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CO_2 Capture from PC Boilers with O_2-Firing

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ABSTRACT

The U.S. Department of Energy (DOE) is investigating CO_2 recovery from fossil-fuel-based power cycles as a greenhouse gas mitigation strategy. Given that the most common such cycle is the conventional pulverized coal-fired plant, a system that could be retrofit to such boilers for CO_2 recovery would have broad applicability. Argonne National Laboratory (ANL) was an early investigator of such systems, proposing them as a source of CO_2 for enhanced oil recovery. Removal of CO_2 from conventional pulverized coal plant flue gas requires treatment of a large volume of gas dilute in CO_2. By substituting nearly pure O_2 for air as oxidant, the volume of the gas is greatly reduced and the CO_2 concentration increased, permitting more economical CO_2 recovery. Such a system was originally proposed by researchers at ANL as a source of CO_2 for enhanced oil recovery (EOR), but it has recently been studied as a means of reducing CO_2 emissions from coal-fired power plants. There is particular interest in this CO_2 recycle boiler as a means of retrofitting CO_2 control to existing plants. ABB Power Plant Laboratories (Windsor, Connecticut, now Alstom Power) has performed a CO_2/O_2 retrofit design study of the Trans Alta Corporation's Sundance Unit 1 boiler. Significantly, that study found that no major modification of the boiler was required for the retrofit. These results confirm technical viability. The high cost of air separation will determine economic viability. Our interest has been in extending this analysis to U.S. plants that might be retrofit candidates. A key difference is that the Sundance plant uses extremely low sulfur coal and requires no post-combustion sulfur removal. A major retrofit at a U.S. plant will result in post-combustion sulfur cleanup requirements. We have therefore focused on the fate of sulfur in the gas path and the performance of sulfur removal equipment. We have also extended previous analyses by considering upstream and downstream processes with the ultimate goal of completing a life cycle assessment. Product transport, in particular, introduces important technical and economic challenges.

Background - Argonne National Laboratory (ANL) Investigations

Carbon dioxide has found extensive use for enhanced oil recovery (EOR). Early applications of this technology were concentrated in regions with natural sources of CO_2. Combustion flue gas is an obvious artificial source of CO_2. However, the cost of extracting CO_2 from flue gas, which is highly diluted by atmospheric nitrogen from combustion air (as well as by excess air), renders direct use of flue gas CO_2 uneconomic in most circumstances. A key difficulty is the presence of sulfur dioxide (SO_2), which interferes with CO_2 separation by its interaction with reagents and absorbents. As an alternative, researchers at ANL proposed the removal of nitrogen prior to combustion — that is, the use of high-purity oxygen rather than air in the boiler [Wolsky, 1985]. This would result in a flue gas stream of nearly pure CO_2, avoiding the need for post combustion gas separation. We refer to this technology as CO_2 recycle because a portion of the flue gas stream is recycled to the boiler to replace the nitrogen, thereby maintaining design level gas flow rates and gas path temperatures in the boiler.

Argonne's investigations demonstrated technical feasibility of the CO_2 recycle boiler. Two combustion tests, one at laboratory scale [Weller et al., 1985] and one at a small commercial stoker furnace [Kumar, 1987], demonstrated that coal can be burned in a CO_2/O_2 and in a flue gas/O_2 atmosphere. Efficient combustion (low residual carbon) was achieved in the laboratory tests conducted at CO_2/O_2 molar ratios between 2.18 and 3.65. Target molar ratios in these tests were 3.76, 2.44, and 2.23. A base case using air with 17% excess was run on the same apparatus. The CO_2/O_2 ratios were selected to achieve, respectively, the same oxygen concentration as that in the base case, the same adiabatic flame temperature as that in the base case, and the same flame radiation as that in the base case. These values provide guidance for setting the recycle rates in subsequent modeling, as discussed below. Following the success of the laboratory-scale combustion tests, a commercial, 2.2-million-Btu/h stoker boiler was retrofit with flue gas recycle, oxygen feed, and the necessary controls. This demonstration confirmed the feasibility of applying the recycle system in a utility setting, but substantial air in-leakage in the coal feed, ash removal, and other systems limited total CO_2 concentration in the flue gas to 48.5%.

At its 10-million Btu/h utility boiler pilot facility near Irvine, CA, Energy and Environmental Research Corporation (EERC) also performed tests for ANL [Abele, 1987]. The objectives of these tests were to (1) determine the ratio of recycle gas to oxygen that would achieve heat transfer performance similar to that realized with conventional firing, (2) quantify observable operational changes (such as flame stability), and (3) provide a basis for scaling experimental results to commercial scale. Tests were performed for baseline performance (coal with air) and for flue gas recycle with and without dehydration. With wet recycle, overall furnace heat transfer efficiency for the recycle case matched the performance with air by using a recycle gas-to-O_2 ratio of 3.2. With the recycle stream dehydrated, the baseline performance was matched with a recycle ratio of 2.7 [Payne et al., 1987]. The EERC project also involved computer modeling of the CO_2 recycle plant boiler. Important conclusions from the EERC modeling project are (1) performance with CO_2 recycle can be reliably predicted with heat transfer models and (2) standard boiler operating practice can be used to compensate for deviations in recycle ratios of ±0.4 away from the optimal values.

EERC measurements were used to construct mass balances around the furnace for comparison with expected values as verification of the flue gas measurements. Flue gas mass out was exceeded by water, fuel, and oxidant stream inputs by 0.3 to 10.6%, perhaps reflecting flue gas losses in the pressurized furnace. Measured energy balances were established on the basis of (1) the chemical potential energy of the fuel and sensible heat of the oxidant as inputs and (2) heat transferred in the radiant and convective pass sections of the furnace. The heat balance in the radiant section closed within 3%, and in the convective section, the heat balance closed within 7%. These results give confidence in the test program's results, principally the optimal recycle ratio needed to achieve performance comparable to that with air and coal.

The EERC project further characterized emissions and furnace operational issues. It was found that NO_x emissions declined linearly with increasing recycle ratio. A decrease of 50% was realized over the range of recycle ratios tested. Sulfur emissions were actually reduced relative to the baseline, particularly in the dehydrated recycle cases, in which a water quench is used to condense flue gas moisture. This is a result of scrubbing by the highly alkaline fly ash of the test coal and may not be a general result. Further understanding of any differences between combustion with air and with recycle gas-O_2 mixtures was gained by study of unburned carbon, gas temperatures, furnace gas composition, and radiant heat flux. Carbon burnout appears to be faster with the recycle gas, particularly the dry recycle gas, than with air. Temperature profiles along the furnace tower were very similar for the baseline (air) and for the recycle gas, generally agreeing within 10%. This finding suggests that comparable performance and operability can be expected. Radiant heat flux for the recycle gases is considerably lower in the high- temperature flame zone than that demonstrated with air. This result is consistent with known emissivity of CO_2 and H_2O in contrast with N_2.

While the EERC work generally found no operational difficulties associated with use of the recycle with oxygen in place of air for combustion, the wet recycle did cause operational difficulties when used directly for coal transport. This was due to condensation of the recycle stream moisture on the coal. Therefore, drying of the coal transport stream by quenching is recommended. In the dry recycle case, the full recycle stream is quenched to remove moisture, and this difficulty does not arise. Among the issues that did not present operational problems are slagging and fouling, start-up, and switching from air to recycle gas. The conclusion of the EERC analytical and experimental program is that CO_2 recycle "may be applied successfully as a retrofit to a wide range of utility boiler and furnace systems." [Abele, 1987]

Another contribution of the ANL effort was a preliminary design study of issues in CO_2 compression and transport [Lynch, 1985]. The transport of CO_2 product must be recognized as extremely important to technical and economic success. This ANL design study recognized the need to transport CO_2 at supercritical conditions and to dehydrate the CO_2 to avoid condensation, freezing, and the formation of corrosive acids.

Specifications for the ANL study set the moisture limit at a mole fraction of 50 parts per million. At that level, glycol drying was the recommended technology, since solid desiccant adsorption towers would be very large and expensive compared with a glycol absorber. A four-stage compression system was recommended, with the glycol drying applied between the third and fourth stages. The more recent design study done by ABB for Trans Alta also recommends drying between the third and fourth stages of compression, but specifies a far lower moisture content of only 1 ppm molar [Palkes and Liljedahl, 1999]. For this specification, a molecular sieve dryer is recommended.

Canadian Studies

In the early 1990s, NRCan's CANMET Energy Technology Centre undertook combustion tests of various O_2-CO_2 mixtures at a vertical combustion test facility constructed for the purpose. After initial tests on bottled gas, flue gas recycle tests were performed to evaluate gas composition (NO_x, SO_2, and H_2O). Trans Alta Corporation then commissioned an engineering study of the Sundance Unit 1 plant to evaluate the feasibility of its conversion from air to O_2 with flue gas recycle. The Power Plant Laboratories of ABB Combustion Engineering performed the study with assistance from Air Liquide and ABB Lummus Global, Inc. The objectives of the engineering study were to (1) identify any expensive retrofits required in the pressure components of the furnace, (2) consider reusability of fans and other major auxiliary systems, and (3) estimate impacts on plant output and efficiency and the implied cost of recovered CO_2.

The outcome of the engineering study was consistent with the earlier ANL work, in that no major boiler modifications were required to achieve comparable heat duty performance. However, not surprisingly, the cost of air separation assured that the recovered CO_2 would be very expensive. The ABB study is very useful in that it provides engineering analysis and design details based on extensive practical experience with utility systems. Therefore, it does not simply confirm the Argonne results, but it lends them greater credibility. Among the engineering details of use in our model building effort are the addition of an oxygen heater, a supplementary feed water heater, and a gas cooler. The ABB study provides useful detail in the design and costing of these auxiliary systems, and their results provide a useful benchmark.

System Description

Our analysis extends earlier work with the addition of upstream and downstream processes to the PC plant. We assume the feasibility of a flue gas recycle conversion, and we use the results of earlier studies to calibrate a process model of the PC-recycle plant. The resulting mass and energy flows through that plant then determine input requirements from upstream processes and output to downstream processes. The overall system under study is summarized in Table 1 and Figure 1.

In previous studies, we have investigated a series of coal gasification options with CO_2 recovery. One common element among those studies has been the use of an IL No. 6 coal, which is commonly used as a design basis for gasifier studies. To remain consistent with those studies, we include that coal in the present study. However, those that are used closer to the destination oil field may be of more interest. Western sub-bituminous coal is one example. Therefore, we have expanded this study to include several coal options.

Table 1 includes conventional cryogenic air separation, as well as an unspecified alternative system, which may be a lower-conversion-efficiency cryogenic system, a membrane system, or some other alternative. Use of such a system will reduce the cost of air separation but increase the downstream treatment cost required to obtain a pure product for EOR. As listed in Table 1, this treatment includes removal of impurities (particularly O_2 and N_2), compression, and dehydration. The main difficulty here has been identified by ABB in the design study for Trans Alta: condensing CO_2 mixtures in the presence of impurities. This condensing step is necessary for separation of the impurities.

Table 1. Flue Gas Recycle PC Plant System with CO_2 Recovery

Raw Materials Supply	Coal Mining	Deep-mined, high-sulfur IL bituminous coal
		Surface-mined, low-sulfur western coal
	Coal Cleaning	Jig plant cleaning for IL coal
		No cleaning of surface-mined coal
	Coal Transport	Rail transport
	Air Separation	Cryogenic process, 99% purity
		Alternative systems with lower purity
	Water	Local availability assumed
Power Plant	Coal Preparation	Recycled flue gas used as drying medium and for coal transport
	Combustion	Conventional furnace retrofit to use mix of O_2 and recycled flue gas as oxidant
	Power Generation	Rankine cycle with high-, intermediate-, and low-pressure turbines
	Gas Separation and Cleanup	Particulate removal via ESP or fabric filter
		Sulfur removal via limestone scrubber
	CO_2 Product Preparation	Drying
		Removal of noncondensable gases
		Compression
Product and Waste Distribution	Power Transmission	Transmission lines and losses are not included in the system
	CO_2 Transport	CO_2 dehydration
		CO_2 compression
		Supercritical pipeline delivers CO_2 to oil field
	Solids Disposal	Ponding of scrubber sludge, ash and slag recovered for sale
	Emissions	Conventional air emissions from combustion are avoided

DESIGN ISSUES

Sulfur Dioxide Effects

Sulfur dioxide is of great importance for flue gas recycle systems. First, SO_2 is destructive to solvents used for either chemical or physical absorption of CO_2. Thus, successful removal of CO_2 from flue gas requires pretreatment by highly effective removal of SO_2. It was this fact that originally led Argonne researchers to consider separating nitrogen from CO_2 before combustion. The problem then becomes separating SO_2 from CO_2 rather than separating CO_2 from N_2 in the presence of the damaging SO_2. A second consideration is the performance and fate of SO_2 during EOR operation. Under some circumstances, SO_2 (and even NO_x) does not have to be removed. In general, the presence of SO_2 and NO_x will not hamper EOR operations. The presence of these gases may even improve EOR performance. Also, it has been demonstrated that for some formations, these gases are not always passed through to the extraction wells but are absorbed by the mineral formations through which the oil passes. However, a third consideration dictates the need for SO_2 removal. Acid gases in the presence of moisture can accelerate corrosion of the pipeline and other equipment. A fourth consideration is that these gases add to the burden of noncondensable gases, reducing pipeline capacity.

Scrubber Performance

The high sulfur concentration in U.S. coals was part of the motivation for this study as a follow-up to the design work done by ABB for Trans Alta, Corp. A series of sulfur-related concerns have been investigated. One of these concerns is that alkali scrubber-type SO_2 removal systems might experience performance degradation as a result of the high partial pressure of CO_2 in the recycle gas (compared to that in conventional flue gas). The formation of carbonates might reduce reagent utilization and reduce collection efficiency. We have resolved this concern with a series of laboratory-scale scrubber tests. Baseline performance for the laboratory scrubber was established by using a gas mix comparable with that used in conventional plant flue gas (14% O_2, 17.5% CO_2, 3600 ppmv SO_2, and about 200 ppmv NO, with the balance N_2). Flue gas from a CO_2 recycle plant was simulated with premixed gas of 12.2% N_2, 3% O_2, 3300 ppmv SO_2, 170 ppmv NO, and the balance CO_2. These gas streams were passed through the scrubber until the SO_2 removal efficiency declined because of saturation of the recirculating scrubber reagent. The principal test result is that the high CO_2 concentration does not impede SO_2 removal. It was also observed that the time elapsed until reagent performance declines is extended in cases of high CO_2 concentration.

Another significant result is that scrubber sludge oxidation due to sulfate is very limited in the high CO_2 cases. This is due to the low oxygen content of the test gas rather than the high CO_2 concentration. The observation, however, is relevant to a CO_2 recycle plant, since oxygen content must by strictly limited in the CO_2 product gas. Therefore, excess O_2 and air in-leakage will be tightly controlled, resulting in low flue gas oxygen content. Forced oxidation of the scrubber sludge will probably be required.

Clean or High-Sulfur Recycle?

Another sulfur-related issue that we have considered in some detail is the potential for the flue gas recycle system to greatly reduce the volume of gas that must be treated by the SO_2 abatement system. Because the recycle system has eliminated nitrogen as a diluent, the net amount of flue gas produced is only about 20% of that produced in a conventional plant. The substitute of recycled flue gas is merely an expedient way to preserve boiler temperature profiles. It does not affect the net gas generation. If the recycle is extracted prior to gas cleanup, the remaining gas volume going to treatment is small. This could result in considerable savings in the construction and operation of gas cleanup equipment. Of course, such an approach will result in the buildup of SO_2 and other impurities in the boiler. The recycle might even be extracted prior to particulate removal, resulting in ash recycling. This is not generally expected to be appropriate in a retrofit situation, and existing plants have already made the investment in highly efficient particulate removal. Most likely, a dirty recycle stream will be extracted after particulate removal.

We have incorporated a split recycle in our model to investigate the buildup of SO_2. The split recycle system extracts some of the full recycle before sulfur removal and some following sulfur removal. In this way, the buildup of SO_2 can be controlled as required by materials limitations for a particular boiler. This is a fundamental design issue for the CO_2 recycle retrofit. The best option is likely to be coal- and plant-specific.

Recycle Specifications

The background provided above includes several references to the flue gas/O_2 recycle ratio. This is a fundamental design parameter. Nitrogen has a very important role in combustion and heat transfer. It tempers the combustion by absorbing heat. This determines the relative amount of heat transfer in the radiant and convective sections of the boiler. CO_2 and N_2 differ substantially in both heat capacity and emissivity, so a simple volumetric substitution of CO_2 for N_2 is expected to result in a change in the heat transfer balance within the furnace. This could require boiler modifications, such as changes in the number of tubes and in the steam flow patterns within the boiler. To avoid such expensive retrofits, the ratio of recycled flue gas to O_2 must be adjusted to preserve the original boiler heat balance. It is not possible to have precisely the same heat balance throughout the boiler with such a dramatic change in gas composition. Furthermore, differences in boiler design suggest that the right gas mix will be boiler-specific. The earlier ANL studies recommended a recycle ratio of about 2.7 (moles CO_2 + H_2O per mole O_2) for a dehydrated recycle stream and 3.2 for a wet recycle stream. That work also found that boiler operating adjustments could accommodate variation of up to ±0.4. The more recent design study by ABB assumed a recycle ratio of 3.0. The recycle for the ABB design is dry, but a substantial amount of moisture is introduced with the coal, so the ABB assumption is consistent with the earlier findings.

CO_2 Product Specifications

High purity is required for the CO_2 product. ABB assumes 98% CO_2 with the remaining any combination of O_2, N_2, argon, NO_X, and SO_2. Water content is limited to 1 mole ppm. This specification is an important determinant of retrofit design features. The limit on N_2 implies that high-purity O_2 must be used. A reasonable assumption is 99%. Higher purity is obtained only at substantial additional cost. To meet these specifications, it is essential to limit excess O_2 and air in-leakage. Combustion requires excess O_2. A conventional plant will use approximately 20% excess air and will expect 10% or more additional air through leakage. Even a substantial reduction in these values for the retrofit plant will clearly violate the product specifications. ABB concluded that separation of O_2 and N_2 through condensation of CO_2 followed by stripping of the dissolved oxygen and nitrogen was essential. They also determined that lower O_2 quality (e.g. 95%) would make it impractical to condense CO_2 for this separation process.

MODEL DEVELOPMENT

ASPEN Plus

As in any modeling exercise, the modeling tool must be selected according to the project objective. Our objective is to characterize mass and energy flows in sufficient detail that changes in coal composition, O_2 purity, recycle strategy, and process equipment effectiveness will be reflected in predictions of product composition, power output, and residual emissions. To meet these objectives, we have used ASPEN Plus to model processes at the power plant, from coal preparation through CO_2 product compression and delivery via pipeline. Convergence algorithms in ASPEN Plus are particularly important for the recycle calculations to obtain steady-state flows and compositions. The physical property models available in ASPEN Plus are essential to represent the pipeline system performance. Because of continuing interest in these advanced power plant systems, models developed in this program are expected to have application to future programs. Thus, there is interest not only in model results, but also in the modeling process itself. With this in mind, a description of some modeling issues specific to ASPEN Plus is provided here.

Coal Specifications and Coal Preparation

ASPEN Plus treats bulk coal as a non-conventional solid. This is ASPEN Plus terminology for solids that do not participate in chemical reactions. For such materials, the composition is unimportant. This treatment of coal is adequate for physical processes, such as grinding and screening. Before coal can participate in a chemical process, such as combustion, a conceptual reactor is used to convert the bulk coal into its constituents, which are conventional chemical components. Thus, the coal specifications required by ASPEN Plus include physical and chemical properties.

Coal preparation at the power plant includes drying and pulverizing, which are accomplished simultaneously in modern plants. We have modeled coal drying in a separate process from crushing because of the nature of ASPEN Plus. ASPEN Plus includes some solids handling unit operations, including crushing, which can be used to represent pulverizing. Required input includes coal particle size distribution and a grindability index, such as the Hardgrove Grindability Index. The model calculates the pulverizer power requirement, which is useful for estimating net plant power. For solids handling calculations coal chemical composition is not a consideration.

Coal specifications used elsewhere in the model for mass balance and chemical calculations additionally include the ultimate analysis, proximate analysis, and sulfur analysis. Because a consistent set of values for these analyses must be used, it is not possible to perform a sensitivity study on a single parameter, such as sulfur content. Instead, results are obtained for several alternative coals exhibiting the variations of interest.

Coal drying is important from our materials flow perspective because it is one strategy to limit the moisture that must be removed from the flue gas. It is also important from a plant operations perspective. For instance, it may affect ignition characteristics. That is, the plant operator may establish specifications for maximum coal moisture content. Plant conversion to CO_2 recycle may require changes in drying practice at some plants. The most common drying systems are direct or semi-direct. In these systems, the drying medium (normally, heated air) follows the pulverized coal into the furnace carrying the moisture with it. Clearly, we do not want to introduce air into the furnace, so a portion of the recycled flue gas is used as the drying medium. In an indirect drying system, the pulverized coal is stored and the drying medium is released to the atmosphere. In this case, heated air or nitrogen may be used as the drying medium, but some residual from this drying medium will be introduced to the furnace with the coal. Thus, conversion to a direct system may be required to limit the introduction of air or nitrogen.

Combustion

Prior to combustion, a conceptual reactor is used to convert coal into its constituent elements. At this point, the coal is converted from a non-conventional solid into a mix of conventional chemical components and ash, another non-conventional solid. The conventional components can then be processed in a chemical reactor. Combustion, for instance, is modeled as a reaction between the coal constituents and the oxidant stream (O_2 with recycled flue gas). The combustion reactor is modeled as a non-adiabatic Gibbs reactor. The Gibbs reactor achieves equilibrium based on Gibbs free energy calculations. If the reactor were adiabatic, very high flame temperatures would be attained, and the equilibrium composition would include substantial amounts of carbon monoxide. A more realistic furnace representation sets a target temperature for the combustion products and calculates heat released in the process. This heat release represents radiant heat transfer to the water walls of the furnace. Again, our models characterize gas flows and gas stream properties. We have not developed a thermal equilibrium heat transfer model that can estimate gas path heat transfer based on radiative properties, convective flows and furnace geometry, and surface characteristics. However, we can incorporate the results of such detailed modeling as performed for ANL by the EERC [Abele, 1987]. By using the recommended recycle ratio from that study, we can have some confidence that the retrofit furnace will achieve performance comparable to that of the original plant.

Since our model uses an equilibrium reactor, perfect mixing is implicitly assumed, and reactor kinetics (reaction rates) does not enter into the calculation. Therefore, we cannot directly estimate excess oxidant requirements. In a conventional pulverized coal plant, 15–20% excess air is required to assure complete combustion. Nothing in the use of flue gas recycle suggests that less excess air will be required in the retrofit plant. The ABB design for Trans Alta includes 25% excess O_2 when all oxygen-bearing streams entering the furnace are taken into account. For our model, the use of the design specification features of ASPEN Plus is very useful. A design specification or target value is set for the O_2 flow rate of the furnace exhaust. For instance, that target might be set at 20% of the molar flow of carbon in the feed coal. A variable parameter, in this case the feed rate of the pure O_2 stream, is adjusted until the design specification is met. It is significant that a 20% excess O_2 requirement does not mean that 20% excess O_2 is consumed. The use of the recycle returns most of the excess O_2 to the furnace. Some exits with the product CO_2 and is largely removed by the subsequent stripping operation.

Gas Treatment

The flue gas exiting the furnace is cooled by heat exchange with recycled gas oxygen going to the furnace. Following this heat recovery, particulates are removed in the electrostatic precipitator (ESP), and sulfur dioxide is removed in a wet scrubber. ASPEN Plus includes an ESP unit operation. The ESP model can be used in a design mode, in which the incoming particle size distribution and a target removal efficiency are used to estimate basic design parameters. Particle separation is then accomplished at the specified efficiency.

We introduce the first recycle stream separation after the ESP. As noted above, any recycle extracted at this point reduces the treatment burden of the scrubber or other sulfur removal process. If this system is being retrofit to an existing plant currently without sulfur control, this strategy could be beneficial. The portion of the stream not sent to the first recycle is treated in a limestone scrubber for sulfur removal. An alternative sulfur recovery system was also investigated. That system employs the Beavon process to reduce SO_2 to H_2S, which is then treated in a Claus-SCOT system for recovery of elemental sulfur. Such a system has the considerable advantage that it produces a salable by-product and minimal waste. However, capital costs for such a system are higher than those for a limestone scrubber. These capital costs would be substantially reduced if the treated volume is reduced by extraction of the recycle before sulfur removal.

Sulfur separation in the scrubber is represented simply by a splitter, which diverts a fraction of the SO_2 flow to a waste stream. We have experimented with an electrochemical scrubber model built in ASPEN Plus, but at this point, only the simpler approach has been implemented.

Recycle

A second recycle stream is extracted after the scrubber. The flow rate of this stream is determined by a design specification that sets the total recycle flow consistent with the recycle ratio target. The proportions of clean and dirty recycle will depend upon boiler component tolerance for SO_2, cost of the SO_2 scrubber, coal sulfur content, and site-specific plant characteristics. We have simply used our model to estimate flue gas composition and flow rates under various recycle scenarios. Consideration of these other issues will be included in subsequent research.

CO_2 Product Preparation and Transport

Our ASPEN Plus model is being extended to include gas separation, compression, and dehydration required for final preparation of the CO_2 product. Gas separation is required to eliminate excess O_2 and N_2 that has entered the process with feed coal, through infiltration, and because of the need for excess oxidant. Gas separation is a complex system requiring condensation of the CO_2 followed by stripping of dissolved O_2 and N_2. This process will be modeled in sufficient detail to estimate the energy balance around the system so that its effect on net plant energy and cooling water requirements can be predicted.

The purified CO_2 is compressed and dehydrated prior to pipeline transport to the oil field as a supercritical fluid. We have modeled the delivery and deep-well injection in detail to be certain that the compression pressure is adequate to maintain super-critical fluid throughout the process. A number of ASPEN Plus physical property models were evaluated by using the models to calculate density horizontally along the pipeline and vertically to the injection point. Comparison of these model predictions against reported values from industry has allowed us to calibrate the ASPEN Plus property model that best represents the product stream.

RESULTS

The cost of process equipment and the cost of its operation can generally be estimated as a sum of exponential terms, where one term represents the physical size, and a second term represents effectiveness or efficiency. For SO_2 scrubbers, the size parameter is normally gas flow rate, and the effectiveness term is SO_2 removal rate, which depends on removal efficiency and SO_2 concentration in the feed stream. For capital cost, the gas flow rate is by far the most important parameter. There is an exponential scaling effect, so costs do not increase precisely linearly with increasing flow rate, but the increase is nearly linear. Operating costs also depend upon gas flow rate because of fan energy use and reagent pumping required for an effective liquid-to-gas ratio. Since the flue gas recycle plant has eliminated nearly 80% of the net flue gas volume, there is an opportunity to reduce scrubber costs by extracting some or all of the recycle prior to the scrubber.

One concern with this strategy is the buildup of SO_2 in the gas path. A portion of the SO_2 is converted to SO_3. This combines with water to form sulfuric acid vapor, which can condense on gas path surfaces at temperatures below the sulfuric acid dew point of around 250–300˚F. Normal plant operation allows for minimum metal temperatures in the air heater to be somewhat less than this dew point in order to optimize plant efficiency (by extracting the greatest possible sensible heat from the flue gas). In the presence of high sulfur concentrations, minimum metal temperatures must be higher, and an efficiency penalty is incurred. Our model does not yet have the sophistication needed to tie all of these factors together and estimate the penalty. However, the gas path mass balance and chemistry models reveal the SO_2 concentrations as a function of the amount of recycle drawn before and after sulfur removal. These results are summarized in Table 2 for a 1.5% sulfur coal. Note that even the use of 100% clean recycle results in a slight elevation of SO_2 concentration relative to the non-recycle case.

Table 2. Influence of Recycle Strategy on SO_2 Concentration in the Flue Gas

Fraction of Total Recycle Extracted before Sulfur Removal	Sulfur Concentration in Flue Gas (ppmv)
0.7	3110
0.6	2370
0.5	1920
0.4	1650
0.3	1390
0.2	1230
0.1	1080
0	1000

Additional results from the mass and energy balance calculations will be presented at the conference. These include net emissions, product gas composition for selected coals, and net energy.

COMPARATIVE COSTS OF POWER CYCLES

Table 3 includes the results of this assessment for the CO_2 capture from a pulverized coal boiler, including the costs of transportation and CO_2 sequestration in a table showing the comparative costs of several fossil-based and non-fossil-based energy cycles. In 1996, the California Energy Commission [CEC] undertook a broad survey of pricing for various power-generating technologies [CEC, 1996] that was combined with CO_2 inventory data for the same power-generating technologies from the U.S. DOE Energy Information Agency [DOE, 2000]. Consistent with these numbers, a recent EPRI study has compared all the cost estimates for 90% CO_2 capture systems that also appear in Table 5 [Holt and Booras, 2000]. Transportation costs for the CO_2 assume a fully developed infrastructure cost of \$7.82/tonne CO_2 [Doctor, et al., 2001], as compared against first system costs of \$25/tonne CO_2 [Doctor et al., 1994}. No adjustments were made for the 1996 costs of natural gas because the CEC did not structure its report so that fuel costs could be manipulated separately, but with the necessary adjustments, the costs of turbine combined-cycle systems would be comparable with or higher than those if CO_2 sequestration is to be included. The costs of sequestration in the field are based on the observation that during 1999–2000, the breakeven for CO_2-flooding EOR required crude prices higher than \$12/bbl oil. If the typical utilization of 5,600 standard cubic feet/bbl of oil is employed, this equates to \$2.14/1,000 standard cubic feet of CO_2, or \$34/tonne CO_2.

	Cost - Base	Cost - CO_2	Transport	Reservoir	TOTAL
PC Coal with retrofit recycle	20.0	33.3	6.8	29.8	69.9
Natural Gas with co-gen [1]	41.5	59.5	2.6	11.4	73.5
Hydroelectric	82.0	0.0	0.0	0.0	82.0
IGCC Coal	52.4	65.7	5.7	25.1	96.5
PC Coal with co-gen	50.5	82.5	6.8	29.8	119.1
Nuclear	125.5	0.0	0.0	0.0	125.5

1. Natural Gas pricing from 1996 assumed

CONCLUSION

This process design employs an ASPEN model for developing a system with a pulverized coal-fired boiler and CO_2 recycle. This approach is technically feasible without major boiler modification. However, if the CO_2 is being produced for commercial use in EOR, the economics of the retrofit are unfavorable, unless incentives or requirements for CO_2 emission reduction are implemented. In addition, while experimentally investigating the possible impact on flue gas desulfurization of high CO_2 concentration in the flue gas, this presents no difficulty for the proposed concept.

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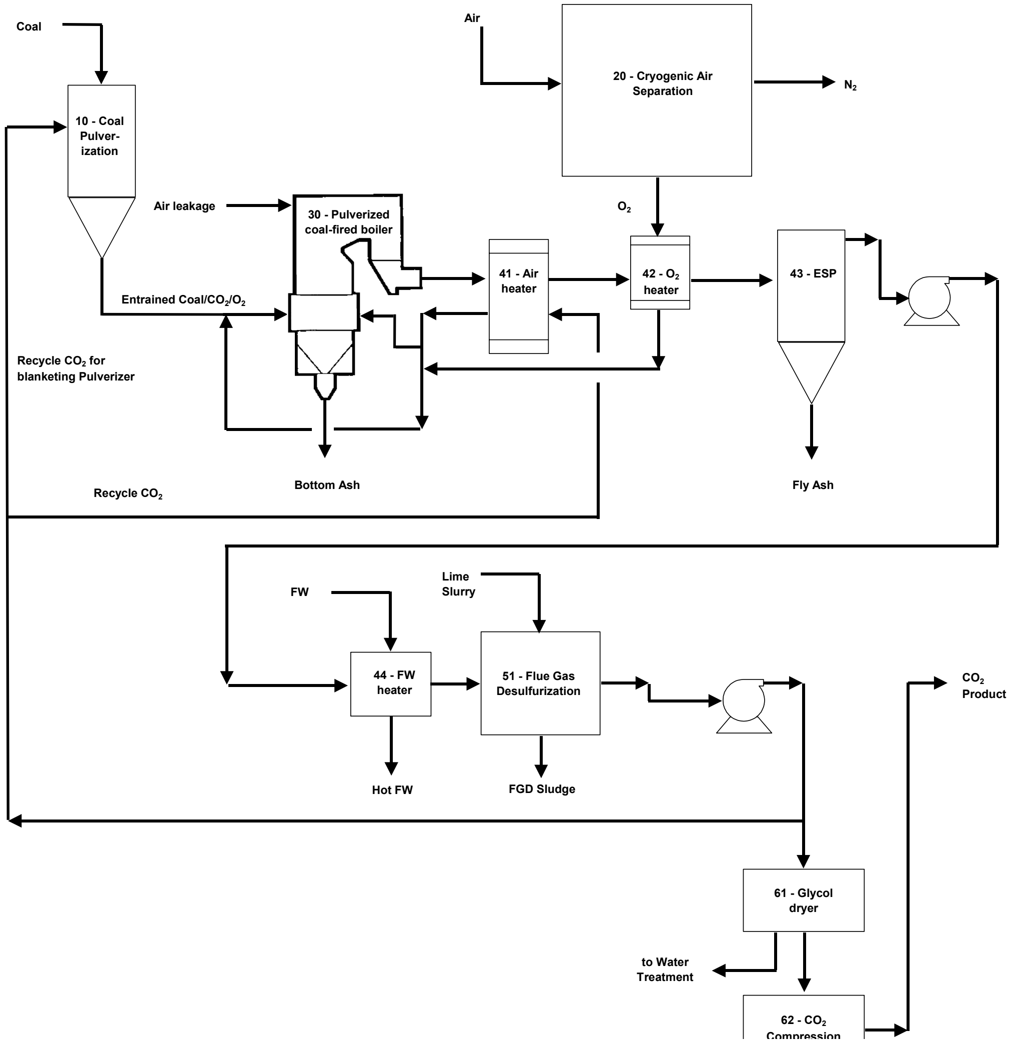

Figure 1. Pulverized Coal-fired Boiler with CO_2 Recycle