Comparison of Processes for Capture, Separation, and Sequestration of CO_2 from the Combustion of Coal

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Introduction

CO_2 is a major “greenhouse” gas and combustion of carbonaceous fuels is one of the primary sources of its addition to the atmosphere. The sequestration of CO_2 from combustion or other sources is not currently prescribed by law and has no direct economic advantages but a recent study by the National Academy of Engineering has listed it among the principal technological challenges of the 21st century. Since the time that this work was completed, hardly a day has gone by without the appearance of a technical report or a news item concerning the sequestration of CO_2. However, none of this new information and opinion negates or renders our study obsolete, because it had the narrow objective of examining the technical feasibility and relative economic costs of several processes for the recovery, separation, and sequestration of CO_2 from the combustion of a particular coal in a particular location to generate electricity at a representative rate. The optimality of the choices made in our work can be questioned but the results are nevertheless presumed to remain valid as a point of reference.

Location

Our analysis is based on the combustion of coal, which, despite widespread opposition, is generally expected to be a major source of energy for the generation of electricity in the next several decades. The Illinois Basin, which underlies Illinois, western Indiana, and western Kentucky was arbitrarily chosen for this analysis because (1) it has both large and accessible holdings of bituminous coal, (2) includes a number of geological formations suitable for sequestration, and (3) produces 255 million metric tons of CO_2 per year (11.7% of total US emissions), 92% of which are from coal-fired power plants. (The other major sources of emission in this region include petroleum refineries, cement plants, and miscellaneous heavy industries.)

Venues for Capture and Separation

The possible venues for sequestration include algae, the ocean depths, depleted oil and gas reservoirs, coal seams that are not suitable for mining, and deep saline formations. Algae are one of the most productive users of CO_2 in the plant world, and after saturation the can be burned, which produces thermal energy with no net production of CO_2. However, before it can be burned the mass of algae must be dried, which is very energy-consumptive. The rate, potential capacity, and economic feasibility of this form of sequestration are difficult to estimate at the present time because of a lack of the necessary technical information or industrial experience. Accordingly, this alternative was not examined in detail.

Venues for Sequestration

The choice of the Illinois Basin as a site eliminated the possibility of sequestration in the ocean. The oil and gas reservoirs, the coal seams, and the saline formations of the region have an estimated capacity of 282 million metric tons, 282 million metric tons, and 4.14 billion metric tons of $\mathrm{CO_2}$, respectively. Insertion of CO_2 in the oil and gas reservoirs may enhance their recovery and in coal seams may replace and release methane that can be recovered, but the fractional offsets of the cost of sequestration appear to be negligibly small. In saline formations, the CO_2 dissolves in part and may react in part to form carbonates, but the latter do not appear to have any economic potential. If all the electricity produced in the Illinois Basin were produced by the combustion of coal, and if 90% of the corresponding production of CO_2 were sequestered in the oil and gas reserves there they would become saturated in only 1.12 years. The coal seams have an almost equal capacity, while the saline formations have a capacity of 16.2 years. Because of its greater capacity, attention was focused on sequestration in the saline formations.

This total of only 18.4 years is discouraging at first sight, but it is an unrealistic extreme case. Over this period of time sequestration, as a new technology, would be expected to be applied to only a very small fraction of the coal burned to produce electricity in the basin. This time period provides a window of opportunity in time to seek solutions other than sequestration and other than the particular processes for sequestration examined herein.

Representative Conditions

For the quantitative calculations of the combustion of coal and of the separation and sequestration of CO_2, “Old Ben 26” coal and a conventional furnace and turbine capable of producing 500MW of electricity at an overall efficiency of 36% and operated at 65% capacity on an annual basis, were chosen as representative. The corresponding annual requirement of coal and the annual production of CO_2 were calculated to be 1.0 and 2.47 million metric tons, respectively. These quantities amount to approximately 1% of those for the whole basin. The flue gas produced by the combustion of this low-sulfur bituminous coal contains a significant amount of ash in the form of particulate matter, CO, and N_2, and traces of NO_x and SO_2. The ash is postulated to be removed by electrostatic precipitation, and the NO_x and SO_2 catalytically. However, these costs were not assigned to that of sequestration because they are required in its absence. The concentration of CO was based on the use of 25% excess air, which results in a mass concentration of 58% N_2 in the flue gas. Gaseous N_2 is not ordinarily considered a pollutant but it is in terms of the sequestration of CO_2. It can be almost totally avoided in the stack gas by burning the coal with oxygen rather than air, but at the price of a whole set of new problems, including the high cost of oxygen and the necessity of a massive recirculation of flue gas, or of some other expediency, to reduce the flame temperature. This alternative was not examined. The separation of CO_2 from the flue gas was thereby considered to be a necessary part of the process of sequestration.

Processes for Separation

Three processes for separation of the CO_2 from the resulting "clean" flue gas were examined, namely absorption in an aqueous solution of monoethylamine (*MEA*), adsorption by and reaction with calcium oxide (*CaO*) , and adsorption by and reaction with magnesium oxide (*MgO*). Because the use of aqueous *MEA* as an absorbent for various chemicals is a well-developed technology, it was adopted as the base case for fiscal comparisons. The use of *CaO* as an adsorbent and reactant is also well developed but it has a serious shortcoming when used for CO_2, namely the temperature of 1175K required for the decomposition of the calcium carbonate (Ca_2CO_3) that is formed. Magnesium oxide (*MgO*) was considered as a possible alternative to *CaO* because the decomposition of magnesium carbonate (Mg_2CO_3) occurs at the much lower temperature of 700K, but the physical handling of powdered *MgO* was immediately recognized to be a possible serious impediment. The discovery of a Department of Energy report by Breault and Reasbeck that described the impregnation of *MgO* on solid granules of alumina (Al_2O_3) resulted in our reconsideration of *MgO*, and the subsequent detailed calculations revealed the resulting process to be more economical than those utilizing *MEA* or *CaO*. These calculations were based on the recovery of 90% of the CO_2 in 245 kg/s of cleansed stack gas at 311K and slightly above 1bar.

***MEA*.** The *MEA* system was conventional except for its size, consisting of 12 identical 18-tray columns, 4.27m in diameter and 15.85m high. The estimated utility costs, which included 1.7kg of steam per kg of CO_2 totalled \$21 per metric ton of CO_2, which corresponds to the value of 21% of the output of the generator. Liquid absorption might be more favorable if a solvent could be found that degrades more slowly, is less corrosive, has a higher capacity for CO_2, and requires less energy for regeneration than *MEA*.

***MgO*.** As contrasted with the conventionality of the *MEA* system, the *MgO* system has three unique characteristics: 1) the preparation of the impregnated tablets; 2) the utilization of six identical packed-bed adsorber/reactor/desorbers 5.0m in diameter, and 6.3m high; and 3) their operatin in parallel in batch cycles at different stages. To produce the combined MgO/Al_2O_3 granules, alumina tablets 5mm in diameter are soaked for up to 3 days in an aqueous solution of 20-80 gm/l of magnesium hydroxide. The water of hydration is then driven off by heating, resulting in a mass ratio of *MgO* to Al_2O_3 of 1.2. The total requirement for *MgO* supported on alumina in the six adsorbers was estimated to be 990Mg with an annual replacement of 10% due to attrition. Clean stack gas at 384K and 1.5bar is compressed to 14.6bar before entering a packed bed in the first step of the cycle. The adsorption and reaction are very exothermic and recirculation and cooling of a portion of the exiting gas by heat exchange with water is necessary to avoid an excessive temperature. The recirculation to feed ratio is 12/5. The stream exits the adsorber/reactor at 475K with 10% of the CO_2 , 90% having been converted to Mg_2CO_3. The desorption is very endothermic and the same of rate of recycling, in this case with heating by exchange with flue gas, is necessary to maintain a sufficient temperature.

Storage and Compression

The captured CO_2 at 600K, after being cooled to 311K, passes at a flow rate of 254 Mg/hr into two 11,300m^3 holding tanks, which are maintained at slightly above atmospheric pressure. Before its sequestration the CO_2 must be converted to a condition within the supercritical phase, which requires a pressure of at least 73.3bars at 311K. A still higher pressure, namely 95.2bar is required to compensate for the pressure drop over a representative distance of 167 km in a 10-in (0.254-m) pipeline leading to the site of sequestration.

Because the this compression accounts for over half of the total cost, several alternative schemes were considered. The final choice was a centrifugal blower and three-stage centrifugal compression with intercooling. Two-stage compression would require a slightly lower fixed capital investment (by 0.7%) but a slightly higher operating cost (by 4%) as well as more electricity and cooling water. The use of holding tanks for the compressed gas in the event of a temporary shutdown of the compressors was examined but concluded to be impractical.

Estimated Overall Costs

The estimated operating and fixed costs of separation and sequestration of CO_2 using the *MEA* and MgO/Al_2O_3 processes for the indicated conditions (a 500MW combustor/generator in the Illinois Basin) are summarized in Table 1.

Table 1. Comparative Costs

Process	Operating cost			Capital investment
	10^6\$/yr	\$ per metric ton of CO_2	¢ per KWH	10^6\$
Capture				
MEA	139	56.70	8.9	463
MgO	44	17.80	1.5	178
Sequestration	34	14.00	1.2	108

Conclusions

Based on the preceding analyses, it is concluded that separation using *MgO* is superior to that using MEA in every respect except operational experience. The fixed and variable costs are both much less and the plant equipment is more compact. Even for this new process, the overall costs for capture, separation and sequestration are huge and no easy means of accomplishing their reduction is apparent. Because it produces no income, this process of sequestration can be justified only for environmental reasons or legal requirements. Our estimate of a total cost of 3.7 ¢ per KWH provides some guidance in those respects.