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CO_2 Capture from the Flue Gas of Conventional Fossil-Fuel-Fired Power Plants

A. M. Wolsky, E. J. Daniels, and B. J. Jody

Energy Systems Division, Argonne National Laboratory,
9700 South Cass Avenue, Argonne, Illinois 60439

Research has been conducted at Argonne National Laboratory to identify and evaluate the advantages and deficiencies of several technologies, both commercially available and alternative technologies, for capturing CO_2 from the flue gas of utility boilers that use air as an oxidant (the current universal practice). The technologies include chemical solvent, cryogenic, membrane, physical absorption, and physical adsorption methods. In general, technologies for capturing CO_2 are expensive and energy-intensive. Therefore, they result in a substantial overall increase in the cost of power generation. Research to improve the performance and economics of these technologies is discussed.

INTRODUCTION

Methods for avoiding emission of CO_2 to the atmosphere are receiving increasing attention because CO_2 emissions are expected to contribute to future climate change via the greenhouse effect. This paper reviews several technologies that can (or promise to) capture CO_2 from the flue gases of today's fossil-fuel-fired utility boilers. These technologies have reached their present state of development for reasons other than avoiding climate change. Indeed, they were developed for several reasons: acid-gas cleanup, CO_2 capture for enhanced oil recovery, and CO_2 capture for use in the beverage industry. Now it is appropriate to ask if these technologies might be put to a related but different use—avoiding CO_2 emissions.

To further that inquiry, estimates are given of the amount of CO_2 emitted by power plants, and various methods of CO_2 capture are described. These methods are chemical solvent, physical absorption, cryogenic, membrane, and physical adsorption.

Several topics pertinent to avoiding emissions of CO_2 and other greenhouse gases, including CH_4 and N_2O, have been addressed elsewhere [*1, 2, 3*] and are not discussed here. Strategies for controlling CO_2 emissions, other than CO_2 recovery from conventional flue gas, are also discussed elsewhere [*4*]. These strategies include: (a) combustion of fuels in oxygen, yielding a mixture that is primarily H_2O and CO_2 from which water can be easily separated [*5*], and (b) partial (or total) elimination of carbon from fuels before combustion [*6*].

CO_2 EMISSIONS

Amount of CO_2 Emitted from Power Plants

Data on electric power generation by fossil-fuel-fired power plants in the United States are compiled by the Energy Information Administration (EIA) [*7*]. We used these data to estimate the thermal efficiencies at which the power was generated from various fossil fuels and to estimate the quantities of CO_2 generated by fossil-fueled power plants, using the following assumptions and approximations:

1. Chemical composition of the hydrocarbon content of coal is assumed to be $CH_{0.8}$.
2. Chemical composition of petroleum is assumed to be $CH_{1.59}$, and its density is assumed to be 849 kg/m^3 (53 lb/ft^3).
3. Heating values of the different fossil fuels used for power generation were taken to be the averages reported by EIA for July 1991 [*7*]:
   Coal —23,870 kJ/kg (20,525,135 Btu/ton). This implies that about 60 percent of the coal mass is represented by the formula $CH_{0.8}$.
   Petroleum —6.6×10^6 kJ/bbl (6,313,470 Btu/bbl).
   Natural Gas—38,174 kJ/m^3 (1,025,250 Btu/1000 ft^3).
4. Power production from September through December 1991 was estimated relative to production in 1989 by using the same ratio for the first eight months of the two years.

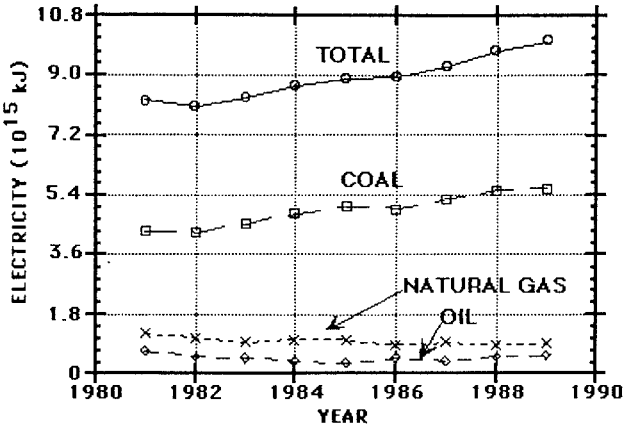

FIGURE 1. Power production in the United States during the 1980s using different fuels.

The results were found later to be in good agreement with the data published by EIA [*8*].

Figure 1 shows the total electric power (fossil, hydro, and nuclear) produced in the United States over the last decade and the amounts produced by coal, petroleum, and natural gas. The data indicate that over 50 percent of the electric power was generated using coal. While generation using natural gas and petroleum fuels did not increase in the 1980s, generation using coal increased by over 20 percent during the same period. This is significant because, of the three fossil fuels considered, coal generates the most CO_2 per unit of fuel energy. Figure 2 shows approximate material and energy balances for a typical coal processed by a conventional coal-fired power plant. This figure shows that a conventional coal-fired power plant produces over 0.25 gram of CO_2 for each kilojoule of electrical energy (or 0.9 tonne/MWh). The generation of CO_2 also liberates about 80 percent of the coal energy (the other 20 percent comes from burning the H_2 in the coal).

Figure 3 shows the power generation efficiencies attained in the 1980s. Coal-fired plants are more efficient than either petroleum- or natural-gas-fired facilities, primarily because many of the oil- and natural-gas-fired plants are older, and consequently less efficient, units that are used only for meeting peak demands. Properly designed natural-gas-fired base-load units should be able to achieve higher efficiencies. Furthermore, as natural gas is mostly CH_4, over 50 percent of the energy of combustion of natural gas comes from its H_2 content, while H_2 provides only about 20 percent of the energy of combustion for coal. Therefore, a significant reduction in CO_2 emissions can be realized if coal is replaced with natural gas. However, CH_4 is itself a greenhouse gas that, mole for mole, is suspected to contribute between 10 and 30 times more to the greenhouse effect than does CO_2 [*1*, *2*]. Thus, comparison of coal with natural gas must take CH_4 leaks into account.

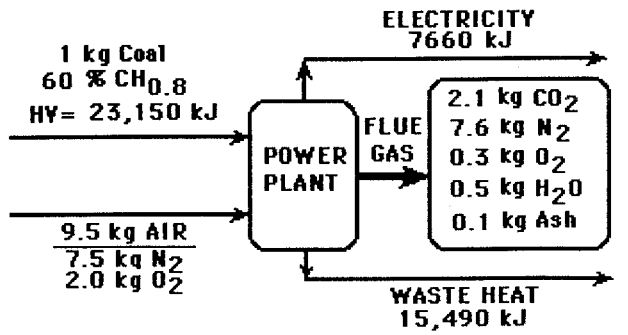

FIGURE 2. Products of coal combustion with 110 percent theoretical air.

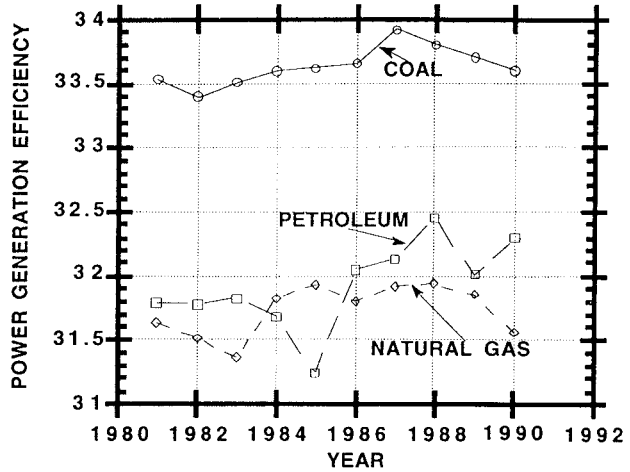

FIGURE 3. Power generation efficiency using different fuels.

On the basis of the information shown in Figures 1, 2, and 3, and on the assumptions stated above, the total amount of CO_2 produced by fossil-fueled power plants was estimated with the results shown in Figure 4. In 1990, the total amount of CO_2 produced was about 1.68 billion metric tons, an increase of 20 percent over the 1982 levels. Coal-fired facilities account for more than 90 percent of the CO_2 produced by fossil-fueled power plants. While the coal-fired power plants emit a tremendous amount of CO_2, similar amounts of CO_2 are also emitted by transportation vehicles and by industrial and residential heating equipment. The difference is precisely that there are few power plants and many automobiles, thus making power plants an obvious place to hope for economies of scale in CO_2 capture.

Several technologies for the capture of CO_2 from flue gas exist. These include chemical solvent methods, physical absorption methods, cryogenic methods, membranes, and adsorption methods. All of these CO_2-capture processes are expensive and energy-intensive [*4*, *9*]. We evaluated these methods and found that the chemical-solvent method is commercially available and could be the most economically favorable.

Considerations of the Flue-Gas Stream

The recovery of CO_2 from the flue-gas stream is made more difficult by the nature and composition of that stream. Several factors must be considered, as discussed below.

CO_2 is present at low concentrations and thus low partial pressures. For example, when coal is burned with 110 percent

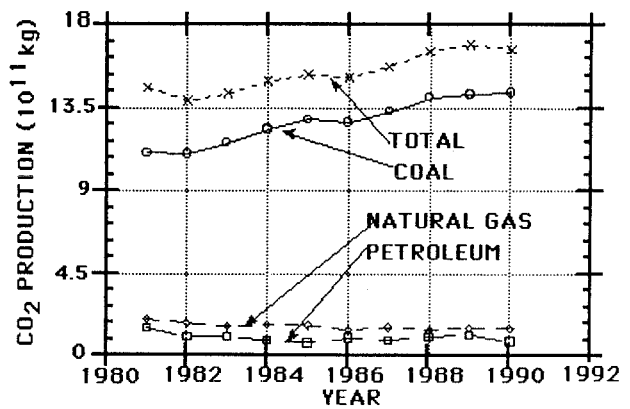

FIGURE 4. CO_2 production by U.S. Power Generation Plants.

theoretical air, CO_2 represents about 15 mol percent of the flue gas. The CO_2 percentage is lower if one considers the moisture that is already present in the coal, but the CO_2 percentage is even lower for oil (~13 percent) and natural gas (~9 percent). Most capture methods require compression of the flue gas to increase the CO_2 partial pressure for more efficient separation.

The dominant compound in the flue gas is N_2. Both N_2 and CO_2 require very low temperatures and/or high pressures to be condensed. These conditions complicate the process of separating N_2 and CO_2 from each other using cryogenic methods, both because of the potential for forming solids and because the solubility of N_2 in the condensing CO_2 will become appreciable; distillation of the N_2 from the liquid CO_2 would be required to attain high purities, if they are needed.

The large number of species present (N_2, CO_2, H_2O, H_2S, COS, CO, O_2, particulates), and the corrosive nature of some of these species, complicate all applicable separation processes. For example, even though sulfur-bearing compounds and O_2 are present in only small quantities, they undergo irreversible chemical reactions when amine scrubbers are used. These reactions result in the degradation of the solvent. When membranes are considered for the separation, difficulty in developing a single-component-selective membrane increases as the number of species present increases.

The presence of appreciable water vapor in the flue gas increases the cooling load on the separation process, and the water vapor could, under the right conditions, interact with the CO_2 to form clathrates. These solid-type (slush) structures could cause transport problems within the recovery system. Condensing water could also contain appreciable quantities of dissolved acid gases, including CO_2, which makes the water acidic and corrosive. Most of the water can be separated at much higher temperatures than are needed to separate CO_2 and N_2. Figure 5 shows the pressures to which the flue-gas stream must be compressed to permit condensing 90 percent of the water using cooling fluid at different temperatures. Figure 5 also shows the approximate cooling-fluid inlet temperature required to achieve 90 percent condensation of the H_2O and the flue-gas exit temperature required at the different pressures. For example, cooling towers supply cooling water at about 300 K (80°F) to the condenser. To achieve 90 percent drying of the flue gas stream using the 300 K (80°F) water, the flue gas has to be compressed to about 1,010 kPa (10 atm). (Incidentally, the amount of water generated when burning coal is about 4 percent of the amount of steam raised by the coal for power generation, excluding the moisture present originally in the coal). Furthermore, the necessary drying levels to prevent corrosion are achieved using desiccants, after the bulk

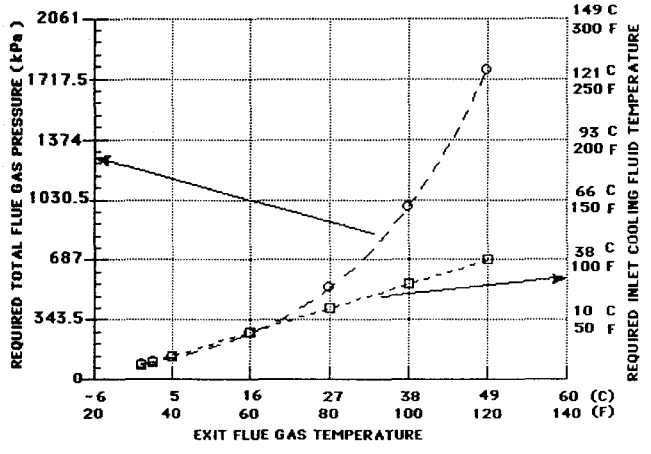

FIGURE 5. Pressure to which flue gas must be compressed to achieve 90 percent drying using different cooling-fluid-inlet temperatures.

of the water is condensed by conventional means. Solid desiccants and liquid absorption materials, such as ethylene glycol, diethylene glycol, triethylene glycol, and glycerol, can be used [*10, 11*]. These materials are normally used at high pressures and are regenerated at temperatures in excess of 450 K (350°F).

Both CO_2 and N_2 are relatively inert gases, so only at excessively high temperatures will they react to form environmentally stable solid or liquid products that are easy to separate and dispose of.

The captured CO_2 must be under pressure in order to transport it to its final sequestering destination. Today, there is an extensive network of CO_2 pipelines in west Texas and adjacent areas. Present practice maintains the pipeline pressure high. Since the CO_2 would be sequestered either underground or under water, the energy for compression cannot be wholly captured at the sequestering destination. (For example, when the pipeline pressure is 11,000 kPa (110 atm), the CO_2 can be submerged to a 1,100-m depth below the ocean surface. Note that, at 11,000 kPa, CO_2 is essentially a liquid, its critical point being 74 atm and 88°F.) The compression energy applied to the N_2 portion of the flue-gas mixture, prior to the CO_2 separation, may be recoverable in certain designs. The N_2-rich stream may be heated and expanded in a gas turbine to produce mechanical work.

The quantity of CO_2 emitted by power plants is more than 30 times greater than the quantity used in commerce. Therefore, the cost of recovering CO_2 cannot be offset by the market value of the recovered CO_2.

TECHNOLOGIES FOR CO_2 CAPTURE

Chemical Solvent Method

Chemical stripping of CO_2 involves one or more reversible chemical reactions between CO_2 and another material (such as: mono-, di-, or tri-ethanol amine; di-isopropanol amine; sodium hydroxide; sodium carbonate; or potassium carbonate) to produce liquid or solid species that, upon heating, break down to liberate CO_2 and regenerate the material reacted with CO_2. For example, the monoethanol amine (MEA) processes can be represented by the following reversible reaction:

$$C_2H_4HNH_2 + CO_2 + H_2O \rightleftarrows C_2H_4HNH_3^+ + HCO_3^-$$

Typical processes include ADIP, ALKAZID (M & DIK), BENFIELD, CATACARB, ECONAMINE, MDEA, SEABORD, VACUUM CARBONATE, SNPA-DEA, SULFIBAN, SULFINOL, GIAMARCO-VETROCOKE, and STRETFORD. Many of these processes are in full-scale operation for the removal of acid gases, including CO_2, from natural gas reservoirs, ammonia production plants, and other applications. Dow Chemical Co. developed the Gas/Spec FT-1 amine-based process in the late 1970s, which was later known as the ECONAMINE process [*12*]. In 1978, an 800-metric tons per day amine-based plant was started up to recover CO_2 from flue gases. A 200 metric tons per day unit was started up in January 1991, and another 300-metric tons per day plant was started up in March of the same year [*13*]. Two other amine-based pilot plants were constructed and tested in Canada [*14*]. Experience with these and other plants identified three major problems: (1) solvent degradation due to irreversible reactions with O_2, SO_x, and COS; (2) corrosion in the absorber and the regenerator; and (3) operating difficulties caused by variable operating conditions. Corrosion inhibitors and additives brought these problems under acceptable control.

Most of the energy consumed by these processes is used to raise steam at pressures about 445 kPa (50 psig). The steam is used in the solvent regeneration process. The energy-intensiveness of these processes is partly related to the low absorption capacity (CO_2 absorbed per unit mass) of the solvents,

because all of the aqueous solutions have to be heated to higher temperatures to liberate the captured CO_2. These systems require electric energy for pumping the solvent and for compressing the gas and cooling water for cooling the weak solution after the regeneration process.

Energy savings can be realized if a solvent that has high absorption capacity at moderate temperatures and atmospheric pressure can be developed. The higher the absorption capacity, the less the quantity of solvent required to handle a given gas stream, and, consequently, less energy will be required for regeneration, pumping, and cooling. Smaller and less costly equipment will be required. A solvent that can be regenerated at lower temperatures stands a better chance of being able to use waste heat instead of extracted steam from the turbine, which results in lower turbine power output.

Regeneration at elevated pressures could also be advantageous because the resulting CO_2 will be at high pressure. This requires a solvent that can be regenerated at high temperatures without thermal degradation.

Another alternative for saving energy would be to develop a thermally stable solvent that can be converted to superheated vapor, after regeneration at high pressure, and used as the working fluid in an organic Rankine bottoming cycle, instead of wasting the heat used in the regeneration process. This concept will be particularly attractive when used to recover CO_2 from the exhaust of a gas turbine, because the solvent bottoming cycle could be driven by heat available in the turbine exhaust. A simplified schematic diagram of such a concept is shown in Figure 6.

Published data [9] indicated that retrofitting a coal-fired power plant with an ECONAMINE FG^{SM} system for 90 percent CO_2 removal and a Thiosorbic® magnesium-enhanced wet-lime (mag-lime) SO_x removal process resulted in a 35 percent reduction in net power production and that the cost of the retrofitting process was greater than the initial capital cost of the power plant itself. The cost included that of CO_2 transportation to and disposal in large depleted natural-gas fields or in the ocean. The results are given in Table 1. However, to facilitate comparison with other technologies, the cost of avoiding CO_2 emissions, while providing the same electrical service, is relevant. Because MEA processes need steam, whose production generates CO_2, more CO_2 must be captured than is avoided. As a result, the cost of CO_2 avoided by MEA is approximately twice the cost of CO_2 captured. On the basis of available data [9], we estimated that the net economic impact of capturing and disposing of CO_2 was an increase in the cost of electricity production from 46 to 140 mills/kWh, an important part of which is due to compression and drying.

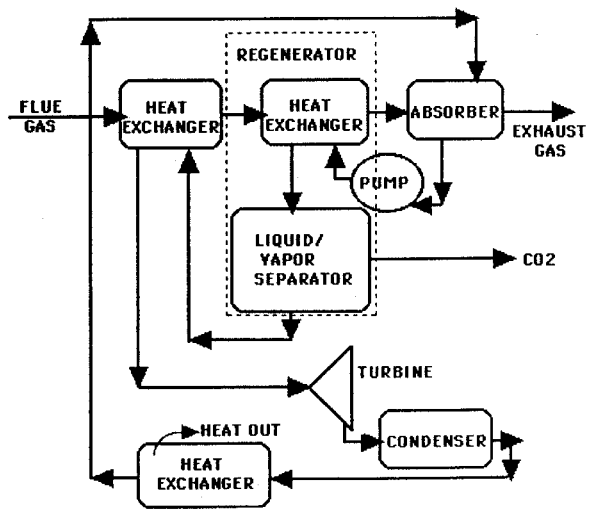

FIGURE 6. Integrated solvent regenerator/bottoming cycle.

Table 1 An ECONAMINE System Integrated with a Conventional Pulverized Coal-Fired Power Plant

Process Subsystem	Cost[a] ($1,000 kg of CO_2 Captured)		
	Capital	Energy	Total
ECONAMINE	10	12	22
CO_2 Compression and Drying	4	5	9
Cycle Modifications	6	—	6
CO_2 Pipeline	15	—	15
CO_2 Disposal	5	—	5
Total	40	17	57

[a] Discount rate is assumed to be 17.4 percent, and electricity cost is $0.05/kWh.
Source: Reference 9; data used with permission of the authors.

Physical Absorption Methods

Physical absorption is a bulk phenomenon. Inorganic and organic liquids may be used to preferentially absorb some gaseous species, separating these species from the gas mixture. The absorption process depends on the operating temperature and pressure, as well as on the nature of the gases and the absorption liquid. The absorption liquid is regenerated by increasing its temperature (temperature swing) and/or reducing its pressure (pressure swing). High-boiling solvents that have high partial pressures are preferred in order to keep solvent losses to a minimum and to prevent contamination of the released gas with solvent vapors.

Organic solvents that are commonly used include propylene carbonate (Fluor process), n-methyl-2-pyrrolidone (NMP-PURISOL), methanol (RECTISOL), dimethyl ether of polyethylene glycol (DMPEG-SELEXOL), and sulfolane (SULFINOL). Many of these processes are practiced on commercial scale. The inorganic solvent suggested for CO_2 scrubbing is sea water.

The absorption capacity of organic and inorganic solvents for CO_2 increases with increasing pressures and with decreasing temperatures. Because the partial pressure of CO_2 in the flue gas is low and the flue-gas temperature is high, the physical absorption method does not appear likely to compete with the chemical solvent method for processing the flue-gas stream. Dissolution of other flue-gas constituents, mainly H_2O, is expected to adversely impact the absorption of CO_2. Particulates in the flue gas could also cause rapid degradation of the absorption material unless the material is frequently filtered to remove the trapped particulates. These particulates could also create pumping problems.

This process could, however, be a very efficient approach for processing such high-pressure CO_2-rich streams as those encountered in advanced power generation systems, as for example, the O_2-blown coal gasification/combined cycle [4, 9]. Therefore, as we stated earlier, even though the physical-absorption process does not appear promising for capturing CO_2 from the flue gas of conventional power plants, it could be a very efficient and economical approach for CO_2 capture from emerging power-plant streams.

Cryogenic Methods

Cryogenic separation of CO_2 from combustion-gas streams involves multistage compression, followed by cooling of the compressed gas after each stage, and eventually, expansion of the cool, high-pressure stream to cause a large drop in temperature. As a result of this process, CO_2 will be liquefied, or even solidified, and separated from the gas phase. A distillation (fractionation) column may be required to separate dissolved impurities from the product CO_2. After CO_2 separation is

accomplished, the effluent stream will consist essentially of N_2 and the excess O_2 used in the combustion process. Therefore, the key question is how does this method compete, in terms of energy consumption and cost, with conventional (cryogenic) upstream air-separation systems, as both lead to essentially the same results.

The gas compression/cooling process may be supplemented with a conventional refrigeration cycle to provide additional cooling to permit the condensation of CO_2 with less gas compression at higher temperatures. The liquefied CO_2 can be pumped to the desired sequestering pressure, if necessary. This combination will also eliminate the risk of solid CO_2 formation in systems that are not designed to handle solids. The residual N_2 gas, which will be at high pressure, can be heated to high temperature and expanded in a multistage gas turbine with reheat to provide at least part of the power consumed in the compression process. This alternative is expensive in terms of capital cost, but it will minimize any reduction in output of the power plant as a result of CO_2 recovery.

The optimum total stream-pressure/refrigeration temperature may be determined analytically. Determination of the optimum pressure should take into consideration the solubility of N_2 and the acid gases (SO_x, COS, NO_x) in the liquefied CO_2. Even though the acid gases will have a minimal impact on the mass and volume of the CO_2 product, they could reduce the usefulness of the CO_2 as a commercial product or impact the final disposal. In the presence of water, the acid gases exacerbate the corrosion problem in the transportation pipelines. The presence of SO_x, COS, and NO_x may also limit the options available for sequestering the CO_2 after it is captured. Design of cryogenic systems for the separation of CO_2 should also address the potential formation of such solid structures as CO_2/H_2O clathrates, ice crystals, and solid CO_2 particles or flakes, which could cause plugging problems. The presence of particulates is another concern when considering cryogenic separation of flue gases, especially those gases generated by coal combustion.

Cryogenic separation of CO_2 from the combustion products of utility boilers is an energy-intensive process. It is estimated that the energy penalty will be in excess of 50 percent of the coal energy. Therefore, this process appears uneconomical [*4*]. However, it has received little attention and deserves more before it is dismissed.

Membrane Systems

Membranes are generally solid porous, semiporous, or nonporous structures through which some of the species in a mixture permeate, leaving other species behind. In another type of membrane, called a liquid membrane or a gas-absorption membrane, the solid porous structure is impregnated with a liquid that is capable of dissolving the species whose permeation is desired, to assist in permeation of that species through the membrane [*15, 16*]. Microfiltration membranes are in the widest application, while gas separation membranes are used the least [*17*].

Two criteria of a good membrane separator are selectivity and high permeability or mass flux (the rate at which species permeate or cross from one side of a unit area of the membrane to the other). Unfortunately, the two criteria are inversely related for the common polymeric membranes. The permeability of the membrane depends on many factors, including the chemical potential gradient of the species being separated [*17, 18, 19, 20*]. This gradient develops as a result of gradients in one or more of the following parameters: partial pressure, concentration, electrical potential, or a combination of these. The mass flux depends also on the nature and design of the membrane itself.

Flue-gas streams generated by coal combustion are composed of many species present in different concentrations, each having its own solubility in and diffusivity through a membrane material. For example, CO_2, H_2S, and H_2O have reasonably high solubilities in membrane materials at moderately low partial pressures, but O_2 and N_2 have high diffusivities in the same materials. Therefore, because the overall permeability depends on both of these parameters, the development of a CO_2-selective membrane system for such a stream is not an easy task. Multistage separation will definitely be required to achieve adequate separation. The flue gas can be cooled, dried, and compressed prior to coming in contact with the membrane system. This reduces the problem of separating CO_2 from N_2. Several polymeric materials can be used to achieve substantial separation of CO_2 from N_2. Sluijs [*20*] reported that cellulose acetate polymers can have a CO_2/N_2 selectivity (ratio of the permeability of CO_2 to the permeability of N_2) of about 66.5. This value means that substantial separation of these two gases is technically feasible, although a selectivity of over 200 is needed for adequate separation. Concentration of the CO_2 in the flue-gas stream by such membranes is probably a more realistic goal. The presence of fine (approximately micron size) particulates under acidic conditions could result in performance degradation and increased maintenance requirements.

Membranes can be energy-efficient when excessive compression is not required to attain a practical pressure gradient of the targeted species across the membrane. The relatively low CO_2 pressure in the flue gas dictates the need for compression to increase the partial pressure of the gas constituents and to compensate for the pressure loss across the membrane. Another concern with membranes is the ability to maintain a sufficiently low partial pressure of CO_2 on the downstream side of the membrane. With the exception of water vapor as a potential sweeping gas for the separated CO_2 (which could be easily separated by condensation), evacuation of the downstream chamber or absorption of the CO_2 into a chemical or physical absorption material appear to be the only obvious alternatives. These alternatives are energy-intensive. However, the evacuation alternative could be integrated with the product compression step, provided that the product is low in non-CO_2 constituents.

PHYSICAL ADSORPTION SYSTEMS

Physical adsorption of gases on solid adsorbents (such as activated carbon) is a surface phenomenon in which a few layers of the adsorbed gas are held by weak surface forces. The capacity of an adsorbent for a given gas depends on the operating temperature and pressure. The higher the partial pressure and/or the lower the temperature, the greater the adsorption capacity of the system. The key issue for commercial application of these systems is the surface area required per unit mass or volume of adsorbed gas. However, these systems are simple, and their operation and regeneration (temperature swing or pressure swing) can be energy-efficient.

Because the adsorption process is highly sensitive to temperature and pressure, cooling and compression of the flue-gas stream is necessary. Competition between CO_2 and other flue-gas constituents, especially H_2O, for the active adsorption sites is expected to reduce the adsorption of CO_2, because water vapor will be adsorbed preferentially. In addition, the capacity of such systems for CO_2 adsorption is small. Particulates in the flue gas could also cause gradual deactivation of adsorption sites because of the difficulty expected in removing particulates by heating or depressurization. Therefore, we do not expect this system to compete very well with absorption systems for the separation of CO_2 from the flue gas of conventional coal-fired power plants.

CONCLUSIONS

We believe that the chemical absorption system is the most

promising technology for separation of CO_2 from the flue gas of a conventional coal-fired power plant. Cryogenic methods appear to be a distant second, but deserve more attention. Although physical absorption systems appear very promising for Integrated Coal Gasification-Combined Cycle and fuel-cell plants because of the higher pressures of CO_2 encountered in plants operating with these technologies, physical absorption systems lag behind chemical absorption methods when considered for application in conventional power plants.

The chemical absorption (amine) process is still expensive and energy-intensive, and substantial improvements in performance and cost are necessary to minimize the increase in the price of electricity that would result from the use of this technology.

RECOMMENDATIONS

The development of an improved chemical solvent is essential to improving the performance and reducing the cost of CO_2 capture. Such a solvent should be environmentally safe and should have:

- High absorption capacity at moderate temperatures and pressures. An absorption capacity of about 1 kilogram of CO_2 per kilogram of solvent is a reasonable target.
- High vapor pressure and thermal stability at elevated temperatures. The pregnant solution can then be pumped to high pressure, followed by heating to high temperature, to liberate the CO_2 at high pressure and thus avoiding recompression of the CO_2, all without losing appreciable amounts of solvent. A high-vapor-pressure solvent that can also be used as the working fluid in an organic Rankine bottoming cycle will have the added benefit of allowing the regeneration energy that was applied to the solvent to be used for power production instead of wasting it.

ACKNOWLEDGMENT

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