Coal Liquefaction: Recent Findings in Occupational Safety and Health

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ABSTRACT

Coal liquefaction materials contain potentially hazardous and biologically active substances, many of which have not been characterized as to their composition and/or health effects. Animal studies have shown that certain fractions of coal liquefaction process streams may cause tumors at the site of application. Recent industrial hygiene data show worker exposure at low concentrations of suspected carcinogens. Current control technology assessments of coal liquefaction processes indicate potential exposure of plant maintenance and repair personnel to hazardous materials. This report presents the results of recent NIOSH industrial hygiene studies at two coal liquefaction pilot plants and reviews recent health and process aspects of this technology.

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SUMMARY

Some coal liquefaction materials are potentially hazardous because of similarities to materials in other other related coal processes that have been associated with a high cancer risk. Limited survey data obtained by NIOSH at two coal liquefaction pilot plants have shown that workers were exposed to low concentrations of certain polynuclear aromatic hydrocarbons (PNA's) and aromatic amines, some of which are suspected carcinogens. The degree of risk incurred by such exposures cannot be determined because toxicologic data allowing for the evaluation of effects at low exposure levels are unavailable.

This status report includes the results of recent NIOSH industrial hygiene studies at two coal liquefaction plants and an assessment of the health and process aspects of this technology. The findings of the study are summarized below.

Industrial Hygiene Surveys

- Independent NIOSH and DOE industrial hygiene surveys at the same coal liquefaction pilot plant have shown low airborne concentrations of benzene, toluene, xylene and benzo(a)pyrene. In addition, the NIOSH studies have shown low levels of airborne concentrations of aromatic amines and PNA's.

- Samples were taken for seven aromatic amines. Exposure levels were below current OSHA standards. Samples were also taken for 29 PNA's. Exposure levels do not currently exist for these compounds.

Health Aspects

- Limited information exists on the effects of occupational exposure to coal liquefaction materials because most work in the United States has been with bench-scale units and pilot plants, which have minimum processing capacity (less than 50 tons per day) and minimum operating time (less than 1 week of continuous operation).

- Epidemiologic studies, both retrospective and prospective, are needed to determine to what extent workers at all coal conversion facilities have an increased risk for developing chronic occupational diseases such as cancer and respiratory illness.

- Animal toxicity tests, both acute and chronic studies, should be continued to determine the potential health

effects of identified contaminants. Chronic studies should include determination of carcinogenic, mutagenic, teratogenic, and reproductive effects.

- Additional studies are needed to determine the occupational health risk for chronic low-level exposure to the combination of PNA and aromatic amine emissions that occur in direct and indirect coal liquefaction processes.

Control Technology

- The high temperature and pressures, combined with the slurry handling required in the coal liquefaction plants, are unique in industry. These operating conditions accelerate the erosion/corrosion effects and may result in an increased potential for worker exposure due to leaks and fugitive emissions.

- The control technology derived from pilot plant operations can be used in the design of advanced plants in a qualitative rather than a quantitative manner. Although this technology can be used to minimize the potential for worker exposure, it cannot be used to project the potential degree of exposure of the workers in the advanced facilities.

- Current assessment of pilot plant technology has shown specific areas where improvement is necessary to minimize worker exposure. These areas include materials of construction, piping and pressure vessels, rotating equipment, seals, valves, instrumentation, solid/liquid and design techniques.

- Engineering controls emphasizing containment of contaminants should be designed into plants. This is essential because the type and amount of contaminant emitted can change with the process, coal type, or process parameters.

Incorporation and implementation of the above recommended measures will provide protection for the health and well-being of the American worker, avoid costly downtime and unnecessary costs of retrofit controls. The NIOSH-reported surveys are preliminary efforts to characterize worker exposure in coal liquefaction pilot plants. In order for the exposure data to support the development of standards for future production facilities, surveys need to be updated on a continuing basis and expanded concurrently with the development of coal liquefaction technology.

INTRODUCTION

NIOSH believes that there are potential worker hazards associated with coal liquefaction technology and that these hazards have not been adequately and independently addressed in the ongoing debate over a national energy policy. The reason for this belief is that similarities exist between coal liquefaction materials and materials of high toxic potential from related industries where a high cancer risk has been identified [1-3].

NIOSH Programs

The explicit implementation of a well-designed program to identify, evaluate, and control potential hazards must be an essential part of a national energy policy. NIOSH recognized this need in 1975 and initiated a research program concerned with assuring the health and safety of workers in energy-related industries. This program was part of an interagency effort with the Environmental Protection Agency, the Department of Energy, and the National Institute of Environmental Health Sciences. In the area of coal liquefaction and gasification, the NIOSH program includes the following work which is in progress:

- An occupational hazard assessment for coal liquefaction pilot plants;
- An industrial hygiene characterization of coal gasification plants;
- A medical monitoring protocol for new energy industries;
- An industrial hygiene characterization of direct coal liquefaction plants; and
- A control technology assessment of coal gasification and liquefaction processes, which will include related technology from other industries.

The following reports have been published:

- Recommended Health and Safety Guidelines for Coal Gasification Pilot Plants published in January 1978 (NIOSH publication 78-120) and
- Criteria for a Recommended Standard . . . Occupational Exposures in Coal Gasification Plants published in September 1978 (NIOSH publication 78-191).

Coal Liquefaction Processes

Coal liquefaction is the process by which a dirty carbonaceous solid is transformed into a relatively clean hydrocarbon liquid by the application of heat and the addition of hydrogen. There are basically two technologies used--first generation, or indirect processes; and second generation, or direct processes. Each technology presents its own health problems.

In indirect liquefaction, coal is first gasified by one of several techniques. The gas is then cleaned, and the resulting synthesis gas (carbon monoxide and hydrogen) is catalytically transformed either to paraffinic and olefinic hydrocarbons, to methanol, or to gasoline, depending on the system used.

In direct liquefaction, prepared coal is slurried with a coal-derived oil (donor solvent) and reacted with or without catalyst in the presence or absence of hydrogen. Undissolved solids are separated from liquids; and the products, liquid or solid, are recovered by low-pressure processing. These products may require additional refining.

Scope of Report

This report summarizes survey data from two coal liquefaction plants and assesses health and safety problems from current liquefaction studies. The survey characterized worker exposure to some known and suspected carcinogens which included PNA's and aromatic amines. Exposure to air emissions, such as carbon monoxide and hydrogen sulfide, which may produce acute health effects, and for which Occupational Safety and Health Administration (OSHA) standards exist, are not addressed in this report.

MAGNITUDE OF THE POTENTIAL OCCUPATIONAL SAFETY AND HEALTH PROBLEM

The synthetic fuel industry is expected to grow, and along with this growth the number of workers involved in coal liquefaction processes will increase. Of the 2.5 million barrels of synthetic fuels per day which the President called for in his July 15, 1979 speech, an estimated 1.5 million barrels per day was to be produced from coal. Production of this amount by coal liquefaction processes would require approximately 30 plants each yielding 50,000 barrels per day. These plants may require 400-600 workers for daily operation, or a projected 1990 work force of 12,000-18,000.

The increased demand for coal in synthetic fuel plants will require additional miners, assuming the production per miner remains steady. If coal production is estimated at 5-10 tons per man day for deep-mined coal and 50-100 tons per man day for strip-mined coal, assuming an arbitrary 40-60% split between the two mining operations, respectively, and a conversion of 2 barrels of oil per ton of coal, there will be approximately 47,000 additional miners required by 1990. While these miners will not be exposed to coal conversion health hazards, they will be exposed to mining hazards that will increase occupational diseases, injuries, and deaths. This concomitant increase in the number of coal miners must be considered as part of the total impact on occupational safety and health for coal liquefaction technology.

Samples were obtained at two facilities; one used the donor solvent (DS) process and the other the donor solvent/catalytic hydrogenation (DS/CH) process. Although a limited number of personal and area samples were taken, the data characterize worker exposure in these facilities.

Area and Personal Samples

Area samples taken at the two facilities were qualitatively compared for organic materials with a computer data base of known compounds. PNA's, benzene, toluene, xylene, and aromatic amines were present in the workplace environment.

Worker exposure to airborne contaminants at the two coal liquefaction pilot plants is given in Table 1. The data are representative of 8-hour samples. Several related job categories were pooled to form the more generalized categories of Field Technician and Maintenance. The former category includes operators assigned to the coal preparation, solids separation, solvent recovery, and product solidification areas. The latter category includes millwrights, pipefitters and welders.

Polynuclear Aromatic Hydrocarbons (PNA's)

Exposures were determined for 29 PNA's (Table 1) for which analytically pure standards were available. Total PNA exposure of workers at the two facilities was determined as the sum of the concentration of 29 PNA's. In general, PNA exposure was greater for workers of the DS process.

The higher exposure levels at the DS plant may be attributed to the processing of more coal over a longer period of time relative to the DS/CH plant. This suggests that a greater deposition of process stream material may have occurred in the workplace through leaks, spills, and maintenance activities. Volatilization of these materials may have contributed to the higher worker exposure.

Of the PNA's found in the air samples, 97% were two- and three-ring PNA's; 3% were four- and five-ring PNA's. The two- and three-ring PNA's are not currently known to be carcinogens whereas some of the four- and five-ring PNA's are considered potent carcinogens. PNA's were observed in the process streams of the two facilities. Quantitative analysis of one plant showed the two- and three-ring PNA's ranged from 85 to 90%; the balance were four- and five-ring compounds. The predominance of these low molecular weight PNA's in air samples could be due to their higher volatility relative to the higher molecular weight PNA's.

Benzene, Toluene, and Xylene (BTX)

Exposure levels for field technicians at the DS plant were less than 0.02 ppm for benzene, 0.1 ppm for toluene, and 0.04 ppm for xylene.

Table 1

WORKER EXPOSURE TO AIRBORNE CONTAMINANTS AT TWO COAL LIQUEFACTION PILOT PLANTS

Contaminant	OSHA Health Standards (NIOSH Recommendations)	Donor Solvent/Catalytic Hydrogenation Process						Donor Solvent Process					
		Field Technicians		Maintenance		Lab Technicians		Field Technicians		Maintenance		Lab Technicians	
		No. of Samples	Range (Mean)[d]	No. of Samples	Range (Mean)[d]	No. of Samples	Range (Mean)[d]	No. of Samples	Range (Mean)[d]	No. of Samples	Range (Mean)[d]	No. of Samples	Range (Mean)[d]
Total PNA ($\mu g/m^3$)[a,e]	-	12	0.1-0.3 (0.2)	6	0.02-0.5 (0.2)	3	0.05-0.3 (0.2)	12	3-260 (63)	2	85-130 (108)	3	1-22 (9)
Benzene (ppm)[b]	10 (1)	-	-	-	-	3	<0.04	7	<0.01-0.02	1	- (<0.02)	3	<0.01 -
Toluene (ppm)[b]	200 (100)	-	-	-	-	3	<0.08 -	7	<0.01-0.1 -	1	- (0.08)	3	<0.01 -
Xylene (ppm)[b]	100 (100)	-	-	-	-	3	<0.01 -	7	<0.01-0.04 -	1	- (0.04)	3	<0.01 -
Aniline (ppm)[c]	5	15	<0.1 -	5	<0.1 -	3	<0.1 -	12	<0.01-0.02 -	1	- (0.3)	3	<0.02 -
N,N-Dimethylaniline (ppm)[c]	5	15	<0.08 -	5	<0.08 -	3	<0.07 -	12	<0.01 -	1	- (<0.02)	3	<0.01 -
2,4-Dimethylaniline (ppm)[c]	-	15	<0.05-0.1 -	5	<0.08 -	3	<0.07 -	12	<0.04 -	1	- (<0.02)	3	<0.02 -
p-Nitroaniline (ppm)[c]	1	15	<0.07 -	5	<0.07 -	3	<0.06 -	-	-	-	-	-	-
o-Toluidine (ppm)[c]	5	15	<0.09 -	5	<0.09 -	3	<0.1 -	12	<0.02 -	1	- (<0.02)	3	<0.01 -
o-Anisidine (ppm)[c]	0.1	15	<0.09 -	5	<0.1 -	3	<0.1 -	12	<0.02 -	1	- (<0.03)	3	<0.02 -
p-Anisidine (ppm)[c]	0.1	15	<0.1 -	5	<0.1 -	3	<0.1 -	12	<0.02 -	1	- (<0.03)	3	<0.02 -

[a] Sampling was conducted with a 37 mm cassette containing a silver membrane filter sandwiched between two methyl cellulose gaskets backed by 200 mg of Chromosorb 102 in a 1/4-inch diameter glass tube. Analyses were made by gas chromatograph/mass spectrograph and high-pressure liquid chromatography. Sensitivity of the 29 PNA's were within the 1-6 nanogram range. Anthanthrene was the least sensitive at 44 nanograms.

[b] Sampling was by charcoal tube at 100 ml/min with analyses by gas chromatography using the NIOSH standard method P & CAM 127. 30-40 nanograms can be detected in a 20-liter air sample.

[c] Silica gel tubes were used at a sampling rate of 100 ml/min. Analyses were made by gas chromatography using the NIOSH method P & CAM 168. The level of sensitivity ranged from 0.05 ppm for o-anisidine and 2,4-dimethylaniline to 0.2 ppm for p-anisidine.

[d] Less than values (<) indicate that the compound is present but at concentrations which cannot be accurately quantified. The number given represents the concentration which can be accurately measured based on the sensitivity of the analytical method and the sample size. Where the less than value (<) is given in place of the range, this indicates that all samples were below this concentration.

[e] This total includes 29 PNA's:

Acenaphthalene
Acenaphthene
Acridine
Anthanthrene
Anthracene
Benz(a)anthracene
Benzo(a)fluorene
Benzo(b)fluorene
Benzo(g,h,i)perylene
Benzo(a)pyrene
Benzo(e)pyrene
Carbazole
Chrysene/Triphenylene
Coronene
Dibenz(a,j)acridine
Dibenzanthracene
Dibenz(a,i)carbazole
Dibenzpyrene
Dimethylbenz(a)anthracene
Fluoranthene
Fluorene
Indeno(1,2,3-cd)pyrene
2-Methylnaphthalene
1-Methylnapthalene
Naphthalene
Quinoline
Perylene
Phenanthrene
Pyrene

Maintenance workers at this facility had exposures of a similar order of magnitude to the same contaminants. At the DS/CH facility, area samples indicated that the concentration of these low molecular weight aromatics were of the same order of magnitude or lower. In all cases, measured BTX levels were lower than current OSHA standards.

Aromatic Amines

Worker exposure was determined for seven aromatic amines for which OSHA standards exist. These amines included aniline and its three derivatives, o-toluidine, and o- and p-anisidine (Table 1). Exposure to o-toluidine and to aniline and its derivatives was of the same order of magnitude at the two facilities, with exposure levels less than 0.1 ppm. A comparison with current OSHA standards indicates that the measured levels were lower than the standards.

Particulates

Particulates were sampled at the coal preparation area. Total particulates ranged from 0.7 to 10 mg/m^3. The respirable dust levels ranged from 0.1 to 1.4 mg/m^3 (less than 3% silicon dioxide). The OSHA coal dust standard is 2.4 mg/m^3 (respirable fraction less than 5% silicon dioxide). The Mine Safety and Health Administration (MSHA) standard for respirable coal-mine dust is 2 mg/m^3.

Heat Stress and Radiation

All high-temperature equipment was insulated. Measurements of gross alpha, beta, and gamma radiation at these facilities were determined to be at background levels.

Noise

A noise survey of the facilities was conducted with a Type 2 sound-level meter. Levels were below 90 dBA and most areas were below 85 dBA.

Department of Energy Industrial Hygiene Data

The Department of Energy (DOE) has conducted an extensive industrial hygiene survey of the DS facility for specific compounds [4]. This survey began in 1975. The workplace environment and worker exposure were characterized through the collection of area and personal samples. Differences in sampling and analytical procedures preclude a comparison of the DOE and NIOSH data except on a qualitative level. However, benzo(a)pyrene, benzene, toluene, and xylene were measured in both the DOE and NIOSH surveys.

HEALTH ASPECTS

Limited information exists on the effects of occupational exposure to coal liquefaction materials because most work in the United States has been with bench-scale units and pilot plants, which have minimum processing capacity (less than 50 tons per day) and minimum operating time (less than 1 week of continuous operation). The number of operating pilot plants and the number of workers employed at these plants are small. Due to frequent startup and shutdown, workers engaged in experimental operations generally will have a higher probability of exposure to potentially hazardous materials than workers in a commercial plant.

Coal liquefaction materials contain potentially hazardous and biologically active substances. Many of these materials have not been characterized as to their composition and/or health effects. Although some of the available information has demonstrated a direct association between coal liquefaction materials and carcinogenicity, NIOSH believes that future investigations may find a greater occupational hazard than is currently documented in the literature. This belief arises from the apparent similarities of the potential toxicities of coal liquefaction process materials and those materials known to be produced in coke oven and coal tar processes, which have been associated with a higher cancer risk [1,2]. Other potential adverse health effects associated with constituent chemicals in coal liquefaction products include acute effects from inhalation, severe respiratory irritation, and chemical and thermal burns. Fire and explosion hazards are potentially significant as most coal liquefaction processes operate at high temperature and pressure and contain flammable materials.

Information Related to Carcinogenic Effects

There is one U.S. epidemiologic study performed by an industrial firm at a coal liquefaction plant that operated for 7 years [5]. This study was based on 359 workers, of whom 50 had skin abnormalities. Of these 50 workers, 10 were diagnosed as having skin cancer. NIOSH subsequently reviewed the data from this study and followed up 49 of these workers [6]. Only 5 of the 10 were confirmed as having had skin cancer. These findings did not support the initial hypothesis that workers exposed to toxic materials from the coal liquefaction process, particularly those with evidence of skin abnormalities, were at increased risk of developing systemic cancer, compared with the general population.

The significance of these studies relative to today's direct coal liquefaction processes is questionable because the technology used in the above plant was different from current process technology (operating pressures were 5- to 10-fold greater than the current processes), and the hygiene, sanitation, work practices and controls were not implemented as rigorously as in current pilot plants.

This greater emphasis on occupational health may account for the absence of occupational disease in the newer pilot plants over an equivalent operating period. However, numerous cases of thermal burns and dermatitis have been reported for these plants [7].

The longest exposure period for U.S. workers for whom health effects have been reported is approximately 11 years [8]. In this case no systemic cancers were reported, but basal cell carcinomas reportedly occurred on exposed parts of the body (lip, ear and nose) in 3 of 190 employees. However, the significance of these cases is questionable because of the small number of employees involved and the lack of reference data available at the time. These data are currently being acquired by plant personnel.

Foreign plants have operated for a number of years (from 1927 to 1945 in Germany, and from 1955 to the present in South Africa), but no epidemiologic studies have been undertaken. The Sasol coal gasification/liquefaction plant at Sasolburg, South Africa, is presently the largest commercial process in operation; it has been operating for over 23 years and currently employs approximately 5600 workers [9]. After NIOSH personnel visited the Lurgi gasification and tar handling sections of the plant and discussed health problems with the plant doctor and workers, they reported that skin cancer was not observed among the gasifier and gas clean-up crew, nor among the workers of the coal tar separators. However, it was indicated that the plant record keeping did not include chronic disease such as cancer, and follow-up studies on workers were not undertaken.

Short-term irritant effects of various fractions in coal liquefaction processes have been demonstrated in animal studies. Certain fractions of the process have been identified as moderately to severely irritating to eyes of rabbits [10]. Evidence from animal experiments indicates that local carcinomas may result from some direct coal liquefaction process products when they remain on the skin for extended periods [11-13].

Results of experiments on bacteria and rodents indicate that a number of fractions from coal liquefaction processes are mutagenic in microbial test systems and cause neoplastic changes following cutaneous application and intramuscular injection into rodents [11,14,15].

PROCESS ASPECTS

Coal liquefaction processes are designed to operate as closed systems because of high-temperature and high-pressure requirements. Employee exposures result from fugitive emissions, leaks and spills; the potential for further employee exposure exists during equipment repairs and maintenance. Three-phase systems (solid, liquid, and gas) occur frequently within the direct liquefaction processes and lead to an increase in the severity of maintenance problems. This in turn results in increased potential exposure of the plant maintenance and repair personnel to hazardous materials.

The direct coal liquefaction bench-scale units, process development units, pilot plants, etc., which are operating today may be used to establish a data base of potential hazards and toxic materials for commercial operations. But the prime purpose of these precommercial plants, particularly the pilot and demonstration plants, is to establish operating parameters for the commercial plant such as process stream composition, flow rates, temperature and pressure; materials of construction; and equipment requirements. Establishment of these parameters will help to define the requirements for commercial plant design specifications and to indicate those areas where basic or supporting research is necessary. The control technology data for this phase of commercial plant design, as it relates to diminution of worker hazard, is qualitative rather than quantitative. It can be used to minimize the potential for worker exposure in advanced facilities by reducing potential emissions into the workplace and by reducing equipment maintenance needs. However, it cannot be used to project the degree of worker exposure reduction which will occur. Some control technology improvements which are needed to reduce worker exposure to toxic and hazardous materials include:

- More design and process engineers experienced in coal conversion--There is a limited pool of expertise available today as the combination of pressure, temperature, solids, liquid, and handling found in this industry is not found in any other single industry. Without such expertise, errors in both the design and operation of the equipment may lead to frequent and high levels of worker exposure.

- Materials of construction--Because of the tendency for erosion, corrosion, and cavitation to occur within the processes at the elevated temperatures and pressures required, there is a need for more resistant materials of construction for pipes, elbows, valves, rotating equipment, etc. There is also a need for innovative design in implementing these improvements. With the equipment normally available on the commercial market today, redundancy and frequent maintenance is required.

Greater reliability of both design and materials of construction will minimize the preventive maintenance requirements and the potential for exposure in the workplace.

- Instrumentation--There is a substantial requirement for accurate and dependable instruments to measure density, flow, viscosity, and other process parameters. For example, density measurements at present are estimated, based on process samples (these sampling operations present additional potential for worker exposure). Flows cannot be measured accurately without knowledge of stream densities. Reduction in flow may lead to solids settling and consequent plugging; an increase in flow may increase erosion rate. In either case additional maintenance would be required, which would increase the potential for worker exposure.

- Rotary seals and valves--New materials of construction for purposes such as hard-facing and innovative design techniques are needed to minimize leaks around rotary shafts in stirrers, centrifugal pumps, valves and other process equipment. Mixtures of solids, liquids, and dissolved gases processed at pressures and temperatures not normally encountered in industry make this a difficult problem. Available equipment requires higher maintenance rates than are desirable from either an occupational health or an economic viewpoint. Better designs and materials of construction for this equipment will reduce worker exposure.

- Solid/liquid separation techniques--The combination of temperature and pressure required to separate undissolved coal and mineral matter from the product exceeds the capability of the separation industry today. The use of filters, centrifuges, and hydroclones for this segment of the process results in higher worker exposure both from an operating and a maintenance viewpoint. New physical chemical separation techniques appear promising, and, if successful, will reduce worker exposure in this area significantly.

Although the design and selection of equipment for pilot and commercial plants may differ, the sources of exposure (such as leaks, spills, maintenance and repair) will be similar. The above recommended engineering controls will be an important part of any program designed to protect workers against hazardous materials in coal liquefaction plants.

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