and the Consolidation Coal Co. research of more than a decade ago that developed the concept of a coal pipeline which now feeds the Mohave plant from the Black Mesa mine. Essentially, however, these beginnings of new demand for western coal do not reflect results of any recent research into new methods of mining and using coal.

It is time, therefore, for the Nation to seriously address itself to the steps necessary to allow fuller utilization of western coal. Indeed, it is far past time, and we must redouble our effort to make up some of the years we have lost.

Lignite, like some fine French wines, does not travel well. Neither is it readily substitutable for other coals in existing powerplants. Therefore, it has not benefited so directly from the rising demand for fuel in other areas. But lignite has some great advantages--it is present in marvelous abundance, and it is easily accessible at reasonable cost.

Gasification Research

For lignite, then, as well as for other western coals, intensive research is overdue. As you know, modest gasification research is underway. I was privileged to attend the dedication last August of Consolidation Coal Co.'s pilot plant at Rapid City, S. Dak., where the CO_2-Acceptor Process is undergoing tests and further development. Early engineering estimates indicate this will be one of the most economical methods of creating high-Btu gas from coal, and we are following its progress with great interest. That pilot plant will operate for 2-1/2 years, and then a decision will be made on whether to go ahead with a larger unit. Construction of that commercial unit will take additional time.

The CO_2-Acceptor Process is only one of several processes under development. The Institute of Gas Technology is operating a pilot plant for the HY-GAS process at Chicago, and the National Coal Association's affiliate, Bituminous Coal Research, has developed the BI-GAS process for which a pilot plant is being built at Homer City, Pa. All these are financed by the Office of Coal Research and the American Gas Association. The Bureau of Mines is in the pilot plant stage with its Synthane Process at Bruceton, Pa., and has another process, called Hydrogasification, under test in its laboratories.

The Division of Engineering of the National Research Council recently prepared a report for the Office of Coal Research evaluating coal gasification processes. It recommended that pilot plant work should go forward on these processes in the pilot plant stage and more work be done on the Hydrogasification process. It set a timetable that would bring these projects to a point of decision in 1975--the decision being construction of a large-scale demonstration plant. The plant should operate satisfactorily for 1 year, the National Academy said. Presumably, at that point government research would cease and industry would step in with commerical-scale plants.

This is to my mind a fairly leisurely schedule, though the National Academy apparently considers it consistent with its first recommendation, which says, "Modern technology should be developed and applied as rapidly as possible to coal-gasification plants."

There is a saying in Washington these days that you cannot make problems disappear by throwing dollars at them. I do not know how we can be sure of that in coal research--it certainly has never been tried. Some dollars for bets of research long shots and to speed up the promising research already underway might well be the best investment our Nation has made since the Louisiana Purchase.

The need for more research is underscored, paradoxically, by the fact that major companies are going ahead without it. At least two major gas firms are investing hundreds of millions of dollars in coal gasification plants in New Mexico that are based on the German-developed Lurgi process. They will add a methanation process to the Lurgi system if one can be developed, but they are going ahead regardless. Obviously, their need for gas is so great they cannot wait to see if a more efficient or economical system can be developed.

Need for Synthetic Crude Oil From Coal

Gasification is not the only new prospect for western coal. We also need research to develop processes for producing synthetic petroleum from coal. Coal-based petroleum is vital to America's future not only for the domestic-based energy supply it would provide us but because it would give us needed leverage in our negotiations with the oil-producing states of the Middle East.

Unless we have an adequate alternative to our steadily increasing dependence on oil from the Middle East and our pending heavy reliance on liquefied natural gas from Algeria and perhaps elsewhere in that region--and from Russia, we will stand in grave danger of losing our independence in foreign affairs. When the volatile and often hostile governments of the Middle East have the power to control a major part of our energy supply, it is putting great trust in human nature to believe they will not use that control for their own purposes.

Even if we are not subjected to that kind of pressure in terms of policy, we are certain to be under heavy economic pressure. Already the nations of the Organization of Petroleum Exporting Countries are negotiating higher prices and demanding more control of their petroleum production through either increased shares of ownership or outright nationalization. Moreover, we are competing for that oil against Western Europe and Japan, which not only sends the price up but raises the prospect of some damaging future confrontation either with the supplying nations or between the consuming nations.

Finally, and at the very least, we are already spending so much for foreign oil that our international payments are dangerously out of balance. This has already led to devaluation of the dollar--which in turn leads the OPEC nations to raise prices again, and the spiral continues.

I began this speech with a remark about a figurative visit to Fort Knox. If we continued along the path we have begun and import oil and gas at the rates now being forecast, anyone who wants to inspect the contents of Fort Knox a few years from now will have to visit the Persian Gulf.

These are the reasons, or at least some of them, that the President called for a new energy policy in which we depend as much as possible upon our domestic resources. Although we in the coal industry do not believe he went far enough nor fast enough, he has recognized the prime importance of developing and using our coal reserves. We hope the Congress will take him at his words rather than his deeds.

Reclamation of Mined Lands

President Nixon did say one more thing about coal that I have not mentioned. He called for speedy passage of a strip mining and reclamation bill, so that the industry will know the rules under which it must operate and can proceed with its plans.

We at the National Coal Association support this stand. We support Federal legislation to help the States do a better job of reclamation, and we hope Congress passes a reasonable bill as soon as possible. We know that mined land can be effectively reclaimed because responsible coal companies are doing it, not only in the East but in the more challenging conditions of the

West. Of course they have made mistakes in the past when they were learning not only the necessity but the methods of returning land to productive use. They have also learned from those mistakes and resolved not to repeat them.

An outstanding example of industrial good citizenship was the joint declaration last fall by all the companies surface mining lignite in North Dakota, pledging themselves to good reclamation practices. As an example of one company's policy, I was struck by reading the testimony of my good friend Otes Bennett, President of the North American Coal Corp., to the North Dakota Commission last February. Mr. Bennett said that North American as a matter of policy removes and saves the topsoil before mining. Afterward, any remaining highwall is reshaped to match the surrounding terrain, the topsoil is replaced, tilled, fertilized, and seeded. The result is gently rolling terrain that blends with adjacent land and can be traversed by farm machinery. His company has announced a policy of restoring land to an equal if not more valuable condition than before mining, and in the course of this plan, it intends to spend far more than the surface value of the land.

I would like to call this policy to the attention of those environmental critics of surface mining who claim surface coal mine operators are out only to loot the land and at the most will create only a few showplaces to exhibit to visiting Congressmen. If these are showplaces, they will be some of the biggest on record, covering multiple thousands of acres.

Impact of Gasification Plants

North American has agreed to reserve 2.5 billion tons of lignite to supply future gasification plants of Michigan Wisconsin Pipe Line Co. in five coal reserve areas.

Michigan Wisconsin has applied for water rights from Lake Sakakawea and the Missouri River to supply its proposed gasification plants, and I understand that the State is considering this application along with other possible applicants. Without endorsing or lobbying for the Michigan Wisconsin case to the detriment of any others, I would like to review some of the highlights of the proposal to show the dimensions of the future for North Dakota lignite.

The company estimates the cost of developing the necessary mines would exceed $1 billion, and the investment in coal gasification plants would surpass $5 billion. Just one plant, producing 250 million cu ft of gas per day and consuming 10 million tons of lignite a year, would represent a capital cost of $459 million for the plant, the mine, and for gas and water pipelines. It would have a total payroll of more than $12.5 million a year and would employ more than 1,000 persons.

The pipeline company says its first plant may be in operation by 1980, and it might build at least three more plants at 2-year intervals. On the basis of the first four plants, this coal gasification complex would furnish more than 7,700 jobs, create $117 million a year in direct payrolls and indirectly generated funds, and furnish more than $14 million a year in additional tax revenues to the State of North Dakota. This would have a terrific impact in North Dakota, where both population and employment declined by more than 2 pct in the 1960's and where median family income is less than 82 pct of the national average.

This can be the shape of the future--and I am sure this vast project will not be the only one using North Dakota lignite. Multiply it by the other plants that can be based on the great coal reserves of the Powder River and Fort Union deposits, and on other western coal deposits. Consider gasification plants, synthetic oil plants, electric utility plants, and general industry.

But the future will not arrive unbidden. We need accelerated research to make it possible. We need a strip mine law that regulates but does not prohibit responsible surface mining. We need the Department of the Interior to resume coal leasing on public lands. We need to rethink our attitude toward the environment, so that we give proper weight also to concerns of national security and economics in attaining an adequate energy supply. We need national policies that give reasonable assurance of a market for synthetic fuels if we are to attract the necessary financing.

But we as a nation can do these things. We should have started earlier, but at least we can start now. We can make a truly great new day dawn for western coal--great for the West, and great for America.

UTILIZATION OF LIGNITE AND SUBBITUMINOUS ASH

by

Oscar E. Manz[83]

Introduction

The United States and Canada are on the threshold of making significant progress in the utilization of lignite and subbituminous ash. Total ash production is growing rapidly, and there is no indication that the growth rate will diminish during the next few years. The production of lignite fly ash alone increased from 326,900 tons in 1971 to 550,165 tons in 1972.

Since the 1967 Fly Ash Utilization Symposium held in Pittsburgh, the National Ash Association has done an excellent job of promoting the use of ash from all types of coal. The subsequent ash symposia held at Pittsburgh in 1970 and 1973, as well as many other meetings, have done much to promote the utilization of fly ash, bottom ash, and wet bottom slag.

For about a decade, the Civil Engineering Department of the University of North Dakota has been actively engaged in research directed towards practical application of lignite ash in road construction, in concrete, and in other manufactured products. In 1972 the scope of the research was broadened to include the ever-increasing production of subbituminous ash. Since June 1972, the Basic Fly Ash and Slag Testing and Research Institute,[84] as a division of the University of North Dakota Engineering Experiment Station, has been operating under the direction of the author. "Basic" ash is that ash in which the sum of the CaO and MgO is greater than the Fe_2O_3, a characteristic of most western coals including lignite and subbituminous coals.

One of the major directions of testing and research at the University of North Dakota has been towards ASTM acceptance of basic fly ashes for use in portland cement concrete. Extensive data has been gathered and six presentations have been made before the appropriate ASTM committee. Further work is continuing in the substitution of cement with basic fly ash.

Research on roadbuilding applications has already stimulated several roadbuilding jobs in North Dakota. In 1971, the first lignite fly ash-lime-aggregate base (poz-o-pac) was placed in the State.

The institute is conducting performance tests on a subgrade A-7 soil stabilization project using 3-pct lignite fly ash and 3 pct lime on I-29 in

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[83] Professor of Civil Engineering, University of North Dakota, Grand Forks, N. Dak.

[84] National Ash Association. Basic Ash Research and Testing Institute Activities Begin in North Dakota. Ash at Work, v. 14, No. 4, 1972, p. 4.

eastern North Dakota. The combination of fly ash and lime has proven superior to a 6-pct addition of lime.

A study to determine the effect of fly ash particle size on suitability of ash for mineral filler is also underway. There is evidence that the finer the fly ash the less desirable it is as a mineral filler.

Preliminary investigations with fly ash and clay mixtures indicate the feasibility of producing excellent extruded brick containing 25 pct fly ash and 75 pct clay.

This paper will report on basic ash research exclusive of that found in either of the author's previous papers at lignite symposia[85] or the proceedings of the 1967 and 1970 Pittsburgh ash symposia.[86] In addition to work being done at the University of North Dakota, some other research organizations in the United States and Canada are also involved in research on lignite and subbituminous ash.

Production and Utilization of Lignite and Subbituminous Ash

Quantities of lignite and subbituminous ash utilized throughout the United States and Canada, compared with production, are limited as shown in table 25. Although the production of lignite fly ash increased from 326,900 tons in 1971 to 550,165 tons in 1972, the percent of utilization was nearly constant. There are many lignite- and subbituminous-fired powerplants being built or in planning stages, as well as additional capacity being added to present facilities. Many units that have been using higher rank coals have been forced by limits on stack emission to use subbituminous coal. The installation of electrostatic precipitators has increased the production of fly ash at many plants, as well as producing fly ash with different chemical and physical properties. Analyses given in table 26 for Otter Tail fly ash indicate that precipitator ash has a higher CaO, SO_3, Al_2O_3, and Na_2O content and reduced concentrations of SiO_2 and Fe_2O_3. Surface area and percent retained on a No. 325 sieve are much greater for electrostatic-precipitator-collected fly ash than those obtained from mechanical collectors. During the

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[85] Manz, O. E. Utilization of Lignite Fly Ash. Paper in Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Bismarck, N. Dak., April 29-30, 1965, compiled by J. L. Elder and W. R. Kube. BuMines IC 8304, 1966, pp. 66-78.

Manz, O. E. Lignite Fly Ash Utilization. Paper in Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., May 1-2, 1969, compiled by J. L. Elder and W. R. Kube. BuMines IC 8471, 1970, pp. 138-149.

[86] Faber, J. H., J. P. Capp, and J. D. Spencer. Fly Ash Utilization Proceedings: Edison Electric Institute-National Coal Association-Bureau of Mines Symposium, Pittsburgh, Pa., March 14-16, 1967. BuMines IC 8348, 1967, 345 pp.

Manz, O. E. Ash From Lignite. Paper in Ash Utilization, Proceedings: American Public Power Association-Edison Electric Institute-National Ash Association-National Coal Association-Bureau of Mines Symposium, Pittsburgh, Pa., March 10-11, 1970. BuMines IC 8488, 1970, pp. 282-299.

next few years, many plants will be retrofitted with flue gas scrubbing or SO$_2$ control devices. Such treatment reduces the fly ash produced but will almost double the amount of bottom ash. As a result of the success of cyclone burners at the Minnkota lignite-fired plant in North Dakota, it appears that many subsequent large lignite-fired plants will have cyclone burners. Cyclone firing produces about 75 pct black glassy slag and 25 pct fly ash. Therefore, it would be rather difficult to predict the future production of lignite and subbituminous fly ash, bottom ash, and slag.

TABLE 25. - Lignite and subbituminous ash; collection and utilization, 1972[1] (United States and Canada)

	Fly ash, tons		Bottom ash, tons		Cyclone burner slag, tons	
	Lignite	Subbituminous[2]	Lignite	Subbituminous[2]	Lignite	Subbituminous[2]
Total ash collected.......	550,165	1,912,560	181,210	726,900	170,675	4,530
Ash utilized:						
Mixed with cement clinker or cement (pozzolan cement)......	-	1,002	-	-	-	-
Partial replacement of cement in concrete.....	12,284	64,590	-	-	-	-
Stabilizer for road bases, parking areas, etc....................	2,915	32,100	-	-	-	-
Fill material for roads, construction sites, etc....................	-	-	5,000	-	12,435	-
Filler for asphalt mixes.................	13,850	23,000	-	-	-	-
Oil well cementing......	800	11,101	-	-	-	-
Miscellaneous...........	200	1,495	300	-	52,173	([3])
Total ash utilized...	30,049	133,288	5,300	-	64,608	-

[1] In 1971, 326,900 tons of lignite fly ash were collected, and 18,227 tons were utilized.

[2] Figures for United States only.

[3] Blasting grit and roofing granules.

TABLE 26. - Estimated mineralogical composition of fly ashes

Laboratory No.[1]	M-6328	M-6329	M-6330	M-6331	M-6332	[2]9978
Mineral, pct:						
Lime, CaO	7	3	2	2	4	-
Hematite, Fe_2O_3	3	5	6	4	5	3
Magnetite, Fe_3O_4	2	3	2	2	4	7
Quartz, SiO_2	9	7	2	6	4	4
Mullite, $3Al_2O_3 \cdot 2SiO_2$	[3]8	-	-	[3]5	-	2
Periclase, MgO	[3]3	[3]3	[3]4	[3]3	[3]7	-
Carbon, C	-	9	-	3	-	9
Glass	58	60	74	65	66	65
Miscellaneous and unidentified	10	10	10	10	10	10

[1] Identification of fly ash samples:

Laboratory No.	Source
M-6328	Montana Power and Light Co. (subbituminous ash), Billings, Mont.
M-6329	Minnkota Power Co., Center, N. Dak.
M-6330	Basin Electric Power Coop., Stanton, N. Dak.
M-6331	Boundary Dam Powerplant, Estevan, Saskatchewan, Canada
M-6332	Otter Tail Power Co., Fergus Falls, Minn.

[2] Bituminous coal fly ash, Chicago area.

[3] Estimated.

Chemical and Physical Properties of Lignite and Subbituminous Fly Ashes

During 1971, the author provided the Bureau of Reclamation research laboratory at Denver, Colo., with seven samples of lignite fly ash and one of subbituminous fly ash. Tables 25 and 26 and figures 113-118 contain a portion of the resulting test data provided by Ralph Elfert.[87]

Estimated mineralogical compositions are given in table 26 for four of the lignite fly ashes, one subbituminous ash, as well as the bituminous fly ash. The identification of the fly ashes is also given. Figures 113-118 are photomicrographs from typical microscopic fields at X 200. The ashes were mounted in oil of refractive index of 1.55.

Generally, lignite and subbituminous fly ashes are mainly spherical glass particles ranging in size to about 20 or 50 micrometers, with variable but lesser amounts of irregular crystalline grains and unburned lignite and soot. The crystalline constituents include quartz, lime, magnetite, hematite, and

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[87] Elfert, R. J., Jr. Civil Engineer, Bureau of Reclamation, Engineering and Research Center, Denver, Colo. Personal Communication, Mar. 14, 1972, 20 pp.; available on request from O. E. Manz, University of North Dakota, Grand Forks, N. Dak.

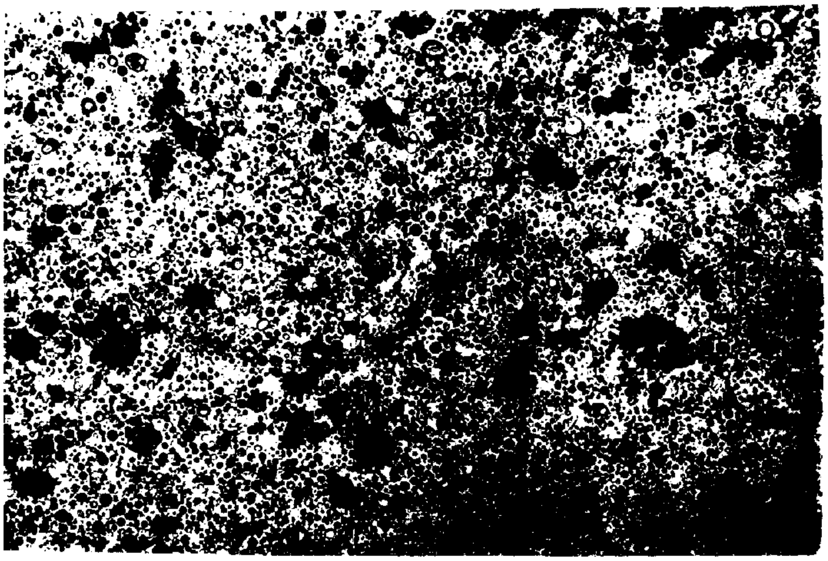

FIGURE 113. - Bituminous coal fly ash, Chicago area, laboratory No. 9978, X 200.

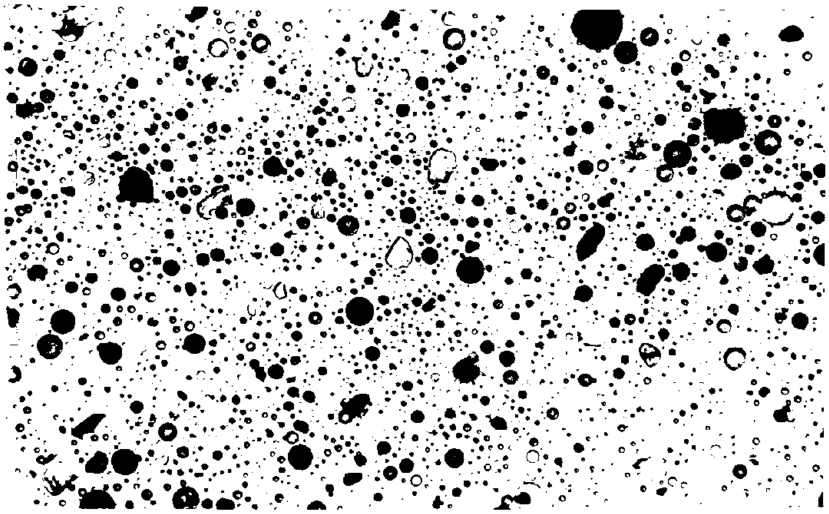

FIGURE 114. - Lignite fly ash, Montana Power and Light Co., laboratory No. M-6328, X 200.

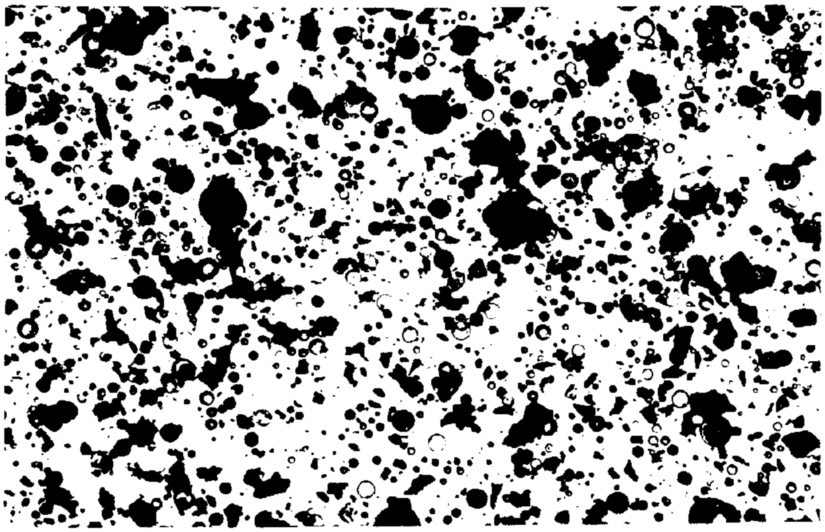

FIGURE 115. - Lignite fly ash, Minnkota Power Coop., laboratory No. M-6329, X 200.

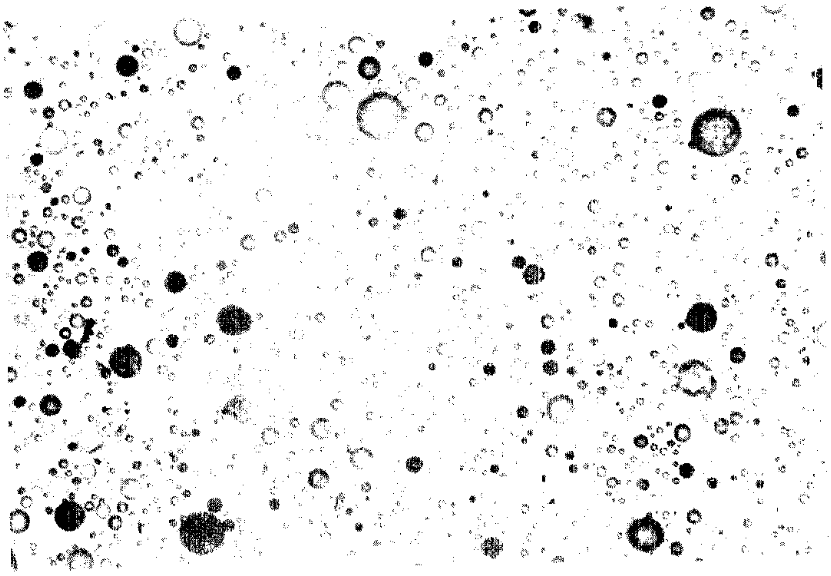

FIGURE 116. - Lignite fly ash, Basin Electric Coop., laboratory No. M-6330, X 200.

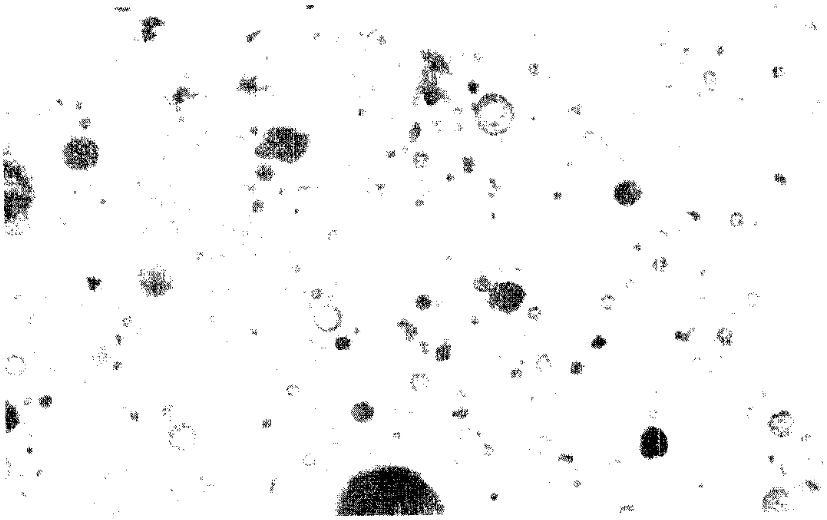

FIGURE 117. - Lignite fly ash, Boundary Dam powerplant, laboratory No. M-6331, X 200.

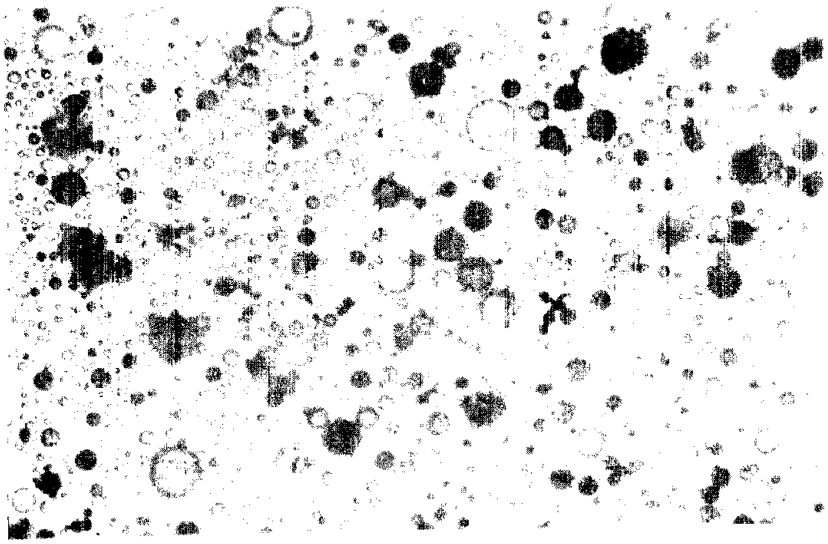

FIGURE 118. - Lignite fly ash, Otter Tail Power Co., laboratory No. M-6332, X 200.

probably mullite and periclase. The glass is the active constituent in evaluating the pozzolanic activity. A pozzolan is a siliceous or siliceous and aluminous material that in itself possesses little or no cementitious value but will, in finely divided form and in the presence of moisture, chemically react with calcium hydroxide at ordinary temperatures to form compounds possessing cementitious properties.

Differences between the samples may be noted as follows:

M-6328.--Very low carbon content. Glass index of refraction varies from 1.56 to 1.66. Glass particles are as large as 20 micrometers in diameter.

M-6329.--Considerable unburned lignite present. Glass index about 1.56 to 1.66. Glass particles are to 20 micrometers in diameter.

M-6330.--Very low carbon. Index range of glass, 1.52 to far above 1.66. Maximum glass particle size about 30 micrometers.

M-6331.--Some carbon present. Index range, 1.53 to 1.66. Maximum particle size about 50 micrometers.

M-6332.--Low in carbon. Index range of glass, 1.54 to 1.66. Maximum glass particles size about 30 micrometers.

9978.--(Bituminous coal fly ash.) Considerable carbon and iron oxides. Range of index of refraction of glass, 1.53 to 1.59. Maximum particle size of glass about 12 micrometers.

The two major differences between the lignite fly ash and the specimen of bituminous fly ash are the following: First, the index of refraction of the glass-lignite fly ash is higher, and second, the average particle size of the lignite fly ash is considerably coarser. Another difference is the presence of free lime in all the lignite fly ashes. This lime is probably "dead burned" and slow to hydrate. One effect of this would be the same as free lime in the cement clinker and another would be a tendency for lignite fly ash to set after being moistened because of reaction of the hydrated lime with the glass.

Data given in table 27 provides chemical and physical data for one subbituminous and five lignite ashes. Also included are the Federal and ASTM specifications for fly ash for use as replacement of a portion of the cement in concrete. There is considerable variation between fly ash samples with respect to fineness, strength, specific gravity, and chemical composition. None of the fly ashes comply with the present ASTM specification for Fly Ash and Raw or Calcined Natural Pozzolans for Use in Portland Cement Concrete. However, excellent results have been obtained by various researchers when fly ashes with similar properties were used to replace part of the cement in concrete. The basis of the author's presentations before ASTM has been to provide supporting data for fly ashes that do not meet the present specifications.

The chemical and physical properties given in table 28 are for selected samples of five lignite and three subbituminous fly ashes. The samples were selected to show the wide variation of basic fly ashes obtained from different powerplants. None of these analyses meet the present ASTM specification for use in concrete.

TABLE 27. - Lignite and subbituminous fly ashes tested at Bureau of Reclamation laboratories

	Saskatchewan M-6331	Montana M-6328	Minnkota M-6329	Otter Tail M-6332	Basin M-6330	Otter Tail[1] M-6386	Federal specification SS-P-570B	ASTM C618-72
SiO_2....................wt-pct..	40.92	37.68	38.50	26.42	39.04	19.22	-	-
Al_2O_3..................wt-pct..	24.41	24.69	16.61	14.95	17.10	17.70	-	-
Fe_2O_3..................wt-pct..	4.83	3.85	8.57	14.35	7.38	5.02	-	-
$SiO_2 + Al_2O_3 + Fe_2O_3$.....wt-pct..	70.16	66.22	63.68	55.72	63.52	41.94	[2]75.0	[2]70.0
CaO.....................wt-pct..	16.92	24.44	19.70	23.78	19.90	28.38	-	-
MgO.....................wt-pct..	4.36	5.81	7.03	7.26	5.90	9.45	[3]5.0	-
Na_2O....................wt-pct..	6.0	.2	1.4	5.8	7.6	7.7	-	-
K_2O.....................wt-pct..	.6	.3	1.7	.5	1.3	.5	-	-
Available alkalies as Na_2O wt-pct..	1.99	.11	.62	2.25	2.13	-	[3]2.0	[3]1.5
SO_3.....................wt-pct..	.77	1.19	2.09	4.12	1.69	9.88	[3]4.0	[3]5.0
Loss on ignition.........wt-pct..	.56	.30	3.58	.09	.16	.40	[3]6.0	[3]12.0
Specific gravity................	2.28	2.55	2.47	2.86	2.71	2.88	-	-
Retained on No. 325 mesh....pct..	15.3	11.3	8.7	15.2	8.5	7.8	-	[3]20.0
Surface area...........cm^2/cm^3..	4,592	9,167	5,303	4,939	4,737	12,082	[2]6,500	[2]6,500
Pozzolan activity index, cement, 28 days.........pct of control..	73	105	81	61	76	70	[2]75	[2]85
Pozzolan activity index, lime, 7 days..........pct of control..	1,013	1,196	1,014	1,068	808	1,616	[2]900	[2]800
Autoclave expansion.........pct..	0.018	0.131	0.013	0.128	0.045	0.164	[2]0.5	[2]0.5

[1] Electrostatic preipitator.

[2] Minimums.

[3] Maximums.

TABLE 28. - Physical and chemical properties of selected lignite and subbituminous fly ash

	[1] 71-39	[2] 71-40	[3] 71-53	[4] 71-72	[5] 71-95	[6] 72-5	[7] 72-14	[8] 72-16
SiO_2..........wt-pct..	31.5	59.0	36.2	41.2	52.1	52.8	19.7	41.2
Al_2O_3.........wt-pct..	10.8	24.6	15.1	22.0	16.7	23.6	13.3	22.2
Fe_2O_3.........wt-pct..	13.4	5.5	9.6	4.8	5.0	4.8	12.7	5.0
$SiO_2 + Al_2O_3 + Fe_2O_3$ wt-pct..	55.7	89.1	60.9	68.0	73.8	81.2	45.7	68.4
TiO_2..........wt-pct..	.2	1.0	.5	.8	1.2	1.0	.5	.5
P_2O_5..........wt-pct..	.1	.1	.2	.4	.1	.6	1.6	.3
CaO...........wt-pct..	25.0	4.9	24.0	17.1	20.8	13.5	33.2	21.4
MgO...........wt-pct..	7.1	.9	7.3	4.7	3.4	1.6	9.1	5.7
Na_2O..........wt-pct..	6.3	1.4	1.1	6.2	.3	1.2	2.2	.2
K_2O...........wt-pct..	.6	.8	1.8	.5	.4	.3	.3	.1
SO_3...........wt-pct..	4.6	.5	3.9	1.0	1.2	.7	5.3	2.2
Loss on ignition wt-pct..	.3	.2	1.8	.4	.4	.7	1.9	.5
Retained on No. 325 mesh............pct..	27.7	44.9	5.4	14.6	19.2	10.0	12.7	11.2
Specific gravity......	2.77	1.67	2.48	2.28	2.44	2.24	2.58	2.48
Surface area cm^2/cm^3..	2,606	1,777	5,156	3,739	2,101	9,115	6,435	7,802

[1] Lignite fly ash, Leland Olds plant, Stanton, N. Dak.
[2] Subbituminous fly ash, Four Corners plant, Fruitland, N. Mex.
[3] Lignite fly ash, Milton R. Young plant, Center, N. Dak.
[4] Lignite fly ash, Boundary Dam plant, Estevan, Saskatchewan, Canada.
[5] Lignite fly ash, Big Brown plant, Fairfield, Tex.
[6] Subbituminous fly ash, Mohave plant, Laughlin, Nev.
[7] Lignite fly ash, Hoot Lake plant, Fergus Falls, Minn.
[8] Subbituminous fly ash, J. E. Corette plant, Billings, Mont.

Lignite and Subbituminous Ash Research, the University of North Dakota

Since 1970 there has been a concerted effort to have lignite and subbituminous fly ash accepted by ASTM for use in portland cement concrete. Beginning June 1970, and every 6 months thereafter, a presentation has been made before ASTM Section C09.03.08.01 on Pozzolans. In 1971 an Ad Hoc Task Force on Lignite Fly Ashes was formed, with Ralph J. Elfert of the Bureau of Reclamation as chairman. The author is also a member.

Supporting data has been gathered from many sources including the Institute at the University of North Dakota. A major portion of the data presented has been similar to that shown in tables 29-30. Chemical and physical data, as given in table 29, is required to determine if the fly ash complies with the ASTM specification. The presentation in table 30 of data resulting from actual replacement of cement by fly ash in concrete is the best supporting data for ashes that do not meet the present specification. The nonspecification lignite fly ashes produced higher corresponding compressive strengths in many cases than did specification Northern States Power Co. bituminous fly ash. The Ad Hoc Task Force is confident that the ASTM Specification C618-72 will be modified in the near future to permit some lignite and subbituminous fly ashes to be used as replacement for 20 to 25 pct of the cement in concrete.

TABLE 29. - Chemical and physical data for fly ashes used in portland cement concrete

Source of lignite	Basin Electric Power Coop., Stanton, N. Dak.	Northern States Power Co., Minneapolis, Minn.	Saskatchewan Power Co., Estevan, Saskatchewan	Minnkota Power Coop., Center, N. Dak.	ASTM C618 Specification Class F
Fly ash sample No.	71-39	72-18	71-72	72-19	
Chemical composition, wt-pct:					
SiO_2	31.5	46.4	41.2	41.3	-
Al_2O_3	10.8	19.6	22.0	14.2	-
Fe_2O_3	13.4	18.4	4.8	8.2	-
Total	55.7	84.4	68.0	63.7	[1]70.0
SO_3	4.6	2.1	1.0	1.7	[2]5.0
CaO	25.0	6.2	17.1	18.0	-
Na_2O	6.3	.3	6.2	1.4	-
K_2O	.6	1.1	.5	1.6	-
MgO	7.1	1.6	6.2	6.5	-
Loss on ignition	.3	1.4	.4	5.1	[2]12.0
PHYSICAL TEST RESULTS					
Fineness:					
Surface area..cm^2/cm^3	2,606	8,359	3,739	3,714	[1]6,500
Retained on No. 325 sieve..pct.	27.7	5.0	14.6	13.4	[2]20.0
Specific gravity	2.78	2.40	2.28	2.29	±5 pct
Pozzolan activity index, cement, 28 days pct of control	67.4	112.3	104.6	65.4	[1]85

[1] Minimums.

[2] Maximums.

TABLE 30. - Summary of results for air-entrained concrete[1]

Fly ash replacement, pct	0	10	15	20	25	30
FLY ASH 71-39 (BASIN ELECTRIC POWER COOP.)						
Compressive strength test results, psi:						
1 day strength	840	664	549	603	469	396
7 day strength	2,725	2,433	2,388	1,964	1,752	1,693
28 day strength	3,785	3,378	3,148	2,753	2,705	2,465
90 day strength	4,226	3,510	2,812	3,139	3,177	2,880
FLY ASH 72-18 (NORTHERN STATES POWER CO.)						
Compressive strength test results, psi:						
1 day strength	840	777	731	637	717	615
7 day strength	2,725	1,983	2,023	2,005	1,892	1,904
28 day strength	3,785	2,955	3,321	3,325	3,469	3,345
90 day strength	4,226	4,050	4,250	4,392	4,591	4,429
FLY ASH 71-72 (SASKATCHEWAN POWER CO.)						
Compressive strength test results, psi:						
1 day strength	840	754	899	812	738	698
7 day strength	2,725	2,425	2,535	2,151	2,281	2,063
28 day strength	3,785	3,743	3,837	3,719	3,920	4,070
90 day strength	4,226	4,279	4,523	4,577	4,689	4,488
FLY ASH 72-19 (MINNKOTA POWER COOP.)						
Compressive strength test results, psi:						
1 day strength	840	904	683	773	579	576
7 day strength	2,725	2,600	2,293	1,986	1,516	1,683
28 day strength	3,785	3,734	3,619	3,310	2,882	3,177
90 day strength	4,226	4,046	4,205	3,777	3,651	4,034

[1] All mixes were 4-1/2 bags cement per cu yd.

Figure 119 indicates that, in general, concrete containing fly ash replacement requires between 90 and 180 days to surpass the compressive strength of corresponding concrete with no pozzolan replacement. However, there has been many instances in which concrete with lignite fly ash replacement has had higher strengths at all ages of test, including 1 and 3 days. When performing comparative concrete tests, it is important that there be close control of slump, air content, water-cement ratio, and curing.

In North Dakota and Minnesota there have been some problems involved using lignite fly ash in concrete. Where the sodium oxide content of the ash is 6 to 6.5 pct and sulfide oxide content, 2.5 to 3.0 pct, a white efflorescence has occurred on the surface of finished concrete. However, it has disappeared in a day or two with no resulting harmful effects. On hard troweled finished slabs with the ambient temperature above 85° F, an alkali-siliceous reaction has occurred between fine shale particles in the aggregate and the sodium oxide in the fly ash. The solution of this problem will require either the selection of nonreactive aggregate or removal of fly ash from the concrete.

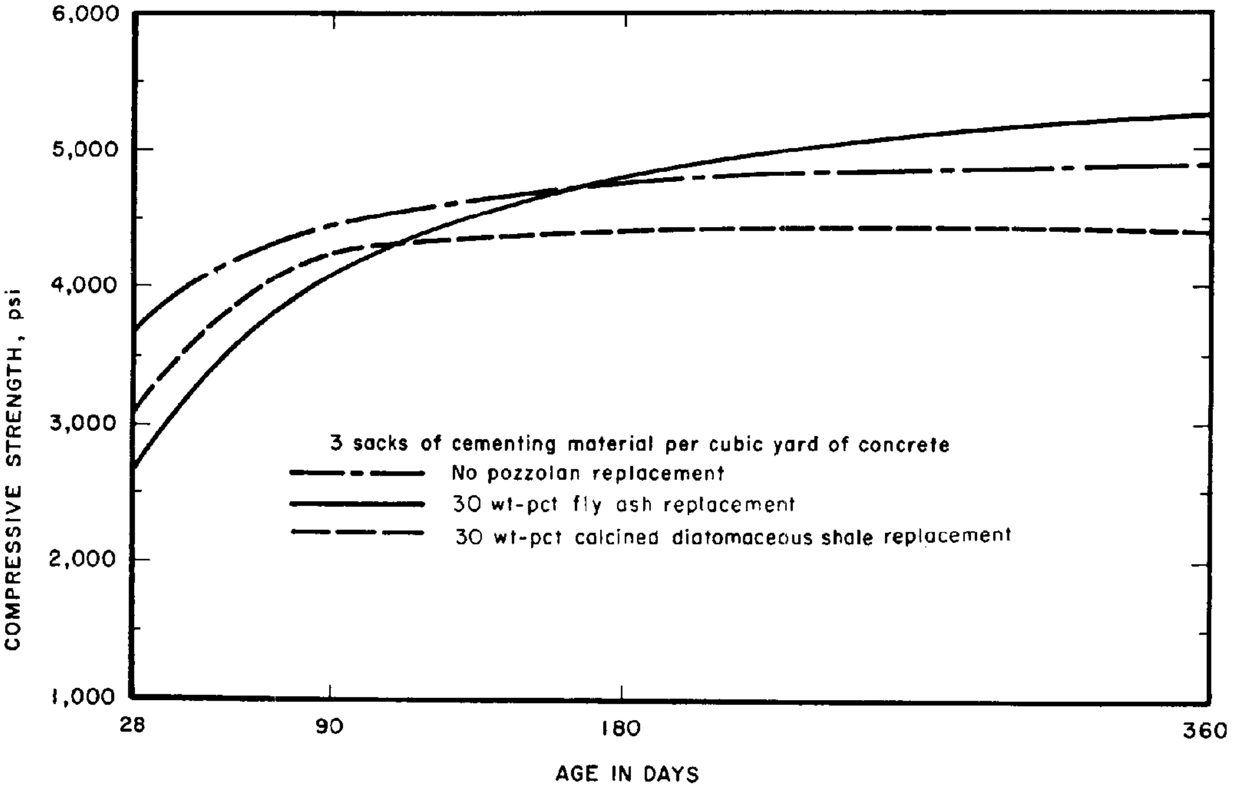

FIGURE 119. - Effect of Pozzolan on compressive strength of concrete.

The author is not advocating the use of all available lignite or subbituminous fly ashes in every concrete application.

Roadbuilding applications of lignite fly ash in North Dakota have resulted in the largest tonnage utilized to date. Poz-o-pac road base mixtures, composed of 3/4-inch maximum aggregate, fly ash, and lime have been used for several years in the eastern half of the United States. In 1971, the first lignite fly ash-lime-aggregate base (poz-o-pac) was placed in North Dakota near the site of the Basin Electric powerplant, a few miles south of Stanton. Approximately 13 pct fly ash and 2 pct hydrated lime were mixed with 85 pct aggregate and sufficient water to obtain maximum density. The laboratory procedure for determining the proper mix proportions is given in ASTM C593--Specifications for Fly Ash and Other Pozzolans for Use With Lime. During 1972, a second North Dakota project involving lignite fly ash poz-o-pac was completed.

A combination of 3 pct lignite fly ash and 3 pct lime has proven superior to a 6-pct addition of lime on a subgrade A-7 soil stabilization project on I-29 in eastern North Dakota. The institute has conducted extensive performance tests involving stabilization of A-7 soils with lignite fly ash and lime. A Master of Science thesis involving the stabilizing effect of various

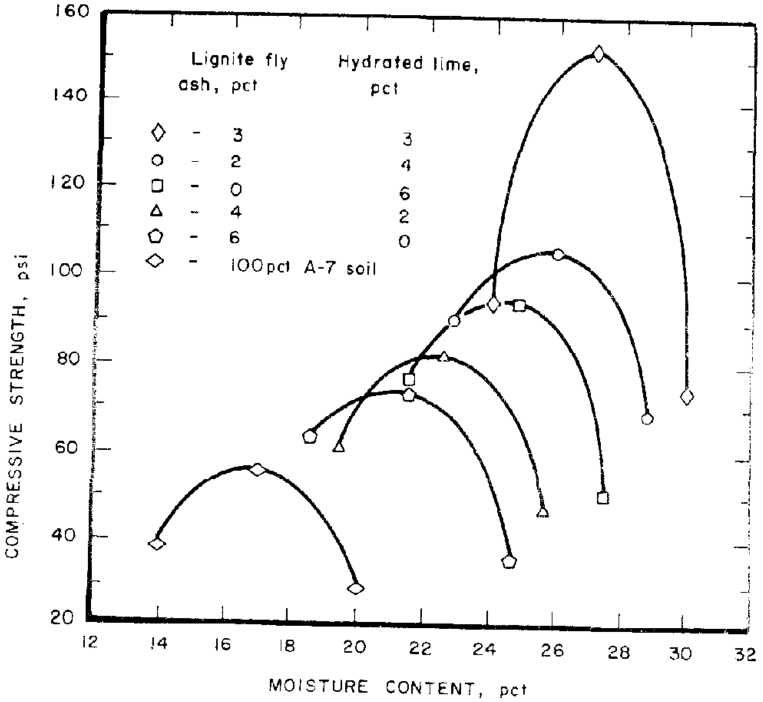

FIGURE 120. - Compressive strength versus moisture content (7 days' moist curing).

combinations of lignite fly ash and lime with an A-7 soil from I-29 has been completed by a civil engineering student at the University of North Dakota.[88]

The following conclusions were obtained:

1. Specimens containing 3 pct hydrated lime, 3 pct lignite fly ash, and 94 pct A-7 soil produced maximum compressive strength.

2. Specimens containing 3 pct hydrated lime, 3 pct lignite fly ash, and 94 pct A-7 soil produced the least amount of swell in the freeze-thaw test and the least amount of shrinkage in the wet-dry test.

3. In general, heat applied to the specimens results in maximum compressive strength.

4. Specimens compacted to maximum dry density at optimum moisture content produced maximum compressive strength.

In figure 120 the superiority of the 3-pct fly ash and 3-pct lime mixture is very evident. The I-29 project involved a 15-mile portion and utilized 7,000 tons of lignite fly ash at a savings of approximately $80,000 compared with use of lime.

In 1969 the North Dakota State Highway Department included a special provision for use of lignite fly ash as a mineral filler; in the most recent standard specifications for road and bridge construction,[89] mineral filler is specified as follows:

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[88] Derucher, K. N. Stabilization of A-7 Soil With Lignite Fly Ash and Hydrated Lime. M.S. Thesis, University of North Dakota, 1973, 48 pp.; available for consultation at the University of North Dakota Library, Grand Forks, N. Dak.

[89] North Dakota State Highway Department. Standard Specifications for Aggregates for Bituminous and Asphaltic Construction. Section 810 in 1971 Standard Specifications for Road and Bridge Construction. Bismarck, N. Dak., July 1971, pp. 576-581.

810-5 MINERAL FILLER

810-5.1 GENERAL REQUIREMENTS. Mineral filler shall be approved by the Engineer, shall be nonplastic, and shall consist of limestone dust, portland cement, hydrated lime, crushed rock screening, or fly ash.

810-5.2 GRADATION. Not less than 98 pct of the mineral filler shall pass a No. 30 sieve, not less than 85 pct shall pass a No. 100 sieve, and not less than 65 pct shall pass a No. 200 sieve.

810-5.2.1 For the purpose of these specifications, fly ash is defined as the finely divided residue that results from the combustion of lignite and is transported from the boiler by flue gases and shall conform to the following:

Moisture content Maximum percent.............3.0
Loss on ignition Maximum percent.............6.0

Subsequent to the 1969 special provision, the loss on ignition was changed from 1.25 to 6.0 pct, maximum. This was due in part to data presented in table 31. Lignite fly ashes collected from conventional powerplants have ignition losses under 1 pct, whereas the new cyclone burner at the Minnkota plant produces fly ash with much higher ignition losses. Variations in ignition loss from 1.9 to 9.1 do not seriously affect the Marshall stability or air voids values of compacted asphaltic concrete.

Since 1969 the North Dakota Highway Department has utilized over 25,000 tons of lignite fly ash as mineral filler. All the fly ashes used complied with the mineral filler specification, but two of the finer ashes produced rather gummy mixes that were difficult to lay. This prompted investigation of the effect of fly ash particle size gradation on performance as a mineral filler. Twelve different fly ashes were tested, including two bituminous fly ashes. It was found that the fly ashes with the highest percentage of uncoated particles, when subjected to the water asphalt preferential test, produced the gummiest mixes. Although most of the finer ashes were included in this group, there were some exceptions, so that the study is being expanded to include more fly ashes.

Several brick test panels were made up and have been subjected to exterior weathering for over 2 years. The bricks were composed of 65 pct Otter Tail lignite fly ash and 35 pct cyclone burner slag and had been made by dry pressing with a pressure of 3,000 psi. In an attempt to prevent efflorescence, the bricks were dipped in a silicone solution prior to being placed in the panels. Unfortunately, the weathering process has reduced the effectiveness of the silicone sealer and caused portions of most of the bricks to scale. It would appear that one must select fly ashes with very low Na_2O content (about 1 pct) in order to prevent efflorescence.

TABLE 31. - Minnkota fly ash (Center, N. Dak.)[1]

Sample	Specific gravity	Pct retained on 325 mesh	Ignition loss, pct	Marshall stability	Air voids
70-59	2.46	13.3	5.7	1,080	9.6
71-42	2.54	7.1	1.8	1,023	10.3
71-43	2.17	20.7	8.4	720	10.3
71-44	2.41	8.2	4.0	780	7.6
71-45	2.22	11.4	5.4	852	10.2
71-46	2.38	12.5	6.1	1,018	9.9
71-47	2.26	15.9	9.1	994	10.3
71-48	2.24	13.0	6.6	1,197	6.7
71-51	2.44	6.6	2.7	1,071	10.3
71-52	2.24	11.9	4.6	1,042	6.9
71-53	2.48	5.4	1.8	955	6.9
71-54	2.28	12.4	6.6	1,029	8.9
71-55	2.45	6.0	1.9	1,039	9.6
71-58	2.27	14.8	4.6.	925	11.6
71-59	2.31	11.9	4.6	955	10.4
71-61	2.39	10.4	3.7	955	7.5
71-63	2.21	16.9	6.3	1,056	10.7
71-65	2.40	9.1	3.0	1,128	7.6
71-67	2.22	14.3	5.1	1,051	8.8
71-68	2.42	10.4	5.1	1,103	6.0
71-69	2.24	13.7	6.4	1,080	8.8
71-71	2.21	11.0	7.4	1,310	7.6
71-41[2]	2.88	17.0	.3	1,041	5.9

[1] 6 pct SC-3,000, 3 pct fly ash.
[2] Otter Tail fly ash.

Further investigation into the feasibility of making bricks from fly ash has involved fly ash and clay mixtures. If the proportions are about 25 pct fly ash and 75 pct clay, the mixture can be extruded from a die as a stiff mud, and the bricks can be cut from the column by wires. Since most brick plants use the stiff mud process, the utilization of fly ash as another ingredient would not involve much capital investment. The addition of 25 pct fly ash can produce some pleasing colors and textures while still meeting the ASTM specifications for clay brick. To be economical, the source of fly ash must be very near to the brick plant because cost for clay is also low.

In an attempt to utilize dry bottom ash, wet bottom slag, and scrubber sludge, it is planned to initiate an extensive testing program the summer of 1973. Mixtures of fly ash, lime, and powerplant scrubber sludge will be compacted for possible road base material. The mixture used at Transpo '72 to pave some 100 acres of parking areas and access roads was composed of lime, fly ash, and sulfate or sulfite sludge. Combinations of dry bottom ash, wet bottom slag, and emulsified asphalt will be tested for possible use for base and surfacing of secondary roads. Also, the feasibility of using mixtures of dry bottom ash, wet bottom slag, and portland cement will be investigated.

Research by Other Organizations Involving Lignite and Subbituminous Ash

There are several organizations in the United States and Canada that are involved in lignite and/or subbituminous ash research.

The Texas Transportation Institute,[90] Texas A & M University, College Station, Tex., has completed a study involving the production of a synthetic aggregate from lignite fly ash.

The Twin City Testing and Engineering Laboratory, Inc., St. Paul, Minn., is conducting a laboratory investigation of lignite fly ash for the North Dakota Department of Highways.

The Research Division of the College of Engineering at Washington State University, Pullman, Wash., has been conducting limited research involving subbituminous ash from the Centralia powerplant.

The Bureau of Reclamation Engineering and Research Center at Denver, Colo., has conducted some chemical and physical testing of lignite and subbituminous fly ashes to provide supporting data for the ASTM Ad Hoc Task Force on Lignite Fly Ash. Ralph J. Elfert, Jr., civil engineer with the Bureau of Reclamation, is chairman of the Task Force.

Manitoba Hydro, Winnipeg, Manitoba, Canada, under the direction of K. A. Lenz, concrete engineer, is conducting a thorough laboratory investigation of both a lignite and subbituminous fly ash for possible replacement of cement in concrete to be used in a hydro dam project.

The Canada Department of Regional Economic Expansion, Prairie Farm Rehabilitation Act Engineering Service, located at the University of Saskatchewan campus at Saskatoon, Saskatchewan, Canada, has been monitoring weight and length change of concrete specimens that have been immersed in both 0.15 molar and 5 pct mixed sulfate solutions for as long as 10 to 11 years. Many of the specimens contain lignite fly ash. A concrete cylinder exposure plot is maintained near the South Saskatchewan River Dam. The cylinders are embedded in soil with one surface exposed and are removed and rated visually at 3- to 5-year intervals.

Karim Nasser of the Civil Engineering Department of the University of Saskatchewan at Saskatoon, Saskatchewan, Canada, is conducting a series of creep tests on concrete containing lignite fly ash.

Successful utilization of lignite and subbituminous ash will require the cooperation of many organizations throughout Canada and the United States for many years to come.

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[90] Bonifay, D. W., W. W. Scott, J. A. Epps, and B. M. Gallaway. Rotary Kiln-Fired Synthetic Aggregates Manufactured From Texas Lignite Fly Ash. Highway Research Board: Highway Research Record 353, 1971, pp. 25-30.

LOW-BTU FUEL GAS FOR POWER GENERATION

by

P. S. Lewis,[91] R. J. Belt,[92] and A. J. Liberatore[93]

Introduction

Gasification and gas cleanup must be considered jointly in view of today's clean environment regulations because their combined action is required if clean gas is to be obtained from coal. Much of the coal sulfur appears in the gas, and in addition, solid and tar particulates are present in concentrations that vary with the gasification process and coal composition. All gasification concepts undergoing development include gas cleanup in the overall processing scheme. Innovations are usually introduced in the gasification step, and in some cases desulfurization may be incorporated at this point, for example, by adding half-calcined dolomite to a fluidized fuel. Purification could also be accomplished after gasification, and existing commercial systems could be used by cooling the gas and scrubbing with liquid solutions. Disposal of liquid and solid wastes is required, and provision for disposing of coal tar could be another requirement. A third possibility is to clean the gas at temperature and pressure using a regenerable solid absorbent for accepting sulfur from the gas.

Low-Btu fuel gas for power generation is receiving serious consideration by the electric utilities because it offers a timely solution to the shortage of low-sulfur fuel. High-sulfur coals can be converted into gas and cleaned of sulfur before being burned for power generation. Commercialization of this practice will make large quantities of high-sulfur coal environmentally acceptable in generation of electricity.

Probably the least complicated system for converting coal into low-Btu fuel gas is the one described in this paper, named MORGAS (Morgantown gas). It incorporates pressure gasification in a stirred bed of mine-run coal, which may have any free-swelling index from low to high, including lignite. Hydrogen sulfide is removed by contacting the hot gas with a bed of solid sorbent containing iron oxide, and elemental sulfur is recovered during regeneration of the sorbent.

Gasification in a fixed bed has been widely used commercially for over 100 years and is still used today on a minor scale. Historically, the fixed-bed gas producer has required a feed of noncaking, lump-sized coal or coke. Recently, the Bureau of Mines at Morgantown, W. Va., has gasified many coals

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[91] Research Supervisor.

[92] Chemical Research Engineer.

[93] Mechanical engineer.

All authors are with the Morgantown Energy Research Center, Bureau of Mines, U.S. Department of the Interior, Morgantown, W. Va.

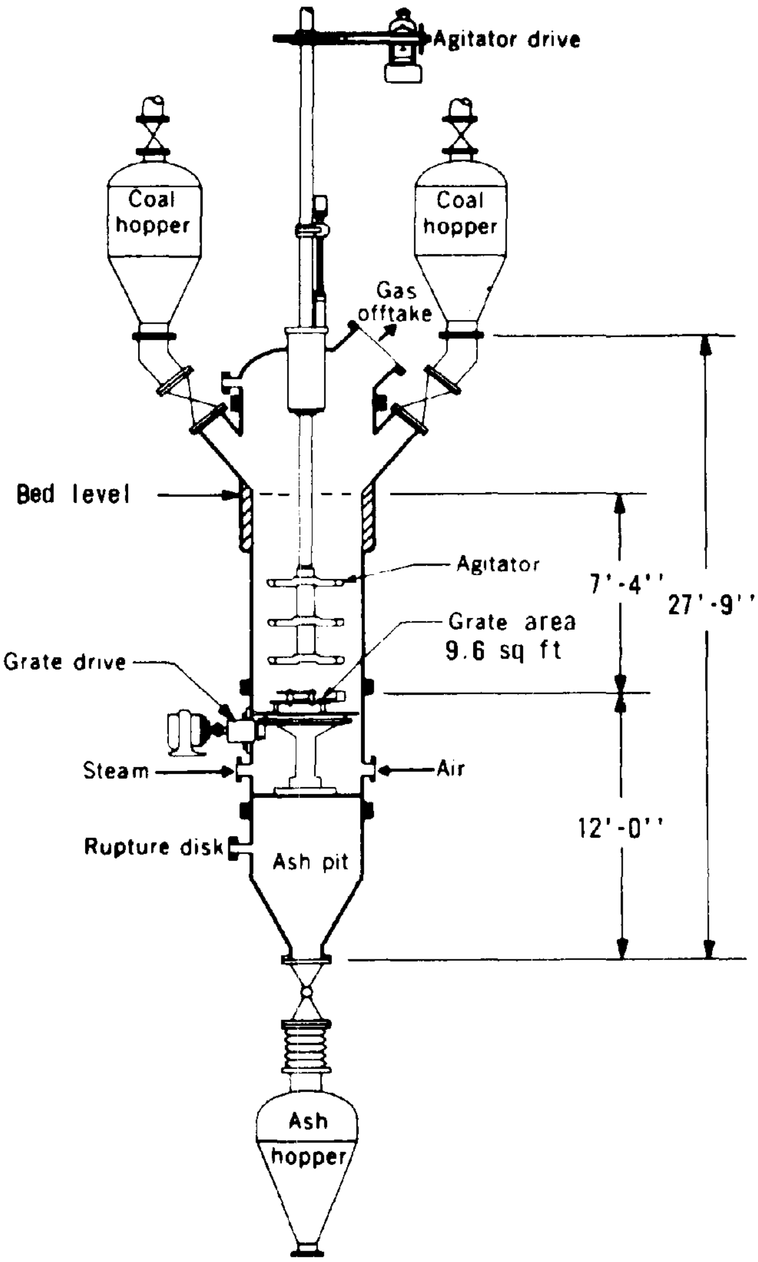

FIGURE 121. - Schematic drawing of MORGAS producer.

including strongly caking Pittsburgh seam,[94] high-volatile A bituminous (free-swelling index 8-1/2), and coal 50 pct smaller than 1/4-inch screen size (run-of-mine).[95] These pioneering developments have demonstrated that deep, continuous stirring or agitation of the fuel bed is essential and beneficial to the gasification process. The bed stirring promotes gasification reactions and gas quality by breaking massive coke formations and maintaining density of the fuel bed. The stirring employed in this work is much more extensive than ever used previously, and it warrants being distinguished by being called the stirred fixed bed, or simply the stirred bed to distinguish it from the conventional fixed bed. The fixed bed provides for only shallow stirring of the topmost layer of fuel where plastic flow and agglomeration are not prevelant.

Equipment

The Bureau's stirred-bed producer resembles the conventional fixed-bed producer except for the use of mechanical deep-bed agitation. Principal dimensions and layout are shown in figure 121. The fuel is supported on a revolving grate with area of 9.6 sq ft. The bed depth varies between 6 and 7 feet; the depth is maintained by

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[94] Lewis, P. S., A. J. Liberatore, and J. P. McGee. Strongly Caking Coal Gasified in a Stirred-Bed Producer. BuMines RI 7644, 1972, 11 pp.

[95] Shultz, F. G., and P. S. Lewis. Hot Sulfur Removal From Producer Gas. Proc., 3d Internat. Conf. of Fluidized-Bed Combustion, Hueston Woods State Park, Ohio, Oct. 29-Nov. 1, 1972, 6 pp.

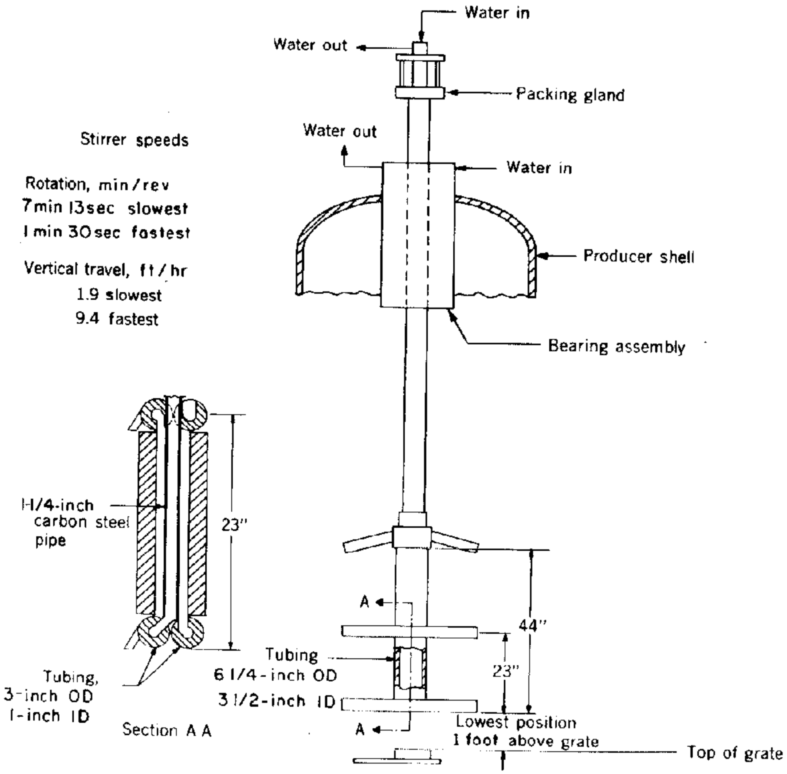

FIGURE 122. - Stirrer arrangement for MORGAS unit.

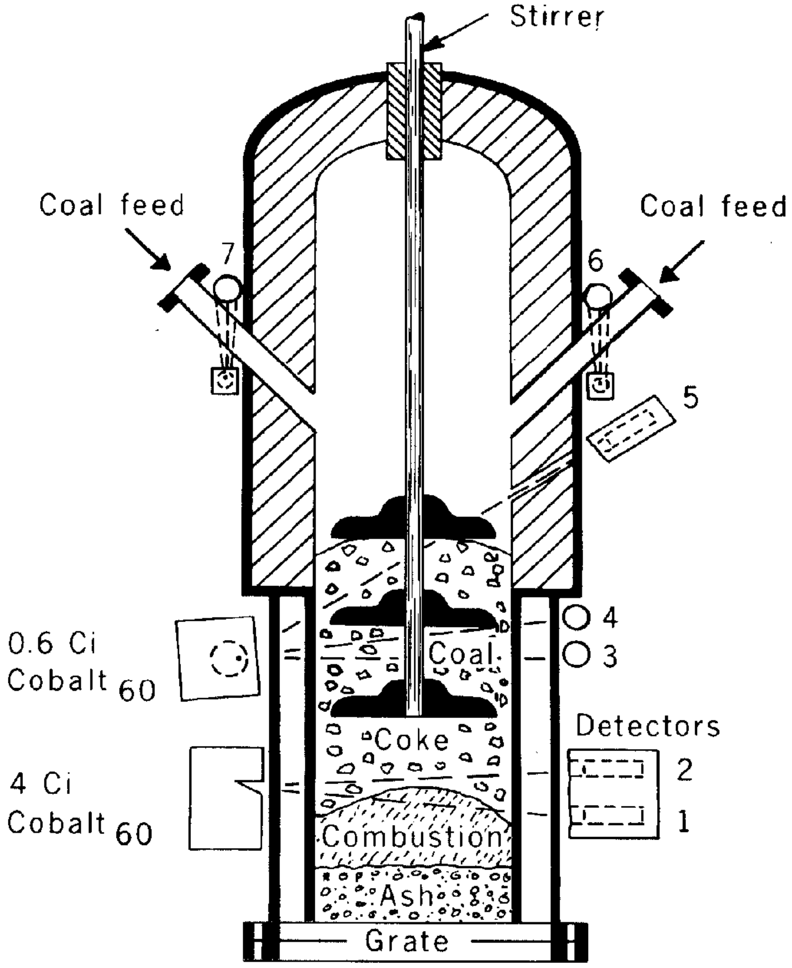

FIGURE 123. - Nuclear density gages applied to gas producer.

frequently adding coal in batches of 200 to 250 pounds, and cinder is removed as needed. The water-cooled stirrer is balanced by a counterweight and supported in the pressure vessel by a thrust bushing sealed by a packing gland. Compound motion is imparted by combined horizontal rotation and vertical reciprocation, which can be controlled with respect to speed of rotation and vertical movement. Figure 122 shows the stirrer in greater detail. The two lower arms are water cooled, but the top arm is not as it normally remains in a low-temperature zone. Steady gasification conditions usually have been obtained by rotating the stirrer at 1/2 rpm and limiting the vertical travel to 2 feet. In practice, the stirrer passes through the bed in 15 min, but this rate can be slower or faster, as the optimum travel rate varies with coal properties. The lowest point reached by the stirrer is usually set 2 feet above the top of the grate, but the limit of travel is within 1 foot of the grate.

Nuclear density gages are used as shown in figure 123 to indicate conditions within the pressure vessel. Ash zone, bed level, and voids in the bed are detected. Control of operating conditions are simplified by the use of these instruments, and centralized, fully automated controls seem to be feasible for multiple units.

Continuous stirring of the bed maintained a dense fuel bed, giving gas having good quality and constant composition. Vertical movement was found to be equally important as rotation for operating the experimental producer, but vertical movement may not be necessary for full-size units. Stirring was needed to break large clinkers that formed in the combustion zone as well as coke in the gasification zone. As shown in figure 124, the torque applied to rotate the stirrer varies directly with the bed depth covering the stirrer.

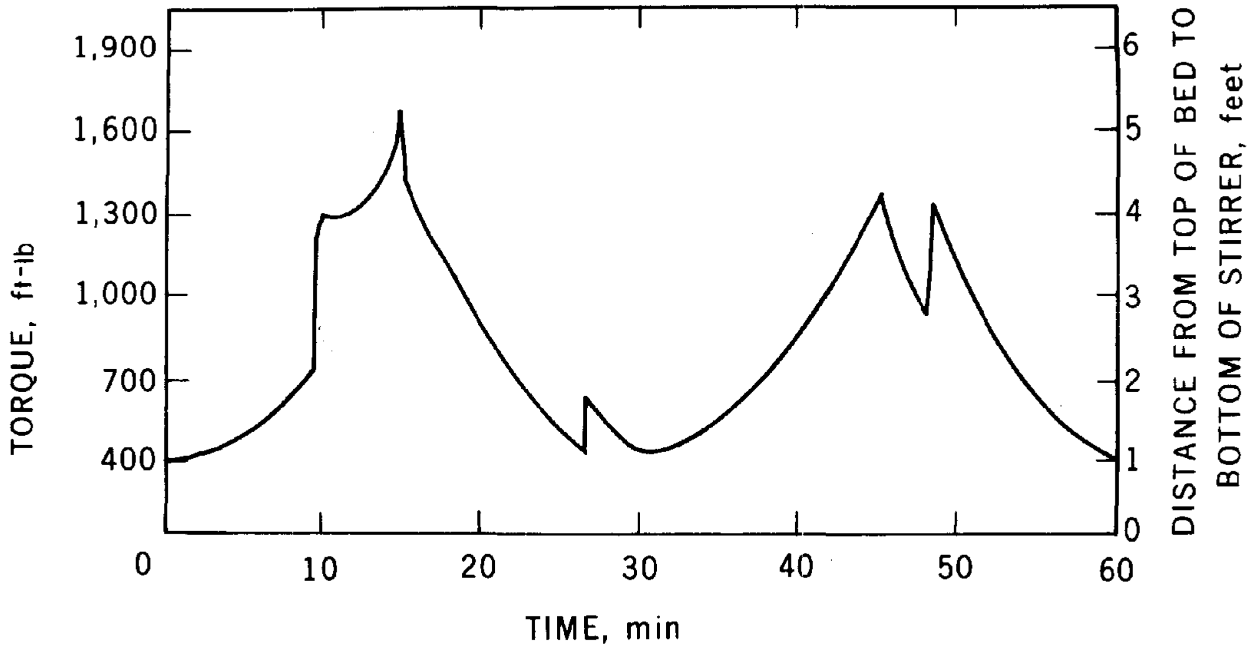

FIGURE 124. - Torque applied depending on depth of stirrer.

At maximum depth, normal torque was 1,300 ft-lb but reached momentary peaks of 1,700 ft-lb. Torque decreased during withdrawal to a minimum of 400 ft-lb. These data were obtained using Upper Freeport coal, which gives a very hard coke. No significant difference in the torque load was found for double-screened 1/4- by 1-1/2-inch Upper Freeport and run-of-mine 0 by 1-1/2-inch. Gasifying run-of-mine coal in a stirred bed was a significant advancement because the limitation of particle size, heretofore believed necessary, can be eliminated. More of the market supply will be available for gasification, and preparation will be less costly. A screen analysis of run-of-mine Upper Freeport coal is given in table 32. Twenty-five pct of the sample passed through a 1/16-inch sieve and 5 pct through 100 mesh. Some fine particles were entrained in the gas, but most was removed in a cyclone separator. Gas vented to the atmosphere and burned had a dust loading of about 0.1 to 0.5 lb/Mcf.

TABLE 32. - Screen analysis, Upper Freeport coal[1]

Screen size	Analysis, pct	
	Direct	Cumulative
2-1/2 by 2 inch	2.5	2.5
2 by 1 inch	12.1	14.6
1 by 1/2 inch	12.2	26.8
1/2 by 1/4 inch	17.6	44.4
1/4 by 1/16 inch	30.2	74.6
1/16 inch by 50 mesh	16.9	91.5
50 by 100 mesh	3.5	95.0
100 by 200 mesh	2.1	97.1
200 mesh by 0 inch	2.9	100.0

[1] Free-swelling index No. 8-1/2.

Results

Gasification

Lignite from the Beulah field, Mercer County, N. Dak., was gasified at 40 and 80 psi and an hourly rate of 1,000 pounds using 3,230 pounds of air and 535 pounds of steam. Gas yield amounted to 57,800 scfh and the moisture-free gas had a heating value of 115 Btu/scf. Optimum yields were not determined because the supply of lignite was limited and inexperience caused some handling losses from decrepitation. Gas production per square foot of grate area was about two-thirds of the amount obtained for a high-volatile coal as a result of lignite's reduced content of fixed carbon and volatiles. Low fixed carbon and volatile contents also contribute to the reduced heating value compared with gas from coals of higher classification. This test demonstrated that lignite could be gasified in the stirred-bed gas producer and is in addition to the slagging gasifier described in an earlier Bureau Report of Investigations.[97]

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[97] Gronhovd, G. H., A. E. Harak, M. M. Fegley, and D. E. Severson. Slagging Fixed-Bed Gasification of North Dakota Lignite at Pressures to 400 Psig. BuMines RI 7408, 1970, 40 pp.

Gas yields and composition are compared in figure 125 for two high-volatile coals, western Kentucky (hvbb) No. 9 seam and Upper Freeport seam (hvab) from northern West Virginia. For both coals, drastic changes in gasification rate and system pressure had virtually no effect on gas composition.

Gas Cleanup

A mixture of iron oxide (hematite, Fe_2O_3) and fly ash was the best sorbent found among more than 20 materials tested. Primary requirements were that the sorbent be readily available and relatively inexpensive, have reasonable sorption capacity and useful life, be easily regenerated for repeated use, and resist fusion or disintegration over the useful temperature range. Fly ash as received could be formed into a durable and regenerable sorbent, but its sorption capacity was improved by adding iron oxide, increasing the concentration to 36 pct from 15 pct originally present. Other oxides present and inactive included silica 35 pct, alumina 18 pct, and small percentages of oxides of calcium, magnesium, sodium, potassium, and titanium. Iron oxide concentrations greater than 40 pct were unsatisfactory because the bed fusion temperature was lowered and fusion took place during normal operations.

Pilot quantities of the fly ash-iron oxide sorbent were made by two catalyst manufacturers by mulling and extruding the mixture to form 1/4-inch-diameter cylinders with 1/4- to 1/2-inch lengths, which were then sintered to develop hardness. Mercury porosimeter measurements showed pore volume of new

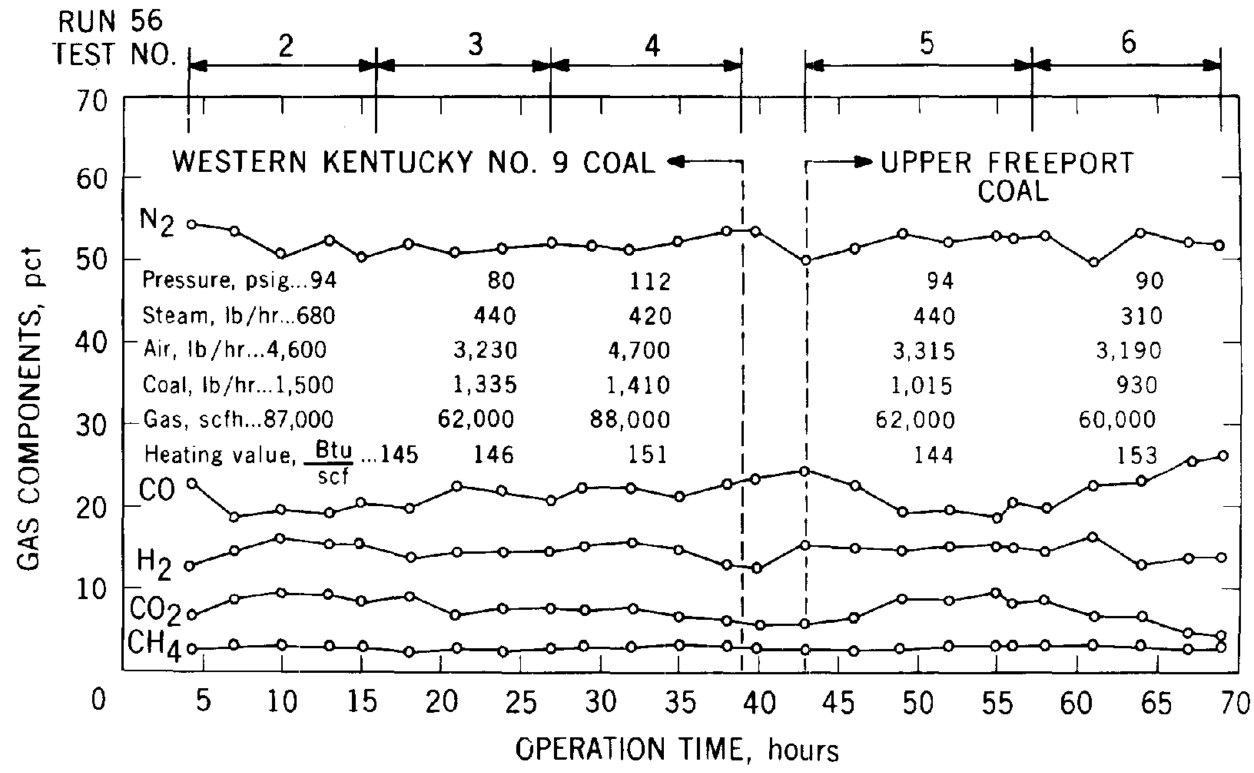

FIGURE 125. - Product gas composition during producer operation.

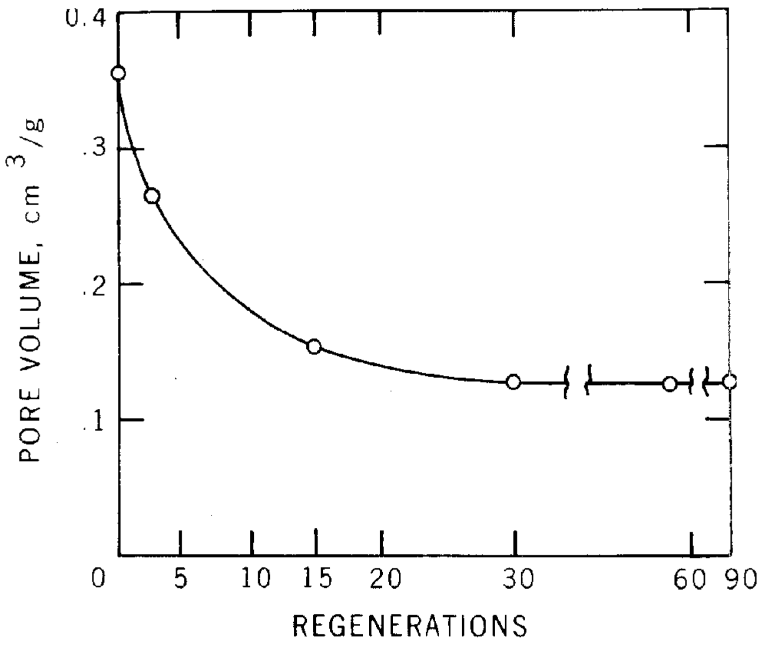

FIGURE 126. - Pore volume reaches constant value after 30 regenerations.

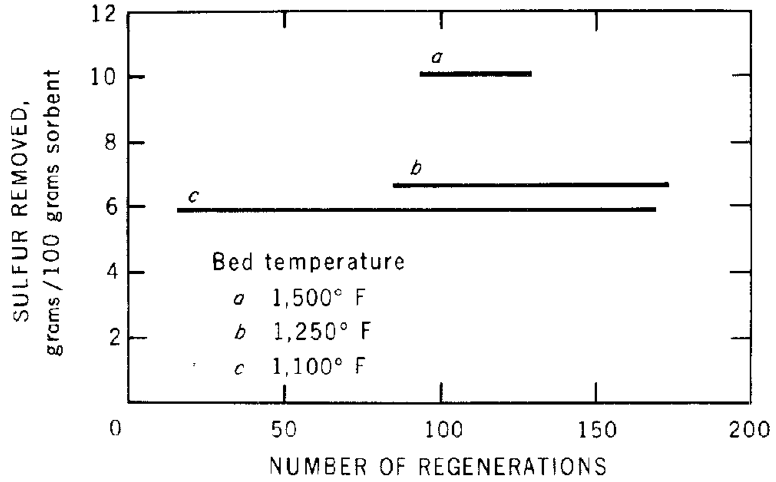

FIGURE 127. - Sulfur sorption increases with bed temperature.

sorbent was 0.36 cm^3/g, but this decreased to 0.13 cm^3/g and remained constant after 30 regenerations, as shown in figure 126. Surface area measured by nitrogen absorption ranged from 4.2 to 6.5 m^2/g. Sorption of hydrogen sulfide from dry simulated producer gas is given in table 33 for materials of essentially the same composition but made by different laboratories.

The sorbent made by Morgantown Energy Research Center was tested through 175 regeneration cycles using simulated producer gas and bed temperatures of 1,100°, 1,250°, and 1,500° F. Producer gas contains about 5 to 10 pct steam by volume, as excess steam is used to reduce temperature in the combustion zone, and the gas leaves the generator at a temperature around 1,200° F. Steam amounting to 7 vol-pct was added to the gas for many of the above tests to closely simulate producer gas. Results obtained with gas containing steam (fig. 127) indicate a reduction in capacity when compared with capacities for dry gas as shown in table 33. This was attributed to the lowering of the hydrogen sulfide concentration at the gas-solid interface by the added steam. Improving the mass transfer coefficient by raising the bed temperature was effective in increasing the capacity from 6 grams of sulfur per 100 grams of sorbent at 1,100° F to 10 grams at 1,500° F.

Iron oxide catalyzes the water-gas shift reaction, $H_2O + CO = H_2 + CO_2$, and steam in producer gas affected the composition of the producer gas in passing through the sorption bed. The composition change resulting from the shift reaction was determined at 300 psig and temperatures of 1,100°, 1,300°, and 1,400° F by passing producer gas containing 18 mole-pct steam through a bed of iron oxide-fly ash sorbent at a space velocity of 1,000 hr^{-1}. Heating value was decreased by dilution from the carbon dioxide added to the gas; increased hydrogen and decreased carbon monoxide concentrations resulted in virtually no net change in heating value because these gases have nearly the same value of 319 and 316 Btu/scf, respectively. The shift would be beneficial if pipeline gas is the end use because additional shifting would be needed to bring the hydrogen:carbon monoxide ratio close to 3:1. Increasing temperature favors higher carbon monoxide concentration at equilibrium. Results are shown in table 34.

TABLE 33. - Sorption of H_2S from dry producer gas by sintered iron oxide-fly ash[1]

	Surface area,[2] m/gram	Bed temperature, ° F	Sulfur removed per 100 grams sorbent, grams	
			From sorption data	From regeneration data
Commercial lab. No. 1............	6.5	1,000	12.5	12.4
		1,250	14.7	13.9
		1,500	22.2	22.0
Commercial lab. No. 2............	4.2	1,000	7.5	6.7
		1,250	11.5	11.0
		1,500	22.5	17.4
Mechanically formed by MERC[3].....	4.2	1,000	10.5	10.9
		1,500	27.6	25.6

[1] All at 3 psig sorption pressure.
[2] B.E.T. nitrogen sorption method.
[3] Morgantown Energy Research Center, Morgantown, W. Va.

TABLE 34. - Change in gas composition at 300 psig and 1,000 hr^{-1} space velocity in absorbent bed

	Hydrogen, pct	Carbon dioxide, pct	Nitrogen, pct	Carbon monoxide, pct	Heating value, Btu/scf
Feed gas..................	17.0	6.7	50.9	25.4	134
Effluent gas:					
1,100° F (dry)..........	16.4	7.9	51.0	24.7	130
1,100° F (wet)[1].........	22.9	13.2	51.1	12.8	113
1,300° F (wet)[1].........	21.3	11.0	51.0	16.7	121
1,400° F (wet)[1].........	19.9	9.7	51.0	19.4	125

[1] Steam content 18 vol-pct. Composition and heating value on dry basis.

Two sorption-regeneration cycles were completed, cleaning gas generated in the pressurized gas producer using Upper Freeport coal; the results are

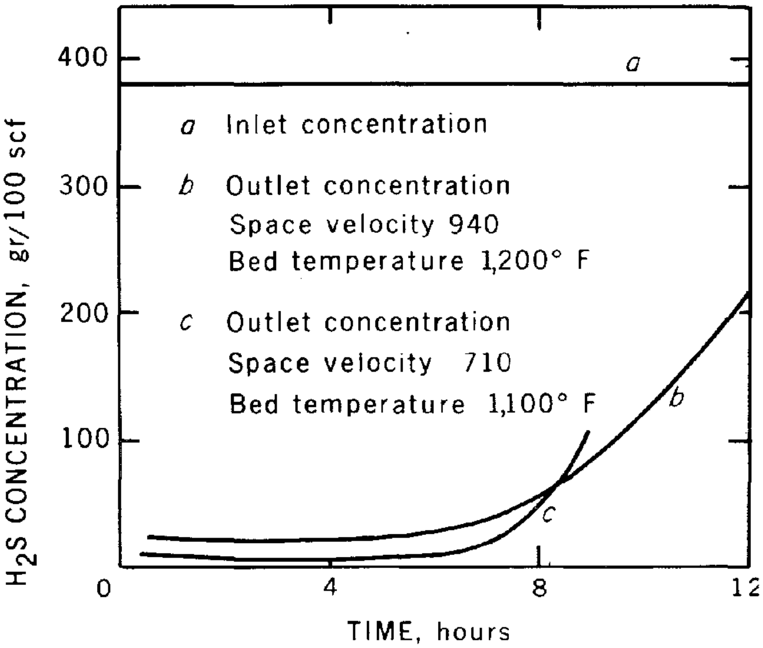

FIGURE 128. - Removing H_2S from producer gas.

shown in figure 128. Gas from the producer was transferred to the sorbent bed at system pressure of 120 psig via a heated pipeline. Bed temperatures were controlled to give 1,100° and 1,200° F, and flow rates were adjusted to give space velocities of 710 and 940 hr^{-1}, respectively. Hydrogen sulfide concentration averaged 380 gr/100 scf, and the gas contained approximately 1/2 pound of dust, 1 pound of tar, and 5 pounds of steam per 1,000 scf. Hydrogen sulfide in the gas leaving the sorbent bed had its concentration reduced to 10 and 20 gr/100 scf and did not increase until after 6 hours on steam. Removal was 95 pct effective with respect to hydrogen sulfide. Tar was not removed by the sorbent.

Reaction mechanism is chemisorption, whereby hydrogen sulfide diffuses throughout the sorbent and reacts with Fe_2O_3 forming FeS and FeS_2. Analyzing the spent sorbent indicated the empirical composition was $FeS_{1.3}$. Iron oxide, Fe_2O_3, was regenerated and the sulfur released as SO_2 by passing air or oxygen over the hot bed. With oxygen regeneration, the effluent gas was pure SO_2 until some oxygen passed through unreacted after regeneration was 90 pct complete. Rather than recovering the SO_2 as sulfuric acid or ammonium sulfate, commercial processes are available to reduce SO_2 to elemental sulfur.

Conclusion

Sulfur-free low-Btu gas can be obtained from coal by gasifying at low pressure using steam and air and removing sulfur at system pressure and temperature. This concept is of particular importance for power generation because operating costs and capital investment are probably the lowest for clean-burning substitute fuel.

CO_2-ACCEPTOR PROCESS: STATUS OF DEVELOPMENT

by

Carl E. Fink[97] and Marion H. Vardaman[98]

Introduction

The CO_2-Acceptor Coal Gasification Process is under development by Consolidation Coal Co. through a contract with the U.S. Office of Coal Research and the American Gas Association. Stearns-Rogers Corp. built and is currently operating a pilot plant at Rapid City, S. Dak. The process performs gasification of lignite coal with steam at 150 psig (11 atm) and about 1,500° F to produce a synthesis gas. The synthesis gas produced at these conditions is expected to have the appropriate ratio of hydrogen and carbon oxides to be the feedstock to a methanation unit without requiring a "Shift Converter." Upon purification (removing trace sulfur compounds) and methanation, one would have a high-Btu pipeline gas of about 900 to 950 Btu/scf.

The unique feature of this process is the use of dolomite (a stone that is a combination of magnesium and calcium carbonates) or limestone (which is nearly all calcium carbonate) to supply heat for the endothermic steam-carbon gasification reaction. Once calcined to the $MgO \cdot CaO$ form and added at high temperature, the dolomite will carbonate exothermally to $MgO \cdot CaCO_3$ in the gasifier to provide chemical as well as sensible heat. Its "acceptance" of CO_2 at the same time serves to reduce the CO_2 level in the gasifier to minimize CO_2 removal in the purification section.

The process is keyed to providing future pipeline-gas requirements for the Midwest, using lignite or subbituminous coal as the starting material. The subject pilot plant has been in operation about a year and will provide data for the design of a commercial plant with a capacity of 250 MMscfd pipeline gas.

Process Description

The process utilizes two fluidized bed reactors with flow of both lignite char and acceptor (either dolomite or limestone) between the gasifier and acceptor regenerator. Ground, dried, and preheated lignite coal from the coal storage and grinding area is fed into the gasifier via lockhoppers.

In the gasifier, the fresh lignite enters into a fluid bed of lignite char. Both devolatilization and gasification will be accomplished in a single reactor. Figure 129 shows that the gasifier consists of a main vessel section and a smaller "boot" section. The reason for this configuration is that

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acceptor is "showered" down through the fluidized bed of lignite char and is collected in the boot. The higher gas velocity in the boot section is sufficient to fluidize the acceptor and strip out any entrained char. The boot thus provides a reservoir for acceptor withdrawal.

The gasifier is operated at 1,500° F and 150 psig. At these conditions, a number of reactions occur as shown in table 35. The principal reactions of gasification are the highly endothermic steam-carbon reaction, $C + H_2O = CO + H_2$, and the water gas shift reaction, $CO + H_2O = CO_2 + H_2$. The heat duty required to maintain the desired temperature is supplied by sensible heat of the acceptor, which flows by gravity from the elevated regenerator to the gasifier, and by the exothermic carbonation of the acceptor as it "accepts" CO_2.

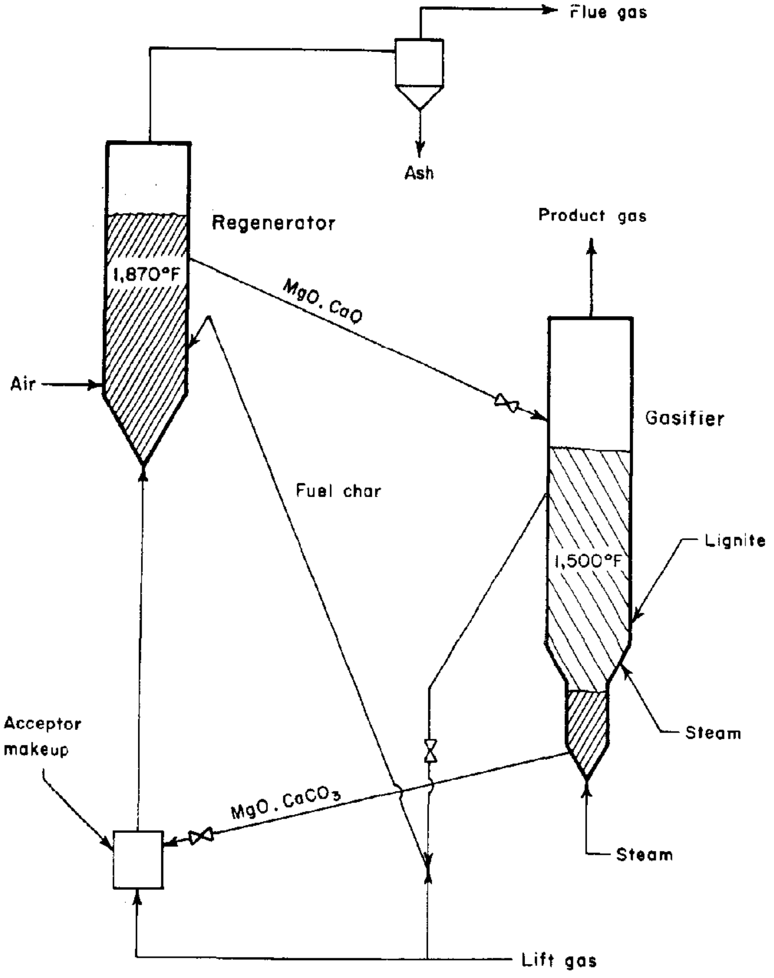

FIGURE 129. - Diagram for CO_2-Acceptor Process.

TABLE 35. - Chemical reactions for CO_2-Acceptor Process

Chemical reaction		Type of reaction
Gasifier:		
(1) $C + H_2O$	$= CO + H_2$	Endothermic.
(2) $2C + H_2O + H_2$	$= CH_4 + CO$	Do.
(3) $CO + H_2O$	$= CO_2 + H_2$	Exothermic.
(4) $CH_4 + H_2O$	$= CO + 3H_2$	Endothermic.
(5) $CaO + CO_2$	$= CaCO_3$	Exothermic.
Regenerator:		
(6) $C + O_2$	$= CO_2$	Do.
(7) $C + CO_2$	$= 2CO$	Endothermic.
(8) $CaCO_3$	$= CaO + CO_2$	Do.

Fresh lignite enters the gasifier from one of two lockhoppers. The point of entry is low in the vessel to obtain maximum vapor retention time for the volatile material from the coal. All of the volatiles are cracked to CH_4, H_2, and oxides of carbon in the gasifier bed. A char bed of about 25 feet is maintained above the transition section of the vessel. During operation, calcined acceptor enters the gasifier vapor space and "showers" through the char bed where it reacts with carbon dioxide. Recarbonated acceptor then collects in the "boot" section from where it is transferred, char free, to the engager pot. Steam, which enters the bottom of the boot as well as at the transition section above the boot, is used both as fluidization gas and as a reactant in gasification. After purification, the gas is a suitable feedstock for methanation. The product of methanation would be pipeline-quality gas.

The recarbonated acceptor is "picked up" in the engager pot by recycle flue gas or "lift gas" and pneumatically conveyed 90 feet into the regenerator. Here a portion of the lignite char from the gasifier is burned with air to produce the heat necessary for recalcination of the acceptor. A fluid bed of acceptor is maintained in the regenerator by a gas velocity high enough to elutriate the very light char ash particles as well as any acceptor formed by attrition. The flue gas leaving the regenerator flows through a cyclone to remove the entrained particulate matter. Further cleaning of this gas is accomplished in a water scrubber. The ash is collected in one of two ash lockhoppers, then slurried to a large settling pond. The acceptor inventory is maintained by continual addition of makeup to the engager pot.

Figure 129 indicates use of recycle flue gas as the lift gas for the acceptor. However, a commercial plant would use air. The pilot plant has the facility to substitute air for flue gas to lift the acceptor.

Pilot Plant Operations

The pilot plant at Rapid City has been in operation for a year. Thus far, eight runs have been made with the objective of demonstrating the process using char as the gasifier feedstock. Although none of these runs has been successful from the standpoint of achieving a sustained gasification run, each has supplied information that has made the succeeding run more successful.

In the first year, much time and effort was spent in modifying the physical aspects of the plant to make it operative at the unusual conditions required for gasification. The refractory designs in both the gasifier and regenerator were inadequate from the standpoint of physical strength. Both refractory systems failed and replacement required over 3 months downtime. In addition, major design changes were made in the regenerator quench system, the gasifier internal cyclone, the regenerator cyclone, the ash lockhoppers, and the hot line design. Also, the startup procedure has been modified to minimize corrosion of the fired heaters.

Since these problems are basically not process oriented, they will not be discussed in detail here. Their detection and solution do, however, represent a large portion of the accomplishments during the first year of operation.

The startup involves five major steps, each of which has its own potential difficulties. These problems can be overcome by the proper control of one or more key variables. Table 36 lists the startup steps, the potential difficulties, and the key process variables associated with each step.

TABLE 36. - Outline of startup sequence

Startup step	Problems experienced	Key process variables
(1) Acceptor addition and circulation.	Pressure balance, loss of seals.	Differential pressure between gasifier and regenerator.
(2) Char addition to gasifier.	Loss of char acceptor interface.	Gasifier boot fluidizing velocity. Gas distribution. Acceptor size consist.
(3) Acceptor circulation with char acceptor interface.	Excessive acceptor holdup in gasifier char bed.	Char bed density. Fluidization velocity. Gas distribution.
(4) Char combustion to calcine acceptor in regenerator.	Deposit formation........	Control of regenerator gas composition. Fluidizing velocity. Gas distribution.
(5) Gasification supplying heat of reaction via acceptor.	Acceptor transfer line between regenerator and gasifier plugged.	Fines content of calcined acceptor. Line purges. Acceptor circulation rate.

The first step involves adding the acceptor inventories in the regenerator and gasifier boot and then circulating the acceptor. The system pressure balance must be maintained to provide solids seals between the gasifier and the regenerator vessels. Each standleg of solids is associated with a maximum allowable pressure drop. Should the system pressure balance require a pressure drop in excess of this maximum, the seal of solids would be lost.

Figure 130 illustrates the principle of the solids seal. A pneumatically operated butterfly valve controls the rate of solids transfer through the line. To facilitate transfer, the differential across the valve should be maintained near zero. This is accomplished with a purge gas that is controlled by the differential pressure. The maximum pressure drop in the standleg is proportional to the weight of solids and occurs when the purge gas flow corresponds to the incipient fluidization velocity.

As the regenerator bed is filled, the pressure drop across the lower standleg will increase. By controlling the regenerator pressure at a differential lower than the gasifier, the pressure drops across the two standlegs can be balanced and circulation maintained.

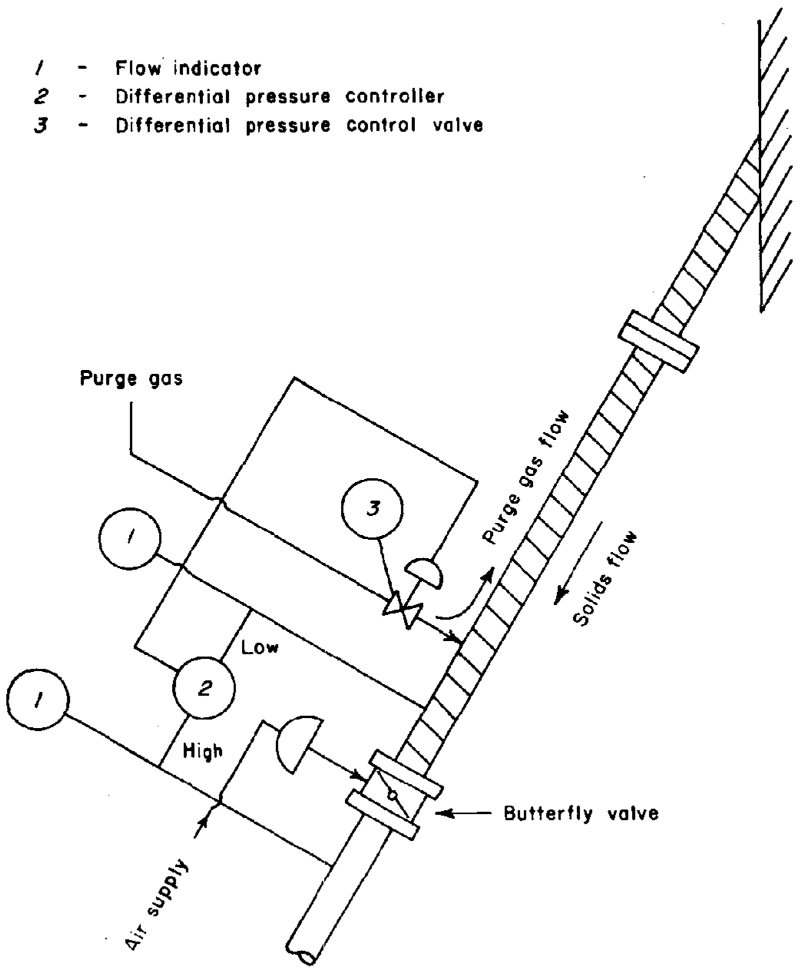

FIGURE 130. - Diagram of solids transfer line.

Three runs were made before acceptor circulation was reliable. Improved pressure measurement techniques and modification of the purge gas system were required in order to monitor the system pressure balance and successfully circulate acceptor.

The second step is the addition of char to the gasifier. The major problem in this step is maintaining a separation or interface between the char and acceptor in the boot. Char was added unsuccessfully in the third, fourth, and fifth runs before an interface was maintained in the sixth run.

Two obvious causes for the loss of the interface were the failure of the gasifier refractory (run 4) and the presence of excessive fines in the acceptor (run 5). However, the final solution to the problem was obtained by studying the interface problem in both a glass model and in the gasifier itself.

A stable interface requires operation of the boot at or slightly above the incipient fluidizing velocity of the acceptor. Techniques were developed to experimentally determine the incipient velocity. In addition, a distributor for the boot gas flow was developed. This distributor consists of a vertical closed end pipe with horizontal holes. During the gasifier tests, the interface was maintained after char addition and acceptor was showered batchwise through the bed. These tests were used to devise instrumentation to detect the presence and locate the level of the interface.

In each of the runs made subsequent to these gasifier tests, the acceptor interface has been successfully maintained while feeding the char bed to the gasifier.

Circulation of the acceptor with a char bed present is the third process step. The major difficulty in this step is avoiding an excessive buildup of acceptor in the char bed. The only way of reducing the acceptor holdup is by reducing the char bed density. This is done in two ways: (1) Increasing the fluidizing velocity, and (2) gasifying the char bed to reduce the density of the char particles.

Since the interface stability requires that the boot flow be near the incipient velocity of the acceptor, the char bed fluidizing velocity must be increased by adding secondary gas above the interface level. The original plant design provided for the addition of secondary gas, but installation of a distributor was necessary to make the flow effective. Another difficulty is that there is no adequate means of directly measuring the expanded char bed density. Experiments made both in the plant and in the glass model show that the differential pressure measurements are insensitive to fluidized bed density changes. The differential pressure measurements are, however, sensitive to changes in the density of the material in the bed.

The particle density of the calcined acceptor is only slightly greater than that of the ungasified char. Therefore, to enable calcined acceptor to shower, the gasifier bed is pregasified by adding air with steam, as the secondary fluidizing gas. Combustion of the char carbon supplies the heat requirement for the carbon-steam reaction. Pregasification was practiced in the seventh and eighth runs and greatly reduced the holdup of acceptor in the bed.

The fourth startup step is char combustion in the regenerator to supply heat for calcination of the regenerator bed. Char combustion has been carried out in the fifth, seventh, and eighth runs.

Significantly, there has been no evidence of ash fusion in the regenerator. However, in the two earlier runs, deposits were formed apparently by a mechanism involving a low-melting CaS-$CaSO_4$ mixture. The reactions involved are as follows:

$$CaS + 2\ O_2 = CaSO_4. \qquad (9)$$

$$CaS + 4\ CO_2 = CaSO_4 + 4\ CO. \qquad (10)$$

$$CaS + CaSO_4 = \text{Liquid (at temperatures} >1{,}700^\circ\ \text{F)}. \qquad (11)$$

$$1/4\ CaS + 3/4\ CaSO_4 = CaO + SO_2. \qquad (12)$$

CaS is oxidized to $CaSO_4$ by either oxygen from the air (equation 9), or CO_2 from the recycle gas (equation 10). The primary source of CaS entering the regenerator is the char ash. At temperatures above 1,700° F, the mixture melts (equation 11), trapping char ash and acceptor fines. As CaO is formed (equation 12), the liquid resolidifies leaving a deposit on the reactor wall. The bench-scale work had shown that the presence of CO in the recycle gas suppresses the oxidation to $CaSO_4$. Originally, it was nearly impossible to maintain the desired CO concentration in the recycle gas because irregularities in pressure balance caused erratic fuel char feed.

Prior to the eighth run, instrumentation was installed that operated the char feed valve to control a set pressure drop across the fuel char lift line. This control was able to adjust for normal changes in the system pressure balance and maintain a constant fuel char rate.

The fluidization characteristics of the regenerator bed also affect deposit formation. Bed expansions in excess of 50 pct are required. The gas distribution at the bottom of the bed where the combustion is carried out is also important. To improve fluidization, a ring-type gas distributor was added to the air inlet prior to the eighth run.

During the eighth run, char was burned in the regenerator for about 18 hours. The CO concentration in the recycle gas was kept near the desired level of 4 pct at all times. Examination of the regenerator bed following shutdown showed that no deposit had formed.

The final startup step is continuing acceptor circulation with calcined acceptor flowing from the regenerator to the gasifier where it will supply the heat requirements for gasification. During the eighth run, the acceptor return line from the regenerator plugged soon after calcination was complete. Since no agglomerated material was found in the line, the apparent cause for the plug was an excessive amount of fines in the calcined acceptor. These fines tend to be fluidized by the standleg purges. During the run the transfer of acceptor from the regenerator to the gasifier was momentarily discontinued. This allowed the fines in the standleg to segregate. An accumulation of fine material at any point in the line will prevent the coarser material from moving down the leg.

The calcined acceptor is much weaker than its $CaCO_3$ counterpart, particularly after the first calcination. High attrition rates are expected during first few calcination-recarbonation cycles. On repeated cycling, the acceptor becomes hard and the attrition is expected to drop to essentially zero.

Several modifications are planned to minimize the plugging problem in the next gasification run. Thermocouples will be added to detect flow stoppages in the line. If the acceptor flow is maintained, there will be less chance for fines to segregate. The purge flows will be decreased to a minimum, and the pressure drop across the line will be controlled at zero. The instrument controlling ΔP between the gasifier and regenerator will be made more sensitive. Finally, the regenerator fluidizing velocity will be raised to maximize fines elutriation from the bed.

Solution of the plugging problem in the acceptor return line is expected to lead to a successful demonstration run of sufficient duration to evaluate the acceptor performance. The next step in the program would then involve the use of lignite as feedstock. Eventually, two lignites and at least one subbituminous coal will be evaluated. The acceptor used for the demonstration runs has been Tymochtee dolomite from Ohio. Future runs will be made to evaluate dolomites and limestones available in Montana, Wyoming, North Dakota, and South Dakota.

LABORATORY STUDIES ON SOLUTION—HYDROGENATION OF LIGNITE

by

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and Wayne R. Kube[101]

Introduction

The United States is presently faced with an energy crisis for both short and long term. In the case of electric power, the Nation has experienced brownouts and blackouts and reduction of voltage at time of peak demand. Natural gas is in short supply as indicated by restrictions in sales and limitation on adding certain classes of customers. Shortages in fuel oil supplies developed during the past winter and there are indications that gasoline could be in short supply during the summer. All factors seem to indicate that the short-term situation will worsen.

The future energy situation is even less favorable. Many projections of future requirements have been reported[102] although not all estimates use the same basis. Usually, the estimates are made using historical data with certain assumptions concerning possible national gross production and per capita consumption of energy with an exponential increase assumed to A.D. 2000. Conservatively, an average 3.5 pct compounded annual growth rate from A.D. 1970 to 2000 would occur. At this average growth rate, energy consumption would increase threefold. A threefold increase in energy consumption in 30 years presents problems in processing and distribution even if reserves were adequate. Difficulty will be experienced in finding the energy to meet the projected demand. All sources must be investigated, including coal, gas and oil, nuclear fission and fusion, oil shale, tar sands, and solar, geothermal, and water power. All practical sources will have to be utilized as part of a total energy concept without extensive competition between sources. Thus, a comprehensive national energy program must be developed.

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[101] Professor of Chemical Engineering.

All authors are with the University of North Dakota, Grand Forks, N. Dak.

[102] Winger, J. C., J. D. Emerson, G. D. Gunning, R. C. Sparling, and A. J. Zraly. Outlook for Energy in the United States to 1985. Energy Economics Division, The Chase Manhattan Bank, New York, June 1972, 55 pp.

McLean, John G., Chairman. U.S. Energy Outlook: An Initial Appraisal 1971-1985. V. 1, 75 pp., report by the National Petroleum Council's Committee on U.S. Energy Outlook, Washington, D.C., July 15, 1972, 38 pp.

U.S. Department of the Interior. United States Energy, A Summary Review. Washington, D. C., January 1972, 42 pp.

The short-term situation may be alleviated more readily than the long-term situation because the present crisis is not caused by shortages in the total supply of energy but by distribution problems, preference for convenience fuels, and environmental and safety policies. In some instances the demand for electric power had been underestimated and insufficient capacity planned. Shortages have been intensified by Federal and State antipollution restrictions that were not in existence when most existing powerplants were constructed. The technology for pollution control has not yet been developed into economical commercial processes. Consequently, historical supply patterns have been displaced, and local shortages have developed in low-sulfur residual oil, natural gas, and low-sulfur coals. Increased pressure has been exerted to relax import restrictions for fuels, and plans have been developed for importation of liquefied natural gas (LNG). Unit train shipments of low-sulfur western coals from the Northern Great Plains coal Province to Midwest population centers are being used to allow meeting of stack emission standards from powerplants. Gasification of liquid hydrocarbons is being developed to produce synthetic natural gas (SNG).

Since liquid feedstocks for gasification are relatively limited, gasification of coal followed by methanation seems an appropriate route for the long-range fulfillment of part of the demand for natural gas. Although no commercial process is in operation in the United States to produce synthesis gas, several prototype processes are being developed in large-scale pilot plants. Reportedly, the first coal gasification plants will use the German Lurgi process, which has been in commercial operation abroad, to produce town gas. Michigan-Wisconsin Pipeline Co. and Peoples Gas Co. of Chicago have obtained mining rights to significant reserves of lignite in North Dakota and plan on having gasification plants in operation sometime in the 1980's.

Although gasification appears to be the first technology that will be commercialized for coal conversion, liquefaction and solvent refining will be required to meet other energy requirements. Processes for producing these forms from coal have not been demonstrated on large scale, although petroleum-type liquids have been made by coal hydrogenation, and coal solvation has given an upgraded low-ash solid product. A 50-tpd pilot plant for coal solution-hydrogenation is under construction at Fort Lewis, Wash.

For many years, research at the University of North Dakota has been directed toward utilization of lignite. Some projects have been in cooperation with the U.S. Department of the Interior, Bureau of Mines, and some, with private sponsors. In April of 1972, a 5-year research contract was signed with the U.S. Department of the Interior Office of Coal Research for a comprehensive investigation and integration of lignite technology.

Project Lignite

The objective of Project Lignite is to develop the information needed for a lignite refinery to produce high-quality solid, liquid, and gaseous fuels from Northern Great Plains Province lignite. The current project is intended to provide detailed data necessary for the design of a pilot plant to demonstrate the feasibility of the refinery concept. The products obtained from a lignite refinery complex could include pipeline gas, synthetic crude oil for refinery processing, solvent-refined lignite for solid or liquid fuels applications, and low-ash coke. In addition, raw materials for chemicals manufacture might be produced from the various fractions.

The complex would be large and would consume millions of tons of lignite each year. Many social, economic, and ecological benefits could result from the establishment of processing complexes in areas of low population density where environmental control should be readily achievable.

The development of a conceptual lignite refinery implies that many subprocesses must be investigated for possible integration into the operational scheme. Five process steps initially seemed to need additional experimental work:

1. Production of solvent-refined coal (SRC) from lignite.

2. Hydrogenation of SRC to produce liquids, and distillation to characterize the liquids.

3. Carbonization to produce char, liquids, and gases.

4. Direct hydrogenation of lignite to produce liquids, gases, and solid residues.

5. Drying of lignite as a separate process step.

These steps will be taken individually if required through the sequence of bench-scale research and process development unit (PDU) evaluation. After each stage, technical and economic evaluations will be made to determine the desirability of continuation. For the projects carried through the PDU stage, the system will be operated over ranges of variables including feed rate, temperature, pressure, and lignite source to optimize the process. Some tests will be made on coals other than lignite for comparison purposes. The first subproject to advance to the process development unit stage is lignite solvent-hydrogenation. Equipment capacities, process design, and some equipment specifications have been completed for a continuous unit for processing 50 lb/hr of lignite.

Solvent-Hydrogenation of Lignite

Coal is a complex organic substance consisting essentially of carbon, hydrogen, and oxygen with some hetero structures containing nitrogen and sulfur. Since about 1860, many investigators have given attention to solvent extraction of coal. Much of the work was directed toward separating and identifying coal molecules and the material that gives a coking coal caking properties. Apparently, the coal substance is not a definite compound but is a dispersed micelle consisting of solid humic substances of generally large molecular weights.

The large-molecular-weight material can be broken down to lighter fragments by solubilization-hydrogenation processes. Depending on extent of conversion, products may be liquid, solid, or gaseous. Addition of hydrogen into the unsaturated and generally aromatic coal structure by various methods of hydrogenolysis assists in fragmenting the large molecules and stabilizing the fragments. Hydrogenation may be carried out at high hydrogen partial pressures and elevated temperatures to produce liquid products. Hydrogenation catalysts, such as cobalt or nickel or less expensive iron or tin, may be used to promote the reaction.

Many modifications of the hydrogenolysis process have been attempted. One approach is hydrogenation of the coal substance in the presence of an organic solvent that may be a hydrogen donor. The undecomposed coal and the mineral matter is separated from the product slurry. The resulting deashed product has potential application directly as a low-ash fuel for combustion processes, as a synthetic crude oil for the production of conventional liquid fuel products and chemical raw materials, or for the manufacture of industrial carbons.

Several proposals for coal liquefaction utilizing solvents are currently under investigation. Projects funded by the Office of Coal Research, in addition to Project Lignite, include two for the production of heavy liquid fuels from coal.[103] The Project Gasoline pilot plant may be reactivated to study the production of low-sulfur fuel oil. The Pittsburg and Midway Coal Co.'s SRC process will be demonstrated in a 50-tpd pilot plant under construction near Fort Lewis, Wash.

Work on solvent-hydrogenation of lignite at the University of North Dakota was initiated in 1965 under the sponsorship of the Great Northern Railway Co. (now merged into the Burlington Northern). Support was also received from the Pittsburg and Midway Coal Mining Co. until a contract was negotiated with the Office of Coal Research for Project Lignite.

Solution-Hydrogenation Reactions

Solution-hydrogenation reactions are complex and depend upon the solvent, the coal, composition or reacting atmosphere, reaction pressure, and temperature, as well as on time for reaction. Reaction is considered to be quite rapid but usually can be modified by the presence of catalysts. The macrocoal substance is fragmented both by hydrogenation and thermal decomposition. The solvent may serve as a dispersant for the molecular fragments and as a hydrogen donor. The organic solvent should have a fairly high boiling range so as to remain a liquid under processing conditions of 1,000 to 5,000 psi and temperatures from 375° to 500° C. With lignite, temperatures over 500° C increase thermal decomposition and reduce solvation. Some of the organic compounds resulting from the solution-hydrogenation of coal have properties similar to those required for solvent; for a commercial process it must be possible to recover sufficient recycle solvent from the reaction to minimize the

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[103] Office of Coal Research, U.S. Department of the Interior. Annual Report 1973. Office of Coal Research, Washington, D.C., 1973, 124 pp.

use of makeup solvent. Most solvent-hydrogenation work has concentrated on use of a relatively high boiling fraction of anthracene oil because availability, hydrogen donor properties, and relatively low price make it advantageous.

The various coal macerals respond somewhat differently to the solution reaction. The coal substances in lignite, being less condensed and less aromatic and having more cross chains and oxygen-hydrogen linkages, react differently from those of higher rank coals. For all coals, fusain appears to be inert, and the other macerals appear to be more or less readily dissolved. Usually, the coal is processed without any attempt to separate macerals since separation is difficult.

Hydrogen is important to the overall reaction. It can cause hydrogenolysis and also combine with free radicals and unsaturated compounds formed during depolymerization to reduce repolymerization. It is not necessary to have molecular hydrogen present initially, since the reducing agent may consist of carbon monoxide-steam mixtures that work well, particularly with low-rank, high-moisture coals such as lignite.[104] Synthesis gas (H_2 and CO) also will give a high degree of coal solution and has been proposed for commercial operation to avoid the necessity of providing pure hydrogen or pure carbon monoxide.

Equipment and Procedures

In the batch studies, the reactor is a 1-gal stainless steel stirred autoclave capable of processing coal slurries (coal plus solvent) with added hydrogen and/or carbon monoxide at pressures to about 5,000 psi and temperatures to about 900° F. This apparatus and its operation have been described previously.[105]

The solvent used was the fraction of anthracene oil boiling between 100° and 230° C at a pressure of 1 to 2 mm of mercury. A useful parameter determined to characterize the solvent is the infrared ratio (IRR) and represents the ratio of absorbance at 3,070 cm^{-1} to that at 2,950 cm^{-1}. The absorbance at 3,070 cm^{-1} is a measure of C-H stretching of hydrogen on double-bond

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[104] Appell, H. K., I. Wender, and R. D. Miller. Liquefaction of Lignite With Carbon Monoxide and Water. Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Bismarck, N. Dak., May 12-13, 1971. BuMines IC 8543, 1972, pp. 32-39.

[105] Skidmore, D. R., D. S. Gleason, and D. E. Severson. Low-Ash Carbon From Lignite. Technology and Use of Lignite, Proceedings: Bureau of Mines-University of North Dakota Symposium, Grand Forks, N. Dak., April 27-28, 1967. BuMines IC 8376, May 1968, pp. 137-143.

Severson, D. E., D. R Skidmore, and D. S. Gleason. Solution Hydrogenation of Lignite in Coal-Derived Solvents. AIME Trans., v. 247, June 1970, pp. 133-136.

Wright, C. H., and D. E. Severson. Experimental Evidence for Catalyst Activity in Coal Minerals. 163d Nat. Meeting ACS, Boston, Mass., Apr. 10-14, 1972, Preprints, v. 16, No. 2, 1972, pp. 68-92.

carbons, and that at 2,950 cm^{-1} the C-H stretching of hydrogen on single-bond carbons. Thus, the IRR ratio indicates the relative unsaturatedness or aromaticity of the solvent; the higher the value the greater the proportion of aromatics present. With limitations, the lower the IRR ratio, the better the potential hydrogen donor characteristics of the solvent. The IRR is also determined on the solvent boiling range portion of the liquefied coal product.

Usually, a relatively small quantity of lignite, enough for two autoclave runs, was pulverized to pass 100 mesh. The normal lignite charge to the autoclave was a little over 300 grams, corresponding to 200 grams of moisture- and ash-free (maf) coal. In some tests, water was added to replace moisture lost during pulverization to maintain a constant amount of water in the slurry.

Upon completion of a run, the autoclave was cooled to about 400° F, and the product gases were exhausted through a series of cold traps into a collection balloon. The gas was mixed in the balloon, sampled, analyzed, and metered. Hydrogen, methane, ethane, carbon monoxide, carbon dioxide, and nitrogen were determined by gas chromatography, and hydrogen sulfide was determined separately by zinc ammonium sulfate absorption. Heavier hydrocarbons are usually not present in the gas stream. In addition to gas, water and light oil fractions were collected.

After discharge of product gas, the autoclave was repressured with nitrogen and the slurry, still at 400° F, discharged through a dip-tube onto a heated Buchner funnel where the solvent and coal-derived liquids were separated from the mineral constituents and unconverted coal. The products collected were filtrate, filter cake, and autoclave residue.

The collected filtrate was distilled to recover a light oil fraction, a solvent fraction, and a vacuum bottoms. The solvent-refined coal product is a major part of the vacuum bottoms. In most runs not quite as much solvent was recovered as had been charged. In continuous operation, there should be a slight net solvent production, but if not, recycle solvent would have to be prepared by widening the distillation cut to include some of the front end of the vacuum bottoms. The filter cake and other residues were extracted with pyridine to determine the amounts of adhering filtrate, and ash contents were determined on the filter cake, the residue, and the filtrate.

Complete material balances were made on each run, utilizing an IBM 370/165 system. The computer program calculates the amount of liquid products in the filter cake and residues, and distributes this quantity among the liquid fractions according to the distillation cuts of the filtrate. Also calculated are normalized gas compositions and consumption of hydrogen and/or carbon monoxide.

Experimental Results

Over 450 tests have been made investigating the influence of many variables such as time, temperature, solvent, test atmosphere composition, filtration temperature, catalytic effect of cations, solvent-coal ratio, and variety of lignite at different moisture contents.

The results of some of the more recent tests have been discussed.[106] The conditions determined from these tests to be near optimum with the batch autoclave for evaluating hydrogenation-solvation using anthracene oil solvent and synthesis gas (50/50 H_2/CO mixture) included temperature of 750° F, reaction time at temperature of 30 min, filtration temperature of 400° F, and an initial total pressure of 1,000 psi. This initial pressure generally resulted in a maximum pressure during conversion of slightly over 3,000 psi.

Slight differences in solvent quality as revealed by the IRR (infrared ratio) were also investigated. The solvent with the higher IRR (having a higher aromaticity) was theorized to have lower hydrogen donor properties. The higher IRR solvent seemed to give increased fragmentation of both coal and solvent molecules, perhaps because the lower availability of hydrogen resulted in less stabilization of fragments and free radicals and allowed them to react to a greater extent.

Effect of Coal Type

In some earlier tests, various coals, mostly lignites, were solubilized in a hydrogen atmosphere using both anthracene oil and tetralin as solvent. Lignite was obtained from working mines and from drill hole samples.

Solvent/maf coal weight ratio was 3.6, time at reaction temperature of 400° C was 2 hours, and initial hydrogen pressure was 1,500 psi. Moisture content was less than 5 pct. Parameters used for evaluation were percent yield of vacuum bottoms, and percent of coal substance converted, both on an maf basis. Where replicated runs were made, average data are summarized in table 37.

Generally, differences existed between coals of different rank and even between various lignites. In one instance, where two beds of lignite were being mined at the same location, differences existed between the two seams. Response of subbituminous coals were quite similar to some lignites, but the yields from bituminous coals were higher. All tests were at the same conditions that were not optimized for any given coal; in fact, no attempt was made to define optimum conditions.

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[106] Souby, A. M., and D. E. Severson. Project Lignite: Convenience Fuels From Northern Great Plains Province Lignite. Unpublished report, University of North Dakota Engineering Experiment Station, Grand Forks, N. Dak.; available upon request from A. M. Souby.

TABLE 37. - Effect of coal type on solubilization

Coal identification	Anthracene oil solvent		Tetralin solvent	
	Vacuum bottoms yield, pct of maf coal	Coal substance converted, pct of maf coal	Vacuum bottoms yield, pct of maf coal	Coal substance converted, pct of maf coal
Lignite:				
Breezy Flat.....	44.0	68.0	66.4	82.5
Velva..........	54.7	78.5	67.7	88.0
Zap............	52.7	76.1	70.8	86.7
North Beulah....	52.9	77.4	66.9	85.3
South Beaulah...	72.2	86.0	68.6	83.2
Glenharold, upper seam.....	56.3	76.0	72.5	87.7
Glenharold, lower seam.....	62.8	87.6	54.3	95.4
Larson.........	63.3	82.1	71.5	90.7
Kincaid.........	70.5	87.1	74.6	91.4
Cedar Coulee....	56.8	85.8	66.9	96.0
Subbituminous:				
Rosebud.........	67.4	83.5	73.3	80.7
Boomer..........	61.2	80.6	63.8	82.7
Bituminous:				
Kentucky No. 1..	79.3	91.0	79.2	94.5
Pittsburgh seam.	79.0	93.5	85.6	95.7

Influence of Solvent Type

Many organic solvents have been proposed or used in hydrogen solution of coal. Several solvents were investigated in micro-batch autoclave.[107] In these experiments, about 0.6 gram of dried pulverized lignite was reacted with selected solvent for 2 hours at 400° C under an initial hydrogen pressure of 1,500 psi. Fifteen solvents were tested including aromatic, aliphatic, amino, phenolic, heterocyclic, and cycloparaffinic compounds. Weight ratio of solvent to lignite was 4:1. Percent solubilization (maf lignite converted to liquids or gases) was used as a parameter. Results are summarized in table 38.

Solvents exhibiting over 70 pct solubilization were considered very effective (tetralin and 1,2,3,4-tetrahydroquinoline), and those below 40 pct, least effective (benzene, phenol, o-cresol, and n-butylamine). A fraction of anthracene oil gave a solubilization of 62.8 pct. However, in the batch autoclave work under different conditions and with a different coal, anthracene oil in some cases gave over 90 pct solubilization.

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[107] Desai, S. N. Thermal Dissolution of Lignite Under Hydrogen Pressure. M.S. Thesis, University of North Dakota, Grand Forks, N. Dak., June 1969, 45 pp.

TABLE 38. - Effect of solvent on solubilization of lignite

Solvent	Average solubilization, pct maf lignite
Benzene	28.3
Phenol	31.6
o-Cresol	36.7
n-Butylamine	37.2
Cyclohexane	41.3
1-Naphthylamine	43.2
o-Phenylphenol	45.0
Quinoline	46.5
Phenanthrene	48.1
Anthracene	49.6
Naphthalene	49.9
Alpha-naphthol	62.8
Tetralin	71.7
1,2,3,4-Tetrahydroquinoline	93.4
Anthracene oil	62.8

Influence of Cations on Extraction

Earlier work had indicated that the naturally occurring minerals in lignite had a catalytic effect on the reaction of hydrogen or carbon monoxide with the lignite.[108] This work was done with a number of lignites of varying mineral content and composition and with lignite to which sodium bicarbonate or potassium carbonate was added. The amount of hydrogen reacting and the conversion of the lignite could be related to the amount of sodium, potassium, and iron present in the lignite. The current work in which various cations are added to extracted lignite is an extension of these previous catalytic studies.

In lignite, cations, particularly those of sodium and calcium, are associated with the coal substance rather than with the mineral ash. These cations are bound to the coal in such a fashion that they can be removed by an ion-exchange process. If lignite is washed with an acid solution, sodium and calcium concentrations are greatly reduced. Other cations of soluble salts can then be added directly to the coal substance by placing the acid-extracted coal in a solution of the desired cations. The concentrations of cation added can be controlled by varying solution concentration and time of contact.

A bulk sample of North American lignite was extracted in a glass column with 1 N HCl, and the ash content on a dry basis was reduced from 10.0 to 3.2 wt-pct. Over 95 pct of the ash after extraction was alumina, silica, and iron oxide. Various concentrations of cations such as sodium, potassium, iron, calcium, cobalt or nickel were added to the extracted lignite by reverse ion exchange. In cases where the cation formed a soluble acetate, this salt was used since the acetate does not add any "foreign" material to the lignite. In

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[108] Third reference cited in footnote 105.

many cases individual cation concentration can be increased over the naturally occurring level without increasing other mineral components.

Partial results from the tests under the aforementioned conditions with cation addition are given in table 39. More complete data are presented in table 40. The highest yield of vacuum bottoms under the conditions used was obtained with the extracted coal as well as a relatively low yield of gas. The catalyst ions generally produced more conversion to lower-molecular-weight products, such as light oil and gas.

Sodium and potassium, which are water gas shift catalysts, gave high consumption of water during the reaction, and the untreated lignite had high water consumption because of its alkali metal content. Water consumption with the hydrogenation catalysts--cobalt, nickel, and iron--was much lower.

The effect of a given cation was quite insensitive to the amount added in the ranges studied, which for the alkali metals were of the same order of magnitude as the amounts naturally present in coal mineral matter.

Solvent recovery was less than 100 pct of solvent charged for all of these experiments. The shift catalyst additions gave closest to self-sufficiency of solvent without loss of vacuum bottoms yield over the extracted lignite.

The conversion of lignite to gases and liquids was about 90 pct for most cases. Addition of cations causes greater production of light oil and gas than would be desired for SRC production.

TABLE 39. - Selected tests indicating effect of added cation on solubilization

Run	455	454	425	423	416	428	429	433	436
Cation	Raw	Extracted	Na	K	Ca	Fe	Co	Ni	Mo
Catalyst level, mM/kg[1]	(2)	(3)	360	170	220	230	320	310	450
Reclaimed solvent IRR	-	-	0.63	0.55	0.49	0.52	0.38	0.53	0.79
Conversion[4]	92	91	94	94	91	95	88	92	89
Vacuum bottoms[4]	59	72	64	57	63	67	68	68	65
Light liquids[4]	21	14	13	19	14	24	19	25	30
Gas[4]	41	25	36	39	38	27	21	22	41
Water consumption[4]	14	2	10	14	2	2	-4	1	9
Solvent recovered[5]	92	92	96	93	89	88	88	89	81

[1] Millimoles of cation per kilogram of raw lignite.

[2] Raw lignite, no cations added.

[3] Extracted lignite, no cations added.

[4] Pct of maf lignite charged.

[5] Pct of solvent charged.

TABLE 40. - Influence of added cations on solution hydrogenation of lignite: Summary of operating conditions and yields

Run	408	414	415	422	425	426	416	431	456	423	453	424	428	427	429	437	432	433	435	434	436	438	439	454	455
Test conditions:[1]																									
Time, hr	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Av temp, ° F	772	765	768	754	750	749	766	748	757	754	751	750	748	747	751	750	753	749	750	748	744	753	748	750	752
Maximum pressure, psi	3,195	3,340	3,395	3,125	3,175	3,165	3,280	3,205	3,320	3,135	3,170	3,255	3,175	3,135	3,225	3,215	3,165	3,155	3,045	2,875	2,975	3,120	2,880	3,230	3,210
Gas charged	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	H_2	CO/H_2	CO/H_2	H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2	CO/H_2
IRR[2]	1.29	1.29	1.29	1.46	1.46	1.46	1.46	1.52	1.52	1.46	1.52	1.46	1.46	1.46	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52
Lignite	Raw	Extracted		Raw	← Extracted North American Lignite →																	Raw	Extracted	Extracted	Raw
Cation added	-	-	Na	-	Na	Na	Ca	Ca	Ca	K	K	Fe	Fe	Co	Co	Co	Ni	Ni	Ni	Mo	Mo	HDS-3A	HDS-3A	-	-
mM/kg[3]	-	-	228	-	265	369	221	166	-	167	-	152	232	107	320	320	140	314	314	150	447	(4)	(5)	-	-
Material in, g:																									
Coal and H_2O	305.2	306.6	317.3	319.7	310.2	313.5	316.0	310.9	309.5	308.8	314.8	307.5	308.6	306.4	310.9	311.4	308.2	311.2	310.9	308.4	307.9	329.6	323.2	307.1	322.6
Solvent	400.0	400.2	400.2	399.3	400.1	399.9	400.5	399.9	400.4	399.3	400.2	399.5	400.2	398.9	399.6	400.4	399.6	399.9	399.9	400.2	399.6	399.4	399.9	400.2	399.6
Gas	118.7	121.2	128.0	123.0	122.3	118.1	98.2	119.8	117.1	119.2	118.8	124.8	116.1	116.9	119.4	14.7	120.1	120.3	14.7	121.4	119.3	122.0	119.9	118.5	119.9
Total	823.9	828.0	845.5	842.0	832.6	831.5	814.7	830.6	827.0	827.3	833.8	831.8	824.9	822.2	829.9	726.5	827.9	831.4	725.5	830.0	826.8	851.0	843.0	825.8	842.1
Material out, g:																									
Filtrate	432.5	445.2	454.0	416.4	467.5	450.0	450.0	460.0	428.7	474.5	408.0	467.5	472.5	470.0	436.5	451.5	455.0	464.0	464.5	384.5	356.0	382.0	386.5	435.0	434.9
Filter cake	88.0	41.3	35.5	93.1	43.0	33.5	40.5	64.5	69.1	30.5	32.5	35.5	45.0	51.5	81.5	72.0	61.0	61.5	54.5	107.0	118.0	129.0	105.0	68.5	71.4
Gas	159.6	164.0	183.8	184.9	187.4	194.9	169.1	157.1	177.8	189.6	207.7	174.0	165.9	160.8	157.7	38.8	164.3	160.0	33.9	192.5	186.8	189.6	190.6	161.7	196.4
Residues	21.5	17.0	22.0	30.0	22.5	45.5	21.5	14.5	30.7	24.5	29.5	18.5	15.5	19.0	21.0	17.0	20.0	13.5	10.5	32.0	24.5	31.5	32.0	24.0	31.4
H_2O	53.5	93.5	82.0	67.0	78.4	66.1	98.3	98.5	101.2	68.6	56.4	87.2	91.8	97.3	104.3	123.4	95.2	95.0	122.1	67.3	69.4	57.8	60.6	91.2	73.1
Light oil	7.5	13.0	13.0	13.5	5.1	12.9	11.7	11.7	12.5	8.9	15.1	13.8	10.2	7.7	10.7	11.1	8.3	11.5	15.9	11.7	13.1	8.2	39.4	13.3	9.9
Total	762.6	774.0	790.3	804.9	803.9	802.9	793.1	806.3	820.0	796.6	749.2	796.5	800.9	806.3	811.7	713.8	803.8	805.5	701.4	795.0	767.8	798.1	814.1	793.7	817.1
Liquid products, g:																									
Light oil	24.6	23.3	31.1	25.0	24.5	32.1	27.4	40.2	43.7	36.6	39.0	30.4	47.6	28.8	37.2	49.9	44.9	47.7	42.2	50.6	55.0	43.6	52.7	26.5	40.9
Solvent	345.6	331.9	355.9	328.5	369.3	365.4	347.2	348.2	347.8	371.8	301.0	357.8	342.7	365.6	343.1	336.7	341.0	344.2	350.3	356.5	302.0	337.6	341.7	350.2	357.0
Vacuum bottoms	138.0	130.4	114.6	158.9	123.5	120.7	122.6	137.1	122.8	109.9	125.2	129.6	130.9	130.1	133.7	140.7	133.3	132.4	132.5	102.3	120.0	126.8	128.6	139.2	114.1
100 pct recovery basis, products, g:																									
Light oil	26.6	24.9	33.3	26.1	25.4	33.2	28.1	41.4	44.1	38.0	43.4	31.8	49.0	29.4	38.0	50.8	46.3	49.2	43.6	52.8	59.2	46.5	54.6	27.6	42.2
Solvent	373.4	355.1	380.7	343.7	382.5	378.4	356.7	358.7	350.8	386.1	335.0	373.6	353.0	372.8	350.8	342.7	351.2	355.3	362.3	372.2	325.2	360.0	353.8	364.4	367.9
Vacuum bottoms	149.1	139.5	122.6	166.2	127.9	125.0	125.9	141.2	123.9	114.2	139.3	135.3	134.8	132.6	136.7	143.2	137.3	136.7	137.1	106.8	129.2	135.2	133.2	144.8	117.6
Total liquid	549.1	519.5	536.6	536.0	535.8	536.7	510.7	541.3	518.8	538.3	517.7	540.7	536.8	534.8	525.5	536.7	534.8	541.2	543.0	531.8	513.6	541.7	541.6	536.8	527.7
Net liquid	149.1	119.3	136.4	136.7	135.7	136.8	110.2	141.4	118.4	139.0	117.5	141.2	136.6	135.9	125.9	136.3	135.2	141.3	143.1	131.6	114.0	142.3	141.7	136.6	128.1
Net gas	53.7	54.2	68.6	70.4	71.8	83.7	75.5	42.0	62.2	77.7	112.3	56.9	54.8	47.1	41.9	24.8	49.1	44.9	20.4	79.6	81.9	80.2	77.5	49.7	82.5
Net H_2O and ash	-30.6	2.5	-19.4	-27.2	-19.3	-34.5	-4.3	1.2	2.5	-28.1	-39.5	-8.9	-4.9	0.6	7.2	26.0	-1.8	-1.6	27.1	-27.9	-17.6	-43.9	-36.9	-4.7	-27.3
Unconverted coal	25.3	24.3	14.5	19.9	12.0	13.8	18.8	15.4	17.4	11.2	9.9	10.9	13.7	15.0	25.0	13.1	17.3	15.6	9.3	16.8	21.6	16.0	17.8	18.7	16.7
Yields, wt-pct maf coal:																									
Net gas	27.2	27.1	34.3	35.2	35.9	41.9	37.7	21.0	31.0	38.9	56.1	28.4	27.4	23.7	21.0	12.4	24.6	22.4	10.2	39.8	41.0	41.2	38.7	24.8	41.3
Light oil	13.5	12.4	16.6	13.1	12.7	16.6	14.0	20.7	22.0	19.0	21.7	15.9	24.5	14.8	19.0	25.4	23.2	24.6	21.8	26.4	29.6	23.9	27.3	13.8	21.1
Vacuum bottoms	75.5	69.6	61.3	83.2	63.9	62.6	62.9	70.5	61.8	57.2	69.6	67.6	67.3	66.8	68.4	71.5	68.7	68.3	68.6	53.3	64.7	69.4	66.6	72.3	58.8
Net liquid	75.5	59.6	68.2	68.4	67.8	68.5	55.0	70.6	59.1	69.6	58.7	70.6	68.2	68.4	63.0	68.1	67.6	70.6	71.6	65.7	57.1	73.1	70.8	68.2	64.1
Conversion	87.2	87.9	92.8	90.0	94.0	93.1	90.6	92.3	91.3	94.4	95.0	94.6	93.2	92.4	87.5	93.5	91.3	92.2	95.4	91.6	89.2	91.8	91.1	90.7	91.6
Solvent recovered:																									
Wt-pct	93.4	88.7	95.1	86.1	95.6	94.6	89.1	89.7	87.6	96.7	83.7	93.5	88.2	93.5	87.8	85.6	87.9	88.8	90.6	93.0	81.4	90.1	88.5	91.0	92.1
IRR[2]	0.32	0.32	0.58	0.61	0.63	0.50	0.49	0.50	1.00	0.55	0.93	0.61	0.52	0.51	0.38	0.39	0.50	0.53	0.65	0.85	0.79	0.76	0.76	0.90	0.90

[1] The solvent used was University of North Dakota middle fraction run anthracene oil.

[2] IRR: Infrared ratio, ratio of absorbance at 3,070 cm^{-1}. Higher values indicate higher aromaticity.

[3] Millimoles of cation per kilogram of raw lignite.

[4] American Cyanamid HDS-3A nickel-molybdenum catalyst: 55 mM Ni and 138 mM Mo per kg raw lignite.

[5] American Cyanamid HDS-3A nickel-molybdenum catalyst: 18 mM Ni and 45 mM Mo per kg extracted lignite.

THE LIQUEFACTION OF SUBBITUMINOUS COAL

by

Dat Nguyen,[109] Warren Scarrah,[110] and Lloyd Berg[111]

PART I: THE USE OF CARBON MONOXIDE AND WATER

Introduction

The carbon monoxide-water approach to coal liquefaction was conceived by Fischer[112] in the 1920's, but recently it has received renewed interest following the work of Appel and coworkers.[113] Research has been conducted in the past few years at Montana State University with the objective of adapting the process to Montana subbituminous coal.[114] With possible commercialization of the process as the ultimate objective, the present work deals in particular with the effects of temperature, pressure, and concentration of a number of selected catalysts and solvents on the batchwise liquefaction of Colstrip coal using a 5-min reaction time.

Experimental Procedure

Runs were made in a 500-cm^3 Inconel 600 autoclave. Where possible, the catalyst was dissolved in water, and freshly ground (minus 40 mesh) coal was soaked in the solution before being mixed with the solvent. Where the catalyst was water insoluble, it was manually mixed with the coal, and the autoclave was charged with the mixture. The autoclave was sealed and pressurized with carbon monoxide to an initial pressure of 1,000 to 1,500 psi. After being brought to the final temperature in a rocking heater and maintained at this temperature for the required "operating time," the autoclave was removed from the heater and let cool to room temperature. Soxhlet extraction was used

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[111] Professor and Head.

All authors are with the Chemical Engineering Department, Montana State University, Bozeman, Mont. Research partially under a Bureau of Mines-Montana State University Fellowship.

[112] Fischer, F. The Conversion of Coal Into Oils. D. Van Nostrand Co., Princeton, N.J., 1925, pp. 174-201.

[113] Appel, H. R., and I. Wender. The Hydrogenation of Coal With Carbon Monoxide and Water. Div. Fuel Chem. Preprints, ACS, v. 12, No. 3, 1968, pp. 220-222.

[114] York, W. J. Reduction of Subbituminous Coal and Lignite Using Carbon Monoxide and Water. Ph.D. Thesis, Montana State University, Bozeman, Mont., 1971, 153 pp.

to determine the amount of moisture- and ash-free (maf) coal that had been converted into benzene-soluble compounds, allowing calculation of percent conversion.

Solvent and Catalyst Selections

Two- and three-ring compounds including decalin, tetralin, naphthalene, anthracene, and phenanthrene were tested for solvent effects. "Coal solvent" from FMC was also tested. Nitrogen-base solvents were avoided because of potential environmental problems in large-scale application.

Two types of catalysts were tested. The first type, catalyzing the water-gas shift reaction between carbon monoxide and water to form hydrogen, included sodium bicarbonate, sodium carbonate,[115] sodium formate, and ferric oxide. The selection of the sodium compounds was on the basis of (1) solubility in water, which insured their even distribution among the coal particles, and (2) relative cheapness, a requirement if they were to be used on a once-through basis.

The selection of the second type of catalysts, which have been hypothesized to catalyze the hydrogenation reactions,[116] was much limited by factors such as cost, water solubility, and corrosiveness to process equipments. As a result, only stannous chloride has been tested. Because of environmental considerations, ammonium chloride, which has been reported to aid in the fragmentation of the coal before the hydrogenation reactions could take place,[117] was not used.

Results and Discussion

Effects of Temperature and Pressure

Figure 131 shows the effect of reaction temperature on conversion. Very little benzene-soluble product was obtained at 300° C, whereas the conversion was over 70 pct at 475° C. Some of this increased conversion is attributed to the heating time from 300° C onward, which was close to 25 min, excluding the final 5-min operating time.

In addition to final temperature, pressure also has a significant effect on conversion. When no carbon monoxide was used, the solvent dissolved close to 40 pct of the coal, with no hydrogenation having taken place (fig. 132). Conversion increased to roughly 80 pct at 1,500 psi initial carbon monoxide pressure. At higher pressures, the effect of increasing pressure was minor. It is reasonable to assume from this behavior that pore diffusion, important in coal hydrogenation, is no longer a controlling factor above an initial pressure of 1,500 psi.

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[115] Work cited in footnote 114.

[116] Weller, S., E. L. Clark, and M. G. Pelipetz. Mechanism of Coal Hydrogenation. Ind. and Eng. Chem., v. 42, No. 2, 1950, pp. 334-336.

[117] Work cited in footnote 116.

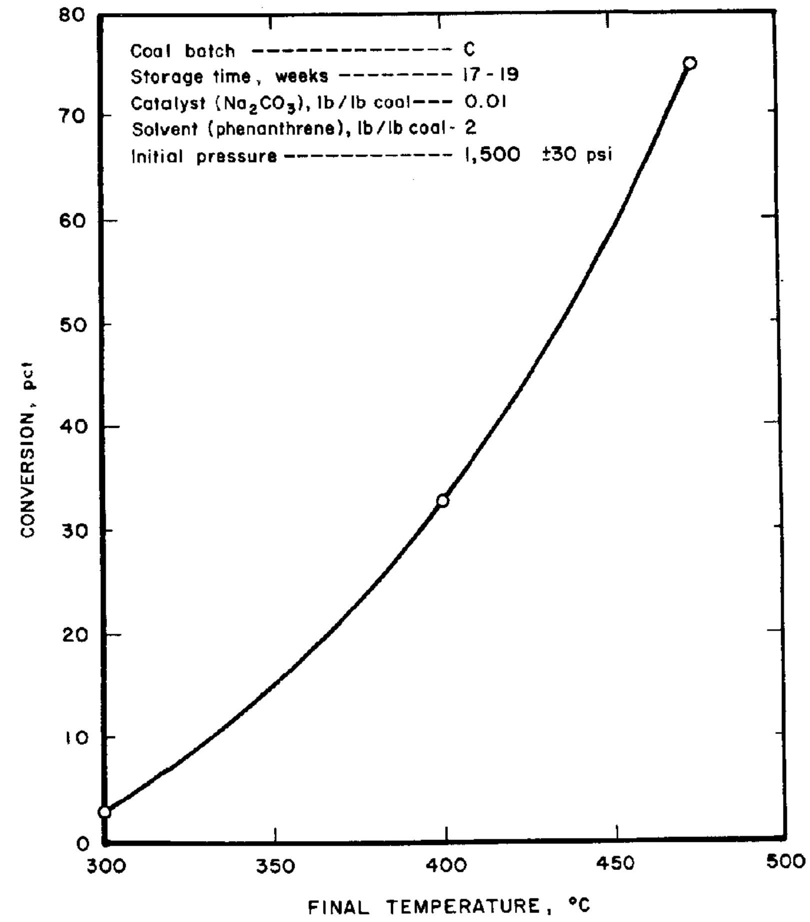

FIGURE 131. - Effect of reaction temperature on conversion.

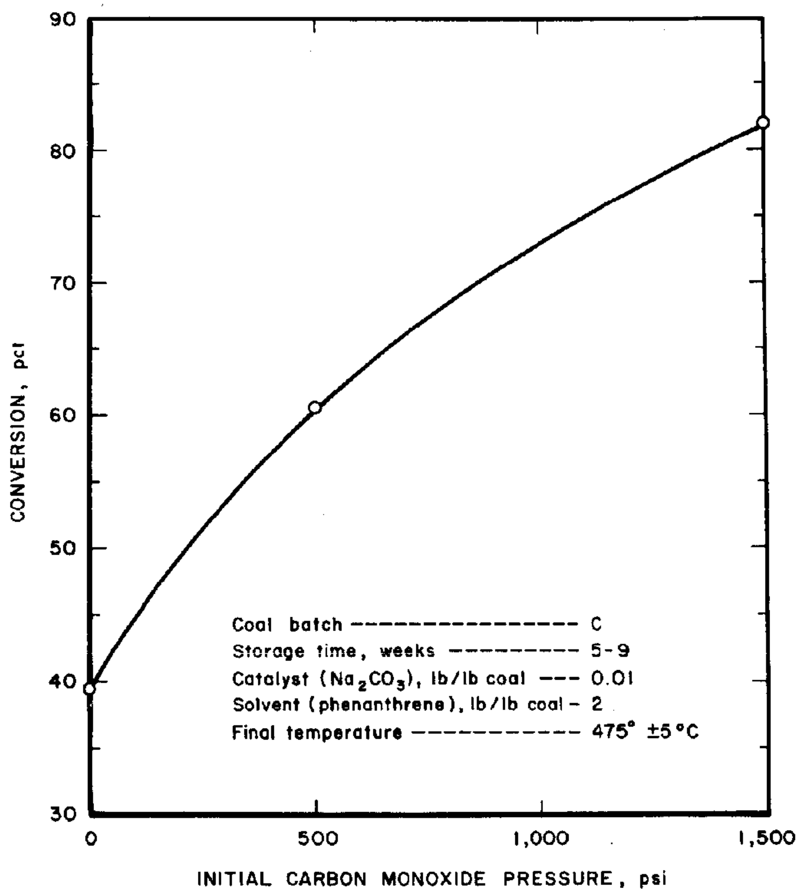

FIGURE 132. - Effect of initial pressure on conversion.

Effects of Type and Concentration of Solvent

Figure 133 shows the decrease in conversion with increase in H/C ratio (double) ring solvents. This decrease tends to confirm a basic assumption that coal contains compounds mostly of the aromatic types. Three-ring compounds gave higher conversions than did double-ring solvents, with phenanthrene giving about 20 pct higher conversion than did anthracene (fig. 134). Phenanthrene gave the highest conversion when present at a 2:1 ratio with respect to coal (fig. 135). This behavior could be attributed to a compromise between having sufficient solvent for the solution reaction and an excess, in which case the extra solvent would compete with the coal for the available hydrogen, and conversion decreased.

FMC "coal solvent" was found unsuitable because of conversions lower than 50 pct with considerable carbon lay-down being observed in the unconverted portion of the coal.

Effects of Catalyst Type and Concentration

Results on conversion using sodium carbonate are still being confirmed. The general trend with both sodium formate and sodium bicarbonate as catalysts was that conversion increased with increasing concentrations of the salts (figs. 136-137). In the particular case of sodium formate, there was a significant decrease in conversion at very low formate concentrations, followed by a gradual increase. It would seem that at these higher concentrations of the formate, the catalytic action of the salt overcomes the negative effects that resulted from alkalinity of the salt on conversion. For sodium bicarbonate, no such counteracting behavior was observed.

The general trend observed with the sodium salts was also exhibited with ferric oxide, though not as pronounced (fig. 138).

Experiments with stannous chloride did not show any significant catalytic effect by this compound on conversion, as shown in the following tabulation:

Order of run	Weight $SnCl_2$/weight coal	Conversion, pct
1	0	51.7
3	0	61.6
2	.0006	58.2
5	.0006	61.4
4	.0012	58.6
6	.0012	58.4

The reaction autoclave was not equipped with a glass liner because such design would be unrealistic for large-scale operations. Consequently, the presence of chlorine compounds from the $SnCl_2$ could have altered the "neutrality" of the reactor wall, causing interactions in consecutive runs and rendering the results inconclusive.

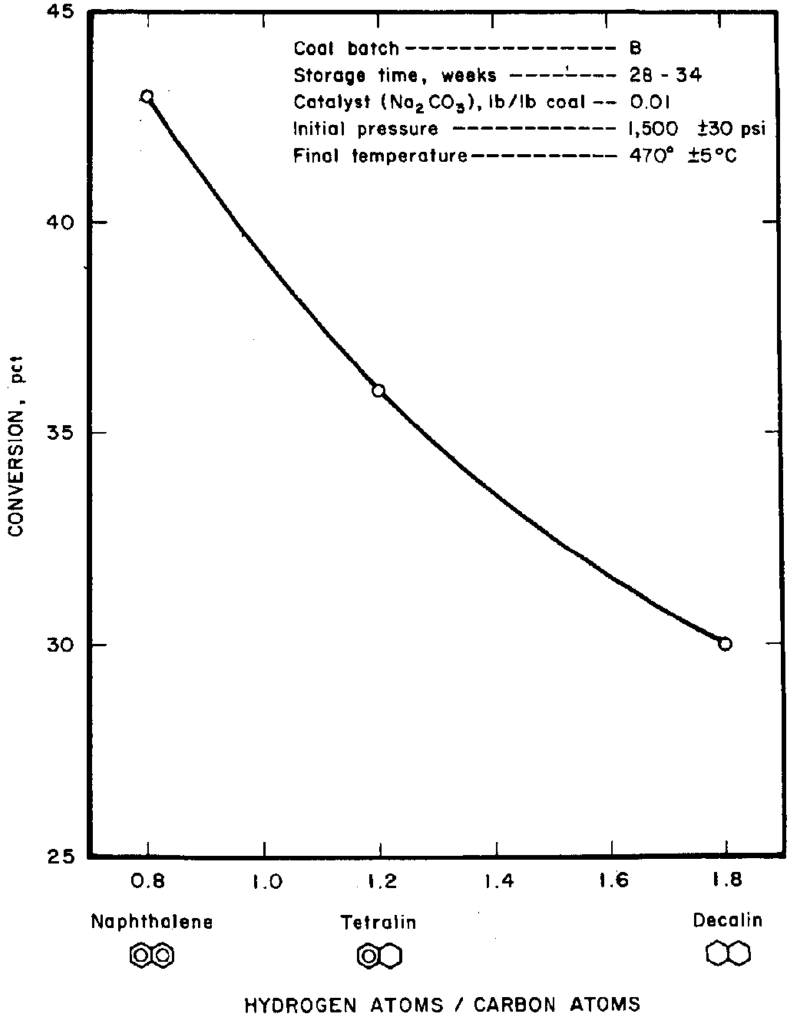

FIGURE 133. - Effect of solvent aromaticity on conversion.

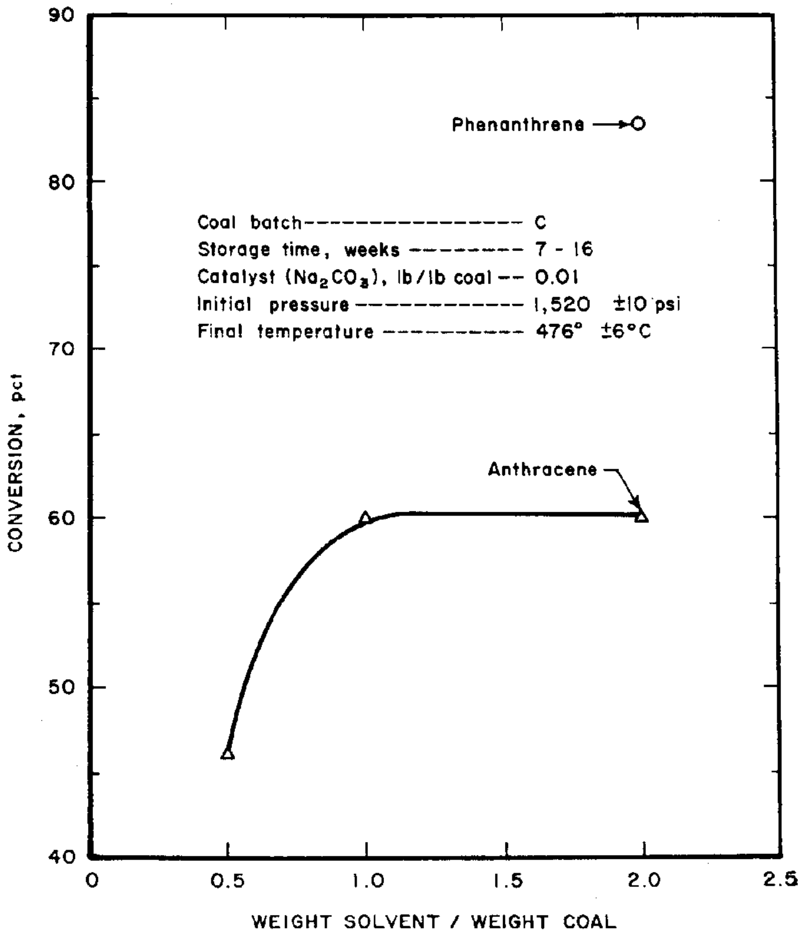

FIGURE 134. - Effect of anthracene concentration on conversion.

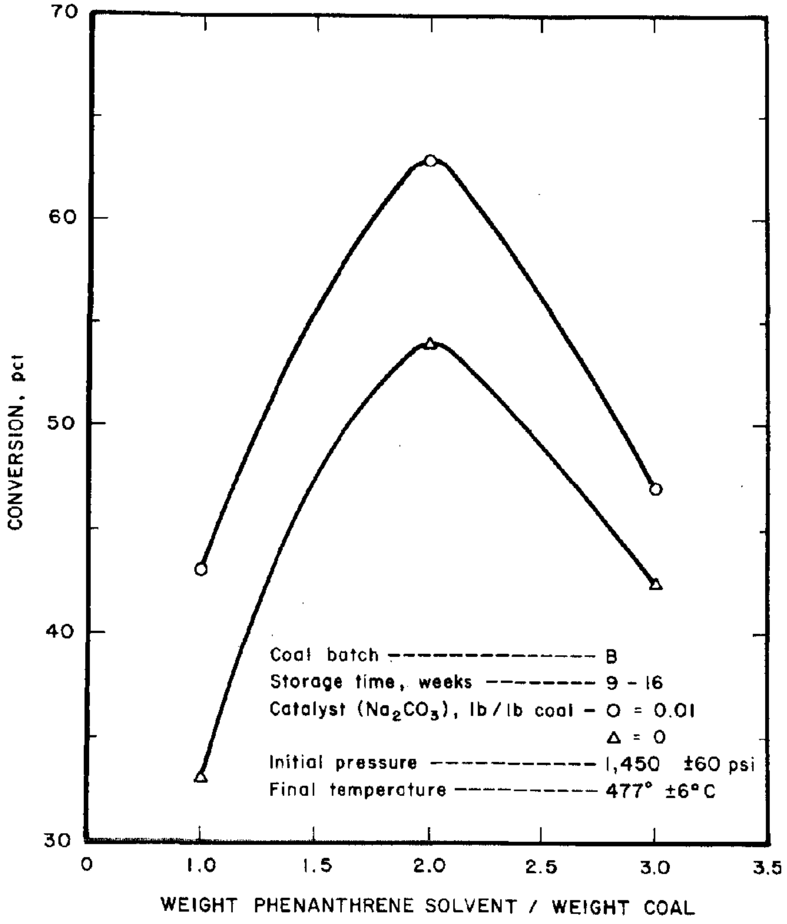

FIGURE 135. - Effect of phenanthrene concentration on conversion.

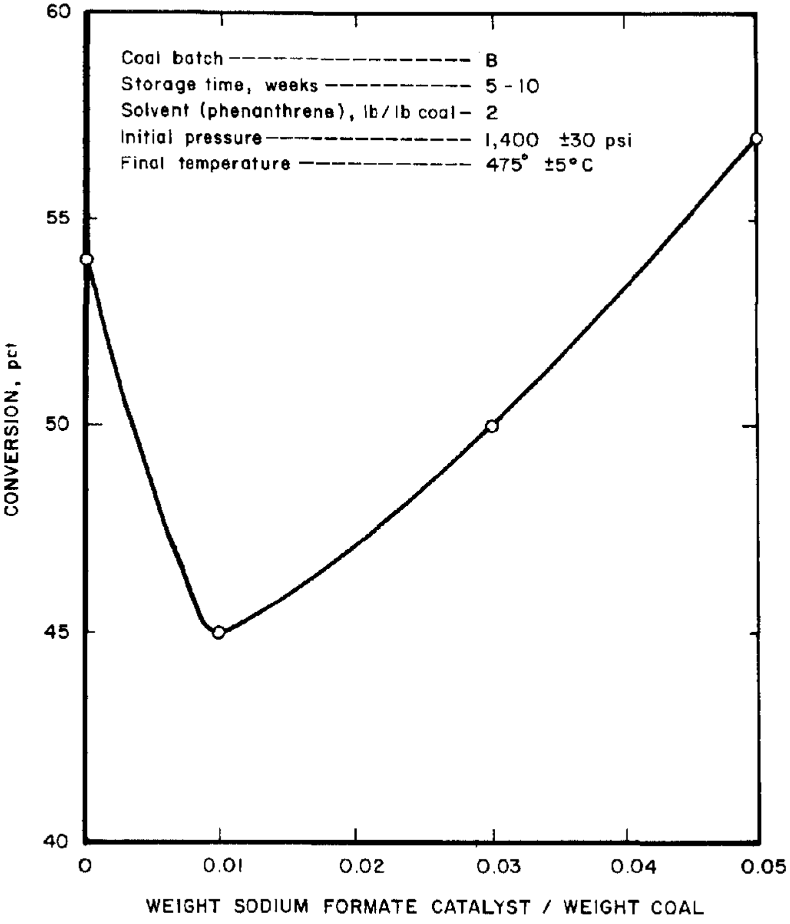

FIGURE 136. - Effect of sodium formate concentration on conversion.

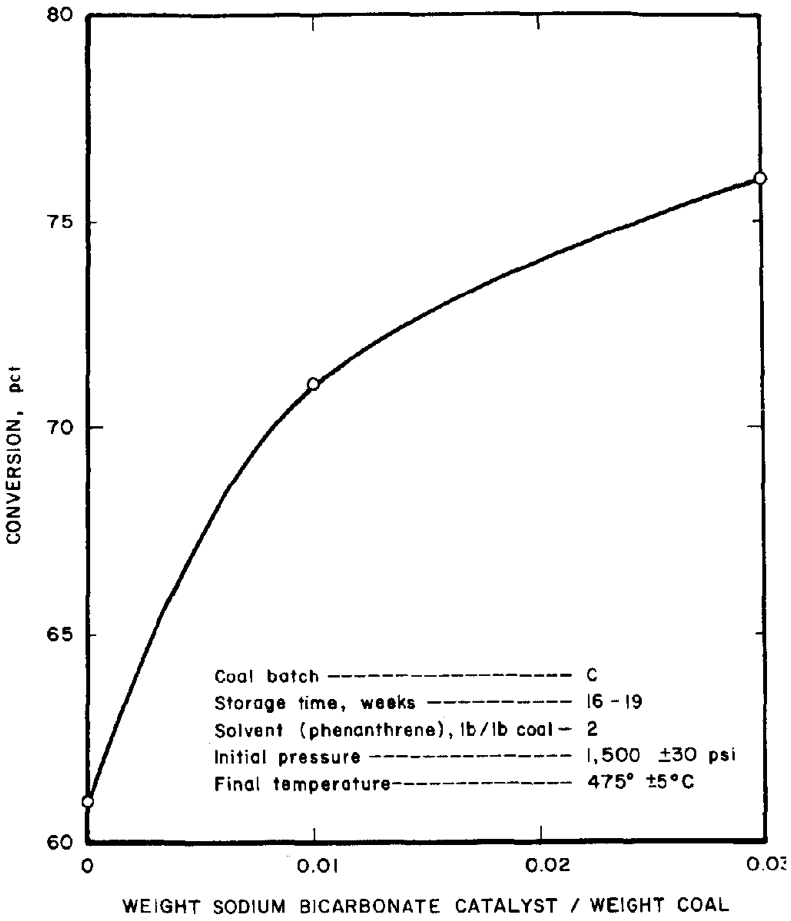

FIGURE 137. - Effect of sodium bicarbonate concentration on conversion.

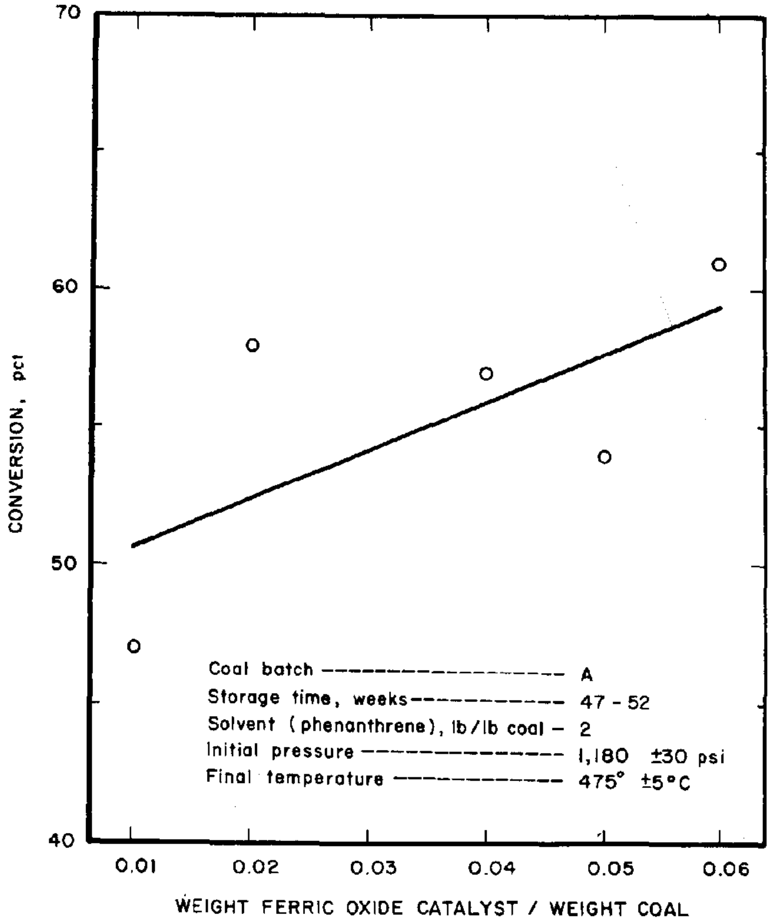

FIGURE 138. - Effect of ferric oxide concentration on conversion.

Conclusions--Part I

At a reaction time of 5 min, it is possible to convert 70 to 80 pct subbituminous coal into benzene-soluble products by hydrogenation with carbon monoxide and water. To warrant the highest conversion possible at such a short operating time, a final temperature of 475° C and final pressure of about 5,500 psi are necessary. Phenanthrene, present at a 2:1 ratio with respect to coal, appears to be the best solvent for the process, when used with 1 to 3 pct of a sodium salt (bicarbonate or possibly carbonate) as a catalyst.

PART II. THE USE OF MASSIVE QUANTITIES OF MOLTEN SALT CATALYST

Introduction

The investigation of hydrocracking catalysts for coal conversion by the Consolidation Coal Co.[118] led to the selection of massive quantities of molten zinc chloride ($ZnCL_2$). Zinc chloride is essentially a cracking catalyst and was found to be superior to hydrofining and dual-function (cracking and hydrogenation) catalysts in these respects: (1) Higher conversion rates, (2) higher levels of conversion, (3) greater yields of gasoline-range distillates, and (4) milder operating conditions of temperature and pressure.

The principal disadvantage in using a molten zinc chloride catalyst was that the hydrocarbon and salt phases did not separate after the reaction was concluded. However, this problem was solved by research at Montana State University.[119] Potassium chloride (KCl) was added to the zinc chloride to reduce the viscosity of the molten salt; the hydrocarbon and salt phases separated cleanly and no deleterious effects were observed relative to the reduction of catalyst activity.

The experiments described in the present report were performed (1) to investigate the effects of process parameters on the conversion of two Montana coals and (2) to explore methods for extending the catalyst life to combat poisoning caused by the nitrogen in the coal.

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[118] Zielke, C. W., R. T. Struck, J. M. Evans, C. P. Costanza, and E. Gorin. Molten Salt Catalysts for Hydrocracking of Polynuclear Hydrocarbons. I&EC Process Des. and Develop., v. 5, No. 2, April 1966, pp. 151-157.
Zielke, C. W., R. T. Struck, J. M. Evans, C. P. Costanza, and E. Gorin. Molten Zinc Halide Catalysts for Hydrocracking Coal Extract and Coal. I&EC Process Des. and Develop., v. 5, No. 2, April 1966, pp. 158-164.
Consolidation Coal Co. Research on Zinc Chloride Catalyst for Converting Coal to Gasoline-Phase I-Hydrocracking of Coal and Extract With Zinc Chloride. V. 3, Book 1, Res. and Develop. Rept. No. 39, Office of Coal Research, Washington, D.C., 1968, 317 pp.

[119] Malsam, J. S. Coal Hydrogenation Studies Using a KCl-$ZnCl_2$ Molten Salt Mixture as a Catalyst. M.S. Thesis, Chemical Engineering Department, 1970, Montana State University, Bozeman, Mont., 45 pp.

Experimental

Experiments were made in a 500-ml Parr rocking-bomb reactor. Two Montana coals were used: Colstrip subbituminous and Savage lignite. The coal and salts were dried for at least 24 hours at a temperature of 105° C, after which the solids were charged to the reactor. Enough hydrogen was added to attain the desired pressure at the reactor operating temperature. After cooling, the gas was discharged from the reactor, and the remaining solids and liquids were extracted with benzene (Soxhlet apparatus) for 24 hours. Coal conversion (moisture- and ash-free basis) was defined as that quantity of coal that was soluble in benzene after reaction. This procedure was modified when making the catalyst-life extension tests; only one batch of salts was used for a number of sequential runs. After extraction, the original salt mixture and any remaining coal residue were charged to the reactor along with the new coal for the next sequential run. Also, during some tests the gas was removed at the reactor operating temperature immediately after the experiment was completed.

Process Parameter Tests

The effects of seven process parameters were investigated in parameter screening tests (table 41). Fractional factorial experimental designs were used to reduce the number of experimental runs required to determine the effects of the parameters and their interactions. One-sixteenth fractional factorials (eight experimental runs) made on each coal showed the following: (1) The process parameters were found to have essentially the same effects on conversion for both coals, (2) the variation due to experimental error was determined, and (3) a one-sixteenth fraction factorial was too small to separate the effects of the parameters.

TABLE 41. - Parameters (factors) and their values (levels) in parameter screening tests

Factor	Factor levels	
	-	+
Temperature........................° C..	350	450
Pressure........................psig..	2,000	3,000
Time............................min..	15	60
Mixing, reactor agitation............	Static	Rocking
Salt:coal (weight ratio)[1]............	2:1	4:1
KCl:$ZnCl_2$ (mole ratio)...............	1:2	1:1
Coal size (U.S. sieve series).........	Minus 40 plus 100	Minus 100

[1] 20 grams of coal were charged to the reactor for each run.

As a result only Colstrip subbituminous coal was used in subsequent runs and another different set of eight experimental runs was made in a one-eighth fractional factorial design.

The experimental runs and corresponding conversions for the one-eighth fractional factorial are given in table 42. The analysis of variance (table 43) can be interpreted as follows:

(1) At the particular levels of the factors investigated in this study, the following factors have no significant effect on conversion:

(a) Pressure,

(b) salt:coal weight ratio,

and (c) $KCl:ZnCl_2$ mole ratio.

(2) Two groups of interactions show significance between two factors (this fractional factorial design does not allow the complete separation of interaction effects but the most likely interactions in each group are marked with an asterisk):

(a) Temperature - time*; pressure - particle size*; mixing - $KCl:ZnCl_2$ mole ratio.

(b) Temperature - $KCl:ZnCl_2$ mole ratio; pressure - salt:coal weight ratio; time - mixing*.

Tests were run to compare sodium chloride and potassium chloride as components to reduce the viscosity of the molten salt. Because the less expensive sodium chloride was found to be as effective as potassium chloride, it was used in subsequent tests.

TABLE 42. - One-eighth fractional factorial experimental design and conversions for parameter screening tests

Factor	Experimental design factor level[1]															
Temperature	-	+	-	+	-	+	-	+	+	-	+	-	+	-	+	-
Pressure	-	-	+	+	-	-	+	+	+	+	-	-	+	+	-	-
Time	-	-	-	-	+	+	+	+	+	+	+	+	-	-	-	-
Mixing	+	-	-	+	+	-	-	+	-	+	+	-	-	+	+	-
Salt:coal	+	-	+	-	-	+	-	+	-	+	-	+	+	-	+	-
$KCl:ZnCl_2$	+	+	-	-	-	-	+	+	-	-	+	+	+	+	-	-
Coal size	-	+	+	-	+	-	-	+	+	-	-	+	-	+	+	-
Conversion of Colstrip subbituminous[2] ....wt-pct..	8	41	16	33	29	33	13	90	52	24	61	17	30	20	34	8

[1] See table 41 for values corresponding to the factor level.
[2] Based on maf coal.

TABLE 43. - Analysis of variance for one-eighth fractional factorial experimental design for parameter screening tests

Source	Degrees of freedom	Sum of squares	Mean square	F-value[1]
Total	16	23,099.00	-	-
Mean	1	16,192.56	-	-
Treatments	15	6,906.44	-	-
Temperature	1	3,570.07	3,570.07	131
Pressure	1	138.06	138.06	5.06
Time	1	1,040.06	1,040.06	38.2
Mixing	1	495.07	495.07	18.2
Salt:coal	1	1.56	1.56	.0572
KCl:$ZnCl_2$	1	162.56	162.56	5.97
Coal size	1	495.07	495.07	18.2
Temperature X pressure Time X coal size Salt:coal X KCl:$ZnCl_2$	1	39.06	39.06	1.43
Temperature X time Pressure X coal size Mixing X KCl:$ZnCl_2$	1	280.56	280.56	10.3
Temperature X mixing Salt:coal X coal size Time X KCl:$ZnCl_2$	1	76.56	76.56	2.81
Temperature X salt:coal Pressure X KCl:$ZnCl_2$ Mixing X coal size	1	1.56	1.56	.0572
Temperature X KCl:$ZnCl_2$ Pressure X salt:coal Time X mixing	1	495.07	495.07	18.2
Temperature X coal size Pressure X time Mixing X salt:coal	1	60.06	60.06	2.20
Pressure X mixing Time X salt:coal KCl:$ZnCl_2$ X coal size	1	33.06	33.06	1.21
3d-order interactions[2]	1	18.06	18.06	.663

[1] F-value are calculated by dividing the treatment mean square by the error mean square = 27.25 from the one-sixteenth fractional factorial. Any F-value larger than 5.59 indicates the factor is significant at a level of 5 pct.

[2] 3d-order interactions refer to the interdependence of the effects of three factors at a time.

Additional parameter tests were run to determine if pressure lower than 2,000 psig would be suitable and if minus 100 mesh coal would have sufficiently small particle size. Tables 44-46 summarize the factorial design used to investigate these two variables and the analysis of the resulting data. The results were the following:

1. The conversions at 500 and 1,500 psig were significantly lower than those at 2,500 psig.

2. The conversions of minus 140 mesh material were significantly higher than those of minus 100-plus 140-mesh material.

3. At the high pressure (2,500 psig) there was an interaction between pressure and particle size; that is, the conversions of the small particle size were much higher than would be anticipated from observations made at the lower pressures.

TABLE 44. - Parameter levels for additional parameter tests

Factor	Factor levels
Temperature....................° C..	450
Pressure......................psig..	500, 1,500, 2,500
Time..........................min..	15
Mixing (reactor agitation)..........	Rocking
Salt:coal (weight ratio)[1]...........	4:1
$NaCl:ZnCl_2$ (mole ratio).............	1:1
Coal size (U.S. sieve series)........	Minus 100 plus 140, minus 140

[1] 10 grams of coal were charged to the reactor for each run.

TABLE 45. - Conversions in additional parameter tests

Pressure..........psig..	500		1,500		2,500	
Mesh (U.S. series)......	Minus 100 plus 140	Minus 140	Minus 100 plus 140	Minus 140	Minus 100 plus 140	Minus 140
Conversion, wt-pct:[1]						
1st replicate.........	26	24	31	36	44	58
2d replicate..........	19	27	32	33	43	67

[1] Based on maf coal.

TABLE 46. - Analysis of variance for additional parameter tests

Source	Degrees of freedom	Sum of squares	Mean square	F-value
Total	12	18,327.27	-	-
Mean	1	16,096.69	-	-
Sub total	11	2,230.58	-	-
Treatments	5	-	-	-
Pressure	2	1,756.56	878.28	[1] 66.6
Coal size	1	215.90	215.90	[2] 16.4
Pressure X coal size	2	178.98	89.49	[1] 6.78
Error	6	79.14	13.19	-

[1] An F-value larger than 5.14 indicates the factor is significant at a level of 5 pct.

[2] An F-value larger than 5.99 indicates the factor is significant at a level of 5 pct.

Still more tests were needed to investigate coal particle size. Sizes of minus 140 plus 200 mesh and minus 200 mesh were compared (table 47). The test statistic for comparing the means of the conversions of the two particle sizes was calculated to be 0.443. At a 90-pct significance level, the test statistic would have to be larger than 2.92 for the means to be significantly different. Therefore, it was concluded that there was no increase in conversion to be gained by using particles smaller than minus 140 mesh.

TABLE 47. - Parameter levels and conversions for tests comparing minus 140- plus 200-mesh fraction and minus 200-mesh fraction

Temperature ° C..	450
Pressure psig..	3,000
Time min..	15
Mixing (reactor agitation)	Rocking
Salt:coal (weight ratio)[1]	4:1
NaCl:$ZnCl_2$ (mole ratio)	1:1
Coal size (U.S. sieve series)	Minus 140 plus 200 and minus 200
Minus 140 plus 200 coal size (U S. sieve series):	
Conversion in first trial wt-pct..	67.4
Conversion in second trial wt-pct..	74.6
Minus 200 coal size (U.S. sieve series):	
Conversion in first trial wt-pct..	76.5
Conversion in second trial wt-pct..	71.2

[1] 20 grams of coal were charged to the reactor for each run.

Catalyst Life Extension Tests

During the tests used to compare sodium chloride and potassium chloride for reducing viscosity of the molten salt, the same initial salt charge was used for a number of sequential runs. As the number of runs increased it was observed that conversion decreased, phase separation became poorer, and the product changed from a light distillate type of material to a heavy tarlike type of material. Investigators from Consolidation Coal Co.[120] had earlier noted that nitrogen in the coal poisoned the zinc chloride catalyst.

It was observed that the process being used to hydrocrack coal had many conditions similar to processes for removal of nitrogen and sulfur from petroleum, such as hydrogenation at high temperatures and pressures. It was hypothesized that the catalyst poisoning might be the result of the experimental technique used; perhaps the poisonous nitrogen compounds were present in the gaseous phase at reactor operating temperature and were condensed and accumulated in the salt mixture as the reactor cooled.

Equipment modifications were made so that the gaseous phase could be removed at reactor operating temperatures. The gas discharge valve was replaced with a high-temperature valve, and an aluminum jacket was machined to fit all fittings on the reactor head. Electrical cartridge heaters were inserted in the aluminum jacket so that the head and all associated fittings could be maintained at the reactor operating temperature. This was necessary to prevent blockage of the discharge line by condensing nitrogen compounds and to minimize the possibility of chloride stress-corrosion cracking caused by HCl that is formed in the hydrocracking process.

Two series of sequential runs were made in which the gas was removed at ambient temperature after cooling in one series and at the reactor operating temperature immediately after completing the run in the other series. However, the initial runs in the ambient-temperature gas removal series would have given higher conversions than the high-temperature gas removal series because additional conversion would take place as the reactor was cooled. To compensate for this additional conversion, the reaction temperature and time were reduced for the ambient-temperature gas removal runs (table 48).

TABLE 48. - Parameter levels for tests comparing ambient-temperature gas removal and high-temperature gas removal

Factor	Factor levels	
	Ambient temperature	High temperature
Temperature........................° C..	425	450
Pressure..........................psig..	3,000	3,000
Time..............................min..	0	15
Mixing (reactor agitation)............	Rocking	Rocking
Salt:coal (weight ratio)[1].............	4:1	4:1
NaCl:ZnCl$_2$ (mole ratio)...............	1:1	1:1
Coal size (U.S. sieve series).........	Minus 140	Minus 140

[1] 20 grams of new coal were charged to the reactor for each run.

Unfortunately, the conversion data for the two runs were not directly comparable. The adjustment of reaction temperature and time did not exactly compensate for the conversion that took place as the reactor was being cooled

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[120] Third reference cited in footnote 118.

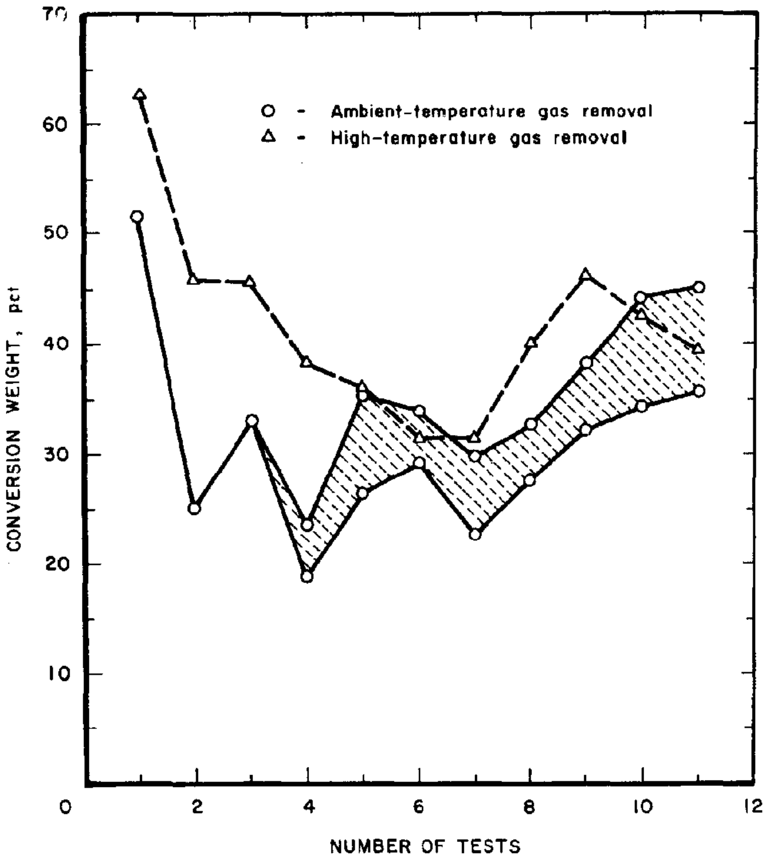

FIGURE 139. - Conversion versus number of tests for high-temperature gas removal and ambient-temperature gas removal.

during the ambient-temperature gas removal series of runs. In addition, there was a problem introduced by trying to simulate a continuous process with a series of batch runs. Undoubtedly, some of the uncoverted coal residue was converted when it was charged again with the salt mixture. Therefore, the conversion should be based on the amount of new coal charged plus part of the coal residue that was charged. Some previous experimental work indicated that for the high-temperature gas removal runs, the conversion should be based on the new coal plus coal residue from the two previous runs. For the ambient-temperature gas removal runs, it was not clear if it would be better to base conversions on new coal and coal residue from the two previous runs or the three previous runs.

In figure 139, the two methods of gas removal are compared relative to conversion. The shaded area represents the uncertainty caused by using coal residue from two or three previous runs to calculate conversions for the ambient-temperature gas removal runs. As conversions leveled off or increased slightly as the number of runs increased for both methods of gas removal, it cannot be concluded that the high-temperature gas removal was significantly better. However, the high-temperature gas removal was better than the ambient-temperature gas removal in two aspects other than conversion: (1) It took longer for the phase separation to worsen, and (2) the product was lighter and less tarlike. The observation comparing phase separation is quite qualitative, but quantitative data is available to support the belief the products were different. The hydrogen:carbon mole ratio of the product from the final high-temperature gas removal run was 1.15:1 and for the ambient-temperature gas removal was 1.03:1; therefore, more hydrogenation occurred using the former method. An analysis made earlier[121] on a different sample of coal indicated

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[121] York, W. J. Reduction of Subbituminous Coal and Lignite Using Carbon Monoxide. Ph.D. Thesis, Chemical Engineering Department, Montana State University, Bozeman, Mont., June 1971, 153 pp.

the hydrogen:carbon ratio of the starting coal was 0.81:1. Additional support for the high-temperature gas removal method was gained by analyzing the salts after they had been extracted with water following the final sequential runs; the nitrogen content of the salt from the ambient-temperature gas removal process was 1.20 wt-pct and that of the high-temperature gas removal process was 0.59 wt-pct. Therefore, more of the nitrogen catalyst-poisoning compounds were removed using high-temperature gas removal than ambient-temperature gas removal.

Conclusions--Part II

Based on the experimental data obtained from these experiments, the following conclusions were reached:

1. At the parameter levels tested--

(a) The parameters have essentially the same effect on conversion of both Colstrip subbituminous and Savage lignite coals.

(b) Pressure, salt:coal weight ratio, and $KCl:ZnCl_2$ mole ratio have no significant effect on conversion.

(c) Some interaction effects which appear likely are temperature-time, pressure-particle size, and time-mixing.

2. Pressures below 2,000 psig significantly reduce conversion.

3. Particle sizes smaller than 140 mesh (U.S. sieve series) are required to eliminate the effect of particle size on conversion.

4. Molten salt mixtures using zinc chloride as a coal hydrocracking catalyst eventually become poisoned with resultant lower conversions, poorer phase separations, and more tarlike products.

5. Gas removal at reactor operating temperatures retards the trend towards poorer phase separation and more tarlike products; the effect on conversion must be studied more thoroughly.

6. More nitrogen compounds are removed using high-temperature gas removal than ambient-temperature gas removal.

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