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ADVANCED TECHNIQUES FOR RADON GAS REMOVAL

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prepared for

U. S. Department of the Interior
Bureau of Mines

by

Arthur D. Little, Inc.
Cambridge, Massachusetts 02140

May 1975

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The purpose of this research was to obtain an objective assessment of the feasibility of several unconventional techniques for removing radon-222 gas from mine atmospheres. The probable efficiency, reliability, safety, and cost of several possible methods were investigated. Most of the techniques were feasible, but none was considered practical under current or foreseeable mining operations. It is not likely that any of the methods evaluated in this research will be applied to the removal of radon from mine atmosphere because they are not cost competitive with fresh air ventilation techniques. The value of this research lies in putting these various removal techniques in their proper perspective with current radon control practices.

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FOREWORD

This report was prepared by Arthur D. Little, Inc., Cambridge, Massachusetts, under USBM Contract Number H0230022. The contract was initiated under the Metal and Non-metal Health and Safety Research Program. It was administered under the technical direction of the Denver Mining Research Center with Mr. Robert Droullard acting as the Technical Project Officer. Mr. Monte D. Camp was the contract administrator for the Bureau of Mines.

This report is a summary of the work recently completed as part of this contract during the period May 22, 1973 to April 15, 1975. This report was submitted by the authors on May 8, 1975.

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I SUMMARY AND CONCLUSIONS

The following report summarizes the results of engineering feasibility studies whose purpose was to assess the probable efficiency, reliability, safety and cost of a number of possible methods for removal of radon gas from the ambient air in underground mines. Fourteen candidate techniques were evaluated. Seven of these were based on specific individual principles, and seven were based on combinations of these principles. The individual principles were as follows:

1. Activated charcoal
2. Organic fluids
3. Semipermeable membranes
4. Molecular sieves
4. Cryogenics
6. Chemical reaction
7. Centrifuge

The combinations were:

8. Cryogenics and activated charcoal
9. Cryogenics and molecular sieves
10. Cryogenics and chemical reactions
11. Membrane filters and organic fluids
12. Membrane filters, cryogenics and activated charcoal
13. Membrane filters and chemical reactions
14. Silica and an Organic Fluid

The categorization of techniques as "individual" or "combined" was found to be somewhat artificial and, in some cases, inimical to the development of the most promising embodiments of the various principles into conceptual systems for purification of mine air. For instance, the use of chemical reagents to remove radon from air is virtually infeasible unless the air is pre-cleaned to remove water and all oxidizable impurities that otherwise consume or poison the radon reagent. For this purpose, either air liquefaction or adsorption on molecular sieves would be required, which, together with the chemical reagent, might be considered a "combination" or "hybrid" system. Once this conclusion had been reached, it became apparent that a more useful form for this study to take would be that of an initial assessment of basic techniques, followed by an examination of various possibilities for combining or modifying those techniques so as to improve their effectiveness and/or lower their cost.

Thus, we realized that it would be advantageous to permit some latitude in the interpretation of the meaning of the term "combinations of individual techniques" so as to achieve the most favorable conceptual designs possible. For example, the usual meaning of the term "cryogenics" was expanded to include any kind of refrigeration, and techniques used "in combination" were understood to include those that were used in an accessory way, such as the above-mentioned need to remove moisture from the air prior to chemical radon scrubbing.

Our initial search of the technical literature showed that very little experimental evidence has been developed in regard to any method for removal of radon from gas mixtures. Published reports were found for techniques based on adsorption on activated charcoal, for chemical reactions, and for a combination technique involving absorption in an organic liquid, followed by adsorption on the surface of a solid particulate substance suspended within the liquid phase. An unpublished research report from a commercial firm was obtained which described an inconclusive test of the latter concept in an underground mine. One U.S. patent for a radon removal method based on cryogenic distillation was found to be conceptually incorrect.

In view of this scarcity of documentation, we were compelled to originate conceptual systems, each of whose active principle was one or more of the cited techniques. Since no experimental work was permitted in connection with this project, we found it necessary to estimate some of the physical data (such as for the permeability of certain polymer films to radon). We realize that the choice of materials or the design of the system might be altered to enhance some characteristics at the expense of others. We were not able to explore all of these alternatives in detail, but when substantial improvements could be made by exercising such choices, we have tried to indicate what their effects and consequences would be.

In at least one instance, that of the hybrid system combining semipermeable membranes with organic fluids, the lack of available data to support the choice of the most favorable combination of materials led to a compromise that rests on the assumption that satisfactory materials exist and need only to be identified. The materials used for our hypothetical model in this instance are admittedly somewhat unrealistic choices, in that they lack certain essential properties such as chemical compatibility. Nevertheless, we believe that they are adequately representative of the types of materials that would be used, and that their deficiencies can be corrected without adding significantly to the cost or complexity of the system.

For each technique, a candidate air-cleaning system was designed, based on the conditions specified in the contract. It was agreed at the outset that, for purposes of standardized comparison, we would consider each cleaning system to be associated with a single secondary air supply system such as might be expected to provide 2000-5000 cfm of air to a work place. The criterion for radon removal effectiveness was limited to a tenfold reduction of radon concentration with the provision that the output of cleaned air should be at least 90% of the volume of the input air.

Of the seven individual techniques studied, only one was judged to be infeasible on purely technical grounds, and even in this single instance - centrifugation - the infeasibility was due to insurmountable engineering problems rather than to any theoretical impossibility of the principle. This is not to say, however, that the feasibility of the remaining six techniques could be said to be comparable with that of familiar underground mining practices. Anyone who is familiar with the problems involved in maintaining the present ventilation systems in underground mines, not to mention the seemingly simple but rarely practiced technique of air filtration to remove radon-daughter ions, would probably find the degree of sophistication required for removal of radon itself by any of the proposed methods to be beyond the present capabilities of the mining industry.

The combinations or "hybrid" techniques presented a somewhat different problem in estimating feasibility. In general, those hybrid systems we were able to conceive were ones in which the essential task of separating radon from the mine air was performed by a single technique, and the effectiveness or efficiency of that process was enhanced by the use of one or more accessory techniques in any of several different ways. Our first criterion in evaluating these hybrid methods was that the resulting process would represent some sort of improvement over the basic technique used in the way we had already proposed. On this basis, three of the combinations identified for study in the original list were soon shown to be without benefit and therefore to be unworthy of detailed examination. Those which showed more positive prospects have been treated individually in the following discussions.

Solid adsorbents, especially charcoal, were recognized to benefit from operation below ambient temperature, and three different kinds of candidate systems were studied using this approach, two of which were shown to be technically feasible and advantageous. One of these involved a temperature-cycled adsorption-desorption system which could be substantially smaller when operated at an adsorption temperature of 2°C than in the absence of refrigeration. The other two systems involved cooling to temperatures well below 0°C and therefore required essentially complete removal of atmospheric moisture. One of these systems, designed for operation at ambient pressure, required the use of molecular sieves to dry the air, and was thus judged to have no advantage over a room temperature system. The other system employed compression and reversing heat exchangers, and permitted the use of a non-cycled adsorption bed.

The concept of a solid sorbent being suspended in a liquid solvent was added to the list of topics originally assigned for study, but only because of the existence of two independent reports of favorable results from experiments with this kind of combination. Because the reported results were not well substantiated and were in contradiction to other experimental and theoretical evidence, we concluded that the technical feasibility of this system had not been (and probably could not be) demonstrated. We decided that it would therefore be inappropriate to use these reports as a basis for development of a conceptual model and its associated details and costs.

A system based on the use of a liquid solvent (organic) together with semipermeable membranes was found to have a number of advantages over either of these techniques alone. Accuracy of our estimate of cost and size of this hybrid system was limited by the lack of necessary computer programs to perform the calculations to optimize the design of the system. By making some not-unreasonable assumptions, however, we believe we have arrived at a system design that is fairly representative of what might be achieved in practice, and that permits at least an approximate estimate of the cost for such a hybrid.

The three hybrid systems which were rejected on the basis of initial consideration were:

10. Cryogenics and chemical reactions
12. Membrane filters, cryogenics and activated charcoal, and
13. Membrane filters and chemical reactions

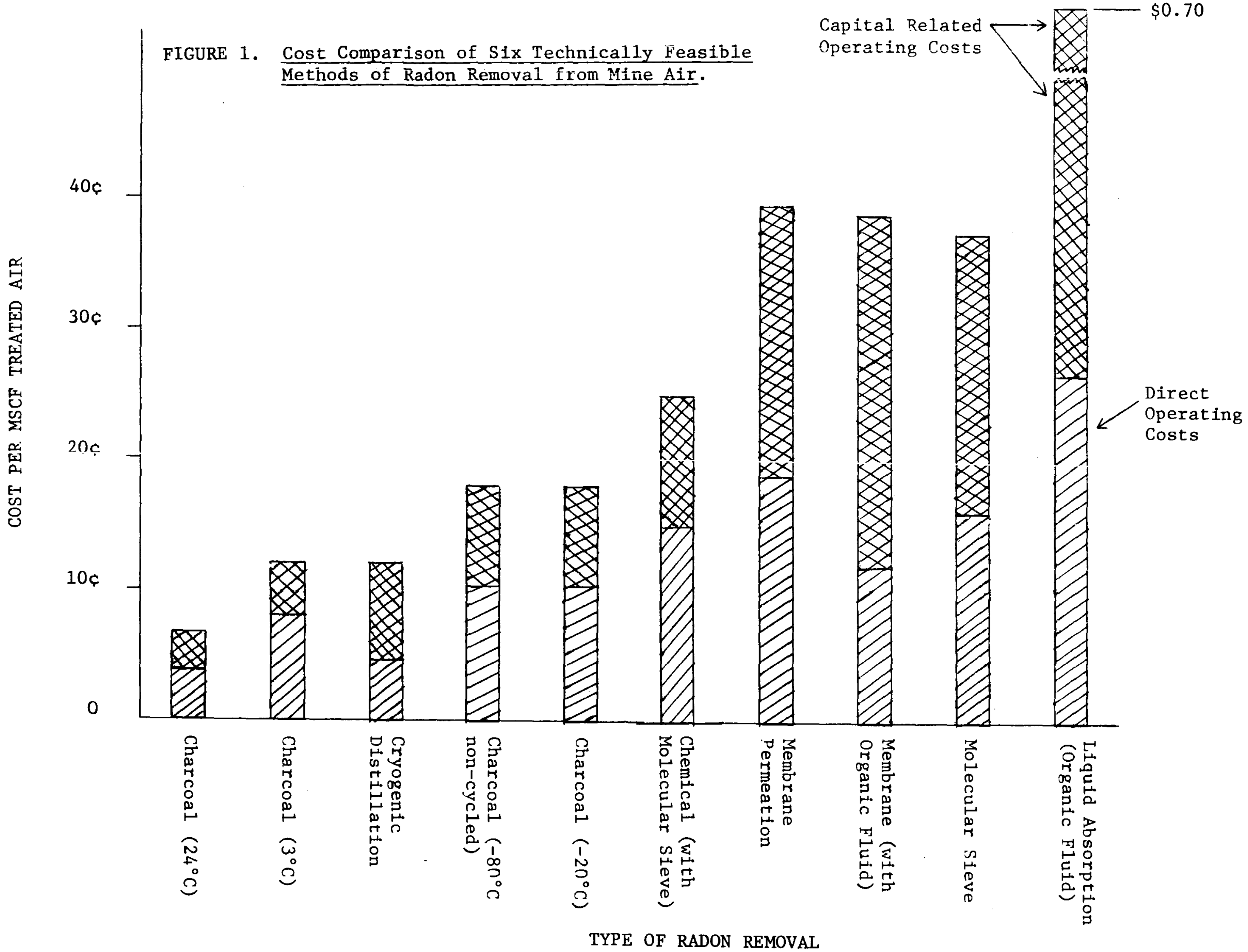
The combination of chemical reactions with either cryogenic cooling or membrane filters (preferably designated "membrane permeation"), was found to have no practical advantage over either technique used alone. The use of a chemical "scrubber" for radon does require assistance from some kind of pre-scrubber for water and oxidizable impurities, but this is best achieved with molecular sieves, after the bulk of atmospheric moisture has been removed by refrigeration to about 3°C. This necessary combination (without which the chemical scrubber system would be infeasible) is considered in Section V-F of this report. Membrane permeation has been found to be more effective at elevated temperatures than at reduced temperatures, thus ruling out any effectiveness of combination with cryogenics. No deficiency of membrane permeation is likely to be amenable to improvement by combination with an adsorption technique such as charcoal. Similarly, neither charcoal adsorption nor cryogenic distillation can be shown to benefit from combination with membrane permeation. Some of these topics have been discussed in greater detail in Section V-K.

Figure 1 below shows the relative costs of implementing and operating the ten technically feasible radon removal techniques. The operating costs are broken down into the direct operating cost and the capital charges relating to the investment required for the facility.

The bases for the operating cost estimates are:

- Labor, Supervision & Overhead at $20,000/Man Year
- Electricity at $0.01/kwh
- Water at $0.05/M gal
- Capital charges at 22% of initial capital investment per year
- Maintenance at 3, 4, or 5% of initial capital investment per year depending on type of process used
- Operations assumed for two shifts per day for 260 days per year
- Unit size is 5,000 SCFM
- Annual volume of air purified is 1.25×10^9 SCF

FIGURE 1. Cost Comparison of Six Technically Feasible Methods of Radon Removal from Mine Air.



Capital cost estimates were developed from estimated equipment costs and the associated installation costs which include all the costs of piping, foundations, electrical, instrumentation, insulation, etc. It has been assumed that a multiplicity of the radon removal units of any one design will be built so that the engineering, overhead, fee and contingency costs are lower than if a single unit were to be built. We estimate that these costs will be 15% of the installed cost.

The maintenance costs have been chosen to reflect the expected requirements depending on the type of air purification system used. For example, maintenance of the charcoal adsorption system has been estimated to require 3% of capital cost per year, since it is a simple low-pressure system utilizing only two relatively small air blowers. On the other hand, the system utilizing membrane permeators operates at a pressure of 20 atm and incorporates three reciprocating compressors. The maintenance charges for this system have been estimated at 5% of capital investment per year to reflect the higher maintenance associated with the operation of reciprocating compressors. The other systems studied are estimated to require maintenance costs of 4% of capital investment per year, since they are more complex than the charcoal absorption system, but do not require the use of reciprocating compressors.

A detailed analysis of the centrifuging system for radon removal was not prepared since all of the operating costs related to that method of radon removal are orders of magnitude greater than the ten systems on which detailed estimates were prepared. If an investment of $20 billion is assumed for that separation system with power requirements of 1,000 megawatts and cooling water requirements of 10 tons/second and assuming continuous annual operation, the treating cost per MCF of gas would be over $1700.

The systems we have designed represent the most economical way in which each technique, whether used alone or in combination, could be made to satisfy reasonable standards of safety, reliability and efficacy. Comparison of the techniques on this basis shows that adsorption on activated charcoal at ambient temperature (24°C) is clearly preferable to all other methods, but that the additional cost of either cooling the charcoal to 3°C or of using the cryogenic distillation technique is small enough to warrant consideration of these methods as alternatives. The remaining techniques are estimated to incur annualized operating costs (including 22% annual capital charges, assuming a five-year amortization period) in excess of $200,000 for a 5000 SCFM output. Inasmuch as these costs approach the present value of the ore which might be produced by a five-man work place, justification of such an expenditure would presumably have to wait upon a substantial increase in the value of uranium ore, or upon the discovery of an enormously rich body, or upon achievement of a major technical breakthrough. An example of such a needed breakthrough would be in the development of low-cost air compressors which could produce the required 20-to-1 compression of 5000 SCFM of air for the membrane separation technique. The present estimated cost for three such compressors, being in excess of one million dollars, is by far the largest cost element in an otherwise relatively inexpensive and generally attractive method.

In summary, ten different techniques were found to be technically feasible and worthy of study in sufficient detail to permit the development of engineering cost estimates for conceptual systems. A question that is of considerable practical importance, but which was not within the scope of the present study, is that of the comparison of cost of radon removal against the cost of diluting the existing radon with additional ventilation air from the surface. Our rough estimates, gathered from previous studies and from our own sources in the mining industry, suggest that the total cost of additional ventilating air for a mine that is less than 1000 ft below ground level is less than one cent per thousand standard cubic feet. None of the cost estimates for advanced techniques that we have developed in the present task are competitive with conventional ventilation methods when compared on this basis. Conceivably, ways might be found to reduce the cost of the least expensive radon removal method (adsorption on charcoal at ambient temperature), and special circumstances might occur in which the effectiveness of conventional ventilation might be inadequate or its cost unusually high. Thus, the removal of radon from mine air appears to be technically feasible, but generally rather expensive, and it will therefore probably not be widely used in the near future.

Table 1 presents in condensed form some of the salient characteristics of each system. Inasmuch as the candidate systems were designed to meet reasonable standards of efficacy, reliability and safety, the characteristics listed under those headings show little variation. A similar constraint was applied to the dimensions of the total system package (except for the centrifuge, whose size could not be made to conform with the dimensions of underground mine working) so that a space no larger than a typical work area would be required to house a 5000 CFM unit. Except for the one system based on Absorption in an Organic Liquid, in which the liquid solvent is to be pumped to the surface for discharge of its radon in a stripping tower, all components were designed to be brought in through the main shaft and haulageways. Thus, the principal dependent variables were the cost of the elements: capitalization and operating costs.

Comparison of all the techniques on an item-by-item basis showed some to be outstanding in certain respects. The following general observations were made:

1. Effectiveness of Radon Removal

Four of the techniques - charcoal adsorption (using alternating beds), molecular sieves, cryogenic distillation and chemical reaction - would be able to remove more than 99% of radon from the air, even if concentrations much higher than the 500 pCi/liter specified limit were encountered. Charcoal adsorption at -80°C, using a single bed with 12.7 day hold-up of radon, makes use of the 3.8 day half-life of ^{222}Rn to allow 90% of the input to decay to solid daughters before the remainder escapes in the output air. The remaining techniques are based on enrichment processes, and are designed to have a minimally acceptable radon removal effectiveness of 90%.

TABLE 1

COMPARISON OF COSTS AND SIZE REQUIREMENTS FOR FEASIBLE METHODS OF RADON REMOVAL

				5000 CFM Unit			
Technique	Efficiency	Reliability	Safety	Approximate Physical Dimensions	Capital Cost	Annualized Operating Cost	Cost per 1000 SCF
Charcoal Adsorption	>99% of Rn removed; >90% of removed Rn returned to waste air	Probable life-time >1 year; little maintenance	Minimal hazards to health & safety. Small hazard of fire in charcoal bed				
24°C				6' x 12' x 16' (1152 cu. ft.)	$ 164,200	$ 84,500	$0.067
2°C				6' x 12' x 20' (1440 cu. ft.)	$ 207,800	$148,300	$0.119
-20°C				5' x 10' x 16' (800 cu. ft.)	$ 320,850	$209,700	$0.168
-80°C (single bed)	>90% of Rn removed & retained permanently in charcoal bed			5' x 10' x 18' (900 cu. ft.)	$ 343,100	$211,500	$0.170
Molecular Sieves	>99% of Rn removed; >90% of removed Rn returned to waste air	Probable lifetime >1 year; little maintenance	Minimal hazards to health & safety. No fire hazard	12' x 20' x 70' (17,000 cu. ft.)	$1,234,000	$468,800	$0.376
Cryogenic Condensation	>99% of Rn removed & retained permanently in liquefier	Probable lifetime >5 years; high reliability, equivalent to large air-conditioning plants	Small hazard of explosion if acetylene is allowed to accumulate. Small hazard of gamma radiation from stored Rn, & of Rn escape if liquid air vessel should rupture	5' x 11' x 20' (1100 cu. ft.)	$ 432,400	$146,800	$0.117
Chemical Reaction (with Molecular Sieve	>99% of Rn removed & retained permanently in scrubber	Probable lifetime of Rn reagent, molecular sieves & activated carbon, about 6 mos. Lifetime of reagent determined by effectiveness of pre-drying & purification steps	Gamma radiation from accumulated Rn daughters requires shielding. Reactivity of Rn reagent with oxidizable matter & with water presents potential fire hazard & toxic gas hazard	3 x (12' x 30' x 16') (17,300 cu. ft.)	$ 613,000	$305,000	$0.245
Organic Fluids	>90%; >95% of removed Rn returned to waste air	Probable lifetime >5 years; little maintenance	Minimal hazards to health & safety with Freon 12; other liquids may have greater hazards (eg. CS_2)	15' x 20' x 40' (12,000 cu. ft.)	$2,283,000	$871,000	$0.696
Semipermeable Membranes	>90%; >99% of removed Rn rejected to waste air which is 10% of input air.	Probable lifetime >5 years; little maintenance	Minimal hazards to health & safety	11' x 16' x 68' (12,000 cu. ft.)	$1,444,000	$502,000	$0.402
Semipermeable Membrane w/ Organic Fluid	>90%; >95% of removed Rn ventilated to waste air	Probable lifetime >5 years, depending on stability of membrane in contact with fluid; little maintenance	Some fire hazard if toluene were used as the fluid; ideal liquid should not be flammable	Absorber unit: 9 x 15' x 20' (2700 cu. ft.) Purging Unit: 30' x 30' x 12' (11,000 cu. ft.)	$1,554,000	$483,300	$0.388
Centrifuge	90% of Rn removed & rejected to waste fraction which is 10% of input air	Extremely low; unlikely that 5000 SCFM unit could ever be made operative	Extreme hazard of violent mechanical failure	1.6 x 10^7 cu. ft.	$18,000,000,000	$2,200,000,000	$1,700.00

2. Disposal of Removed Radon

Three of the techniques - single bed adsorption on charcoal at -80°C, chemical reaction and cryogenic distillation - retain all of the radon until it is transformed by successive radioactive decay into long lived ^{210}Pb or ultimately to stable ^{206}Pb. In three of the techniques - organic liquids, two-bed charcoal adsorption, and molecular sieve adsorption - the radon is desorbed periodically to a waste air stream, or to air at the surface. In the remaining techniques - semipermeable membranes, membranes with organic fluids, and centrifuges - radon is continuously enriched in a small fraction (10%) of the throughput air (or in this case in the recirculating solvent), and this fraction is vented to the surface.

3. Physical Size of the Device

All of the techniques require relatively large pieces of requipment, the smallest being that required by the refrigerated charcoal adsorption system (alternating bed, operated at -20°C) - about 800 cubic feet with a total weight of about ten tons. The cryogenic distillation system is only slightly larger at 1100 cubic feet and 23,600 pounds. The room-temperature charcoal adsorption system requires about 2400 cubic feet of space, but the substitution of molecular sieves for charcoal requires a twelve-fold increase in the volume of the adsorption bed, resulting in total bed volume alone in excess of 16,800 cubic feet, which might be difficult to accommodate underground. The chemical reaction system is also estimated to require about 17,000 cubic feet of space underground, which presents a similarly difficult problem. The multiple-element centrifuge assumes truly horrendous proportions, requiring a minimum volume of 16,000,000 cubic feet, and thus becoming patently infeasible for use in an underground mine.

4. Reliability

Aside from the centrifuge, whose enormous size and complexity undoubtedly exceeds that of any single mechanical device ever constructed, the suggested systems all appear to be technically feasible, although some are far more susceptible to breakdown than others. As we have noted earlier, combinations of techniques were justifiable and even necessary in a few specific limited instances, but in most cases they offered no advantage over the individual techniques. Probably the most straightforward and therefore the most reliable of all is the cryogenic distillation technique, which can be accomplished by the use of a conventional and commercially available air-liquefaction system of modest capacity. Several of the suggested techniques led to embodiments that were relatively complicated and relied on one or more components whose reliability would be essential but whose characteristics cannot be readily predicted under actual operating conditions. Perhaps the most technically complex of the feasible systems is the chemical reaction system, whose reliance on the use of a powerful oxidizing agent (dioxigenyl antimony hexafluoride) which is also highly susceptible to hydrolysis by atmospheric moisture, requires that air be pre-dried and pre-cleaned by extraordinary methods. Failure of any of these pre-cleaning systems would bring about rapid - possibly catastrophic - failure of the reagent beds. In addition, effective continuous operation of the chemical reaction system depends on uniform dispersion of the throughput air across the entire facial area of the bed. Any non-uniformity in porosity of this bed will cause channeling and early deterioration of radon-removal efficiency in the path of least resistance to the throughput air. This hazard applies equally to the charcoal and the molecular sieve adsorption beds.

All the systems have been designed to cope with the expected variations in atmospheric conditions (pressure, temperature, and air composition). None of them, however, with the possible exception of the semipermeable membrane, is designed to withstand instantaneous overpressures of 60 psia. This extreme conditions appears likely to occur only in the immediate vicinity of blasting operations, while radon-removal systems are expected to be installed near main haulageways and thus to be at some distance from the location of intense pressure waves. Providing such unusual additional protection to the equipment would generally multiply the capital cost by substantial factors.

Provided that failure of components in the various systems does not exceed normal expectations, the amount of service and maintenance is likely to be nominal for those systems that have few interdependent components and few moving mechanical parts. Solid adsorption beds, whether molecular sieves or charcoal, will probably require replacement on an annual basis to avoid irreversible contamination and "poisoning." The chemical reagent beds are likely to require replacement more often than once a year, perhaps every six months. The radon separation media in the remaining systems will probably continue to be effective for the active useful life of the mechanical components. All the systems require pre-filtration of incoming air to remove particulates. These filters will require frequent replacement in mines whose air is laden with moisture, diesel exhaust and dust.

5. Safety

The systems having the lowest inherent safety hazard are the ones using semipermeable membranes, molecular sieves and cryogenic distillation. None of these introduces any significant pollution to the air, or significantly alters the oxygen/nitrogen balance. The large reciprocating compressors needed for the semipermeable membrane system will produce some noise and vibration, but not at levels that would prevent their use underground. Both the chemical reaction and the organic liquid systems can emit some noxious or toxic substances to the air, but only if accessory purification systems fail. Unless re-humidification procedures are used, air emitted from the chemical reaction, the molecular sieve, the cryogenic still and the charcoal adsorption systems will be uncomfortably dry. Provision for removal of accumulated acetylene is normal in the design of air liquefaction plants. Failure to make such a provision can cause severe explosions. A potential fire hazard exists wherever charcoal traps are in contact with air, especially at elevated temperatures. This situation occurs in accessory components of the organic fluid and chemical reaction systems as well as in the charcoal adsorption system itself.

The hazard of ionizing radiation from accumulated radon decay products is well known and has often been cited as the principal argument against the use of any form of "radon trap" in the mine. It can easily be demonstrated that concentration of 500 pCi of radon-222 per liter of air from a 5000 SCFM air stream (2360 l/s) eventually results in the development of about 0.5 Ci of penetrating gamma radiation from each of the short-lived decay products ^{214}Pb and ^{214}Bi. Unshielded, this would deliver about 4.5 rads of gamma radiation per hour at a distance of one foot, and would have to be surrounded by about 2 feet of concrete in order to reduce the dose at 3 feet to a tolerable level of 100 mr/week for a 40-hour week.

Fortunately this problem is associated with only three of the techniques under consideration: cryogenic distillation, chemical reaction, and charcoal adsorption on a single bed at -80°C. In all other systems the radon is either discharged promptly to waste air (residence time less than a minute) or it is cyclically discharged to waste air at intervals of one hour or less. In either of these cases the accumulation of daughter products is calculated to be orders of magnitude lower than would be attained in the "infinite hold-up" situation, and the resulting radiation hazard is therefore negligible. The accumulated radioactive decay products in the cryogenic system are likely to be dispersed around the walls of the liquid air container and in the 100 liters of liquefied air. This dispersion of the radiation source, plus the self-shielding achieved by the liquid air and its containment vessel, will substantially attenuate the radiation flux at the surface, permitting considerable reduction or elimination of the additional shielding. The possibility of accidental rupture of the liquid air vessel would result in spillage of all the contained radon, plus whatever daughter products are not attached solidly to the container wall (for the conditions cited, $\sim$0.5 Ci of each nuclide).

In the case of the chemical reaction system and the refrigerated single charcoal bed, the attached radon daughter products would be dispersed over a much larger area and would be subject to self-shielding by the mass of the reagent (or adsorbent) itself (approximately one ton of solids in either case). Further shielding is therefore not likely to be needed in either of these systems; however, it should be noted that the radiation hazard calculation has been based in part on the assumption that the average concentration of ^{222}Rn in the input air does not exceed 500 pCi/liter from day to day. This may not be true in actual circumstances, since ^{222}Rn concentrations well in excess of this value are known to occur in the air of operating uranium mines. If air contaminated with radon at concentrations much higher than 500 pCi/liter is processed continuously for several days by either the cryogenic or the chemical reagent system, the radiation hazard from accumulated decay products may well become troublesome.

It should be remembered, of course, that every one of the candidate systems requires the use of an impact filter to remove particulates from the incoming air. A large fraction of pre-existing radon daughters will be retained on these filters, which, after the first few hours of use, will exhibit a significant flux of gamma radiation, whose intensity will depend on the daughter product concentration in the input air from hour to hour. Radiation levels from this source are not likely to be high enough to require special shielding.

II. BACKGROUND

Small amounts of radon-222, the first radioactive decay-product of radium-226, are present in sandy and granitic soils throughout the world. Concentrations as low as one part in 10^{16} are said to be high enough to present hazards to human health and are found as air-pollutants in certain underground mines, especially in uranium mines. Because radon is a gaseous element at atmospheric pressure and temperature (b.p. 211°K at one atmosphere), and because of its chemical inertness, individual atoms of radon-222 tend to migrate rather freely through minerals in which they are formed. Those that are formed close to exposed surfaces of the ore, or at somewhat greater depths in the porous sandstone deposits characteristic of many domestic uranium deposits, may escape into the open air at the ore surface if they do not first undergo a further radioactive decay to become atoms of polonium-218, and thus acquire more sedentary habits. In sandstone formations in New Mexico, Colorado, Utah and other parts of the southwestern U.S.A., the high pore-volume of the rock ($\sim$20% in many areas) contributes to a correspondingly high radon emanation rate on the order of 10^{-14}Ci/cm^2/sec from surfaces of stopes, drifts and haulageways of underground mines. Although the decay of radon-222 (half-life = 3.8 days) produces a 5.5 MeV alpha-particle, the principle health hazard associated with it is due to the alpha-radiation resulting from the decay of two of its short-lived daughter-products, polonium-218 and polonium-214. Most of the detectable gamma-radiation in radium and uranium ores is emitted by the other two short-lived daughter-products: bismuth-214 and lead-214. While the daughter-product atoms are much more susceptible to removal from mine air by chemical and physical means than is radon itself (inasmuch as they are highly reactive atoms of elements that are metallic solids at normal temperature and pressure) the possibility of preventing their occurrence by eliminating the parent radon from the air is a logically appealing strategy. (If a method could be devised for staunching the flow of radon from the mine-walls and the broken ore, then the ultimate objective would be achieved. Trapping the radon after it has escaped into the mine air is clearly an objective of secondary value.)

Any safe and practical technique that can result in the reduction of this insidious hazard to the health of underground miners, and that can substantially reduce the need for massive primary ventilation of the mines would be of considerable value to the industry and to the society.

III PROGRAM OBJECTIVE

The objective of the Bureau of Mines in undertaking this program was to obtain an objective assessment of the feasibility of a number of unconventional techniques for removal of radon-222 gas from mine atmospheres. Inasmuch as these advanced techniques have never been reduced to practice in actual operating mines, but have generally been limited to theoretical propositions and laboratory-scale demonstrations, it was desired to assess their feasibility in terms of efficiency and reliability for radon decontamination of mine air, as well as the anticipated safety and cost of specific embodiments and practical full-scale systems. Quantitative and qualitative information on each suggested technique was to be developed, such that a number of conceptual designs could be developed which could be compared on a common basis.

It was the intent of the Bureau of Mines that this study should be limited to projections of feasibility based on available information from any and all sources, but that no actual tests or experiments be conducted to obtain data or to verify our estimates. This final report, which is the principal product of the study, is intended to show what the practical consequences would be of implementing each of the suggested techniques, either alone or in whatever combination might appear to be most advantageous.

IV SCOPE OF WORK AND METHOD OF APPROACH

Assessment of the feasibility of removing radon from mine air was to involve a thorough search of sources of published and unpublished data to discover any facts bearing on the possible use of each of seven advanced techniques alone and in combination, for that purpose. By using these accepted principles, facts, and data, and without resorting to laboratory experiments or field tests, hypothetical air cleaning systems were to be designed which would be capable of achieving at least a ten-fold reduction of radon concentration in a typical flow of mine ventilation air sufficient to supply an ordinary stope where from 2 to 5 men might work.

The following standard conditions were specified in order to provide a realistic basis for intercomparison of the suggested techniques:

A. Radon concentration - 30 to 500 pCi/liter

B. Air volume from 2,000 to 5,000 cubic feet per minute (cfm)

C. General atmosphere

1. Relative humidity from 70 to 100 percent
2. Air temperature 50 to 100 degrees Fahrenheit
3. Air velocity 10 to 3,000 feet per minute
4. Aerosol particles 3,000 to 100,000 per cubic centimeter
   Organic fraction 0.1 to 0.8
5. Carbon monoxide (CO) to 500 parts per million (ppm)
   Carbon dioxide (CO_2) to 1 percent
   Nitrogen oxides (NO_x) to 1,000 ppm
   Sulfur dioxide (SO_2) to 3 ppm
6. Air pressure - 10 to 15 pounds per square inch absolute (psia)
   Instantaneous overpressures up to 60 psia

D. Power sources

1. Primary electric power - Commercial AC 60 Hz
2. Compressed air

Candidate systems designed to meet the stated requirements were then to be evaluated for:

A. Efficiency and Reliability of Radon Removal
B. Safety, and
C. Estimated Cost

Our approach to this assignment was as follows. From members of our professional staff and consultants we assembled a working team of physical scientists and engineers, each of whom was singularly qualified to deal with one or another of the assigned techniques. With the cooperation of a literature research specialist, this team reviewed abstracts of technical literature published over the past 30 years. The bibliography attached as Appendix I to this report represents the results of that review. Working in part from consideration of basic physical principles (since very little useful literature exists on the separation of radon from air), and in part from their personal ingenuity and judgment, the team members then set about to devise systems for removal of radon from mine air. Each specialist was assigned one of the proposed techniques for which to develop the most technically effective and promising air purification system to fit the stated specifications. Whenever alternative schemes were proposed, or alternative materials were available, the one offering the highest cost-effectiveness was chosen, provided that reasonable standards of safety, reliability and adaptability to underground operation could be met. Wherever the choice was not obvious, all reasonable alternatives were described.

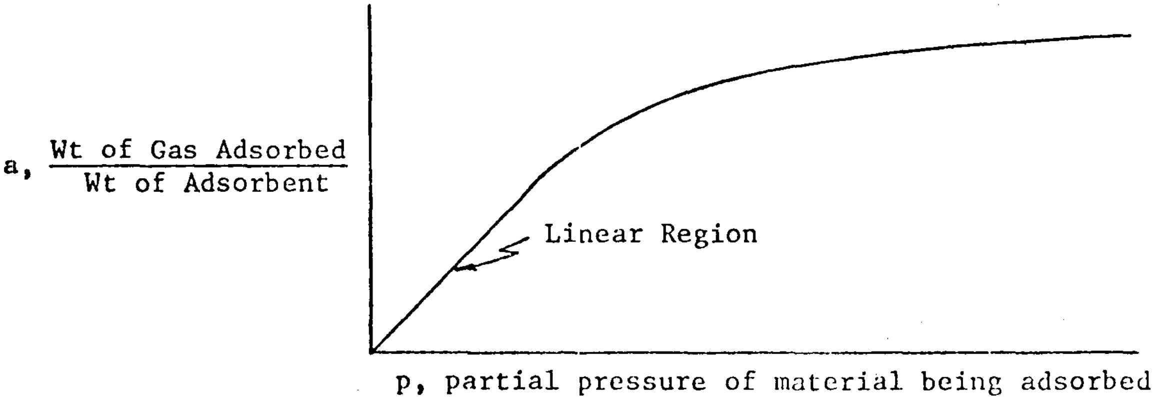
When the determination of technological feasibility had been established, and a conceptual plan produced for a system for purification of 5000 CFM of air, this plan was turned over to a plant design engineer for development of cost estimates for construction and operation of each system. This process inevitably necessitated a second and sometimes a third revision of the conceptual design, since the cost estimates were generally based on the use of system components that are commercially available, and this constraint usually required some alteration of the original system concept. No provision was made in the cost estimates for research and development efforts, although some degree of development work would surely be required to bring any of the techniques to practical realization for underground use.

V FEASIBILITY OF INDIVIDUAL TECHNIQUES

A. ADSORPTION ON ACTIVATED CHARCOAL

1. Description of a Preferred System

The amount of a material which can be held on the surface and in the pores of a solid adsorbent, in equilibrium with a given gas phase at the same temperature, is related to the concentration of that material in the gas phase. The relation is commonly expressed as an adsorption isotherm, i.e., a plot of amount held versus concentration (partial pressure) in the gas phase, all at some constant temperature. For physical (i.e., reversible, non-chemical) adsorption, such isotherms are generally characterized by being linear at low partial pressure, and flat at high partial pressure:



At high partial pressures, the value of a approaches that corresponding to a complete filling of all adsorbent space with an adsorbed phase similar to a liquid.

In the case of radon, however, we are dealing with adsorption from an extremely dilute gas stream; at, say, 100 picocuries/liter, the partial pressure of radon gas is only 7×10^{-17} atmosphere. At such low partial pressures, the adsorption is linear (indeed, the experiments of J. Thomas[1] of the AEC's Health and Safety Laboratory confirm such linearity for radon on charcoal at room temperature up to at least 38,000 pCi/l, well above the levels we are dealing with).

When adsorption is linear, a vast simplification to trap design becomes possible. Consider a trap through which a "carrier" gas (air) containing a trace impurity passes. The impurity is adsorbed initially near the inlet of the trap, then later at successively greater distances from the inlet, until finally the impurity "breaks through" the trap.

At this point, if kinetics are rapid, essentially the entire trap is saturated with respect to the impurity at its inlet partial pressure. If the trap was large enough to pass V liters of carrier gas before this breakthrough occurred, it will turn out to pass the same volume (V liters) up to the breakthrough point even if we raise or lower the inlet impurity partial pressure, so long as we remain in the region of isotherm linearity. Thus, for dilute (linear) gases, we can characterize the adsorption performance of a given adsorbent in terms of the volume of carrier gas it can handle before breakthrough. This volume is called dynamic adsorption coefficient, k, and commonly has the units cm^3(STP) of carrier gas/gram of adsorbent. It can be used for trap design if the adsorption front (i.e., the boundary between saturated and fresh adsorbent) is fairly sharp as it passes through the trap.

The data of Kapitanov[2], of Gubeli[3], and of Fusamura[4] for charcoal are plotted as solid lines in Figure 2. The data from J. Thomas's experiments with charcoal gas mask canisters[1] can be used to develop a room-temperature value for dynamic adsorption coefficient, shown as a single point.

The agreement among the various investigators is satisfactory considering the differences in charcoals, as shown in the legend. All agree, in that a rather striking dependency on temperature is evidenced; thus, adsorption at -50°C is about 40 times greater than at room temperature, while at 120°C it is close to 20 times less than at room temperature. The same sort of temperature dependency has been noted in the case of the other heavy noble gases; see, for example Förster[5] for an analogous plot of k vs. t for krypton and xenon from air on charcoal, showing similar slopes.

Kapitanov indicates that moisture affects radon adsorption on charcoal; he states that saturating the inlet gas with water vapor lowered k by about 50%.

The above was used as our basis for the design of a preferred system of charcoal for use in mines, to allow cost estimation. As a first step in this process, we calculated the size of equipment required to operate at near-ambient conditions.

The beds, then, are sized for operation at 5000 SCFM, saturated with H_2O, containing 500 pCi/liter of Rn. We use the plot in Figure 2 together with Kapitanov's warning about moisture, to arrive at a dynamic adsorption coefficient of 4000 cm^3 (STP) air/g carbon at 75°F (24°C). This requires 4660 lb of carbon for 1 hour's worth of flow. This can be accommodated in a vessel 6 feet in diameter and about 7.5 feet long, allowing the flow to enter the center of the vessel and split through two carbon beds each 2.75 feet thick. Each vessel would be equipped with a pair of 150 KW heaters for the regeneration step. By heating the bed to at least 230°F (110°C) in a stream of untreated air, the data on dynamic adsorption presented above show that the bed's radon content should be reduced by at least a factor of 15 - making it quite possible to achieve an overall decontamination factor of at least 10, as required.

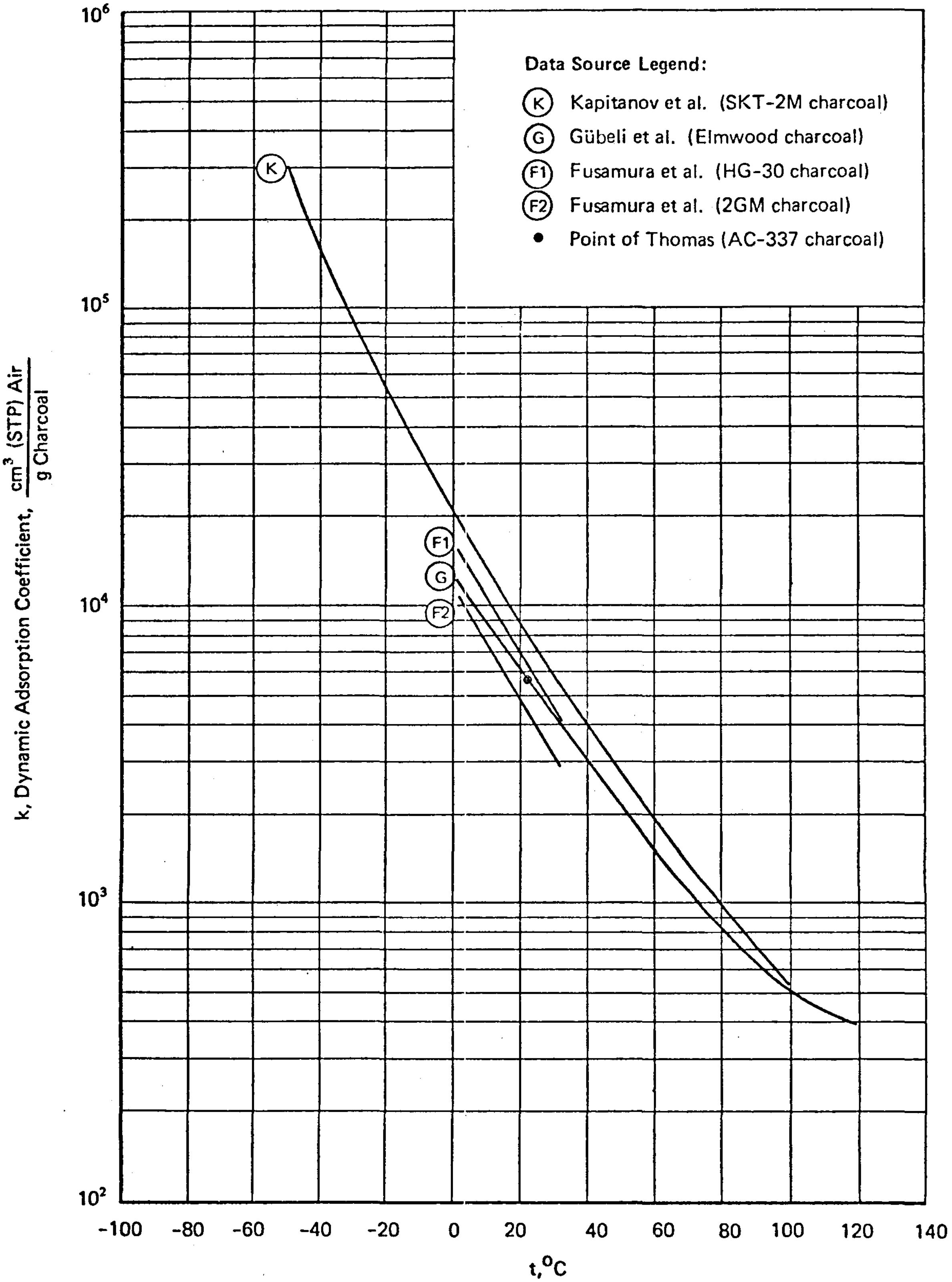


FIGURE 2 DYNAMIC ADSORPTION COEFFICIENT – RADON (FROM AIR) ON CHARCOAL

TABLE 2

EQUIPMENT SIZES AND UTILITY REQUIREMENTS

RADON ADSORPTION AT NEAR-AMBIENT CONDITIONS

Equipment Needs

2 Filters, 5000 CFM each

2 Blowers, 5000 CFM x 4.4 psi (75 HP) each

2 Pre-coolers, 2700 ft^2 each

2 Beds, 6 ft dia x 7.5' long, each containing 4700 lb carbon and each with a pair of 150-KW heaters

2 After-coolers, 330 ft^2 each

1 Blower (regeneration) 1000 cfm x 2" H_2O (10 H.P.)

1 Filter (1000 cfm)

Utility Requirements

Electricity	-	450 KW
Water (20°F rise)	-	140 gal/min

The equipment sizes and utility needs would be about as shown in Table 2.

Such an array could conceivably be placed underground, more or less as shown in Figure 3, but it would not be portable. The accumulated radioactivity and the consequent shielding requirements have also been considered.

2. Efficiency and Reliability

a. Total Concentration of Radon Retained

We have allowed for cooling the air to 75°F if it is supplied at any higher temperature; therefore the temperature range of interest is in this case not 50°F to 100°F, but rather 50°F to 75°F. Radon retention will be essentially quantitative (>99%) as long as we remain below the "breakthrough" volume. Breakthrough would be expected to occur after passage of 4 liters of gas/gram of activated charcoal (at 75°F) to 8 liters of gas/gram of charcoal (at 50°F). These figures allow for 100% relative humidity; reduction to 70% relative humidity could increase the above breakthrough volumes by 30%. These breakthrough volumes are assumed unaffected by changes in radon concentration, air flow rate or velocity, aerosol particles, or CO, CO_2, NO_x and SO_2 contents in the ranges given.

With the above in mind, the concentration of Rn retained on the charcoal bed at saturation would be as shown in Table 3.

b. Weight, Volume, and Width of Device

For the "worst" case - 5000 cfm of 100°F, 100% RH air at 15 psia - we estimate a requirement of two charcoal beds, each containing 2.3 tons of activated charcoal and each 6 feet in diameter by 7.5 feet in length. The total dimensions of the entire system, including blowers, heat exchangers and filters would be about 6' x 16' x 12'. The pressure drop through an operating (split) charcoal bed would be about 10 inches of water.

c. Pressure Drop

TABLE 3

Radon Retention on Charcoal

Feed Concentration	Feed Temperature		Rn Retention at Saturation*	
	°F	°C	70% Relative Humidity	100% Relative Humidity
30 pCi/1	50	10	310. pCi/g charcoal	240. pCi/g charcoal
	75	24	155.	120.
500	50	10	5200.	4000.
	75	24	2600.	2000.

*The above figures are for 15 psia. If the air pressure were 10 psia, then the retention concentrations would be 10/15 as large as the figures in the table.

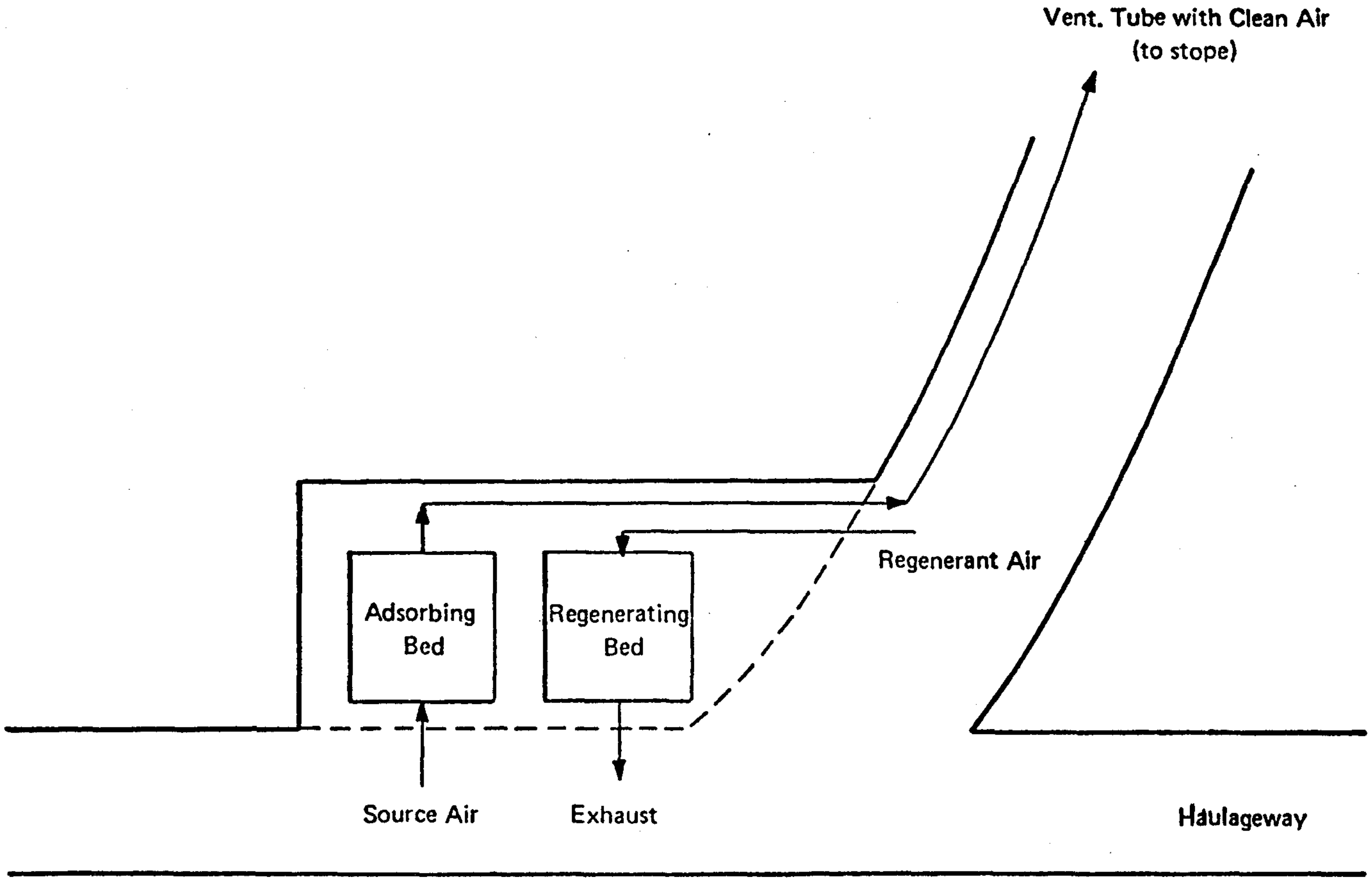


FIGURE 3 LOCATION OF RADON ADSORBER UNIT

d. Residence Time of Radon and Air

The residence time ("holdup") of Rn in the charcoal beds is designed to be 1 hour; that of air is about 2 seconds.

e. Rate of Deterioration

The charcoal should last through thousands of cycles without serious decrease in performance; the temperature will never be high enough to cause appreciable loss by oxidation.

f. Possible Recycling Processes

If it is desired to dispose of the charcoal in a bed, it could be buried safely.

3. Safety

a. Possible Oxygen Removal Rates

None

b. Noxious or Toxic Byproducts

None

c. Maximum Expected Noise Levels

The 75 HP blowers needed should lead to no noise problem.

d. Possible Storage, Use and Maintenance Hazards

Two principal hazards are foreseen in the use of charcoal adsorption for radon removal: fire and gamma radiation. The hazard of fire in charcoal air-purification beds is well known and its abatement has been the subject of intensive effort on the part of government and industry for many years. As a result, charcoal filter-bed design has been improved with particular attention to reducing fire hazard, so that only in extraordinary circumstances can ignition be produced. Fires and sparks cannot be allowed to reach the charcoal, especially when it is heated during purging. If this provision is realized, the fire hazard will be virtually absent.

The second cause for concern is that of accumulation of gamma-emitting radon daughter products in the charcoal bed. It is often pointed out that an irreversible radon accumulator will, in the course of a few day's operation, attain a level of short-lived radon daughter activity that can present a significant radiation hazard. Both RaB (^{214}Pb) and RaC (^{214}Bi) emit gamma radiation which can be hazardous if present in sufficient intensity. It can be shown that a 5000 CFM air stream containing 500 pCi of ^{222}Rn per liter will produce over 0.5 Ci of each daughter product in equilibrium with the total accumulated radon. By using a periodic discharge cycle in the radon trap, however, as is suggested for the charcoal adsorption technique (and for the molecular sieve system described in Section V-D below) the residence time of radon on the bed is too short to permit such a build-up of daughter-product activity. To produce a satisfactory estimate of the actual level of RaB and RaC on the charcoal bed, a series of calculations were made, based on a modification of R.D. Evans' procedure, as given in "Engineer's Guide to the Elementary Behavior of Radon Daughters" and in G. L. Schroeder and R. D. Evans, "Some Basic Concepts in Uranium Mine Ventilation".

The following conditions were assumed:

- Air volume processed: 5000 SCFM
- ^{222}Rn concentration: 500 pCi/1
- Initial Daughter Concentration: nil
- Efficiency of ^{222}Rn retention: 100%
- Efficiency of retention of daughter products: 100%
- Operating Cycle: One hour of adsorption, followed by instantaneous discharge of adsorbed ^{222}Rn, followed by one hour wait. (Actually, discharge will not be instantaneous, but at least 90% of the initially adsorbed radon will be discharged in each cycle.)

The results of these calculations are plotted in Figure 4, with the separate daughter activities (RaA ≡ ^{218}Po, RaB and RaC) shown superimposed on that of ^{222}Rn over four successive cycles. Note that after four hours the cycles of activity (i.e., growth and decay caused by adsorbing and purging the Rn) become regular. Operating the filter in this cyclic method for further time yields no greater increases in activity of these daughters (the long lived daughters will of course gradually grow in). The leveling off of the activity cycle after four hours is predictable from the consideration of the decay of Rn daughters following the removal of the Rn. Even assuming equilibrium of Rn and its short lived daughters (RaA,B,C), three hours after the Rn is removed, no RaA activity remains, about 1% of the RaB activity remains, and about 3½% of the RaC activity remains. Hence, by the time that we start the fifth hour of operation (i.e., three hours after we purged the first hour's Rn accumulation) essentially all of the daughter products that were grown on the filter by the Rn adsorbed in the first hour have decayed. Operating the filter now during the fifth hour we essentially repeat the activity cycle that we observed during the third hour. During later cycles this approximation becomes almost exactly true. Note that for each cycle of purging, the maximum activity of RaB, and RaC occurs after the Rn is purged. This continued growth is the result of decay of the daughters preceding RaB and RaC, respectively.

The graph depicts an instance which is intermediate between the examples cited in Table 3 above. Radon accumulation in each cycle is about

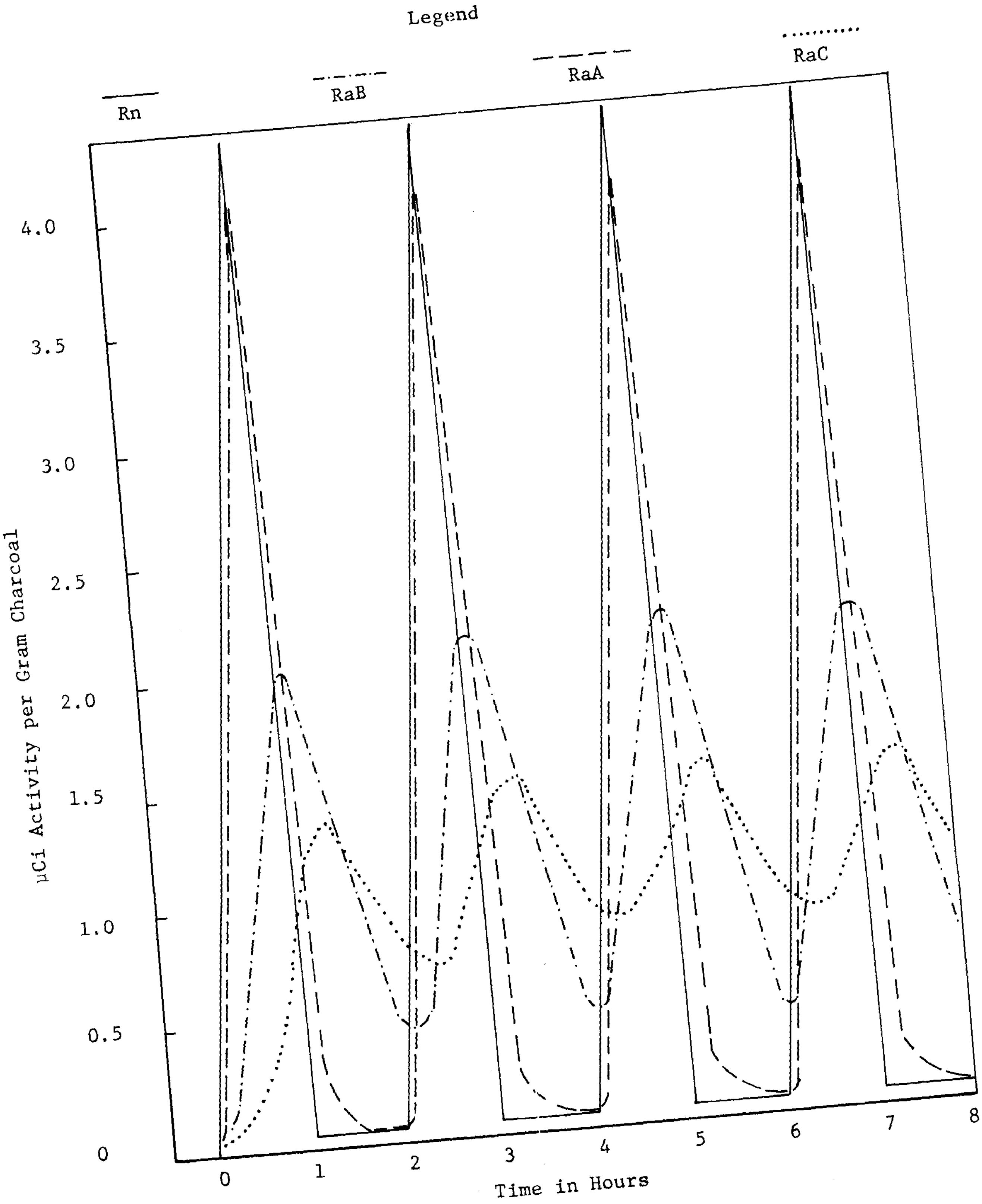


FIGURE 4. Growth and Decay of Daughter Product Activity in Cyclically Purged Radon Trap.

4.4 nCi/g of charcoal. In a bed containing 2.3 tons of charcoal (2.1×10^6g) the total radon retained at the end of the adsorption portion of the cycle will be $(2.1 \times 10^6) \times (4.4 \times 10^{-9}) = 9.2$ mCi. Nearly all of this is discharged in the purging operation, leaving average RaB and RaC concentrations of about 2.6 mCi and 2.4 mCi respectively, or a total of 5 mCi of gamma emitters. This is less than 1% of the amount expected from a non-purging collector, and, if concentrated in a point and not shielded, would deliver about 40 mr/hr at a distance of one foot (30 cm). Inasmuch as the 2.3 tons of charcoal in a steel container provides not only a generous amount of self-shielding, but also places an average distance of about a meter between the source and the exterior wall of the charcoal bed, no further shielding would be required to bring the surface radiation level down to a tolerable 2.5 mr/hr for a 40-hour/week continuous exposure. It should be recalled however that this calculation is based on the assumption of a 500 pCi/liter Rn concentration in the input air. If the Rn concentration were to exceed this value by several times, the radiation hazard might conceivably become noticeable.

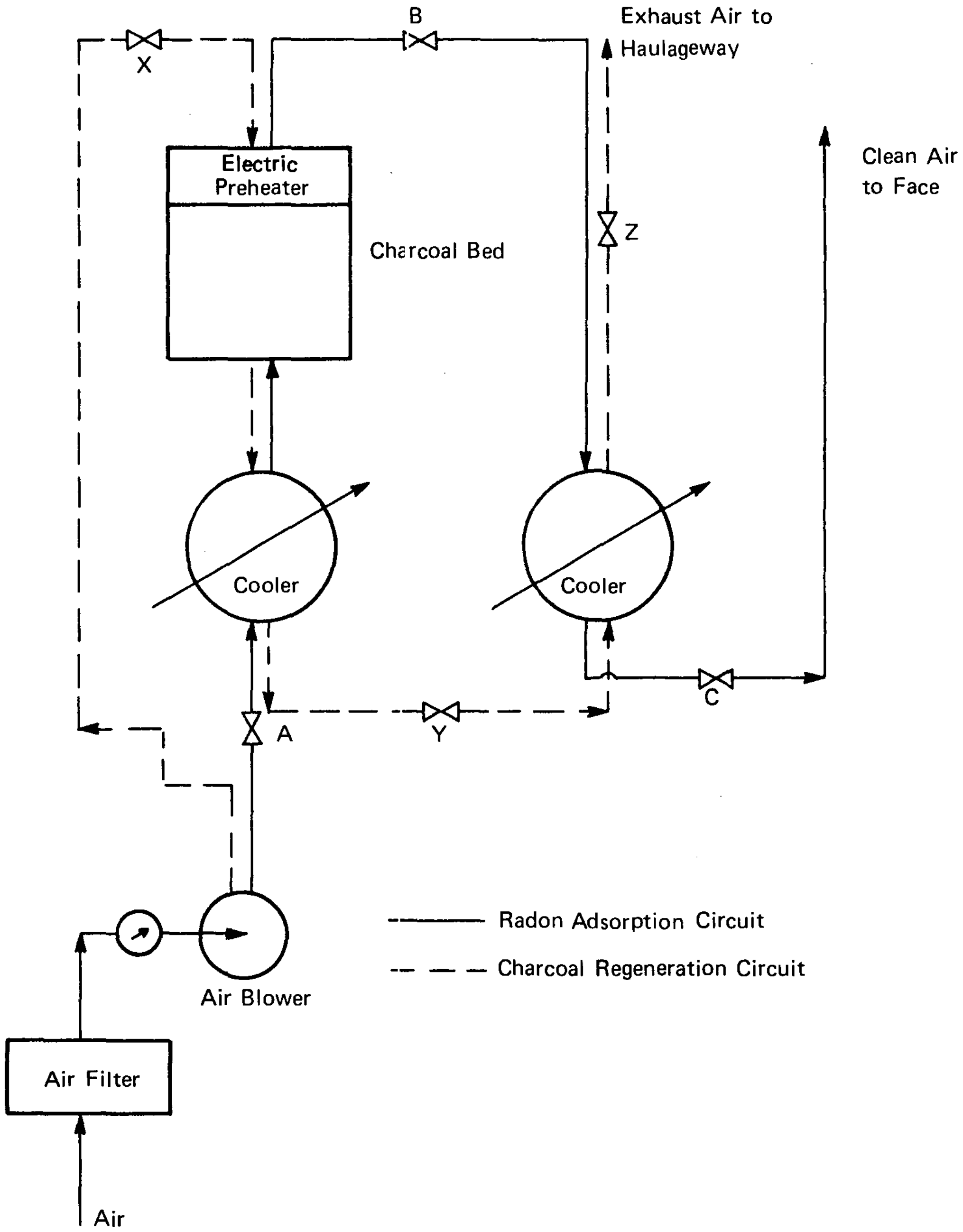
e. Remedial Measures

None would seem needed.

4. Cost

On the basis of the system described above, the following capital and operating costs are estimated. The system consists of two identical and interchangeable segments, one of which is sketched in Figure 5. When a segment is used as a radon adsorber, filtered air is pre-cooled to enhance adsorption efficiency and prior to introduction into the charcoal bed. Following adsorption, the resultant "clean" air is again cooled to render it suitable for inhalation at the working face. For optimum charcoal regeneration, ambient air is preheated at a temperature of 200-230°F and must therefore be cooled to near-ambient conditions prior to exhaustion into the mine haulageway. A system of automatic time-activated dampers is incorporated in the scheme to permit each segment to act as both an adsorber and a regenerator.

The capital costs for such a system are shown in Table 4 (1973 basis). It is considered preferable not to mount the assemblage on a skid largely because of potential problems in moving such a skid-mounted system in and out of the mine haulageway. Accordingly, all pieces of equipment are taken to the underground site and assembled there. Similarly, the system is dismounted at the site when the need arises to move it to a new location. The cost of charcoal has been capitalized since this material is capable of continuous regeneration for at least the estimated life of the major capital equipments (5 years). It is also expected that charcoal losses during this period (and therefore the need for make-up charcoal) will be minimal.



Note: Valves A, B, and C are open during radon adsorption and closed during charcoal regeneration when X, Y, and Z are open.

AIR FLOW PATTERNS IN (ONE-HALF OF) THE RADON ADSORPTION AND CHARCOAL REGENERATION SYSTEM

FIGURE 5

TABLE 4

Adsorption on Charcoal (Ambient Temperature)

CAPITAL COST - 5000 SCFM

Equipment	Size	Installed Cost
2 ea. Blowers	75 HP	$ 51,000
2 ea. Air Filters	5000 SCFM	2,000
4 ea. Heat Exchangers	2700 ft^2	65,000
2 ea. Adsorbers w/Heaters	6' diameter x 7-1/2'	18,000
Charcoal	9200 lbs.	3,800
		$142,800
Engineering, Overhead, Fee & Contingency (15%)		21,400
Total Capital Cost		$164,200

Table 5 itemizes the elements of operating cost. A 2-shift-per-day schedule is assumed for 250 days per year. The system incorporates automatic flow-switching and flow-control devices and therefore requires relatively little attention. An allowance of 1/3-man-equivalent per shift for labor and maintenance, and 1/8-man-equivalent for supervision has been made. An operating cost of about 5.2 cents per 1,000 cubic feet of air processed is estimated.

TABLE 5

Adsorption on Charcoal (Ambient Temperature)

ANNUAL OPERATING COSTS

5000 SCFM, 2 Shifts, 260 Days/Yr

(Capital Cost $164,200)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/Yr	$20,000
Utilities			
Electricity	1.87 MM kwh	$0.01/kwh	18,700
Water	34,000 M gal	$0.05/M gal	1,700
Supplies			
Filters			1,600
Charcoal & Miscellaneous			1,500
Maintenance	3% of Capital Cost		4,900
Direct Operating Cost			$48,400
Capital Charges	22% of Capital Cost		36,100
Total Annual Operating Cost			$84,500
Total Cost per MSCF			6.7¢

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B. ORGANIC FLUIDS

1. Description of a Preferred System

Radon is significantly more soluble in organic liquids than are the major gaseous components of air;[1,2] consequently, selective absorption of radon in such solvents would appear to be a possible basis for radon removal from uranium mine air. In a complete radon removal system, selective absorption of the radon could be followed by:

(1) storage or disposal of the resulting solution,

(2) pumping of the solution to the surface, stripping of the dissolved gases, and return of the solvent to the mine for reuse,

(3) stripping of the dissolved radon and air from the solvent within the mine, and pumping of the enriched gases to the surface for dispersion in air.

System 1 produces "infinite" hold-up of the radon. It would be feasible only if the volume of solvent required were small; otherwise the storage requirements and solvent costs would be prohibitive. Systems 2 and 3 transport radon from the mines to the surface for dispersal.

a. Solvent Selection

A solvent for selective absorption of radon from air should have the following characteristics:

- High capacity for radon
- Low nitrogen and oxygen solubility
- Low vapor pressure at the absorber temperature
- Non-flammable and non-toxic
- Chemically unreactive
- Low viscosity
- Low cost

High capacity for radon is desirable to minimize the quantity of solvent required. Low nitrogen and oxygen solubility is needed for selectivity. Low vapor pressure at the absorber temperature will minimize loss of solvent. Safety requires that the solvent be non-flammable and preferably non-toxic. The solvent should be chemically unreactive so that it will not be easily oxidized by air or react with the materials of construction. Low viscosity is desirable for ease in contacting the gas and liquid phase. Solvent cost is important since a large quantity of solvent will be required.

The solubility of radon in liquids may be conveniently correlated in terms of Hildebrand's solubility parameter, $\delta^{1/2}$, which has energy units of (energy/volume)$^{1/2}$, and may be thought of as a measure of the binding energy of a liquid.[3] Solutes which have values of $\delta^{1/2}$ close to that of the solvent have the greatest solubility, or in other words, "like dissolves like." Figure 6 illustrates this behavior for radon in a number of common solvents. Solubility data for Figure 6 was taken from references 1, 2, 3, and 4. The solubility parameter for radon is approximately 7(cal/cm^3)$^{1/2}$. In the figure, solubilities are expressed as Bunsen coefficients (the volume of gas, reduced to standard conditions, which will dissolve in a unit volume of solvent at a particle pressure of one atmosphere). The behavior of carbon disulfide is anomalous in that radon is significantly more soluble in it than would be expected.[6] The figure indicates that radon is most soluble in "nonpolar" solvents.

The solubility parameters for oxygen and nitrogen are 4.0 and 2.6 (cal/cm^3)$^{1/2}$ respectively,[5] so solvents which have high radon solubility should have relatively low solubility for air.

On the basis of flammability, the silicone oils, and the halogenated hydrocarbons, would appear to be good candidate solvents. However, the silicone oils are very expensive, especially those with low viscosities. Their solubility parameters are in the 5 to 6 range[7] which indicates possible problems with selectivity. Common nonflammable halogenated solvents are the chlorinated hydrocarbons and the fluoro-chloro-hydrocarbons of the Freon or Genetron groups. Many components of the latter group have very favorable solubility parameters and are believed to be relatively non-toxic.[8] However, they do have high vapor pressures at room temperature. Higher boiling compounds of this type are extremely expensive and are commercially available only in small quantities. The chlorinated solvents have no significant advantages over the Freon group and are believed to be significantly more toxic.[8] The high boiling esters and ethers such as dioctyl phthalate, dibutyl phthalate, butyl carbitol, etc. which have high flash points are also possibilities.

Preliminary estimates of the equipment size necessary for absorbing 95% of the radon in air at the rate of 5000 cubic feet per minute, using dibutyl phthalate as a solvent, and operating at a pressure of one atmosphere and room temperature showed that the equipment would be impractically large (the absorption tower where the dissolution of the radon would take place would be greater than 400 feet in height and about 15 feet in diameter). Similar size estimates were obtained for carbon disulfide which is the best radon solvent (from a solubility point of view) though not really a practical one for this application because of its flammability and toxic properties.

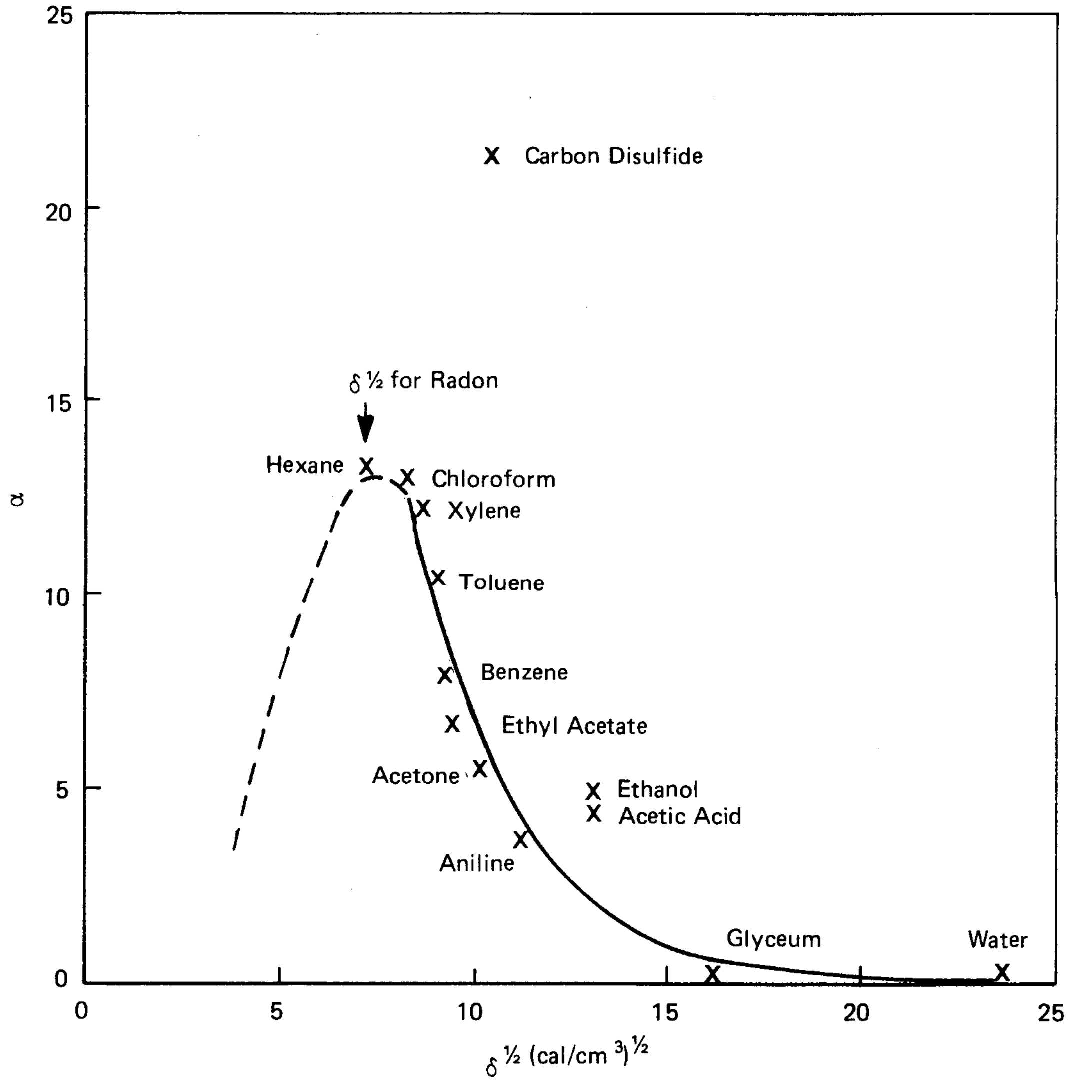

FIGURE 6. DEPENDENCE OF RADON SOLUBILITY IN VARIOUS SOLVENTS AS A FUNCTION OF SOLUBILITY PARAMETER

Operation at higher pressures and reduced temperatures was considered to reduce the size of the absorption equipment (both of these strategies result in increased absorption of radon by a given volume of solvent). Dichlorodifluoromethane (CCl_2F_2, Refrigerant 12, Freon 12) was chosen as a model for these considerations because of its favorable solubility parameter (7 $(cal/cm^3)^{1/2}$), its low toxicity, and nonflammability. This compound is a gas under normal conditions, so operation at low temperatures and/or high pressures is mandatory. The solubility of radon in dichlorodifluoromethane was computed by the method of Prausnitz and Shair,[5] since measured solubilities were not available.

b. Solvent Flow Requirements

The removal of a component of a gas mixture by absorption requires the contacting of the gas and liquid phases and their subsequent separation into clean gas and contaminated liquid streams. The quantity of gas that may be transferred between the phases is affected not only by the solubility of the gas in the liquid but also by the gas-liquid contacting scheme.

If the solvent enters the top of a vertical packed column, so that it runs down over the packing and the gas enters the bottom so that it passes upward through the column, the contacting scheme is said to be "countercurrent." The effluent gas last contacts the entering solvent. If the gas were also to enter the top and pass downward, the contact is "cocurrent." In this scheme the effluent gas last contacts effluent liquid. In each case the gas concentration of contaminant in the effluent gas can be no lower than that which is in equilibrium with the concentration of contaminant in the liquid it last contacts.

Figure 7 illustrates these processes graphically. Here y is the mole fraction of radon in the gas phase and x is the mole fraction of the liquid phase. For simplicity, it will be assumed that the remaining components of the gas phase are not significantly soluble in the solvent, that the solvent is not volatile, and that the concentration of radon is low. These assumptions imply that the molar liquid flow rate, L, and the molar gas flow rate, G, are constant throughout the absorber. The bulk compositions of gas and liquid phases at any point along the column are given by the operating line running from (x_i, y_i) to (x_o, y_o) for cocurrent contact and from (x_o, y_i) to (x_i, y_o) for countercurrent contact. Subscript i indicates inlet mole fractions of radon, and subscript o indicates outlet mole fractions. It may be shown that the equations of the operating lines are:

$$-\frac{L}{G} = \frac{y_i - y}{x_i - y} = \frac{y - y_o}{x - x_o}$$

for cocurrent operations, and

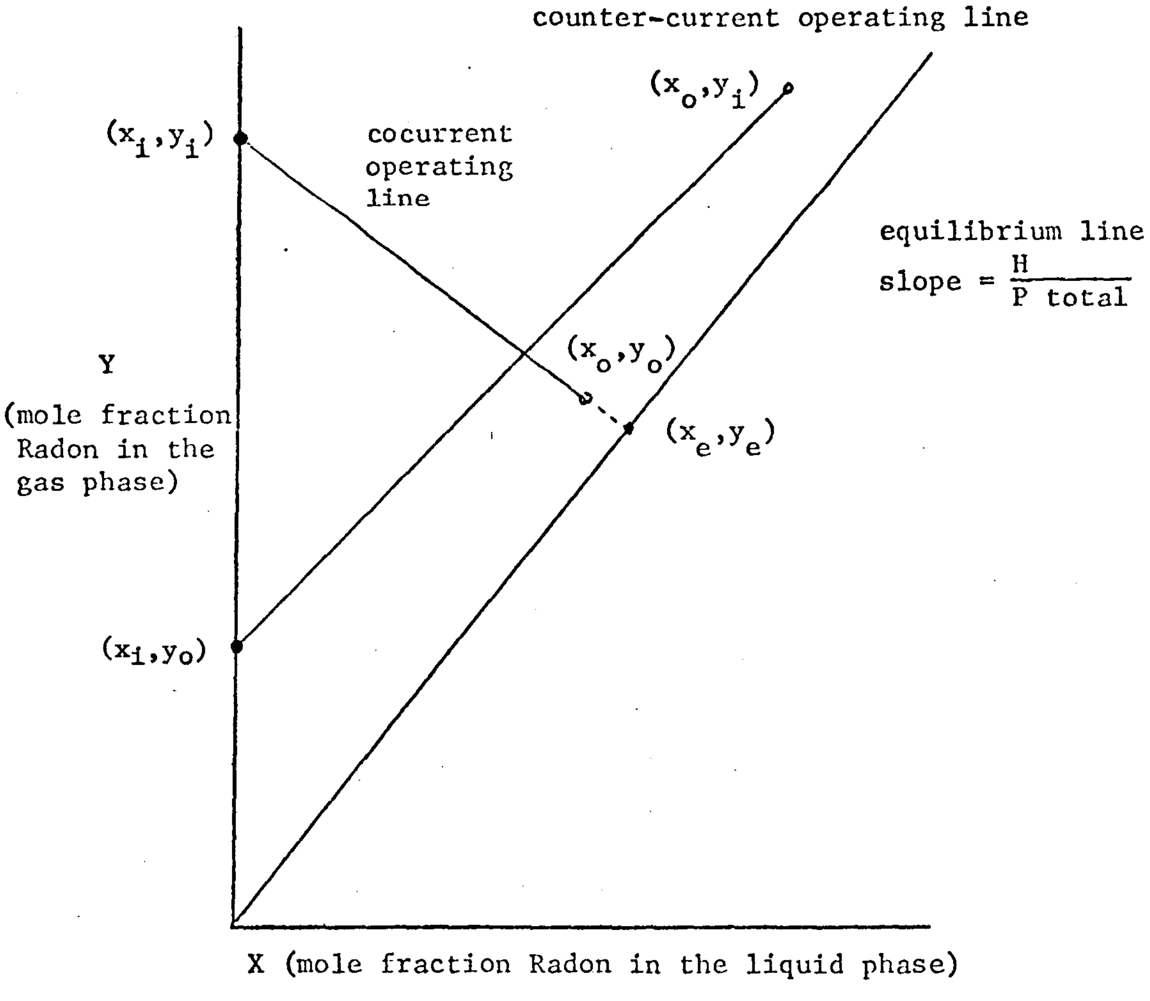

Figure 7. Absorber Operation Characteristics

$$\frac{L}{G} = \frac{y_i - y}{x_o - x} = \frac{y - y_o}{x - x_i}$$

for countercurrent operation.[9]

The slope of the equilibrium line is

$$\frac{H}{P_{total}}$$

where P_{total} is the total gas pressure and H is the Henry's law constant of proportionality. This follows from the definition of y and from Henry's law:

$$y = \frac{P}{P_{total}} = \frac{Hx}{P_{total}}$$

(Henry's Law states that the partial pressure of a solute above a solution is proportional to the mole fraction of solute in the solution, or P = Hx, where P is the partial pressure, x is the mole fraction, and H is the constant of proportionality.)

Once the operating conditions are fixed, they, together with the equilibrium relation, determine the ultimate performance possible from an absorber. For a cocurrent scrubber, no matter how long the phases are kept in contact the gas composition can never go below y_e or the liquid composition above x_e indicated in Figure 7. In a countercurrent scrubber more efficient contact (implying more efficient solvent usage) moves the operating line closer to the equilibrium line while the slope remains $\frac{L}{G}$. The best performance would be obtained when the operating line touches the equilibrium line. If $\frac{L}{G}$ is less than $\frac{H}{P_{total}}$ for countercurrent contact, equilibrium limit is reached when the outlet liquid is in equilibrium with the inlet gas. This simply says that the operating line will intersect the equilibrium line at some point (x_o, y_i), and once this occurs there will be no further driving force for transferring radon from the gas phase to the solution. Reducing $\frac{L}{G}$ reduces the solvent flow requirements. For a given fractional radon reduction, f, in the gas phase, the minimum solvent flow required will be that at which the operating line intersects the equilibrium line. Figure 8 shows several operating lines corresponding to different values of $\frac{L}{G}$ (operating line slopes) which will produce a given fractional reduction of radon of f. The operating line with the minimum value of $\frac{L}{G}$ will intersect the equilibrium line.

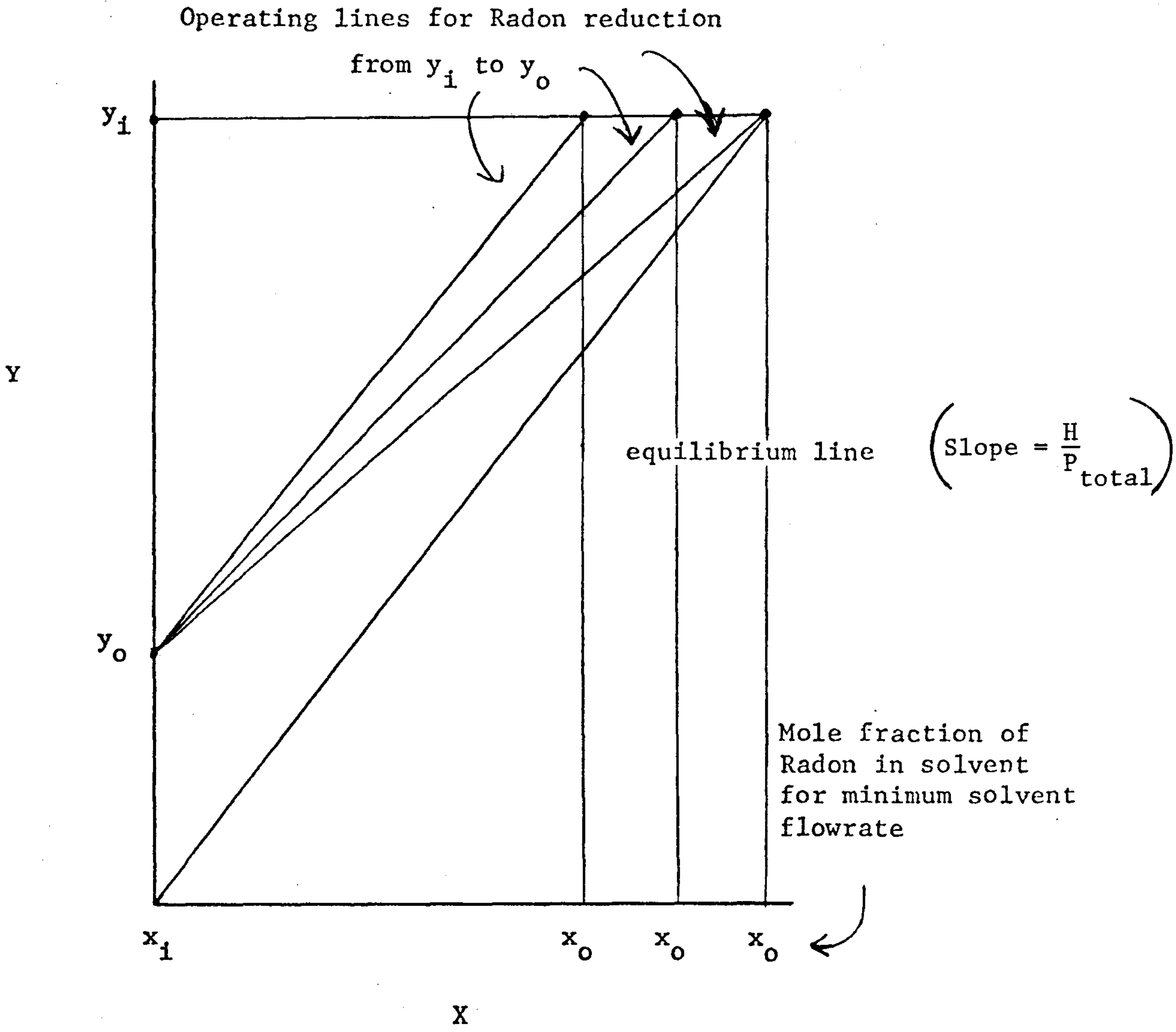

Figure 8. Graphical Determination of Minimum Solvent Flowrate

Using

$$y_i = \frac{H}{P}x_o$$

$$f = (y_i - y_o)y_i$$

$$x_i = 0$$

one finds that the minimum value of L is

$$f \frac{GH}{P_{total}}.$$

For cocurrent flow the analogous equation for minimum L is

$$\frac{GH}{P_{total}} \left(\frac{f}{1-f} \right).$$

It is easy to see that as f approaches unity (large radon reduction, L approaches $\frac{GH}{P_{total}}$ for countercurrent operation, but L increases without limit as f approaches unity for cocurrent contact.

c. Choice of Absorption Systems

Minimum solvent flowrate requirements computed from fGH/P_{total} show that System 1 is much less attractive than the solvent recycling systems (2 and 3). Carbon disulfide will be used to illustrate this, since it has the highest measured solubility for radon.

The Henry's Law constant for CS_2 is 19.9 atmospheres/mole fraction at 25°C.[2] At a gas flowrate of 5000 cubic feet/minute (5722 moles/minute), a total pressure of 1 atmosphere, and 90% reduction in radon, the minimum solvent flowrate required is 103,000 moles/minute (6200 liters/minute). Compression of the air by a factor of 10 would reduce the solvent flow requirements by a factor of 10. At 0°C the Henry's law constant for carbon disulfide is 12.1. Cooling to 0°C and compression to 10 atmospheres would reduce the solvent flowrate to about 380 liters/minute, 3.8×10^6 liters per week, or about 10^6 gallons per week. In practice, solvent flowrates would probably be greater than those calculated from fGH/P_{total}.

Costs for Systems 2 and 3 are expected to be similar. System 2 was selected for further work since the amount of equipment which must be placed in the mine itself is less than for System 3. Operation at low temperatures and high pressures was selected to reduce the size of the radon absorber to practical dimensions even though this necessitated additional equipment such as a compressor which would not be needed for operation under normal conditions. Further optimization of the system was not attempted because no significant reductions in size or cost were expected. For example a less volatile solvent could have been selected which might eliminate the need for charcoal absorption of solvent vapor at the expense of greater solvent costs and large scrubber dimensions.

d. Process Description & Basis of Design (See Fig. 9)

The proposed process design involves the following steps:

1. Compressing, dehydrating, and cooling the 5000 cfm input of mine air to 100 psia and -22°F (-25°C).
2. Removal of radon by absorption in dichlorodifluoromethane in a packed tower.
3. Removal of dichlorodifluoromethane from the gas by carbon bed adsorption.
4. Heating and expanding the clean air to 63°F (17°C) and ambient pressure.
5. Stripping the radon from the solvent leaving the absorption column.
6. Removing the radon from the stripper vapor and ejecting it to the atmosphere.

Capital investment for a system to process 5000 cfm of air would amount to $3,117,000. Based on 4160 hours per year (16 hrs/day, 260 days/yr), the annual operating costs would be approximately $1,089,000 or $0.87/1000 SCF of cleaned ventilation air.

The absorption system is sketched in the schematic flow diagram (Figure 9). This system is based on handling 5000 cfm of input air at 15 psia, a maximum ambient temperature of 100°F, and 100% relative humidity.

Prior to entering the absorption column, the mine air is filtered, compressed to 100 psia, cooled to 35°F (2°C), dried in a glycol dehydration unit, then chilled again to -22°F. The gas then enters the absorption column, a packed tower approximately 4' 6" diameter

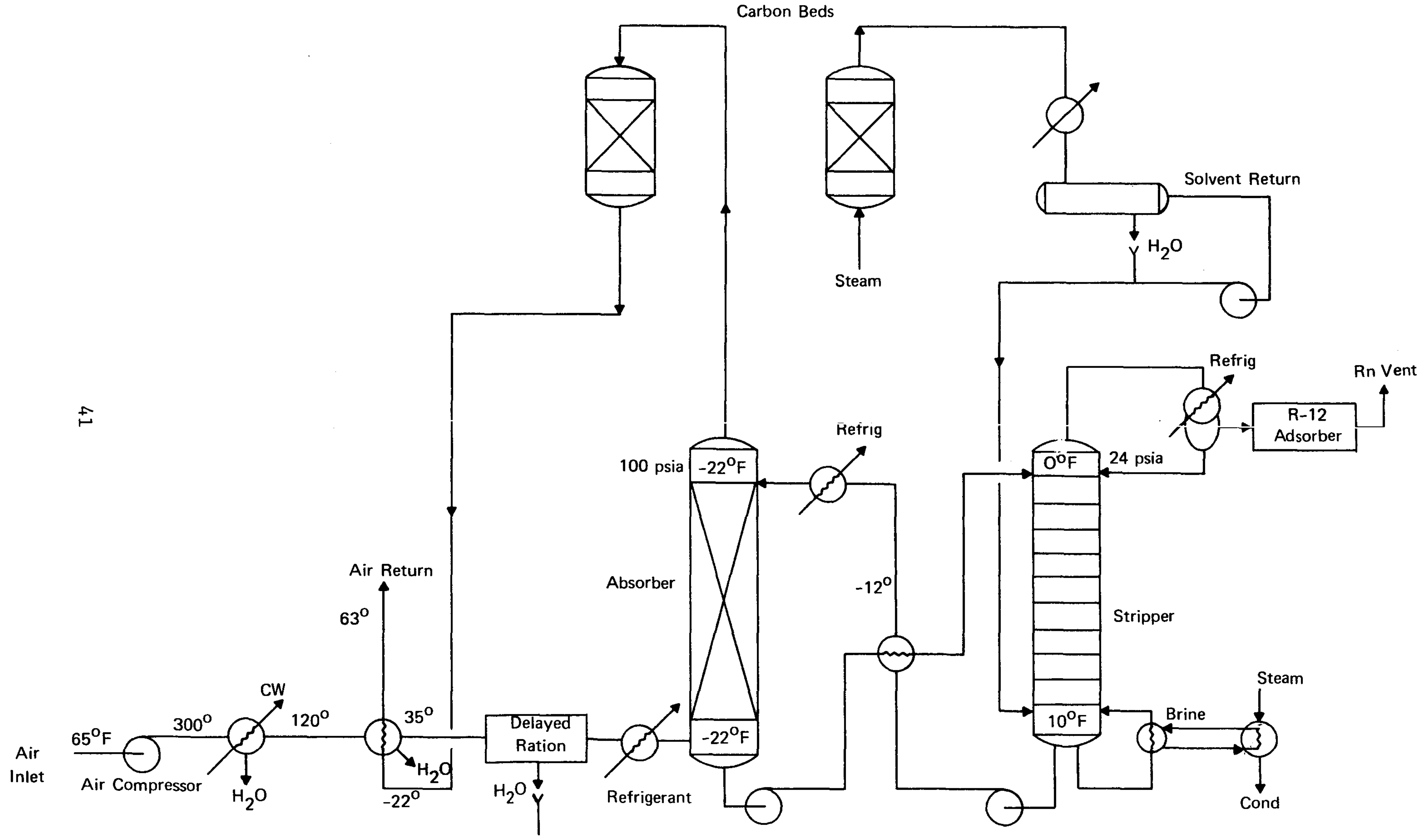

FIGURE 9. Schematic Arrangement of Organic Fluid System

x 50' filled with 2" Raschig rings, where it contacts a countercurrent of dichlorodifluoromethane at 22°F. Here 95% of the radon in the gas is absorbed by the liquid solvent as are insignificant quantities of N_2 and O_2 (0.45 vol. % of total throughput); additionally, some of the solvent vaporizes and becomes part of the gas stream. To avoid losing this valuable material in uneconomic quantities, the gas passes through a carbon bed system to absorb the vapors. The purified air then goes to a heat exchanger and is expanded to ambient pressure at $63^{o}F$ ($17^{o}C$).

The enriched solvent at the base of the absorber is pumped continuously up to a trayed stripping column at the surface. The column, approximately 7' 6" diameter x 50' high, operates at 24 psia and $-5^{o}F$ ($-20^{o}C$). Here radon is removed from the solvent and emitted to the atmosphere.
A small carbon adsorption bed recovers the small quantity of dichlorodifluoromethane ("Refrigerant 12") that would otherwise be vented with the off-gases.
Auxiliary equipment in the system includes an electrically heated system for regenerating the carbon beds, a thermosiphon reboiler for the stripper, and a refrigeration system to cool the liquid and gas feed streams to the absorber.

2. Efficiency & Reliability of Radon Removal

a. Effectiveness of Radon Removal

The organic fluid system is designed to achieve a radon removal efficiency of at least 90% of whatever concentration occurs in the incoming air. This efficiency will be maintained for volumes of air up to 5000 SCFM, and irrespective of changes in temperature, barometric pressure relative humidity or the presence of various air contaminants. Auxiliary methods are used to achieve necessary control of temperature and humidity.

b. Size and Weight of Equipment

Of equipment in the mine, the most bulky is the absorption column (split into four 7.5' diameter x 13' sections weighing 8,000 lbs. each) and the carbon beds (two 6' diameter x 17' horizontal tanks weighing 80,000 lbs. each). All equipment in the mine should fit in a space 15' x 20' x 40' with a total weight near 100 tons. Two 6" diameter pipes running to the surface will handle the solvent flow to and from the stripper. A 15 hp pump will handle the flow to stripper.

c. Residence Times of Radon and Air

Residence time of radon is on the order of a few minutes (maximum), depending on pumping flow rates to the stripper unit at the surface. Residence time of air is on the order of a few seconds.

d. Rate and Cause of Deterioration

The principal cause of deterioration of the system is expected to be the gradual loss of activity of the charcoal beds which remove vapors of the solvent (dichlorodifluoromethane) from the processed air. These beds must be regenerated frequently by heating with steam, a process which is never perfect, and which will eventually result in gradual accumulation of impurities on the charcoal. When and if this occurs the charcoal must be replaced. The solvent itself is expected to last indefinitely.

e. Regeneration of Absorber

As indicated in the system description, low pressure steam is intended to be used to periodically purge the charcoal beds of accumulated solvent deposits as well as any other air pollutant which may be adsorbed to them. The charcoal itself is used to prevent loss of solvent to the mine air, and, to a lesser extent to outdoor air at the surface stripping unit.

3. Safety

The major safety hazard is in avoiding overpressure of the process equipment and/or too rapid vaporization of the solvent. The vapor pressure of dichlorodifluoromethane is only 132 psia at 100°F (37°C) so the system can easily be designed to withstand that overpressure above the operating value of 100 psia. Additionally the process equipment should be thermally insulated to reduce refrigeration requirements and to retard boil-off in the event of a power failure. Since radon is vented continuously from the system there is no serious radiation hazard.

The system should be fairly quiet. Noise would be generated by the various liquid pumps, the large air compressor, and by steam regeneration of the carbon bed and other fluid flows. However, this noise should not be obnoxious and can easily be reduced by acoustic insulation.

4. Cost

a. Capital Costs

Capital costs are outlined in Table 6 as estimated by conventional equipment factoring cost procedures. Total plant capital cost is estimated to be $3,117,000.

b. Annual Operating Costs

Annual operating costs are outlined in Table 7. Electric power requirements total 1800 Kw. Water requirements are 1000 gpm. Chemical losses should be insignificant. Total operating costs are estimated to be $1,089,000/year or $0.87/1000 SCF of air processed.

TABLE 6

Absorption in Dichlorodifluoromethane

CAPITAL COST - 5000 SCFM UNIT

Item	Size	Installed Cost
Steam Compressor	500 hp	$ 125,000
Dichlorodifluoromethane (solvent)	20,000 lbs	10,000
Heat Exchangers		480,000
Absorber	4'6" dia. x 50'	90,000
Stripper	7'6" dia. x 50'	155,000
Carbon Beds	6'0" dia. x 17', two	265,000
Compressor	900 hp	360,000
Refrigeration System	700 tons	450,000
Dehydration System		100,000
Electric Water Heaters	4,900 Kw	75,000
TOTAL EQUIPMENT COST		$1,985,000
Engineering Overhead and Contingencies (15%)		298,000
TOTAL CAPITAL COST		$2,283,000

TABLE 7

Absorption in Dichlorodifluoromethane

ANNUAL OPERATING COST

5000 SCFM, 2 Shifts, 260 days/year

(Capital Cost $2,283,000)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/man-year	$ 20,000
Utilities			
Electricity	24.5×10^6 Kwh	$0.01/Kwh	245,000
Water	250,000 Mgal	$0.05/Mgal	12,500
Maintenance		4% of capital cost	91,500
Direct Operating Cost			$ 369,000
Capital charges		22% of capital cost	502,000
TOTAL ANNUAL OPERATING COST			$ 871,000
TOTAL COST PER MSCF			$0.70

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C. SEMIPERMEABLE MEMBRANES

1. Description of a Preferred System

a. Introduction

Membranes may be used in gas separation processes for two main purposes:

(1) to separate materials on the basis of differential permeation properties, and

(2) as an interface between a fluid absorbent and a process stream.

In the first case, where the permeation properties of the membrane itself are used to separate two or more species, the membrane material is selected on the basis of the relative permeabilities of the species concerned. In addition, a driving force across the membrane must be maintained for the species which is expected to permeate most rapidly. This driving force is commonly expressed as the partial pressure difference of the permeating gas; it may or may not be associated with a total pressure gradient in a system of more than one component.

In the second case, the membrane may or may not permit the absorbed material to permeate more rapidly than the non-absorbed material; the separation is achieved primarily by employing a selective absorbent at the downstream side of the membrane. This sets up a gradient across the membrane for the removed species only.

Inasmuch as this second method requires a combination of techniques, it will not be considered in detail in this section but will be treated separately in Section I below.

b. Separations Based on Membrane Diffusion

The use of membranes to separate gas mixtures has been studied widely, mostly in relation to:

(1) the separation of helium from natural gas

(2) purification of hydrogen

(3) separation of methane from natural gas

The first two examples are special cases in which membranes are available whose permselectivity (semipermeability) with respect to the desired component is virtually ideal. Thus, dry silica glass is permeable to helium because the lattice dimensions of the glass are larger than the atomic diameter of helium. (No other atom or molecule that is stable under normal conditions is small enough to behave in the same manner.) In the second example, hydrogen gas rapidly permeates thin membranes of palladium by a mechanism involving catalytic dissociation into hydrogen atoms which are small enough to penetrate the metal structure.

Clearly it would be optimistic to expect such an idealized situation in the case of radon, and no such system has been identified. In this study, a more common situation will be considered in which the differential membrane permeability of radon with respect to air components will lead to enrichment of the (permeated) air stream with respect to radon.

Values for the permeability of radon through membrane materials have not been reported in the literature. However, judging from the permeability of other gases through polymeric membranes, we would expect silicone rubber to have the highest radon permeability of all currently available membrane materials. We have estimated the permeability of radon through silicone rubber by plotting the logarithms of the permeabilities of a range of gases (see Table 8) against their boiling points and extrapolating to appropriate values for radon (see Figure 10). The reasonable linearity of this plot lends some confidence to a value of about 270 x 10^{-9} cm^3, cm/cm $^2_;$ sec, cmHg* for the permeability of radon.

Two basic schematics for membrane enrichment of radon in an airstream are shown in Figures 11 and 12. The first supposes that the partial pressure gradient required for gas transport is provided by vacuum pumps at the downstream side of each permeator; the second by compression of the gas at the upstream side. The relative merits of these alternates were evaluated.

c. Single Stage Permeation Cell Calculations

A computer program was developed to calculate the membrane area and concentrations of the permeate achieved under different feed stream conditions. The program is general for any number of species in the feed gas. Data necessary to operate the program are feed flow rate and concentrations, permeability constants, membrane thickness, and pressure on each side of the membrane. The following is a description of the calculation procedure.

The permeation of a multi-component gas through a membrane may be simply represented by a modified form of Fick's law.(1,2) For the permeation cell shown in Figure 13,

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* The unit of permeability is such that a gas with a value of 1 will pass at a rate of 1 cm^3 per second through a membrane 1 cm thick and 1 cm^2 in area under a driving force of 1 cm Hg pressure.

TABLE 8

Permeability of Silicone Rubber to Gases

Gas	Permeability*	MW	Boiling Point (°K)
H_2	55	2	20
He	30	4	4
CH_4	80	16	111
N_2	25	28	63
O_2	50	32	90
CO_2	270	44	194 (subl)
Ar	50	40	87
Kr	99	84	120
Xe	171	131	166
Rn	∿ 270 (estimated)	222	211

* Units of permeability: (10^{-9} cm^3, cm/cm^2, sec, cm Hg)

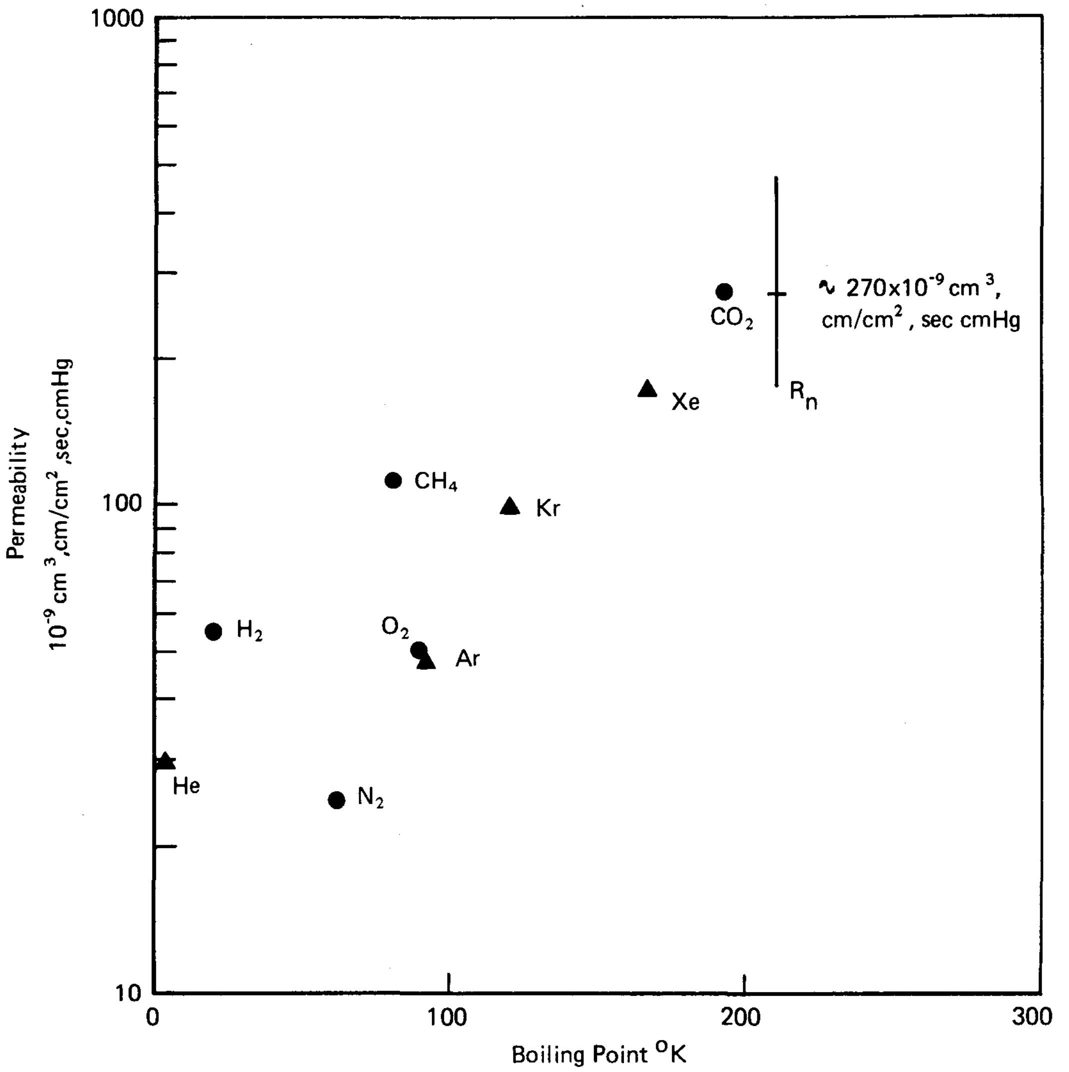


FIGURE 10. VARIATION OF GAS PERMEABILITY THROUGH SILICONE RUBBER WITH BOILING POINT

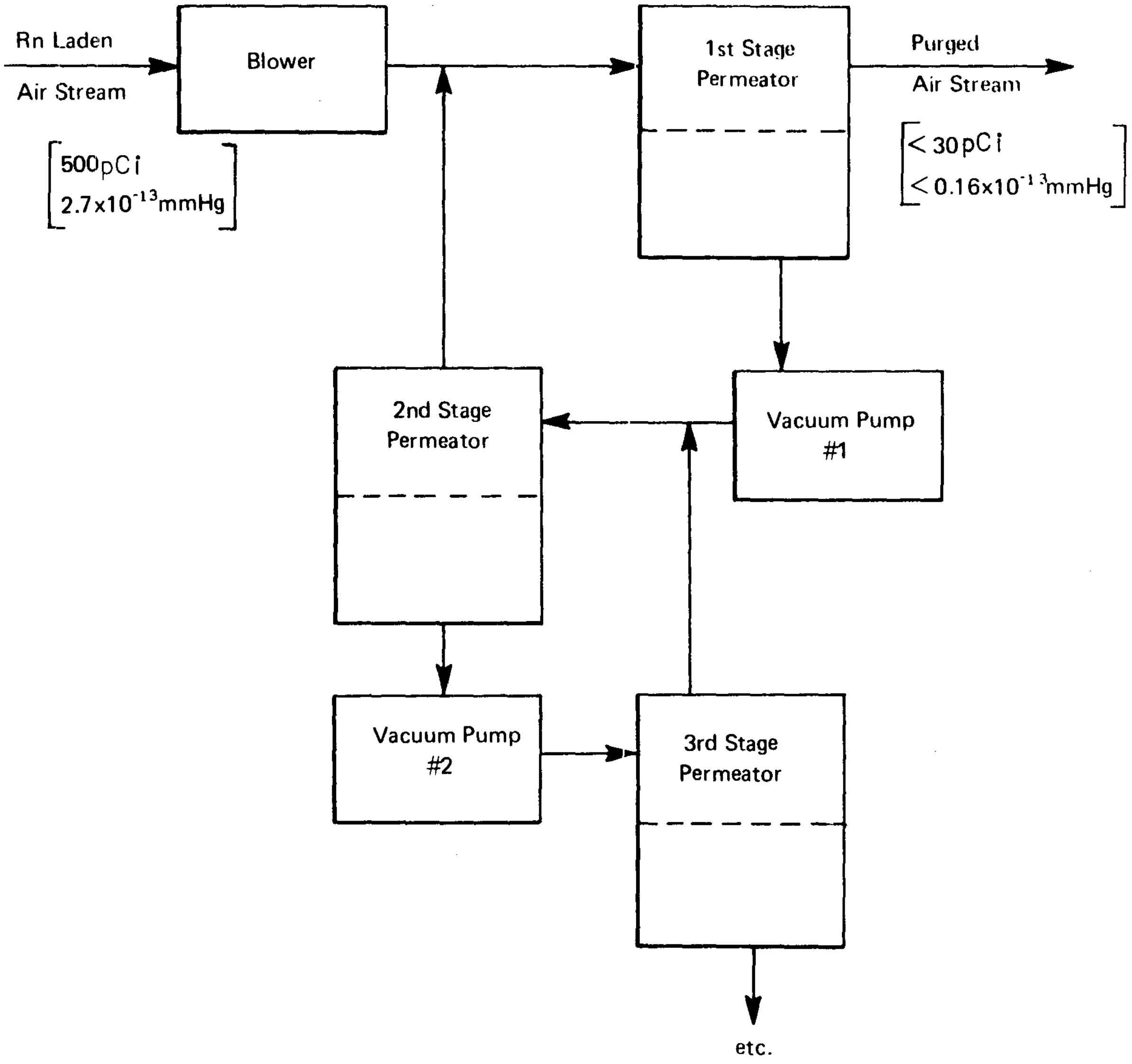


FIGURE 11. SCHEMATIC NUMBER OF A MEMBRANE PROCESS FOR RADON ENRICHMENT OF AIR

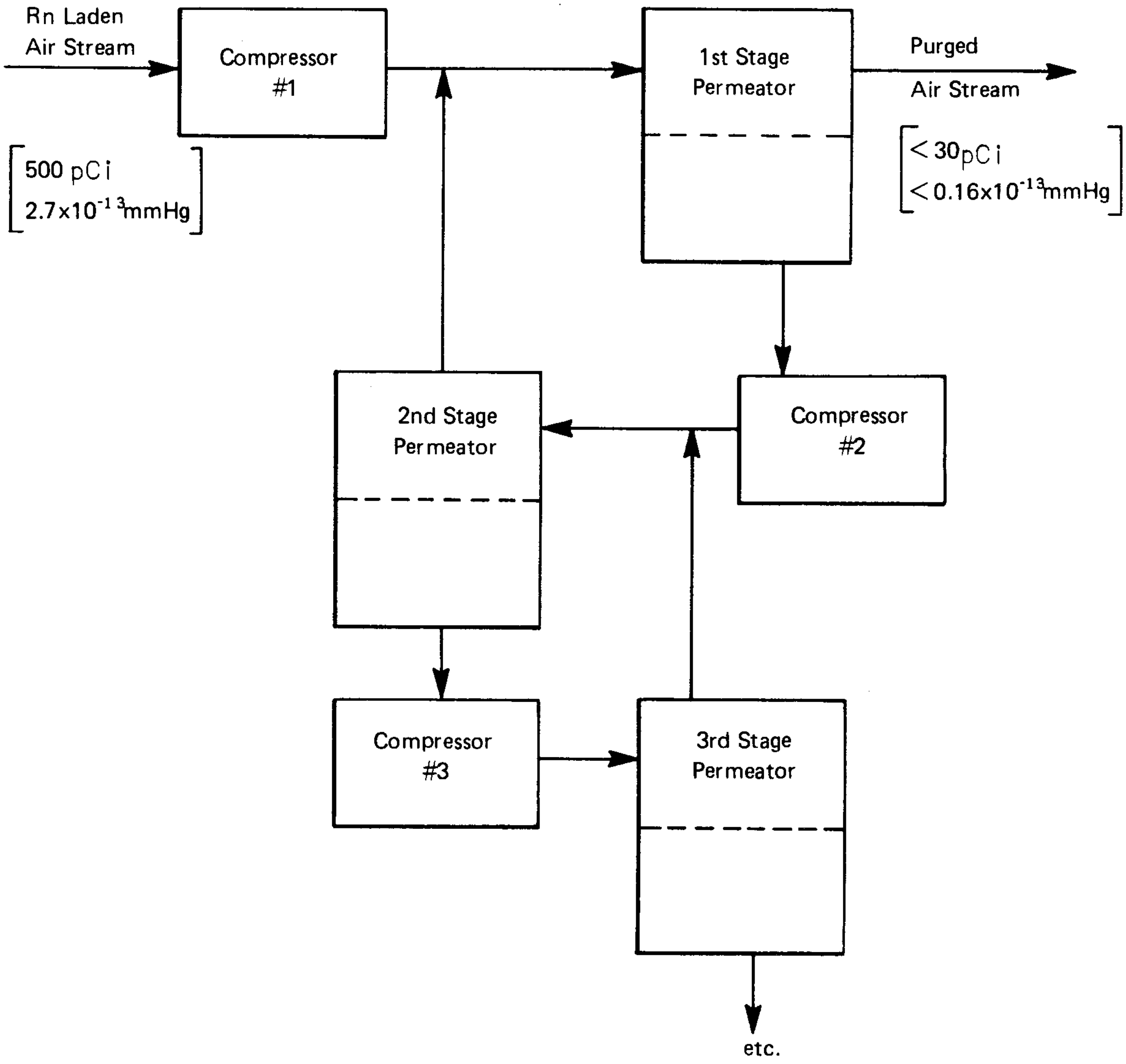


FIGURE 12 SCHEMATIC NUMBER 2 OF A MEMBRANE PROCESS FOR RADON ENRICHMENT OF AIR

$$q_i = \frac{A}{t} \underline{P}_i \left(P\hat{u}_i - P\hat{d}_i\right) \quad (1)$$

where q_i = mass flux of component i

A = membrane area

t = membrane thickness

$\underline{P}_i$ = permeation constant for i

$P\hat{u}_i - P\hat{d}_i$ = partial pressure driving force for i

Equation (1) may be changed to a form more suitable for computation:

$$Vy_i = \frac{A}{t} \underline{P}_i \left(P\hat{u}z_i - Pd\hat{y}_i\right) \quad (2)$$

where V = total rate of permeation

$P\hat{u}$ = total upstream pressure

$P\hat{d}$ = total downstream pressure

z_i = mole fraction of i in permeate

y_i = mole fraction of i in raffinate

Performing mass balances over the cell yields the equations:

$$F = U + V \quad (3)$$

$$Fw_i = Uz_i + Vy_i \quad (4)$$

where F = total feed rate into the cell

U = total removal rate of raffinate

W_i = mole fraction of i in feed

and by definition,

$$\sum_{i=1}^{n} y_i = 1 \quad (5)$$

Combining equations (2) and (4) for component i = 1,

$$A = \frac{V y_1 t}{\underline{P}_1 \left(P\hat{u} \left(\frac{F}{U} w_1 - \frac{V}{U} y_1\right) - P\hat{d} y_1\right)} \qquad (6)$$

and for component i,

$$y_i = \frac{\frac{F}{U} P\hat{u} w_i}{\frac{Vt}{\underline{P}_i A} + P\hat{u} \frac{V}{U} + P\hat{d}} \qquad (7)$$

Rearranging equation (4),

$$z_i = \frac{F w_i - V y_i}{U} \qquad (8)$$

Given values for w_i, $\underline{P}_i$, F, U or V, $P\hat{u}$, $P\hat{d}$, and t, equations (3), (5), (6), (7) and (8) may be used to find V or U, A, y_i, and z_i.

The algorithm for a computer program to perform the above calculations is as follows:

1. From equation (3), calculate U or V.
2. Assume a value, y_1, for the largest component of the feed.
3. Calculate A from (6).
4. Calculate y_i (for i = 1 to n) from (7).
5. Calculate $\sum_{i=1}^{n} y_i$.
6. Repeat steps 2-5 until $\Sigma y_i = 1$ within a preset limit; a simple iterative procedure such as interval halving is satisfactory.
7. Calculate z_i from (8).

One should note that there are inherent limitations in the calculations of the algorithm. It must be assumed that there is perfect mixing on both sides of the membrane. Also, mixing on the high pressure side of the membrane must be rapid enough so that the raffinate has the same concentration as gas within the cell.

After the final y_i's have been determined, the entire algorithm may be restarted using these values as feed concentrations for another cell. This process may be continued through a series of stages in a cascade approach. Maintaining mass balance throughout the system, provisions for recycle streams have been included in the computational procedure.

d. Quantitative Aspects

The program was tested for the simple system of successive stages in Figure 13. Feed gas was assumed to be radon in air, i.e., nitrogen and oxygen. Data for a silicone rubber membrane, with preferential permeability for radon, was used for the examples. A series of representative runs is shown in Table 9. Since radon is the component of interest, concentrations for this species alone are presented.

There are several interesting results indicated by Table 9. One of the most important factors in membrane permeation is the fraction of the feed stream allowed to permeate through the membrane, i.e., V/F. The desired goal is to reduce the ratio of radon in the effluent to radon in the feed (z/w) so that the effluent may be safely purged. Small permeation rates (V/F) do not produce sufficient permeation to reduce the radon in the raffinate to acceptable concentrations. Larger permeation rates improve the radon permeation but result in increased membrane area; such rates also require equipment capable of handling large gas flows. Clearly, optimization of the system would minimize V/F, z/w and membrane area.

In data not shown on the table, the upstream pressure, P_u, was shown to be of some importance; downstream pressure is usually maintained at atmospheric for ease of operation. Decreases in P_u result in increased membrane area and decreased radon concentration in the raffinate. The concentration effect is not as large as the increase in area.

There are two other dependencies also worthy of note. First, the required area of the membrane is directly proportional to its thickness, but thickness has no effect upon radon concentrations. Second, the area is directly proportional to the feed flow rate, but the rate has no effect upon radon concentrations.

e. Results of Computation

Feed flow rates, pressure drop across the membrane, and membrane material were investigated initially for a constant temperature of 25°C (77°F) and subsequently at 15°C (60°F), 38°C (100°F) and 93°C (200°F). Along with determination of the optimum number of stages and individual stage cuts (the ratio of permeate to feed volume) the study produced cascade systems capable of meeting specified conditions. The two most important conditions were: (1) the high radon and (2) the low activity product stream's radon concentration must be reduced to 10% of that of the feed stream. Two membrane materials were studied, silicone rubber, whose effectiveness and cost are high, and polyethylene, whose effectiveness is somewhat lower and whose cost is much lower.

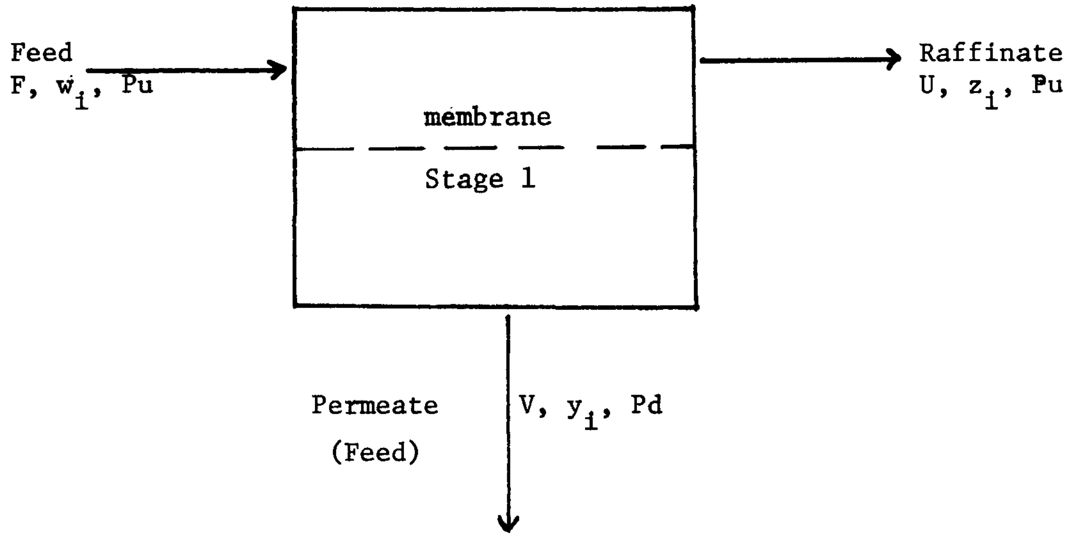


Figure 13. Permeation Cell

TABLE 9

Sample Results for System of Figure 13

Permeability Constants

$P_{N_2} = 25 \times 10^{-9}$ cm^{3}cm/sec cm^2 (cmHg)

$P_{O_2} = 50 \times 10^{-9}$

$P_{Rn} = 270 \times 10^{-9}$

t = 0.001 in.
Pu = 60 atm.
Pd = 1 atm.
F = 2000 CFM 9500 pCi/1 Rn)

V/F	Stage	A (sq ft)	Incoming Radon level w (pCi/1)	Outgoing Radon Level z (pCi/1)	Permeated Radon level y (pCi/1)	z/w
0.10	1	3,140	500	294	2,330	0.59
0.20	1	6,340	500	206	1,660	0.43
0.90	1	30,000	500	65	544	0.13
0.98	1	32,700	500	60	505	0.12

Silicone Rubber

A seven stage cascade with silicone rubber membranes is shown in Figure 14. The Product 1 stream is the high activity, low volume stream and the Product 2 stream is the low activity, high volume stream. Table 10 presents concentrations and flows for every stream and the required membrane area for each stage. The values presented are for a feed flow of 2000 CFM. For larger flows the stream concentrations do not change, but the area is directly proportional to the flow. Table 11 shows results for the cascade of Figure 13, with the same conditions as Table 10 except at a flow of 5000 CFM.

The pressure drop across a membrane does not follow a simple relation for either the stream concentrations or the membrane area of a particular stage. Table 12 presents data for a system with an upstream feed pressure (P high) of 10 atmospheres, as opposed to the same system of Table 11 with 60 atm. In both cases the downstream permeate pressure is 1 atmosphere, and the system temperature is 25°C (77°F).

It should be noted that the raffinate stream is assumed to be at the same pressure as the feed. A reduction in upstream pressure is desirable to reduce capital investment and operating costs of the required compressors. However, comparing the results of Tables 12 and 11, it is seen that at the lower operating pressure for a seven stage cascade, a total membrane area of almost seven times that for the high pressure system is required. Also, in seven stages the desired low specific activity for stream product 2 is not achieved. Therefore, more stages would be necessary - using even more membrane material, and more compressors.

Polyethylene

Due to the relatively high cost of silicone rubber membranes ($\$20/ft^2$), an alternate membrane material was investigated. It was decided that the very low cost of polyethylene (about $0.23¢/ft^2$) made it a good candidate for use in the radon removal cascade. As was the case for silicone rubber the membrane permeability for radon was not available and had to be estimated. It is known that:

$$P = D \times S$$

where P = permeability constant

D = diffusion coefficient

and S = solubility parameter

Extrapolating from known values of D and S for gases of lower molecular weight and for a low density polyethylene membrane[3], a permeability constant for radon at 25°C (77°F) was obtained. It should be stressed that such a value is approximate and actual permeation tests would be

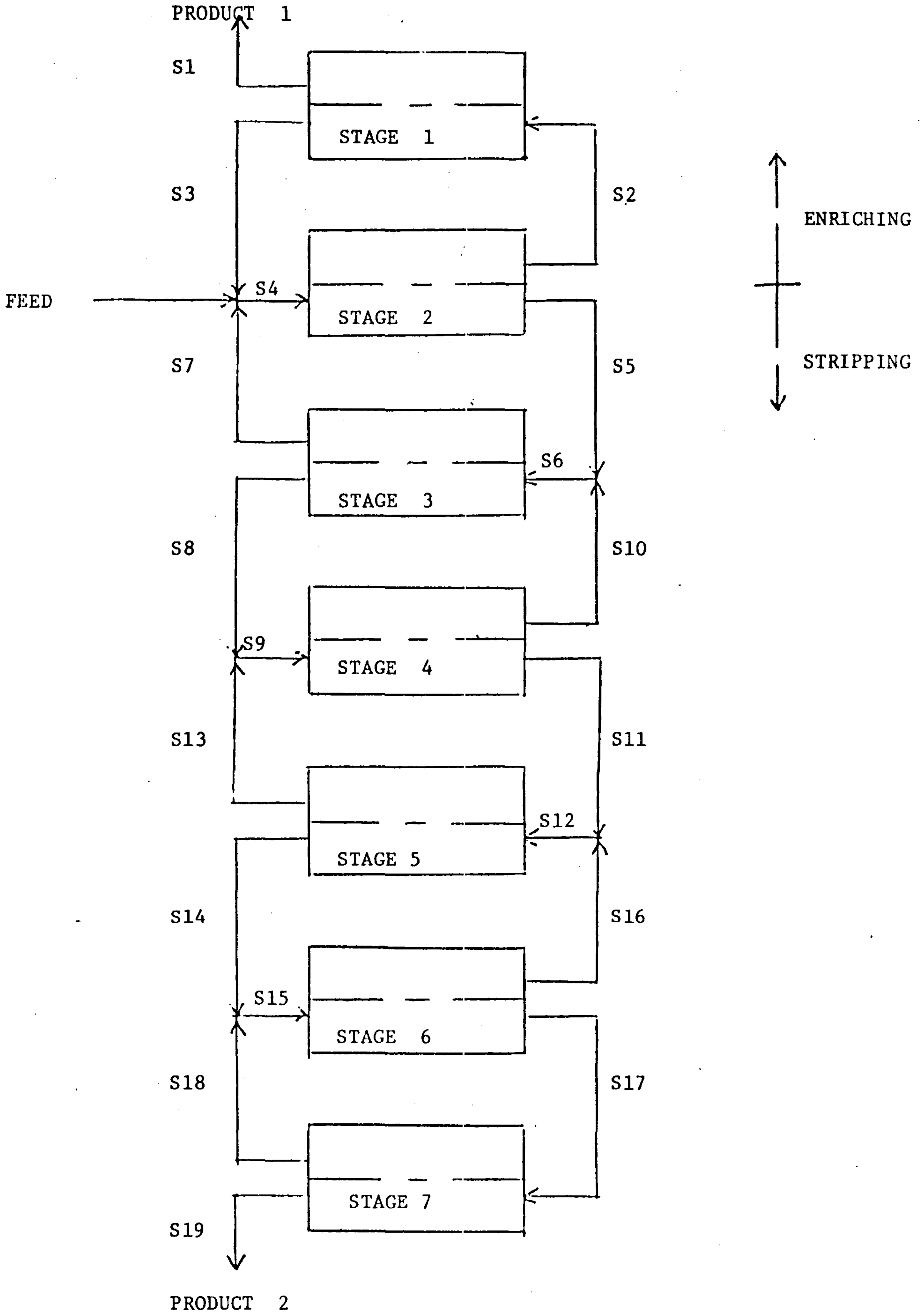


Figure 14. Seven Stage Cascade for Silicone Rubber Membrane

TABLE 10. RESULTS FOR SEVEN STAGE CASCADE (Figure 14)

(2000 CFM Feed)

Membrane Thickness = 0.001 in.
P high = 60 atm.
P low = 1 atm.
Feed = 2000 CFM (500 pCi/l Rn)
Temp. = 25°C (77°F)

Silicone Rubber Permeability Constants

P_{N_2} = 25 X 10^{-9} cm^3 cm/sec cm^2 (cmHg)
P_{O_2} = 50 X 10^{-9}
P_{Rn} = 270 X 10^{-9}

Stage Number	Membrane Area (ft^2)	Stream Number	Feed Flow (CFM)	Feed Conc. (pCi/ℓ)	Permeate Flow (CFM)	Permeate Conc. (pCi/ℓ)	Raffinate (Unpermeated) Flow (CFM)	Raffinate (Unpermeated) Conc. (pCi/ℓ)
1	1891	S1			205	4424		
		S2	315	3077				
		S3					110	577
2	3004	S4	2426	740				
		S2			315	3077		
		S5					2110	390
3	3004	S6	2426	561				
		S7			315	2335		
		S8					2110	296
4	3002	S9	2425	410				
		S10			315	1706		
		S11					2109	216
5	2998	S12	2418	282				
		S13			314	1174		
		S14					2103	149
6	2954	S15	2372	176				
		S16			308	732		
		S17					2063	92
7	2605	S17	2063	92				
		S18			268	387		
		S19					1795	48

TABLE 11. RESULTS FOR SEVEN STAGE CASCADE (Figure 14)
(5000 CFM Feed)

Membrane Thickness = 0.001 in.
P high = 60 atm.
P low = 1 atm.
Feed = 5000 CFM (500 pCi/l Rn)
Temp. = 25°C (77°F)

Silicone Rubber Permeability Constants

$P_{N_2} = 25 \times 10^{-9}$ cm^3 cm/sec cm^2(cmHg)
$P_{O_2} = 50 \times 10^{-9}$
$P_{Rn} = 270 \times 10^{-9}$

Stage Number	Membrane Area (ft^2)	Stream Number	Feed Flow (CFM)	Feed Conc. (pCi/l)	Permeate Flow (CFM)	Permeate Conc. (pCi/l)	Raffinate (Unpermeated) Flow (CFM)	Raffinate (Unpermeated) Conc. (pCi/l)
1	4,728	S1			512	4424		
		S2	788	3077				
		S3					276	577
2	7,509	S4	6064	740				
		S2			788	3077		
		S5					5276	390
3	7,509	S6	6064	561				
		S7			788	2335		
		S8					5276	296
4	7,507	S9	6061	410				
		S10			788	1706		
		S11					5273	216
5	7,495	S12	6044	282				
		S13			786	1174		
		S14					5258	149
6	7,384	S15	5929	176				
		S16			771	732		
		S17					5158	92
7	6,513	S17	5158	92				
		S18			671	387		
		S19					4488	48

TABLE 12. RESULTS FOR SEVEN STAGE CASCADE (Figure 14)

(P_{high} = 10 atm)

Membrane Thickness = 0.001 in.
P high = 10 atm.
P low = 1 atm.
Feed = 5000 CFM (500 pCi/l Rn)
Temp. = 25°C (77°F)

Silicone Rubber Permeability Constants

$P_{N_2} = 25 \times 10^{-9}$ cm^3 cm/sec cm^2 (cmHg)
$P_{O_2} = 50 \times 10^{-9}$
$P_{Rn} = 270 \times 10^{-9}$

Stage Number	Membrane Area (ft^2)	Stream Number	Feed Flow (CFM)	Feed Conc. (pCi/l)	Permeate Flow (CFM)	Permeate Conc. (pCi/l)	Raffinate (Unpermeated) Flow (CFM)	Raffinate (Unpermeated) Conc. (pCi/l)
1	31,417	S1			512	3336		
		S2	788	2405				
		S3					276	676
2	49,676	S4	6064	731				
		S2			788	2405		
		S5					5276	481
3	49,676	S6	6064	677				
		S7			788	2239		
		S8					5276	447
4	49,675	S9	6061	609				
		S10			788	2006		
		S11					5273	401
5	49,584	S12	6044	518				
		S13			786	1704		
		S14					5258	340
6	48,821	S15	5929	400				
		S16			771	1318		
		S17					5158	263
7	42,979	S17	5158	263				
		S18			671	868		
		S19					4488	172

required for a true permeability.By trial and error it was found that the four stage system of Figure 15 would produce an acceptable separation of radon using polyethylene membranes. For 2000 CFM and 5000 CFM systems, the results in Tables 13 and 14, respectively, were obtained. It is readily seen that although the number of stages is less than required for a silicone rubber cascade, each stage needs about 200 times the membrane area. In total, the polyethylene system necessitates using almost 120 times the membrane material (at comparable feed flows).

The reason for the differences between the polyethylene and silicone rubber cascades lies in the permeability constants - as listed in Tables 10 and 13 respectively. The absolute magnitude of the permeability of silicone rubber to radon is about 80 times greater than that of polyethylene (PE). Thus more area is required to pass comparable flows of radon through PE membranes. However, since the permeation rate is also dependent on partial pressures, the relation between permeability constant and species' stream concentrations is not simple. Placing stages in cascades with recycle streams further changes the situation. The radon selectivities for PE, i.e., the ratio of permeability constants for radon to nitrogen or oxygen, are superior to those of silicone rubber. Therefore, each stage of PE membrane produces a better separation and less stages are required.

The four stage polyethylene system designed for operation at 25°C (77°F) is an alternative to the silicone rubber system. A preferred alternative was subsequently developed by taking into consideration the effect of temperature on the permeability of polyethylene and of silicone rubber membranes. Based on available data for the effect of temperature on the permeability of these materials to other gases, especially the "rare" gases (He, Ne, Ar, Kr, Xe), the behavior of radon, nitrogen and oxygen was calculated for a variety of temperatures. As a practical low limit for ambient operating temperature, 15°C (60°F) was chosen, and for a practical maximum 38°C (100°F) was used. Since compression of the gases to 20 atmospheres is contemplated, however, an equilibrium temperature in the neighborhood of 93°C (200°F) -- a practical continuous service temperature -- may be obtained with cooling. At this temperature, the permeability of polyethylene to all the gases (Rn, N_2, O_2) increases dramatically, suggesting a possible reduction of membrane area by about a factor of 30, compared to our previous estimate. This is also the maximum temperature which polyethylene and silicone rubber will tolerate for continuous service. For silicone rubber, the effect of increased temperature is less marked, and, since the permeability increases for oxygen and nitrogen but not significantly for radon, the effect appears to be disadvantageous. Permeability ratios were calculated for the three temperatures, representing the ratio of the permeability for each gas through each membrane, relative to that at 25°C (77°F). The results were:

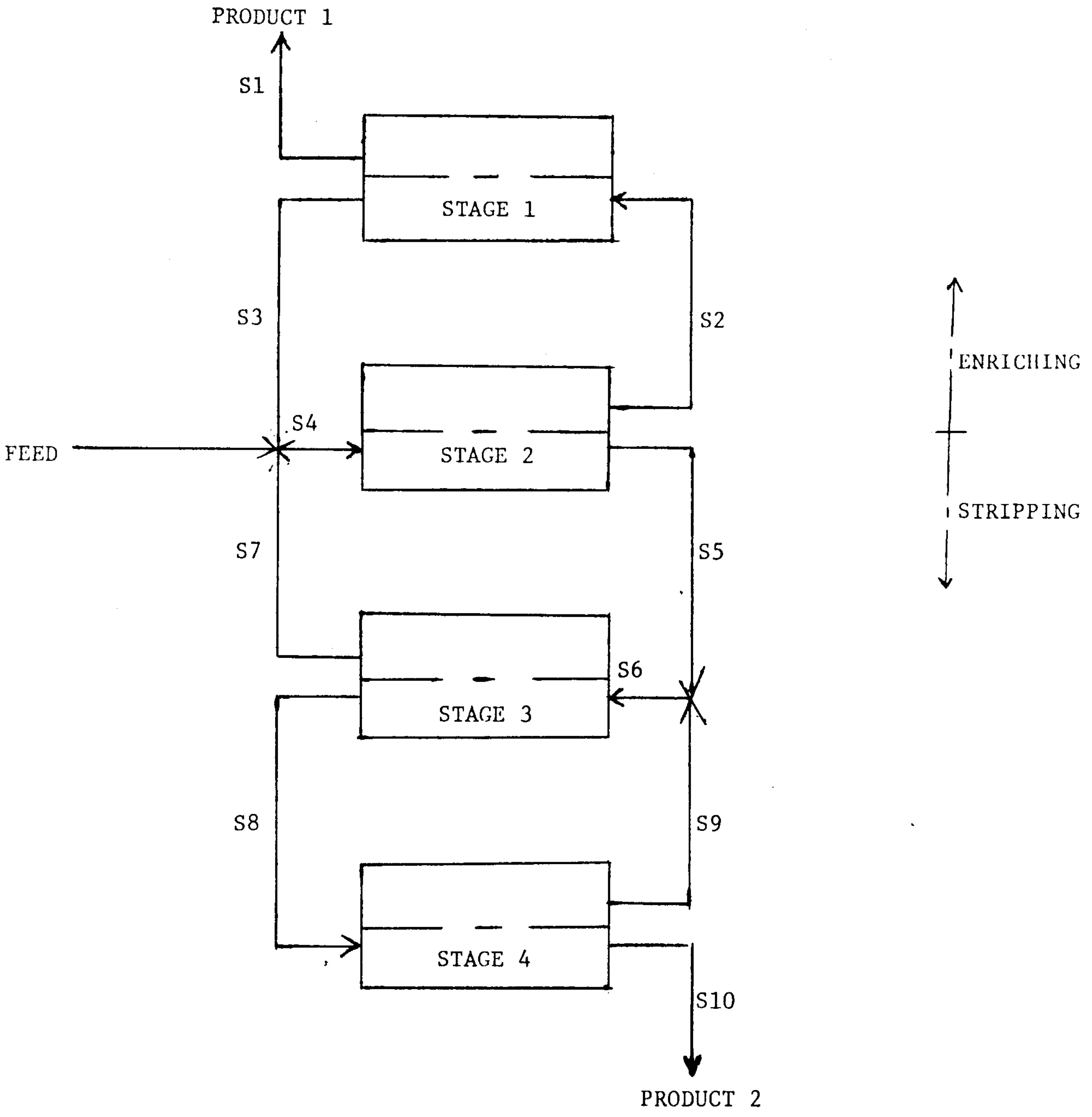


FIGURE 15. Four Stage Cascade.

TABLE 13. RESULTS FOR FOUR STAGE CASCADE (Figure 15)

(2000 CFM Feed)

Membrane Thickness = 0.001 in.
P high = 60 atm.
P low = 1 atm.
Feed = 2000 CFM (500 pCi/l Rn)
Temp. = 25°C (77°F)

Polyethylene Permeability Constants
$P_{N_2} = 0.97 \times 10^{-10}$ cm^3 cm/sec cm^2(cmHg)
$P_{O_2} = 2.9 \times 10^{-10}$
$P_{Rn} = 35 \times 10^{-10}$

Stream Number	Membrane Area ($ft^2 \times 10^{-5}$)	Stream Number	Feed		Permeate		Raffinate (Unpermeated)	
			Flow (CFM)	Conc. (pCi/l)	Flow (CFM)	Conc. (pCi/l)	Flow (CFM)	Conc. (pCi/l)
1	3,948	S1			204	4707		
		S2	314	3156				
		S3					110	276
2	6.642	S4	2418	562				
		S2			314	3156		
		S5					2104	174
3	6.580	S6	2372	192				
		S7			308	1083		
		S8					2064	59
4	6.580	S8	2064	59				
		S9			268	335		
		S10					1796	18

TABLE 14. RESULTS FOR FOUR STAGE CASCADE (Figure 15)
(5000 CFM Feed)

Membrane Thickness = 0.001 in.
P high = 60 atm.
P low = 1 atm.
Feed = 5000 CFM (500 pCi/1 Rn)
Temp. = 25°C (77°F)

Polyethylene Permeability Constants
P_{N_2} = 0.97 X 10^{-10} cm^3 cm/sec cm^2 (cmHg)
P_{O_2} = 2.9 X 10^{-10}
P_{Rn} = 35 X 10^{-10}

Number	Membrane Area (ft^2 X 10^{-5})	Stream Number	Feed		Permeate		Raffinate (Unpermeated)	
			Flow (CFM)	Conc. (pCi/l)	Flow (CFM)	Conc. (pCi/l)	Flow (CFM)	Conc. (pCi/l)
1	9.840	S1			511	4707		
		S2	786	3156				
		S3					275	276
2	16.54	S4	6046	562				
		S2			786	3156		
		S5					5260	174
3	16.42	S6	5931	192				
		S7			771	1083		
		S8					5160	59
4	14.88	S8	5160	59				
		S9			671	335		
		S10					4489	18

Polyethylene

Temperature		60°F	100°F	200°F
$P_T/P_{77°F}$	(all species)	0.52	2.17	30.3

Silicone Rubber

Temperature		60°F	100°F	200°F
$P_T/P_{77°F}$	N_2	0.86	1.20	2.24
	O_2	0.89	1.16	1.91
	Rn	1.00	1.01	1.07

Through the courtesy of Professor James Porter of the Massachusetts Institute of Technology, an improved computer program was made available to us for optimization of parameters for design of a polyethylene membrane permeator. Using this program, and assuming an operating temperature of 93°C (200°F) throughout the system, a three-stage cascade was found to be the most cost-effective arrangement, as shown in Figure 16. The optimum pressure differential across the membrane was found to be twenty (20) atmospheres. The flow rates and radon concentrations for this preferred system are shown in Table 15. The total membrane area required at 20 atm. is estimated to be 685,000 ft^2, compared to 147,000 ft^2 for operation at 60 atm., but the cost-saving effected by eliminating the extra compressor required for the fourth stage more than offsets the additional expense of the larger amount of membrane. Figure 17 shows schematically the very simple design of the system, which consists essentially of a series of three permeator compressors. Although we considered a number of alternative configurations for the permeators, especially that which Stern, et al.[2] recommended for separation of helium from natural gas, our preferred design is based on the use of hollow fibers. Devices of this kind, incorporating tens of millions of fine fibers (.001" I.D., .003" O.D.), closely spaced in parallel array, and enclosed in a heavy-walled cylindrical steel vessel, are manufactured commercially (E.I. duPont de Nemours Corp., "Permasep") for separation of H_2 from CO and other uses. Our calculations, whose results are confirmed by the manufacturer, indicate that a pressure drop of about 2.5 atmospheres (43 psia) will be required to overcome frictional forces in the gas flowing inside hollow fibers of .010" I.D. to produce a total permeate flow of 1800 SCFM through the longest of the three permeator units. (This assumes the high pressure feed/raffinate to be in contact with the exterior walls of the fibers, so that the low pressure permeate will flow through the interior of the fibers.) Thus, an actual raffinate pressure of 20 + 2.5 = 22.5 atm. would be required to maintain permeation through the membrane and flow through the fiber cores.

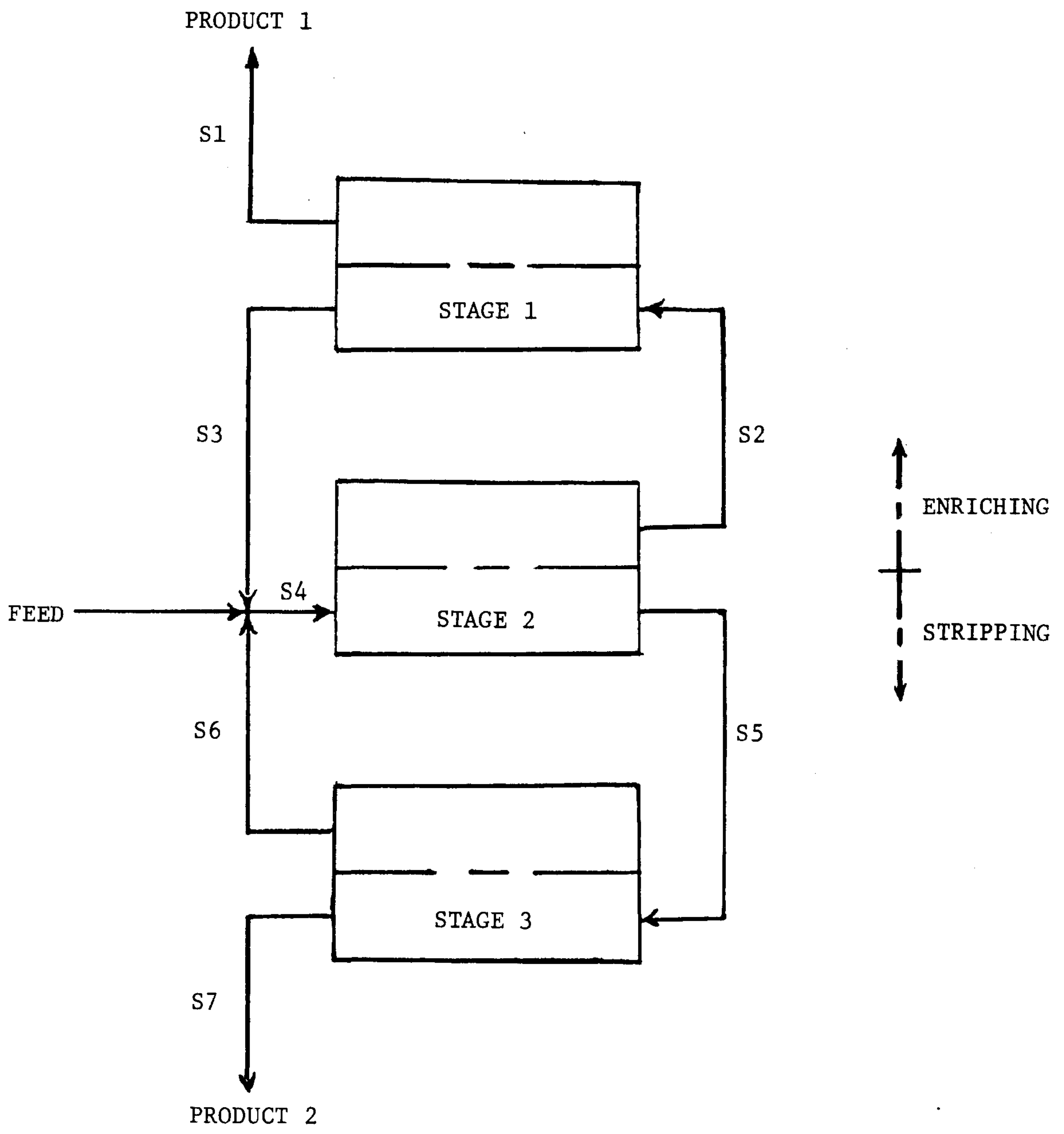
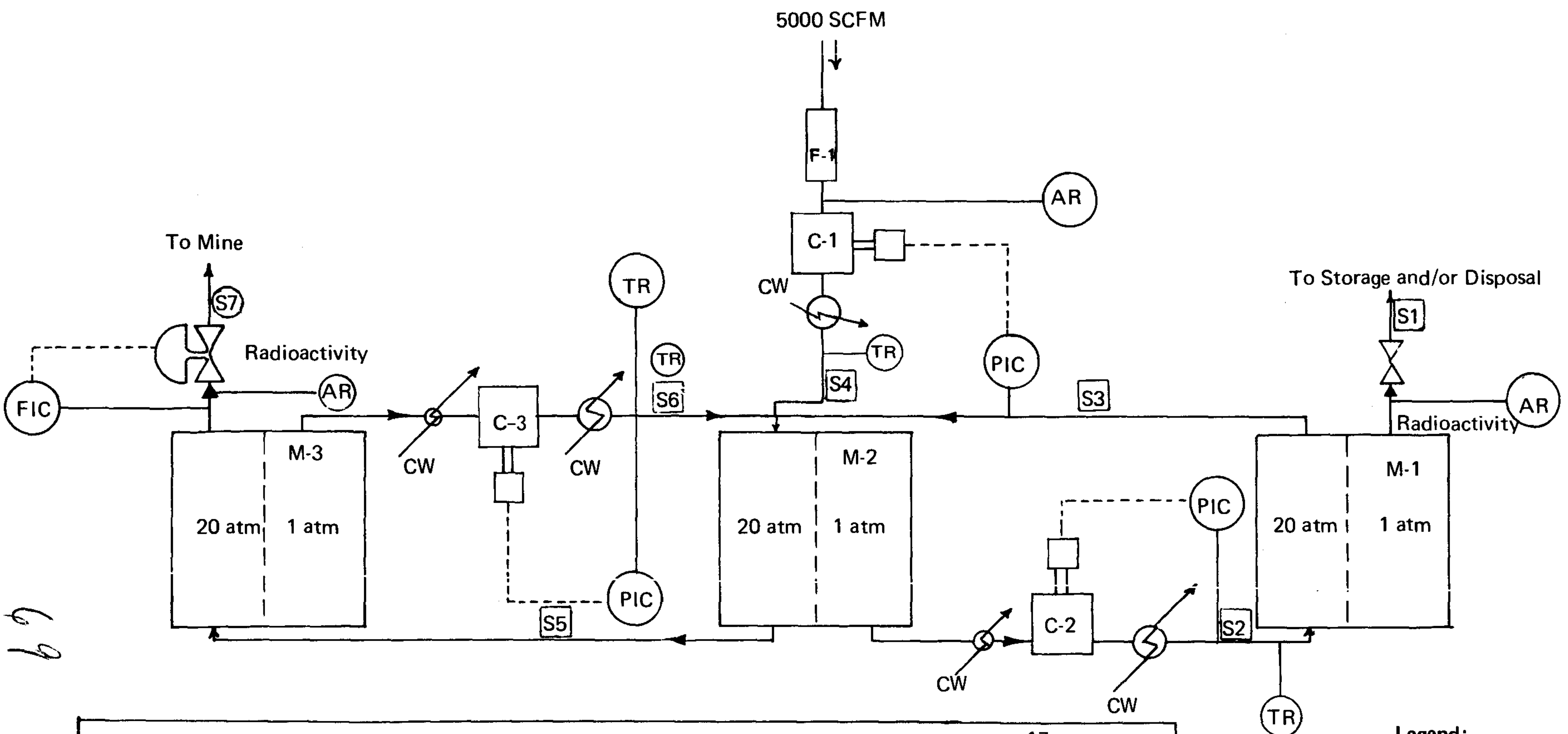


FIGURE 16 -- THREE STAGE CASCADE



Legend:
F – Filter
C – Compressor
M – Membrane Permeator
AR – Radioactivity Analyzer Recorder
TR – Temperature Recorder
PIC – Pressure Indicator Controller
FIC – Flow Indicator Controller
CW – Cooling Water

Material Balance for 5000 SCFM System in lb moles/hour; Comp in Mole Fraction X 10^{17}

Stage No.	3		2		1	
	Flow	Comp	Flow	Comp	Flow	Comp
Permeate	183.255	31.612	293.925	101.87	80.65	287.67
Raffinate	725.85	3.16	909.1	8.8963	213.275	31.612
Total	909.105	–	1203.025	–	293.925	–
Feed	909.1	8.8963	1203.05	31.612	293.925	101.87

Difference in Permeate and Raffinate Vs Feed Due to Computer Round-Off Errors

FIGURE 17 SCHEMATIC DIAGRAM OF 3-STAGE PERMEATOR

TABLE 15 RESULTS FOR THREE STAGE CASCADE

Membrane Thickness = 0.001 in.
P high = 20 atm.
P low = 1 atm.
Feed = 5000 CFM (500 pCi/1 Rn)
Temp. = 200°F

Polyethylene Permeability Constants

$P_{N_2} = 2.9 \times 10^{-9}$ cm^3 cm/sec cm^2 (cmHg)
$P_{O_2} = 8.8 \times 10^{-9}$
$P_{Rn} = 106 \times 10^{-9}$

Number	Membrane Area (ft^2 X 10^{-5})	Stream Number	Feed		Permeate		Raffinate (Unpermeated)	
			Flow (CFM)	Conc. (pCi/1)	Flow (CFM)	Conc. (pCi/1)	Flow (CFM)	Conc. (pCi/1)
1	0.727	S1			500	4551		
		S2	1822	1612				
		S3					1322	500
2	3.774	S2			1822	1612		
		S4	7458	500				
		S5					5636	141
3	2.353	S5	5636	141				
		S6			1136	500		
		S7					4500	50

Assuming .012" O.D. fibers (.010" I.D., .001" wall), the necessary surface area of the membrane for each stage will be provided by using bundles of 2.3 million parallel fibers. Permeator #1 will then be 12.5 feet long, Permeator #2 will be 20 feet long and Permeator #3 will be 4 feet long. Assuming an inter-fiber spacing of .003", the total bundle will be about 25" diameter, and can thus be housed in a 30" I.D. steel pipe.

2. Effectiveness of Radon Removal

The system described above has been designed to remove 90% of the radon present in the incoming air, irrespective of the initial concentration. The removed radon is concentrated in a small fraction of the processed air which is designed to be 10% of the input volume. This fraction is waste air and is presumed to be vented to the surface.

The membrane cascade system is capable of handling changes in the mine atmosphere to be processed. Differences in radon concentrations and air volumes present no problems as long as such changes are monitored. For example, an increase in flow rate requires more membrane area, and thus the addition of extra stages to reach acceptable concentrations. (Air velocity is important only as it relates to compressor sizing and flow rate as described above.) For changes in ambient temperature either heating/cooling devices or different membrane area may be necessary. In this, it should be remembered that the permeability of gases is usually related exponentially to temperature.

Aerosol particles must be eliminated completely due to a possible "deactivating" effect induced by the buildup of such particles on the membrane surface. Gaseous constituents of the air other than radon - such as carbon monoxide and nitrogen oxides - will alter the distribution of radon in the product streams. Such changes are dependent upon the specific components, their concentrations, and the particular membrane material. A true multi-component system is more difficult to handle than a radon/air mixture, but the principles described still apply. The membranes considered thus far are very permeable to water vapor. Therefore, changes in the relative humidity of air may be treated as for any other gaseous component.

b. Size and Weight

The preferred filtering system consists of a viscous impingement self-cleaning filter followed by a glass fiber filter. Both could be contained in a package 6' x 4' x 4'.

Reciprocating compressors with coupled electric motors will be used for air compressions. The feed compressor/driver will weigh ∿65,000 lbs. The dimensions of this compressor will be about 11' x 16' x 6'. The dimensions of the electric motor are about 5' x 3' x 3'. The two smaller compressor/

driver systems will weigh about 20,000 lbs each. Dimensions for the compressor are 11' x 6' x 6' and for the drivers 4' x 2½' x 2½'. Ordinarily, these would be mounted on concrete foundations. The larger system would require a foundation of 180 cubic yards of concrete. The two smaller systems would require about 60 cubic yards each. Presumably, the rock floor of the mine would be sufficiently strong to permit elimination of much or all of this foundation.

The permeators chosen for this process are hollow fiber permeators. These are 30" steel pipe sections packed with bundles of hollow polyethylene fiber. Lengths and total weight are as follows:

Permeator #1	12.5 feet
Permeator #2	20 feet
Permeator #3	4 feet
Total Weight	17,000 lbs

The entire assembled system would occupy a volume whose dimensions would be 11' x 6' x 68' and would consist of space for the inlet air filter, followed by the first-stage compressor and the first permeator, the second-stage compressor and the second permeator, and finally the third-stage compressor and the third permeator.

c. Residence Time of Radon and Air in the Device

The entire throughput time for processed air in the three-stage system is less than one minute.

d. Deterioration and its Remedies

The only integral machinery required by the cascade system are the high pressure compressors. The membrane support structures contain no moving parts. Thus, the failure of the system would occur most likely through malfunction of a compressor or rupture of a membrane. Since the desired gas separation can be achieved most efficiently at very high pressure differentials, e.g., 20 atmospheres, the effects of such pressures should be of primary concern in preventing system failure. Suitable compressors with the desired pressure capacity are available and need not be discussed.

With proper design, the cascade stages can operate with little maintenance other than checks for pinholes and surface contamination by particulates. The compressors would also be subject to a regular preventive maintenance schedule. The life of the membrane material and the compressors would depend upon the particular operating conditions and actual tests would be necessary.

3. Safety

The radon activity retained in the example seven stage silicone rubber and four stage polyethylene cascades has been calculated on the basis of radon solubility and partial pressure differences across the membrane. Breakdowns of the level of activity for each stage (at 2000 and 5000 CFM) are presented in Tables 16 and 17 for silicone rubber and polyethylene, respectively. It is clear that the larger membrane area required by high flows increases the radon retention per stage. Computations at 5000 CFM show that the space time for a silicone rubber stage is about 20 seconds; for a polyethylene stage it is about 0.5 second. Therefore, there is no significant holdup or decay of radon within each permeation cell. Since the activity levels in Tables 16 and 17 are in the microcurie region, residual radiation from the cascade itself should be negligible.

There are no known toxic byproducts produced by the cascade. There is some oxygen removal occurring in the high flow rate/low radon specific activity product stream, but this does not appear to be of a magnitude which would present a problem. It should be noted that there is an accompanying enriching effect on the oxygen of the low flow/high radon product. However, this stream is not of great concern.

There are no fire or explosive hazards except those caused by abuse of the electric motors. Therefore the compressor/drivers should be housed in water-tight enclosures so as to prevent short-circuiting caused by water or other electrically conducting media finding their way into the circuitry of the compressor/drivers.

The principal source of noise will be the reciprocating compressors and their associated driving motors, but these machines are not expected to be objectionably noisy. If necessary, however, accoustically insulating enclosures could be installed around the compressors.

4. Cost

The proposed process design includes the following:

1. Compression and heating of the air stream for efficient operation of the permeation devices.

2. Removal of 90% of the radon in a staged cascade of membrane permeators.

TABLE 16. RETENTION OF RADON IN SILICONE RUBBER MEMBRANES

Feed = 5000 CFM (see Table 11)

Stage	Weight (g x 10^{12})	Activity (μCi)
1	2.95	0.454
2	3.19	0.492
3	2.41	0.371
4	1.76	0.271
5	1.21	0.186
6	0.741	0.114
7	0.342	0.053

Feed = 2000 CFM (see Table 10)

Stage	Weight (g x 10^{12})	Activity (μCi)
1	1.18	0.182
2	1.28	0.197
3	0.964	0.148
4	0.705	0.108
5	0.484	0.075
6	0.296	0.046
7	0.137	0.021

TABLE 17. RETENTION OF RADON IN POLYETHYLENE MEMBRANES

Feed = 5000 CFM (see Table 14)

Stage	Weight (g x 10^{10})	Activity (μCi)
1	1.58	24.3
2	1.67	25.7
3	0.560	8.62
4	0.155	2.39

Feed = 2000 CFM (see Table 13)

Stage	Weight (g x 10^{10})	Activity (μCi)
1	0.633	9.73
2	0.668	10.30
3	0.224	3.45
4	0.062	0.96

A system to process 5000 SCFM of air would cost $1,410,000. The total annualized cost would be about $493,000. On a 4150 hours per year (16 hrs/day, 260 days/year) basis, the operating cost of this system is about 39.5¢ per 1000 SCF of air.

Utilities requirements for a 5000 SCFM system are as follows:

Water	340 GPM
Power	2091 KW

Capital cost estimates were made using the best of the three following methods:

- Vendors' quotes or estimates
- Cost-capacity relations in the indicated literature
- Lange factors

Total plant capital cost is estimated at $1,444,000. (Refer to Table 18.)

Annualized cost estimate was based on the following:

- 5 year straight line depreciation
- Taxes and insurance at 1.5% of capital
- Water at 5¢/1000 gals and power at 1¢/kwh
- Labor at $5.00/man hr

Total annualized cost is estimated at 40.2¢ per 1000 SCF of air. (Refer to Table 19.)

TABLE 18

TABLE 18. MEMBRANE PERMEATION

CAPITAL COST -- 5000 SCFM UNIT

Equipment	Size	Installed Cost
Inlet Air Filter	5,000 SCFM	$ 2,500
3 Compressors	2800 HP Total	1,012,000
3 Permeators	685,000 ft^2 Total	123,500
Humidifier		10,000
		$ 1,256,000
Engineering, Overhead, Fee & Contingency (15%)		188,000
Total Capital Cost		$ 1,444,000

TABLE 19. MEMBRANE PERMEATION

ANNUAL OPERATING COSTS

(Capital Cost $1,444,000)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/Yr	$ 20,000
Utilities			
Electricity	8.7×10^6 kwh	$0.01/kwh	87,000
Water	85,000 M gal	$0.05/M gal	4,200
Supplies			1,000
Maintenance	5% of Capital Cost		72,200
Direct Operating Cost			$184,400
Capital Charges	22% of Capital Cost		317,700
Total Annual Operating Cost			$502,000
Total Cost per MSCF			40.2¢

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1. Li, Norman N. (ed.), Recent Developments in Separation Science (Vol II), The Chemical Rubber Co., Cleveland, Ohio, 1972, pp. 129-130.

2. Stern, S.A., et al., "Helium Recovery by Permeation," Industrial and Engineering Chemistry, Vol. 57, No. 2, February 1965, pp. 49-60.

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D. MOLECULAR SIEVES (Ambient Temperature)

1. Description of a Preferred System

The term "Molecular Sieve" is a descriptive expression that was coined by J. W. McBain[1] in 1926 to characterize the behavior of certain naturally occurring zeolites - solid hydrated aluminosilicates - in adsorbing molecules of certain size and/or polarity from fluid vapor or liquid mixtures. According to C. K. Hersh[2] about forty natural crystalline zeolites are known, and, thanks mostly to the interest of the Linde Company, Division of Union Carbide Corp., a number of synthetic crystalline zeolites are now available for industrial use. They are generally known by the designation: Types 3A, 4A, 5A, 13X, which were originally patented proprietary products of the Linde Company. These and others are manufactured by several companies for separation of gases or liquids and are particularly effective for certain kinds of substances.

The mechanism by which molecular sieves operate combines surface adsorption, either chemical or physical, with a fixed and uniform pore size which is of molecular dimensions. This mechanism results in a preferential adsorption of polar (and therefore readily adsorbed) molecules and of atoms and molecules of relatively small dimensions. Thus, it is well known that traces of H_2O can be removed very effectively from various gases such as argon by the use of molecular sieves, and that traces of low-molecular weight straight-chain hydrocarbons can be removed from branch-chained isomers because the slightly smaller molecular volume of the linear alkanes allows them to be contained within the pores of the zeolite structure. For removal of traces of radon from air, however, the special advantage of the molecular sieve, namely its selectivity for small atoms and molecules in the presence of larger ones, cannot be used, since an atom of radon is larger in diameter than a molecule of N_2, O_2, or H_2O. Thus, any molecular sieve whose pore size is large enough to accommodate radon atoms will also trap these other impurities. The result is that the molecular sieve must be made to rely only on its ability to physically adsorb the radon in preference to oxygen, nitrogen, water and various impurities. Under these circumstances its adsorptive performance must be compared directly with that of any other solid adsorbent, especially charcoal, and in that comparison it is definitely inferior to charcoal.

The system was designed to remove 95% of the radon present in mine air at 100°F, 100% relative humidity and 15 psia. In order for the molecular sieve to adsorb a significant quantity of radon, the temperature and humidity must be low. Thus, as indicated on the attached schematic flowsheet, the air is first filtered, compressed, and cooled. This removes approximately 71% of the water in the air. The remaining water vapor is removed by two parallel beds of type 5A molecular sieve, each around 4 feet in diameter and 4 feet long. The large heat of adsorption of water on molecular sieve (1800 Btu/lb.) necessitates cooling the air after it leaves the drying beds. The cool, dry air can now be fed to the main molecular sieve adsorption beds.

After the radon is adsorbed, the clean dry ventilation air passes through a packed bed of 1" Raschig rings 6 feet in diameter and 2 feet thick in order to rehumidify the air before blowing it to the mine face.

After one hour of operation, the system is saturated with water and radon, so the air is switched to a duplicate system while the beds are regenerated in a thermal-swing cycle. During the first part of the regeneration, the drying and radon-adsorbing beds are heated to an average temperature of 225°F. Two separate compressors are required for this. To heat the beds, air is heated electrically to 250°F, blown through the beds, cooled, and exhausted to the haulageway. This heating step takes 50 minutes for the drying bed and 30 minutes for the radon-adsorption bed. To cool the adsorbent, closed air circuits are set up around the beds with heat exchangers at each end of the beds to cool the air coming from the compressors and to cool the air coming from the beds. It takes about 30 minutes to cool the radon-adsorption bed and 10 minutes for the drying bed. These regeneration steps are outlined on the schematic flow sheet. A schematic arrangement of the location of the entire system is shown in Figures 18 and 19.

2. Efficiency and Reliability

a. Total Concentration of Radon Retained

There do not appear to be published data readily available on the adsorption of radon on molecular sieve materials. We can extrapolate, however, based on the available data on charcoal, using proprietary data obtained in our own laboratories for krypton adsorption on charcoal and on molecular sieve (13X and 5A types). These suggest that the dynamic breakthrough capacity (volume of carrier gas resulting in krypton breakthrough, per gram of adsorbent) is about 20 times lower on molecular sieve than it is on charcoal. As a rough approximation, we shall assume this to be true for radon as well. In the absence of further data, we shall also assume the effect of water vapor on molecular sieve adsorption to be the same as in the case of charcoal adsorption.

The temperature range of interest after precooling is 50°F to 75°F. Radon retention would be essentially quantitative (>99%) as long as we remain below the "breakthrough" volume. Breakthrough would be expected to occur after passage of 0.2 liter of gas/gram of Molecular Sieve (at 75°F) to 0.4 liter of gas/gram of Molecular Sieve (at 50°F). These figures allow for 100% relative humidity; reduction to 70% relative humidity might increase the above breakthrough volumes by 30%. These breakthrough volumes are unaffected by changes in radon concentration, air flow rate or velocity, aerosol particles, or CO, CO_2, NO_x, and SO_2 contents in the ranges given.

With the above in mind, the concentration of Rn retained on the molecular sieve bed at saturation would be as shown in Table 20.

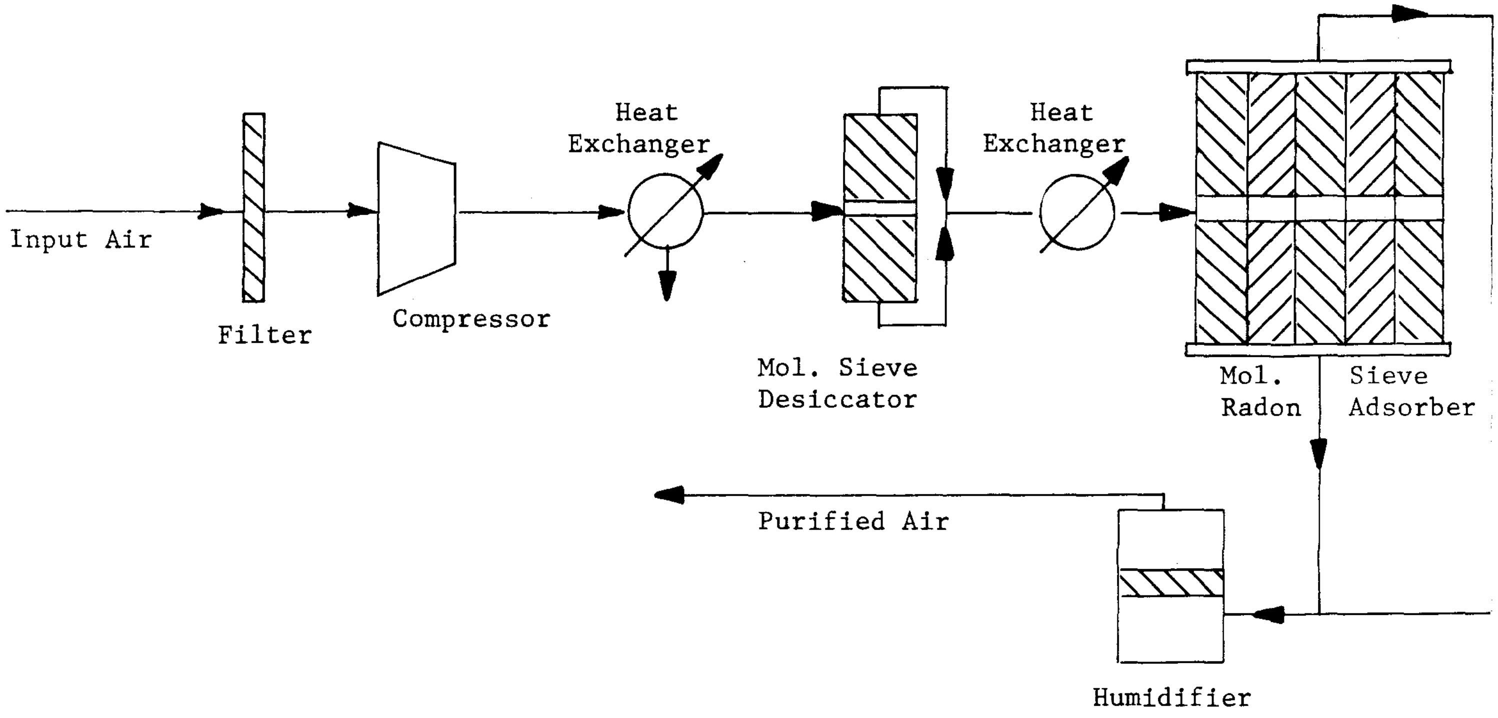

FIGURE 18. Schematic Arrangement of One Half of Molecular Sieve Radon Adsorption System.

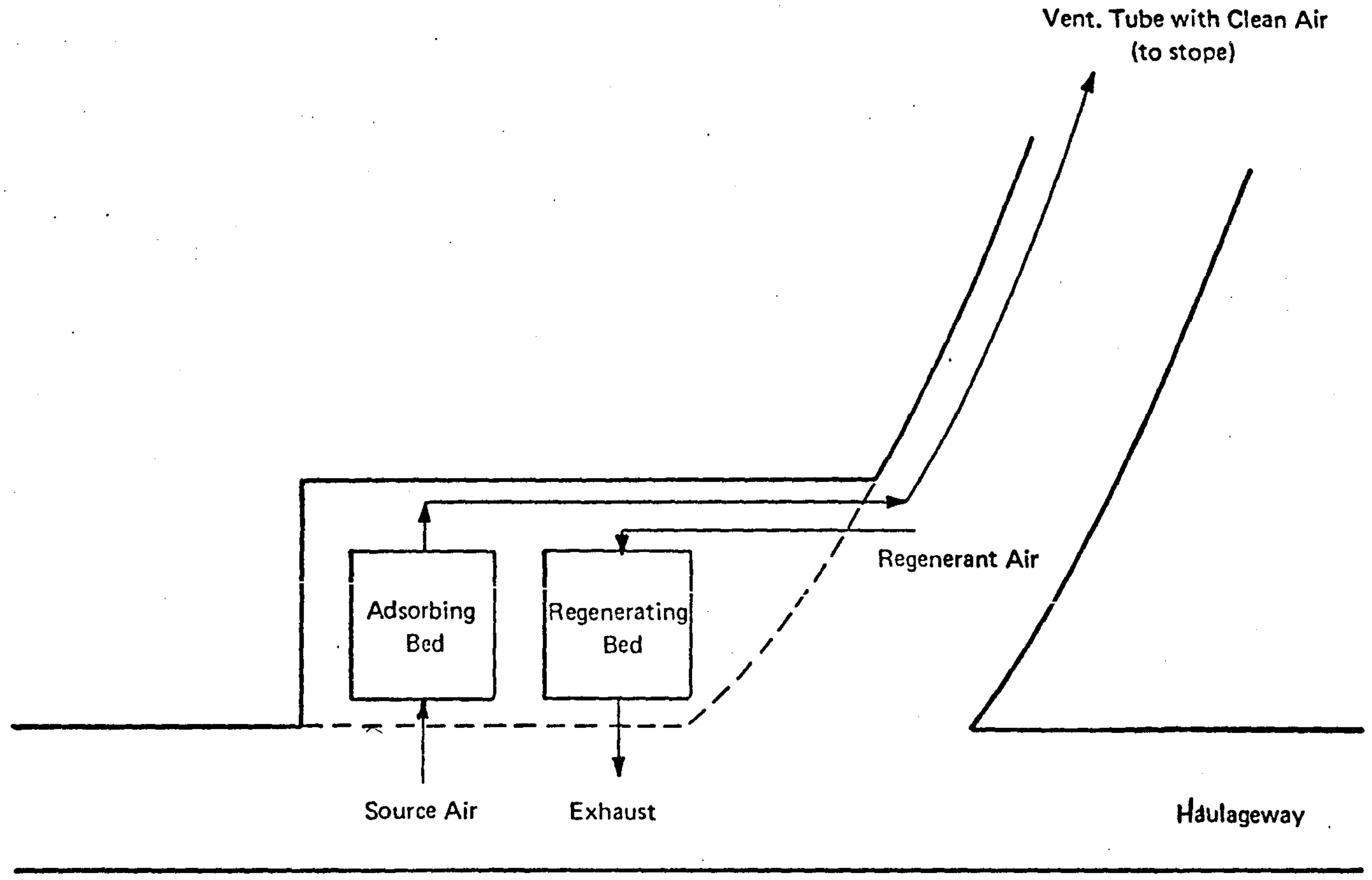

FIGURE 19. LOCATION OF RADON ADSORBER UNIT

TABLE 20

ESTIMATED RADON RETENTION ON MOLECULAR SIEVE

Feed Concentration	Feed Temperature	Rn Retention at Saturation*	
		70% Relative Humidity	100% Relative Humidity
30 pCi/l	50°F	16. pCi/g Sieve	12. pCi/g Sieve
	75	8.	6.
500	50	260.	200.
	75	130.	100.

* The above figures are for 15 psia. If the air pressure were 10 psia, then the retention concentrations would be 10/15 as large as the figures in the table.

b. Weight, Volume, and Width of Device

For the "worst" case - 5000 CFM of 100°F, 100%RH air at 15 psia - we estimate a requirement of two sets of beds, each containing 46 tons of molecular sieve.

In order to maintain a reasonably small pressure drop through this large quantity of granulated solid matter, each of the two alternating beds should be divided into ten smaller beds, all connected in parallel. Each of the smaller beds would be a cylinder six feet in diameter and ten feet long. One additional Molecular Sieve bed of similar dimensions will be required for each of the two alternating bed arrays, making a total of 22 vessels, 6' diam. x 10' long, with a total weight of 268,000 lbs. Eight heat exchangers, each one four feet in diameter and 12 feet long, weighing a total of 58,000 lbs, will make up the remainder of the principal mass and volume of the equipment. The total system will occupy a space 12' x 20' x 70' = 16,800 ft^3 and have a total mass near 350,000 lbs.

c. Residence Time of Radon and Air

The residence time ("holdup") of Rn in the beds is designed to be 1 hour; that of air is about 30 seconds.

d. Rate of Deterioration

The adsorbent should last through thousands of cycles without serious decrease in performance; the temperature during desorption will, however, have to be higher than it would be for charcoal, say 250°F. This material is more difficult to regenerate.

e. Possible Recycling Processes

If it is desired to dispose of the adsorbent in a bed, it could be safely buried.

3. Safety

a. Possible Oxygen Removal Rates

There are no significant safety hazards associated with this design. The molecular sieve is non-toxic, non-flammable, and will not adsorb a significant fraction of atmospheric oxygen.

b. Noxious or Toxic By-Products

None.

c. Maximum Expected Noise Levels

The air compressors are the major source of noise and they should be acoustically insulated.

d. Possible Storage, Use and Maintenance Hazards

Because of the fact that the molecular sieve bed accumulates radon continuously during the one-hour adsorption portion of the operating cycle, some decay to gamma-emitting short-lived daughter products will occur until the regeneration or "back-flushing" operation removes the accumulated radon and discharges it to a waste air stream. Exactly the same situation occurs in the use of a charcoal adsorption bed that is operated on a one-hour-on, one-hour-off cycle, and the accumulation of gamma-emitting decay products was predicted for that situation in Section VA3 above. For a molecular sieve system only the mass and volume of the adsorbent bed are different, since each of the two parallel beds contains 46 tons of molecular sieve in an array measuring about 30' x 12' x 10'. Table 20 above shows that as much as 260 pCi of ^{222}Rn per gram of sieve might be accumulated from a 5000 SCFM air stream having an initial Rn concentration of 500 pCi/1. Since 46 tons = 4.2×10^7g, the total ^{222}Rn burden of the bed would be 10 mCi at the end of each one-hour adsorption cycle. More than 90% of this is removed during the regeneration process, and the residue decays <u>in situ</u>, leading to an average concentration of RaB and RaC of 2.6 mCi and 2.4 mCi or a total of 5.0 mCi of gamma-emitters respectively, by the end of the third complete cycle of operation. If concentrated at one point and not shielded, this would result in an absorbed dose of about 44 mr/hr at a distance of one foot (30 cm). Inasmuch as 46 tons of molecular sieve not only provides a generous amount of self-shielding but also distributes the radiation source widely throughout the entire volume of the the bed, the system is judged to be substantially free of radiation hazard. It remains only to be noted that these calculations apply to an input radon concentration of 500 pCi/1. Very much higher concentrations encountered over periods of many hours or days could conceivably produce a small radiation hazard, but the possibility of a major hazard seems remote.

e. Remedial Measures

None would seem to be needed.

4. Cost

Capital costs are outlined in Table 21 as estimated by conventional equipment-factoring cost procedures. Total plant capital cost is estimated to be $1,234,000.

Annual operating costs are outlined in Table 22. Most per-unit rates are outlined in the table. Electric power requirements total 1365 Kw including pumps, compressors, and electric air heaters. Total cooling water consumption is around 563,000 gallons/day based on 60°F ambient water temperature. Due to thermal cycling, it is suggested that the molecular sieve material be replaced every four years. Total annual operating costs are around $468,800 per year or 37.6¢ per 1000 SCF of air processed.

TABLE 21. MOLECULAR SIEVE ADSORPTION (Ambient Temperature)

CAPITAL COST 5000 SCFM UNIT

Equipment	Size	Installed Cost
Air Filter	5000 SCFM	$ 2,000
3 each Compressors	825 Hp total	330,000
8 each Heat Exchangers	20,400 ft^2 total	250,000
Electric Heaters	1200 KW	25,000
Molecular Sieve Beds	---	250,000
Humidifiers	---	10,000
Type 5A Sieve	187,000 lbs.	206,000
		$1,073,000
Engineering Overhead Fee & Contingency (15%)		161,000
TOTAL		$1,234,000

TABLE 22. MOLECULAR SIEVE ADSORPTION (Ambient Temperature)

ANNUAL OPERATING COST

5000 SCFM 2 Shifts 260 days/year

(Capital Cost $1,234,000)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/yr	$ 20,000
Utilities			
Electricity	54.6×10^6 KWH	$0.01/KWH	54,600
Water	140,000 m gal	$0.05/m gal	7,000
Mine Supplies			3,800
Maintenance	4% of capital cost		49,000
Molecular Sieve			62,500
Direct Operating Cost			$197,300
Capital Changes	22% of capital cost		271,500
TOTAL ANNUAL OPERATING COST			$468,800
TOTAL COST PER MSCF			37.6¢

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E. CRYOGENIC CONDENSATION

1. Description of a Preferred System

a. Introduction

Radon is a heavy monatomic noble gas, and because of this it can be expected to conform quite strictly to physical and thermodynamic principles applicable to "ideal" gases, liquids and solutions. This is fortunate, because although very few measurements are available for this rare substance, it is nevertheless possible to calculate its thermodynamic properties with considerable confidence.

The normal boiling point of pure radon is 211°K (-62°C). Its vapor pressure has been measured to the critical point (377.2°K, 62.4 atm)[1], and from these measurements we are able to derive the heat of vaporization from the Clapeyron-Clausius equation in the form:

$$\frac{d\ln p}{dt} = \frac{\Delta H}{RT^2}$$

in which p is the vapor pressure at temperature T, and ΔH is the heat of vaporization. In integral form, this becomes:

$$\ln p = c - \frac{a}{T}$$

The two constants c and a have been calculated from the vapor pressure data between -10.3°C and -70.5°C resulting in C - 9.722 and a = 2053. ΔH is calculated to be 4.1 kilocalories per mole.

Radon in air having a radiation level of 30 pCi per liter would have a concentration of 5×10^5 atoms per liter. This is equivalent to a mole fraction of 2×10^{-17}. Using the vapor pressure formula above, we can find the dew point (temperature for condensation of Radon liquid from a gas having a partial pressure of 2×10^{-17} atm.); and it calculates to be 42.5°K. This means, of course, that radon cannot be separated out as a liquid from such a dilute air mixture because the air itself will liquefy at the much higher temperature of 81. °K, and the radon will condense along with it. We see from this that a purely cryogenic process for removal of radon from air requires at least partial liquefaction of the air, and we can immediately conclude that such a plant will be very similar to a standard air separation plant, the only important difference being that we have no need to separate the oxygen from the nitrogen as is done in an air separation plant.

b. The Cryogenic Process

The basic processes required for air liquefaction and revaporization are shown diagramatically in Figure 20.

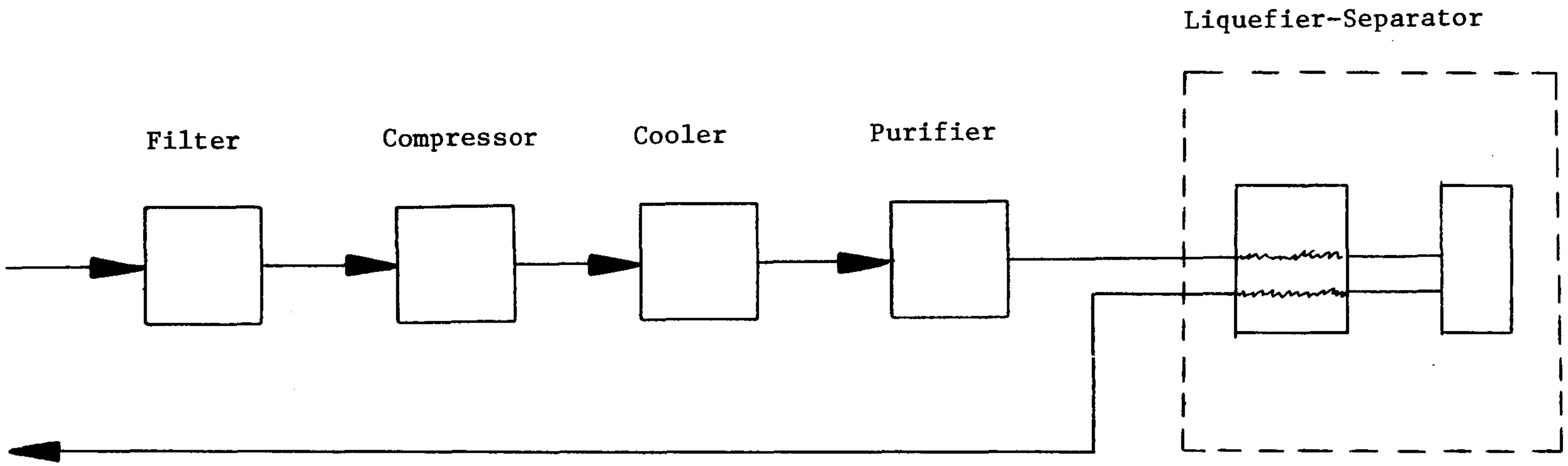

Figure 20. Basic Cryogenic Process

If the mine air contains an appreciable amount of dust, it will be necessary to pass it through a filter before it enters the compressor. After compression, the air must be cooled back to room temperature using either water or air to remove the heat of compression. The amount of heat rejected is very substantial, and the choice of ambient air or circulated water will have to be made after careful consideration of local environmental factors. After cooling, the compressed air must be treated to remove any condensible contaminants such as H_2O and CO_2; otherwise, these would accumulate as solids in the cryogenic plant and block flow passages. Alternatively, reversing heat exchangers may be used in which untreated compressed air is cooled down in countercurrent flow to the returning cold air. Water and carbon dioxide are frozen out on the heat transfer surfaces of the passages; but before there is too much accumulation, the ingoing and outgoing streams are switched so that the passage containing the H_2O and CO_2 deposits is now exposed to returning purified air, and the solids reevaporate into the low-pressure air stream leaving the plant. The two flow streams are switched back and forth sufficiently frequently to avoid any long-term accumulation of impurities in either of the passageways.

Two alternatives are available in the design of the liquefier: either

1. it completely liquefies all the input air before admitting it to the reboiler, where the oxygen and nitrogen are continuously boiled away (leaving the radon behind), or

2. the input air is compressed and admitted as fine bubbles (sparging) to a pool of previously liquefied air into which the radon condenses.

The latter technique, which is based upon intimate contact between vapor and liquid by means of sparging, is shown schematically in Figure 21. The advantage of the sparging scheme over the continuous liquefaction and reboiling method is that it requires compression of the gases to only two atmospheres, while complete liquefaction requires further compression to four atmospheres.

Cooled, compressed air enters the plant and is further cooled to the dewpoint in the reversing heat exchanger. It then enters the sparger tank with the boiling liquid which surrounds them. As we shall see later, most of the radon is retained in the reboiler liquid in the steady state, whereas the oxygen and nitrogen are continuously boiled away. The revaporized air leaves the reboiler at the established pressure, is warmed up a few degrees in a non-reversing passageway in the heat exchanger and is then expanded adiabatically through a cold expansion turbine to produce the required refrigeration for the plant. This air, almost free of radon contamination, leaves the plant through the reversing heat exchanger and is warmed to room temperature by the incoming compressed air.

An air separation plant, with no net output of liquid products, requires an expansion ratio of approximately two-to-one in order to produce sufficient refrigeration to balance the heat leakage into the plant and to make up for inefficiencies of the heat exchanger. Since the expander exhaust pressure is approximately one atmosphere (absolute), the inlet pressure will be approximately 2.0 atmospheres. The expander exhaust is at the saturation temperature (8.18°K); and if the expander efficiency is taken as 85% (ΔH actual/ΔH ideal x 100), the inlet temperature will be 97.3°K. The expander inlet gas must have the composition of pure air because it is the only stream leaving the plant. Since this vapor results from boiling liquid in the reboiler at 2 atmospheres' pressure, vapor-liquid equilibrium data for oxygen-nitrogen mixtures requires the composition of the liquid to be 50% O_2 - 50% N_2, boiling at a temperature of 88°K. This is also the dewpoint of normal air at 2.0 atmospheres.

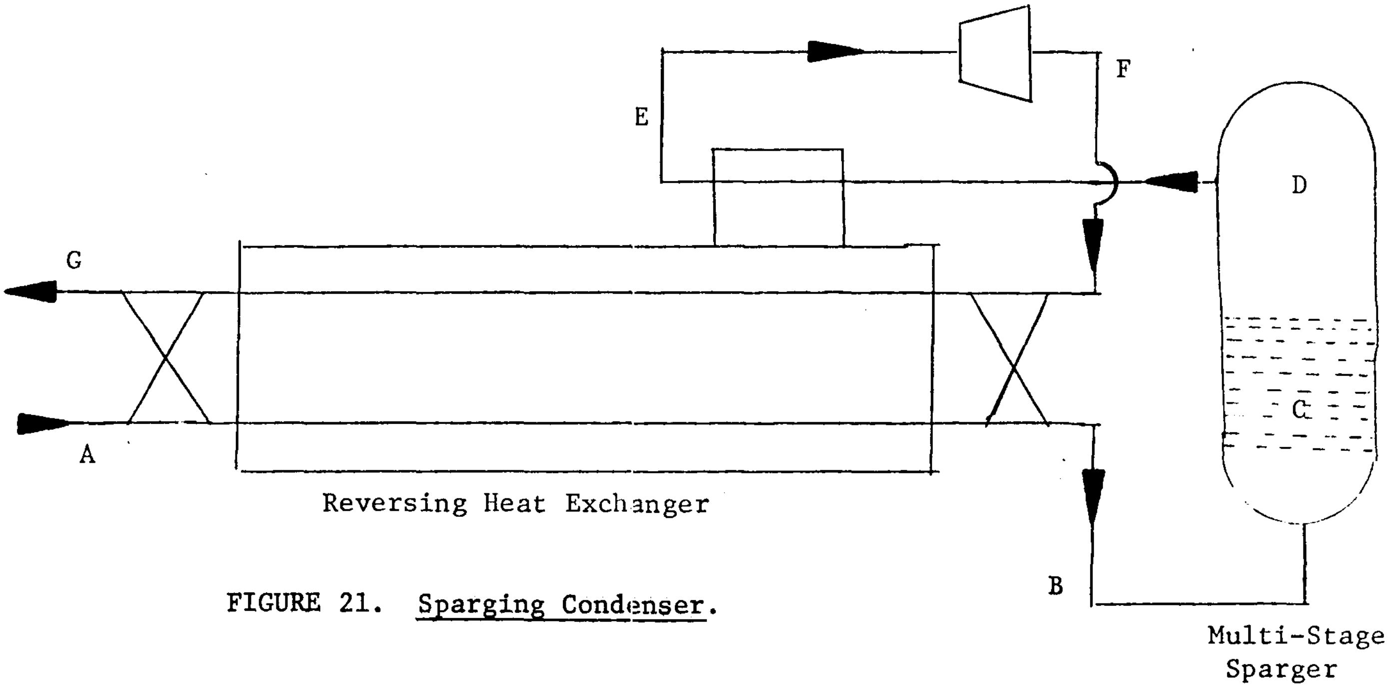

FIGURE 21. Sparging Condenser.

Location	Phase	Pressure	Temp.	Composition
A	Gas	2.0 atm.	300°K	21% O_2
B	Gas	2.0 atm.	88°K	21% O_2
C	Liq	2.0 atm.	88°K	50% O_2
D	Gas	2.0 atm.	88°K	21% O_2
E	Gas	2.0 atm.	97.3°K	21% O_2
F	Gas	1.0 atm.	81.8°K	21% O_2
G	Gas	1.0 atm.	295°K	21% O_2

In order for a sparger to be as effective as a total-condensation-reboil process it would have to achieve complete vapor-liquid equilibrium. Sparger designs (for example, a typical bubble plate or sieve plate of a conventional distillation tower) as a rule of thumb, can be expected to have an efficiency of 90%. That is, a liquid and vapor subjected to contact through a conventional sparger plate will have their compositions altered toward equilibrium to the extent of 90% of the total theoretical composition change. A second pass will result in 99% and a third will promote a vapor-liquid composition within 0.1% of equilibrium. It is evident, therefore, that a well designed multiple plate sparger can be operationally equivalent to the total-condensation-reboil process with considerable saving in cost.

c. Entrapment of Radon

Radon present in the air will, of course, condense in contact with the liquefied air in the condenser reboiler. Most of it will remain in the liquid phase in the reboiler and will accumulate there unless otherwise removed. As it accumulates, the concentration will rise to a point where, eventually, the rate of radioactive decay of Rn will equal the rate of arrival minus the rate at which it escapes in the boiled-off air.

$$\text{Rate of arrival} = pA \text{ moles per day} \qquad (1)$$

where A is moles of air per day, and p is the partial pressure of Rn in the entering air.

$$\text{Rate of escape} = p^{*}A \text{ moles per day} \qquad (2)$$

where p^{*} is the equilibrium partial pressure of Rn in the boiled-off vapor leaving the plant. The decay half-life of Rn is 3.8 days; therefore, a steady-state concentration x^* will develop in the reboiler liquid which is calculated as follows. At any given time, t, the Rn concentration, is x, the rate of decay is:

$$\frac{dx}{dt} = -kx \tag{3}$$

For a half-life of 3.8 days, k is found to be $k = 0.182 \text{ day}^{-1}$. A steady state will be established when the net rate of addition plus the decay rate equals zero.

$$M\left(\frac{dx}{dt}\right)_{x = x} + A(p-p^*) = 0 \tag{4}$$

where M is the number of moles of liquid air held in the reboiler, and A is the number of moles per day of air circulated. Combining (3) and (4) leads to:

$$x^* = \frac{A(p-p^*)}{Mk} \tag{5}$$

Henry's law states that:

$$p = xp_o \tag{6}$$

where p is the partial pressure of Rn in the vapor phase; x is the mole fraction in the liquid phase; and p_o is the vapor pressure of pure liquid Rn at the temperature of interest. Using the vapor pressure equation for pure Rn derived in the Introduction section, we calculate p_o for the reboiler temperature (88°K) to be $p_o = 1.2 \times 10^{-6}$ atm. Hence, from (6):

$$p_{eq} = x^* \times 1.2 \times 10^{-6} \text{ atm.}$$

where p_{eq} is the equilibrium partial pressure of Rn in the reboiler vapor. This indicates that the mole fraction of Rn in the liquid phase is a million times as great as that in the gas phase, a not unexpected result, but completely at variance with the subject matter of a recent United States patent.[2]

Since the total pressure in the reboiler is 2 atmospheres, $p_{eq} = 2\,p^*$ because p^* is the partial pressure of Rn in the air leaving the plant after having been expanded to one atmosphere pressure. Therefore,

$$p^* = x^* \times 0.6 \times 10^{-6} \text{ atm.} \tag{7}$$

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2. U.S. Patent 3606760 - Method of Purifying Radon Containing Air.

Combining (7) and (5) and substituting the numerical value 0.182 day^{-1} for k:

$$E = \frac{p-p^*}{p} = \frac{M}{M + 3.3 \times 10^{-6} A} \tag{8}$$

This equation is an expression of the effectiveness of removal of radon from air by the cryogenic process. The fraction of radon removed is seen to be independent of the degree of contamination p, and depends only on the rate of flow of air A and quantity of liquid held in the reboiler M.

Consider a plant which processes 5,000 SCFM of air containing 500 pCi/l (3.3×10^{-16} atm.) of Rn. A reasonable reboiler volume would be 100 liters and would contain 5.5×10^4 moles. Under these circumstances, the fraction of radon removed can be calculated from Equation (8):

$$E = \frac{p-p^*}{p} = 0.99 \tag{9}$$

Combining this result with Equation (5) leads to:

$$x^* = 5 \times 10^{-12} \tag{10}$$

This is the equilibrium (steady-state) concentration of Rn in the 100-liter volume of reboiler liquid. It corresponds to a concentration of 1.7×10^{-10} moles per liter and to a local radiation level of 5.6 mCi/l.

The rate of buildup of radon in the liquid phase can readily be calculated from the fact that the rate of buildup,

$$M\frac{dx}{dt}$$

at any given time is the sum of the addition rate and decay rate. Using the same notation as before,

$$M\frac{dx}{dt} = (p-p^*)\ A - Mkx$$

p^* may be omitted because it has a value of zero at the beginning and never exceeds a value of 0.01 p even when $t \to \infty$.

$$\frac{dx}{dt} = \frac{A}{M}p - kx$$

$$\frac{dx}{\frac{A}{M}p - kx} = dt$$

Upon integration,

$$\ln\left(\frac{A}{M}p - kx\right) = -kt + C$$

where C is a constant of integration. C is determined from the fact that $x = 0$ at time zero:

$$C = -30.77$$

We now calculate the time required to build up the radon concentration to a value of 90% of the steady-state value which occurs for $t \to \infty$:

$$t_{90\%} = 12.6 \text{ days}$$

In summary, it appears that the cryogenic method of radon removal, in which radon-containing air is liquefied and revaporized, is technically feasible. The effectiveness of the process is excellent. Approximately 99% of the Rn is removed and is entrapped in a reservoir of liquid air. The Rn content in the liquid builds up over a period of approximately two weeks to a steady-state equilibrium concentration of approximately 1.7×10^{-10} moles/liter of liquid, corresponding to a radioactivity level of 0.56 Ci per 100 liters of liquid.

The steady-state performance for radon removal is shown in the following table for a number of flow rates and contamination levels. Actually, at any given time early in a run, or for runs of short duration (say 8 hours) the performance is much better than indicated in the table, because the concentration of Rn in the liquid is less than the steady-state value.

Operating Performance

Flow rate air-SCFM	Radon Content pCi/l	Liquid Volume liters	Removal Efficiency % - Removed	Radon Content effluent air pCi/l
5000	30	100	99%	<0.3
5000	100	100	99%	<1.0
5000	500	100	99%	<5.0
2000	500	100	99.6%	<2.0
2000	30	100	00.6%	<0.2

d. Reversing Heat Exchangers

The main purpose of the reversing heat exchangers is to cause the returning cold air to absorb heat from the incoming compressed air thereby cooling it progressively from room temperature to the liquefaction temperature as a means of conserving refrigeration. As the compressed air is cooled condensable impurities such as water vapor, carbon dioxide, sulfur dioxide and oxides of nitrogen will condense on the heat exchange surface and gradually accumulate. Before there is sufficient accumulation to impede flow, however, the low pressure stream is switched into the passageway which had been carrying the impure compressed air and vice versa. Since the low pressure returning air has a greater volume at any given temperature its evaporative capacity is sufficiently great to evaporate all the impurities deposited in the previous half-cycle. This greater-than-needed evaporative capacity is essential for successful operation; otherwise there will be gradual accumulation of solids which will eventually block the passageways and interrupt operation. The purpose of the third channel on the cold end of the reversing exchanger is to cool down the portion of the exchanger where solid CO_2 deposits so as to assure complete removal.

Traces of carbon monoxide are not condensed in the heat exchanger and will cause no difficulty in the proposed process. Methane, if present, would tend to accumulate in the sparger liquid since its boiling temperature is approximately 30°K higher than that of air. However,the volumetric concentration in the liquid would rise only to approximately ten times that in the gas phase after which equilibrium would occur and it would then boil off at the same rate as it arrives. Consequently small concentrations would cause no foreseeable problem.

Trace amounts of acetylene (and possibly other unsaturated hydrocarbons) have caused severe explosions in cryogenic air separation plants. It is thought that traces of acetylene too dilute to freeze out in the heat exchangers arrive at the distillation column and accumulate in the reboiler liquid. Eventually solid crystals separate out of the liquid oxygen and are sensitive to detonation by liquid turbulence. It is not known if solid acetylene would explode in the sparger liquid (50% O_2, 50% N_2), but in any event, the accumulation of acetylene can beprevented by circulating a small side stream of the liquid over a suitable molecular sieve to adsorb out any acetylene present. These adsorbers are usually used in tandem so that one is in service while the other is warmed up for reactivation.

2. Efficiency and Reliability

The Cryogenic Process, as a separator of radon from air, is highly efficient. We have seen from a previous section that with a reasonable choice of design parameters, the separation efficiency may be as great as 99%. Also with a reasonable set of parameters the radon may be retained in continuous operation for an indefinite period of time because the rate of decay will become equal to the rate of entrapment. Since it is not a cyclical process there is no radon disposal problem except in the event of a complete plant warm-up. When that occurs the residual liquid could be drained into an auxiliary thermally insulated vessel and disposed of outside the mine.

The reliability of the plant should be excellent. Air separation plants of similar design have operated for many years with essentially total reliability and with only routine maintenance. Since the only major moving parts are the electric motor, a centrifugal compressor and an expansion turbine, a plant constructed to this design should have a reliability equivalent to a large industrial air conditioning installation.

a. Power Requirement

Almost the entire energy requirement of the cryogenic radon removal plant will be the power required to compress air to 2.0 atmospheres absolute. The energy will have to be supplied by electric power since it is not reasonable to burn fuel in the confined workings of the mine; moreover, the heat rejection would probably be prohibitive. The thermodynamic work required to compress air from p_1 to p_2 is given by:

$$W = RT_1 \left(\frac{\gamma}{\gamma - 1}\right) \left[\left(\frac{p_2}{p_1}\right)^{\frac{\gamma - 1}{\gamma}} - 1\right] \text{ cal/mole}$$

where R is the gas constant, T_1 is the inlet temperature in degrees Kelvin, and γ is the ratio of the constant-pressure to constant-volume heat capacity for air. In this particular case, $p_2 = 2$, $p_1 = 1$, $T_1 = 288°K$ (60°F), $\gamma = 1.41$, and $R = 1.987$ cal/°K/mole.

$$W = 439 \text{ cal/mole}$$

A flow of 5000 SCFM corresponds to 0.379×10^6 gram-moles per hour. Therefore the work of compression is:

$$W = 166 \times 10^6 \text{ cal/hr}$$

which is equivalentto 259 hp. Assuming the compressor is 80% efficient, the shaft power is <u>324</u> bhp and the motor would require <u>240</u> kilowatts of electrical power.

The expansion turbine will produce a net power output which can be calculated in the same manner as the work of compression. Alternatively, the enthalpy change in the air can be obtained from an entropy-temperature diagram by carrying out an adiabatic expansion between two and one atmospheres resulting in the expanded gas reaching the dew-point temperature. Using either method, and assuming 80% efficiency of the turbine, we find:

$$W = 53 \text{ bhp.}$$

This power output is available for operating an electrical generator or auxiliary pumps or blowers as needed. As a practical matter, however, we believe the extra power will be needed for one purpose or another and it would probably not be wise to plan to use the expander power to reduce the power required by the compressor motor. Hence, we will proceed on the estimate that the net electrical power requirement of the plant is 240 Kw.

b. Cooling Requirement

All of the heat of compression must be removed in the cooler. If air is used as the cooling medium and if we assume an initial temperature of 60°F and a final temperature of 80°F, the circulation rate would be 40,000 SCFM. Clearly, this air would have to be brought in from the outside and continuously discharged from the mine in order to maintain an ambient temperature below 80°F. However, if such a large flow of fresh air was to be made available for cooling purposes, it would more than suffice (by almost an order of magnitude) the ventilation requirement of 5,000 SCFM. Therefore, it appears that air cooling of a cryogenic plant would be completely unreasonable.

If the plant is to be water-cooled, a source of water would have to be piped in and discharged outside the mine. Assuming again that the water inlet temperature is 60°F, and allowing for more favorable heat transfer conditions for water, a discharge temperature as high as 100°F may be tolerable. Under these circumstances, a flow rate of 45 gallons per minute would be required. If a more elaborate, counter-flow, water cooler is used, a higher water discharge temperature could be achieved (120°F) thereby reducing the water requirement to 30 gallons per minute.

3. Safety

The safety record for air separation plants of design very similar to the proposed radon separation plant is excellent. Over the past three decades, hundreds of these plants have been put into service all over the world with no known serious accidents reported. Very early in the history of development, there were several minor explosions in the liquid oxygen reboiler section of the distillation column which have since been attributed to the presence of trace amounts of acetylene. In the radon separation plant, the highest concentration of oxygen which occurs is 50% and it is not known whether or not solid acetylene crystals would explode under these conditions. In any event, a hydrocarbon adsorber appended to the reboiler section of the sparger would solve the problem. The possibility of accumulating other impurities is discussed in the section on Reversing Heat Exchangers and the conclusion is that no hazard exists.

The most serious hazard which can be identified arises from the fact that the plant in steady-state operation contains 100 liters of liquid air contaminated with 3.4×10^{10} pCi of radon for mine air containing 30 pCi/l, and as much as 5.5×10^{11} pCi of radon for mine air containing 500 pCi/l. Any accident which results in sudden spillage of this liquid would cause a rapid release of radon to the local atmosphere. On the other hand, it is difficult to visualize any accident (barring a chemical explosion in the sparger) which would cause mechanical disruption of a simple metal tank. If the hazard is judged to be intolerable,,however, the sparger could be placed within a separate containment vessel, both of which would be surrounded by thermal insulation. This would delay the evaporation of spilled liquid for several hours and greatly reduce the hazard of sudden release of gaseous radon.

Accidental release of the stored radon could conceivably happen without any malfunction of the equipment or rupture of the liquid container. If the plant is shut down intentionally and is not put back into operation within a specified time, the 100 liters of stored liquid will evaporate and the last trace of liquid to evaporate will release the entire radon content. The critical time lapse between shutdown and restart will depend on several design parameters, the most influential of which are the quality of thermal insulation and the volume of liquid stored. If, for example, it is decided to operate the plant for only one eight-hour shift per day, it will be necessary to design a sparger with insulation capable of preserving the liquid charge for at least 16 hours of shut-down time. This can certainly be done, but it may require vacuum insulation and higher cost of construction. Moreover, the operating costs (per 1000 SCF of air processed) will be much higher than if the plant were to be used in continuous service.

An instantaneous overpressure of 60 psia poses two distinctly different types of hazards to the cryogenic radon separation plant. The first of these applied whether or not the plant is in operation and is simply the hazard of collapsing one or more of the sealed or semi-sealed vessels comprising the physical plant. Design features to mitigate this hazard are to provide blow-in check valves for all enclosures which need not be hermetically gas-tight, such as cold-box enclosures, and to provide adequate structural strength for gas-tight vessels, such as the sparger vessel, so that the collapse pressure will exceed 60 psia.

If an overpressure of 60 psia were to occur during operation, the density of the compressor suction stream would be four times normal for the duration of the pressure surge. This would put a correspondingly large mechanical stress on the motor, coupling, gear box, and compressor components. Many industrial machines are normally designed with a four-to-one safety margin so it should not be difficult to procure a suitably designed compressor-motor package from the normal suppliers.

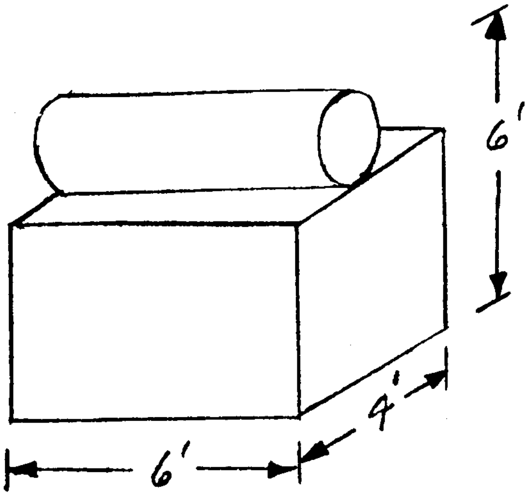
The principle source of noise will be the electric motor and centrifugal compressor. Neither of these components is particularly noisy, but if necessary the motor-compressor unit could be contained within an accoustically insulated box. Another scurce of noise is the reversing heat exchangers, arising from the fact that each reversal, every one to two minutes, is accompanied by a loud blow-down noise. The reversal noise could never be eliminated, but it could be greatly reduced by means of a well designed muffler.

4. Size, Weight, and Cost

The entire plant separates naturally into five packages as follows:

a. Compressor Intake Filter and Aftercooler

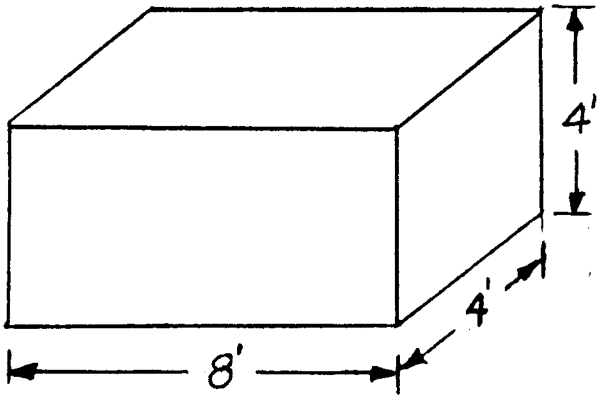
The preferred intake filter consists of a viscous impingement self-cleaning filter followed by a glass fiber filter. Both would be contained within a package 6' x 4' x 4', weight = 2500 lbs, cost $2500. Attached to this package and becoming an integral part of it would be a cylindrical shell-and-tube aftercooler 18" diameter, 6' long, weight = 1600 lbs, cost $3500. For total package:



height	6'
length	6'
width	4'
weight	4100 lbs

b. Compressor and Motor

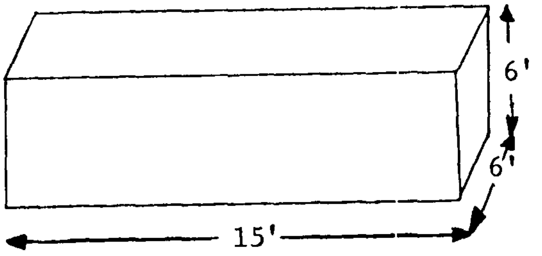
The compressor/motor package consists of a 325 hp, 3600 rpm (460 volts, 60 cycles) electric motor coupled to a single-stage centrifugal rotary compressor.



height	4'
length	8'
width	4'
weight	10,000 lbs

c. Reversing Heat Exchanger

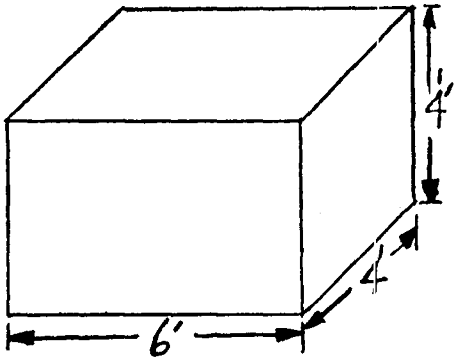
The reversing heat exchanger is a plate-fin type aluminum brazed assembly of extended surface. It consists of four cores, each 2' x 2' x 10', assembles in parallel and contained within a sheet metal cold box. Allowing for 10-12 inches of thermal insulation, this package will be the largest of the five.



height	6'
length	15'
width	6'
weight	6000 lbs.

d. Expansion Engine

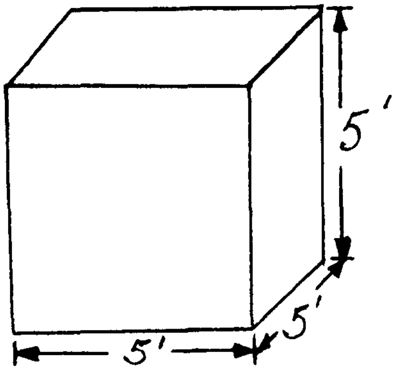
The expansion engine itself is a relatively small piece of equipment. It would most likely be a radial inflow type as manufactured by Airco Cryogenics or Rotoflow Corporation. The power output could be used to operate a centrifugal supercharger ahead of the main compressor or alternatively, the power output could be dissipated as heat. In any case, the size and weight of the expansion engine package would be little affected.



height	4'
length	6'
width	4'
weight	2000 lbs

e. Sparger Cold Box

The sparger itself would consist of a cylindrical aluminum tank 3' in diameter and 3' tall. Since it must be very well insulated, the cold box would have to be at least 5' in diameter and 5' tall, or alternatively, a 5' cube.



height	5'
length	5'
width	5'
weight	1500 lbs.

In summary, the total plant will occupy a volume of about 1100 cubic feet, will have a total weight of 23,600 pounds, and will consist of components having an aggregate purchase cost of $186,000. In addition to component costs we must add approximately $200,000 for miscellaneous valves, piping, controls and installation costs, making the total plant cost $386,000.

The large amount of capital required to construct a 5000 SCFM cryogenic purification plant suggests that the operating costs will be quite sensitive to plant usage. More specifically, we would expect operating cost per hour to be much less if the plant is operated 24 hours per day as compared to a one shift per day schedule. In conformity with our standard practice for all systems studied, we have estimated the operating cost on the basis of two-shift operation, 260 days per year.

TABLE 23. CRYOGENIC CONDENSATION

CAPITAL COST - 5000 SCFM UNIT

Equipment	Size	Installed Cost
Air Filter	5000 SCFM	$ 2,500
After Cooler		7,000
Compressor	325 HP	130,000
Reversing Heat Exchanger	4 Cores (24,000 ft)	80,000
Expansion Engine	53 HP	140,000
Sparger Cold Box	3 ft diameter x 3 ft	16,500
		$376,000
Engineering, Overhead, Fee & Contingency (15%)		56,400
Total Capital Cost		$432,400

TABLE 24. CRYOGENIC CONDENSATION

ANNUAL OPERATING COST

5000 SCFM, 2 Shifts, 260 Days/Year

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	\$20,000/Yr	\$ 20,000
Utilities			
Electricity	10.4 x 10 kwh	\$0.01/kwh	10,400
Water			3,000
Miscelaneous Supplies			1,000
Maintenance	4% of Capital Cost		17,200
Direct Operating Cost			\$ 51,600
Capital Charges	22% of Capital Cost		94,800
Total Annual Operating Cost			\$146,400
Total Cost per MSCF			11.7¢

References

1. Lange's Handbook of Chemistry 1952 - p. 1480.
2. U.S. Patent 3606760 - Method of Purifying Radon-Containing Air.

F. CHEMICAL REACTION (With Molecular Sieves)

1. Description of a Preferred System

Only a few reactions of radon are known. A list of substances which react with radon is given in Table 25. The radon containing products of the reactions with these compounds are nonvolatile; so in principle, at least, they provide a means for removing radon from air.

a. Radon Chemistry

Microcurie quantities of radon react with an excess of fluorine at 400°C. This reaction has been carried out by condensing radon in a nickel vessel, pressurizing with fluorine, and then heating the vessel. A nonvolatile radon-containing product remains in the vessel after the fluorine is pumped off.[1,2,3] When confined to a small volume, millicuries of radon may react with fluorine at room temperature as a result of activation by intense alpha radiation from the radon.[4] By analogy with the fluorides of krypton and xenon, the product of the fluorine-radon reaction is thought to be RnF_2. The radon-fluorine reaction product reacts with water to release radon, oxygen, and hydrogen fluoride. XeF_2 and KrF_2 also react with water to yield oxygen, hydrogen fluoride and xenon or krypton.

Solutions of radon-fluorine compounds have been prepared by the spontaneous reaction of radon with solutions of fluorinating agents (BrF_3 in HF, BrF_5 in HF, K_2NiF_6 in HF, BrF_3 in IF_5) and with pure liquid fluorinating agents (ClF, ClF_3, ClF_5, BrF_3, BrF_5 and IF_7).[5] These reactions were carried out in in a metal vacuum system by condensing radon into tubes containing small quantities of the reagents. After standing for one and one half hours at room or lower (in the case of the volatile reagents) temperatures, the excess fluorinating agent was pumped off, leaving a nonvolatile radon-containing residue.

BrF_3 (a liquid at room temperature) has been used to scrub radon from air.[6] BrF_3 was placed in a simple gas bubbler and dry air containing about 2-3 millicuries/liter of radon passed through. By placing two traps in series it was shown that BrF_3 could efficiently scrub radon. It has also been shown that a BrF_3 trap will scrub a large fraction of the ambient radon from air (picocurie/liter level).

A number of solid ionic complex compounds have been found to react with radon.[7,8] Air containing radon (about 1 to 20 millicurie/liter) was passed through small (6.3 mm dia by 50-100 mm long) powder beds of these reagents. ClF_2SbF_6, BrF_2SbF_6, $BrF_4Sb_2F_{11}$, $IF_4(SbF_6)_3$, BrF_2BiF_6 and O_2SbF_6 were found to capture 100% of the radon. Several other compounds ($KBrF_4$, $CsBrF_6$, $KClF_4$, BrF_2TaF_6, and IF_4SbF_6) were found to retain radon with lower efficiency. Further experiments have shown that O_2SbF_6 is effective in removing radon at the picocurie per liter level (40 pc/liter) from air.

TABLE 25

SUBSTANCES KNOWN TO REACT WITH RADON

F_2

BrF_3

BrF_5

ClF

ClF_3

ClF

IF_7

O_2SbF_6

$KBrF_4$

K_2NiF_6

$CsBrF_6$

$KClF_4$

$RbClF_4$

ClF_2SbF_6

BrF_2SbF_6

$BrF_4Sb_2F_{11}$

$IF_4(SbF_6)_3$

IF_4SbF_6

BrF_2TaF_6

BrF_2BiF_6

The reactions involved are thought to be of the type

$$Rn + ClF_2^+ SbF_6^- \rightarrow RnF^+ SbF_6^- + ClF$$

$$Rn + 2O_2^+ SbF_6^- \rightarrow RnF^+ Sb_2F_{11}^- + 2O_2$$

Attempts to induce radon to react with oxygen, ozone, and chlorine have been unsuccessful.[2] A claim that radon will react with strong aqueous oxidizers[10] has been disproven.[11]

b. Problems with Chemical Removal of Radon from Mine Air

There are two difficulties associated with chemical removal of radon from mine air. These are the reactivity of the reagents with water and the hazardous nature of the reagents. Reaction with moisture in the air will deplete the reagent supply, possibly produce toxic products[7,8,12,13] and release previously captured radon (by reaction with the trapped radon compounds). There is no known substance which will react with radon but not not with water. Considering the inert nature of radon, it is unlikely that one will be found. Safety problems are least severe with the solids since they are nonvolatile. The liquid reagents and the solvents which have been proposed are sufficiently volatile that dangerous concentrations of toxic gases may be produced at room temperature. This problem is obviously most severe in the case of fluorine since it is a gas under usual conditions.

c. Selection of a Scrubber Agent

The $Rn + F_2$ reaction requires elevated temperatures. It was eliminated from further consideration on this basis and on safety grounds. Although liquid scrubbing reagents have a number of desirable features including the possibility of a low pressure drop across the scrubber and ease of replacing the reagent, the safety problems associated with the liquids and solvents which have been proposed are also severe. At the present state of the art, solids appear to be the best candidates for chemical removal of radon from mine atmospheres. Of the solids, O_2SbF_6 has a number of desirable features; it reacts smoothly with water (instead of violently) to yield oxygen and small quantities of ozone and hydrogen fluoride[9]; it has a relatively high decomposition temperature (240°C)[14]; and it is less corrosive than the complexes with fluorine containing cations. Thus attention will be focused on O_2SbF_6 for the remainder of this report.

d. Design of the System

The only data on which to base an O_2SbF_6 scrubber design are those from small scale laboratory experiments. Published data are available on a small O_2SbF_6 powder bed scrubber. Experiments have also been conducted with O_2SbF_6 pellet beds. The pellet beds have the advantage of a much lower pressure drop.[8] Scale-up of laboratory beds was used to estimate the dimensions of a full scale scrubber. These dimensions are given in Table 26. The scale-up was based on the parallel operation of a number of small scrubbers to produce a flow of 5000 CFM. From Table 26 it is clear that the pellet trap is preferable.

Consumption of O_2SbF_6 by moisture in the air will be substantial. 5000 cubic feet of air at 100°F and 100% relative humidity contains 14.3 lbs of water.[15] On a one to one molar reaction ratio, this quantity of water would consume 212 lbs of O_2SbF_6. Such a consumption rate is clearly unacceptable. Reduction of water content to saturation at 0°C by refrigeration would reduce this consumption rate to 5.9 lbs of O_2SbF_6 per minute (357 lbs/hour) for 5000 CFM of air. This is also quite large. At -20°C the consumption rate would be reduced to 2.4 lbs/minute (143 lbs/hour). It is obvious that extremely efficient water removal will be required. For the comfort of miners the air should be rehumidified after passage through the O_2SbF_6 scrubber.

CO, NO, NO_2, and SO_2 will also react with O_2SbF_6; CO_2 does not.[8] The reaction products are listed in Table III. At 1000 ppm, NO_x would consume 3.4 pounds per minute of O_2SbF_6 at an air flow of 5000 CFM, SO_2 at 3 ppm would consume 0.01 pounds per minute, and CO at 500 ppm would consume 1.7 pounds per minute (assuming one to one molar reaction ratio in all cases). It is quite likely that the concentrations of these substances would be reduced by the water removal process so special traps may prove unnecessary. Organic particulate matter could also react with the O_2SbF_6. The effectiveness of the O_2SbF_6 pellet trap is about 99% (fraction of radon retained) even at the picocurie/liter level.[8] An air filter should be provided to remove all large particles.

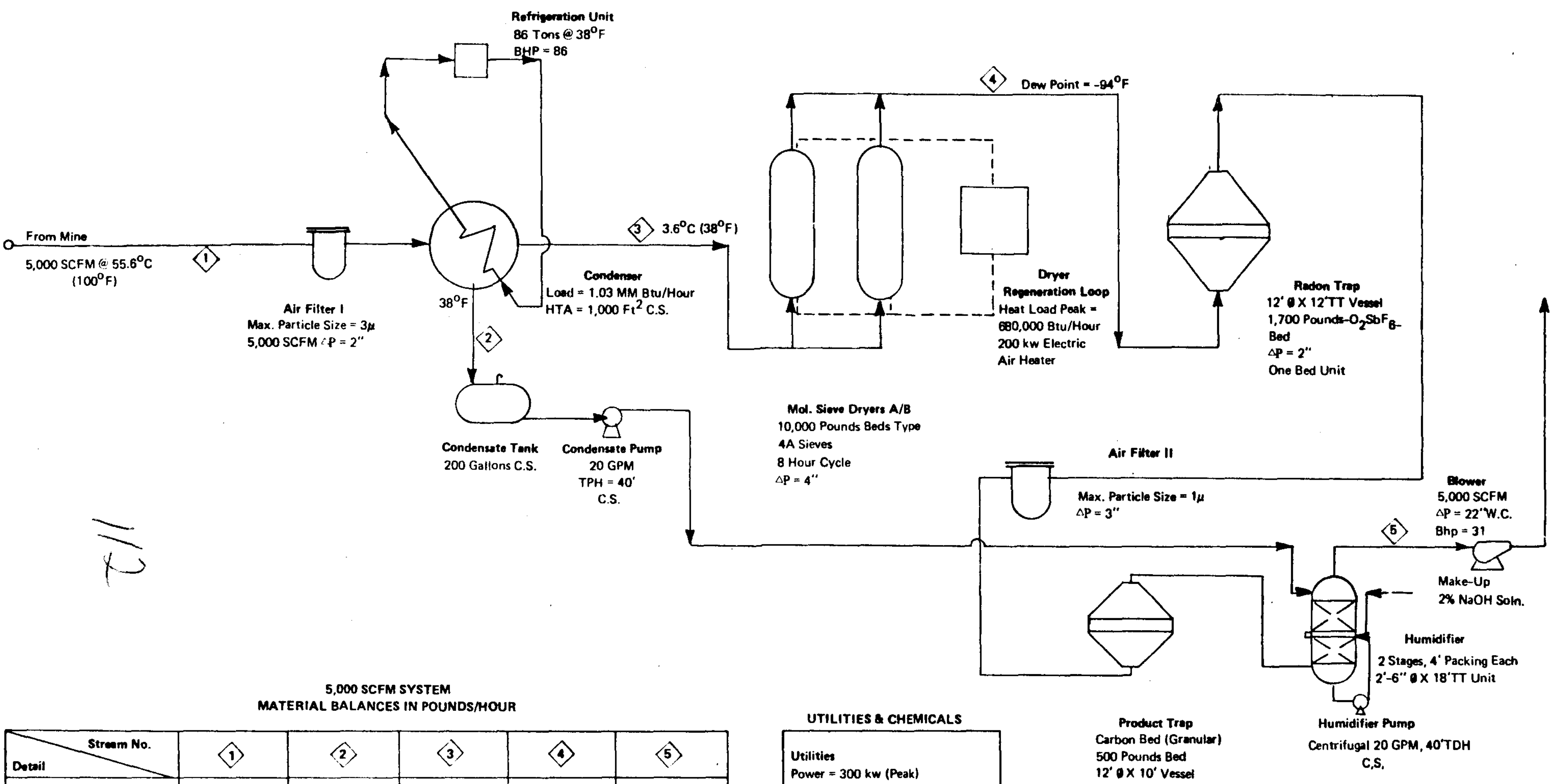
As shown in Figure 22, the radon-laden air would pass through an Air Filter I to remove all particulates down to 3 μm diameter. This would be a conventional cartridge type filter.

In attempting removal of water, various combinations of refrigeration, molecular sieves and dessicants were considered. Based on approximate evaluations and discussions with vendors, the following conclusions were reached:

i. Economics dictate that as much of the water as possible be removed by refrigeration. This means a condenser operating at about 2-4°C (36-38°F), since lower temperatures produce icing.

ii. Air saturated at 2-4°C can be dehydrated to a dew point of -70°C (-94°F) by Type 4A molecular sieves. This was chosen as the most efficient method of dehydration.

The air after Air Filter I would enter a condenser. In the condenser, the refrigerant would be maintained at about 2°C (34.6°F) and the air would be cooled to about 3.3°C (38°F). The condenser would be of the finned type and have a heat transfer area of over 1000 ft^2. The refrigeration load would be supplied by a packaged mechanized refrigeration unit rated at 86 tons. The water collected in the condenser would flow to a 200 gallon carbon steel condensate tank. From here it would be pumped to the rehumidification section of the humidifier unit.



5,000 SCFM SYSTEM
MATERIAL BALANCES IN POUNDS/HOUR

Stream No. / Detail	1	2	3	4	5
Air Water Others	23,700 712 –	– 598 –	23,700 114	23,700 .008 –	23,700 120 –
Total	24,412	598	23,814	23,700.008	23,820
Temperature (°F) °C	(100°F) 55.5°	(38°F) 3.6°	(38°F) 3.6°	–	–

UTILITIES & CHEMICALS

Utilities
Power = 300 kw (Peak)
CW. = 131 GPM

Chemicals (Annual)
Mol Sieves (4A) = 10,000 Pounds
O_2SbF_6 = 950 Pounds
Carbon = 500 Pounds
NaOH = 60 Pounds

May 15, 1974

Radon Gas Removal O_2SbF_6 Trap System Schematic Flow Diagram		
5,000	SCFM	Unit
75,946	F-1	Rev. B

FIGURE 22. Schematic Arrangement of Chemical Scrubber System.

System: O_2SbF_6 Trap System

No.	Detail	Cost Basis	Cost Ref.	Equipment Cost
1	Blower	5,000 SCFM, 100^{o}F, ΔP = 24" C.S.	Estimate from Files Including Motor (50 H.P.)	$6,000
2	Air Filters I & II	5,000 SCFM, 40-100^{o}F, for 3μ Size Particles, ΔP = 2"	Estimate from Files	$1,000
3	Condenser	1.03 MM Btu/Hour, 1,000 Ft^2 HTA C.S./Al-Fins	Estimate @ $ 6/Ft^2	$6,000
4	Refrigeration System	GHCOL Based, 86 Tons/38^{o}F Packaged Unit	Package Unit Estimate from File	$35,000
5	Cond. Tank & Pump	200 Gallon C.S. Tank 20 GPM Centrifugal Pump, C.S.	Estimate from File	$2,000
6	Mol Sieve Dryer	10,000 Pound Beds, Type 4A, Electric Regeneration System (200 kw) Fully Automatic, ΔP = 4", C.S.	Vendor's Estimate	$160,000 Installed
7	Radon Trap	12' Ø X 12' Vessel, 1,700 Pounds O_2SbF_6 Bed ΔP = 2", C.S. Disposable Trap Material	Bed at $50/Pound from Vendors. Rest is Estimate from File	$110,000
8	Carbon Trap	12' Ø X 10' Vessel, 500 Pound Carbon Bed ΔP = 2", C.S.	Estimate from File	
9	Humidifier System	2 Stage Unit 2'-6" Ø X 18'TT Vessel 1" Ceramic Rings, 8' Paucing Total 20 GPM Pump	Estimate from File	$7,000

FIGURE 22 a. Legend for Figure 22.

The air, saturated at about 3.6°C, would enter a 2-bed molecular sieve dryer system. Each of the two beds would have 10,000 lbs of Type 4A sieves, providing for an 8-hour cycle. Average loading of the beds at the end of the cycle would be about 10% by weight. Half-life of the molecular sieves under normal use would be about two years, requiring replacement every four years.

The air would give up practically all its moisture and would exit the bed at a dew point of -70°C (-94°F). The two beds would be placed on duty alternately. The spent bed (after 8 hours on stream) would be regenerated by a counter-current flow of air at 550°F. This would be done by a separate blower moving clean outside air through an electric heater and then through the bed. Peak heat load requirement would be about 680,000 Btu/hr; the electric heater would be rated at 200 KW. The entire 2-bed dryer would be on an automatic cycle.

The dry air would then be sent to the radon trap. This would be a 12' x 12' vessel of steel containing about 1700 lbs of O_2SbF_6 either as solid pellets or coated on an inert support in a layer about 7 cm deep (about 3"). Radon would be completely absorbed and retained indefinitely. Penetrating gamma rays from the trap would be shielded by a one-foot thick wall of concrete.

It is estimated that loss of the O_2SbF_6 reagent due to reaction with water alone may exceed about 2165 lbs per year. In addition, while the dryer and condenser may remove substantial amounts of CO, NO_x, and SO_2, some may remain, but the latter are not included in this design. A split-bed O_2SbF_6 scrubber arrangement would be used to avoid radon breakthrough. After the first bed becomes ineffective the second section would be shifted to the first position and a new bed placed in the second position.

The dry air would contain a small amount of ozone (reaction product of H_2O with O_2SbF_6) after leaving the radon trap. To remove this, the air would be passed through a bed of activated carbon. This trap would be a 12' x 10' x 2" vessel with a bed of 500 lbs. of granular carbon. It would remove all strongly oxidizing compounds that may have been generated by the action of the O_2SbF_6.

After removal of ozone, the air would be sent to a two-stage humidifier whose purpose would be:

a. Removal of small amounts of HF by NaOH scrubbing.
b. Rehumidification of the very dry air prior to re-injection into the mine.

The humidifier would be a 2'6" x 18' unit with two sections and an external air by-pass. Air would first enter the lower section; the latter would be packed 1" ceramic Raschig rings to a depth of about four feet. Contact would be with a dilute (∿2%) aqueous solution of sodium hydroxide to absorb all HF. The activated carbon bed should remove most HF. The NaOH trap would therefore serve primarily as a safety feature in the event of a breakthrough of HF from the carbon bed.

TABLE 26

Comparison of Size Requirements For Powder Bed and Pellet Bed

	Powder Bed	Pellet Bed
Area (Cross-sectional)	3.33×10^{-4} ft^2 (31.2 mm^2) (from diameter = 6.3 mm)	1.07×10^{-3} ft^2 (78.5 mm^2) (from 1 cm^2 cross-section)
Length	0.16 ft (5 cm)	0.23 ft (7 cm)
Flow rate	5.3×10^{-4} CFM (15 ml/min)	3.74×10^{-2} CFM (35 linear ft/min)
Pressure drop	not given	approx. 2 cm H_2O (0.8 in H_2O)
Number of parallel beds to yield 5000 CFM	9.43×10^{6}	1.34×10^{5}
Area of 5000 CFM scrubber	3140 ft^2	143 ft^2
Length of side of 5000 CFM scrubber (assuming square)	56 ft	11.9 ft
Weight (estimated from 50% open volume and density of 1.6 grams/cm^3)	26,000 lbs	1660 lbs
Residence time of air in device	approx. 0.1 sec	approx. 6×10^{-3} sec
Residence time for Radon	"∞"	"∞"

TABLE 27

PRODUCTS OF O_2SbF_6 REACTION WITH AIR COMPONENTS

Reactant	Product
H_2O	O_3, O_2, HF
CO	COF_2, O_2, SbF_5 (liquid)
NO_2	NO_2SbF_6, O_2
NO	$NOSbF_6$, NO_2SbF_6, NO_2
SO_2	SO_2F_2, SbF_5, O_2

The air would then pass through to the upper section via an external pipe and contact water in another four feet of packing. Humidification would be to about 60-80% at 70°F.

The clean rehumidified air would pass through the blower which would supply the ΔP of 24" W.C. and re-enter the mine.

A schematic layout of the suggested chemical scrubber system for cleaning of a 4000 SCFM air supply is shown in Figure 22. It is estimated that all the equipment in this system can be placed in three (3) 12' x 30' x 16' skids.

2. Efficiency and Reliability

The lifetime of the O_2SbF_6 scrubber will be limited by the presence of reactive materials, chiefly water, in the air. It will not be possible to restore the O_2SbF_6 once it reacts. The only way to minimize deterioration is to trap the reactive substances before they reach the O_2SbF_6.

3. Safety

Oxygen will not be removed by the O_2SbF_6 scrubber. Reaction of O_2SbF_6 with water and other air contaminants will produce oxygen, and small quantities of ozone and other hazardous materials. A trap should be placed after the O_2SbF_6 scrubber to remove the reaction products.

The scrubber itself has no moving parts; however, noise may be generated by the air moving equipment, the dehumidifier, and the rehumidifier.

The O_2SbF_6 trap will retain essentially all radon in the air passed through it; consequently the radon daughter activity will build up to a high level. A trap receiving 5000 CFM of air with 500 picocurie/liter of radon will accumulate activity equivalent to 0.5 gram of radium and would deliver 4.5 r/hr of gamma radiation if it were a point source. Two feet of concrete would be necessary to reduce the dose to 100 mr/work week of 40 hours at a distance of 3 feet (point source).

O_2SbF_6 is a powerful oxidizer. Contact with oxidizable materials such as organics may result in a violent reaction. The O_2SbF_6 should be enclosed in a metal container to minimize the effects of such a reaction.

4. Cost

The proposed process design involves the following steps:

a. Removal of essentially all moisture from the air stream by low-temperature condensation and molecular sieve dryers.

b. Removal of all radon in an O_2SbF_6 trap.

c. Removal of ozone in a carbon trap.

d. Removal of HF and rehumidification in a 2-stage scrubber.

A system to process 5000 SCFM of air would cost about $616,000. The total annualized cost would be about $305,000. On a 4160 hours-per-year (16 hrs/day, 260 days/yr) basis, this amounts to 24.5¢/1000 SCF of cleaned ventilation air. It may be noted that this is necessarily a "hybrid" system, involving the use of molecular sieves to remove moisture prior to contact of the feed air with the radon-removing reagent. In view of the high vulnerability of O_2SbF_6 to moisture, it was necessary to incorporate a highly effective drying process in the system. The best technique available for this purpose appears to be the combination of a refrigerator drier and a bed of molecular sieves, which, as we have shown elsewhere, are relatively poor adsorbents for radon itself.

TABLE 28. CHEMICAL REACTION
CAPITAL COST - 5000 SCFM UNIT

Equipment	Size	Installed Cost
Blower	50 HP	$ 10,000
2 ea. Air Filters	5000 SCFM	5,000
Condenser	100 ft^2	10,000
Refrigeration System	86 tons at 38°F	50,000
Condensate Tank & Pump	200 gallons	5,000
Mol Sieve Drier	20,000 lbs of Sieve	160,000
Radon Trap	1700 lbs O_2SbF_6	276,000
Carbon Trap	500 lbs Carbon	10,000
Humidifier		10,000
		$536,000
Engineering, Overhead, Fee and Contingency (15%)		80,000
Total Capital Cost		$616,000

TABLE 29. CHEMICAL REACTION

ANNUAL OPERATING COST

5000 SCFM, 2 Shifts, 260 Day/Yr

(Capital Cost $616,000)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/Yr	$ 20,000
Utilities			
Electricity	832,000 kwh	$0.01/kwh	8,300
Water	33,000 M gal	$0.05/M gal	1,600
Materials & Supplies			
O_2SbF_6	2,164 lbs	$50/lb	108,200
Type 4A Sieve	5,000 lbs	$1.10/lb	5,500
Miscellaneous			2,000
Maintenance	4% of Capital Cost		24,500
Direct Operating Cost			$170,100
Capital Charges	22% of Capital Cost		134,900
Total Annual Operating Cost			$305,000
Total Cost per MSCF			24.5¢

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G. CENTRIFUGE

1. Description of a Preferred System

a. Introduction

Techniques of gas centrifugation have been developed on a laboratory scale for enrichment of uranium-235 concentration relative to its natural propor-to uranium-238. This difficult and tedious enrichment process which is accomplished by converting the natural uranium to the hexafluoride which is gaseous at room temperature, and by applying centrifugal forces many times gravity, is considered to be potentially justifiable, in spite of its apparent high cost and low yield, because of the extraordinary value of ^{235}U and because of the comparable difficulty and cost of any known alternative.

Inasmuch as the efficiency of centrifugal separation of gaseous molecules is dependent in part on the difference in their atomic masses, the large mass of radon-222 compared to the average molecular weight of air ($\sim$29) would imply an advantage of almost a factor of eight over the efficiency of the uranium isotope enrichment. In considering the use of gas centrifugation as a technique for purification of ventilation air, however, one must take into account the need for processing of enormous quantities of gas (5000 SCFM $\cong 10^{15}$ tons/hour) compared to the relatively miniscule quantities processed in isotope separation work, but probably the most important difference lies in the extremely small concentrations of radon (about one part in 10^{15}) upon which the useful separative work must be done. The same mechanical work which must be applied to the radon atoms to achieve a ten-fold depletion of their concentration in the purified air must therefore also be applied to 10^{15} times as many air molecules, but for no purpose whatever.

In the interest of achieving an optimal system design for an air-purifying centrifuge to act on 5000 SCFM of mine air, and to produce a tenfold depletion of any intput radon concentration between 30 pCi/l and 500 pCi/l without sacrificing more than one-tenth (5000 SCFM) of the throughput air to waste, several different design concepts were explored. At first it was assumed that the centrifuge would consist essentially of a circular cylinder which would be rotated about its axis. The air is assumed to enter at one end and to flow along the cylinder. The clean air would be withdrawn from the central portion of the remote end and the air with excessive radon would be extracted from the periphery of the cylinder.

It was required first to explore a number of parameters that tend to govern the practical dimensions of such a device.

b. Requirement for Laminar Flow

If the centrifuging action is to produce the desirable gradients in radon concentration, it is important that the stratification achieved through centrifugal action should not be nullified through turbulence resulting from the longitudinal flow through the cylinder. It is therefore essential that this longitudinal flow be laminar rather than turbulent. This requires that the Reynolds' Number Vd/ν be less than 2000; here

V is the central longitudinal velocity (feet/sec)
d is the diameter of the cylinder (ft)
ν is the kinematic viscosity of air (about $2 \times 10^{-4} ft^2/sec$)

This requirement is satisfied if

$$Vd < 0.4$$

In laminar flow the velocity distribution is parabolic and the average velocity (on a throughput basis) is V/2. The throughput is accordingly

$$\frac{\Pi}{8} d^2 V = Q \ (ft^3/sec)$$

When this is combined with the requirement that the flow be laminar, one discovers

$$d > \frac{5\,Q}{\Pi}$$

It is unattractive to contemplate a separator which is more than 10 feet in diameter but for such a unit the input would be only 30 Π ft^3/min and the output of clean air would be even less as is discussed later. If an input of 5000 ft^3/min were attempted, one would require 54 such units. It is impractical to contemplate larger units since the permissible throughput increases only linearly with diameter, while the weight and cost will rise with a higher power of the linear dimension. Thus on a tentative basis we may proceed with the analysis, assuming that the diameter of the centrifuging cylinder is no more than 10 feet.

c. The Centrifugal Separation Relationship.

Tentatively it is assumed that the equipment might be operated to attain a stratification equivalent to that at equilibrium under isothermal conditions at zero throughput. Certainly it would be unreasonable to expect superior performance.

Let p_o denote the partial pressure of a gaseous component on the axis; let ρ_o indicate the corresponding density of that component. Then for an ideal gas one finds

$$\rho = \frac{\rho_o}{p_o} p$$

The centrifugal relationship

$$\frac{dp}{dr} = \rho \, \omega^2 r$$

then becomes

$$\frac{dp}{p} = \frac{\rho o}{po} \omega^2 r^2$$

for which the solution with proper boundary conditions is

$$p = p_o \varepsilon^{\frac{\rho_o}{2p_o} \omega^2 r^2}$$

The density of radon is about 0.01 gm cm^{-3} at one atmosphere pressure (i.e., 10^6 dynes cm^{-2}) and 273°K; hence, ρ_o/p_o is about 10^{-8}. The expression for the isothermal centrifugal gradient at T°K then becomes

$$p = p_o \varepsilon^{13.6 \times 10^{-7} \frac{\omega^2 r^2}{T}} \qquad \text{(for radon)}$$

The corresponding expression for air is

$$p = p_o \varepsilon^{1.8 \times 10^{-7} \frac{\omega^2 r^2}{T}} \qquad \text{(for air)}$$

Hence the relative concentration of radon in air will increase with increasing radius in proportion to

$$\varepsilon^{11.8 \times 10^{-7} \frac{\omega^2 r^2}{T}}$$

Note that in the above expressions ω is in radians per second and r is in cm.

d. Fractional Throughput of Clean Air

If one assumes that the concentration of radon in the clean air must be reduced by a factor n relative to the incoming air, then one requires

$$\frac{\int_o^{R^*} 2\pi r \, dr \, \varepsilon^{1.18 \times 10^{-6} \frac{\omega^2 r^2}{T}}}{\int_o^{R} 2\pi r \, dr \, \varepsilon^{1.18 \times 10^{-6} \frac{\omega^2 r^2}{T}}} = \frac{1}{n}$$

where in the limits of integration, R* denotes the radius of the exit duct and R is the radius of the centrifuge. (R = 152 cm for d = 10 ft.)

This is equivalent to

$$n \left\{ \varepsilon^{1.18 \times 10^{-6} \frac{\omega^2 R^{*2}}{T}} - 1 \right\} = \varepsilon^{1.18 \times 10^{-6} \frac{\omega^2 R^2}{T}} - 1$$

If $R^*/R = \alpha$ and $T = 304^{\circ}K$, this becomes

$$n \left\{ \varepsilon^{0.59 \times 10^{-6} \alpha^2 \omega^2} - 1 \right\} = \varepsilon^{0.59 \times 10^{-6} \omega^2} - 1$$

Suppose n=10 (i.e., a ten-fold reduction in concentration were required), one can determine α^2 as a function of ω^2 for the 10 foot diameter centrifuge. The values are tabulated below:

ω^2	α^2
10^6	0.13
2×10^6	0.17
4×10^6	0.28
8×10^6	0.53

e. Limits Imposed by Strengths of Materials

Spinning a 10 foot diameter cylinder to achieve these angular velocities produces a hoop stress in the cylindrical skin. This stress level in psi amounts to

$$\frac{D\,\omega^2\,r}{64}$$

where D is the density in lbs/in^3 (0.28 for steel), ω is the angular velocity in radians per second, and τ is the radius in inches (60 inches radius for 10 foot diameter).

Corresponding to $\omega^2 = 10^6$, this stress level would be 130,000 psi. This stress level, which occurs for the minimum level of ω^2 that could be of interest, is excessive for design with structural steel.

f. Scaling Factors

If the product ωR is maintained constant, the fractional throughput of air will be preserved. On increasing R by a factor 4 while maintaining ωR constant, the hoop stress corresponding to $\alpha^2 = 0.13$ will fall to 32,000 psi. It would be possible to sustain a stress level of 65,000 psi in such an apparatus; this would correspond to $\alpha = 0.17$ in a cylinder of diameter 40 ft. The air input to such a unit would be $120\ \pi = 375\ ft^3/min$.

g. Output of Clean Air and Exhausting of Spent Air

The output of clean air is determined by the value $\alpha = 0.17$ when applied to the density gradient appropriate to the air mass. The fraction of the input air that emerges as "clean" air would then be

$$\frac{\varepsilon^{0.18 \times 0.17} - 1}{\varepsilon^{0.18} - 1} = 0.16$$

This implies that for the 375 ft^3/min which enters the equipment, only 60 ft^3/min emerges as "clean" air (i.e., radon concentration reduced tenfold); 315 ft^3/min of residual contaminated air must be pumped out of the mine. It is clear that it would be simpler to pump 60 ft^3/min of surface air into the mine than to pump 315 ft^3/min out of the mine.

h. Operating Speed

If the 40 ft, diameter centrifuge is to be operated so as to achieve $\alpha = 0.17$, one requires $\omega^2 = 2 \times 10^6 \div 16$ or $\omega = 354$ radians sec^{-1}. This implies a surface speed of <u>480 miles per hour</u>.

The total impracticability of rotating a 40 ft diameter cylinder at a peripheral speed of 480 mph is not worthy of further pursuit, though as an exercise in "over-kill" one might compute the horsepower required to overcome the drag forces and the air-conditioning load this would imply. The momentous engineering task associated with such a mechanical system could never be justified, even as an experimental device.

i. Thermodynamic Considerations

Further consideration was given to the previously stated assumption that a degree of stratification would be necessary in the centrifugal gas corresponding to that at equilibrium under isothermal conditions at zero throughput. The time required to permit the radon concentration to approach the steady state centrifugal gradient turns out to be essentially equal to the time required for the air to attain near-isothermal conditions. This requirement imposes a constraint on the dimensions of the centrifuge. It turns out that for maximum gas throughput the length of the centrifuge must be about 500 times the radius. Thus a tubular configuration is indicated, rather than a disc-like shape. Further, the mass of the equipment required for a given air throughput has been found to increase in direct proportion to the tube radius, leading to the conclusion that a battery of small-diameter tubular centrifuges is preferable to a single massive cylinder. On translating these new requirements to the 40 foot diameter tubes contemplated earlier, we realized that the 2-mile length requirement puts this design entirely out of the question for use underground.

When a gas mixture such as air enters a centrifuge the compression that occurs towards the walls of the centrifuge results in both an immediate increase in density and an increase in temperature. This is the so-called adiabatic process which occurs when there has been insufficient time for heat to flow from the hotter air at the periphery to the cooler air near the axis.

In this adiabatic state there are two factors out of balance. There is a temperature gradient in the air mass and hence a tendency for heat to flow to reduce this gradient. Also the concentration of radon is initially uniform throughout the air and there is a tendency to establish the concentration gradient characteristic of the centrifugal field.

The transfer of heat and the transfer of radon both proceed through a process of molecular diffusion. Since the concentration of radon is low, the rate of diffusion of radon within air is predominantly characterized by the diffusional characteristics of air itself. Thus the rates of radon transport and of heat transfer will be comparable in magnitude. Since the concentration of radon is low, the rate of diffusion of radon within air is predominantly characterized by the diffusional characteristics of air itself. Thus the rates of radon transport and of heat transfer will be comparable in magnitude. Since the object is that of realizing a concentration gradient approaching that attained after prolonged centrifuging, one requires a residence time within the centrifuge comparable to that which will result in a near isothermal state. For practical purposes we may examine the conditions under which the improvement in concentration ratio is about 90 percent of that attainable in infinite time.

The residence time required to approach the isothermal state is determined by the thermal diffusivity of air, which is simply the ratio of the thermal conductivity to the heat capacity per unit volume. Rather than undertake a detailed analysis of the thermal transient it is sufficient to refer to results quoted in Carslaw and Jaeger "Conduction of Heat in Solids", Oxford, 1947, p. 175. These can provide useful guidance relative to the minimum permissible residence time within the centrifuge. The various curves in the graph refer to this residence time in units of a^2/k where a is the radius of the centrifuge in cm and k is the thermal diffusivity which for air is approximately 0.18 $cm^2 sec^{-1}$.

Examination of the curves in Figure 23 shows that a residence time "t" corresponding to

$$\frac{kt}{a^2} = 0.5$$

will be required in order to achieve near isothermal conditions (Tr/Ta $\geqq$ 0.9 throughout the cylinder. This implies $t = 2.8\ a^2$. Ta is the temperature at the periphery of the centrifuge (the inner wall surface) and Tr is the temperature at any value of radius = r.

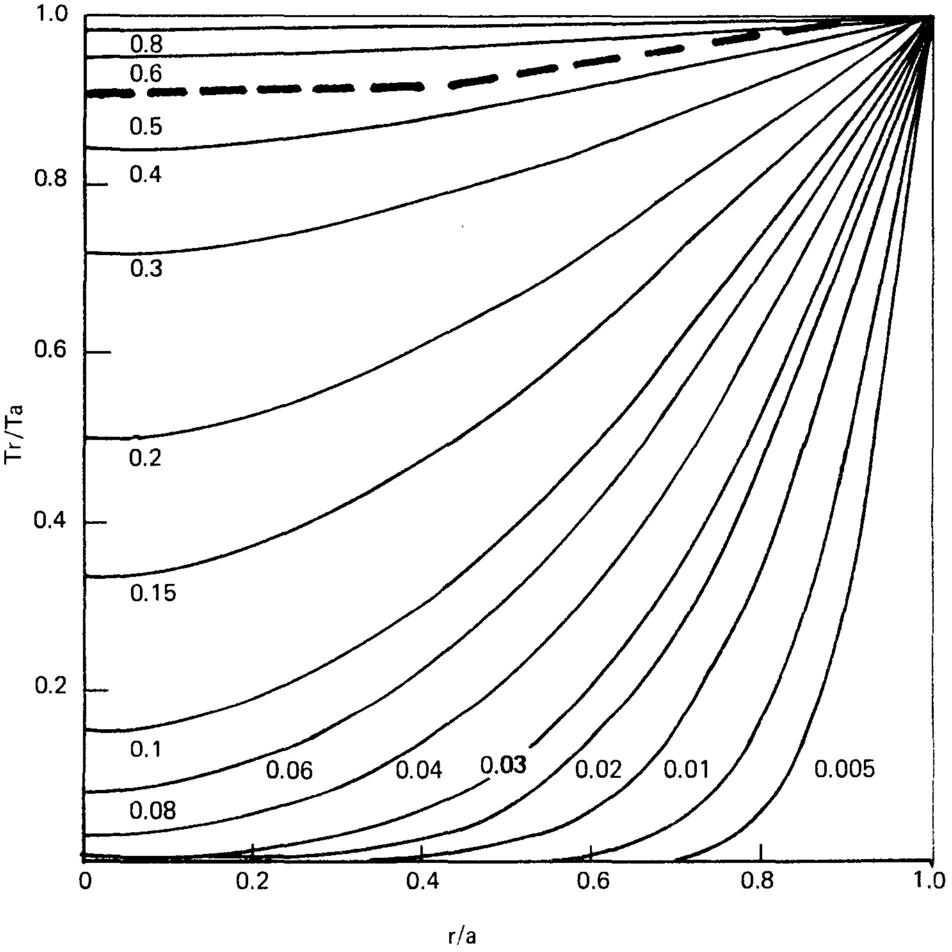


FIGURE 23 RADIAL TEMPERATURE DISTRIBUTION AT VARIOUS POINTS IN A CYLINDRICAL MASS. THE NUMBER ASSIGNED TO EACH CURVE REPRESENTS A SPECIFIC VALUE OF kt/a^2 CORRESPONDING TO A RESIDENCE TIME, t.

In order that the flow through the centrifuge not produce turbulence which would destroy the stratification one requires that the Reynolds number R satisfy

$$R = \frac{2Va}{\nu} \leq 2000,$$ where V is gas velocity (cm sec^{-1})
and ν is kinematic viscosity ($\text{cm}^2\text{-sec}^{-1}$)

For air, the kinematic viscosity is about 0.18 $\text{cm}^2\text{sec}^{-1}$. Hence,

$$Va \leq 180 \text{ cm}^2\text{sec}^{-1}$$

One may now infer a length-to-radius ratio L/a for the centrifuge for maximum throughput, since the residence time (t) will be L/V. At maximum throughput this implies

$$t = 2.8\, a^2 = \frac{La}{180}$$

whence $$\frac{L}{a} = 500$$

The maximum rate of air input is approximately

$$\tfrac{1}{2}V \quad \pi a^2 \quad \rho$$

The factor ½ is associated with the parabolic velocity profile, where ρ is the maximum density. Since $= 1.2 \times 10^{-3}$ cm^{-3}, and $Va \leq 180$ cm^2 sec^{-1}, the maximum air input becomes

$$0.11\, \pi a \text{ g sec}^{-1}$$

The surface area of the centrifuge tube is

$$2\pi aL = 1000\pi a^2 \text{ cm}^2 \text{ (since } L/a = 500)$$

If the wall of the tube weighs 1 g cm^{-2} (i.e., about 1.2 mm thick for steel) then the weight of the centrifuge tube will be $1000\pi a^2$ g. Thus for each g sec^{-1} of air input the processing centrifuge tube will weigh about $10^4 a$ g.

Clearly, there is an incentive to avoid large diameter tubes since the total mass of separation equipment will increase in direct proportion to the tube radius. Also, since for maximum throughput L/a = 500, it appears that a value of a = 1 cm, leaving L = 5 meters, appears reasonable; the air input per tube will then be 0.34 g sec^{-1}. To produce breathing air at 2000-5000 ft^3 min^{-1} (1.1-2.8 kg sec^{-1}), a large number of tubes will be required. For a single stage, 3300 tubes will accept 1.1 kg sec^{-1}. To determine how many will be needed for the desired 10-fold reduction of radon concentration, one must first estimate the centrifuging efficiency under isothermal conditions.

We shall assume that the air fraction into which the radon has been concentrated will be removed from the mine, and that this fraction will be kept below 10% of the clean air to be supplied.

On combining the isothermal requirement relating pressure p and density ρ through Boyle's law

$$\frac{p}{p_o} = \frac{\rho}{\rho_o}$$

with the centrifugal relationship

$$\frac{dp}{dr} = \rho\omega^2 r$$

and integrating the resulting differential equation, one finds

$$\frac{\rho}{\rho_o} = \varepsilon^{\frac{\rho_o}{2p_o}\omega^2 r^2}$$

where the subscript zero refers to conditions at the axis of the centrifuge.

One is now faced with the practicality that the maximum surface speed ωa in centrifuges operating with surrounding air is about 250 ft sec^{-1}. This corresponds to 7500 cm sec^{-1} which is about one quarter of sonic velocity. (cf. McGraw-Hill, Encyclopedia of Technology - Centrifuges). Atmospheric pressure is about 10^6 dynes cm^{-2}, and air density between 10°C and 38°C is about 1.2×10^{-3} g cm^{-3}. Thus the maximum isothermal densification in a centrifuge surrounded by air is given by

$$\frac{\rho}{\rho_o} = \varepsilon^{0.6 \times 10^{-9} \times 5.6 \times 10^7}$$

$$= \varepsilon^{0.034} \qquad \text{(for air)}$$

Radon is 7.7 times denser than air and the maximum gradient for radon then becomes

$$\frac{\rho}{\rho_o} = \varepsilon^{0.26} \qquad \text{(for radon)}$$

At intermediate radii these relationships become

$$\frac{\rho}{\rho_o} = \varepsilon^{0.034\left(\frac{r}{a}\right)^2} \qquad \text{(for air)}$$

$$\frac{\rho}{\rho_o} = \varepsilon^{0.26\left(\frac{r}{a}\right)^2} \qquad \text{(for radon)}$$

The mass concentration within radius r in the centrifuge is proportional to

$$\int_o^r 2\pi r\rho(r)\ dr$$

This is easily shown to be proportional to

$$\int_o^{\left(\frac{r}{a}\right)^2} \varepsilon^{0.034\alpha} d\alpha \qquad \text{(for air)}$$

and

$$\int_o^{\left(\frac{r}{a}\right)^2} \varepsilon^{0.26\alpha} d\alpha \qquad \text{(for radon)}$$

The beneficiation in a single stage of centrifugal separation is then:

$$\frac{\varepsilon^{0.034\beta}-1}{\varepsilon^{0.034}-1} \Bigg/ \frac{\varepsilon^{0.26\beta}-1}{\varepsilon^{0.26}-1}$$

where β is the fractional cross-section area from which beneficiated air is removed.

Clearly, extensive multistaging would be required to achieve satisfactory separation. One might therefore choose $\beta = 0.5$ for which case the beneficiation factor is

$$\frac{\varepsilon^{0.017}-1}{\varepsilon^{0.034}-1} \Bigg/ \frac{\varepsilon^{0.13}-1}{\varepsilon^{0.26}-1}$$

$$= \quad \frac{0.01718}{0.0345} \times \frac{0.297}{0.1384} = 1.065$$

For $\beta = 0.5$ the degradation is such that one further stage of beneficiation permits the effluent to be used for the input to the proceeding stage.

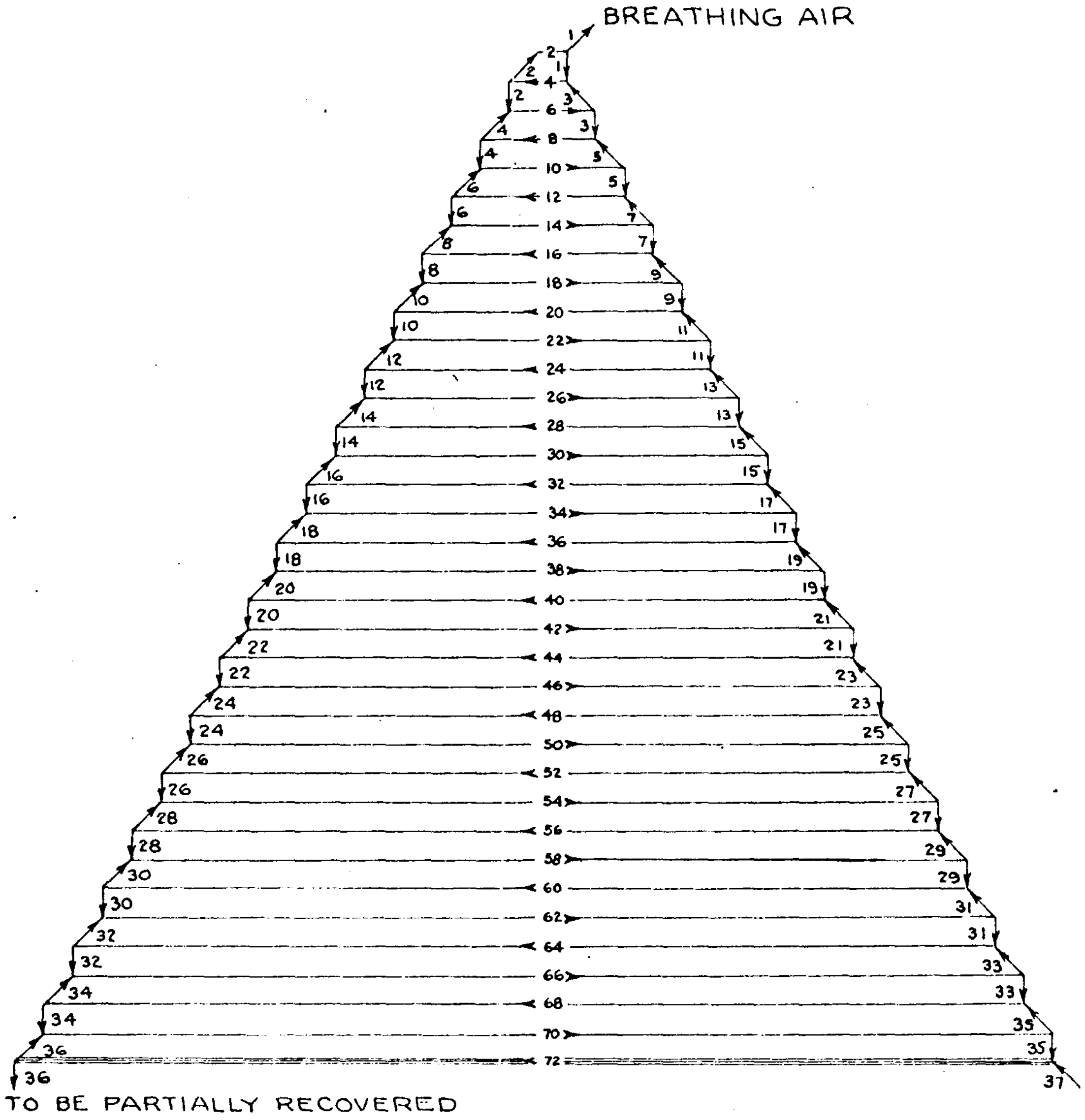
The beneficiation factor is so low that some thirty-six stages (i.e., $1.065^{36} = 10$) in cascade would be required to achieve a 10-fold reduction in radon concentration in the breathing air. Further it is essential that the air into which the radon is concentrated (and which must therefore be removed from the mine) should not exceed 10 per cent of the beneficiated air. The necessary cascading to assure 36 stages of beneficiation and yet reject only 10 per cent of the processed air has been analyzed on the basis of returning degraded air to the input of the preceding stage. This requirement is slightly more conservative than optimum theoretical design but provides a good guide to the magnitude of the centrifugal separation plant. The analysis is depicted in flow charts of Figure 24 and 25.

The number of centrifuges required relative to the number of centrifuges of identical size which deliver the breathing air is found to be 7142, i.e., a total of 14,285 relative to 2. This results from summing the arithmetic progression corresponding to the beneficiation cascade and on summing the arithmetic progression corresponding to the partial recovery of air from the effluent that is contaminated to levels higher than occurs in the input air.

j. Comparison of Air Purification with Uranium Enrichment

As a result of the appearance of a recent textbook dealing with the current theory and practice of gas centrifugation for isotopic enrichment of uranium , we have been able to confirm the essential findings of the foregoing theoretical analysis. Although the specific subject of air purification is not discussed by the authors, a generalized theoretical treatment of the problem is presented which permits a ready comparison of the effectiveness of the technique for air purification and for uranium isotope enrichment. Inasmuch as the centrifuge method is only marginally cost-effective for the small throughput of high value product associated with uranium processing, the results of such a comparison represent a powerful argument against the use of the centrifuge for removal of a trace impurity such as radon from air. In fact, the implication of the Avery and Davies treatment of the problem can be extended to any physical separation procedure in which multiple-stage processing must be employed to achieve the desired separation.

The generalized argument is based first on the comparison of maximum separative power, δU_{max}, for a centrifuge operating on a gas mixture containing equal quantities of two dissimilar species: i.e., radon/air and $^{235}UF_6/^{238}UF_6$. For this special idealized situation, and assuming equal peripheral velocities, the large difference in mass between radon (M=222) and air ($M \cong 29$), compared to the small mass difference between UF_6 molecules containing different uranium isotopes (M=349 vs M=352) give a large apparent advantage to the radon/air separation.



LEGEND

SLOPING ARROW - BENEFICIATED

VERTICAL ARROW - CONTAMINATED
RETURNED TO EARLIER STAGE.

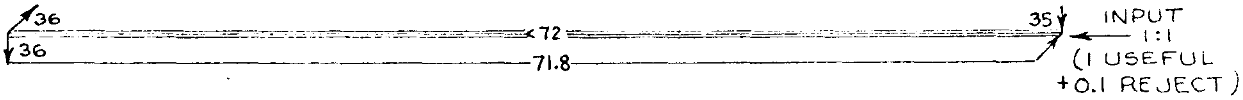
HORIZONTAL ARROW - 50/50 CENTRIFUGE
WITH 1.065 BENEFICATION

N.B. AIR IN THIS PYRAMID IS CLEANER THAN INPUT AIR.

FIGURE 24

Flow Scheme for Cascade Centrifuge

N.B. AIR IN THIS PYRAMID IS DIRTIER THAN INPUT AIR



PARTIAL RECOVERY PYRAMID

TOTAL AIR IN CENTRIFUGAL PROCESSING PER UNIT OUTPUT

$$\underbrace{\frac{36}{2}(72+2)}_{\text{36 STAGES}} + \underbrace{\frac{72-0.2}{0.2} \times \frac{1}{2}(72+0.2)}_{\text{REJECTED 10\% OF USEFUL}}$$

i.e. $18 \times 74 + 179 \times 72.2 = 1332 + 12923.8 = 14255.8$

FIGURE 25.

Flow Scheme for Cascade Centrifuge, Cont'd.

As shown in Appendix I of Avery & Davies[1], the maximum separative power of a centrifuge can be calculated from the following relationship

$$\delta U_{max} = D_{\rho}\left[\frac{\Delta M V^2}{2\ RT}\right]^2 \frac{\pi Z}{2} \qquad \text{g/sec}$$

wherein:

D is the gas diffusion coefficient

ρ is the gas density g/cm^3

ΔM is the difference in molecular weights of the gas species

V is the peripheral velocity in cm/sec

Z is the length of the centrifuge in cm

R is the universal gas constant

T is the temperature in ^{o}K

To compare the maximum separative power of a given centrifuge for different gas mixtures, we can assume that the values of T, R, Z, and V are identical. (In our earlier report we showed that the peripheral speed, V, of a centrifuge for air purification will be limited to 75 m/sec by the fact that it must operate in ambient air. Were it feasible to design such a centrifuge for operation <u>in vacuo</u>, the peripheral speed might be increased to 300 m/sec, equal to that of a uranium-enrichment machine. For the purpose of this calculation we will make the generous, if unrealistic, assumption that vacuum operation is feasible.)

The product, $D\rho$ for uranium hexafluoride is reported to be 2.34×10^{-4}g/cm sec. For air, at a density, $\rho = 1.29 \times 10^{-3}$ g/cm^3, and with D = 0.181 cm /sec, $D\rho = 2.33 \times 10^{-4}$g/cm sec.[2] Thus, it appears that at equal peripheral velocities of δU_{max} varies as a function of ΔM^2. Since ΔM for radon in air is 193 and for the uranium isotope $\Delta M = 3$, the radon system has an apparent advantage of a factor of 4.1×10^3.

The condition of equal molecular populations of the two separate species which is assumed in calculating δU_{max} is not realized in either the uranium enrichment or the radon removal applications. It therefore becomes necessary to take into account the relative abundance of the least abundant species in each case, since the separative performance will be diminished in proportion to the concentration of this species. For air containing 300 pCi ^{222}Rn per liter, there are 5.3×10^5 atoms of ^{222}Rn per liter, or 1.9×10^{-17} atoms of ^{222}Rn per molecule of air. Thus, the mole fraction N, of ^{222}Rn in air is 1.9×10^{-17}. A tenfold reduction of radon concentration in the centrifuge would produce an effluent mole fraction of 1.9×10^{-17}. By contrast, in uranium enrichment the mole fraction of ^{235}U is increased from $N = 7 \times 10^{-3}$ to $N = 5 \times 10^{-2}$. If N is the mole fraction of the less abundant species, the probability of encountering unlike pairs is N(1-N). Consequently the separative performance of a machine will depend upon the relative concentration of species to be separated and the separative processing required will vary as:

$$\frac{1}{N(1-N)} \qquad \text{(c.f., Avery \& Davies, p. 18)}$$

Since in either case the value of N is small, one may replace (1-N) by unity in which case one need only average 1/N over the range of enrichment to determine the influence of relative concentration on the amount of separative processing required.

In reducing the radon concentration from 300 pCi/litre to 30 pCi/litre the average value of 1/N will be

$$\frac{\displaystyle\int_{1.9 \times 10^{-17}}^{19 \times 10^{-17}} \frac{dN}{N}}{\displaystyle\int_{1.9 \times 10^{-17}}^{19 \times 10^{-17}} dN} = \frac{2.3}{17.1 \times 10^{-17}} = 1.35 \times 10^{16}$$

Over the range of uranium from 0.7% to 5% isotopic abundance the corresponding average value of 1/N will be

$$\frac{\displaystyle\int_{0.007}^{0.05} \frac{dN}{N}}{\displaystyle\int_{0.007}^{0.05} dN} = \frac{1.97}{0.043} = 45.7$$

Thus from the point of view of the amount of separative processing required, the abundance ratio in the case of radon removal from air is less favorable than that for uranium enrichment by a factor

$$\frac{1.35 \times 10^{16}}{45.7} = 3 \times 10^{14}$$

While the maximum separative power δU_{max} was shown to be greater by a factor of 4.1×10^3 for centrifugal separation of radon from air than for uranium isotope enrichment, the abundance ratio for the species to be separated is less favorable by a factor of 3×10^{14} in the case of radon removal. On combining these factors one concludes that per gram of gas output, the radon removal problem would require 7×10^{10} times as much separative processing as is done in enriching a comparable throughput of uranium hexafluoride.

This inefficiency of centrifugal removal of radon from ventilation air stems from the fact that the mole fraction for radon in acceptable air is such an extremely small number. This same consideration will apply to every type of separative device which operates through a succession of discrete stages in each of which an incremental improvement is achieved.

When the mole ratio is extremely low as in the case of radon in air the only practical basis for separation will be one which inherently provides the equivalent of an enormous number of separative stages in continuous process (e.g. absorption on passing through a filter, or concentration by freezing out). In such instances each molecular impact with the receiving surface may be considered as a separative stage.

2. Efficiency and Reliability

a. Effectiveness of Radon Removal

While radon concentrations of 30 to 500 picocuries per liter are stated as the range of input concentrations, a beneficiation factor of at least 10 is needed if the equipment is to be realistic. The cost and complexity will increase approximately in proportion to the beneficiation factor. Efficiency of radon-removal is essentially uniform over the stated input concentration range.

The equipment would be insensitive to relative humidity except to the extent that this might influence the dust-precipitation equipment required to remove particulate matter from input air.

Likewise, operation of the equipment would not be greatly influenced by variations in operating temperature over the range 50°F to 100°F.

Air velocity through the centrifuges is limited to 180 cm sec^{-1} for a tube diameter of 2 cm. This is equal to about 35 feet per minute.

Complete removal of aerosol and dust particles from the input air is essential to prevent cumulative loading of the tubes. If 10^5 particles per cm were encountered in a separator producing 3000 $ft^3\ min^{-1}$ ($1.35 \times 10^6\ cm^3\ sec^{-1}$), a removal rate of 1.35×10^{11} particles per second would be required. If the particle density were 2.5 gm cc^{-1} and the mass median diameter were 5 μm, then the average mass per particle would be

$$2.5 \times \frac{125\pi}{6} \times 10^{-12}\ g = 1.6 \times 10^{-10}\ g$$

and the required mass-removal rate would be

$$1.35 \times 10^{11} \times 1.6 \times 10^{-10} = 22 \text{ g sec}^{-1}$$

Hence, particulate matter must be removed at a rate of about 22 g sec^{-1} or 4000 lbs per day.

Impurities of noxious gases which are heavier than air will also be separated from the air in the centrifuging process. This will have little effect on the equipment, provided it is fabricated from a corrosion resistant alloy.

Over-pressures up to 60 psia will be very damaging to delicate centrifuging equipment. The interconnection of multiple stages will undoubtedly suffer from recurrent shocking at 60 psia overpressure.

The excavation and set-up time to relocate a delicate instrument of dimensions 5 meters x 350 meters x 100 meters as mining advances would severely limit the mobility of the system. The power intput of the order of 1000 megawatts will present a significant load on the power that can be generated from the uranium mined.

b. Space Requirement

Each tube of radius 1 cm has a throughput of 0.34 g sec^{-1}. The output of 1 to 3 kg sec^{-1} of breathing air thus requires some 3000 to 9000 tubes in the output stage. Since the number of centrifuges of the total assembly is 7142 times the number in the output stage the total number of centrifuging tubes required will range from 22 to 66 million.

It would be reasonable to allow 16 cm^2 per tube for a compact interconnection. The centrifuging plant would in this case be 5 meters long and have a cross section of some 350 x 100 meters to 350 x 300 meters (i.e., about 20 football fields). Obviously, the cross section can be reduced by increasing the length.

For each g sec^{-1} of air throughput the centrifuge of radius 1 cm will weigh 10 kg. The cascaded centrifuges that will yield 1 g sec^{-} of breathing air beneficiated 10-fold and yet not require renewal of more than 0.1 g sec^{-1} of contaminated air will thus weigh 143 metric tons.

Beneficiated air is required at a rate of about 1 to 3 kg sec^{-1}. Thus, the weight of the centrifuging equipment will be about 143,000 to 430,000 metric tons to support 3 to 5 men on a working face.!

c. Power Requirements for a Centrifuge Operating in Air

At each transition from one tube to the next the coherency of the rotation is lost. The air near the periphery will have attained a velocity of about ¼ sonic; when this ordered motion emerges as turbulence, the absolute temperature will rise by about 6 percent. Air exterior to the centrifuge is also set in motion at comparable speed; accordingly, it is reasonable to account for the energy imparted internally and externally by assuming that all the air inside each tube reaches peripheral velocity and neglect contributions to air from outside each tube.

Air is delivered at 1 to 3 kg sec^{-1} from the final stage, but the air exiting from all centrifuges amounts to 14,285 times as much. If the absolute temperature rise is 6 percent or about 18°C, and since the heat capacity of air is about 1 joule $g^{-1}{}^{\circ}C^{-1}$ the cooling requirements amount to about

$$14 \times 10^6 \times 18 \text{ watts} = 250 \text{ megawatts at } 1 \text{ kg sec}^{-1}$$

$$\text{or} \qquad 759 \text{ megawatts at } 3 \text{ kg sec}^{-1}$$

This is of course the power to be supplied and the heat to be removed. Allowing for the inefficiencies of drives, the need for air conditioning, etc., the power input will probably be in the range of some 1000 to 2000 megawatts.

If heat removal of the air conditioning involves raising the temperature of water by input of 100 joules g^{-1} (i.e., about 43°F rise) the water flow required will be about 10 tons per second. This should be contrasted with the 1 to 3 kg sec^{-1} of air from the surface that would eliminate the requirement for the whole process.

d. Residence Time

The residence time in each stage of centrifuging will be about 2.8 seconds. a period of about 2 hours will be involved for the air traversing the most circuitous route.

e. Rate and Cause of Deterioration

The principal cause of deterioration of the air purification equipment will probably be metal fatigue due to imbalances in any one of the 22 to 66 million spinning tubes.

f. Preventive Maintenance

Preventive maintenance will undoubtedly be required on a routine basis. Inspection at two-week intervals with major overhaul every six months might be contemplated, provided, of course, that such a gargantuan device could ever be made to function at all.

3. Safety

The centrifuge will not result in decreased oxygen concentration in the breathing air.

The principal noxious byproduct is heat which will require a massive commitment to air conditioning.

The noise levels anticipated would probably be at least that associated with normal drilling, blasting and trolley operation, or 110-120 db.

The slightest impact damage to any one of the 22 to 66 million tubes, each 16 ft long and 0.8 inches in diameter, spinning at about 70,000 rpm, would probably result in instantaneous disintegration of the entire device.

4. Cost

This is clearly precision equipment and its cost to manufacture and install cannot conceivably be less than \$20,000 per ton (i.e., \$10 per lb). On this basis the equipment for producing the air supply for a team of 2 to 5 men would cost some \$3 to \$9 billions. Drive motors and interconnections can be expected to double this to some \$6 to \$18 billions.

The financial estimate of \$18 billion for a 5000 ft^3 min^{-1} unit at 10-fold beneficiation is but a crude first cut at the pricing based on \$10 per lb for the centrifuging elements, with a doubling of the price to take account of pumping stations and duct work. Detailed itemization is not feasible without the development of a detailed design.

On the fortnightly inspection the equipment must be shut down. The maintenance could be provided at an insignificant incremental cost. The skills required in servicing this equipment are those of instrumentation engineers. Electrician specialists will be required to do periodic inspection of the electrostatic precipitator; minimal outage times of the order of 1% can be anticipated with four man crews during outages provided automated inspection techniques are employed.

Air conditioning is essential to the system. Standby air conditioning is probably desirable. The water pumping capability must handle about 10,000 times the weight of the air which would have eliminated the need for the centrifugal separator.

A very crude estimate of the cost of providing purified air at the rate of 2000 to 5000 SCFM by this technique indicates a cost of about \$1.70 <u>per cubic foot</u>. The power requirements alone exceed those that could be supplied by the normal yield of uranium from a five-man stope by more than a factor of ten. Inasmuch as the engineering feasibility of the technique appears to be beyond the reach of the present industrial and technological capability, no detailed cost estimate has been prepared.

References

1. D.G. Avery and E. Davies, "Uranium Enrichment by Gas Centrifugation", Mills & Boon, Ltd. London (1973).

2. Am. Inst. of Physics Handbook, 2nd. Ed., p. 236.

H. ADSORPTION ON SOLIDS AT LOW TEMPERATURES

1. General Considerations

We shall consider here some hybrid systems involving adsorption of radon on solids at temperatures lower than room temperature. Primary attention has been devoted to charcoal adsorbents. Molecular sieves would be used in exactly the same manner. Being poorer adsorbents than charcoal, however, they must be used in proportionately larger quantities. Sizes and costs of molecular sieve systems can therefore be estimated by applying an appropriate scale factor to the values for charcoal. Because the use of low temperature is not sufficient to make molecular sieves competitive with charcoal under any circumstances, detailed cost calculations were not judged to be useful. In general, if the temperature is reduced, a reduction in adsorbent bed size is permitted, because of the higher dynamic adsorption coefficient at lower temperatures. The relative costs of the room temperature system, versus one using refrigeration, must be measured in terms of the added costs of cooling balanced against the savings in the amount of adsorbent required.

Cooling the air to temperatures above 0°C can be effected by continuous mechanical chillers, rejecting water easily as a liquid condensate. Cooling to temperatures below the freezing point of water becomes more difficult because the operation becomes discontinuous - either a system of periodically reversing heat exchangers must be used to purge ice from the surfaces, or a periodically regenerated drying bed (molecular sieve, activated alumina, or silica gel) must be used to remove the water prior to cooling. There is also a choice to be made, when considering cooling below 0°C, as to whether the system should be cooled indirectly (through heat exchange with an external refrigerant) or directly (using the air itself as a working fluid, and exchanging heat between the incoming high-pressure air and the emitted low-pressure air). We summarize these choices thus:

a. Chilling to temperatures above 0°C by indirect cooling
b. Chilling to temperatures below 0°C by indirect cooling
c. Chilling to temperatures below 0°C by direct cooling

and will discuss them separately.

2. Adsorption on Charcoal Above 0°C - Indirect Cooling

Here we consider the effect of installing a mechanical chiller to cool the incoming air down to 2°C. For a 5000 SCFM flow saturated at 75°F, this would necessitate a chiller to remove

- sensible heat of air: 220,000 BTU/hr

- latent heat of water condensed: 348,000 BTU/hr

Assuming we can recover about two-thirds of the sensible heat by heat exchange, we need a chiller rate of (348,000) + (1/3)(220,000) = 421,000 BTU/hr, or 35 "tons" of capacity. (To allow for adequate initial cooling when heat exchange is not available, we should use a unit capable of 48 tons peak refrigeration load.) A schematic is shown in Figure 26.

At a temperature of 2°C, in air saturated with water vapor, the value of dynamic adsorption coefficient "k" for radon on charcoal would be about 10,000 cm^3 (STP)/gram. This is 2.5 times the value of k at 25°C (also in air saturated with water vapor). Thus, the bed can be made 0.4 times the size of a bed at 25°C, for the same 1-hour holdup time for radon. Such a bed would contain 1864 lbs of charcoal, which could be divided into four disc-shaped packets, each 3.8 feet in diameter by 1.4 feet long, fitted with supports and spacers and mounted in one tube 3.8 feet in diameter by 7.5 feet long. The discs would be fed in parallel to maintain a desirably low pressure drop.

The size of the heater and the amount of air needed for regeneration of the charcoal bed decrease greatly, relative to that of the room temperature bed, because there is now less water to vaporize, and there is only 40% as much carbon to heat. Because the bed is designed for a one-hour holdup time for radon, an alternating two-bed system is required. All the performance characteristics of this system will be identical to the one intended for use at 24°C, the only difference being the addition of an air chiller, and the considerable reduction in amount of charcoal needed. In summary, the components of such a system would be as shown in Table 30, below.

The average electrical load would be about

13 KW	for the blowers
60	for the heaters
44	for the chiller
117 KW	Total

3. Adsorption on Charcoal Below 0°C - Indirect Cooling

If it is desired to cool the beds further, i.e., below 0°C, then water first must be adsorbed or frozen out as ice. Since, for indirect cooling, the air would remain at a pressure of about 1 atmosphere throughout, we rule out the reversing ("freezeout") heat exchanger which depends on a larger volume of gas flow on the vaporizing side, and instead assume that a molecular sieve bed would be inserted downstream of the precooler to dry the air prior to refrigeration. Such a bed would have to remove 424 lb/hr of water from the gas stream, and, assuming that the molecular sieve can adsorb a quantity of water equal to 10% of its weight, a bed of 4240 lbs of molecular sieve would be needed for water removal. Since this new bed needed for drying is the same order of magnitude in size as the 4660-lb room temperature charcoal bed whose size we are trying to reduce by cooling, the advantage of a system based on indirect cooling below 0°C is negated.

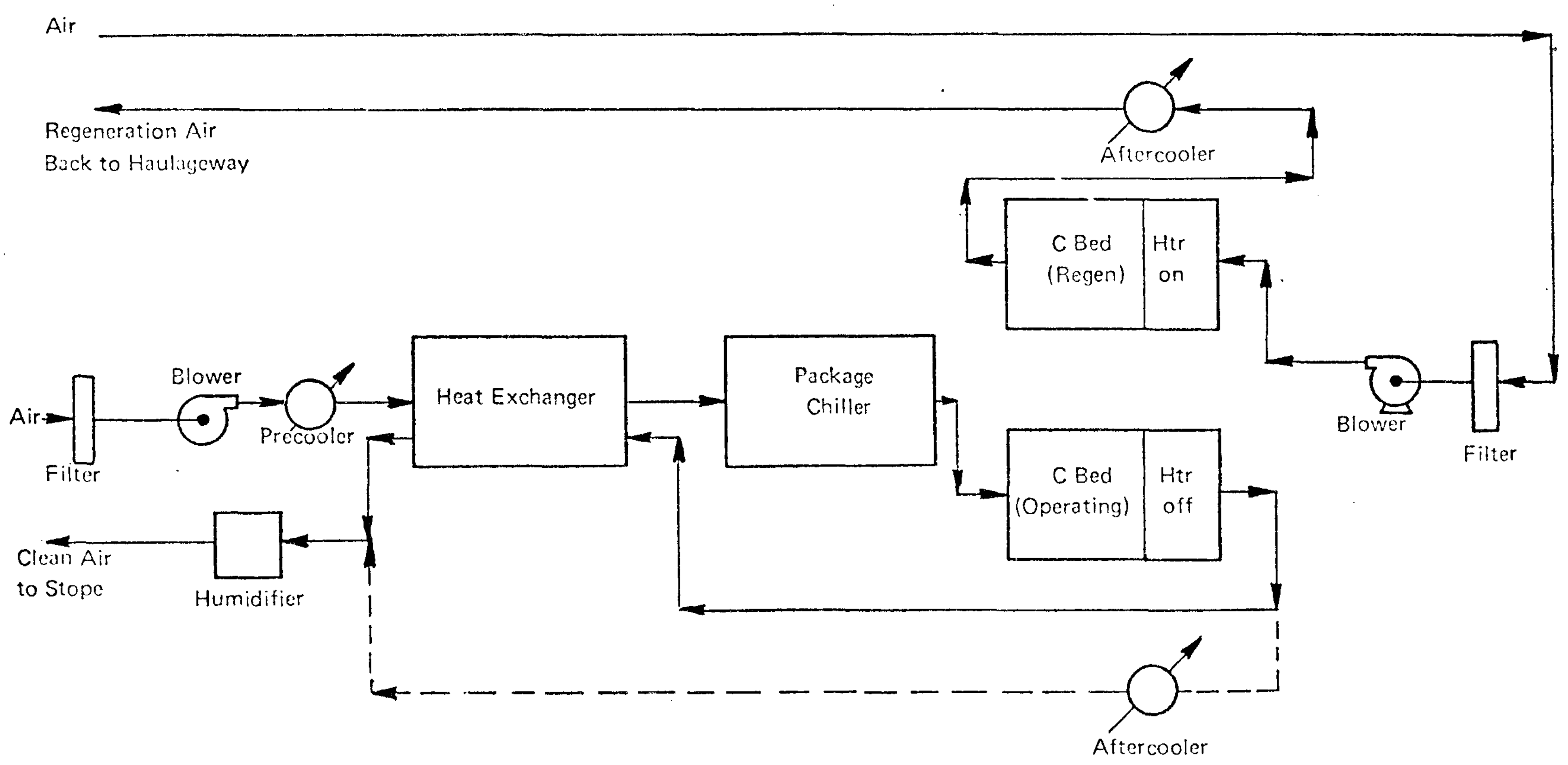


FIGURE 26. CHARCOAL ADSORPTION - INDIRECT COOLING ABOVE 0°C

TABLE 30

Components of 2°C Alternating Bed Charcoal Adsorption System

1	Filter, 5000 CFM
1	Blower, 5000 CFM x 15" H_2O (100 HP)
1	Precooler, 2700 ft^2
1	Package Air Chiller, 48-tons, air out at 36°F
1	Heat Exchanger, Air/Air, 700 ft^2
1	Filter, 1000 CFM
1	Blower (regeneration) 1000 CFM x 2" H_2O (10 HP)
2	Charcoal Beds, 3.8' diam. x 7.5' long, each containing 1900 lb charcoal and each with a 60-KW heater
2	Aftercoolers, 100 ft^2 each
1	Post-Humidifier, to bring air to 65% RH

4. Adsorption on Charcoal Below 0°C - Direct Cooling

If we are willing to compress the air and use it as a working fluid, we can achieve temperatures below 0°C without the need for an external chiller system, and we can take advantage of the reversing-flow heat exchanger. In this respect, the system will be very similar to the cryogenic distillation (liquid air sparging) system described in Section V-E. The major difference is simply that, because a charcoal bed can be operated at much higher temperatures than the liquid air sparger, the heat exchange surfaces will be appreciably smaller. For this reason a system based on charcoal adsorption at -20°C has the advantage of being smaller than any other system considered in this study. Figure 27 shows a low-temperature charcoal system operating on direct refrigeration, using an alternating two-bed system.

We shall work our details for four bed temperatures: -20, -40, -60, and -80°C. Adopting a nominal compression to 2 atmospheres, we shall be able to compare directly with the costs of the cryogenic distillation system. (Higher pressures than this can achieve nothing but further size reduction of an already small charcoal bed, at the cost of extra horsepower.) We shall adopt an 8-hour cycle time, since the cooldown of the beds is more difficult at low temperature and therefore a 1-hour cycle time is no longer satisfactory. The results listed in Table 31 show the effect on heat exchanger size and bed size.

It will be noted that, although bed size decreases as temperature drops, the complexity of the bed design increases. To keep pressure drop reasonable (2 psi) and still maintain L/D ratios of at least 0.3 for good distribution, we are forced to subdivide the bed into smaller and smaller parallel components as the size shrinks. Thus, while at room temperature each bed was split into two parallel-fed discs, at -20°C four are required, at -40°C eight, and at -60°C and below a dual bank of many parallel 2" tubes must be used. For alternate operation, two beds are required, one of which is being warmed and regenerated while the other is cooled and adsorbing. All four of these below-zero cases require, in addition to the reversing heat exchanger and two charcoal beds described above:

1	Compressor, 5000 SCFM to 2 atm, 325 HP
1	Expander, 1500 SCFM, to deliver 20 HP
1	Filter, 5000 CFM
1	Precooler, 2700 ft^2
1	After-cooler, 1700 ft^2
2	Bed heaters

The electrical load is in all four cases about 240 KW, plus the heater power shown in the table. The regeneration air is taken in all cases as a small side-stream of the main flow from the cold end of the heat exchanger, because this gas is dry; room air, while it is warmer than the bed, would be unsuitable for regenerating the bed, as it would deposit ice in the cold bed, perhaps plugging it.

5. Adsorption on Charcoal at -80°C: Single-Bed System

At -80°C, it also becomes feasible to make a bed large enough to achieve a radon holdup time of 12.7 days rather than 8 hours, without exceeding a reasonable bed size. With such a long holdup, a single bed could operate continuously, with no need for regeneration, and always produce an output gas with only one tenth the radon content of the entering gas. Such a charcoal bed would contain 37.4 x 12.7 x 24/8 = 1425 lb of charcoal and thus would be of almost the same dimensions as a -20°C, 8-hour bed, or as 4 discs, each 3.5' diam. x 1.2' long, all in a shell 3.5' diam. x 7'long, with no bed heater. The heat exchanger size would be the same as for the 8-hour case at -80°C, and the power requirement would be about 240 KW.

6. Adsorption on Molecular Sieves at Low Temperature

Operation at low temperatures permits the use of proportionately smaller amounts of molecular sieve adsorbent, but the proportionate gain is not sufficient to overcome the distinct advantage of the higher adsorption power of charcoal at all temperatures. A rough estimate of the costs of molecular sieve radon adsorbers in place of the charcoal units described above, can be made by using the same rule of thumb as that used previously; i.e., such a bed will have about 20 times the weight and 15 times the volume of an equivalent charcoal bed. (Molecular sieve has a bulk density 1.4 times that of charcoal). Thus, the above cases indicate bed sizes as follows:

t_{bed}	Radon Holdup	Molecular Sieve Radon Bed Set (2 Complete Sets Required)	Heater Size KW/Set
2°C	1 (hr)	38,000 lb (6 beds, 6'D x 5.3'L)	1600
-20	8	27,700 (4 beds, 6'D x 5.8'L)	150
-40	8	9,360 (2 beds, 6'D x 3.9'L)	50
-60	8	2,500 (1 bed, 6'D x 2.1'L)	50
-80	8	750 (1 split bed, 2.4'D x 3.6'L*)	50

* Active length; actually composed of 4 discs, so with space, L=6'

In addition, all the molecular sieve cases except that of -80°C will require an auxiliary 15 HP blower for regeneration air. To the power requirements in the table must be added about 250 KW additional for compressor and blower, to obtain total electrical load. The lack of any inherent advantage of molecular sieve over charcoal makes the determination of other system characteristics and costs superfluous.

7. Efficiency and Reliability

The designs discussed above appear to be mechanically feasible, with equipment and materials of known performance available. Efficiency of radon removal is expected to be at least 99% for all alternating-bed systems. For the one case of the long delay single charcoal bed system, radon removal is designed to be 90% of the concentration in the input air. Reliability will be most strongly affected by the effectiveness of regeneration of the adsorbent in the alternating bed systems, and by the useful lifetime of the charcoal in the single-bed system.

The refrigeration equipment for adsorption below 0°C resembles greatly the equipment described in Section E for a cryogenic condensation (liquid air sparging) system. The compressor systems are identical. Indeed, the principal difference, aside form radon retention on a carbon bed rather than in a container of liquid air, is the reduction in size of the very large reversing heat exchanger required for the liquid air system. The charcoal system, since it operates at higher temperatures than the liquid air system, can utilize a more modest-sized heat exchanger. The exchanger sizes in the above table run from 7780 to 14,550 ft^2 and these should be compared to about 24,000 ft^2 for the liquid air case. All exchangers are assumed to work with a temperature difference of 5°C (9°F), this being about the limit for achieving complete re-evaporation of the ice from the exchanger surfaces during flow reversal, when dealing with a pressure ratio of 2:1. We must thus limit the flow to the expander. (We show this on the diagram by a bypassing Joule-Thomson valve across the expander, which allows only 30% or less of the total flow to go to the expander.) This means we can withdraw only about 20 HP of useful work in the expander, so, as in the case of the liquid air system, we ignore the small net work returnable from the expander to the system. One rather considerable advantage of the warmer regime in which the charcoal adsorption operates is the fact that the exchangers would not have to take out the CO_2, as they would in the cryogenic condensation techniques.

Among the uncertainties that may influence the performance of a low-temperature adsorption system are the following:

a. The assumed radon adsorption performance is based on data obtained from laboratory experiments with "clean" air. We do not know how serious would be the effects of the impurities in mine air (CO_2, SO_2, hydrocarbons, oxides of nitrogen).

TABLE 31

Sizes of Heat Exchangers and Charcoal Adsorbent Beds for Operation Below 0°C with 8-Hour Radon Holdup

t_{bed}	Reversing Heat Exchanger Size: Surface Area	Reversing Heat Exchanger Size: Unit Volume	Size of One 8-Hour Charcoal Bed	Bed Shell Size	Average Bed Heater Power
-20°C	7780 ft^2	180 ft^3	1385 lb (4 discs, each 3.5' diam x 1.2' long)	3.5'D x 7'L	8.0 KW
-40	10,040	225	468 lb (8 discs, each 2.0' diam. x 0.6' long)	2.0'D x 7'L	3.0
-60	12,290	270	125 lb (2 banks of 150 2" tubes, each 9.1' long)	3.1'D x 3'L	1.0
-80	14,550	360	37 lb (2 banks of 100 2" tubes, each 4" long)	2.5'D x 2'L	1.0

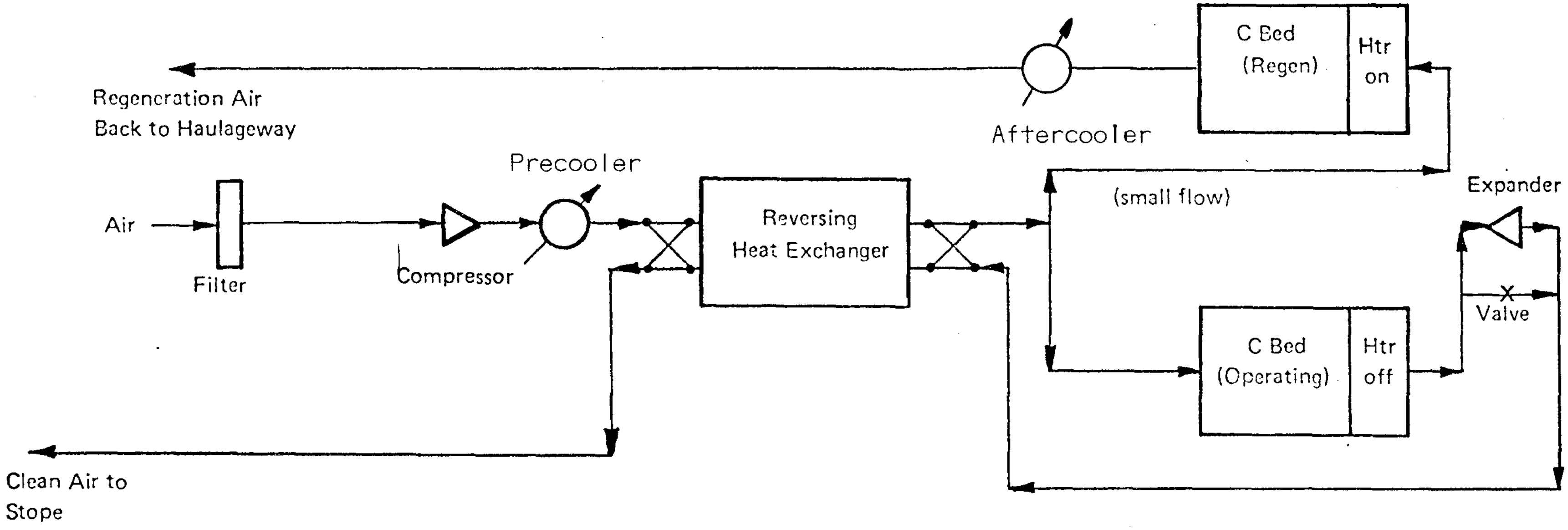


FIGURE 27. CHARCOAL ADSORPTION - DIRECT COOLING BELOW 0°C

b. In the systems that operate below 0°C, we do not know how much of the radon is likely to be adsorbed or otherwise entrapped in the ice freezing out in the reversing heat exchanger. If this turned out to be appreciable, it would mean that the product gas, even if it were radon-free leaving the adsorbent bed, would be recontaminated by this ice-entrapped radon again in its return (warming) passage through the reversing heat exchanger.

8. Safety

With respect to safety, it should be noted that, for the -80°C case with 8-hour beds, if such a bed were suddenly to be warmed to room temperature, it would liberate some 60 SCF of gas which could be contained at, say, 100 psi if a surge volume of $9 ft^3$ were available. (If, on the other hand, the 100 liters of liquid air in the cryogenic case were suddenly to be evaporated, it would give rise to about 2600 SCF of gas, presenting a greater containment problem.) It should also be noted that the sudden combustion of 37 or more pounds of charcoal could lead to safety problems which must be avoided by good design.

A further cautionary note must be added with regard to the radiation hazard associated with longer holdup times. An eight-hour holdup time will eventually produce several millicuries of gamma radiation from daughter product decay. This will decay to an insignificant level during the regeneration cycle, but the average radiation dose would probably be unacceptable without some shielding. The large amount of charcoal required for operation at -20°C will contribute most or all of the needed dose reduction, but at -80°C the bed is much smaller and may therefore need some modest amount of shielding. The 12.7 day holdup system will contain at least a half-curie of gamma activity in continuous operation, but this will be well shielded by distribution in 3/4 ton of charcoal.

9. Cost

The following tabulations give detailed estimates of capital and operating costs for 5000 SCFM units based on three different types of refrigerated charcoal adsorption systems:

a. Indirect cooling to 2°C, alternating beds with 8-hour radon holdup

b. Direct cooling to -20°C, alternating beds with 8-hour holdup

c. Direct cooling to -80°C, single bed with 12.7 day holdup

TABLE 33

Adsorption on Charcoal at 2°C

Annual Operating Cost
5000 SCFM, 2 Shifts, 260 Days/Year

(Capital Cost $207,800)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/yr	$20,000
Utilities			
Electricity	7.33×10^6 kwh	$.01/kwh	73,000
Water	6000 Mgal	$.05/Mgal	300
Supplies			1,000
Maintenance	4% of Capital Cost		8,300
Direct Operating Cost			$102,600
Capital Charges	22% of Capital Cost		45,700
TOTAL ANNUAL OPERATING COST			$148,300
Total Cost per MSCFM			11.9¢

TABLE 34

Adsorption on Charcoal at Various Temperatures Below 0°C
Direct Cooling: Alternating Beds with 8-Hour Radon Holdup; Single Bed with 12.7 Day Radon Holdup

CAPITAL COST - 5000 SCFM UNIT

Standard Equipment (Common to All Systems)	Size	Installed Cost
Filter	5000 cfm	$ 2,000
Compressor	325 HP	140,000
Expander	20 HP	64,000
Precooler	2700 ft^2	8,000
After-cooler	1700 ft^2	6,000
Subtotal		$ 220,000

ADDITIONAL COSTS

Temperature	Radon Holdup Time	Reversing Heat Exchanger	Charcoal	Tanks	Heaters	EOF & Contingency	Total
-20°C	8 hours	$32,400	$1,100	$16,000	$350	$51,000	$320,850
-40°C	8 hours	$43,300	$400	$8,000	$150	$52,000	$328,850
-60°C	8 hours	$51,200	$100	$21,000	$150	$55,000	$347,450
-80°C	8 hours	$60,500	$30	$9,000	$150	$54,000	$343,680
-80°C	12.7 days	$60,500	$600	$8,000	--	$54,000	$343,100

TABLE 35

Adsorption on Charcoal at -20°C

Annual Operating Cost

Alternating Beds, 8-Hour Radon Holdup
5000 SCFM, 2 Shifts, 260 Days/Year

(Capital Cost $320,850)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/yr	$20,000
Utilities			
Electricity	10.4 x 10 kwh	$.01/kwh	104,000
Water	25,000 Mgal	$.05/Mgal	1,300
Supplies			1,000
Maintenance	4% of Capital Cost		12,800
Direct Operating Cost			$139,100
Capital Charges	22% of Capital Cost		70,600
TOTAL ANNUAL OPERATING COST			$209.700
Total Cost per MSCFM			16.80¢

TABLE 36

Adsorption on Charcoal at -80°C

Annual Operating Cost

Single Bed, 12.7 Day Radon Holdup
5000 SCFM, 2 Shifts, 260 Days/Year

(Capital Cost $343,100)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1/2 man/shift	$20,000/yr	$20,000
Utilities			
Electricity	10.0 x 10^6 kwh	$.01/kwh	$100,000
Water	25,000 Mgal	$.05/Mgal	1,300
Supplies			1,000
Maintenance	4% of Capital Cost Cost		13,700
Direct Operating Cost			$136,000
Capital Charges	22% of Capital Cost		75,500
TOTAL ANNUAL OPERATING COST			$211,500
Total Cost per MSCFM			16.95¢

I. SEMIPERMEABLE MEMBRANE/ORGANIC FLUID HYBRID SYSTEM

1. Description of a Preferred System

The selective removal of radon by semipermeable membranes and the absorption of the gas by organic fluids have been known to be technically feasible processes (see Sections B and C), but very costly compared to others that have been studied. Since these are compatible unit operations, a study was made of the possibility of combining them to produce a hybrid. It was felt that there might be a synergistic effect upon radon removal with an accompanying benefit such as a reduction in capital investment and operating cost. The combined membrane/absorber system we have envisioned would place the gas and liquid in contact in a manner similar to that shown in Figure 28.

The advantages of using a membrane as an interface between a fluid adsorbent and the process (air) stream are:

- the absorbent need not be mixed with the air, nor subsequently separated from it.
- the process may be continuous if means are devised to continuously regenerate the absorbent, for example by counterflowing it over the membrane surface and processing it elsewhere.
- the surface area of contact between process stream and absorbent is precisely defined.
- counterflow techniques may readily be employed.
- fluid dynamic conditions determining boundary layer conditions may be controlled.
- the gradient for transport of a species (radon) from the process stream (air) is determined by the concentraion in the process stream and absorption isotherm of the absorbent.
- the differential membrane permeabilities are not critical.

Finding a suitable solvent with respect to chemical, physical and toxicological properties for use in a membrane cascade is, at the very least, difficult.

It can readily be seen that the combined operation of a permeator at 200°F and the use of Freon-12 as an absorbing fluid is not feasible. Operation of the cascade at liquid Freon temperatures would necessitate an increase in membrane area of about 100 times (or an appropriate increase in feed pressure). Both of these possibilities would be economically prohibitive, and thus rule out such a membrane-fluid combination.

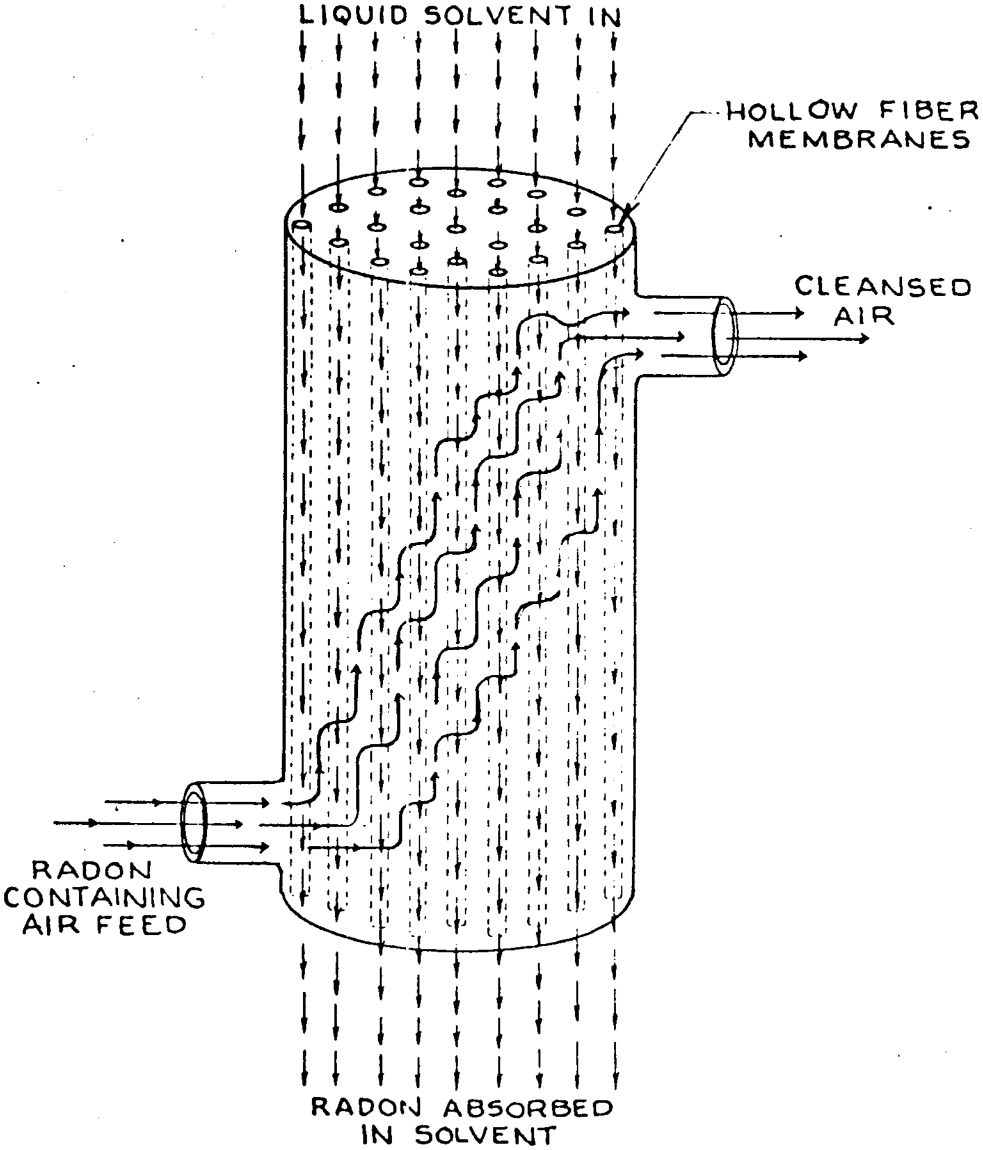


FIGURE 28. SHELL AND TUBE EXCHANGER FOR RADON PERMEATION ABSORPTION

Although costly and probably unsafe for use underground, toluene was chosen as a model solvent because it was judged to have all the other properties necessary for a membrane/solvent hybrid, and because we believe there is a possibility of finding or creating a substance that would be safe and inexpensive as well as effective. Toluene has a solubility parameter similar enough to radon and so different from oxygen and nitrogen that radon is most likely to be preferentially absorbed. There is also the added advantage that radon solubility data are available for toluene over the temperature range 32-140°F. Data for the Bunsen absorption coefficient were extrapolated to 200°F to provide a modified Henry's law constant and the appropriate solubility. As an example of the temperature dependence of the hybrid system, decreasing the temperature from 200°F to 75°F increases the solubility by a factor of about 4, but increases the membrane area by about 30 times. Thus, 200°F was chosen as the temperature for the combined system; one should note that at 200°F toluene (B.P. = 231°F) has a vapor pressure of about 425 mm Hg.

In order to understand the advantages of a membrane/organic fluid process, gas transport through a semipermeable membrane will be described. The equation of mass flux for one particular gaseous species across a membrane is:

$$q = \frac{A}{t} \underline{P} (P_R - P_p)$$

where
q = mass flux
A = area of membrane
t = thickness of the membrane
$\underline{P}$ = membrane permeability constant
P_R = partial pressure in the raffinate (high pressure side)
P_p = partial pressure in the permeate (low pressure side)

Reducing the partial pressure of radon in the permeate increases the partial pressure driving force and requires less membrane area. Usually the permeate and raffinate partial pressures are determined by the feed conditions, flow rates, membrane area, permeabilities, and pressure drop across the membrane. However, the addition of an organic liquid with a low partial pressure of radon on the permeate side aids mass transport. The ideal case for a maximum driving force would occur when the partial pressure in the permeate is zero, or

$$q = \frac{A}{t} \underline{P} P_R$$

The percentage of ideal transport may be thought of as a membrane stage "efficiency" defined as $P_R - P_p/P_R \times 100$.

For example, examination of the data in Table 15 (Section C) shows that the efficiency for Stage 3 in the schematic diagram of Figure 17 would be 50%. In order to reach 97% efficiency, the permeate partial pressure would have to be decreased by 80%. Using Henry's law, calculations were made to determine the concentration of radon in toluene which would correspond to this reduced partial pressure. It was found that 7.5×10^{-18} moles of radon would be absorbed in one liter of toluene. For the sample Stage 3, 1.8 cubic feet of solvent would be required per SCF of permeate gas. There would also be an accompanying decrease in membrane area, or alternatively, a reduction in the feed pressure. Because of the high cost of compressors, such a decrease would be welcome. In this example, the 80% reduction in partial pressure would lead to a decrease in the membrane area of approximately 97%. Alternatively, the feed pressure could be decreased from 20 atm to approximately 1.2 atm., a 94% reduction. It is possible that fans might even be used to replace the compressors at such low pressures. However, there is a problem in that the composition of radon in the raffinate would be increased about threefold, if the permeate partial pressure is reduced 80%. This change would require additional stages to reach the desired effluent specific activity. Alternatively, the raffinate concentration may be decreased by not reducing the membrane area (or pressure) as much as predicted.

In order to examine the merits of this conceptual hybrid system, a hypothetical design was developed to illustrate major considerations. For reasons that have already been cited, the basic design has rested on the assumption that toluene (or a functionally similar liquid) would be used to absorb radon which had first been selectively extracted from mine air by permeation through polyethylene membranes at 93°C (200°F). Initial concentration of the radon in air was assumed to be 500 pCi/l, and the air input was assumed to be 5000 SCFM. For the purposes of this example, the toxicity, flammability, and chemical compatibility of toluene in the model system were assumed to be acceptable. Although in fact toluene would probably not be completely suitable on all these counts, we felt it to be an adequate model for the present purpose.

We have already shown that the use of toluene as an absorber for radon that permeates through the membrane could allow the feed pressure in the permeator to be reduced from 20 atm. to 1.2 atm., permitting a dramatic saving in compressor costs. However, this benefit was accompanied by a disadvantage in that more stages would be required to reduce the radon concentration by the desired factor of ten. Further work in simulating an entire cascade of toluene/membrane absorbers revealed that 1.2 atm. is a theoretical minimum and would require an infinite number of stages. A feed pressure of 3 atm. would require over 50 stages, and 5 atm. would require about 15 stages. The calculations for optimization of a cascade of organic fluid/semipermeable membrane absorbers are quite involved and would require the development of a computer program for accurate solutions. In the absence of such a program, only rough estimates can be made for design optimization.

To keep the number of permeator stages to a reasonable level, the system described here was designed for operation at 5 atm and has a total of 15 permeation stages. The membrane area of each of the stages is assumed to be 300,000 ft^2, making a total of 4.5×10^6 ft^2 of permeable membrane area. Each permeator contains a bundle of 2.3×10^6 polyethylene fibers (ID = .015", OD = .016"). The flow of toluene through all the fibers of a stage is estimated to be 310 ft^3/min. For an average permeator length of 12 feet, there is a pressure drop of 1.3 atm. associated with pumping the fluid through the small capillary tubes.

A solvent regeneration and recovery system is required which will be capable of processing all of the toluene used (6.7 x 10^6 ft^3/24-hour day) in the facility. A total volume of about 80,000 gallons will be required, and all of this will be completely recycled about 84 times in each 24-hour day.

The contaminated liquid toluene is pumped to a purging unit where it is discharged into four bubble tanks, each 15 ft in diameter and 12 ft high, operating at 0.6 atm. Air, radon, and a small quantity of toluene are rejected to the waste air stream. Clean toluene liquid (at its boiling point) is withdrawn from the bottom of each tank. The majority of this liquid is returned to the permeators, but a small fraction is sent to a reboiler which delivers toluene vapor to the bottom of each tank. Vapor is withdrawn from the top of each tank by a liquid-seal vacuum pump through an extended condensing section. Most of the toluene vapor is recovered, liquefied, and returned to the tank.

It is important to note that no provision was made in the calculations for the non-radon portion of the permeate gas. The flow of these gases (N_2, O_2, CO_2, A, etc.) through the membrane will be substantial and could affect our estimates of heat and mass transfer, and of flow conditions within the polyethylene fiber. This is a major consideration, the effect of which requires extensive calculations which are beyond the scope of the present study.

2. Efficiency and Reliability

a. Effectiveness of Radon Removal

This sample system is designed for removal of 90% of the radon in the incoming air. The remaining 10% is to be contained in a gas volume not

b. Size and Weight of Equipment

The absorption equipment is composed primarily of the air compressor and filter (approximately 4' x 4' x 8', 5 tons), the permeators (15-16' x 3' cylinders, total 60 tons), and the air cooler (approximately 3' x 3' x 5', 1 ton). All of this could be combined in a package occupying no more than 9' x 15' x 20' = 2700 ft^3.

The purging system includes: the four buble tanks, each 15'D x 12', containing 209 tons of liquid toluene (58,000 gallons); the reboiler, conde condenser, vacuum pump, and liquid pumps, all of which are relatively small. This would combine to make a unit of about 11,000 ft^3.

c. Residence Times of Radon and Air in the Device

At a feed flow rate of 5000 SCFM, the residence times of radon and air in the cascade are less than one minute. Holdups in the regeneration/recovery unit would be on the order of a few minutes.

d. Deterioration and Its Remedies

A principal cause of deterioration in the system described above would be attack of the membrane surface by the solvent. At 200°F (93°C), polyethylene is not inert to chemical reaction with certain solvents. Selection of a more inert solvent than toluene, or a compromise in operating temperature would eliminate the problem. The membrane itself can easily withstand 5 atm. pressure and would require only regularly scheduled inspection for pinholes and removal of surface contaminants. The membrane life would depend upon actual conditions, but at low pressures and with a compatible solvent it is expected to be on the order of a year. The organic fluid requires no attention or replacement, with a satisfactory solvent recovery and regeneration system. The latter system for recovery/regeneration and the necessary process equipment, i.e., pumps and compressors, would be subject to periodic maintenance.

3. Safety

The major safety hazard of a toluene/membrane system would be due to the flammable nature of the solvent. At room temperature the lower and upper explosive limits are 1.4% and 6.4% respectively. (Note that this range expands with increasing temperature.) Also, since toluene vapor is heavier than air, a real safety problem arises. Therefore, the system would have to be completely sealed and the possible use of explosion proof equipment considered. There is also a health hazard associated with toluene vapors, which the sealed system would eliminate. It should be noted that a non-flammable solvent may already exist, whose ability to dissolve radon is at least as good as toluene's. The feasibility of the system actually depends on the use of such a solvent.

There is no significant holdup or decay of radon in the membrane or in the permeator itself and the contact time of solvent is less than 10 sec. The concentration of radon in the effluent liquid is about 0.1 $\mu Ci/ft^3$ (3500 pCi/l). Virtually all of this radon would be vented to waste air, resulting in a negligible radiation hazard from accumulation of decay in the recirculated solvent.

Other than toluene, there are no toxic byproducts produced in the system. There is some oxygen removal in the purged air, but not of a large magnitude.

Noise would be generated by the compressors and liquid pumps, but the level should be acceptable; if desired, acoustical insulations could be installed.

4. Cost

a. Capital Costs

Capital costs are outlined in Table 1 as estimated by conventional equipment factoring cost procedures. Total plant capital cost is estimated to be $1,678,000.

b. Annual Operating Costs

Annual operating costs are outlined in Table 2. Most per unit rates are outlined in the table. Electric power must be provided for approximately 1000 total motor horsepower including the air compressor, liquid pumps, and the condenser refrigerator. Total cooling water consumption is approximately 120 gpm based on 60°F ambient water temperature. A small amount of toluene will be lost in the air stream leaving the vacuum pump. Total annual operating costs are around $516,000 per year or 41.3¢ per 1000 SCF of air processed.

TABLE 37. MEMBRANE PERMEATION AND ABSORPTION IN IN AN ORGANIC LIQUID

CAPITAL COST - 5000 SCFM UNIT

Equipment	Size	Installed Cost
Air Filter	5000 SCFM	$ 2,000
Air Compressor	650 HP	260,000
Liquid Pumps	389 HP Total	66,000
Air Cooler	3700 ft^2	7,000
Organic Liquid (Toluene)	290 tons	36,000
4 Bubble Tanks	15'D x 12' high	250,000
Vacuum Pump		2,000
Condenser		12,000
Refrigeration	1 ton	4,000
Permeators	4.5×10^6 ft^2	700,000
Pipelines	200 ft	12,000
Sub-Total		$ 1,351,000
Engineering Fee and Contingency 15%		203,000
TOTAL		$ 1,554,000

TABLE 38. MEMBRANE PERMEATION AND ABSORPTION IN AN ORGANIC LIQUID

ANNUAL OPERATING COSTS

5000 SCFM, 2 shifts, 260 days/year

(Capital Cost $1,554,000)

	Requirements	Unit Cost	Annual Cost
Operating Labor & Supervision	1 man/shift	$20,000/yr	$ 40,000
Utilities			
Electricity	3.10 x 10^6 Kwh	$0.01/Kwh	31,000
Water	30,000 Mgal	$0.05/Mgal	1,500
Toluene Make-up	1,245 gal	$0.70/gal	900
Reboiler Fuel	0.7 x 10^6 Btu/hr	$2/MMBtu	5,800
Maintenance	4% of Capital Cost		62,000
Direct Operating Costs			$141,400
Capital Charges	22% of Capital Cost		341,900
Total Annual Operating Cost			$483,300
Total Cost per MSCF			38.8¢

J. ORGANIC FLUID/SOLID ADSORBENT HYBRID SYSTEM

In 1966, R. Reiter[1] reported some surprisingly favorable results from an experiment in removal of radon from ambient air. The results were surprising because they would not have been predicted from theoretical considerations and because they appeared to contradict some earlier experimental work that had been meticulously documented and which was consistent with theory.[2] Reiter's experiment consisted of building a recirculating impingement device, through which air could be passed at a rate of 50 m^3/min over a bed of porcelain Raschig rings, on which was a coating consisting of 360 g of "Aerosil" silica dispersed in 6 kg of tricresyl phosphate. The author did not attempt to explain, nor had he predicted, any specific advantage that might be realized by coating the surface of an active solid sorbent for radon with a viscous liquid such as tricresyl phosphate. Rather, he started with the knowledge that certain organic liquids have a significant capability to absorb radon gas. Then, having observed that one of the best of these, tricresyl phosphate, had a Bunsen coefficient, α, equal to 4.3 at 20°C, he concluded that an organic

$$\alpha = \frac{\text{Concentration of radon in TCP}}{\text{Concentration of radon in air}}$$

liquid alone would not have sufficient capacity to significantly reduce the radon concentration in a total volume of 50-100 m^3 of air. For reasons that he has not made clear, Reiter decided that the addition of a solid adsorbent would permit the radon to be "continuously extracted" from the TCP. For this purpose he chose MOX-60 "Aerosil" silica, which has a surface area of 60 m^2/g. Operation of the assembled system for 20 minutes in a closed room of 60 m^3 volume, containing an initial charge of 222-radon in air, resulted in a reported reduction of the radon concentration to a "limiting value" of 20-25% of its initial concentration. Continued operation thereafter produced no further reduction.

In 1968, two years after Reiter's publication, D. Bird and A. C. Pitsas of the General Electric Company[3] conducted an in-mine test of the TCP-silica radon scrubber as part of a general test of filtration and electrostatic precipitation, most of which was concerned with radon daughter removal. (It is interesting to note that both Reiter's and the G.E. team's reports were primarily concerned with topics other than radon removal, and that the radon removal experiments were reported only in a peripheral way, with few data and little discussion of theory.) The G. E. device included a Reiter-type TCP/silica bed in an experimental filtration system which passed 17 m^3/min of air continuously for a period of 2½ hours. Measurements of 222-radon concentrations at the entrance and exit of the scrubber section showed an essentially constant 30% reduction in radon concentration throughout this period.

Unfortunately, neither Reiter nor Bird and Pitsas felt constrained to account for experimental anomalies between their results and those of other workers. Bird and Pitsas found no apparent 20-minute saturation limit for radon such as Reiter had observed for the TCP/silica bed.

Reiter's claim of 80% limiting removal implies that the bed came to equilibrium having picked up an amount of radon equal to four times that which still remained (at equilibrium) in the 60 m^3 of room air. The corresponding dynamic adsorption coefficient k for radon on silica is 4 x 60 x 10^6 cm^3/360 g silica = 6.67 x 10^5 cm^3 air/g silica. (We neglect here the capacity of the TCP, since Reiter's reported Bunsen coefficient of 4.3 means that the 5 liters of TCP itself could have held the radon from only 4.3 x 5 ℓ/60,000 ℓ = 0.4% of the room air.)

Bird and Pitsas' claim of a steady 2.5-hour reduction of 30% in radon level across the unit implies that the bed picked up the radon from 30% of the total flow over the 150-minute period. At 17 m^3/min, this means that the bed picked up the radon from 0.30 x 150 x 17 m^3 of air. Since the bed was still not apparently saturated, as the performance was not deteriorating, this implies a dynamic adsorption coefficient k for radon on silica of more than 0.30 x 150 x 17 x 10^6 cm^3 air/360 g silica = 2.13 x 10^6 cm^3 air/g silica (again neglecting the capacity of the 5 ℓ of TCP).

The two apparent values of k underlined above are orders of magnitude higher than those reported in the literature for radon on silica (and indeed on charcoal as well). The data of Burtt and Kurbatov[2] show a value of k for radon from air on silica of only 28.5 cm^3 air/g silica. Not even Reiter is willing to argue that the presence of TCP should increase this value; in his paper he states that the TCP acts "only as a means of transportation from the gas/liquid interface to the liquid/solid interface". (Indeed, he never demonstrates just why the TCP should not be expected to act simply as an added obstacle to the contacting of the radon with the silica.)

If we reject as very unlikely the hypothesis that "Aerosil" silica is over 20,000 times more active than Burtt and Kurbatov's silica, or that the TCP really has more than 10,000 times the Bunsen solubility coefficient for radon reported by Reiter, or that the porcelain Raschig rings used for support were really the active agent, then we are faced with the more probable explanation: the radon level at the sample point in Reiter's room may have decreased for other reasons when air was circulated, and the sampling and/or analysis of radon gas may not have been done satisfactorily by either set of investigators.

In view of the wide discrepancies between the scanty data offered by these two investigators and the incompatibility of their results with any theoretical hypothesis based on known solubility coefficients for radon in TCP and dynamic adsorption coefficients for radon on silica, we cannot find in them any valid basis on which to propose a feasible radon removal unit for consideration in this study. Even if we were to grant the extremely unlikely possibility that an effective retention coefficient (combining α and k) in the order of 6 x 10^6 could be achieved -- a conclusion that can be reached by an optimistic extrapolation of Bird and Pitsas' data -- the cost for attaining a ten-fold reduction of radon concentration in a 5000 SCFM air supply would be at least twice that which we have estimated for our least expensive technique, charcoal adsorption.

We have therefore concluded that a hybrid system combining absorption in an organic liquid with adsorption on a solid substrate would not be more effective than either technique used alone, and is therefore not worthy of further considerations.

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K. OTHER PROPOSED HYBRID SYSTEMS

1. Membrane Permeation, Cryogenics and Activated Charcoal

No technologically feasible technique was found which would improve the effectiveness of removal of radon from air by combining membrane permeation with cryogenics and activated charcoal. Cryogenic concentration, as described in Section E, suffers from no limitation that would benefit from the addition of membrane separation or charcoal adsorption at any part of the system. Membrane permeation as described in Section C would benefit from an effective reduction of the partial pressure of radon on the permeate side of the membrane, but this is not practically attainable in continuous operation by the use of a solid adsorbent. Elevated temperature regimes are generally preferable to reduced temperatures for membrane permeation, so that refrigeration does not appear to be an advantageous adjunct to such a system.

One possibility remained which, for a time, appeared to show some advantage. A charcoal adsorption system, operated, as we have described in Section H-5, at -80°C without periodic regeneration to discharge radon and other adsorbed impurities, may suffer a reduced operating life expectancy if impurities such as H_2O, SO_2, NO_2, CO_2 and vapors of various organic substances contaminate and poison the adsorbent's surface. A form of pre-scrubbing or pre-filtration to remove some or all of these impurities might therefore be beneficial in extending the useful life of the charcoal bed. Considerations of the use of membrane permeation for this purpose was based on the hypothesis that while oxygen and nitrogen travel through the membrane much more slowly than does radon, such impurities as H_2O, SO_2, NO_2 and CO_2 would be expected to permeate even more slowly than O_2 and N_2. Hence, the membrane could be used as a prefilter to remove impurities which might otherwise tend to poison the adsorbent; the permeate from the membrane separation would be a lower volume air stream and although enriched in radon, it would presumably be depleted in the impurities which lower the effectiveness of the charcoal.

Contrary to this hypothesis, however, the separation factors through silicone rubber and polyethylene membranes for H_2O, SO_2, and NO_2 versus radon proved to be considerably greater than 1.0 (See Table 39). Thus, radon and the permeate stream would actually be more highly contaminated with these three impurities than the feed stream. It is therefore evident that the use of membrane separation as an accessory to a refrigeration/adsorption concept is *a priori* unworkable.

2. Membrane Permeation and Chemical Reactions

No technologically feasible way was found in which to combine membrane permeation with chemical reactions so as to improve the effectiveness of either technique alone. As indicated earlier, membrane permeation would benefit from any means of reducing the partial pressure of radon on the permeate side of the membrane, but this is not practically attainable in continuous operation by the use of a solid reagent such as dioxygenyl antimony hexafluoride. Liquid or gaseous reagents such as BrF_3 would be

TABLE 39

SEPARATION FACTORS FOR CONTAMINANTS VS. RADON THROUGH POLYETHYLENE AND SILICONE RUBBER MEMBRANES

Component	α_{i-Rn}*	
	Silicone Rubber	Polyethylene
H_2O	11.1	2.9 - 17.2
SO_2	4.6	-
NO_2	2.4	-
CO_2	1.0	0.36
NO	0.18	-
O_2	0.18	0.08
CO	0.11	0.08
N_2	0.09	0.03

* The separation factor, α_{i-Rn}, is the ratio of the permeability constant for component i, to the constant for radon, i.e., P_i/P_{Rn}.

effective in principle, since they have sufficient mobility to serve as a liquid transport medium for the radon, but the necessarily extraordinary chemical reactivity of these substances would make them difficult to contain in contact with a suitable membrane and would present an unacceptable safety hazard.

The possibility of using a membrane filter as an adjunct to the chemical reaction system described in Section F was found to be without apparent advantage. The principal hazard to the performance of that system is due to the competition of water for the highly reactive dioxygenyl antimony hexafluoride. Membrane permeation is not an effective technique for removal of water from air-radon mixtures, and is therefore not preferable to molecular sieves for that purpose.

3. Cryogenics and Chemical Reactions

As indicated in subsection K-1 above, the cryogenic condensation system is not hampered by any specific limitation that could conceivably benefit from the use of chemical reaction. Conversely, the chemical reaction system might benefit from the use of cryogenic condensation for removal of excess water from the input air in order to protect the reagent from early consumption. Any reagent which can effectively remove radon from mine air must have extraordinary reactivity as an oxidizing agent. The only compounds known to be effective for this purpose are at least equally reactive with water, so that input air to the chemical scrubber must be subjected to extremely thorough dehydration before it encounters the active reagent. Cryogenic concentration was considered for this purpose in our study of the chemical reaction system (Section F-1.d) and was found to be economically justifiable only when used in a very rudimentary sense as an adjunct to the molecular sieve. In this context the technique is more properly termed "refrigeration", since the air is cooled only to 2°C, so that excess moisture can be removed as liquid condensate leaving the remaining dehydration to the molecular sieve. A dehydration strategy based on the use of cryogenic condensation alone would require a system essentially identical to the one proposed in Section E for total removal of radon itself. No other technically feasible and advantageous system was proposed.

APPENDIX A

ADVANCED TECHNIQUES FOR RADON GAS REMOVAL

BIBLIOGRAPHY

An updated bibliographical listing of selected references has been compiled herewith. This listing contains the citations from the "Preliminary Bibliography" of November 1973 with some additions.

The Bibliography has been organized by subject - techniques for radon gas removal. Techniques considered include:

- adsorption
- chemical reactions
- organic fluids
- centrifigation
- membrane filters
- cryogenics

There is also a "general" section, which contains references to material related to radon gas removal from uranium mine atmospheres.

Within each subject section of the Bibliography, the citations are listed alphabetically by author. Papers without authors given are listed at the end of each section. Multiple references by an author are listed in chronological order.

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APPENDIX B

PRINCIPAL INVESTIGATORS

The study whose results are reported herein was conducted by a team of professional scientists and engineers, all of whom are staff members or consultants to Arthur D. Little, Inc.. While many of the judgements and conclusions were reached in conferences involving various groups of these participants, the principal research work was performed by the individuals whose task assignments are listed below. Brief biographical resumes of the major investigators are attached.

Program Director: Donald B. Lindsay

Documentation: Ann J. Whittier

Mining Technology and Safety: William I. Watson

Radiation Safety and Radon Technology: Robley D. Evans, Gerald L. Schroeder, Constantine J. Maletskos (consultants) and Donald B. Lindsay

Engineering Cost Assessment: Stanley E. Dale

Report Production: Anne J. Carlisle

Technology Assessment:

Charcoal Adsorption: Edwin L. Field (Basil U. Igwe)*

Organic Fluids: J. Ronald Lawter (Edward Interess, J. Kevin O'Neill)

Semipermeable Membranes: John M. Ketteringham, Lita L. Nelsen, Lawrence N. Davidson (Ronald Johnson)

Molecular Sieves: Edwin L. Field (J. Kevin O'Neill)

Cryogenics: Howard O. McMahon (Basil U. Igwe)

Chemical Reactions: J. Ronald Lawter (C.J. Santhanam)

Centrifuge: Reed H. Johnston

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*Names in parentheses are those of engineers who assisted in preparation of individual system designs and cost estimates.

DONALD B. LINDSAY (Program Manager)

A.B., Chemistry, Wesleyan University, 1949

Tracerlab, Inc., 1949-1951
Technical Operations, Inc., 1951
U.W. Air Force Cambridge Research Center, 1951-52
Arthur D. Little, Inc., 1952-

Areas of Specialization

Analytical Chemistry
Radiation and Radiotracer Technology
Research Planning and Management

Experience

Mr. Lindsay is a Unit Leader in the Chemical Systems Section of Arthur D. Little, Inc. His professional training has been primarily in radiochemistry and analytical chemistry, areas in which he has done original research and has directed applied research and development projects for many years. His experience in the application of these specialties to problem-solving tasks for industrial firms and various government agencies has included the planning and direction of a number of interdisciplinary projects requiring the combined efforts of a variety of technical specialists in physical and biological sciences, economics, engineering and industrial management.

Among the major projects in which Mr. Lindsay has had a significant role are: (1) appraisal of the technical merits, manufacturing and probable market demand for a novel diagnostic x-ray recording system; (2) development of instrumentation for monitoring of radioactive gas emissions from a cryogenic noble gas retention system designed for use in nuclear power reactors and/or fuel reprocessing operations; (3) assessment of business opportunities in the control of health and environmental damage hazards in the nuclear power industry; (4) survey of business opportunities in the development and sale of products for nuclear medicine technology; (5) study of the feasibility and probable cost to the U.S. uranium mining industry of complying with a proposed tightening of government-imposed limits on occupational exposure to airborne radioactivity in underground mines; (6) engineering feasibility assessment of advanced techniques for reduction of ultra-trace-level concentrations of radon gas from mine atmospheres; (7) development of a novel personal alpha radiation monitor for use by underground uranium miners; (8) survey of advances in electro-optical technology and assessment of their probable impact on scientific and medical instrumentation markets in the 1970's; (9) assessment of the nature and extent of needs for analytical chemistry capabilities in the research laboratories of a major U.S. pharmaceutical firm; (10) development of a five-year research plan to improve system maintenance and repair technology for an association of U.S. utilities companies; (11) assessment of the need for applied research and new-product development capabilities for an established manufacturer of household appliances; (12) assessment of available technological options for new-product development for an established manufacturer of industrial photocopying equipment.

Mr. Lindsay is a member of the Society for Applied Spectroscopy, the Health Physics Society and the AAAS. He is also the chief radiation safety officer for Arthur D. Little, Inc.

STANLEY E. DALE

Stanley E. Dale joined Arthur D. Little, Inc. in 1960; presently is a member of the staff of the company's Chemical Engineering Section.

Education

Bachelor of Science in Chemical Engineering, University of Michigan, 1949 (Tau Beta Pi)

Master of Science in Chemical Engineering, University of Michigan, 1950

Pre-ADL Experience

Before joining ADL, he had 10 years' experience with Callery Chemical Company, where he was in charge of the design, procurement, construction and operation of pilot plants for the production of boron hydrides. He was the project engineer for three major process units of Callery's $35 million high-energy boron fuel plant at Muskogee, Oklahoma, and was a process supervisor for the start-up and initial operation of those units.

Experience at ADL

At Arthur D. Little, he has been active in the areas of polymer production, insecticide manufacture, dyestuff manufacture, cooling water and water-treatment systems, environmental control systems, total energy systems, heavy organic synthesis, various petrochemical economic evaluations, cryogenic purification systems, and the like.

He has done equipment development on continuous filtration devices, gas-fired burners, pulsed autoclaves, vacuum distillation units and polymerization systems. He has performed technical and economic analyses of a unique oxidation process to produce a fine organic material, a complete analysis of an operation for producing germicides, waxes and similar materials, and has prepared economic analyses on processes for producing such materials as naphthalene, phenol, acrylonitrile, acetaldehyde, town gas, color films, vinyl acetate, graphite, styrene and liquefied natural gas. He has been active in the project management of nitrogen fertilizer and LNG plants. A technical-economic evaluation of a petrochemical project in Korea and a fertilizer project in Pakistan have been completed by him.

Mr. Dale participated in the process and mechanical design of a caustic scrubbing system to remove sulfur dioxide from boiler flue gases. He assisted in the preparation of the equipment and construction specifications for this scrubbing system.

Memberships

American Institute of Chemical Engineers
American Chemical Society
Registered Professional Engineer, Commonwealth of Massachusetts

LAWRENCE DAVIDSON

Lawrence Davidson joined Arthur D. Little, Inc., in 1972 and is a staff member of the Chemical Systems Section. He obtained his B.S. in chemical engineering in 1970, and his M.E. in chemical engineering in 1971, from Rensselaer Polytechnic Institute. His interests include computer simulation, reaction kinetics, and air and water quality control.

Prior to joining Arthur D. Little, Inc., he was engaged in new product and process development for a radiopharmaceutical firm. He has also carried out process design and pilot plant development of a static mixer for paper pulp, under a fellowship awarded by Westvaco Corp.

Mr. Davidson has worked on a contract for the Environmental Protection Agency to study the chemistry and kinetics of limestone scrubbing for the removal of sulfur dioxide from stack gases. The study was motivated by a need to understand the mechanism of precipitation and scale formation which lead to system unreliability. He was responsible for day to day process control and data analysis of a bench scale scrubber. Also included in the study was the development of a computer model which defines the complex equilibria in the system. Presently, he is working on a similar EPA project investigating operating parameters of the double alkali process for sulfur dioxide removal. This includes supervision of bench scale work in conjunction with ADL pilot plant operations.

He was part of the team working for the Coast Guard to classify oil and hazardous chemical spills and recommend control procedures as part of the Chris manual. A large portion of the work entailed research and data gathering for the list of hazardous substances. For the EPA, Mr. Davidson has also been involved in sampling and analysis of waste water for determining effluent guidelines of the plastics industry. Recently, he has been engaged in evaluating a process for the treatment of Claus tail gas.

Mr. Davidson is a member of Phi Lambda Upsilon, Tau Beta Pi, the American Chemical Society and the American Institute of Chemical Engineers. He received his Engineer-In-Training certificate from the Commonwealth of Massachusetts in 1972.

ROBLEY D. EVANS (Consultant)

Dr. Evans attended the California Institute of Technology, where he received his doctorate in Physics in 1932. In 1934 he came to the Massachusetts Institute of Technology as assistant professor of physics, was advanced in 1938 to associate professor and in 1945 became professor of physics. For his application of radioactive isotopes to wartime problems in World War II, Dr. Evans received this country's second highest civilian award, the Presidential Certificate of Merit, in October of 1948. He was awarded the Silvanus Thompson Medal of the British Institute of Radiology in London on June 9, 1966, in recognition of his outstanding contributions in the field of radiation protection and safety, and delivered at that time the Silvanus Thompson Memorial Lecture on "The Effects of Skeletally Deposited Alpha Ray Emitters in Man".

Dr. Evans' work with the Government includes service as consultant to the Office of Scientific Research and Development, the Army, the Air Force, the Navy, the U.S. Public Health Service, the U.S. Department of State, the Surgeon General, the Secretary of Defense, Research and Development Board of the National Military Establishment, the Federal Radiation Council, and the Division of Biology and Medicine of the Atomic Energy Commission. He is currently a consultant to the Federal Aviation Administration on the radiation-biology aspects of supersonic transport.

Dr. Evans is author or coauthor of more than 150 scientific papers on statistics, interaction of radiation in matter, medical physics, radiation standards, biological effects of radiation, industrial applications of radiation and of radionuclides, and other topics in pure and applied radioactivity; and holds membership in many technical and professional societies.

EDWIN L. FIELD

Mr. Field received a B.Ch.E. from the College of the City of New York and an S.M. in Ch.E. from the Massachusetts Institute of Technology. He is a member of the American Institute of Chemical Engineers, the American Nuclear Society, and the American Institute of Mining, Metallurgical and Petroleum Engineers, and he is a Registered Professional Engineer in the Commonwealth of Massachusetts.

Mr. Field joined the staff of Arthur D. Little in 1962, with some eleven years of prior experience. At Tracerlab, Inc., he had designed and supervised the construction and operation of plant-scale equipment for the cryogenic separation of gases. As a senior supervisor at DuPont's Savannah River Plant near Aiken, South Carolina, he had provided technical assistance in the production of radioactive materials for the U.S.A.E.C.

At Arthur D. Little, Mr. Field has applied his practical chemical engineering experience and his familiarity with computer techniques to studies in the metals, mineral and nuclear industries. Some major assignments have been:

Technical/economic metallurgical feasibility studies. Primarily nickel and copper smelting, iron ore and titanium dioxide beneficiation, and steel sheet manufacture. These involved study of process alternatives, preparation of estimated capital and operating costs, and ultimately of cash flows. In most cases, computer simulation of all or part of the enterprise was required.

Process development studies. Theoretical and laboratory work on the solar-evaporative crystallization of multicomponent salt solutions, on the reactivity of lime/limestone with sulfite/sulfate species in spent SO_2 scrubber solutions, and on recovery of trace amounts of As from solids.

Air pollution control studies. Sampling and analysis of sulfide ore smelter ducting systems, economic analysis of the costs of recovery of sulfur values in alternative forms, costs of recovery of nitrogen oxides from stack gases, and evaluation of petrochemical smog studies.

Nuclear Studies. Inspection of nuclear reactor fuel elements, design and testing of adsorptive traps for the gaseous fission products arising in nuclear reactors, economic analysis of uranium isotope separation by gaseous diffusion, economic modeling of the nuclear power supply industry, and application of PERT techniques to research activities in the U.S. breeder reactor development program.

Computer simulations. Radiant heat networks, refrigeration and ice-making systems, multi-stage leaching, and crystal brining operations.

Mr. Field is also the author of several technical publications.

REED H. JOHNSTON

Dr. Johnston obtained his B.Sc. in mathematics and physics at McGill University in 1943, following which he worked on resonance slots and microwave antennas and in medical electronics. He was engaged in the study of atmospheric diffusion and its applications to chemical warfare and returned to graduate study at Stanford University in 1948, where he received his M.Sc. in physics and his Ph.D. in physical chemistry. As an employee of the Defense Research Board of Canada from 1945 to 1961, Dr. Johnston has participated in a broad range of activities which included shock tube investigations, nuclear measurements, design of instrumentation and interpretation of data in the measurement of the spectra of gamma flash in United Kingdom nuclear weapons tests, applications of digital techniques to cryptology, optimized programming of recirculating memory computers, analysis of effectiveness of military systems.

Dr. Johnston joined the staff of Arthur D. Little, Inc., in the summer of 1961 and his efforts continue to be broad based in the application of physics to technical problems.

JOHN M. KETTERINGHAM

Dr. Ketteringham is a physical chemist who was graduated from Oxford University, England, with the degrees of M.A. and D.Phil. He joined Arthur D. Little, Inc., in 1964 and is now Manager of the Chemical Systems Section. This section comprises about 50 staff members who apply a chemical training to a variety of laboratory based and other technical problems in the environmental, energy, and biomedical areas.

At ADL, he has combined his technical interest in membrane technology, electrochemistry and chemical kinetics with a variety of survey, market research and technical evaluation studies. While he has specialized personally in recent years in the medical products area, he still retains an active involvement in environmental technology assignments, particularly those carried out in his section. He contributed to a recent evaluation of membrane techniques for the removal of radon from mine atmospheres.

He serves on a number of review committees on environmental subjects in Arthur D. Little, Inc.

Dr. Ketteringham is a member of the American Association for the Advancement of Science, the Electrochemistry Society, and the American Society for Artificial Internal Organs.

JAMES RONALD LAWTER

B.S., in Chemistry, University of South Carolina, 1965
Ph.D., in Physical Chemistry, Massachusetts Institute of Technology, 1971
Research Associate, Boston College 1971-1972

Dr. Lawter has been a member of the Analytical Chemistry group of Arthur D. Little, Inc., since 1972.

AREAS OF SPECIALIZATION

Combustion Chemistry
Dispersion Modeling
Air Pollution Analysis
Source Sampling

EXPERIENCE

Dr. Lawter is currently responsible for computing air quality using computer dispersion models for several projects undertaken for industrial clients. He has had extensive experience in computer modeling techniques involving contour mapping and projection of pollutant concentration.

Since joining Arthur D. Little, Inc., Dr. Lawter has been involved in the measurement of gaseous emissions from stationary sources, a study of jet combustor exhausts, and an investigation of the effects of fuel droplet-size on emissions from liquid spray flames. He has also conducted a program for the evaluation of hydrogenation catalysts and is currently involved in programs for evaluation of techniques for the removal of radon from uranium mine atmospheres and for the evaluation of devices for the measurement of emissions from industrial sources.

At Boston College, he used optical and mass spectroscopic methods for the study of flames as a part of a U.S. Navy project for research on the chemical kinetics of propellant reactions. As part of this program he also studied the properties of species produced in electrical and microwave discharges.

His doctoral thesis research was in chemical kinetics and was concerned with the molecular interactions involved in chemical reactions.

He is a member of the American Association for the Advancement of Science and the American Chemical Society.

CONSTANTINE J. MALETSKOS (Consultant)

Dr. Maletskos received the S.B., S.M., and Ph.D. degrees in biophysics, all from the Massachusetts Institute of Technology. He is an acknowledged expert in the areas of radiobiology, radiation measurement, and radiation protection and safety. For the past twenty years, he has served as a consultant or staff member to a number of hospitals, universities, and professional and government committees regarding radiation as applied to biology, chemistry and physics.

His experience includes radiation protection planning (program and plant layout), environmental planning and programs, training and education, radiaation emergency planning, and planning for the radiation aspects of plant medical programs. He designed, organized and directed the radiation protection program of the Massachusetts Institute of Technology Research Reactor, including the in-plant considerations as well as the environmental ones. He designed and organized the radiation protection program for the Yankee Reactor at Rowe, Massachusetts, the first private nuclear power plant. He has since been actively involved with the planning of several other nuclear power plants. Dr. Maletskos has long experience in nuclear instrumentation and calibration, radiochemistry, radiation dosimetry, radiobiology, and the measurement of radioactivity in samples and in human beings. He is familiar with radwaste and transportation activities.

Dr. Maletskos is a consultant to the National Council on Radiation Protection and Measurements in regard to a variety of their subjects under current consideration. He is also a consultant to the Environmental Health Services of the Massachusetts Institute of Technology Medical Department in an aspect of human radiobiology which provides strong input into the setting of radiation protection standards for radioactivity within human beings.

Dr. Maletskos is a member of the Boston Medical Physics Group, the Health Physics Society, American Nuclear Society, American Association for the Advancement of Science, the Massachusetts Medico-Legal Society, American Academy of Forensic Sciences, American Association of Physicists in Medicine, International Conference on Medical Physics, Radiation Research Scoeity. He is certified by the American Board of Health Physics.

HOWARD O. McMAHON

Dr. McMahon, a native of Killam, Alberta, Canada, and a naturalized American citizen, received his B.A. and M.A. degrees from the University of British Columbia at Vancouver in 1935 and 1937, respectively, and his Ph.D. degree from the Massachusetts Institute of Technology in 1941.

Dr. McMahon was a research associate at M.I.T. for several years before joining Arthur D. Little, Inc., in 1943. In 1952, he was named Science Director; and was successively Vice President, Head of Advanced Research Division, 1956; Senior Vice President, 1960; and Head of Research and Development Division, 1962. In 1963 he was appointed a member of the Board of Directors and of the Executive Committee of his company and named Executive Vice President. In 1964, he became President and retained that position until 1972 when he assumed his present position as Senior Vice President on the Corporate Technical Staff.

Shortly after joining Arthur D. Little, Inc., in 1943, Dr. McMahon was instrumental in developing, with Professor Samuel Collins of the Massachusetts Institute of Technology, the Collins-ADL Helium Cryostat, which has become the standard method throughout the world for liquefying helium for low-temperature research. For this work he was awarded the Edward Longstreth Medal in 1951 from the Franklin Institute.

Dr. McMahon has also developed an efficient rectifying column packing, special heat exchangers, a moisture indicator for excessively dry atmospheres, and a cooling cycle, which produces refrigeration at extremely low temperatures by means of continuous closed-cycle operation.

He has carried out basic studies on the molecular structure of glass and on high-temperature, thermal radiation phenomena in glass. In 1952, the American Ceramic Society presented him with the Frank Forrest Award for his work in this field.

Dr. McMahon is a member of the American Association for the Advancement of Science, the American Academy of Arts and Sciences, the American Chemical Society, and the American Physical Society. He is the author of many papers and articles on low-temperature physics and has been granted twenty-two patents on a wide variety of inventions.

LITA NELSEN

Lita Nelsen received a B.S. degree in Chemical Engineering in 1964 from the Massachusetts Institute of Technology, where she was awarded the Robert D. Hunneman Prize for originality in chemical engineering, among other awards. In 1966, she was awarded an M.S. in Chemical Engineering from M.I.T. Her M.S. thesis was on "Sorption and Diffusion of Water Vapor Through Polyvinyl Alcohol Films." After leaving M.I.T., she was employed for some years by the Amicon Corporation, Lexington, Mass., where she continued her work on the mechanisms of gas and liquid transport through semi-permeable membranes, and the engineering applications of such membranes.

Mrs. Nelsen was also employed by the Corporate Technology Center of Tyco Laboratories on projects involving electrochemical sensing of carbon monoxide gas in enclosed atmospheres, desalination of sea water, etc.

Since joining Arthur D. Little, Inc., Mrs. Nelsen's work has included projects on gas and solute transport through semi-permeable polymeric and gel films.

Mrs. Nelsen's publications include two book chapters on transport through semi-permeable ultrafiltration membranes, and a number of technical articles in the area. She has also filed a number of patent applications in the area.

GERALD L. SCHROEDER (Consultant)

Ph.D., Nuclear geochemistry - oceanography

Education:	B.S.	MIT	1959	Chemical engineering
	M.S.	MIT	1961	Geology and geophysics
	Ph.D.	MIT	1965	(January) Interdepartmental: Oceanography-nuclear geochemistry

Professional experience:

Two month research cruise with Woods Hole Oceanographic Institute on R V Chain 1961

Research staff, Department of Physics, MIT, 1962-63, and 1965 through present. Supervise and conduct research in the natural radiation environment, uranium mine prospecting and ventilation, the characteristics of radiations associated with uranium and radium deposits as observed at the earth's surface and in the immediate vicinity of an ore body. Development of portable instrumentation (including a portable three channel analyser) for monitoring such complex decay schemes.

Neutron activation and x-ray fluorescence as related to non-destructive chemical analysis for microanalysis, in situ ore analysis, continuous monitoring of pollutants in or composition of gaseous or liquid streams.

1965 through present. Consultant active in mining industry; ore prospecting and in situ analysis; surface radiations; uranium mine ventilation.

WILLIAM I. WATSON

Mr. Watson is currently a member of the Chemical and Metallurgical Engineering Section of Arthur D. Little. He has had broad experience consisting of some 30 years in both underground and open pit mining operations as well as process and extractive metallurgy. His experience and training qualify him to work on any technical or economic aspects of the location, development, mining and processing of mineral raw materials.

Included in his experience was work on the initial design, engineering and surveying for a large open pit copper mine in Arizona and four years' work in Northern Chile at a large porphyry copper deposit where a block caving mining system was used. In addition, Mr. Watson has had four years' experience in underground mines in California and Montana, where many different systems, techniques and equipment were used.

In addition to mining problems and mineral deposit evaluations, Mr. Watson has been engaged in process development for uranium, precious metal, and base metal ores. In the nonmetallic field, work has been done on coal, phosphate sands, diatomaceous earths, sulfur, glass sands, salt deposits, clays, limestone, granites, slate, and sand and gravel deposits.

Other recent work has included the following:

The development of mining and processing costs and overall economics of a Chilean iron ore deposit. This involved an open pit mining plant and a dry crushing and magnetic separation processing plant.

Complete evaluation of a Canadian iron ore deposit in the Labrador Trough. This included the determination of mining methods, costs and equipment specification, as well as the development of a crushing, fine grinding, spiral and electrostatic concentrating and pelletizing flowsheet and its associated costs and equipment specifications. The open pit systems studied involved conventional shovel-truck operations as well as a potential ore pass - rail haulage method. Other iron ore evaluations have been done in Bolivia and Liberia.

In the phosphate area, he has evaluated deposits in Florida, Tennessee, Western States, and South Carolina, as well as several foreign deposits. Based on our evaluation one of the deposits in northern Florida is now under development.

Work in the coal industry has included a complete study of the coal resources of Oklahoma and Arkansas and their potential development, a study of the present and future construction of shafts, slopes and drifts in the coal regions, the development of mining and processing techniques for the production of charcoal briquettes from lignite, the evaluation of the permissibility of underground equipment, a study of underground coal gasification, and the feasibility of utilizing methane from coal mines.

ANN J. WHITTIER

Mrs. Whittier, who joined Arthur D. Little's Literature Research Group in 1960, received a B.A. degree from Mount Holyoke College in 1957 and an M.Ed. from the Harvard Graduate School of Education. At Arthur D. Little, Inc., she has been concerned with the collection and collation of technical information on a wide variety of subjects. Her experience includes literature searches, the preparation of abstracts and bibliographies and the setting up of information retrieval systems. She translates French fluently and Russian slightly. She has taken IBM courses in FORTRAN and a course in technical writing.

In recent years, Mrs. Whittier has participated in a number of environmental management studies, including a study of methods for sampling and analysis of particulate and gaseous fluorides from stationary sources, a review of the literature to assess the health effects of air pollutants from industrial fuels, and a follow-up study, which emphasized various aspects of combustion emissions and their atmospheric interactions, and the implications of research and legislation in air pollution control. In several previous studies, she reviewed the literature on international waste water treatment, on special equipment for solid wastes and refuse disposal, on less common gaseous pollutants, on ventilation and air conditioning in Soviet industrial plants, on chemical methods of sampling and analysis of gaseous pollutants from fuel combustion, and on methods of removal of SO_2 from stack gases. For another study she gathered and summarized standards and regulations on Federal, State and local levels for air quality industrial and automotive emissions. She is currently participating in a study concerning the disposal of hazardous wastes as well as preparing the ADL staff publication, "Environmental Pollution TECH CHECK", distributed weekly.

For other projects, Mrs. Whittier reviewed the literature relative to the safe handling of beryllium, the handling and storage of ammonium nitrite, and the transport of potentially hazardous materials. In addition, she reviewed the statutory regulations and codes of practice for the storage of liquids in various ports, gathered data on materials handling systems in hospitals, on mercury poisoning, on microwave radiation hazards, on hurricane damage, and on the reaction of fluorine with metal containers and container coatings. Previously she participated in several long-term projects on the properties of glass, plastics, and adhesives. In the past, Mrs. Whittier particpated in projects concerned with Soviet research on high-temperature materials, in the preparation of a directory of East European research institutes, and in a long-term review of the literature related to factors affecting the storage of liquid propellants in the space environment. She has also provided the literature support for numerous projects concerned with data communication systems and with electronic components.