The following manuscript was unavailable at time of publication.

A GENERIC NO$_x$ CONTROL INTELLIGENT SYSTEM (GNOCIS) FOR COAL-FIRED POWER PLANTS

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The Integrated Environmental Control Model

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Introduction

The capability to estimate the performance and cost of emission control systems is critical to a variety of planning and analysis requirements faced by utilities, regulators, researchers and analysts in the public and private sectors. The computer model described in this paper has been developed for DOE to provide an up-to-date capability for analyzing a variety of pre-combustion, combustion, and post-combustion options in an integrated framework. A unique capability allows performance and costs to be modeled probabilistically, which allows explicit characterization of uncertainties and risks.

Modeling Framework

Previous reports have described earlier development and applications of the Integrated Environmental Control Model (IECM).[1,2] The IECM provides a systematic and integrated treatment of emission control options employing pre-combustion, combustion and post-combustion control methods. The model was developed to provide preliminary performance and cost estimates for new coal-fired power plants. Of particular interest were a number of advanced environmental control technologies being supported by DOE. For comparative purposes, however, a set of "baseline" technologies representing current commercial systems also is part of the IECM framework. Table 1 lists the technologies currently included. For each technology, a process performance model was developed to account for all energy and mass flows (including air pollutants and solid wastes) associated with that process. Coupled to each performance model, an economic model was developed to estimate the capital cost, annual operating and maintenance (O&M) costs, and total levelized cost of each technology. The technology models developed for the IECM in the mid-to-late 1980s now are being updated and enhanced to reflect more recent design criteria and associated performance and costs. The status of major IECM components is briefly reviewed in the paragraphs below.

Coal Cleaning Processes. The IECM includes models of both conventional and advanced coal cleaning processes. The conventional processes include four plant designs of increasing complexity, which provide increasing capability for sulfur as well as ash removal. Each of these plant designs (referred to as cleaning levels 2, 3, 4 and 5) can be optimized to achieve a target sulfur or ash reduction while maximizing overall yield (thus, minimizing costs). Data requirements for these models includes coal-specific washability data plus cleaning circuit design parameters such as top size and bottom size for different coal fractions.

Models of several advanced physical coal cleaning processes also have been developed based on limited data for several U.S. coals. While these processes are capable of achieving higher levels of sulfur and ash reduction than conventional processes, their costs also are higher. Several of these processes have been developed to provide "super-clean" coal for use in coal-liquid mixture fuels, which compete with other premium fuels such as oil or gas.

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Table 1. Emissions control technology options for the IECM

Plant Area	Baseline Processes	Advanced Processes
Physical Coal Cleaning	• Level 2 Plant • Level 3 Plant • Level 4 Plant • Froth Flotation	• Selective Agglomeration • Heavy Liquid Cyclones • Coal-Pyrite Flotation • Magnetic Separation
Combustion Controls	• Low NO_x Burners	• Reburning (gas)[a] • Slagging Combustors[a]
Post-Combustion Controls	• Selective Catalytic Reduction (Hot-side and Cold-Side) • Wet Limestone FGD • Wet Limestone with Additives • Wet Lime FGD • Lime Spray Dryer • Electrostatic Precipitator (Cold-side) • Reverse Gas Fabric Filter • Pulse Jet Fabric Filter	• NOXSO • Copper Oxide • Electron Beam • Advanced SO_2/NO_x Removal[a]
Waste Disposal & By-Product Recovery	• Landfill • Ponding	• Sulfur Recovery • Sulfuric Acid Recovery • Gypsum

[a] To be completed in project Phase II.

Base Power Plant. Performance and cost models of a base power plant are needed to accurately characterize the cost of integrated emission control systems, particularly when coal cleaning is employed. The IECM base plant performance model includes detailed mass and energy balances, fuel combustion equations, and thermodynamic relationships to calculate flue gas flow rates, plant efficiency, and net power generation. The environmental performance of the furnace also is determined from mass and energy balances where possible, or from empirical relationships where necessary, as in the case of NO_x emissions. A detailed model of the air preheater also has been developed to properly account for energy credits when advanced environmental control processes are used.

Revised cost models for the base power plant have been developed based on recent data from EPRI for three furnace designs related to coal rank (bituminous, subbituminous and lignites). The new cost algorithms estimate capital costs and annual O&M costs as a function of key design parameters.

NO$_x$ Controls. The IECM includes both in-furnace and post-combustion NO_x control options. Currently, the in-furnace combustion controls include low NO_x burners for a new power plant meeting or exceeding federal New Source Performance Standards. Additional combustion options suitable for NO_x retrofits currently are being developed and will be implemented in the IECM during 1995.

Post-combustion control methods include both "hot-side" and "cold-side" selective catalytic reduction (SCR) systems. New SCR performance and cost models incorporate recent data

and experience from SCR units worldwide. The revised models contain a larger number of system design parameters, a more detailed characterization of catalyst activity, and additional details related to capital cost and O&M cost parameters.[4] While SCR systems on coal-fired plants are only now emerging commercially in the United States, their widespread use in Europe and Japan, often in combination with FGD, represents the benchmark design for comparisons with advanced emissions control systems being developed by DOE.

Particulate Emission Controls. The IECM includes performance and cost models of cold-side electrostatic precipitators and fabric filters. Performance and cost models for both technologies recently have been updated to reflect current applications.[4] The revised ESP performance model calculates total particulate removal as a function of ash composition and flue gas properties, while fabric filter performance is related primarily to the air-to-cloth ratio. The latter models also have been expanded to include both reverse gas and pulse jet fabric filter designs. Recent EPRI studies have been used to update the economic models for all particulate collectors in the IECM.

Flue Gas Desulfurization Systems. Substantial improvements in FGD system design, accompanied by reductions in cost, have been seen over the past decade, and recent enhancements to the IECM modules now reflect these changes.[5] New FGD performance and cost models have been developed for the IECM for four common types of FGD systems: (1) wet limestone with forced oxidation; (2) wet limestone with dibasic acid additive; (3) magnesium-enhanced wet lime system; and (4) a lime spray dryer system. The new cost models reflect the results of recent studies for EPRI, while the new performance models represent the capabilities of modern commercial systems.

Combined SO_2/NO_x Removal Processes. A key element of DOE's Clean Coal Technology program focuses on advanced processes for combined SO_2 and NO_x removal to achieve high performance goals at lower cost than the conventional combination of SCR plus FGD. Models of three SO_2/NO_x control systems have been developed for the IECM: the fluidized-bed copper oxide process, the NOXSO process, and the electron beam process. The copper oxide and NOXSO processes are of continuing interest to DOE, and earlier versions of the performance and cost models for these two processes are being refined and updated at the present time.

Waste Disposal and By-Product Recovery Systems. IECM revisions treat solid waste disposal as a variable cost item associated with a particular control technology, consistent with the costing method used by EPRI and others. Thus, boiler bottom ash disposal is included in the base plant model, fly ash disposal costs are incorporated in the ESP or fabric filter models, and FGD wastes or by-product credits are treated in the FGD cost models.

Advanced processes employing combined SO_2/NO_x removal produce by-product sulfur or sulfuric acid rather than a solid waste. Because the sulfur or sulfuric acid plant is a significant part of the overall plant cost, separate engineering models have been developed for these two components. These models are sensitive to input gas composition and other parameters affecting overall process economics.

Probabilistic Capability

A unique feature of the IECM is its ability to characterize input parameters and output results probabilistically, in contrast to conventional deterministic (point estimate) form. This method of analysis offers a number of important advantages over the traditional approach of examining uncertainties via sensitivity analysis. Probabilistic analysis allows the interactive effects of variations in many different parameters to be considered simultaneously, in contrast to sensitivity analysis where only one or two parameters at a time are varied, with all others held constant. In addition, probabilistic analysis provides quantitative insights about the *likelihood* of certain outcomes, and the probability that one result may be more significant than another. This type of information is generally of greater use than simple bounding or "worst case" analyses obtained from sensitivity studies, which contain no information on the likelihood of worst case occurrences.

The ability to perform probabilistic analysis comes from the use of a new software system which uses a non-procedural modeling environment designed to facilitate model building and probabilistic analysis. In addition to a number of standard probability distributions (e.g., normal, lognormal, uniform, chance), the IECM can accommodate any arbitrarily specified distribution for input parameters. Given a specified set of input uncertainties, the resulting uncertainties induced in model outputs are calculated using median Latin Hypercube sampling, an efficient variant of Monte Carlo simulation. Results typically are displayed in the form of a cumulative probability distribution showing the likelihood of reaching or exceeding various levels of a particular parameter of interest (e.g., emissions or cost).

Model Applications

The IECM is intended to support a variety of applications related to technology assessment, process design, and research management. Examples of questions that can be addressed with the IECM include the following:

- What uncertainties most affect the overall costs of a particular technology?
- What are the key design trade-offs for a particular process ?
- What are the potential payoffs and risks of advanced processes vis-a-vis conventional technology?
- Which technologies appear most promising for further process development?
- What conditions or markets favor the selection of one system design (or technology) over another?
- How can technical and/or economic uncertainties be reduced most effectively through further research and development?

The IECM also has been modified to allow estimation of retrofit costs as well as new plant costs. A series of user specified "retrofit factors" may be applied at the process area level for a particular system to estimate the higher costs of retrofit facilities. To operate the IECM, a new graphical interface has been developed which provides an extremely user-friendly mode of operation. As currently configured, the IECM runs on a Macintosh computer.

Illustrative Results

Here we present results illustrating the probabilistic capabilities of the IECM as applied to a single power plant as shown in Figure 1. This case corresponds to an Appalachian medium sulfur (2.1%S) coal for a power plant with a net capacity of 300 MW. Table 2 shows the input parameter values and uncertainty distributions assumed for this example. The plant is

assumed to achieve 95% SO_2 removal employing a wet limestone FGD system with forced oxidation.

Cost results are shown in Figure 2. These figures show the cumulative effect of uncertainties in different model parameters. For example, for capital cost (Figure 2a),
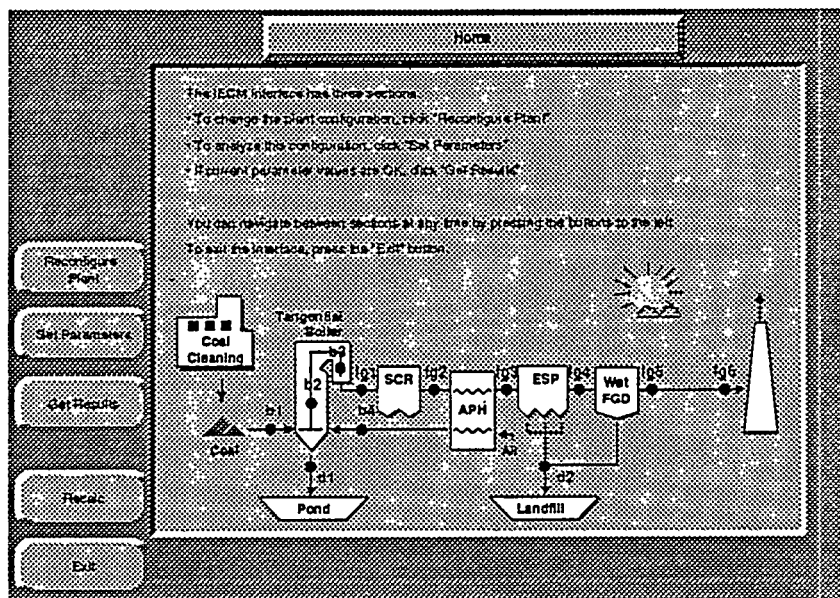

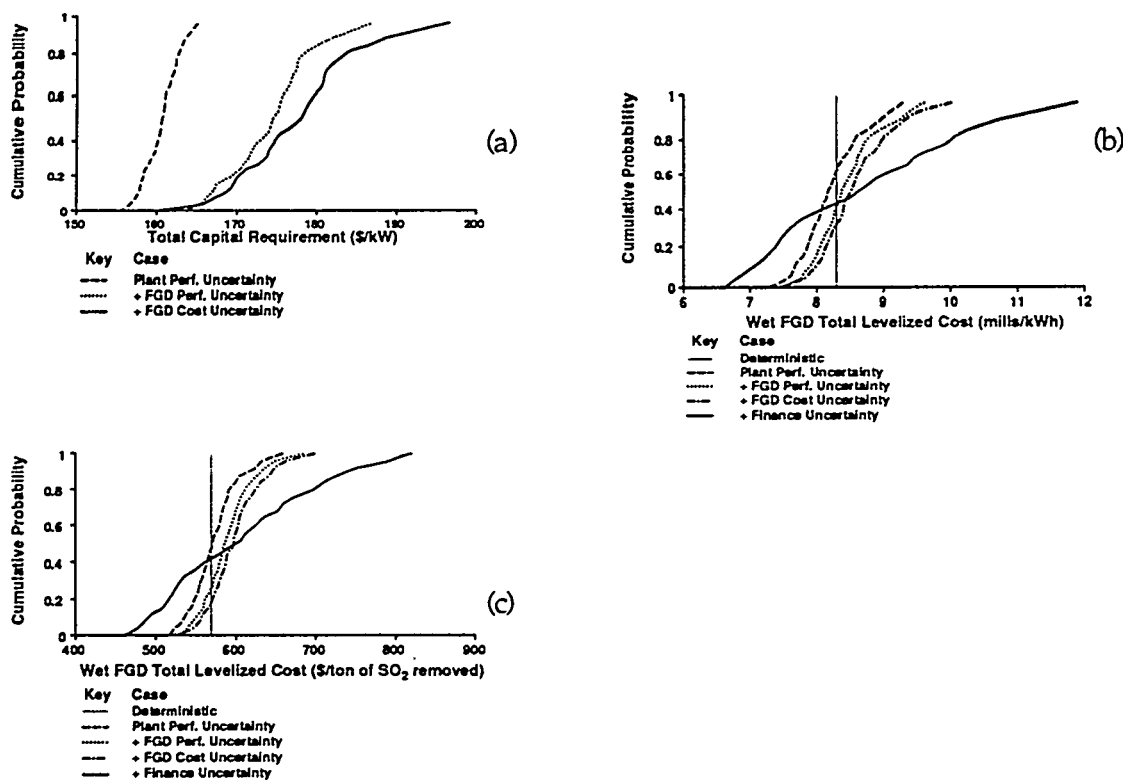Figure 1.   User interface screen showing the case study plant configuration.

Figure 2.   Probabilistic results for case study: (a) FGD capital cost; (b) total levelized cost; (c) cost per ton of SO_2 removed. All costs are in constant 1993 dollars.

Table 2. Performance and cost parameter uncertainties for case study

Model Parameter	Nominal Value	Probability Distribution[a]	Parameter Range[b]
Gross Capacity	330 MW (300 MW net)	None	
Coal Sulfur Content[c]	2.13 wt%	None	
Steam Cycle Heat Rate	7880 Btu/kWh	Neg. Half Normal (1,0.018)	(7599, 7871)
Capacity Factor	65%	Normal (1,0.07)	(58, 72)
Excess Air (boiler)	20%	Uniform (0.75,1.25)	(15,25)
Air Preheater Leakage	19%	Triangle (0.4,1,1.25)	(9, 23)
Economizer Outlet Temp.	700 °F	Normal (1,0.03)	(666, 734)
Air Preheater Outlet Temp.	300 °F	Normal (1,0.03)	(285, 315)
Energy for Coal Pulverizers	0.6% MW_g	Normal (1, 0.03)	(0.57, 0.63)
Energy for Steam pumps	0.65% MW_g	Normal (1, 0.03)	(0.61, 0.68)
Energy for Fans	1.5% MW_g	Normal (1,0.03)	(1.4, 1.6)
Energy for Cooling Towers	1.8% MW_g	Normal (1,0.03)	(1.7, 1.9)
Energy for Misc. Equip.	1.3% MW_g	Normal (1,0.03)	(1.2, 1.4)
SO_2 Removal Efficiency	95 %	None	
Particulate Removal Efficiency	50 %	None	
Reagent Stoichiometry	1.03	Triangular (0.99,1,1.05)	(1.02,1.03,1.08)
Capacity of each Absorber	50%	Uniform(1,1.2)	(50, 60)
Gas Temp. Exiting Reheater	152 °F	None	
Total Pressure Drop	10 in H_2O	Normal (1,0.03)	(9.5, 10.5)
Chloride Removal Efficiency	90 %	None	
Gen. Facilities Capital	10% PFC	Lognormal(1,1.3)	(6.3, 15.9)
Eng. & Home Off. Fees	10% PFC	Half Normal (1,0.17)	(10.1, 13.3)
Project Contingency	15% PFC	Normal (1,0.11)	(12.3, 17.9)
Process Contingency	2% PFC	Half Normal (1,0.5)	(2.0, 4.0)
Limestone Cost	$15/ton	Uniform(0.7,1.3)	(10.5, 19.5)
Disposal Cost	$8.15/ton	Normal(1,0.1)	(6.8, 9.5)
Operating Labor Rate	$20/hr	Uniform(0.75,1.25)	(15,25)
Inflation Rate	0%	None	
Plant Book Life (years)	30 yrs	None	
Real Return on Bonds	4.6%	None	
Real Return on Com. Stock	8.7%	None	
Real Return on Pref. Stock	5.2%	None	
Real Cost Escalation Rate	0%/yr	None	
Federal Tax Rate	36.7 %	None	
State Tax Rate	2 %	None	
Property Tax Rate	2 %	None	
Investment Tax Credit	0%	None	
Fixed Charge Factor	0.0877	Uniform(0.7,1.3)	(0.06, 0.11)
O&M Levelization Factor	1.00	Normal (1,0.03)	(0.95, 1.05)

[a] For normal distributions the values in parenthesis are the mean and the standard deviation normalized on the nominal value. Uniform distributions show the range, and triangular distributions the range and mode also normalized on the nominal value.

[b] The parameter range for the normal distribution encloses two standard deviations or about 95% of the values.

uncertainties in only the base power plant parameters introduces some skewness to the curve, reflecting primarily the input distribution for steam cycle heat rate. That heat rate distribution (see Table 2) reflects a potential for improved plant efficiency relative to the nominal (deterministic) case. Additional uncertainties in the FGD system performance parameters introduce a larger variation in plant capital costs, and this is expanded further when uncertainties in model cost parameters also are added. In this case, the shift to the right toward higher costs reflects the parameter input distributions used for this example, in particular the assumptions of possible vessel oversizing, higher contingency costs, and higher reagent stoichiometry relative to the nominal design values. As a result of these more conservative design assumptions, Figure 2a shows a 95 percent confidence interval ranging from $163 to $196/kW, compared to the nominal (deterministic) value of $165/kW.

Similar results are seen in Figures 2b and 2c for the total levelized cost and cost per ton of SO_2 removal. Here, the cumulative probability distributions also show the effect of additional uncertainties in plant operation and financial parameters which affect annual revenue requirements. Again, the input assumptions in this example reflect a greater potential for higher rather than lower costs relative to the nominal parameter values. Thus, in Figure 2c the cost per ton of SO_2 removed varies from $470 to $805/ton for the 95 percent confidence interval, compared to a nominal value of $570/ton with no uncertainties. Figure 2c shows about a 60 percent chance that the uncertainties assumed in this example will lead to a higher cost per ton than the deterministic value.

While one can see graphically in Figures 2 that certain sets of parameters have a more pronounced effect on overall uncertainty than others, more formal mathematical methods can be used to identify the key parameters whose uncertainty most affects the outcome. The importance of an input variable is derived by assessing its influence on the output variable of interest. For linear relationships this can be accomplished by the use of partial correlation coefficients. For nonlinear relations, such as those involved here, it becomes more difficult to assess the importance of individual variables. However, if the model output is a monotonic function of the input then it is possible to linearize the relationship by using rank transformations on the input and output values. The transformation involves replacing each value of a variable by its rank. Now the partial correlation coefficient on the rank transformed variables (PRCC) can be used to assess the importance of each input variable.

The results of such an analysis are shown in Table 3 for the plant in this example. In general, plant size and coal sulfur content are the variables that most affect plant costs. However, other parameters also strongly affect either capital cost, O&M costs, or total levelized cost in this example. For instance, the design absorber capacity — which varies from 50 to 60 percent of the flue gas flow rate in this analysis — is the second most important parameter affecting FGD capital cost for this plant. Note that the rank order of different variables may vary from case to case, depending on the magnitude and uncertainty in each parameter. Because a large number of technical and economic factors affect the performance and cost of environmental control systems, PRCC analysis can be especially useful for identifying key process variables whose uncertainty might be reduced through targeted research and development, especially for advanced environmental control technologies.

Table 3. Results of partial rank correlation coefficient (PRCC) analysis showing the five most important model parameters affecting FGD cost for the illustrative example

Model Parameter	Capital Cost (M$)	O&M Cost (M$)	Total Levelized Cost (mills/kWh)	SO$_2$ Removal Cost ($/ton)
Gross Capacity	1	1	1	2
Coal Sulfur Content	3	2	3	1
Capacity Factor		4	4	5
Fixed Charge Factor			2	3
Absorber Capacity	2			
Reagent Stoichiometry	4			
Electricity Cost		3	5	4
Limestone Cost		5		
General Facilities Cost	5			

Conclusion

This paper has described an integrated modeling framework for evaluating the cost and performance of power plant emission control systems. While the illustrative examples in this paper focus on modern FGD systems, a key purpose of these models is to facilitate comparisons between alternative systems, particularly advanced technologies that may offer improved performance and/or cost characteristics. In such cases, the probabilistic capability of the models described here can be especially helpful in quantifying the risks as well as potential payoffs of advanced technologies, investment strategies, and R&D priorities.

Acknowledgments

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Hollow Fiber Contactors for Simultaneous SO_x/NO_x Removal

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INTRODUCTION

Control of SO_x and NO_x emissions from coal-fired utility and industrial boilers is a topic of growing national and international importance. Whereas a host of commercial and semi-commercial processes exist (Drummond and Gyorke, 1986), increasingly stringent emission standards mandate that processes be low cost, highly efficient, and, ideally, produce marketable byproducts. Although a wide range of site-specific economic factors dictate the best choice of SO_x and NO_x control, removal of SO_2 by wet limestone scrubbing and removal of NO_x by selective catalytic reduction (SCR) are regarded as the best commercialized technologies for this application. A typical wet limestone plant requires approximately \$100 to \$120 of capital investment per kilowatt of electric power generating capacity and has power and miscellaneous operating costs in the range of 5 to 6 mils per kilowatt hour (kWh). The SCR process requires approximately \$70 to \$100 of capital investment per kilowatt of electric power generating capacity. Its operating costs, including ammonia consumption, is in the range of 1.8 to 2.2 mils per kWh. These costs can add approximately 20% to the final cost of generated electricity, a significant burden on rate payers.

Our proposed research is aimed at developing (to a subscale prototype size) a new process for removal of SO_x and NO_x that is more than 20% less costly than limestone scrubbing of SO_2 and selective catalytic reduction of NO_x, removes at least 95% of the SO_x and 75% of the NO_x, and produces salable byproducts with no waste material. To accomplish this objective, we intend to develop a regenerable wet scrubbing process for SO_x and NO_x that exploits the advantages of a class of novel contacting devices, known as hollow fiber contactors (HFC), and that uses novel SO_x and NO_x absorption chemistry. The HFC devices will bring substantial capital cost savings to our process. The novel use of the contactors for regenerating the SO_2 scrubbing liquor and the novel NO_x absorption chemistry will help reduce the energy consumption.

This development program will include three phases conducted over 60 months. The initial phase, encompassing the first 24 months of the project, will emphasize demonstrating the technical feasibility of each component of the flue gas control process—including process chemistry, mass transfer rates, and the ability of the HFC modules to function in the presence of particulate matter. At the end of this phase, we expect to have demonstrated 99% SO_2 removal efficiency and 85% NO_x removal efficiency in the laboratory. These efficiency targets are greater than the final goals of this project. The second phase of the work is designed to develop scale-up rules for the HFC modules and to test the longevity of the NO_x scrubbing chemistry. In the 15 months of this second phase, we expect to demonstrate 97% SO_2 removal efficiency and 80% NO_x removal efficiency with scalable modules and to show that our NO_x chemistry is

industrially robust. The third phase of the project will take place in the final 21 months wherein we will concentrate on building and operating a sub-scale proto-type system with a capacity of handling about 100 cubic feet per minute of flue gas. We expect to be able to demonstrate 95% SO_2 removal efficiency with 75% NO_x removal efficiency with the sub-scale prototype.

PROCESS DESCRIPTION

Our concept involves SO_2 scrubbing followed by NO_x scrubbing with regeneration of the individual scrubbing liquors (Figures 1 and 2). The desorbed, concentrated SO_2 and NO_x streams are fed to conventional sulfuric acid and NO_x reduction devices that are not part of the proposed research but which we include for costing estimates as part of the overall process. Both absorption and desorption vessels are based on HFC technology. The basic features of an HFC are that it allows phases to <u>contact</u> without <u>mixing</u> and that there is a large amount of interfacial contact area per unit volume of vessel (Qi and Cussler, 1985). The fiber material (e.g., polypropylene) is hydrophobic, and the fiber walls have small pores (e.g., 0.05 μm) so that gas/liquid contact surface area is created on the liquid side of the pores. Because the walls of the fibers are thin (e.g., 25 μm), there is minimal mass transfer resistance added by the wall of the fiber (diffusion time through the stagnant pore gas is about 0.1 milliseconds). So long as the aqueous phase pressure exceeds that of the gas phase, the interface is stable. The aqueous phase pressure must, however, be kept less than the breakthrough pressure of about 150 psi. Commercially available hollow fibers have sufficiently high mechanical strength to withstand more than 100 psig external pressure and 60 psig internal pressure (Bhave and Sirkar, 197; Majumdar et al., 1988). The fiber wall eliminates entrainment of liquid in the gas and allows independent control of the liquid and gas flow rates. However, the key element of this technology is that the surface area per unit vessel volume is 7 to 30 times higher than that of a traditional packed tower contactor. Consequently, the vessel volume for a given amount of mass transfer is substantially decreased, leading to reduced cost of the scrubber vessel.

Scrubbing SO_2 from flue gases with HFC devices is not new (e.g., Ogundiran et al., 1989; Sengupta et al., 1990). Variants of the HFC technology can, however, be used very efficiently for regenerating these scrubber liquors, and it is this aspect that constitutes part of the novelty of our approach.[1] For regenerating the SO_2 liquor, we intend to contact the liquor with an organic solvent in a hollow fiber contacting device. The organic solvent must have high affinity for SO_2 and very little solubility in water (e.g., dimethyl-aniline). The rationale for using organic extractants is that these have approximately half the heat capacity of water so that the SO_2 can be heat stripped at lower costs (Tung and Kuhr, 1989). These organic liquids cannot be used for direct scrubbing of flue gases because of unacceptably large contamination of the flue gas by the organic solvents.

HFC devices are used so the organic extraction can be done without mixing—so there are no emulsions formed and no phase separation steps required. Further, the high specific contact area results in an extraction vessel much smaller than conventional ones. There are two options for conducting this extraction. In Option A, the SO_2-rich scrubber liquor is passed in the lumen of the HFC, and organic solvent flows on the shell. Sulfur dioxide is preferentially extracted by the organic liquid. The organic liquid is taken to a separate vessel and heated to desorb the SO_2. In Option B, the hollow fibers are divided into two bundles. The SO_2-rich liquor passes down the lumen of one of the bundles, and organic solvent occupies the shell space around the hollow fibers. In the other bundle of hollow fibers, a low pressure is maintained with a small flow of water vapor to draw and sweep the SO_2 through the organic liquid (Figure 3). In this option, no further vessel is required for desorbing the SO_2 from the organic solvent. This particular form of a hollow fiber contactor is known as a hollow fiber contained liquid membrane, (HFCLM; Majumdar, et al., 1988; Sengupta, et al., 1988).

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[1] For the NO_x stripper, however, there is little incentive to use anything other than a conventional stream stripping device. Consequently, we will use conventional technology to regenerate the NO_x liquor.

PROCESS CHEMISTRY

SO$_2$ Scrubbing. Substantial information exists on the advantages and disadvantages of various chemistries for both SO$_x$ and NO$_x$ absorption (Harkness et al., 1986; Jethani et al., 1990; Oliver and Greenaway, 1989; Roberts and Friedlander, 1980; Oliver, 1989). For wet regenerable SO$_2$ scrubbing processes, all of the chemistries are essentially different means of providing alkalinity to the solution to induce a high solubility for SO$_2$ (in the form of HSO$_3^-$ and/or SO$_3^=$. Typical absorbent solutions consist of water with dissolved sodium sulfite (Wellman-Lord process), magnesia, ammonia, sodium sulfide, sodium citrate, soda ash, potassium carbonate, or potassium citrate (Pfizer Sultrol process). We intend to study several of these chemistries and pure water for SO$_2$ scrubbing.

Whereas these chemistries are well understood, two novel features of the HFC absorber can be exploited to make these chemistries more cost effective than they can be in traditional contactors. First, there are no losses by entrainment of liquor in the flue gas in an HFC device. Second, the liquid/gas flow rate ratio can be much higher in an HFC absorber than in a conventional absorber because the gas and liquid flow rates are independent of each other. Consequently, the concentration of total dissolved alkalinity can be kept lower and still remove the required fraction of SO$_2$ from the flue gas. Reduced dissolved alkalinity will result in increased lifetime of the SO$_2$ scrubbing compounds.

NO$_x$ Scrubbing. For NO$_x$ scrubbing, selection of suitable chemistry has been historically one of the most difficult problems. Many unsuccessful attempts have been made to design an ideal scrubbing chemistry. Based on developments in the 1970s, a number of commercial processes have used ferrous chelates based on ethylene-diaminetetraacetic acid (EDTA). Nitric oxide forms a coordination complex with Fe^{2+}-EDTA, and the reaction rates are very fast. Unfortunately, however, as a regenerable process, EDTA chemistry has been proven to be quite expensive because the Fe^{2+} species is oxidized easily to the ineffective Fe^{3+} species. Also a large number of unwanted chemical reactions take place in the scrubber liquor, creating the need for very costly and capital-intensive treatment steps of the scrubbing liquor.

We intend to focus instead on novel water soluble phthalocyanine compounds, developed by SRI, and novel water soluble polymeric derivatives of EDTA, similar to those described by Bedell et al. (1988), both of which we will synthesize in our laboratories. We have already shown under limited flue gas conditions that these compounds bind NO and NO$_2$ reversibly with no interference from O$_2$, CO$_2$, or SO$_2$.

Finally, one of the potentially greatest advantages of our process (as it relates to process chemistry) is separate SO$_2$/NO$_x$ scrubbing liquors. This separation will reduce substantially the wide variety of complex side reactions that lead to the need for convoluted regeneration schemes or expensive blowdown in other processes (e.g., Liu et al., 1988).

Extraction of SO$_2$ from Spent Scrubbing Liquor. The third aspect of the process chemistry is an organic solvent for extracting SO$_2$ from spent scrubbing liquor. Several suitable solvents are available with a high SO$_2$ affinity and no propensity to form solid adducts with SO$_2$: dimethylaniline (DMA), quinoline, and various ethers such as diethylene glycol dimethyl ether (diglyme), triethylene glycol dimethyl ether (triglyme), and tetraethylene glycol dimethyl ether (tetraglyme; Demyanovich and Lynn, 1987). Indeed, DMA and the glymes have been used for commercial absorption processes for SO$_2$ and H$_2$S (Kohl and Riesenfeld, 1985). To limit our effort on this topic, we will restrict ourselves to DMA. To overcome small losses of DMA by volatilization, we will synthesize DMA oligomers.

RESULTS

To date we have worked on nine elements of the program: SO$_2$ scrubbing tests, synthesis of SO$_2$ and NO$_x$ extraction compounds, their capacity and reversibility tests, spent SO$_2$ liquor regeneration tests, NO scrubbing tests, integrated NO life tests, particle deposition, economic evaluation, and performance of scalable modules. Previous results on some of these elements are

presented elsewhere (Gottschlich et. al., 1993 and Bhown et al., 1994). More recent results are discussed next.

SO$_2$ Scrubbing. In our previous reports (Gottschlich et al., 1993 and Bhown, et al., 1994), we indicated that SO$_2$ removal rates more than 95% were obtained using 200 fiber, 23 cm long HFC with 0.2 M aqueous Na$_2$SO$_3$ solution. Therefore, we shall not discuss these results here.

SO$_2$ Absorbing Compounds. Bhown, et al., 1994, presented several compounds synthesized by SRI and their performance toward SO$_2$ liquor generation. Amongst these compounds d-DMA was reported to be the leading candidate. Figure 4 manifests the performance of d-DMA under repeated exposure to SO$_2$ at different temperatures. At 50°C, excepting Runs 5 and 6, the results given in the figure show that d-DMA sustains and performs well over repeated exposures to SO$_2$. Therefore, we use d-DMA in all our further studies.

SO$_2$ Spent-Liquor Regeneration. We reported earlier that spent 0.2 M Na$_2$SO$_3$ solution was successfully (>98%) regenerated using DMA in a separate 1000 fiber HFC. Therefore, we shall not discuss these results here.

NO Absorbing Compounds. Earlier we reported several phthalocyanine compounds synthesized by SRI and their ability to absorb NO. Amongst these compounds, 3-ph exhibited superior performance and also showed no oxidation in the presence of O$_2$. The life tests on this compound are presented below under separate task.

NO Scrubbing. NO removal rates more than 85% were obtained using 3-ph solution in a 300 fiber HFC. From the results obtained it was shown that mass transfer is a strong function of gas flowrate and also depends on reaction kinetics at low temperatures. Thus our absorption tests are conducted at temperatures close to 50°C.

Particle Deposition. We generated 0.2 μm silica particles (typical of fly ash material) into gas streams using pneumatic nebulizers. The particles were shown (Figure 5) to agglomerate when dispersed into gas streams from liquid. The lumens of a 1155 fiber HFC were exposed to a gas stream (11.67 L/min) containing 30 mg/m^3 of particles for a period of 80 hours. The pressure drop across the length of the fibers was observed to increase gradually at the beginning of deposition and rapidly afterwards reaching about 100" water in 30 hours (Figure 6). Figure 7 shows the effect of the particle deposition on the mass transfer coefficient (MTC) during the entire operation. We speculate that the overall drop of about 20% in MTC is due to accelerated condition used in the tests as well as difficult nature of experiments.

Integrated NO Life Tests. In our previous report (Bhown et al., 1994), we demonstrated that one of the phthalocyanine compounds (3-ph) exhibited superior mass transfer characteristics for absorbing NO. Therefore, in order to further demonstrate the absorption/desorption behavior of 3-ph, we have carried-out 30 hours worth of integrated absorption and desorption experiments. Figure 8 gives the absorption and desorption results. In these tests, simulated flue gas contained 500 ppm NO, 4.5% O$_2$, and balance N$_2$. The approximate flat nature of both absorption and desorption curves indicate that 3-ph does not undergo any deactivation or deterioration under repeated exposure to NO. Longer exposure tests are currently being conducted.

Economic Evaluation. Based on our laboratory (Phase I of the program) test results, we estimated the costs of major battery limit equipment and associated utility costs. The design basis for the calculations are given in Table 1 and the comparison of the costs of the proposed process with that of the traditional processes are presented in Table 2. The capital costs are low by a factor of two compared to that of widely employed wet limestone and SCR technology. In addition, the processing costs of the present process is also less compared to that of existing technologies.

Performance of Scalable Modules. In this Phase II of the project, in order to readily scaleup the process to commercial level, we intend to use rectangular HFCs as shown in Figure 9. Our preliminary design estimates indicate that the modules can effectively treat gas flowrates up to 200 SLM. We will present the mass transfer results in the meeting.

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Table 1. BASIS FOR EXAMPLE DESIGN OF 500 MW (e) SO_x/NO_x PLANT

Flue gas flowrate	1 million actual ft^3/min
	2.25×10^{10} sccm
Flue gas temperature	160°F
Flue gas pressure	760 mm Hg
Flue gas composition	
SO_x	3000 ppm(v)
NO_x	450 ppm
CO_2	14%
H_2O	8%
O_2	3.3%
N_2	74.4%
Fraction of SO_x/NO_x removed in absorbers	
SO_x	98%
NO_x	85%
Pressure drop	
SO_x scrubber (flange to flange)	10" H_2O
NO_x scrubber (flange to flange)	10" H_2O

Table 2. COMPARISON OF HF PROCESS WITH TRADITIONAL PROCESSES

	Capital Cost ($/kW)	Operating Costs[1] (mils/kWH)	Processing Cost[2] (mils/kWH)
(A) Proposed Option A			
SO_2/NO_x scrubbing/regenerating	$60.50	7.76	9.16
Small scale H_2SO_4 plant[3]	10.40	0.49	0.75
Small scale SCR NO_x reduction plant	7.90	0.55	0.73
Total Option A	$78.80	8.8	10.64
(B) Traditional Process			
(1) Wet Limestone plus SCR[4]	$185.50	7.33	12.14
(2) Wellman Lord plus SCR	$333.00	10.45	16.18
(3) Citrate plus SCR	$303.00	8.99	14.26

[1] Operating costs include utilities, raw materials and labor and other administrative overhead.
[2] Includes amortized capital costs at 20% year plus operating costs.
[3] Source: Elkin, 1985.
[4] Selective catalytic reduction capital cost is $70.80/kw and operating cost is 2.02 mils/kwh in each traditional process.
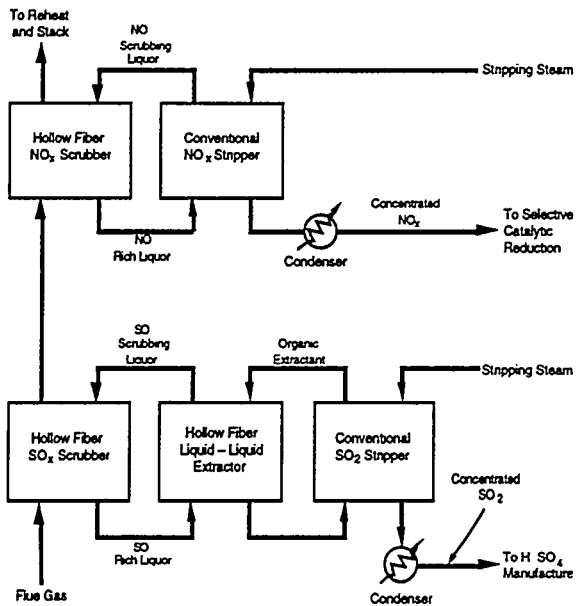
Figure 1. Proposed SO$_x$/NO$_x$ wet scrubbing with hollow fiber contacting devices. (Option A for SO$_2$ liquor regeneration)

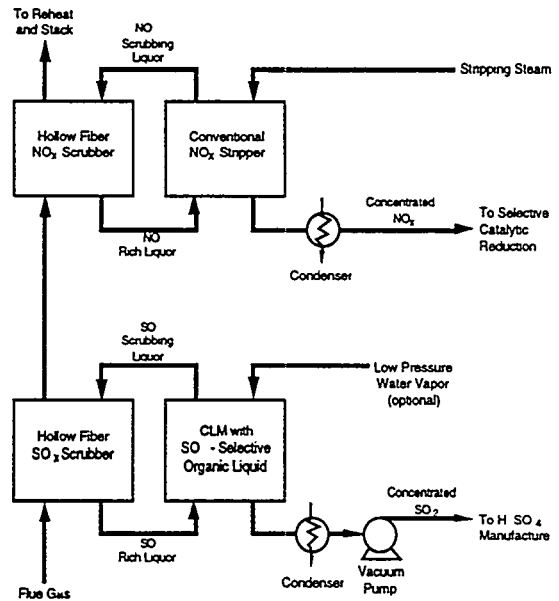SO$_2$ liquor regeneration occurs in hollow fiber liquid – liquid extraction (LLE) device.

Figure 2. Proposed SO$_x$/NO$_x$ wet scrubbing with hollow fiber contacting devices. (Option B for SO$_2$ liquor regeneration)

SO$_2$ liquor regeneration occurs in hollow fiber contained liquid membrane (CLM) device. Condenser/vacuum pump system combine to give driving force for operation of CLM.
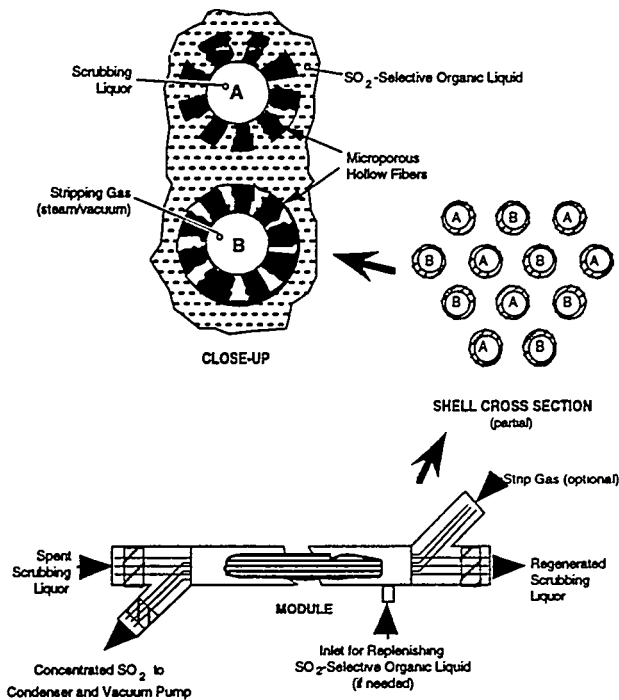
Figure 3. Hollow fiber contained liquid membrane for regenerating SO$_2$ scrubber liquor.
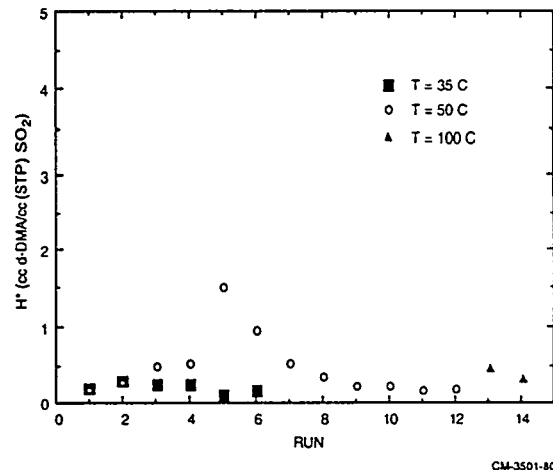
Figure 4. Lifetime tests on d-DMA for SO$_2$ absorption/desorption.

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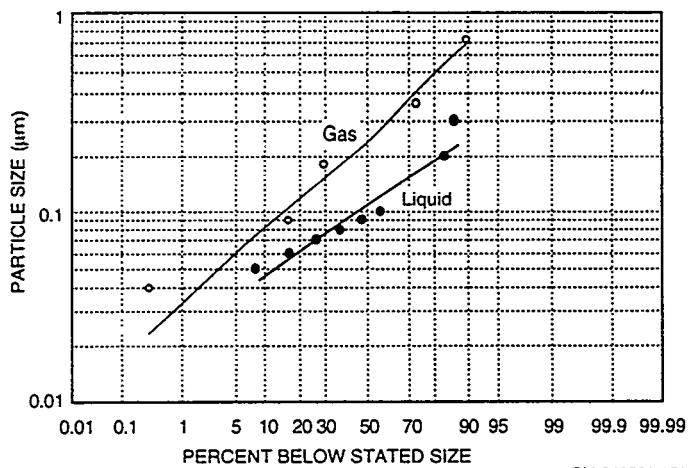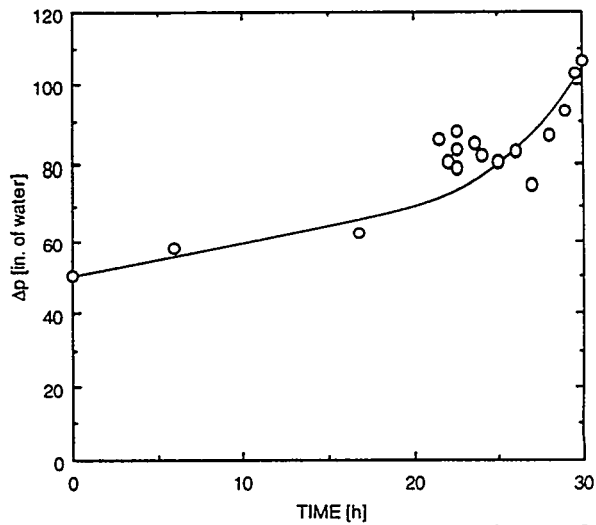
Figure 5. Particle size distribution in gas and liquid phases.

Figure 6. Pressure drop across HFC as a function of deposition time.
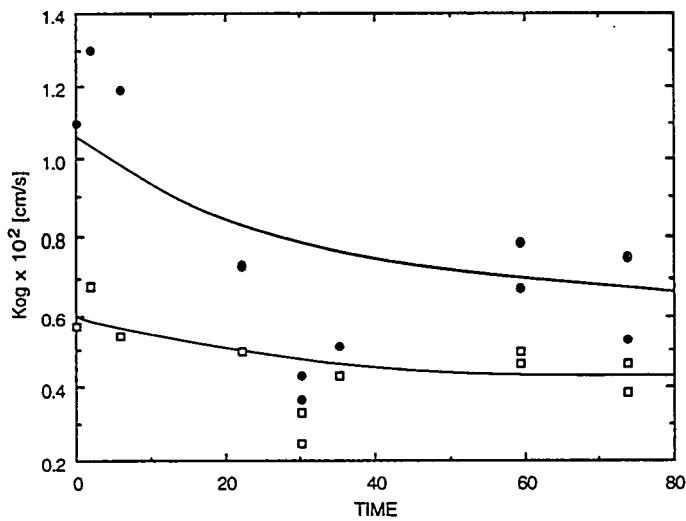

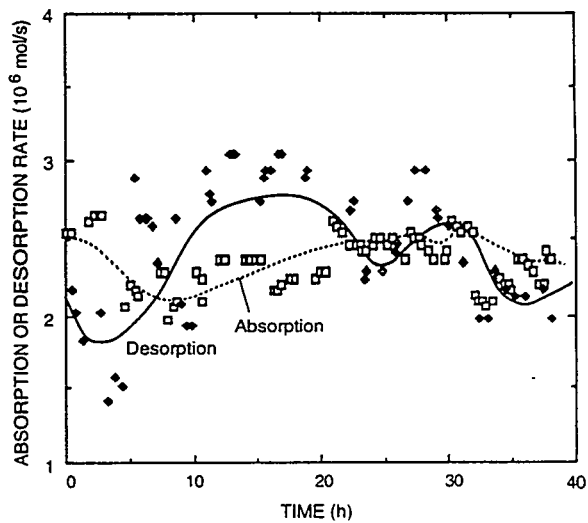Figure 7. 1155-fiber HFC mass transfer coefficient with particle deposition.

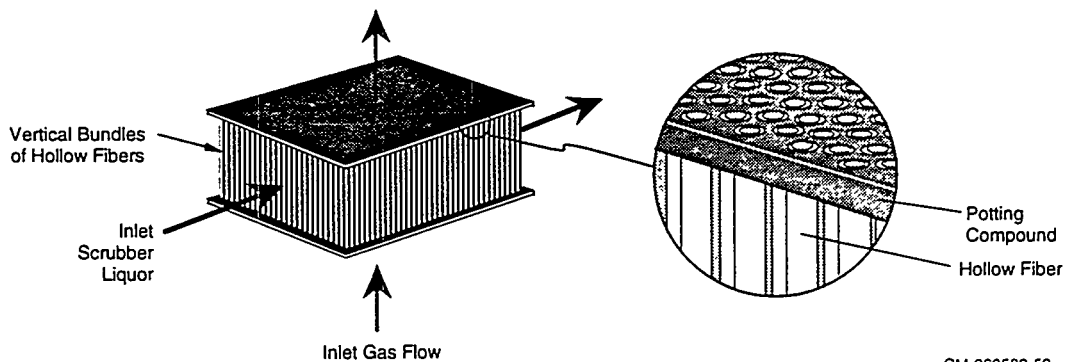Figure 8. Simultaneous NOx absorption and desorption tests using 3-pH solution.

Figure 9. Cross-flow pattern of gas and liquid flow in a rectangular hollow fiber cassette (module).

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DEVELOPMENT OF THE ADVANCED COOLSIDE SORBENT INJECTION PROCESS FOR SO_2 CONTROL

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ABSTRACT

The goal of this work was to develop a low-capital-cost process capable of over 90% SO_2 removal as an economically attractive option for compliance with the Clean Air Act. The Advanced Coolside Process uses a contactor to simultaneously remove fly ash and saturate the flue gas with water, followed by sorbent injection into the highly humid flue gas and collection of the sorbent by the existing particulate collector. High sorbent utilization is achieved by sorbent recycle. The original performance targets of 90% SO_2 removal and 60% sorbent utilization were exceeded in 1000 acfm pilot plant operations using commercial hydrated lime as the only sorbent. Process optimization simplified the process equipment, resulting in significant cost reduction. Recent accomplishments include completion of equipment testing and sorbent optimization, a waste management study, and a long-term performance test. An economic evaluation for the optimized process projects capital costs 55% to 60% less than those of limestone forced oxidation wet FGD. The projected levelized control cost is 15% to 35% lower than wet FGD (25% lower for a 260 MWe plant burning a 2.5% sulfur coal), depending on plant size and coal sulfur content.

BACKGROUND AND INTRODUCTION

In-duct dry sorbent injection technology has been actively developed in the U.S. since the early 1980s. The performance of these processes was established through the development of the Coolside process[1-3] and the HALT process[4] and through the DOE duct injection technology development program.[5] These development efforts included pilot-scale tests, proof-of-concept tests, and a full-scale utility demonstration. Demonstrated performance was in the range of 40-50% SO_2 removal at 2.0 Ca/S molar ratio. Additionally, the 105 MWe demonstration of the Coolside process at the Ohio Edison Edgewater Station[3] showed that an SO_2 removal of 70% can be attained by improving calcium hydroxide sorbent activity with sodium-based additive injection at a 0.2 Na/Ca molar ratio (~32% sorbent utilization).

Process performance data and economic analyses support the attractiveness of duct sorbent injection for a range of retrofit applications.[6] However, the applicability as a compliance option for the Clean Air Act or other regulations can be expanded by increasing SO_2 removals and sorbent utilizations. Higher SO_2 removals became more important for retrofit technologies with the passage of the 1990 Clean Air Act Amendments. Higher sorbent utilization reduces operating cost.

The objectives of the work reported here were to improve the applicability of in-duct sorbent injection technology as a compliance option for the 1990 Clean Air

Act Amendments and to reduce total SO_2 control costs. Specific desulfurization performance targets were to achieve 90% SO_2 removal and 60% sorbent utilization, while retaining the low capital cost and retrofit advantages inherent to in-duct sorbent injection technology. These targets represent a substantial improvement over existing sorbent injection technologies.

Initial pilot plant tests indicated that a process concept, referred to as the Advanced Coolside process, had the potential to achieve the process performance targets stated above.[7] This process is described in the next section. Pilot plant development of the Advanced Coolside process focused on the following areas:

- Optimization of sorbent recycle to improve sorbent utilization efficiency and SO_2 removal capability.
- Optimization of process equipment to reduce capital and operating costs.
- Optimization of sorbent systems for improved performance and reduced cost.
- Evaluation of process operability issues.
- Evaluation of solid waste disposal as well as possible solid by-product utilization.

Process conceptual design and economic evaluation was an ongoing consideration during the development program. This allowed research and development to focus on approaches with the most potential for improving the process design and the process economics.

DESCRIPTION OF ADVANCED COOLSIDE PROCESS

Figure 1 shows a schematic of the Advanced Coolside process. The process achieves greater SO_2 removal and sorbent utilization than previous duct sorbent injection processes because it is operated at a higher flue gas humidity, and more fully exploits the potential of sorbent recycle. The key to the process is a gas/liquid contacting device downstream of the air preheater. The contactor serves two purposes: to nearly saturate the flue gas with water and to remove most of the coal fly ash from the flue gas. The sorbent is injected downstream of the contactor into the highly humid flue gas. Hydrated lime is very active for SO_2 capture near the saturation point, even in the absence of liquid water droplets. Because the flue gas is already humidified prior to sorbent injection, there is no strict residence time requirement for droplet evaporation, although residence time affects SO_2 removal as discussed below. SO_2 is removed by the sorbent in the duct and by that collected in the existing electrostatic precipitator (ESP) or baghouse. The heat of reaction between SO_2 and hydrated lime raises the temperature of the flue gas by roughly 8-10 °F for each 1000 ppm of SO_2 removed. Therefore, the particulate collector can be operated at an increased approach to saturation. However, because hydrated lime activity is highly sensitive to the approach to saturation, this reaction heat effect also acts as a limiting mechanism for SO_2 capture.

The spent sorbent is captured by the existing particulate collector as a dry powder. Sorbent recycle is an integral component of the Advanced Coolside process, allowing the sorbent utilization target of 60% to be exceeded. The potential for recycle is increased because fly ash is removed separately before sorbent injection. Furthermore, process performance can be improved by adding small amounts of water to the recycle sorbent prior to re-injection. The water

acts to maintain a close approach to saturation by evaporating, counteracting the heat of reaction. The moisture addition step is a key to maintaining sorbent activity and, thus, to exceeding the SO_2 removal target of 90%.

Equipment design optimization was focused on the flue gas/water contactor. For the initial pilot plant tests the contactor was a relatively complex device.[7] In the process design optimization program, second and third generation contactors were designed, tested, and optimized. The improved contactor designs significantly reduce capital and operating costs. The third generation design consists of a low-pressure-drop, in-duct venturi followed by a cyclonic separator.

EXPERIMENTAL

This process development program was carried out in a 1000 acfm (ca. 0.3 MWe equivalent) pilot plant. Figure 2 is a schematic of the Advanced Coolside desulfurization pilot plant. It was designed to simulate integrated Advanced Coolside operation, including combined flue gas saturation and fly ash removal by a contactor, sorbent injection downstream of the contactor into the saturated flue gas, and steady-state continuous sorbent recycle with wetting of the recycle sorbent. The pilot plant, operating procedures, and analytical procedures are described in detail elsewhere.[7-9]

Test sorbents were analyzed using a Micromeritics Digisorb 2600 nitrogen adsorption apparatus (for surface area and pore size distributions), a Malvern Instruments 2600C EASY Particle Sizer M5.4, a LECO CHN-1000 analyzer, and a Perkin Elmer 7-Series Thermogravimetric Analyzer.

The equipment for the waste/by-product characterization study includes a laboratory mixer, a mechanical soil compactor and a unconfined compression tester. TCLP and ASTM leaching tests were conducted as specified in the standard methods. The moisture-density relationships and unconfined compressive strengths of Advanced Coolside wastes were determined with laboratory equipment complying with ASTM Standards. Waste samples were pelletized using a Littleford Brothers LM-130 batch mixer, a 36" I.D. rotary disc pelletizer and a 55-gal curing drum. Pellet properties were determined using a sieve shaker and an LA abrasion machine according to ASTM Standards.

DISCUSSION

Recycle Optimization

Process Performance Goals Exceeded. The recycle optimization tests showed that the process performance targets of 90% SO_2 removal and 60% sorbent utilization could be exceeded. The 90% SO_2 removal target was achieved at sorbent utilizations over 70%. At conditions simulating the use of a baghouse for particulate collection, very high SO_2 removal (>99%) was achieved while maintaining at least 60% sorbent utilization. Sorbent recycle was a key to achieving these levels of performance.

The tests conducted to simulate SO_2 removal in a utility boiler unit equipped with an ESP achieved 90% SO_2 removal at sorbent utilizations of up to about 75% (Table 1). These tests were conducted with frequent baghouse pulse cleaning and

with hot air reheat to partially quench the SO_2 removal reaction in the baghouse. Based on mass transfer calculations, ESP removal may be limited to about 30% of the SO_2 in the flue gas at the ESP inlet.

The tests conducted without reheat and less frequent baghouse pulsing to simulate SO_2 removal in a plant with a baghouse showed very high efficiency SO_2 removal (90 to >99%) while maintaining the target of 60% sorbent utilization (Table 2).

Effect Of Fresh Ca/S Ratio and Recycle Ratio. Increasing the fresh Ca/S ratio and/or the recycle ratio (lb recycle sorbent/lb fresh hydrated lime) increases the amount of calcium available for reaction with SO_2. By maintaining a sufficiently high concentration of available calcium in the sorbent, the process target of 90% SO_2 removal can be achieved or exceeded. For example, at a 10 °F approach to saturation in the baghouse and with water addition to the recycle at a ratio of 0.15 lb H_2O/lb recycle, increasing the total available Ca/S ratio from 1.8 to 2.2 increased the in-duct SO_2 removal from 60% to 70% and the system SO_2 removal from 84% to 90% (Table 2). In both cases of this example, 70% of the available calcium came from the fresh hydrated lime; the remainder came from the recycle sorbent.

Effect of In-duct Residence Time. High SO_2 removals and sorbent utilizations were achieved with 1.7 to 2.7 sec in-duct residence time. Below 1.7 sec residence time, SO_2 removals were significantly lower. Between 1.7 and 2.7 sec there was little effect of additional residence time on in-duct SO_2 removal.

Moisture Addition to Recycle. Moisture acts primarily to maintain a close approach to saturation by counteracting the reaction heat effect. Moisture also provides surface water to the sorbent particle which can enhance gas/solid reactions. The addition of moisture to the recycle sorbent had a strong positive effect on desulfurization performance of the sorbent. For example, at a 1.2 fresh Ca/S mol ratio, 5.0 lb/lb recycle ratio and a 10 °F approach in the baghouse, the addition of 0.15 lb H_2O/lb of recycle sorbent increased the in-duct SO_2 removal from 59% to 81% and the system removal from 73% to 88%. The sorbent utilization increased from 61% with no moisture addition to 71% with moisture addition.

The optimum moisture addition level in the pilot plant tests was between 0.10 and 0.15 lb water/lb recycle sorbent. Increasing moisture addition level to 0.20 lb/lb resulted in a decrease in performance because of sorbent agglomeration. However, the optimum water addition level determined in pilot tests does not apply directly to large-scale operation because the ratio of transport air to sorbent is about an order of magnitude greater in the pilot plant than a typical large-scale transport system. Consequently, in the pilot plant more water is required on the sorbent to allow for the evaporation into the dry transport air. Based on heat and mass balance calculations the moisture addition level would be about 0.05 lb H_2O/lb solid for a 250 MW plant burning a 2.5% sulfur coal, which is less than half that used in the pilot plant tests.

Equipment Design Optimization

Equipment design optimization was a key to reducing the cost of the Advanced Coolside process. One focus of design optimization was the contactor. A third generation contactor was designed that is mechanically simpler than the original[7]

and second generation[8-9] designs. Design optimization also was focused on the sorbent recycle equipment.

<u>Contactor Design Improvements</u>. The third generation contactor (Figure 3) was designed to reduce capital cost and a reduced plant footprint. It consists of a low-pressure-drop, in-duct venturi followed by cyclonic separator; the total design Δp was 5" W.C. Water is sprayed by hydraulic nozzles in the throat of the venturi, which reduces water droplet size and provides turbulent contact between droplets and flue gas for efficient particle capture and humidification. The water/fly ash mix is separated from the flue gas by the downstream separator.

The pilot plant venturi contactor was an off-the-shelf design with minor modifications. The contact time between the venturi throat and the cyclonic separator was about 0.1 sec. This contactor allowed reasonably close approaches (ca. 0 to 4 °F) to be achieved.

The fly ash collection efficiency of the venturi contactor was greater than 90% in pilot plant tests conducted with EPA Method 17 sampling. Fly ash collection efficiency was independent of gas flow over a range of 380 to 1025 acfm. The results indicate that a single venturi contactor can handle the range of turndown required for a commercial application to follow changing boiler load.

Operability of the third generation contactor was good throughout the performance tests. There were no problems with fly ash accumulation in the venturi, on the spray nozzles or in the cyclonic separator.

<u>Optimization Of Recycle Sorbent Treatment Equipment</u>. A test was conducted in which the recycle sorbent was wetted using a pilot-scale, continuous pugmill. Performance of the pugmill was compared to the high intensity mixer used in previous pilot plant tests. The results indicate that a pugmill can produce a satisfactory product, from both materials handling and reactivity standpoints. These results are encouraging because a pugmill has substantially lower capital and operating costs than the high intensity mixer.

<u>Other Design Optimization</u>. In addition to the pilot plant optimization testing discussed above, engineering studies were conducted to explore process improvements in all major process subsystems. Key areas identified for process improvement/cost reduction include:

- Use of hydrocyclones instead of a thickener to concentrate the fly ash slurry before mixing with spent sorbent.
- Use of on-site lime hydration of quicklime for larger plants.
- Simplification of the flue gas reheat system.
- Improvements in the recycle handling system design.
- Simplification of the ductwork conceptual design.

Sorbent Optimization

The Sorbent Optimization program included pilot plant evaluation of different sorbents including several commercial hydrated limes and specially prepared high surface area limes, an evaluation of hydration variable effects in a pilot hydrator, and testing of additive promotion. The results showed that process performance is relatively insensitive to hydrated lime source, unlike the

conventional Coolside process. Small amounts of additives incorporated during the recycle wetting step incrementally improved desulfurization in the baghouse, but were not necessary to exceed the performance targets.

Pilot plant tests were conducted on five different commercial hydrated limes. The limes tested were from different geographic areas and were selected from among the largest hydration plants in the country. The BET surface areas of the commercial hydrates tested ranged from 14 to 24 m^2/g. Three specially-prepared, high surface area hydrated limes were tested in the pilot plant; surface areas ranged from 35 m^2/g to 41 m^2/g. The desulfurization results showed only a small variation among the limes tested. Thus, a variety of commercial hydrated limes can be used to achieve the process performance goals of 90% SO_2 removal and 60% sorbent utilization. The relative insensitivity of the process to hydrated lime source is advantageous, allowing the use of the lowest cost available hydrated lime.

A statistically designed test program was conducted to determine the effect of hydration variables on hydrated lime properties and the effects of these properties on desulfurization performance. This program was conducted by Dravo Lime Company using a 100 lb/hr pilot hydration test facility. Data analysis indicates that there was no significant correlation of desulfurization activity with sorbent physical properties or with quicklime source.

Recycle tests were conducted with small amounts of additives incorporated in the combined recycle and fresh sorbent during the moisture addition step. A moderate enhancing effect of small amounts (ca. 0.03 mol/mol fresh Ca) of inorganic chloride compounds (NaCl, $CaCl_2$) on sorbent performance was observed in the baghouse but not in the duct. Therefore, use of small amounts of these additives may be an attractive means of achieving high SO_2 removal efficiencies (90 to >99%) in a plant with a baghouse. Additive incorporation in the recycle pretreatment step is attractive because it uses existing equipment and commercially available hydrated lime.

Once-through tests were conducted with a finely pulverized limestone. Although limestone is not a sufficiently active sorbent for commercial use in the Advanced Coolside process, the results indicated that $CaCO_3$ does have significant desulfurization activity (23% removal in duct and 31% system removal at 2.0 Ca/S mol ratio). This may be a significant observation, since some $Ca(OH)_2$ is converted to $CaCO_3$ in the Advanced Coolside process. The activity difference may be largely a result of the lower surface area of limestone (1.6 m^2/g) compared to that of hydrated lime.

Process Performance Tests

The objective of the performance testing was to generate performance and operability data for design and scale-up of the process. The performance test consisted of about one week of operation with two shifts per day followed by three separate weeks of 24 hour/day operation. The total on-stream time was 295 hours. The purpose of the initial week of testing was to establish near steady-state operating conditions and sorbent composition. The purpose of the around-the-clock operation was to evaluate performance and operability issues during longer periods of continuous operation. Although the test was divided into three periods of 24 hour/day operation, the same sorbent material was used; that is,

the baghouse material collected at the end of one period was used as the recycle material at the beginning of the subsequent test period.

The test conditions for the performance test were selected based on the results of the process optimization tests. The Ca/S ratio was in the range of 1.2 to 1.3 for the test. The recycle ratio was 7 lb/lb fresh lime and the recycle water addition level was about 0.12 lb/lb recycle. The third generation contactor (venturi + centrifugal separator) was employed for all the testing; it was operated to achieve near saturation conditions. The SO_2 removal averaged about 90% during the performance test.

Although the scale of the 1000 acfm pilot plant is not sufficient to completely resolve process operability issues, the performance test provided a positive indication of the operability of the Advanced Coolside process. The key operability issues evaluated were operation of the flue gas duct with sorbent injection at high humidity and operation of the recycle sorbent wetting, handling and transport systems. These and other issues should be further evaluated in larger scale, longer term tests.

There were no major operating problems in the flue gas duct with injection of wetted recycle sorbent at high humidity. The pilot plant had a duct configuration with numerous changes in flue gas direction, presenting more potential for operating problems than typical commercial systems. Because soot blowers are included in the conceptual process design, the duct was periodically air lanced with 50 to 80 psig air to simulate soot blowing. The soot blowing was effective in preventing accumulation of solids in the pilot plant duct. The material which adhered to the duct walls was generally soft and easily removed and carried to the baghouse by the soot blowing. The soot blowing was used primarily at elbows and near the sorbent injection point. The amount of accumulation in straight duct runs was small and tended to level off with time even without soot blowing.

For most of the performance test, the recycle sorbent was effectively wetted, fed, and transported to the flue gas stream. Recycle handling did, however, require frequent operator attention, although much of this attention was specific to the small scale and the specific equipment employed in the pilot plant. There were instances of eductor plugging, a fairly common problem in small-scale systems, because the orifice in the eductor venturi is quite small. It is anticipated that this would not be a significant problem with properly designed commercial scale pneumatic transport equipment.

After longer periods of continuous operation with high sorbent utilization, the wetted recycle material tended to agglomerate more. This increased the difficulty of feeding the material and somewhat reduced desulfurization performance. Agglomeration occurs when more water is added to the recycle sorbent than can be incorporated into the available sorbent pore volume. The pore volume of sorbent decreases at higher utilization levels. Although less sorbent agglomeration would be expected in a full-scale process (because less added water is needed, as discussed earlier), this is an important phenomenon to be addressed for process scale-up.

During the process performance tests, observed levels of CO_2 pickup were higher than observed during short term tests; this may have contributed to sorbent

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agglomeration because $CaCO_3$ formation reduces pore volume and, thus, moisture-carrying capacity.

Waste Management Evaluation

The initial objective of the waste management study was to develop the data needed for designing the waste handling and disposal systems for the process. The test program was expanded to include exploratory tests of by-product utilization options. This involved pelletization tests and preliminary evaluation for production of synthetic aggregate materials.

The Advanced Coolside process generates two waste streams: the dry spent sorbent from the particulate collector and the fly ash/water slurry collected in the contactor that is subsequently concentrated. The proposed concept for disposal or utilization is to mix the two streams, controlling the overall moisture content by proper design of the fly ash slurry concentration system.

Three Advanced Coolside waste samples were prepared for use in the waste management study to simulate waste produced from a boiler using feed coals with 7.5% ash and 3.5%, 2.5% and 1.5% sulfur. Advanced Coolside waste samples were characterized to ensure that adequate information is available on the physical and chemical nature of the waste for the design and construction of safe and stable landfills.

The maximum dry bulk density of Advanced Coolside waste ranged from 75 to 80 lb/ft^3 with increasing fly ash component in the waste. The moisture content which gave the maximum density (optimum moisture) was about 32% (dry basis). At optimum moisture and compacted to 95% of Proctor density, Advanced Coolside waste had unconfined compressive strength that is suitable for landfill disposal. The strength increased from 20 psi (uncured) to 100 psi or higher after 28 days of curing. As a point of reference for unconfined compressive strength values, bulldozers used in landfills exert pressures ranging from about 12 psi to about 19 psi.

Advanced Coolside waste leachates were prepared using both the TCLP and ASTM leaching procedures. The trace element (As, Ba, Cd, Cr, Pb, Hg, Se and Ag) concentrations were well below (by at least a factor of 50) RCRA allowable limits. Thus, the waste can be classified as non-hazardous for landfill disposal. In addition, the concentrations of Fe, Mn, Ca, Na, Al, sulfate, K and total dissolved solids (TDS) in the leachates were similar to those from other dry flue gas desulfurization (FGD) wastes.

Pelletization takes advantage of the cementitious properties of the Advanced Coolside waste to make products that may be applicable for use as synthetic aggregates. Pellets produced from Advanced Coolside waste were lightweight, had low bulk specific gravity, and had a desirably low LA abrasion index, low water absorption, and a coarse size distribution; however, they also had a high soundness index (i.e, low durability). These data indicate that pellets made from Advanced Coolside wastes may have potential for use as lightweight coarse aggregates in concrete masonry units. For this use, there is no soundness index specification. Since waste disposal can be a significant portion of the operating cost, a more thorough evaluation of other pellet characteristics for this application, along with an evaluation of potential economic impacts, is recommended.

Conceptual Process Design and Economic Evaluation

The primary objectives of this task were to develop a conceptual design for a utility-scale application of the Advanced Coolside Process and to assess the economic attractiveness of the process. An additional objective was to identify process areas for potential cost reductions to guide research efforts in areas that would most impact the economics.

Based on the results of an interim economic study, economic targets were established for the process midway through the project. These were to achieve a 20% levelized cost advantage and a 50% capital cost advantage over limestone wet scrubbing for a range of plant sizes and coal sulfur levels. Based on conversations with utilities, these levels of cost advantage would make it attractive to consider a less developed technology.

A final conceptual process design and economic study for the Advanced Coolside Process was performed. It describes a complete conceptual process design for full-scale, coal-fired applications of the process. Advanced Coolside process costs are compared to those of limestone forced oxidation (LSFO) wet FGD technology. The process economics were investigated for coal sulfur levels ranging from 1.0% to 3.5% (as-received) and plant sizes ranging from 160 to 512 gross MW. The final economic study incorporates the results of pilot plant process optimization work which has resulted in a significant reduction in process costs.

The Advanced Coolside process enjoys a capital and levelized cost advantage relative to LSFO in all coals and plant sizes examined in this study. Figures 4 and 5 show capital and levelized cost comparisons for a 2-5% S coal. The figures further indicate that the economic targets established in the first interim evaluation have been achieved for a wide range of coal sulfur contents and plant sizes. The projected capital cost of Advanced Coolside is 55% to 60% lower than limestone forced oxidation wet FGD. Total levelized SO_2 control cost in $/ton SO_2 removed ranged from 15% to 35% lower than LSFO, over the range of plant sizes and coal sulfur contents investigated. For a mid-range plant size (260 MW) and a mid-range coal sulfur content (2.5%), the levelized cost advantage is 25%. The levelized cost is sensitive to sorbent transportation charges and as a result is highly site-specific.

EPRI Technical Assessment Guidelines were followed. To make a fair comparative evaluation, similar design philosophies, equipment cost algorithms, and financial assumptions (Table 3) were used for the evaluation of both Advanced Coolside and limestone forced oxidation technologies. Both processes were evaluated for 90% SO_2 reduction. The process design for the limestone forced oxidation wet FGD process was recently updated based on current commercial trends to reflect the state of the art. This includes the use of a single absorber module with no spares.

CONCLUSIONS

1. The Advanced Coolside process achieved the process SO_2 removal target of 90% at sorbent utilization efficiencies of over 70%. The keys to achieving this performance were achieving near saturation in the contactor and optimizing sorbent recycle, including the moisture addition step.

2. The Advanced Coolside process has the potential for very high SO_2 removal efficiency (90% to greater than 99%).

3. The contactor was redesigned for significantly reduced capital and operating costs and smaller plant footprint compared to the initial design. Other process design improvements were realized, including improvements in the recycle wetting and handling systems, the sorbent preparation and handling system, the waste handling system, and the flue gas duct design.

4. The Advanced Coolside process is relatively insensitive to the lime source. This is an economic advantage, allowing the use of the lowest cost sorbent available.

5. The addition of small amounts of additives to the recycle sorbent during the water addition step can improve desulfurization performance in a baghouse, but this is not required to achieve performance targets.

6. Pilot plant operation provided a positive indication of the operability and retrofit potential of the Advanced Coolside process. Recycle test duration ranged up to 150 hours. A long-term (300 hr) performance test with 24 hr/day operation was conducted under optimized process conditions.

7. The waste management evaluation indicated that the combined spent sorbent/fly ash waste is suitable for landfill disposal.

8. A final process conceptual design and economic evaluation projects capital costs less than one half of those for limestone forced oxidation wet FGD. The projected total SO_2 control cost on a levelized basis is 25% lower than wet FGD for a 260 MWe plant burning a 2.5% sulfur coal. The levelized cost is sensitive to sorbent cost and, thus, is highly site-specific. This cost advantage meets previously established economic goals for increasing the attractiveness of the technology for electric utilities.

RECOMMENDATIONS FOR FUTURE DEVELOPMENT OF THE ADVANCED COOLSIDE PROCESS

- Demonstration of the process on at least the 5-10 MWe scale to confirm the scale up of process performance and operability.
- Further process development of by-product utilization could give the process unique advantages over conventional FGD technology.
- The capability of the process to control air toxics should be investigated. A literature analysis suggests that the Advanced Coolside process has important features for the control of mercury and HCl. Both the gas/liquid contactor and the sorbent entrainment zone provide low temperature and efficient mass transport conditions important for capture of these species. Furthermore, the relatively high recycle ratios employed could increase the feasibility of using a more expensive co-sorbent such as activated carbon.

ACKNOWLEDGMENT

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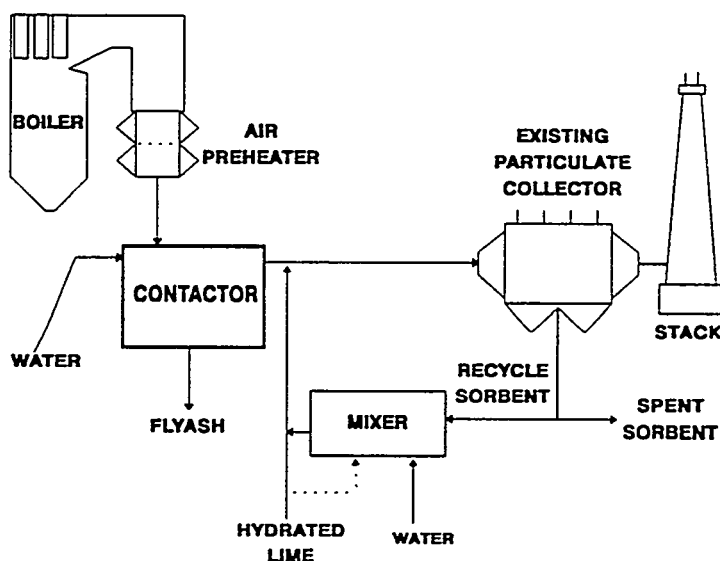
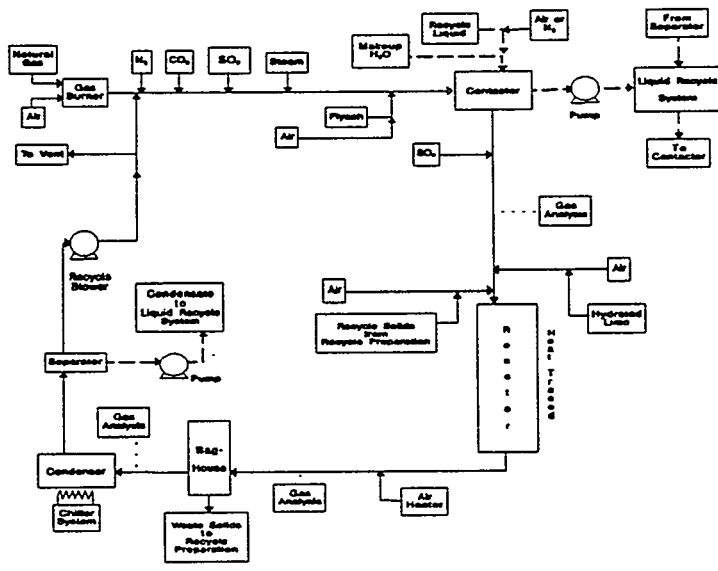Figure 1. Schematic of the Advanced Coolside Process.

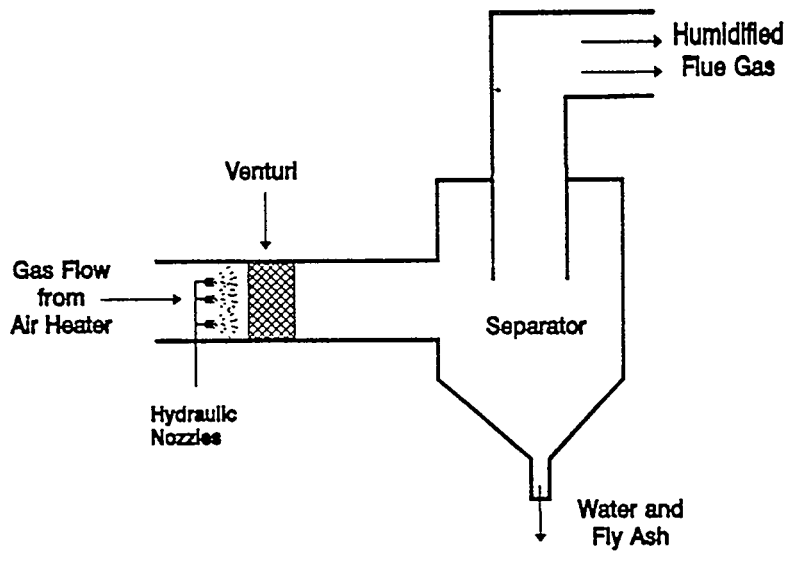Figure 2. Schematic of the 1000 ACFM Advanced Coolside Pilot Plant.

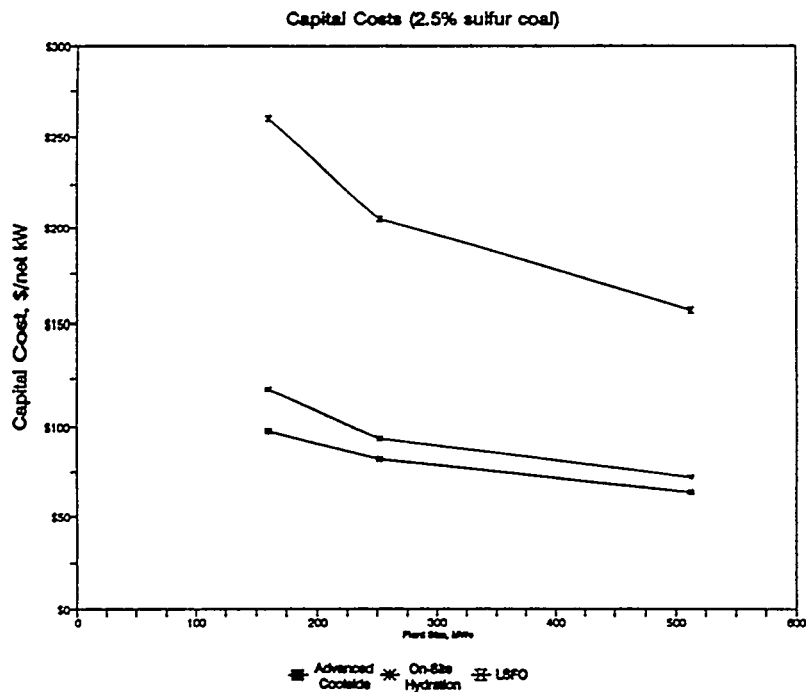Figure 3. Schematic of the Third Generation Contactor.

Capital Costs (2.5% sulfur coal)

Figure 4. Comparison of Capital Costs for the Advanced Coolside Process and Limestone Forced Oxidation Wet FGD (LSFO) for a Range of Plant Sizes Burning A 2.5% S Coal. (For Advanced Coolside, on-site hydration is compared to buying hydrated lime.)
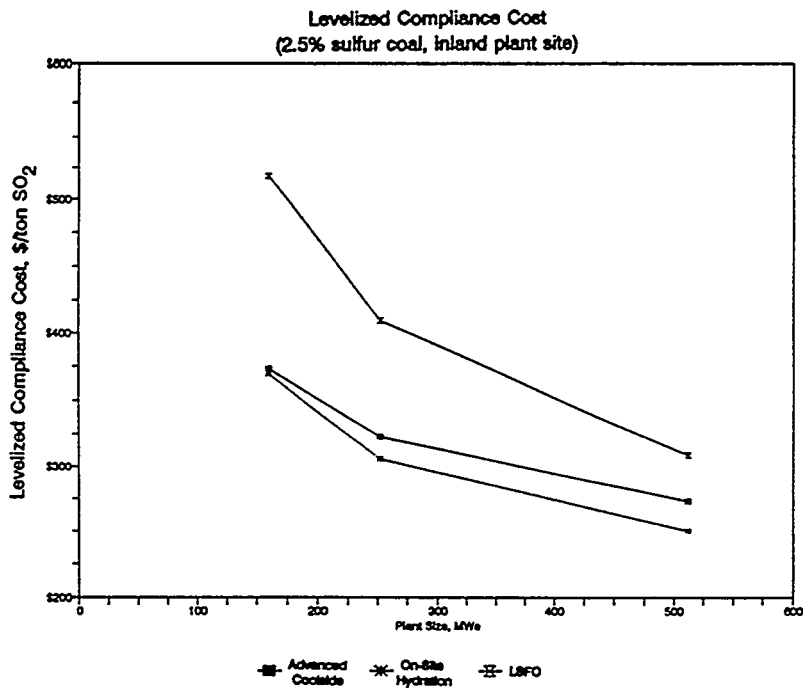
Levelized Compliance Cost
(2.5% sulfur coal, inland plant site)

Figure 5. Comparison of Levelized SO_2 Control Costs for the Advanced Coolside Process and Limestone Forced Oxidation Wet FGD (LSFO) for a Range of Plant Sizes Burning A 2.5% S Coal and Assuming an Inland Plant Site. (For Advanced Coolside, on-site hydration is compared to buying hydrated lime.)

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TABLE 1

SUMMARY OF RECYCLE TEST RESULTS: TESTS SIMULATING SO_2 REMOVAL WITH AN ESP

Test	Fresh Ca/S, mol	Recycle Ratio (a)	Water Addt'n lb/lb Recycle Sorbent	Total $Ca(OH)_2$/S mol Ratio	Approach Temp., Baghouse Inlet Approach, °F	SO_2 Removal, %		Sorbent Util., %	
						Duct	System (b)	Steady State (c)	Solids Analyses
12	1.4	4.5	0.15	2.2	23	83	90	63	62
13	1.2	6.9	0.12	2.1	23	87	90	75	70
12A	1.5	4.3	0.15	2.5	24	84	90	60	59

Common Conditions: SO_2 Inlet Concentration = 1500 ppm (dry); Flue Gas Flow = 340 SCFM

(a)   lb dry recycle/lb fresh lime
(b)   duct + baghouse
(c)   calculated steady-state sorbent utilization

TABLE 2

SUMMARY OF RECYCLE TEST RESULTS: TESTS SIMULATING SO_2 REMOVAL WITH A BAGHOUSE

| Test	Fresh Ca/S, mol	Recycle Ratio (a)	Water Addt'n lb/lb Recycle Sorbent	Total $Ca(OH)_2$/S mol Ratio	Approach Temp., Baghouse Inlet, °F	SO_2 Removal, %		Sorbent Util., %	
						Duct	System (b)	Steady State (c)	Solids Analyses
6A	1.2	5.0	0.00	2.2	10	59	73	61	58
7A	1.3	3.3	0.15	1.8	9	60	84	67	68
8A	1.2	3.4	0.10	1.8	11	64	81	65	66
9	1.5	3.5	0.15	2.2	12	70	90	61	63
10	1.2	4.9	0.15	1.7	9	81	88	71	68
11	1.6	3.9	0.15	2.4	11	91	97	60	58
11A	1.6	3.8	0.15	2.4	12	88	100	61	61
17B	1.2	6.9	0.12	1.4	10	84	92	76	72

Common Conditions: SO_2 Inlet Concentration = 1500 ppm (dry)

(a)   lb dry recycle/lb fresh lime
(b)   duct + baghouse
(c)   calculated steady-state sorbent utilization

TABLE 3

KEY ASSUMPTIONS OF PROCESS ECONOMIC STUDY

	Advanced Coolside	Forced Oxidation Wet FGD
Delivered Sorbent Cost	$65/ton, 7% inerts (hydrated lime) $57/ton (quicklime)	$15/ton (limestone)
Waste Disposal Cost	$6.50/ton	$6.50/ton
SO$_2$ Removal	90%	90%
Capacity Factor	65%	65%
Capital Life	30 years	30 years
Retrofit Factor	Medium (1.22-1.34)	Medium
Location Factor	1.06	1.06
Design Philosophy	"nth" plant, 18% Capital Contingency	"nth" plant, 18% Capital Contingency
Sparing	Auxillary equip. only, no major equip.	Auxillary equip. only, no major equip.
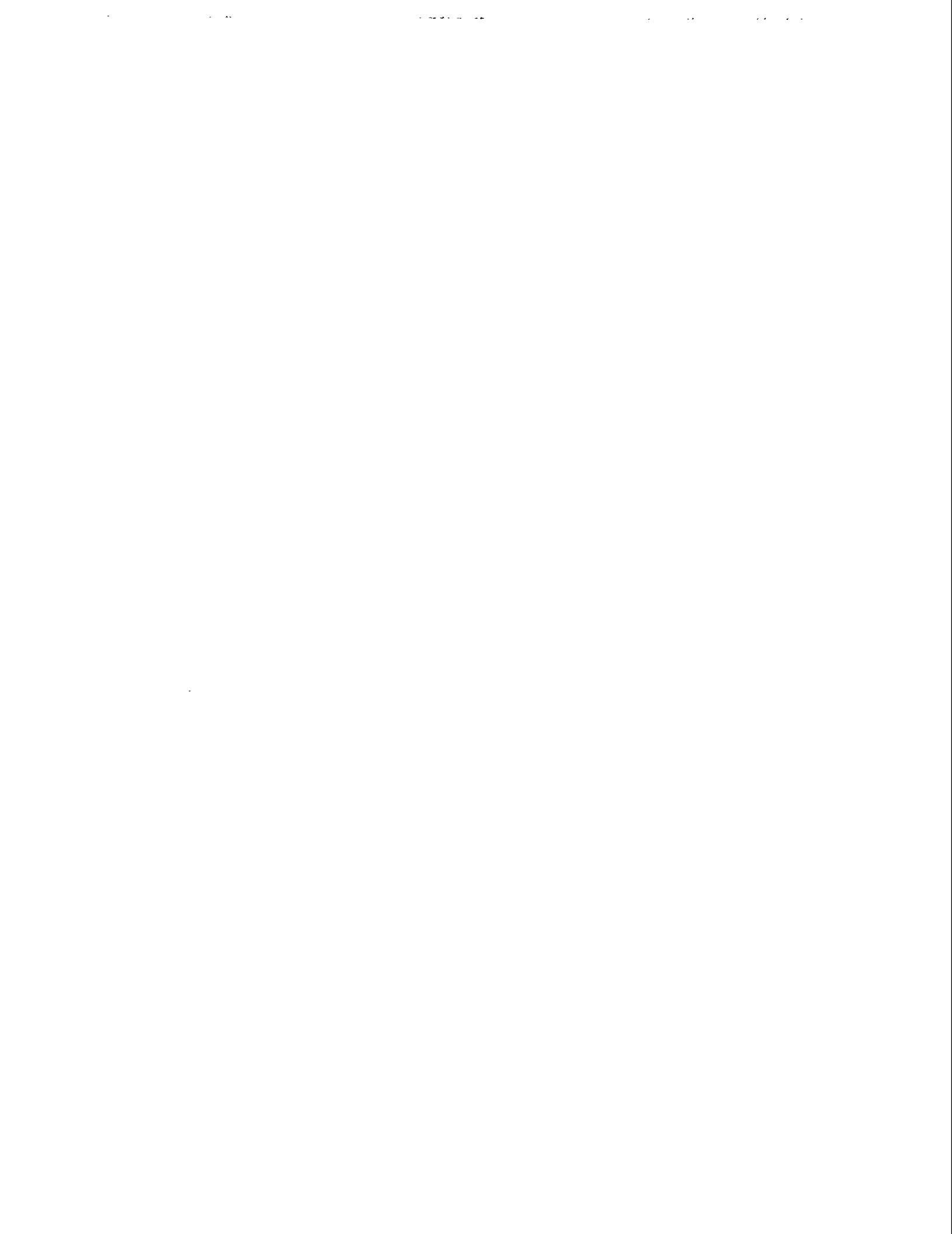
The following manuscript was unavailable at time of publication.

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INVESTIGATION OF COMBINED SO_2/NO_x REMOVAL BY CERIA SORBENTS

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PUSHING THE PULVERIZED COAL ENVELOPE WITH LEBS

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ABSTRACT

In response to challenges from technologies such as IGCC and PFBC, the ABB LEBS Team has proposed removing the barriers to very large advances in environmental and thermal performance of pulverized coal plants. Pulverized coal will continue to be the source of more than half of our electric generation well into the next century and we must develop low-risk low-cost advances that will compete with the claimed performance of other technologies. This paper describes near-term PC technologies for new and retrofit applications which will accomplish this.

INTRODUCTION

Since the inception of the "Engineering Development of Advanced Coal-Fired Low-Emission Boiler Systems" contract (LEBS) the aggressive emissions targets have been gradually tightened and the efficiency target gradually raised in response to pressure from several directions. The contract targets for emissions are now approximately one-half of the original values for NO_x, SO_x and particulates and the efficiency target has been raised substantially - 38% to 42% (HHV, net). The ABB Team believes it would not be difficult to reduce the emissions by one-half again and to raise the efficiency target another 3-4 percentage points, and it proposes to do so.

LEBS is restricted to pulverized coal firing (PC) which is viewed by many as less glamorous than other coal-fired technologies such as IGCC and PFBC, most likely because of the misconception that PC with the Rankine steam cycle has neared its limits of efficiency and emissions performance. In truth, there is considerable room for cost-effective improvements. The path to these improvements is defined and is short. The required development effort is not great and the result will be low-risk low-cost familiar-looking systems which will be readily accepted by the very conservative, risk-averse utility industry. The technologies described below are fuel-flexible and suited to retrofit, repowering and new applications.

SUBSYSTEM TECHNOLOGIES TO ACHIEVE PERFORMANCE GOALS

In-furnace NO_x Control

General Description: The most cost effective way to reduce nitrogen oxides when burning a fossil fuel, coal in this case, is through an in-furnace NO_x reduction process. The foundation for ABB's in-furnace NO_x reduction process is TFS 2000™, a proven technology which is currently being employed on a commercial basis. The TFS 2000™ has been described in detail elsewhere [1]. Briefly, it involves substoichiometric fuel/air operation in the firing zone, the use of concentrically arranged air injection in the windbox whereby the air jets are aimed at a larger imaginary circle than the fuel jets, the use of multiple levels of separated over fire air and the use of a pulverizer with a dynamic classifier. As with all in-furnace NO_x reduction systems, the key is to be able to operate in a mode which produces low NO_x without exacerbating the combustible losses, notably the carbon content in the fly ash. Much of the work in this part of the LEBS program is aimed toward characterizing the performance of firing system configurations that build on the TFS 2000™ concept and which have been formulated by ABB to achieve even greater NO_x reduction without increasing combustible losses.

The objective of ABB's in-furnace NO_x reduction process is to reduce nitrogen oxides leaving the primary furnace to 0.1 lb NO_x/MM Btu (75 ppm @ 3% O_2) or lower while maintaining an acceptable level of carbon in the fly ash. (Further NO_x reduction can be achieved with the downstream SNO_x™ process which is discussed below.)

The process for evaluating the various firing system concepts/configurations which ABB has formulated involves the use of computational modeling, small scale experimental testing and larger scale experimental testing. Additionally, it has involved concurrent characterization of coal pulverization in an ABB-developed pulverizer with a dynamic classifier. As NO_x levels are pushed ever lower it is imperative that the fuel particle size distribution also be more tightly specified as a primary means of controlling combustible losses. Figure 1 is a flowchart showing the interaction of the various activities described above, the primary deliverable being input into the revised system design. The FSBF referred to in Figure 1 is a Fundamental Scale Burner Facility and the BSF is an 80 million Btu/hr Boiler Simulation Facility.

The two primary activities which will be addressed in this paper are preliminary results from testing in the Fundamental Scale Burner Facility (FSBF) and characterization of one of the LEBS coals in ABB's Pulverizer Development Facility. Computational modeling is underway and will be addressed briefly.

Computational Modeling: Two models are being employed to help analyze the various firing systems concepts that have been formulated. A kinetics reaction model, CHEMKIN, is being used to provide a preliminary evaluation of the potential for various concepts to achieve the desired results. It is recognized that results from this evaluation are qualitative at best and can only be used to provide trends; nevertheless its use can be an important screening tool to help prioritize the most promising concepts for further evaluation. A computational fluid dynamics model, FLUENT, is being used to further evaluate concepts under conditions which better simulate actual boiler operation. Unlike CHEMKIN which assumes either well-stirred reactor conditions or perfect plug flow conditions, FLUENT is able to simulate real-world mixing conditions. ABB's large combustion facility, the Boiler Simulation Facility (BSF) has been modeled with FLUENT. Experimental measurements from the BSF compare quite well with those predicted by FLUENT, namely parameters such as gas temperatures and gaseous concentrations like O_2 and CO. Having validated FLUENT with BSF data, the intent is to use it as well as the CHEMKIN model in ways that capitalize on their respective strengths to evaluate and screen various firing system concepts.

Fundamental Scale Burner Facility (FSBF): The LEBS plan calls for evaluation of advanced coal reburning as a supplemental NO_x reduction technology. Reburning is classically thought of as a separate, downstream (from the primary combustor) zone into which "reburn fuel" is injected followed by a "burnout zone" where air is injected

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[1] "Development of ABB CE's Tangential Firing System 2000" presented at the '93 EPA/EPRI NO_x Conference.
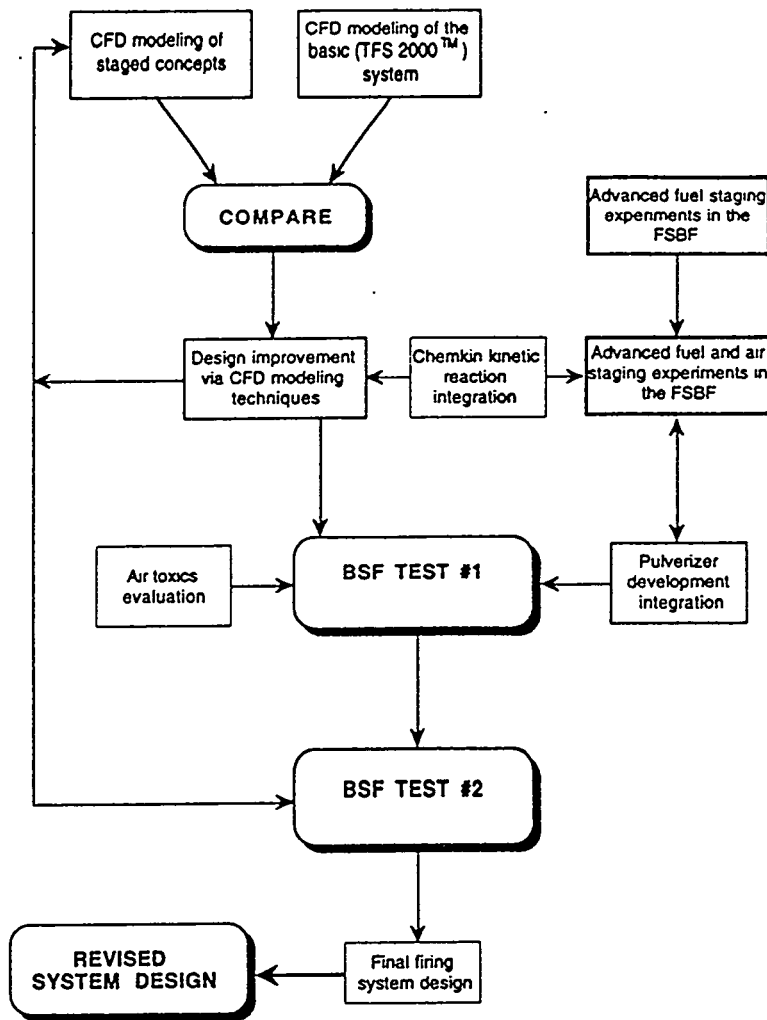
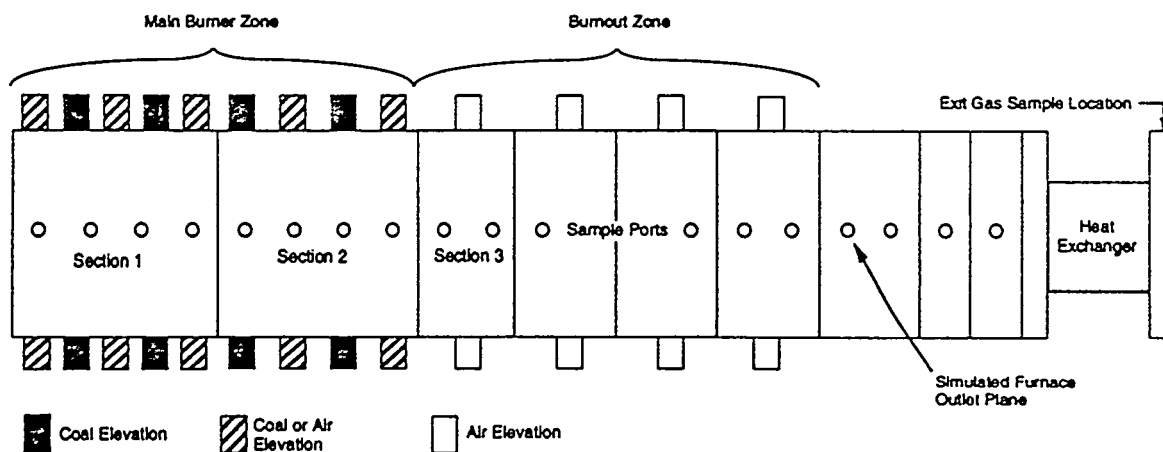Figure 1 - LEBS Work Flow Chart

Figure 2 - Schematic of Fundamental Scale Burner Facility

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to burn out the remaining combustibles from the reburn zone, which typically operates at substoichiometric conditions. Reburning in the classical sense has been shown to be an effective technology for reducing NO_x; however, when used in the traditional fashion greater residence time is needed in the separate reburn and burnout zones. However, NO_x reduction through the reburning process can and does occur within the primary combustor. The objective of testing in the FSBF has been to characterize the NO_x and combustion performance from firing system concepts which do not have the classical, separate reburn and burnout zones, but rather which employ integrated strategies within the main windbox zone that take advantage of NO_x reduction through reburn process chemistry. The advantages are less residence time, a smaller furnace and a favorable cost impact. It is believed that tangential firing, specifically building upon the already-established TFS 2000™ system, is well suited for adaptation to the integrated firing system concept.

The Fundamental Scale Burner Facility (FSBF) is a horizontally fired experimental combustor which has a capacity of 5 million Btu/hr. See Figure 2. It has been configured to simulate tangential firing; air and fuel are injected from four nozzles for each plane, or elevation, the term used to describe a plane in a tangential firing system. As noted in Figure 2 there are a number of planes from which fuel or air can be injected to simulate and evaluate a particular firing arrangement. Additionally there are air-only injectors downstream of the main windbox to simulate over fire air injection.

Low NO_x firing generally requires that the main windbox or burner zone be fired under substoichiometric conditions. Table 1 shows the relative NO_x values as a function of bulk stoichiometry in the main windbox zone as simulated in the FSBF. Configuration 1 represents a base case where all of the combustion air is injected through the main windbox; the NO_x level is arbitrarily shown as 100%. Configurations 2 and 3 show relative NO_x values for substoichiometric firing in the main windbox and with different amounts of Separated Over Fire Air (SOFA) in SOFA levels 1 and 2. As expected, substoichiometric operation results in lower NO_x and the strategy for staging the SOFA also makes a difference in the final NO_x levels.

Table 1. Relative NO_x Levels vs. Main Burner Zone
Stoichiometry (MBZ) and Separated Over Fire Air (SOFA)

Configuration	NO_x%*	MBZ Stoich	SOFA 1 Stoich	SOFA 2 Stoich
1	100	1.15	1.15	1.15
2	66	0.8	1.0	1.15
3	54	0.8	0.8	1.15

*Normalized, with Case 1 assigned 100%

Initial testing in the FSBF was designed to evaluate a number of variables within the main firing zone, including firing system configuration and operating conditions, for their effect on NO_x. Table 2 shows some initial results from a number of firing system configurations, some of which employ integrated firing system strategy. Relative to the base case, configurations 4 and 5 are run to produce lower NO_x without the use of SOFA, while configurations 6 and 7 show relative NO_x levels with the use of SOFA.

Table 2. Relative NO_x Levels for Integrated Firing Configurations vs. Basecase (Configuration 1)

Configuration	NO_x%*	MBZ Stoich	SOFA 1 Stoich	SOFA 2 Stoich
1	100	1.15	1.15	1.15
4	64	1.15	1.15	1.15
5	59	1.15	1.15	1.15
6	54	0.8	0.8	1.15
7	49	0.8	0.8	1.15

*Normalized, with Case 1 assigned 100%

Testing in the FSBF is continuing; plans under our LEBS Program call for continued evaluation of firing system concepts that are hypothesized to produce ever lower NO_x levels while meeting our goal of maintaining combustible losses at minimum levels.

Coal Pulverization : As noted above, specification and control of coal particle size distribution is an important prerequisite for successful operation of a low NO_x firing system. Conditions for achieving low NO_x tend to run counter to those that are favorable for good coal combustion; therein lies the challenge. Paying attention to the proper coal particle size distribution has the obvious effect of facilitating better carbon burnout and the perhaps not-so-obvious effect of enhancing NO_x reduction through earlier release of nitrogen species in the near-burner zone where the opportunity for conversion to molecular nitrogen is increased.

ABB has constructed a Pulverizer Development Facility (PDF) for the study and characterization of coal pulverization and classification. The PDF includes a coal storage and feed system and a fine coal collection system as necessary support equipment for the pulverizer itself. The mill represents a commercial design, based on a size 323 bowl mill, but with the flexibility to change out important components within the mill, such as grinding elements and classifiers. The capacity of the PDF is about 3.5 tons/hr.

The early focus of the LEBS-related work which utilizes the PDF has been to characterize the coal particle size distribution and mill power requirements. Ideally it is desired that the top size of the coal particles be closely controlled and that classification is more efficiently carried out so that sufficiently fine particles are not recirculated needlessly back to the grinding zone of the mill. The use of a dynamic classifier is one way of accomplishing this.

Table 3 shows results from recent testing with various dynamic classifier designs as compared with a base case static classifier design. It is apparent that the goals of greater coal fineness, less coarse material and lower power requirements have all been achieved with at least two of the dynamic classifier designs. Results have been demonstrated with conventional air:coal ratios, namely 1.5 lb air/lb coal.

Future testing in the FSBF will employ the use of coals having various particle size distributions to ascertain and quantify the benefits of using finer coal.

Table 3. Dynamic Classifier Characterization

	Static Classifier	Dynamic Classifier Designation			
		HP1	HP2	RB1	RB2
Product Size (wt%)					
+50 mesh	0.1	0.2	0.0	0.0	0.0
+100 mesh	2.4	1.2	0.6	0.5	0.6
-200 mesh	84.3	85.7	92.7	93.1	90.7
Relative Power Requirements (%)	100	81	98	97	107

Stack NO_x, SO_2, Particulates and Title III Pollutants

General Description of Control Technology: Boiler outlet emissions will be controlled by a modified SNO_x™ process, referred to as the SNO_x™ Hot Scheme. The SNO_x™ process which simultaneously removes nitrogen oxides and sulfur oxides from flue gases, is a licensed technology developed by Haldor/Topsøe A/S, Denmark. The SNO_x™ technology has been demonstrated in several forms, one as a Clean Coal Technology at Ohio Edison's Niles Station, and has been constructed and operated on a commercial scale in Denmark. The SNO_x™ technology consists of five key process areas: particulate collection, NO_x reduction, SO_2 oxidation, sulfuric acid condensation, and acid conditioning. For the LEBS process, the particulate collection and NO_x reduction process are integrated into a single process step.

Particulate/NOx Control: The first step, particulate collection, will have a direct effect on the performance of the downstream SO_2 converter, particularly the frequency of cleaning the SO_2 catalyst. This is due to the inherent ability of the catalyst to retain greater than 90% of all particulate matter which enters the converter. The collection of this particulate matter, over time, will cause the gas draft loss to increase. The virgin draft loss can, however, be restored through catalyst cleaning, called screening. Higher dust loads at the SO_2 convertor inlet therefore require more frequent cleaning, and higher catalyst attrition loses. A target dust level of 0.0008-0.0016 lb/MMBtu (1-2 mg/Nm3) leaving the collector is desired. Consequently, dust emissions from the SO_2 convertor are often an order of magnitude lower.[2]

To achieve the required particulate loadings at the SO_2 converter inlet, a high efficiency collection device must be employed. For the LEBS process, a ceramic filter manufactured by CeraMem will be employed.

The construction of the ceramic filter is based on the use of porous honeycomb ceramic monoliths. These high surface area, low cost materials were developed for, and are widely used as, catalyst supports. The monoliths have many cells or passageways which extend from an inlet face to an opposing outlet face. Cell structure is usually square and cell density can vary from 25 to 1400 cells per square inch (cpi) of face area. Mean pore size can range from 4 to 50 microns.

The superior properties of commercially available monoliths make them ideally suited for applications requiring high thermal stability, mechanical strength, and corrosion resistance. These rigid ceramics have been used for years as NO_x SCR catalyst supports in combustion flue gas applications. The monolith structure used for catalyst support material is readily adapted to function as a particulate filter. The monolith structure is modified by plugging every other cell at the upstream face with a high-temperature inorganic cement. Cells which are open at the upstream face of the monolith are plugged at the downstream face. Flue gas is thereby constrained to flow through porous cell walls, and at appropriate intervals, the filter is cleaned by backpulse air.

CeraMem has developed the technology for applying thin ceramic membrane coatings to the monoliths and controlling the pore size. The thin (approximately 50 microns) membrane coating has a pore size approximately 100-fold finer than that of the monolith support. Thus the filter retention efficiency is determined by the membrane pore size, not the monolith pore size. The ceramic filter will operate as an absolute filter; that is all particulate over a certain diameter will be removed from the gas stream. The split diameter is determined and controlled by the ceramic application.

In the LEBS process, commercially-tested SCR catalyst is applied to the clean side of the particulate filter. As with other low dust SCR applications, concerns about flyash poisoning of the catalyst are eliminated, and catalyst loadings may be reduced as the catalyst will have a "higher" activity. Also, in this application, the reaction kinetics will not be controlled by mass diffusion as in other monolith applications. Instead, the kinetics will be much faster, taking advantage of "forced diffusion", where the flue gas will come into forced contact with the catalyst as it passes through the monolith wall. A third benefit of this technology in relation to SCR performance will come about from elevated conversion temperature. Typical SCR applications operate at about 675°F, whereas the LEBS application will operate at a slightly higher temperature of 750-775°F. Increased temperature should not affect catalyst life, but should improve the efficiency of the reducing reagent, in this case ammonia. The increase in temperature should result in a lower ammonia concentrations at the SCR outlet, often called slip.

Particulate and NOx Emissions Levels: Taking advantage of the clean-side catalyst application, forced diffusion kinetics, and higher reduction temperature should allow for much higher reduction efficiencies and efficient reducing reagent consumption. Early data indicate that at NO_x inlet concentrations of 200 ppm, NO_x reduction should exceed 90% without any measurable ammonia slip.

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[2] These levels are below normal detection limits of EPA Method 5 sampling. The method collection time would have to be extended to be able to detect these emissions.

In particulate collection tests conducted at ABB's Corporate Research Facility, collection efficiency was found to be almost absolute, in most cases greater than 99.99996% with an inlet flyash loading of 4-5 lb/MMBtu (5-6 g/Nm^3). Outlet emissions could not be detected by standard EPA Method 5 techniques, and were determined to be less than 0.0000016 lb/MMBtu (0.0020 mg/Nm^3) by laser-light scattering instrumentation.

SO$_2$ Control: SO$_2$ emissions are controlled by the SO$_2$ oxidation catalyst, sulfuric acid condensers, and acid conditioning system. An oxidation catalyst, which is widely used in the sulfuric acid industry, converts the SO$_2$ to SO$_3$ at greater than 97% efficiency. The efficiency of the catalyst is not affected by presence of water vapor or chlorides in concentrations up to 50% and several hundred ppm, respectively. An additional benefit of the sulfuric acid catalyst is its ability to oxidize carbon monoxide and hydrocarbons present in the flue gas stream to innocuous compounds.

The SO$_3$ in the gas leaving the SO$_2$ converter is hydrated and condensed in two steps. First, the bulk of the SO$_3$ is hydrated to sulfuric acid vapor as the flue gas passes through the Ljungstrom air heater and the temperature drops to approximately 500°F. At this point, the flue gas is still well above the acid dewpoint, thus avoiding acid condensation and corrosion of the ductwork. The flue gas then enters the WSA Condenser, a unique tube and shell falling film condenser with the boiler combustion air used as a cooling medium on the shell side. Borosilicate glass tubes are used to convey and cool the flue gas. In both steps, the hydration and condensation reactions are exothermic, thereby adding heat to the flue gas and subsequently to the boiler thermal system. The design and operation of the WSA Condenser make possible virtually complete condensation and capture of the sulfuric acid at concentrations of 92 to 95 wt %.

SO$_2$, SO$_3$ Emissions Levels: To this point, SNO$_X$™ systems have not been built with SO$_2$ removal efficiencies of greater than 95%, and therefore, data other than that obtained at laboratory scale would not support the ability to achieve higher removal efficiencies. However, ultra-high removal efficiencies (typically greater than 98%) have been studied by Haldor/Topsoe, with the information being used to design, build, and operate high-efficiency systems. As this is a catalytic system, SO$_2$ removal efficiency is fixed and somewhat inflexible. If a system is designed for a specific removal efficiency, it will maintain that degree of control over a wide operating range without any drop-off, unlike chemical reagent systems which tend to become gas-side limited and lose removal capibility as inlet SO$_2$ levels decrease. Increasing removal efficiency would require minor modification of the convertor vessel with catalyst addition.

SO$_3$ emissions will be controlled by the efficient condensation system, in excess of 99.9% condensation. However, some SO$_3$ will pass through the system and will exit the stack, and it is expected that this amount would not be in excess of 20 ppm - a level similar to emissions from present-day wet or dry desulfurization systems.

Title III Pollutants: The Niles demonstration facility was sampled as part of the DOE/EPA Field Chemical Emissions Monitoring. It was found that the SNO$_X$™ technology was able to reduce Title III metal emissions by greater than 98% and Title III organic compounds were not detected at significant levels. The commercial scale facility in Denmark was also sampled by an independent team, with the results reported confirming those obtained from the Niles sampling.

Improved Thermal Efficiency: Heat addition, transfer, and recovery are of significant importance in the SNO$_X$™ process. The process generates recoverable heat in several ways. All of the reactions which take place with respect to NO$_X$ and SO$_2$ removal are exothermic and increase the temperature of the flue gas. This heat is recovered in the air heater and WSA Condenser for use in the furnace as combustion air. Because the WSA condenser lowers the temperature of the flue gas to about 210°F, compared to the 300°F range for wet and dry scrubbers[3], additional sensible heat is recovered along with that from the heats of reaction. In comparison to an NSPS-compliant plant, 38% more heat is recovered from the flue gas stream after the boiler, itself accounting for a 1.9 percentage point increase in the net plant thermal efficiency.

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[3] Although stack temperatures for wet and dry FGD systems range from 125-250°F, heat recovery - that done by the air heater - usually is limited to a minimum temperature of 300°F. After that, the flue gas is quenched, accounting for the temperature difference.

Waste and Byproducts

As shown in Table 4, unlike many other processes, the SNO_x™ process does not generate a waste product or intermediate. Also, the SNO_x™ process does not produce a "commercial grade" product which does not meet the specifications of its intended market, as has been the case with Wet FGD gypsum. The sulfuric acid produced by the SNO_x™ process, a typical analysis of which is presented in Table 5, meets or exceeds U.S. Federal Specification O-S-801E Class 1 and is commercially tradable without limitation.

The ABB project team includes, as an advisor, Peridot Chemicals. Peridot Chemicals operates two sulfuric acid production facilities and distributes acid from several international involuntary acid producers. Peridot Chemicals has provided very useful insight into the domestic sulfuric acid market.

Installation of a SNO_x™ facility, or any large acid production facility, will force a reshaping of the local acid market, and alliances could be made with local brokers, suppliers, and consumers for distribution and consumption of the acid. It is believed by sulfuric acid market experts that domestic involuntary acid production could displace international involuntary acid production.

It is expected that the flyash from the LEBS system could be sold commercially, similar to present day ash disposal. Carbon content in the flyash is expected to be less than 5%, and there is not expected to be any noticeable presence of ammonia.

Table 4. End product and disposition by technology.

Technological Advancement	Product and Disposition, Cost Comparison
1st Generation Technology - consumable reagent processes, usually sodium- or calcium-based, such as lime/limestone Wet FGD, lime Dry FGD, limestone furnace injection, and duct injection.	Landfill of sulfite/sulfate compounds with little commercial or industrial value. Calcium sulfate from wet FGD can be upgraded to commercial-grade gypsum, but at significant cost. Low capital cost, offset by high O&M cost.
2nd Generation Technology - regenerable reagent processes, such as CuO and MgO systems.	Produces chemical intermediate, usually metallic sulfide or sulfite, which must be landfilled or further processed to produce sulfuric acid or elemental sulfur. High capital cost and O&M cost.
3rd Generation Technology - catalytic (no reagent) technologies, such as SNO_x™ process.	Direct production of elemental sulfur or sulfuric acid. High capital cost, offset by low O&M cost.

On whole, it is expected that the amount of landfill material from an NSPS-compliant plant (ESP/limestone WFGD) will be reduced by approximately 85%, expressed on a heat input basis. This figure accounts for the FGD by-product, a wet mixture of calcium sulfite and calcium sulfate, being landfilled as opposed to the SNO_x™ product, commercial grade sulfuric acid, being sold. For every ton of sulfur in coal, 7.7 tons of landfill waste has been converted to 3.3 tons of sulfuric acid. With evaluations of $15 per ton landfill and $30 per ton commercial grade acid, a net swing of $214 is created, from an outlay of $115 for landfill costs to an income of $99 from acid sales (expressed on per ton S in coal basis). Comparing disposal for the LEBS SNO_x™ design to a dry FGD/fabric filter system would yield a differential of $164 in favor of the SNO_x™ plant, primarily in the reduction of landfill costs associated with a dry waste. At a acid price of $20 per ton, it is expected that the air pollution control system would operate at a financial break-even position.

**Table 5. Federal Specification For Commercial Grade Sulfuric Acid
and Actual Acid Analysis from Operating Unit**

Criteria	Federal Specification Maximum Values	Typical Values from Operating Unit
Concentration, wt %	93.2	94.76
Color	Water Clarity, None	ND
Iron, ppm	50	3.4
Copper, ppm	50	0.025
Zinc, ppm	40	0.12
Arsenic, ppm	1	0.012
Antimony, ppm	1	<0.002
Selenium, ppm	20	26
Nickel, ppm	1	0.04
Manganese, ppm	0.2	0.065
Ammonium, ppm	10	2.9
Chloride, ppm	10	ND

HIGH EFFICIENCY POWER CYCLES

Steam Cycle: The most widely used power plant in the United States has been a subcritical single reheat cycle. It features a drum boiler operated to produce 2400psig/1000°F at the turbine throttle. In the late fifties, the industry introduced supercritical steam cycles which enabled higher plant efficiencies and improved operating costs. As the initial problems were resolved and supercritical technology matured, these plants demonstrated availabilities comparable to their subcritical counterparts. However, for a variety of reasons but mainly due to low cost of fuel, there have been no supercritical plants constructed in the United States since the late seventies.

The original incentives for supercritical cycle development in the 1950's are even more critical today. The time has come to take a hard look at cycle options and improvements in heat rate through higher steam conditions. Heat rate improvement means reduced emissions of SO_2, NO_x, CO_2, particulate, etc., per unit of electricity produced.

Figures 3 illustrates heat rate improvements with the increase in pressure and temperature for a single and double reheat plant as compared to a conventional subcritical unit. For example, a steam cycle designed with steam conditions of 3625psig (250 bar) and 1000°/1000°F offers plant heat rate improvement of over 1.4%.

Heat Rate Improvements
with Single Reheat Turbine

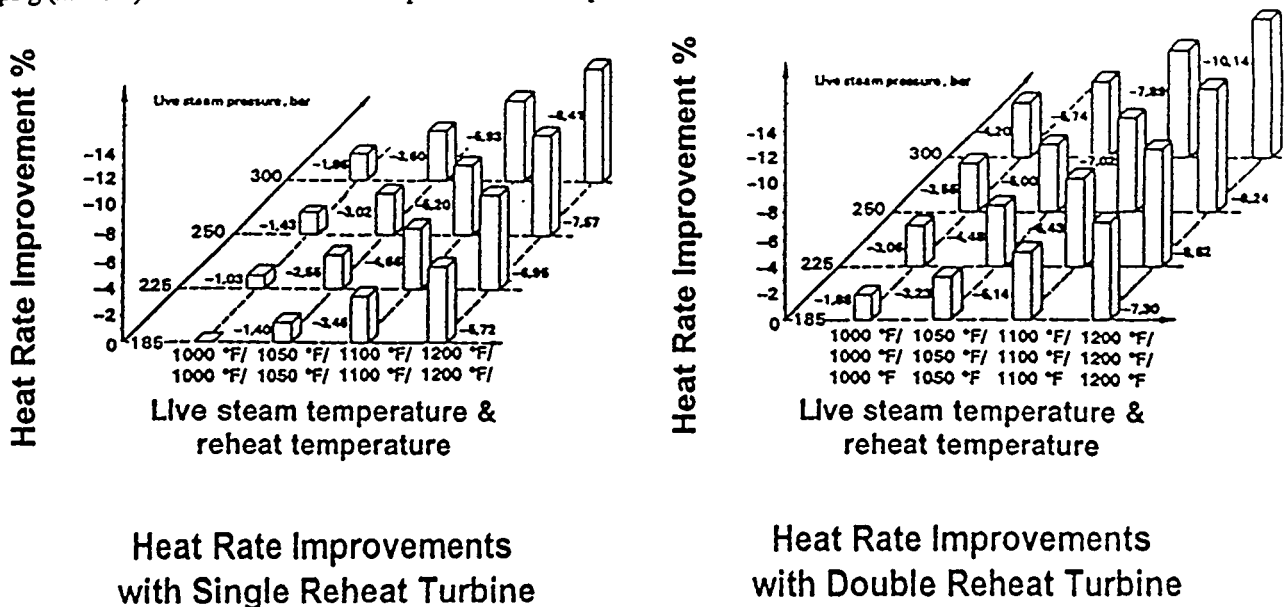Heat Rate Improvements
with Double Reheat Turbine

Figure 3 - Heat Rate Improvement vs Pressure and Temperature

For over a decade, ABB CE with the support from DOE and EPRI has been participating in the development of an advanced steam cycle with throttle conditions of 4500psig/1100°/1100°/1100°F. This plant is commercially available and includes state-of-the-art technical advances made in materials, manufacturing processes, design analyses, and control systems. Plants with very similar steam conditions are in successful commercial service in Japan and Denmark (where high efficiency has greater value.) Depending on the condenser pressure, plant capacity, and type of coal-fired, the net plant HHV efficiency is approximately 41% (8324 Btu/kwhr) to 43% (7937 Btu/kwhr). Although the steam conditions may appear to be advanced, they do not constitute a significant departure from the current experience. One may only need to recall the Eddystone unit of the Philadelphia Electric Company which was commissioned in 1959. With initial steam conditions of 5000 psig, 1200°F and two reheats of 1050°F, Eddystone I had the highest steam conditions and efficiency of any electric plant in the world. Due to some initial problems, very few of which were related to high temperature and pressure, the steam turbine throttle conditions were reduced to 4700 psig and 1130°F. It remains an important unit in Philadelphia Electric's future generation plan as evidenced by life extension beyond the year of 2010. However, in contrast to the Eddystone unit which was designed for base load capacity needs, the state-of-the-art plant would be capable of sliding pressure mode of operation and cycling duty with fast start-up and fast load change rates. These desired plant characteristics are accommodated by introducing a steam generator with spirally wound furnace walls, an integrated start-up system, and a split back pass for steam temperature control. The past experiences are factored into the design of critical components. Major design improvements include improved materials such as advanced ferritic alloys as T91 (9Cr) and modified 12 Cr for piping, headers, and steam turbine rotors. Better analysis techniques combined with advanced monitoring and control systems ensure that the state-of-the-art plant would be able to operate without any loss of component life over and above that expected from current units.

Higher steam conditions, such as 5000psig/1200°/1200°/1200° F, offer the prospect of an additional plant efficiency improvement of approximately 3%. The estimated net plant efficiency is in the range of approximately 42% (8126 Btu/kwhr) to 44% (7757 Btu/kwhr). Design of the high temperature components is not expected to change significantly from the state-of-the-art plant except for some material upgrade in the critical areas. For base load capacity, technology is probably available to build this plant today, particularly, if the reheat temperatures are reduced to 1150° or 1100° F. For cycling duty, the key to success lies in the application of advanced high strength ferritic and austenitic alloys developed in the past decade for such critical components as furnace wall tubing, headers, piping, and steam turbine rotors. It is believed that with some additional R&D effort the 5000 psig steam cycle can be offered commercially in the 2000 to 2002 time frame.

Significant additional thermodynamic gain can be achieved by adopting even higher steam conditions such as 6000psig/1300°/1300°/1100°F. The expected net plant efficiency should be in the range of 43.5% (7846 Btu/kwhr) to 45.5% (7501 Btu/kwhr). Since these steam conditions fall outside the realm of the current experience, there is little doubt that formidable technical problems will need to be solved. To meet the future needs for high efficiency, the industry is beginning to experiment with high steam temperature applications. Conceptual designs and preliminary test results at steam conditions of 1500psig/1500° F have been reported in the literature. To facilitate implementation of the new technology consistent with the needs of the early 21st century, perhaps the time has come for comprehensive assessment of the ultra high steam conditions.

Kalina Cycle: An alternative approach to the use of higher temperatures and pressures to gain Rankine cycle plant efficiency is to change the cycle working fluid. The use of mixtures as the working fluid provides the ability to vary the composition throughout the cycle. This realizes a structural advantage in designing a power plant cycle by providing a degree of freedom to minimize thermodynamic losses throughout the process. The Kalina cycle, currently under demonstration, is one such cycle.

In the Kalina cycle a mixture of ammonia and water is used as the working fluid. In contrast to a single component, the temperature of the ammonia/water working fluid continually changes during the boiling process. The light component (ammonia) boils off first, leaving behind a mixture with a greater concentration of the heavier component (water). As this occurs, the boiling temperature of the remaining liquid increases. This fundamental degree of freedom facilitates the minimization of thermodynamic losses.

For direct fired Kalina cycle applications (those where the source of thermal energy input to the cycle comes from fuel combustion) net plant efficiencies in the range of 45% to 50% (HHV) are possible today. This range can be achieved at vapor conditions of 2400 psig/1050°F/1050°F/1050°F. Similar to the steam Rankine cycle, increasing vapor temperatures will augment the efficiency advantage of Kalina cycles.

Kalina cycle plants can take advantage of all LEBS technological advances in combustion and emissions control. The plant cycle may be structured to accommodate the application of the SNO$_X$™ Hot Process, offering substantial efficiency improvement versus the conventional Rankine cycle. Finally, because the efficiency gains are a result of structural improvements in the plant cycle, the capital cost of the plant may be less than a conventional Rankine subcritical single reheat plant.

PROOF-OF-CONCEPT TEST FACILITY

If the ABB Team is selected to execute Phase IV of the LEBS Project, a 435 million Btu/hr system (coal and flue gas flow equivalent to 50 MWe net @ 42% efficiency) will be constructed as a repowering of Richmond (Indiana) Power & Light - Whitewater Valley Unit 1. Since the existing turbine/generator and most of the boiler and infrastructure may be retained, it may not be possible to demonstrate an advanced cycle. However, the system will be in commercial service during the six months of testing which will permit conclusive demonstration of: all emission control technologies, operability and reliability of a complete generating system in full commercial service, the integration (air, gas and water) of the SNO$_X$™ hot process with the boiler system and turbine/generator, the quality of the byproduct sulfuric acid, etc.

The major items of equipment that will be installed are:

- Complete Firing System (feeders, pulverizers, burners, air supply, burner management system) and required boiler modifications.
- Complete SNO$_X$™ Hot Process including ammonia/urea injection, catalytic filter, SO$_2$ reactor, acid condenser, acid and ash storage and handling.
- Gas bypass to maintain desired gas temperature to the SNO$_X$™ Hot Process.
- Air-to-Condensate Heat Exchanger to augment existing feedwater heater.
- Gas-to-Air heat exchanger.
- Clean gas ductwork to the existing stack.
- Boiler Forced Draft Fan and Drive.
- Induced Draft Fan and Drive.
- Hot air ductwork to the boiler.
- Distributed Control Systems.
- Diagnostic Devices.
- Required BOP, demolition, civil and structural work to provide a complete operating plant.

The existing boiler's air heater and associated ductwork will be removed.

The true value of the LEBS Project lies in the testing and commercial operation of a POCTF of sufficient capacity to convince the markets that LEBS represents low-risk technologies. This is the only sure path to the overall objective of expedited commercialization.

A COMMERCIAL GENERATING UNIT DESIGN

In Phase I of the Project each contractor produced the preliminary design of a commercial generating unit (CGU). The ABB Team's 400 MWe CGU illustrated in Figure 4 is an adaptation of a conventional pulverized coal-fired steam-electric plant. It will be compared to a Kalina design under a separate work effort. For each design

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selected technologies have been introduced to achieve reduced levels of emissions, increased thermal efficiency, reduced waste and improved costs. These technologies involve primarily three areas:

- An advanced low-NO_x combustion system.
- The SNO_x™ Hot Process.
- Advanced supercritical boiler and turbine cycle.

This combination of emission control processes meets or betters all of the target emission levels for the LEBS Project, while producing either benign or saleable by-products from the gas treatment. The advanced cycle and the SNO_x™ Hot Process enable the design to meet the efficiency objective and, indirectly, the cost of electricity objective.

Expected performance of the CGU compared to an NSPS-compliant plant is listed in Table 6. The NSPS plant assumes a 2400 psi/1000°/1000° cycle with wet limestone FGD and an electrostatic precipitator.

Table 6. Emissions Reduction Performance

		NSPS PLANT	LEBS CGU
SO_2,	lb/mm Btu*	0.60	0.10
NO_x,	lb/mm Btu	0.60	0.02
Particulate,	lb/mm Btu*	0.030	0.002
Total Waste,	LB/kWh	0.352	0.117
Net Efficiency (HHV),	%	35.4	45

* 3 lb S and 15.4 Lb ash per million Btu in the coal.

Volatile organic emissions, CO and ammonia slip will be oxidized in the SO_2 oxidizer and there will be no visible stack plume. The CGU produces significantly less waste than the NSPS plant. Part of this is due to the lower amount of ash produced per kWh because of the higher efficiency cycle and the SNO_x™ Hot Process. The major portion of this reduction results from the production of sulfuric acid as a commercially saleable by-product rather than the sludge normally generated by an FGD system. The plant uses a 5500 psig, 1300°F supercritical thermodynamic cycle with two reheat streams. The gross output of the generator is 468 MWe and the net plant output is 445 MWe.

The CGU has a total plant cost that is less than the cost of a current NSPS plant. The total capital requirements estimate includes the time related portions of the project estimate such as allowance for funds used during construction. The improvements come from the adoption of an aggressive commercialization plan that utilizes the concept of a "Consortium" formed to produce a number of these units on a replicated and modularized basis. These factors, coupled with ABB's commitment to a significantly reduced "cycle time" for the boiler and other key equipment results in significantly reduced schedule from award to start-up. This aggressive construction schedule improves the time-related costs.

The CGU will satisfy the objective of having a cost of electricity equal to or less than that for the NSPS plant. The calculated cost of electricity is reduced by the by-product credit received from the sale of the sulfuric acid (using a figure confirmed by an outside market study) and by an aggressive but achievable capacity factor. An independent reliability, availability and maintainability analysis was completed for the CGU. The study was based on performance data obtained from the NERC data base and utilized the industry accepted "Delphi" process to adapt the data for the CGU. One reason enhanced reliability and equivalent availability are achieved is that the SNO_x™ Hot Process is "passive", i.e., it has far less mechanical equipment than is typically found in flue gas desulfurization processes that utilize lime or limestone. The simpler process, absence of mechanical equipment, and the passive character of the process results in higher reliability and availability.

The design also incorporates advanced diagnostics concepts which provide early warning of impending failures in the plant equipment. This advanced knowledge has several benefits that result in improved reliability and availability. Advanced diagnostics should enable maintenance outages to be both more effective by providing maintenance information in areas which might not be readily amenable to inspection, and shorter because preparation will be better due to a reduced number of "surprise" repairs.

Finally, the CGU will have good access and ease of maintenance because it was designed for good access and ease of construction. The plant is laid out with the "ranch" concept. This means that the stacking of equipment is minimized. Rather, it is spread out in the horizontal plane. In addition, the plant design incorporates a "backbone" utility rack for piping, cable, conduit and electrical wiring. The ground level portion of this rack is used as a maintenance access corridor that runs throughout the plant. Also, organizing piping and conduit on overhead racks provides more ground level access to equipment for maintenance. Incorporating these features in the design of the plant, and coupling them with the implementation of a "design for maintainability" approach during the detailed design stage, will result in a plant with superior availability and higher capacity factor which helps reduce the cost of electricity.

CONCLUSIONS AND FUTURE WORK

All of the foregoing are responsive to the technical, regulatory and economic needs of the power generation industry. The advanced performance of ABB's LEBS technologies coupled with efforts to minimize investor risk should make it extremely attractive to the utilities and IPPs. The near term character of the LEBS technologies chosen, coupled with the attractive performance and cost features of the ABB CGU, support a confidence in the Project Team that the proposed CGU design will be acceptable and marketable.

Testing at laboratory and pilot scale will be completed in Phase II. Phase III will consist of updating the CGU and POCTF designs based on the results of the work completed in Phase II. In addition, the licensing of the POCTF will be completed and a detailed test plan will be written.

*   *   *   *   *   *

Figure 4 - 400 MWe Plant
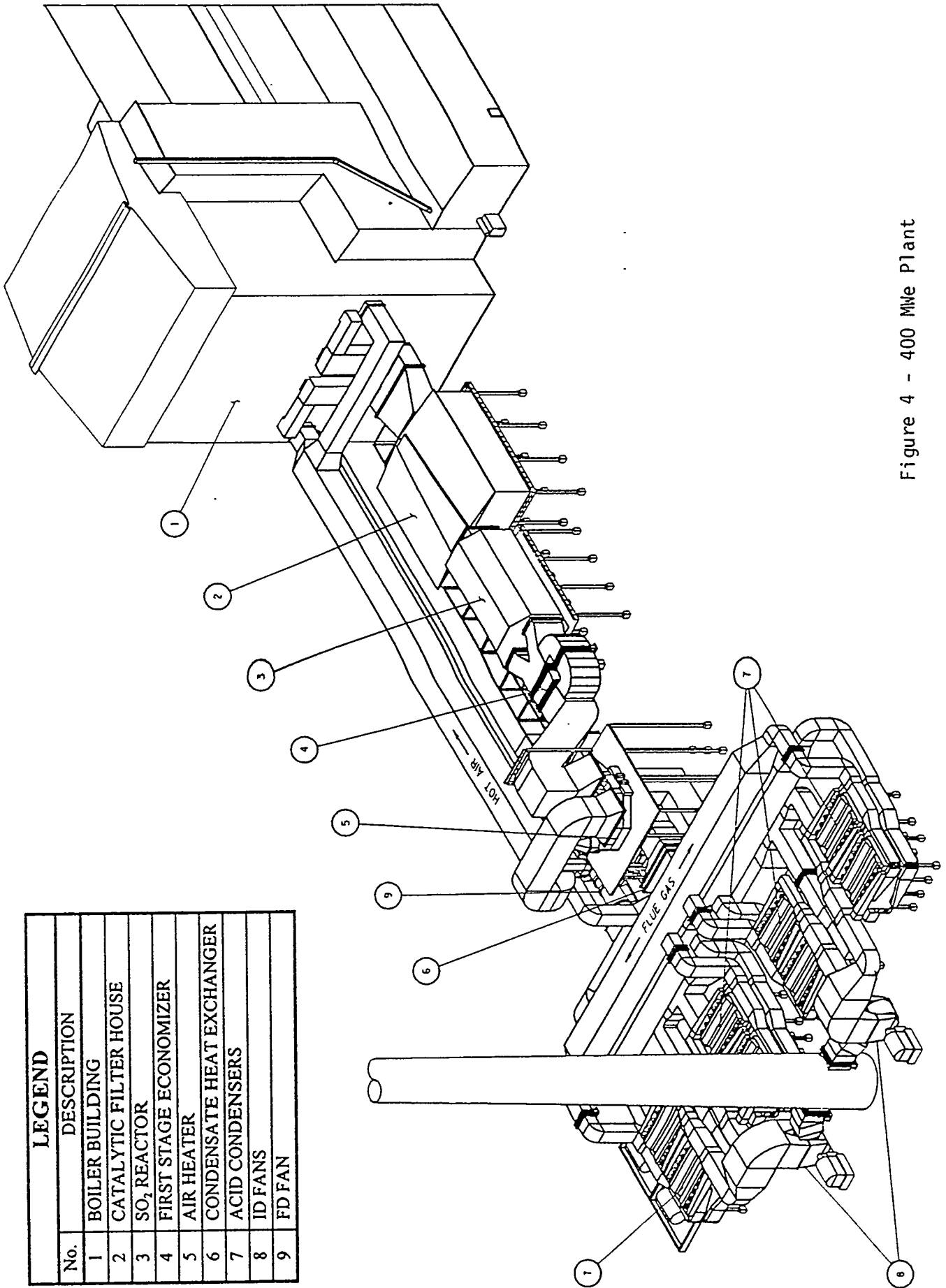
LEGEND	
No.	DESCRIPTION
1	BOILER BUILDING
2	CATALYTIC FILTER HOUSE
3	SO₂ REACTOR
4	FIRST STAGE ECONOMIZER
5	AIR HEATER
6	CONDENSATE HEAT EXCHANGER
7	ACID CONDENSERS
8	ID FANS
9	FD FAN

HOT AIR

FLUE GAS

UPDATE OF PROGRESS FOR PHASE II OF B&W'S ADVANCED

COAL-FIRED LOW-EMISSION BOILER SYSTEM

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Importance of LEBS - Over the past five years, advances in emission control techniques at reduced costs and auxiliary power requirements coupled with significant improvements in steam turbine and cycle design have significantly altered the governing criteria by which advanced technologies have been compared. With these advances, it is clear that pulverized coal technology will continue to be competitive in both cost and performance with other advanced technologies such as Integrated Gasification Combined Cycle (IGCC) or first generation Pressurized Fluidized Bed Combustion (PFBC) technologies for at least the next decade.

In the early 1990's it appeared that if IGCC and PFBC could achieve costs comparable to conventional PC plants, their significantly reduced NO_x and SO_2 emissions would make them more attractive. A comparison of current emission control capabilities shows that all three technologies can already achieve similarly low emissions levels (see *Figure 1*).

Figure 2 shows that by developing technologies that permit utilization of advanced steam conditions in a PC plant, cycle efficiency differences can also be mitigated. In fact, advanced pulverized coal fired plants are already being built in Japan and Europe with ultra-supercritical steam conditions which will achieve net plant efficiencies exceeding 40% (HHV) with very low emissions.

Driven by high fuel costs and low emissions requirements, development has progressed in Japan and Europe ahead of the United States. Present emissions limits (NSPS), fuel costs to the generating company and legislation, such as fuel adjustment clauses, do not currently justify high efficiency or extremely low emissions technologies domestically. However, this is likely to change. In recognition of the increasingly global nature of the power industry and the current lack of market support for advancement of PC technology in markets serviced by domestic suppliers, programs such as DOE's Combustion 2000 LEBS are necessary to prevent domestic technology development from falling too far behind. European and Japanese suppliers who are advancing PC technology driven by the need in their own markets will become domestic competitors with mature advanced PC designs and an experience base unavailable to U.S. suppliers.

In order to maintain relevance, B&W's goals for the LEBS program have been continually refined since the initial work began in 1993. Design performance goals began as no more than 0.2 lb NO_x/MBtu, 0.2 lb SO_2/MBtu and 0.015 lb particulate/MBtu with at least a 38% net plant efficiency (HHV) without increasing cost relative to a conventional NSPS plant. They have evolved over time to the present goals of no more than 0.1 lb NO_x/MBtu, 0.1 lb SO_2/MBtu and 0.005 lb particulate/MBtu with a net plant efficiency of 42% (HHV) while reducing solid wastes and complying with anticipated air toxics regulations at or below the cost of a conventional NSPS plant.

Program Structure - The LEBS program is structured in four Phases to be executed over a seven year period.

Phase I, which is completed, involved system analysis, R&D planning and component definition resulting in a preliminary commercial generating unit design. Phase II provides for pilot and subsystem scale testing to confirm and improve the conceptual designs developed in Phase I. In Phase III designs will be developed for the construction of a Proof-of-Concept (POC) Demonstration Facility. In Phase IV the POC facility will be constructed and operated in order to prove the readiness of the technology for commercial application.

In Phase I B&W organized four teams to focus on design of the major subsystems; NO_x Control, SO_2/Particulate Control, Boiler and Balance of Plant (BOP). As a result of those efforts the importance of controls and sensors to achieving the integration necessary to meet the performance goals was recognized and a fifth subsystem team has been formed for that purpose. Several design options were identified and evaluated by the subsystem teams during Phase I to determine the most economical approach to meet the program goals. The resulting B&W design consists of a boiler which integrates advanced low NO_x combustion and limestone injection dry scrubbing (LIDS) technologies. Figure 3 shows B&W's LEBS Commercial Generating Unit design.

This paper summarizes the status of the work in progress for Phase II of B&W's LEBS program. To better communicate progress, it is presented by subsystems.

NO_x Subsystem - Investigations in Phase I through pilot-scale testing and modeling and at the Combustion Research Facility at the Massachusetts Institute of Technology (MIT) were used to test a promising low-NO_x burner developed by MIT and benchmark it against the state-of-the-art B&W DRB-XCL® burner. While the NO_x emissions from the MIT burner were below the original DOE program goal of 0.2 lb NO_x/MBtu, it was deemed too difficult to develop it to commercial practicality within the time frame of the LEBS program.[1]

In formulating the research, development and testing (RD&T) plan for Phase II, it was decided to pursue development of a new experimental B&W advanced low-NO_x burner which had previously shown improved performance compared to the B&W DRB-XCL® and had been designed with commercialization in mind. A pilot-scale testing campaign for the B&W Advanced Low-NO_x burner was initiated in Phase II to further develop the B&W burner design. Two series of tests have been completed in the Small Boiler Simulator (SBS) Test Facility at B&W's Alliance Research Center and another is scheduled for mid-summer 1995.

The SBS provides a realistic furnace for burner development in which the DRB-XCL® and Advanced Low-NO_x burners had been extensively tested previously. Figure 4 shows a schematic of the SBS configured for PC firing. The water-cooled 5 MBtu/hr furnace has a calculated residence time from the PC burner centerline to the furnace exit of 2 seconds when operating at design capacity. The inside surface of the furnace is insulated to yield a furnace exit gas temperature of 2100 to 2300°F.

The SBS is equipped with an indirect pulverized coal feed system which allows adjustment of the primary air-to-coal ratio. During testing, stack gases were sampled continuously from a location above the convection pass section outlet through a heated sample line, furnace exit gas temperatures were measured by a K-type, high-velocity-thermocouple (HVT) probe and CO, CO_2, O_2, SO_2, and NO_x concentrations were measured in the stack gas, after filtering and drying, and plotted on a Chessell strip chart recorder.

An optical pyrometry system from Diamond Power Specialty Company called FLAMEVIEW™ was used to non-intrusively map the flame temperature two dimensionally via a patented process utilizing two-color pyrometry. Stack fly ash was sampled isokinetically according to an EPA recommended method, using an Anderson Universal Stack Sampler. Cumulative batch samples were obtained by radial traverses and analyzed for carbon utilization.

Initial testing utilized a pilot-scale B&W DRB-XCL® burner, which had previously been tested in the SBS with different coals[2], firing Illinois #6 coal for a benchmark comparison. In addition, extensive field data on the commercial scale DRB-XCL® PC burner[3] was available for comparison. A full complement of data was taken at various loads and excess air levels.

The original overfire air ports on the SBS were tested while firing with the pilot-scale DRB-XCL® burner. Two sets of ports were evaluated and the results were compared to typical field performance. Initial results showed that the lower front wall ports, located at typical residence times, gave good results, however, modifications were needed to improve the air flow direction and velocity. These modifications were incorporated with the third series of testing on the SBS with the Advanced Low-NO$_x$ burner.

The NO$_x$ emissions and unburned carbon (UBC) measurements during benchmark testing were similar to results obtained in the Phase I testing at MIT and previously obtained results firing other coals with the pilot-scale DRB-XCL® burner. Overall, the burner performed as expected based on commercial operation. Stoichiometries of 1.20 to 0.7 were tested with no effect in the stability of the flame and in all cases, the unburned carbon remained below 5%.

Subsequent to benchmark testing with the DRB-XCL® burner, testing of the Advanced Low-NO$_x$ burner began with the burner in a previously tested configuration[4] to establish repeatability and to characterize the burner with Illinois #6 coal. Emission values similar to those previously obtained resulted. Air distribution, excess air, and coal delivery were varied while in this original configuration. All variations produced trends similar to those obtained previously.

Several hardware changes, including various flame stabilizer rings (FSR) and air separation vanes (ASV), were also tested. Again, key parameters were varied and operating data was recorded for each test series. All variations were compared to determine the optimum hardware and settings.

After the optimum hardware (ASV, FSR) and operating settings for the Advanced Low-NO$_x$ burner were determined, further hardware modifications to the air flow distribution and coal delivery system were made. These changes proved to be very promising. NO$_x$ emissions of the Advanced Low-NO$_x$ burner were 10-15% lower than the DRB-XCL® burner benchmark values. UBC and CO emissions were found to be similar with both burners, while the burner pressure increased only slightly with the Advanced Low-NO$_x$ burner.

Since the Advanced Low-NO$_x$ burner was not staged during the second series of pilot-scale testing, field experience was extrapolated. These extrapolations indicate that the NO$_x$ emissions with staging to a burner stoichiometry of 0.85 would fall below the original DOE NO$_x$ emission goal of 0.2 lb NO$_x$/MBtu approaching 0.15 lb NO$_x$/MBtu with combustion only, progressing toward the overall process goal of 0.1 lb NO$_x$/MBtu.

Based on the first two series of pilot-scale testing and the results obtained from numerical modeling of various hardware and operating configurations, further improvements will be made to the Advanced Low-NO$_x$ burner for the third test series. Staged and unstaged versions of the Advanced Low-NO$_x$ burner will be demonstrated in the SBS in preparation for scale-up to 100 MBtu/hr for Subsystem testing scheduled to begin in January 1996. Figure 5 shows the comparison between NO$_x$ emissions and burner configurations tested in the SBS.

SO$_2$/Particulate Control - Figure 3 shows the B&W LIDS system which is a limestone-based furnace injection/dry scrubbing SO$_2$ removal process which cost-effectively integrates three commercially-proven flue gas cleanup technologies: furnace limestone injection, dry scrubbing, and fabric filtration. Sulfur dioxide removal occurs in the boiler, in the dry scrubber, and in the fabric filter.

LIDS reduces cost using a dry scrubber for downstream SO$_2$ removal by permitting the use of limestone as the sorbent (as opposed to the more expensive lime used in most dry scrubbing processes), and reducing the inlet SO$_2$ concentration to the dry scrubber through in-furnace SO$_2$ removal. This latter fact permits the LIDS process to be applied to units firing high-sulfur coals.

In order to prove the predicted performance of this unique system, pilot-scale testing was conducted in B&W's SBS during Phase I. The furnace, dry scrubber, and fabric filter processes were fully-integrated to achieve the SO$_2$ removal goal. During those tests the furnace stoichiometric molar ratio (Ca/S) was 1.4, approach to adiabatic saturation temperature was 10°F, and slurry solids concentration was 40-45%. These conditions were

held relatively constant to obtain data over an extended period of time.

The fully-integrated LIDS system removed 98% of the SO_2 (0.10 lb SO_2/MBtu) based on an average of the data measured during the test series. Achieving the SO_2 emissions goal using the B&W LIDS process required optimal SO_2 removal performance of the entire integrated system. Average values from the individual unit operations included 23% removal in the furnace, 58% in the dry scrubber, and 95% removal in the fabric filter. These values are based on the inlet and outlet SO_2 concentrations of each operation. Figure 6 shows the total SO_2 removal accomplished at the outlet of each piece of the LIDS process.

In addition to sulfur removal, the LIDS system has also proven to be very effective in removing air toxics such as heavy metals, acid gases, and organics. The calcium-based sorbent injected into the furnace which provides numerous reaction temperature windows with high particulate availability for condensation of air toxic species and the low operating temperatures in the dry scrubber and fabric filter are believed to be the primary factors.

Since mercury is a likely target for regulation due to its potential to bioaccumulate in the food chain, screening tests of mercury emissions from the LIDS system were performed by Frontier Geosciences, Inc. to characterize the mercury capture capabilities of the process. Phase I LIDS overall mercury removal results are presented in Table 1. Total mercury removal across both the dry scrubber and fabric filter during Phase I testing consistently averaged 97% across the LIDS system.

LIDS development and demonstration in the SBS continues to further define the process. Where the Phase I LIDS demonstration showed the feasibility of achieving ultra-high SO_2 removal, potential limitations inherent to the basic LIDS system were identified. This led to further pilot-scale testing and enhancement in Phase II. The resulting enhanced LIDS process achieved 98% SO_2 removal (0.10 lb SO_2/MBtu) while eliminating the issues associated with the original LIDS system. Figure 7 shows the total SO_2 removal accomplished at the outlet of each piece of the Phase II LIDS process.

Mercury measurements were repeated to verify the high mercury removal achieved in Phase I. The measurements were once again made in triplicate at the system inlet, dry scrubber outlet, and fabric filter outlet by Frontier Geosciences, Inc. Phase II LIDS overall mercury removal results are presented in Table 2. Total mercury removal averaged 92% across the LIDS system.

Boiler - Investigation of the corrosive conditions that the furnace and superheater metals must endure and identification of potential advanced materials continues in Phase II. Data and ash samples taken from SBS testing are being used in laboratory retort tests to assess the corrosion resistance of several potential advanced materials

Three major areas impacting the boiler design are also being pursued to improve both boiler and cycle efficiencies without increasing cost; the limestone injection system, the airheater and the reheater control method.

In the preliminary design, warm, compressed air taken from the combustion air system was used to convey and inject the limestone into the upper boiler furnace. The conveying system is being re-evaluated utilizing the cool, clean flue gas leaving the fabric filter rather than the combustion air. The new system compresses and preheats the gas and limestone to a significantly higher temperature prior to injecting it into the furnace. Heat and mass balances have been made and are being evaluated. This system is expected to reduce auxiliary power, compressor cost and airheater requirements and permit an increase in economizer feedwater temperature which has been shown to improve steam cycle efficiency.

The regenerative airheater utilized in the preliminary design introduces leakage losses to the cycle and requires power to operate. To improve cycle efficiency, a heat pipe airheater is being evaluated. Initial expectations are reduction in cost, elimination of leakage losses and elimination of the auxiliary power.

Temperature control of the reheaters is critical to achieving cycle performance. A method which can

independently control the heat absorption in each reheater throughout the load range rather than using spray attemperation will result in the highest cycle efficiency. Convection pass and gas path arrangements are being considered which have potential to achieve this result.

Balance of Plant - The net cycle efficiency (HHV) produced in Phase I effort did not approach the present goal of 42%. In addition to improving boiler efficiency, several improvements in the steam cycle and reduction of auxiliary power requirements for balance of plant equipment planned for investigation.

Controls and Sensors - In order to achieve low NO_x production, precise control of the combustion conditions is necessary. Sensors are being developed to provide the ability to measure and control the air to fuel ratio for each burner in a multi-burner system. Coupled with advanced burner design and air staging, on-line control of the individual burner stoichiometry is expected to achieve the minimum possible NO_x levels from the combustion process without adversely affecting UBC. In addition, the control philosophies are being developed to operate the combustion controls required for the advanced low NO_x equipment and LIDS in an integrated manner with the boiler to achieve minimum emissions over the load range.

Future Work - B&W's CEDF will be utilized for the LEBS Phase II Subsystem testing scheduled to commence in October 1995. Preliminary characterization testing was conducted in the winter of 1994-1995 under the LEBS program using the state-of-the-art DRB-XCL® low-NO_x burner.

The CEDF is designed for a heat input of 100 MBtu/hr, and integrates combustion and post-combustion testing capabilities to facilitate the development of the next generation of power generation equipment. The furnace (Figure 8) is designed for testing a single 100 MBtu/hr burner, or multiple wall-fired burner configurations. It has been carefully designed to yield combustion zone temperatures, flow patterns, and residence time representative of commercial boilers. Boiler convection pass and air heater simulators maintain representative conditions through the entire boiler system to facilitate studies of air toxics capture in back-end flue gas clean-up devices. Representative gas phase time-temperature profiles and surface metal temperatures are maintained throughout the convection pass. Convection pass metal temperatures are maintained in the 600-1100°F range by way of a novel double-walled tube design. Following the air heater, the flue gas enters a vertical dry scrubber unit to control sulfur dioxide emissions. The resulting dry by-products are then filtered from the gas by a multi-chamber pulse-jet fabric filter.

Modifications will be made to the existing CEDF during the summer of 1995 to incorporate a LIDS system. Furnace limestone injection, solids recycle, and further instrumentation will be added. The existing dry scrubber, fabric filter and slurry slaking system will be utilized.

Preliminary POC Demonstration Facility Design - In order to satisfy the intent of Proof-of-Concept (POC) testing in a cost-effective manner, B&W has elected to further modify the 100 MBtu/hr CEDF to create the POC Demonstration Facility for Phase IV. A preliminary POC design and RD&T plan have been completed and reported to the DOE. The POC facility, which builds on the modifications made to B&W's CEDF in Phase II for Subsystem testing, will cost drastically less than the original estimates based on using a Utility plant site.

The majority of the modifications to the CEDF will be associated with the NO_x and Controls & Sensors subsystems since most of the equipment for the SO_2/Particulate subsystem will be installed for the Phase II Subsystem testing. The major modifications will include; "hardening" of the systems to permit extended periods of operation, incorporation of advanced controls and sensors integrating control of both the NO_x and SO_2/Particulate subsystems, installation of multiple opposed wall-fired advanced low-NO_x burners and an over-fire air system, and incorporation of two coal feeders to simulate multiple pulverizers.

Conclusion - With the exception of one more series of SBS testing of the Advanced Low-NO_x burner, pilot testing is completed and the design of the Subsystem Test and POC Demonstration Facilities are progress. With the completion of the Subsystem testing schedule to begin in October 1995, B&W expects to further confirm and refine our LEBS design and costs in preparation for proof-of-concept demonstration in Phase IV. With the help

of the LEBS program, B&W intends to develop a commercial PC plant design which is competitive with the other emerging advanced coal-fired technologies in both performance and cost.

Acknowledgements- The authors wish to express thanks to the U.S. Department of Energy's Pittsburgh Energy Technology Center for supporting the B&W LEBS Team's efforts.

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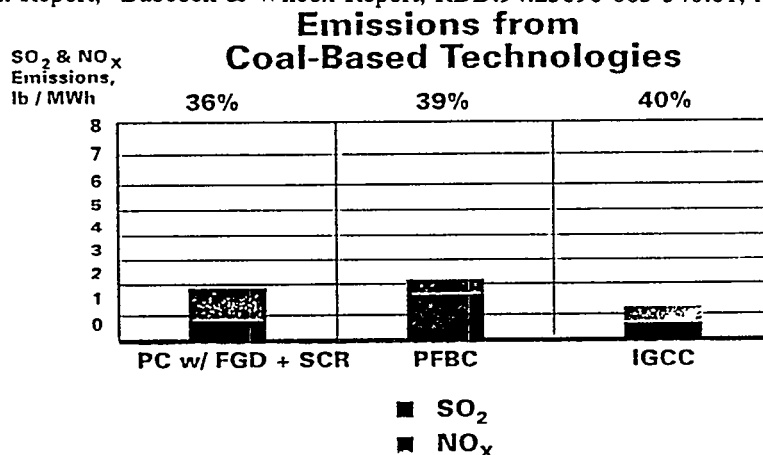
Figure 1 - Current Emissions from Coal Based Technologies.
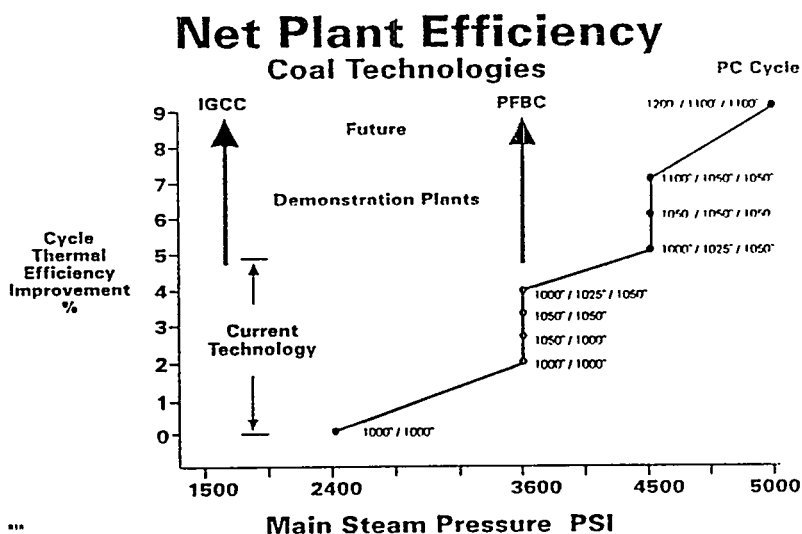
Figure 2 - Net Plant Efficiency

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Figure 4 - B&W Small Boiler Simulator

F: Furnace only
F+DS: Furnace and Dry Scrubber
F+DS+BH: LIDS System

Figure 6 - Phase I LIDS Feasibility Demonstration SO₂ Removal

Figure 3 - LEBS Commercial Generating Unit

Combustion 2000 Pilot-Scale Preliminary Test Results
Firing Illinois #6 Coal

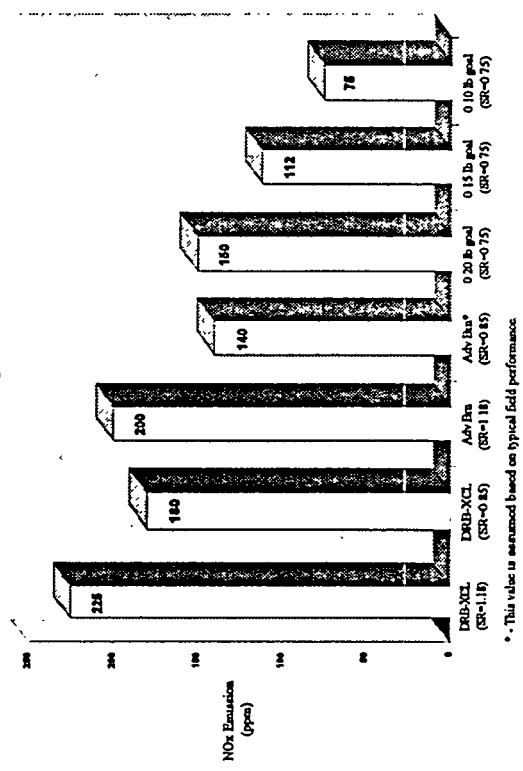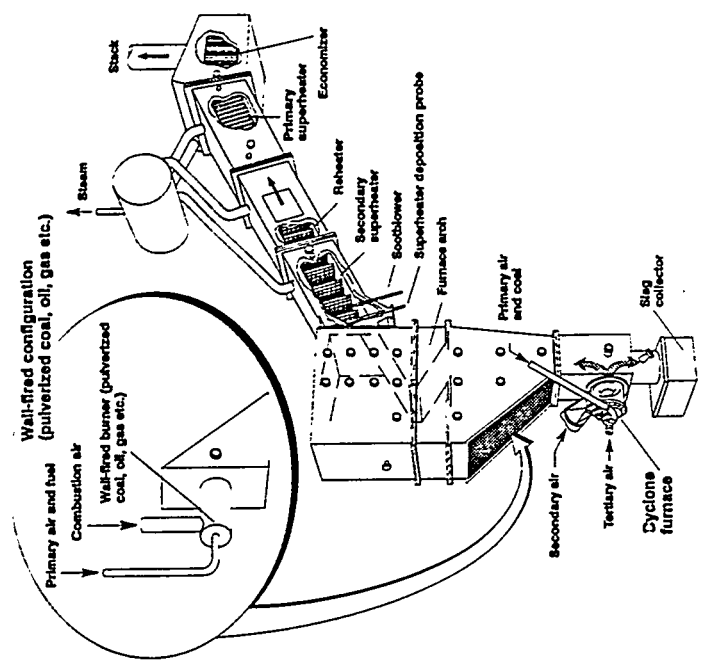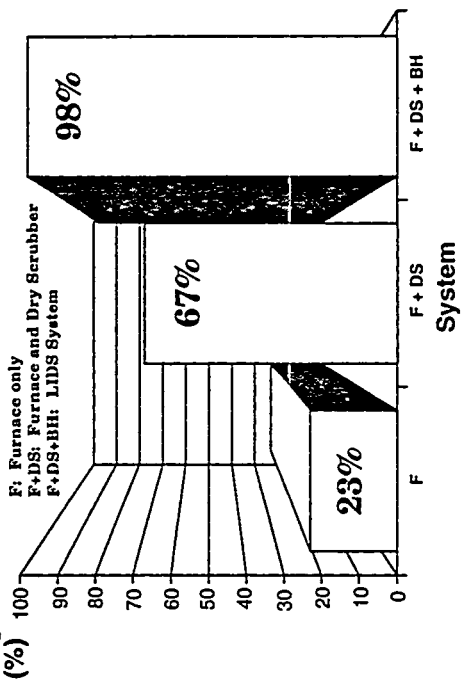Figure 5 - NOₓ Emission versus Burner Configurations Tested in the SBS

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Stream	Total Hg, Ug / Nm3	Hg Removed Across System
System Inlet	19.85	
Inlet Baghouse	4.90	
Outlet Baghouse	2.73	86.25%
System Inlet	16.49	
Inlet Baghouse	3.49	
Outlet Baghouse	1.16	92.97%
System Inlet	17.85	
Inlet Baghouse	3.58	
Outlet Baghouse	0.80	95.52%

Table 2 - Phase II LIDS Total Mercury Removal
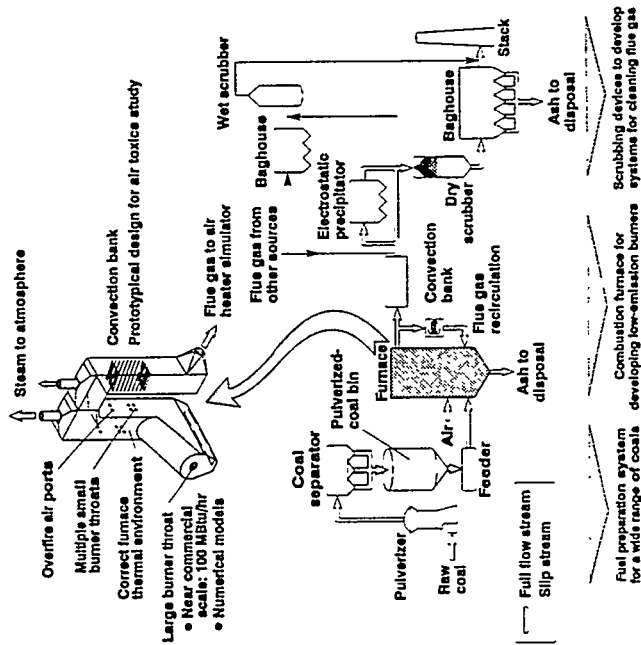
Figure 8 - B&W Clean Environment Development Facility

Stream	Total Hg, Ug / Nm3	Hg Removed Across System
Inlet Scrubber	5.0	
Inlet Baghouse	2.49	
Outlet Baghouse	0.047	99.06%
Inlet Scrubber	4.91	
Inlet Baghouse	2.33	
Outlet Baghouse	0.067	98.64%
Inlet Scrubber	6.19	
Inlet Baghouse	2.17	
Outlet Baghouse	0.408	93.41%

Table 1 - Phase I LIDS Total Mercury Removal

Figure 7 - Phase II LIDS Process Definition SO$_2$ Removal

The following manuscript was unavailable at time of publication.

ENGINEERING DEVELOPMENT OF A LOW-EMISSION BOILER SYSTEM

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DEVELOPMENT OF A HIGH-PERFORMANCE, COAL-FIRED POWER GENERATING

SYSTEM WITH A PYROLYSIS GAS AND CHAR-FIRED

HIGH-TEMPERATURE FURNACE

Jack Shenker
Project Manager
Foster Wheeler Development Corporation
Contract Number: DE-AC22-91PC91154
Period of Performance: March 1992 through April 1995

INTRODUCTION

A high-performance power system (HIPPS) is being developed. This system is a coal-fired, combined-cycle plant that will have an efficiency of at least 47 percent, based on the higher heating value of the fuel. The original emissions goal of the project was for NOx and SOx to each be below 0.15 lb/MMBtu. In the Phase 2 RFP this emissions goal was reduced to 0.06 lb/MMBtu. The ultimate goal of HIPPS is to have an all-coal-fueled system, but initial versions of the system are allowed up to 35 percent heat input from natural gas. Foster Wheeler Development Corporation is currently leading a team effort with AlliedSignal, Bechtel, Foster Wheeler Energy Corporation, Research-Cottrell, TRW and Westinghouse. Previous work on the project was also done by General Electric.

The HIPPS plant will use a High-Temperature Advanced Furnace (HITAF) to achieve combined-cycle operation with coal as the primary fuel. The HITAF is an atmospheric-pressure, pulverized-fuel-fired boiler/air heater. The HITAF is used to heat air for the gas turbine and also to transfer heat to the steam cycle. Its design and functions are very similar to conventional PC boilers. Some important differences, however, arise from the requirements of the combined cycle operation.

Materials considerations limit the air temperature that can be reached in the HITAF. When the effects of temperature and flow imbalances are considered, the upper limit to air temperature is around 1400°F for steel alloy tubes, and 1800°F for ceramic tubes. These limitations require that some fuel be fired in the gas turbine to achieve gas turbine inlet temperatures that are currently over 2300°F. To obtain the most benefits from the HIPPS approach, the Foster Wheeler HIPPS concept uses a pyrolysis process to convert coal into fuel gas and char. Having these two fuels available allows us to push the limits of HIPPS by using the relatively clean fuel gas in critical areas.

HIPPS CYCLES

Two arrangements for greenfields HIPPS plants have been developed. One arrangement uses a ceramic air heater in the HITAF and natural-gas-firing in the gas turbine. This is referred to as the 35 Percent Natural as HIPPS. The other option uses coal-derived fuel gas from the pyrolyzation process as the fuel for the gas turbine. This arrangement requires no natural gas and is referred to as the All Coal HIPPS. A process flow schematic of the 35 Percent Natural Gas HIPPS is shown in Figure 1. In this arrangement the HITAF has two furnaces: one for firing fuel gas, and one for firing char. The fuel gas is fired upstream from a ceramic air heater that is used for the final heating of gas turbine air in the HITAF. This air heater is an AlliedSignal design, and it raises the air temperature from

1400 to 1800°F. The fuel gas is cleaned of alkalies and particulates before it is burned, thereby protecting the ceramic air heater from the effects of alkali corrosion and tube bank fouling.

The char is fired in a separate furnace, and these combustion products transfer heat to the lower-temperature air (1030 to 1400°F) and to steam cycle tube surface. The design of these portions of the HITAF are very much like conventional PC boilers. All of the tubes that are exposed to the char flue gas are steel. A TRW slagging combustor is used for the combustion of char. It will remove 70 to 90 percent of the ash in the form of slag. This type of operation more than compensates for the increased ash content of the char relative to parent coal.

Natural gas is fired in the gas turbine to raise the air temperature to 2350°F entering the gas turbine, and some of the gas turbine exhaust is used as combustion air in the HITAF. The remaining gas turbine exhaust goes through a heat recovery steam generator before going to the stack. Back-end emissions systems will be used in the HITAF exhaust stream as required to meet specific plant requirements.

A process flow diagram for the All Coal HIPPS is shown in Figure 2. In this type of HIPPS plant, the fuel gas from the pyrolyzer is used as fuel for the gas turbine instead of natural gas. This arrangement requires that the pyrolyzer subsystem be run at higher pressure, but it simplifies the HITAF and lowers fuel costs.

In the 35 Percent Natural Gas HIPPS, it is necessary to heat the gas turbine air to 1800°F in the HITAF to limit the natural gas heat input to 35 percent of the plant heat input. This limit on natural gas consumption is a project requirement, and it forced the use of a ceramic air heater. If coal-derived fuel gas is used as the gas turbine topping combustion fuel, it is no longer necessary to push the HITAF air outlet temperature that high. The air temperature leaving the HITAF can be 1400°F which eliminates one furnace and the ceramic air heater. The entire HITAF can then be fabricated with standard, proven materials of construction.

The remaining systems of the All Coal HIPPS are similar to the 35 Percent Natural Gas HIPPS. A TRW slagging combustor is also used for char combustion, and backend emissions systems will be used as necessary to meet specific plant emissions requirements.

SYSTEM DEVELOPMENT

As part of the Phase 1 effort, engineering analysis and small-scale research have been done on HIPPS plant designs. Pilot plant testing would be part of a Phase 2 project. Some areas that have been investigated in Phase 1 follow:

Char Combustion - Char generated under conditions that would be typical of HIPPS operation was obtained and characterized. This characterization included standard laboratory tests and flat flame burner tests that were run at Brigham Young University (BYU). The results of these tests were used by TRW in the computer modeling of the slagging combustors.

The char test results were encouraging, both from the aspect of its suitability as a fuel and from the ability to adapt current combustor models for use with char. Thermogravimetric tests (TGA) showed that the weight loss for char started around the same temperature as for typical anthracite coals. This is a higher temperature than the parent Pittsburgh No. 8 coal. However, the rate of weight loss at this point was similar to that of the parent coal and much greater than anthracite coals.

In the flat flame burner tests, paticles of coal and char were passed through a flame of controlled temperature and composition. These particles were collected after different residence times to check carbon burnout, sulfur release and nitrogen release. A summary of some of the results is shown in Table 1. It can be seen that the characteristics of the char combustion are very similar to those of the parent coal combustion after the volatiles are consumed. Apparently the pyrolyzation process has an effect on the coal that is similar to the initial stages of combustion. This situation is beneficial because it will minimize the model modifications and allow the use of existing data on various coals.

Fuel Gas Combustion - Fuel gas combustion will be somewhat different in the 35 Percent Natural Gas HIPPS and the All Coal HIPPS. In the All Coal HIPPS, the fuel gas will be burned in the gas turbine topping combustor. Westinghouse has done considerable testing firing this fuel with there multi-annular swirl burner (MASB) arrangement. This combustor has been specifically designed for this fuel, and test results have shown that less·than 30 percent of the ammonia in the fuel gas will become NOx [1]. The All Coal HIPPS has conditions very similar to the MASB test conditions, so we are confident that it can be applied.

In the 35 Percent Natural Gas HIPPS, the fuel gas is fired in an atmospheric pressure furnace. Tests were run to determine the characteristics of this type of combustion with a typical industrial burner. T-Thermal Corporation conducted tests with a 3MM Btu/h vortex burner. Fuel gas and the vitiated combustion air were syntesized and heated to the anticipated burner inlet temperatures. The ammonia content of the fuel gas was varied to determine the effect of ammonia on NOx generation. The burner was fired into a refractory lined furnace, and flue gas temperatures and composition were measured.

Combustion under the HIPPS conditions was stable and complete. The low-Btu fuel and relatively low oxygen content of the combustion air (19 wt%) did not present any combustion problems. Thermal NOx generation was also low (0.04 lb/MM Btu), as evidenced by runs with no ammonia in the fuel gas. There was concern that the elevated temperatures of the fuel and combustion air might increase thermal NOx. This did not turn out to be the case. The situation with the conversion of ammonia to NOx was not as good. At the anticipated ammonia content of 0.16 wt%, about 75 percent of the ammonia was converted to NOx.

This situation will require either some burner development or the use of Selective Catalytic Reduction (SCR). Although there are commercially available low-NOx burners, these burners are designed for low thermal NOx. A burner similar to the MASB could probably be developed for furnaces, but at this point we do not feel that this is necessary. Even with the ammonia conversions measured, the HIPPS NOx goals can be met with the application of proven SCR technology.

Gas Turbine - Both of our HIPPS arrangements and those of competing concepts require that gas turbine compressor discharge air be taken off the machine, heated and then brought back for topping combustion. As part of this project, General Electric has developed a preliminary design to achieve this type of operation with their Model 7FA gas turbine. The design is based on the 35 Percent Natural Gas HIPPS conditions, which include an air return temperature of 1800°F.

The basic GE design is shown in Figure 3. There are two torus-shaped headers. The inner header collects the compressor discharge air after it is used as cooling air for the combustors. This air then goes to the HITAF, where it is heated to 1800°F. The air then returns to the outer torus from which it is distributed to the 14 combustors.

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As previously mentioned, Westinghouse has also developed a system that incorporates the necessary HIPPS functions. Their system was based on fuel gas combustion and the lower air return temperatures that are more typical of our All Coal HIPPS. The Westinghouse 501F gas turbine is being used as the base-case turbine for the All Coal HIPPS conceptual plat design. It is also likely that the GE design could be adapted for the application, but the necessary analyses have not yet been done.

HITAF Design - The design of the HITAF for the 35 Percent Natural-Gas HIPPS has been previously reported [2]. The HITAF for the All Coal HIPPS is shown in Figure 4. The HITAF is designed to be very similar to a conventional PC boiler. In this manner we can take advantage of existing design, fabrication and operating experience.

There are a few areas where the HITAF design deviates a little from standard boiler designs. Three walls of the furnace will be refractory-coated, primary superheater tubes. This is done to improve the thermodynamic efficiency of the system. Refractory lining of furnace evaporator tubes has been done in the past, and on a few occasions superheater technology, and the use of the TRW slagging combustor will improve the situation by lowering furnace heat fluxes and ash loadings.

With an air outlet temperature of 1400°F, the tube metal temperature will be around 1550°F. Several alloys have established allowable stresses for this temperature range. Tests would be run in a Phase 2 effort to determine which alloys have the best corrosion properties for the air heater.

Summary - The FWDC team HIPPS concept can be applied in two basic arrangements. One arrangement uses a ceramic air heater to get the maximum indirect heating of air for the gas turbine. Natural gas is then fired to achieve the required inlet gas temperature. Our ceramic air heater arrangement has benefit of operation with a relatively clean fuel, because the coal is converted to fuel gas and char. The cleaned fuel gas is used as fuel for the ceramic air heater, thereby reducing the effects of corrosion ad ash deposition.

The second arrangement of our HIPPS concept limits the indirect heating of the air to temperatures that can be achieved with steel alloy tubes. With this concept, the coal is also converted to fuel gas and char, but only the char is fired in the HITAF. The fuel gas is used as fuel for the gas turbine topping combustor. This arrangement of HIPPS will allow achievement of the ultimate program goal of a totally coal-fueled HIPPS within the scheduled time frame of the program.

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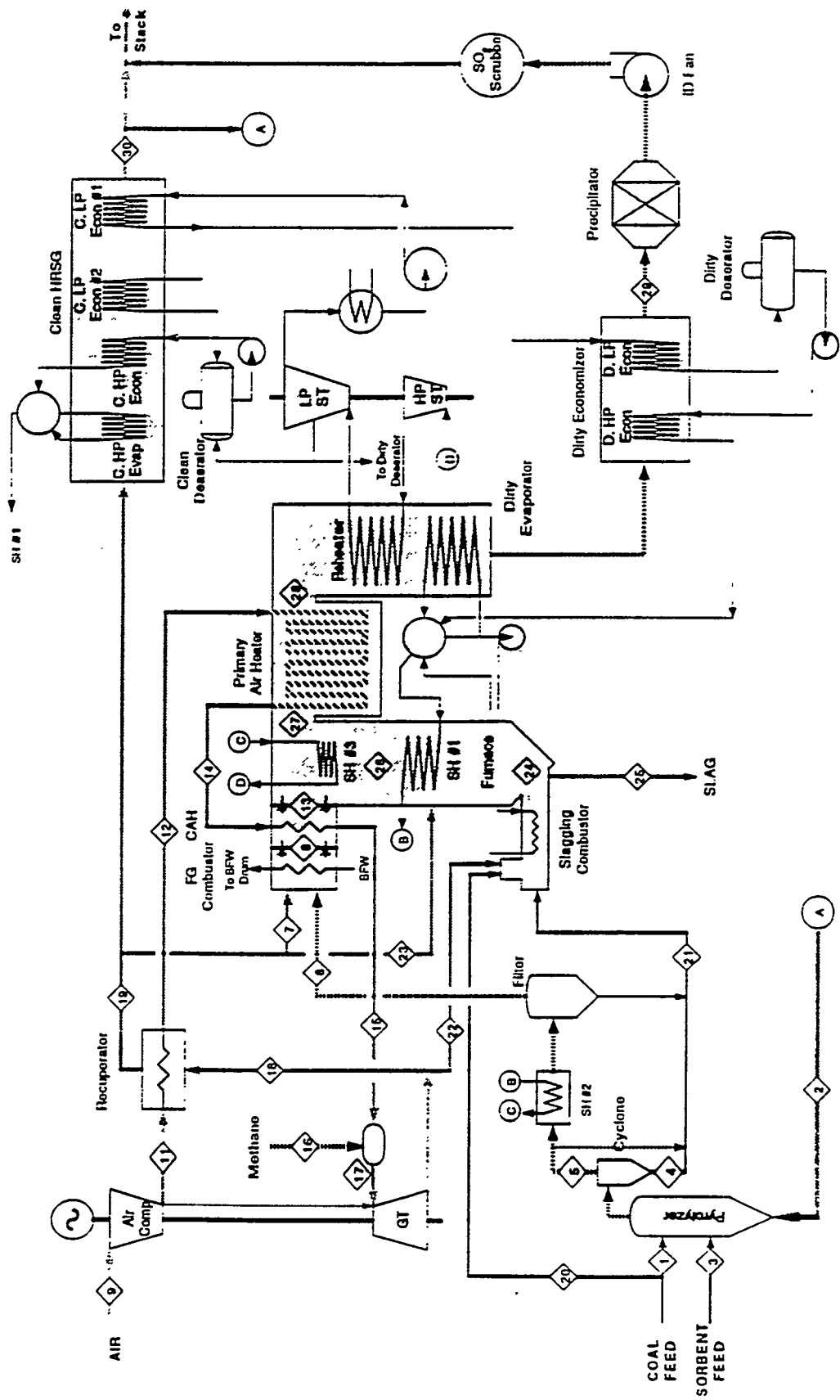
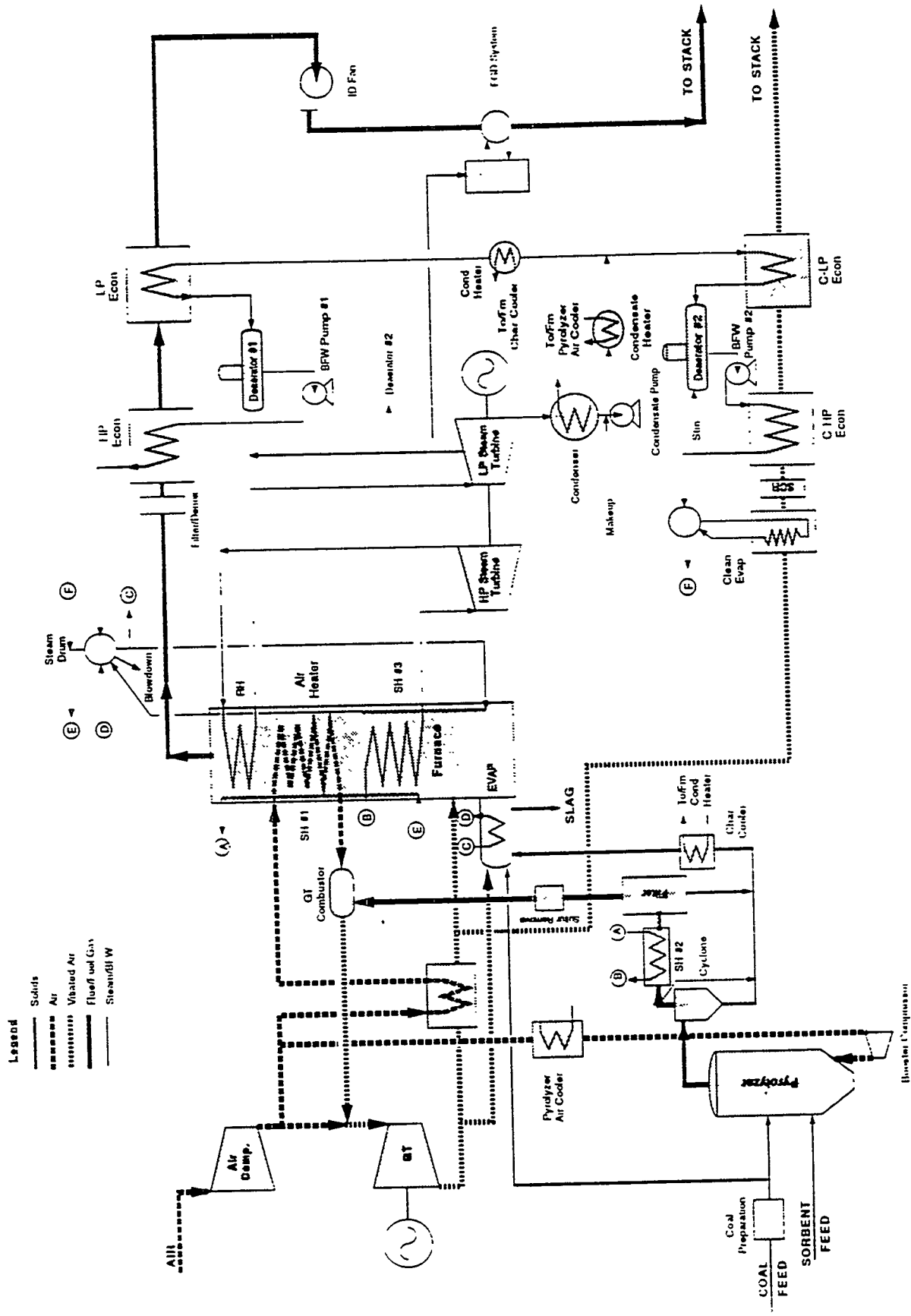Figure 1 - 35 Percent Natural Gas HIPPS Process Flow Diagram

Figure 2 - All Coal HIPPS Process Flow Diagram

Table 1 - Char Burnout Results (1800 K/5% O_2)

Pittsburgh No. 8	Sampling Distance, in.	Average Burnout (% DAF)		Average Apparent Density, g/cc	Surface Area	
		Volatiles Included	Char Only		N_2, m²/g	CO_2, m²/g
Coal	2	50.4	0	0.38	77	241
Coal	4	58.6	14.1	0.37	44	241
Coal	6	60.5	29.6	0.42	37	188
Coal	8	63.5	30.3	0.42	33	134
Char	2	N/A	10.6	0.70	31	146
Char	4	N/A	20.4	0.65	28	149
Char	6	N/A	29.2	0.67	33	135
Char	8	N/A	34.7	0.68	31	101
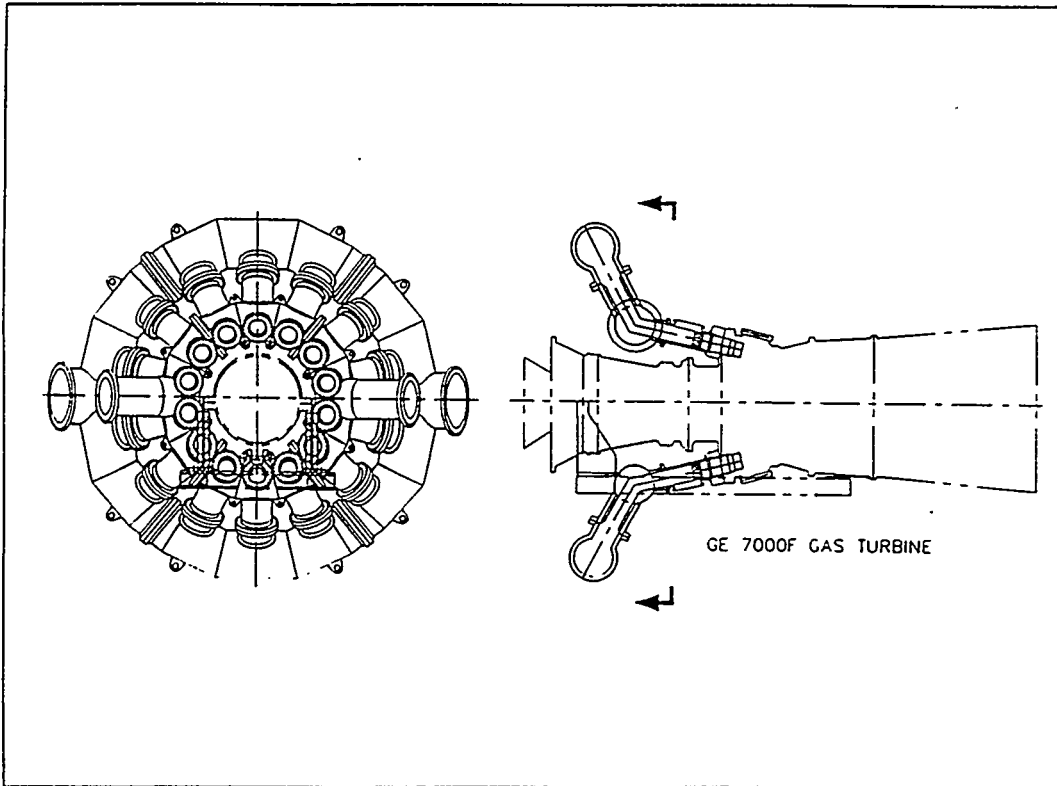
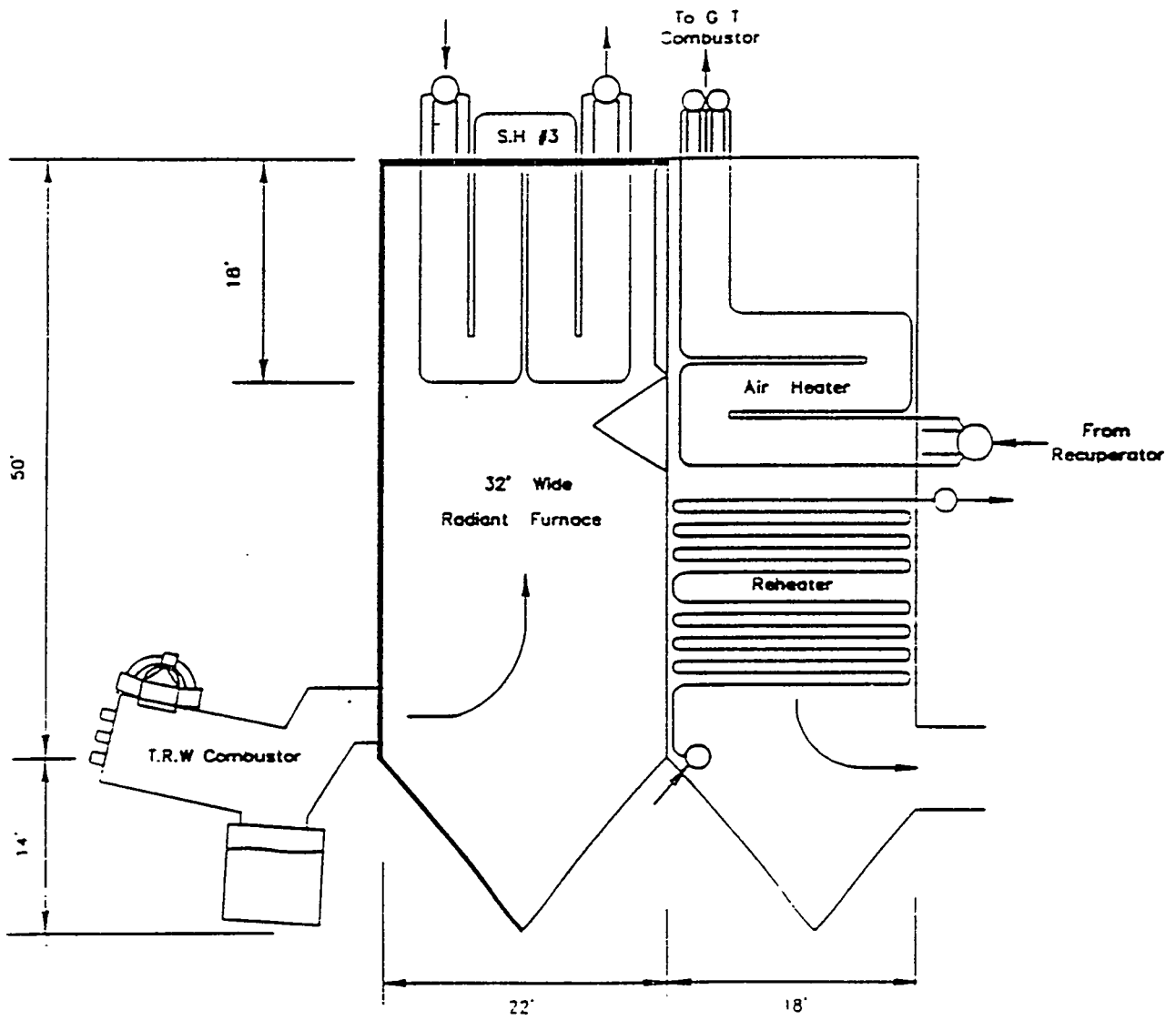Figure 3 - GE HIPPS Gas Turbine Piping

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Figure 4 - HITAF for All Coal HIPPS

The following manuscript was unavailable at time of publication.

THE PHYSICS OF COAL LIQUID SLURRY ATOMIZATION

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FUNDAMENTAL STUDY OF ASH FORMATION AND DEPOSITION: EFFECT OF REDUCING STOICHIOMETRY

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1. Objectives and Approach

This project is designed to examine the effects of combustion stoichiometry on the fundamental aspects of ash formation and ash deposit initiation. Emphasis is being placed on reducing stoichiometries associated with low-NOx combustion, although a range of oxidant/fuel ratios are being considered. Previous work has demonstrated that ash formation depends strongly upon coal mineralogy, including mineral type, size, amount, and the presence of organically associated inorganic species. Combustion temperature and the oxidation state of iron also play a significant role. As these latter items will vary with changes in stoichiometry, research to determine the net effect on deposition is required.

To achieve these goals, a research program with the following technical objectives is being pursued: 1) identify the partitioning of inorganic coal constituents among vapor, submicron fume, and fly ash products generated from the combustion of pulverized coal under a variety of combustion stoichiometries, 2) identify and quantify the fundamental processes by which the transformation of minerals and organically-associated inorganic species occurs. Identify any differences from standard excess air pulverized coal combustion conditions, 3) modify, to incorporate the effects of combustion stoichiometry, an Engineering Model for Ash Formation. The model is capable of predicting the size and chemical composition distributions of the final ash products under standard pulverized coal combustion conditions of 20% excess air. These modifications will extend the model to include phenomena that may be dominant under a broad range of stoichiometries.

Experiments, sample analyses, and modeling are being conducted at several facilities as part of this program. Detailed coal and ash sample analysis using Mössbauer spectroscopy, X-ray absorption fine structure spectroscopy (XAFS), and computer controlled scanning electron microscopy are being carried out at the University of Kentucky (UKy). Small-scale drop tube combustion tests using size and density classified coal samples are being carried out at the

Massachusetts Institute of Technology (MIT) to determine the extent of mineral coalescence and inorganic vaporization as a function of combustion stoichiometry. Combustion experiments utilizing utility grind coals are being conducted at Physical Sciences Inc. (PSI) to examine the effects of stoichiometry on mineral interactions. Deposition experiments using ash generated from combustion experiments and using pure minerals are also being conducted to investigate deposit initiation as a function of combustion conditions. PSI's Engineering Model for Ash Formation (EMAF) is being modified to include effects of combustion stoichiometry as part of this effort. Self-sustained combustion experiments are being conducted in the University of Arizona (UA) 100,000 Btu/h facility to address issues of scaling in combustion processes. The interaction of iron with aluminosilicates as a function of changing combustion conditions will be the focus of this effort. Modeling of iron-aluminosilicate interactions is being conducted as part of several tasks.

2. Coal Selection and Characterization

Four U.S. coals and one U.K. coal have been studied during this program: 1) Black Thunder sub-bituminous coal from the Powder River Basin, 2) a run-of-mine Pittsburgh #8 bituminous coal from the Appalachian Basin, 3) a washed commercial version of a Pittsburgh #8 coal obtained from DOE/PETC, and 4) a physically beneficiated product produced from Pittsburgh #8 coal, 5) Silverdale bituminous coal from the U.K., obtained as part of a collaborative effort with a program led by the utility Powergen. Coal and mineral analyses have been provided in previous reports.

3. Effect of Combustion Stoichiometric Ratio on Ash Formation

As a major element of this research program, the effect of combustion stoichiometric ratio on ash particle composition and size distributions is being examined. To date, work in laminar flow drop tube furnaces has been completed; work in the 100,000 Btu/h self-sustained combustor at the University of Arizona is ongoing. In the drop tube experiments conducted at PSI and MIT, the formation of chemically complex ash particles was found to be dependent upon combustion conditions. Less extensive formation of chemically complex phases was observed under oxygen lean conditions. Relative to the coal, however, significant amounts of chemically complex mixtures were observed under all combustion conditions. Differences may therefore simply be related to differences in stage of combustion. This trend is most clearly seen in ash generated from combustion of beneficiated Pittsburgh #8 coal. In the coal, iron-free aluminosilicate minerals were abundant, with no evidence for iron-aluminosilicate particles (Figure 1a). Considering particles that are rich in iron, aluminum, and silicon (particles that are comprised of greater than 80 mole percent Fe+Al+Si on an oxygen free basis), formation of iron aluminosilicates was observed at stoichiometric ratios of 0.6, suggesting that iron interacted with aluminosilicate minerals early in the combustion process when the residual carbon content was high. More extensive iron aluminosilicate formation, as evidenced by the broader mixture of compositions present in a ternary diagram (Figure 1), was observed at a stoichiometric ratio of 0.9. Ash generated during combustion of the coal at SR = 1.2 showed the broadest mixture of Fe-aluminosilicate compositions and in turn the highest concentration of Fe-Al-Si particles, as determined by CCSEM analysis at the University of Kentucky (Table 1). For comparison,
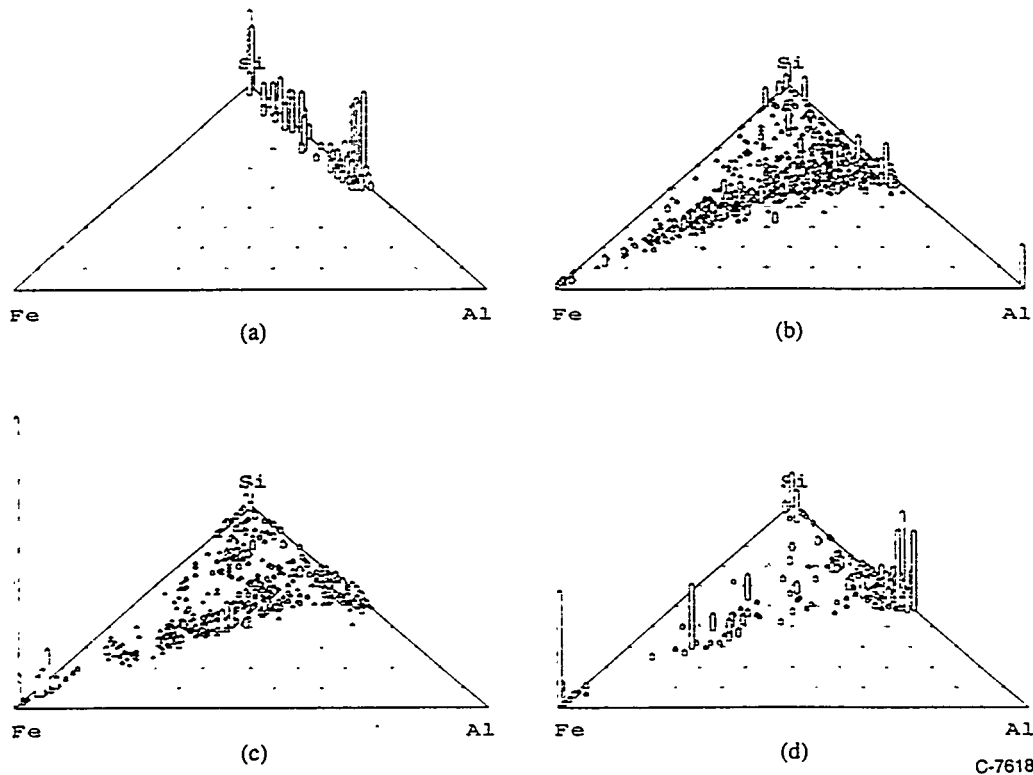
Figure 1. Fe-Al-Si ternary diagram for ash generated from combustion of a beneficiated Pittsburgh #8 coal in the PSIT drop tube reactor. Wall temperature = 1500°C, residence time = 2.6 s. Each point represents an ash composition detected by CCSEM; the height of each peak represents the normalized column of ash particles with the indicated composition. (a) coal minerals (b) ash generated at SR=1.2 (c) ash generated at SR = 0.9 (d) ash generated at SR = 0.6.

Table 1. CCSEM analysis of laboratory-generated combustion ash (volume percent) beneficiated Pittsburgh #8 coal; $T_{wall} = 1500^{oC}$, $t_{res} = 2.6$ s.

Ash composition	SR = 0.6	SR = 0.9	SR = 1.2
Si	7.3	9.1	5.3
Si + Al	6.8	5.5	12.5
Si + Al + Fe	30.2	52.2	59.6
Fe + S	3.8	0.7	0.0
Fe + Si + S	5.9	4.3	1.2
Fe	0.6	1.2	0.7

Table 2. CCSEM analysis of laboratory-generated combustion ash (volume percent) Silverdale coal; $T_{wall} = 1500^{oC}$, $t_{res} = 2.6$ s.

Ash composition	SR = 0.6	SR = 0.9	SR = 1.2
Si	6.6	8.0	10.7
Si + Al	8.3	4.9	7.1
Si + Al + Fe	25.7	32.2	34.5
Fe + S	6.1	2.8	0.6
Fe + Si + S	4.2	1.4	1.8
Fe	0.0	4.2	3.8

results for Silverdale bituminous coal are presented in Table 2. Although the percentage of ash that is Fe+Al+Si rich increased with increasing SR, the changes were small relative to those observed for the beneficiated Pittsburgh #8. Very little change with SR was observed for Silverdale coal.

The effect of carbon in the ash on the "stickiness" or adhesion efficiency of inertially impacting ash particles has also been examined. At the exit of the PSIT drop tube furnace, a ceramic converging section is inserted. This section contains uncooled ceramic tubes that are placed orthogonal to the flow direction and act as inertial impaction and collection devices. In experiments with the three Pittsburgh #8 bituminous coals, the collection efficiency (fraction collected / fraction passing through projected tube cross section) increased with increasing
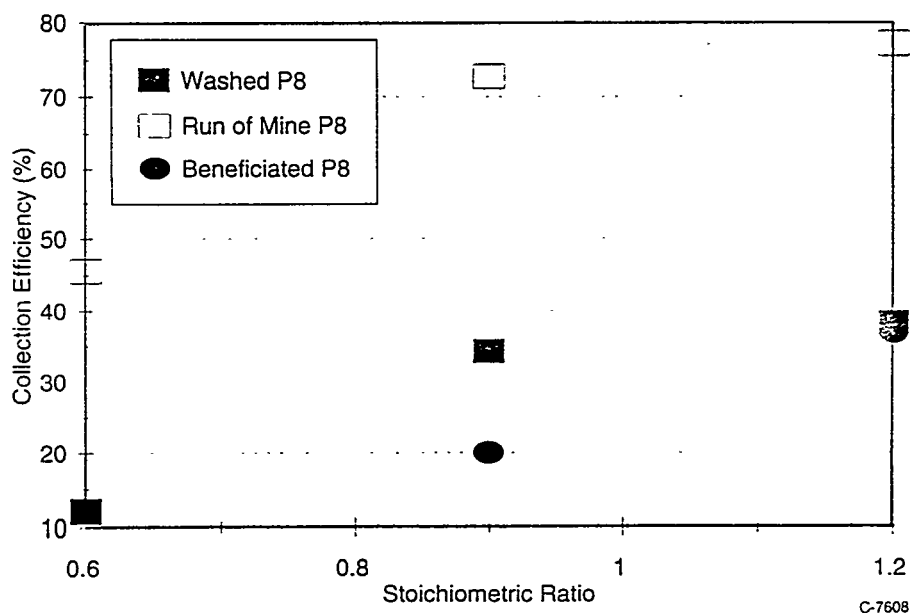

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Figure 2. Ash collection efficiency for three Pittsburgh #8 bituminous coals as a function of combustion stoichiometric ratio. PSIT reactor, 2.6 s residence time, reactor wall temperature of 1500°C.

combustion stoichiometric ratio (Figure 2). This is a result of the decreasing carbon content in the ash as oxygen concentration is increased, as shown in Figure 3. In this figure, the curves represent calculated values assuming that collection efficiency under conditions of residual carbon present in the ash can be expressed as the product of X_c, 1-fraction carbon remaining, times $\eta_{\text{"pure" ash}}$, the collection efficiency under conditions of minimum carbon present in the ash,

$$\eta_{\text{effective}} = \eta_{\text{"pure" ash}} \, X_c.$$
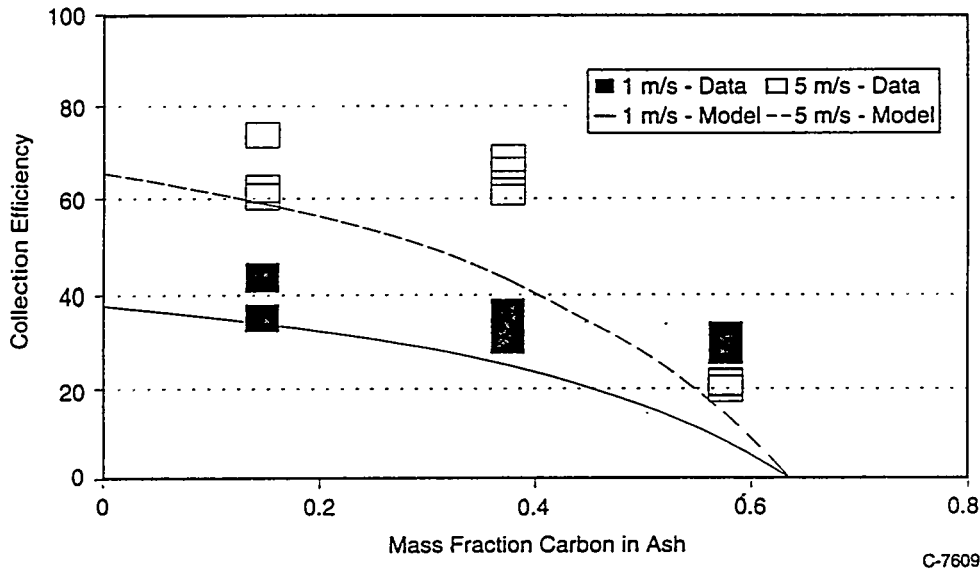

Figure 3. Ash collection efficiency for ash derived from combustion of Silverdale bituminous coal. Note that data are plotted v. the mass fraction of carbon present in the ash.

4. Effect of Oxygen Partial Pressure on Formation of Iron-containing Ash Particles

At MIT, a series of combustion experiments under varying oxygen partial pressure have been conducted with a sieved 63/75 μm sample of the washed Pittsburgh #8 coal. Ash samples collected under p_{O2} equal to 0, 0.10, 0.30, 0.60, and 1.00 (wall temperature = 1477°C in all cases) were then analyzed by Mössbauer spectroscopy at the University of Kentucky for forms-of-iron. Results are compared with the iron phase distribution in the sieved parent coal and in the bulk ash (so-called ASTM ash) in Figure 4. As oxygen partial pressure is increased, the percentage of iron in glassy phases first increases (0 to 30% oxygen) and then remains relatively constant (30 to 100% oxygen). The ratio (Fe^3/Fe^{+2}) in glass, important as a measure of ash particle stickiness, continues to increase with increasing SR, however. This suggests the existence of a threshold temperature below which iron-glass interactions will be less than the theoretically achievable level, based upon mineral distributions in the parent coal.

Mössbauer analysis of ash samples generated during combustion of the program coals in the PSIT drop tube furnace showed similar trends. As seen in Table 3, the ratio (Fe^{+3}/Fe^{+2}) for iron in glassy phases increased with increasing stoichiometric ratio for all coals. For these experiments, note that the total fraction of iron present in the glassy phases remained relatively unchanged with changing stoichiometric ratio. The lower values of (Fe^{+3}/Fe^{+2}) in glass relative

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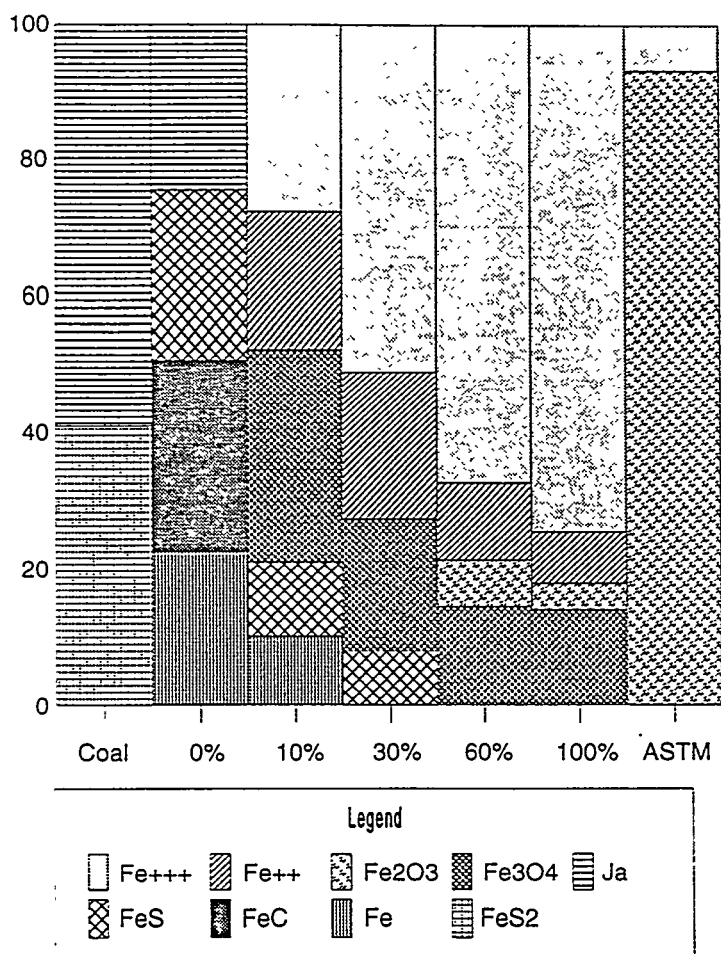
Figure 4. Iron speciation (percentage of iron in each phase) for ash samples generated from combustion of 63/75 µm washed Pittsburgh #8 bituminous coal at different oxygen partial pressures in the MIT drop tube furnace. Wall temperature equal to 1477°C for all experiments.

to the MIT data (Figure 4) are believed due to the lower particle temperatures and more dilute oxygen levels encountered in the PSIT reactor.

5. Engineering Model for Ash Formation

The Engineering Model for Ash Formation (EMAF) is designed to predict the ash composition and size distribution of a given coal based on its coal particle size and mineralogy. This model was originally designed to make these predictions based on complete combustion (e.g. super-stoichiometric conditions). To model the ash size distribution and composition for combustion under reducing conditions it was necessary to modify the model to account for the presence of unburned char. This modification was accomplished by developing a kinetic submodel, based on information in the literature, to predict the fraction of unburned char in each size range. This submodel has been discussed in program Quarterly Reports.

The output of the kinetic sub-model is used to define the fraction of minerals in each simulated coal particle that is **not** exposed during the combustion process. These minerals will generally maintain the same size and composition, as they cannot interact with other minerals. The remaining minerals are assumed to coalesce and interact with other minerals - similar to the original model. Based on these mechanisms, the model predicts the size and composition distributions of the ash that would be measured by CCSEM. Figures 5 and 6 illustrate the predicted changes in ash size and composition as a function of stoichiometric ratio for the ROM Pittsburgh No. 8. On one extreme is the one ash per mineral case, which would match those conditions where very little char burnout has taken place, such as low residence times or stoichiometric ratios. The other extreme is the completely burned out case (SR=1.2). The model predicts a smooth transition of ash mass to the larger sizes with increasing stoichiometric ratio - indicating an increase in ash coalescence with stoichiometric ratio. This increase in coalescence is also evident in the composition predictions (Figure 6). The major ash particle categories (reported on an oxygen

Table 3.

Iron distribution in ash generated from combustion of program coals at PSIT.
$T_{wall} = 1500°C$, $t_{res} = 2.6$ seconds

	SR	Fe_xS	Fe^{+2}	magnetite	Fe^{+2} glass	Fe^{+3} glass
run-of-mine Pittsburgh #8	0.6	19	0	0	81	0
	0.9	8	0	23	51	19
	1.2	0	0	22	58	20
washed Pittsburgh #8	0.6	30	25	13	32	0
	0.9	10	25	29	36	0
	1.2	6	24	36	25	9
beneficiated Pittsburgh #8	0.6	18	0	0	82	0
	0.9	4	0	11	64	21
	1.2	0	0	15	41	44
Silverdale	0.6	37	0	0	63	0
	0.9	13	0	16	71	0
	1.2	0	0	44	39	17
Black Thunder	0.6	0	22	0	24	54
	0.9	4	17	0	11	68
	1.2	0	13	0	7	80

and hydrogen-free basis) are initially Si Al K, Si, Si Al, and lesser amount of Si Al Fe and Si Al Ca. As these ash particles coalesce, the amount of Si (quartz) and Si Al K (illite) decreases, while the amount of Si Al increases. This is due to the product of Si and Si Al K mixing having a lower K content and therefore being binned in the Si Al category. There is also evidence of increased Fe - silicate coalescence as a function of increasing stoichiometric ratio (burnout).

6.   Plans

In the remaining months of the program, two major tasks are to be completed. These are (1) conclusion of experiments in the UA self-sustained combustor, and (2) completion of the analysis of XAFS data collected by UKy during examination of Silverdale ash and deposit samples.
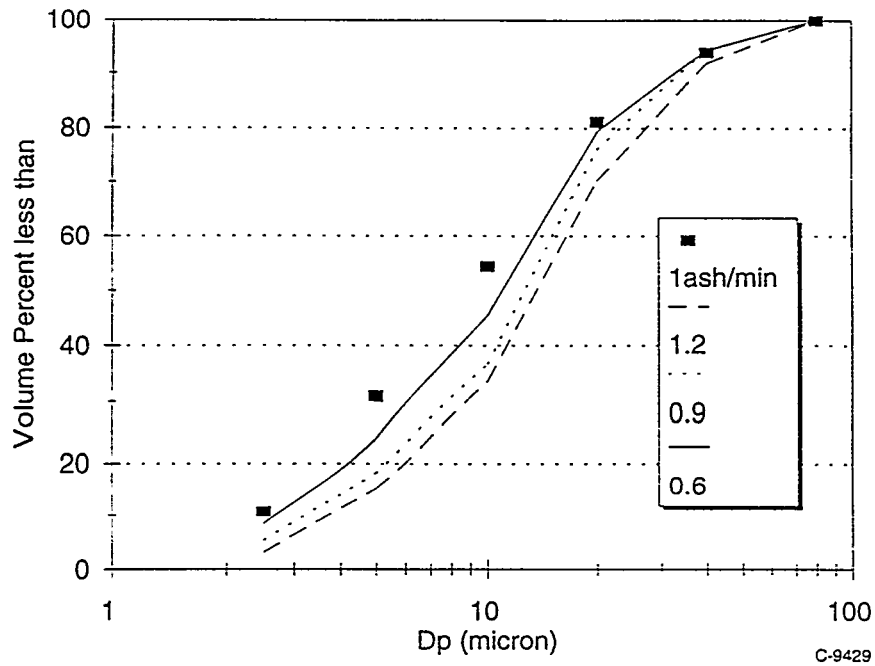
Figure 5. Predicted ash PSD as a function of stoichiometric ratio. Mineral size distribution (1 ash/min) shown for comparison. Coal: ROM Pittsburgh No. 8.
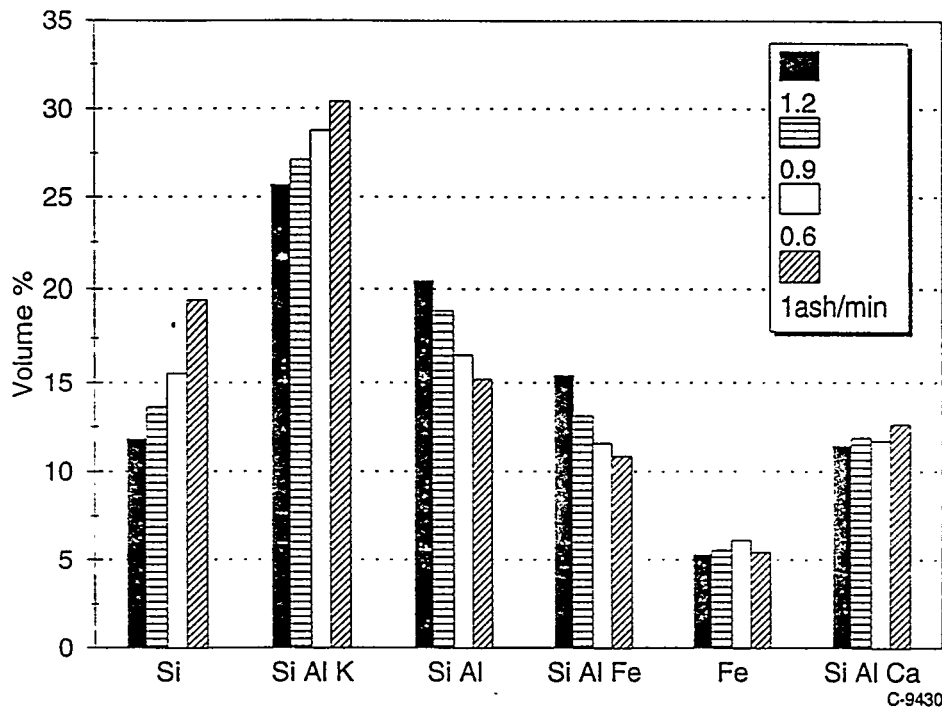

Figure 6. Model prediction for ash compostion - ROM Pittsburgh No. 8.

DEPOSIT GROWTH AND PROPERTY DEVELOPMENT

IN COAL-FIRED FURNACES

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PROJECT OBJECTIVES AND DESCRIPTION

The objectives of this research project are: (1) to provide a self-consistent database of simultaneously measured, time-resolved ash deposit properties in well-controlled and well-defined environments and (2) to provide analytical expressions that relate deposit composition and structure to deposit properties of immediate relevance to PETC's Combustion 2000 program. This project is distinguished from related work being done elsewhere by: (1) the development and deployment of *in situ* diagnostics to monitor deposit properties, including heat transfer coefficients, porosity, emissivity, tenacity, strength, density, and viscosity; (2) the time resolution of such properties during deposit growth; (3) simultaneous measurement of structural and composition properties; (4) development of algorithms from a self-consistent, simultaneously measured database that includes the interdependence of properties; and (5) application of the results to technologically relevant environments such as those being planned under the Combustion 2000 program.

Work completed during FY94 emphasized diagnostic development. During FY95, this development work will be completed and we will emphasize application of the diagnostics to meet the other project objectives. Included in this work are the development and application of two *in situ*, real-time diagnostic systems for monitoring the properties of inorganic materials on Heat transfer surfaces and in the gas-phase during controlled combustion of selected coal samples in Sandia's Multifuel Combustor (MFC). Also, several diagnostics are being incorporated into the MFC that will eventually be used to characterize ash deposit properties.

The project comprises six subtasks, as follows:

Subtask 2.1   Diagnostics for Coal Combustion Environments

The objective of this subtask is to develop and demonstrate diagnostics capable of *in situ* measurement of: (1) condensed-phase species on surfaces in combustion environments, and (2) inorganic vapors in turbulent, particle-laden, combustion gases.

Subtask 2.2 Experimental Determination of Transport, Thermal, and Structural Properties of Ash Deposits

The objective of this subtask is to provide self-consistent, simultaneous measurement of ash deposit properties under combustion conditions that simulate commercial-scale environments such as those expected to occur in Combustion 2000 technologies. Properties to be measured simultaneously and in real time include thermal conductivity, emissivity, porosity, mass and

volume rate of growth, surface composition, and tenacity. Additional properties to be measured, but not in real time or *in situ*, include bulk elemental composition, bulk species composition, shear strength, true density, and detailed morphology.

Subtask 2.3   Analysis of Deposit Properties

The objective of this subtask is to provide an analytical capability for describing the development of deposit properties in combustion systems. Deposit properties to be predicted by this analytical method are similar to those discussed in Subtask 2.2 and include: (1) thermal conductivity, (2) emissivity, (3) porosity, (4) mass and volume rate of growth, (5) surface composition, (6) tenacity, (7) bulk elemental composition, and (8) major species composition.

Subtask 2.4   Chemical Reactions in Deposits

The objective of this subtask is to determine rates and mechanisms that describe chemical reactions in coal ash deposits that alter their properties or their morphology. This subtask is more limited in scope than the previous subtasks. We do not intend to conduct a comprehensive study of inorganic chemistry as it relates to ash deposits. We do intend to review available literature and perform calculations that allow us to capture the first-order terms that describe changes in deposit chemistry with time. This work will result in usable results, but is limited to global kinetics and simplified chemical mechanisms that outline the nature of the reactions and their dependence on operating parameters. The details of the kinetics will not be determined.

Subtask 2.5   Application to Combustion 2000 Program

The objective of this subtask is to exchange technology developed under other subtasks with ongoing Combustion 2000 efforts by other PETC contractors.

Subtask 2.6   Documentation

The objective of this subtask is to provide timely and accurate documentation of project progress, major milestones, and publishable results.

BACKGROUND

Mineral matter in coal is cited as "the nemesis of coal-burning industries" and as playing "the dominant role in fuel selection, in setting the design and size of the furnace, and in establishing how that boiler furnace will be operated" (Raask, 1985). However, the fate of mineral matter during pulverized coal combustion is relatively poorly understood, despite this importance to coal conversion systems. Significant progress in past years has been made through large measure because of PETC-sponsored research describing transformations of inorganic material during combustion. This new research project focuses on ash deposit formation and the development of deposit properties.

This research project supports the overall DOE mission of nurturing the effective and efficient use of national energy resources. It also supports the specific objective of the PETC mineral matter program to develop qualitative and quantitative understandings of mineral matter

transformations in environments typical of pulverized-coal combustors and to incorporate these into tractable, mechanistic models. Major aspects of the project are designed to support DOE's Combustion 2000 program.

RECENT PROGRESS

The focus of this discussion is on the development of a diagnostic for detecting alkali vapors in combustion diagnostics being performed as part of Subtask 2.1.

Tunable Diode Laser Spectroscopy

During this year, a diagnostic based on a tunable diode laser (TDL) was developed to detect alkali vapor in combustion environments. NaCl is one of the species of prime interest to alkali transport in coal combustion systems and is also the most difficult of the major alkali-containing species to monitor spectroscopically. While NaCl is the focus of discussion below, the same technique can be used for sulfates and other alkali-containing vapors of interest.

System Description

A tunable diode laser probes the vibrational spectrum of alkali-containing vapors such as chlorides, hydroxides, and sulfates of sodium and potassium. The vibrational bands for the hydroxides and chlorides are in the region from 330 to 400 cm^{-1} (30 to 25 μm), a region requiring more specialized equipment than is common for most molecular spectroscopy. Figure 1 illustrates the major components of this system. The following description describes the optical and essential electronic components of the system.

A tunable diode laser (TDL) provides the probe beam for analyzing the vapors. This lead-salt diode operates at low temperature (7-35 K), and is less well behaved than traditional TDLs. A Laser Photonics cryogenic (helium) compressor and cold finger maintains the laser temperature at a set value. The cold finger can accommodate up to four diode lasers simultaneously. Three are currently installed, but all of the data reported below were collected with a single diode.

The laser is tuned over a range of about 0.5 cm^{-1} by varying the diode current between its lower threshold value (typically ≈ 100 mA) and its upper limit (typically 500 mA to 1 A). The lower limit is dictated by the current required to induce lasing in the diode. The upper limit is dictated by the safe current carrying capacity of the laser. The specific frequency of the laser light is determined by a combination of temperature and current. In practice, these diodes emit approximately twelve frequencies simultaneously, depending on current and temperature. The beams from the laser are more divergent than is common in most lasers. The beams are collimated with an off-axis paraboloidal mirror, but they still diverge somewhat. The beam is chopped either by chopping the current to the diode or by passing the beam through a mechanical chopper. Beam chopping has a practical upper limit of around 5 kHz. The current controller installed with the TDL has a maximum current modulation rate of 1 kHz, but more rapid modulation can be controlled by interfacing the controller with external signal generators.

The beam then passes through a furnace used to generate reference, test, and calibration spectra. This furnace operates at temperatures up to about 1600 °C and is open at both ends. A version with windows is being developed to allow lower pressure (20 Torr), more controlled calibration

data to be obtained. The material of choice for these windows is thallium bromoiodide, generally referred to as KRS-5. Its advantages include its transparency at these wavelengths and reasonable transparency at visible wavelengths. Traditional infrared materials such as potassium bromide and sodium chloride are opaque at the wavelengths we are interested in (25-30 μm). Means of placing KRS-5 windows on either end of the furnace are still being pursued. The windows must be maintained at relatively cool temperatures, well under 100 °C for both physical and safety reasons. Thallium bromoiodide softens at low temperature and is toxic. There are known ways to protect the windows from heat or condensing inorganic vapors while providing an gas-tight seal for the furnace, but they require both composition and temperature gradients in the test cell. The furnace walls are removable and are composed of available ceramic materials, generally either mullite or silicon carbide.
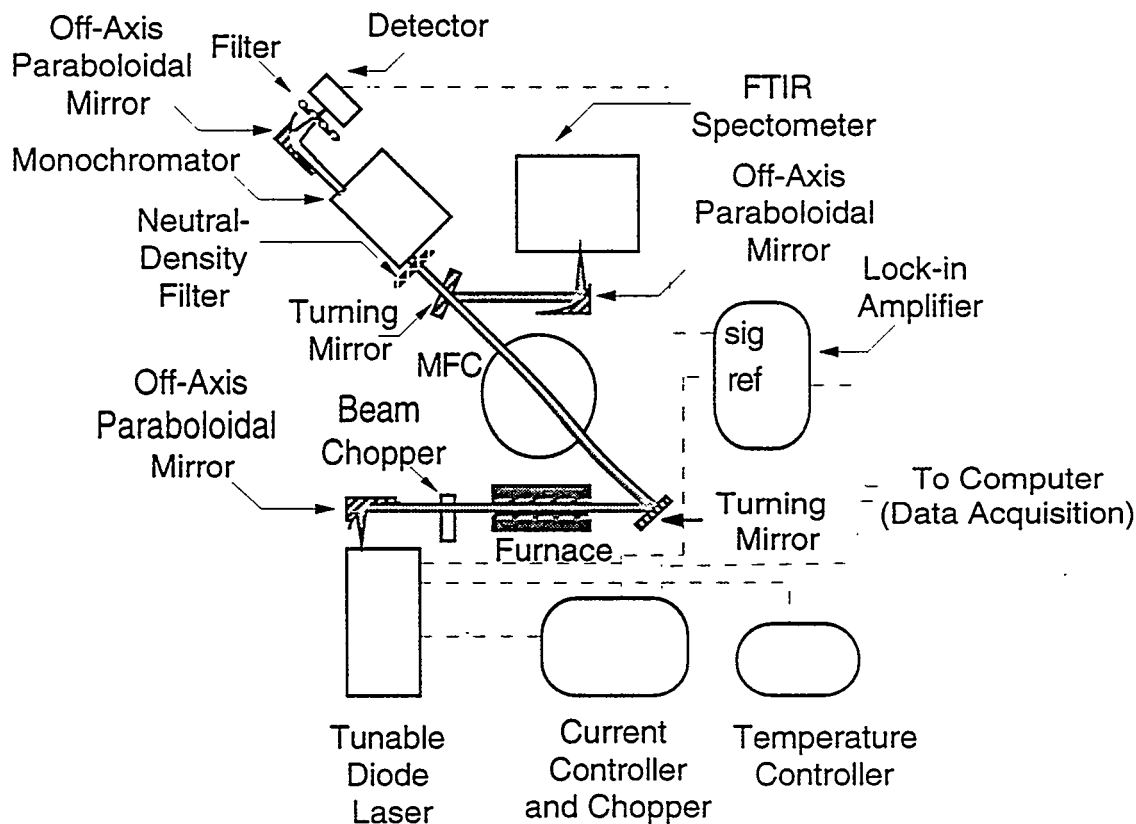

Figure 1   Schematic diagram of the TDL system used to monitor alkali-containing vapors. Optical components are illustrated as rectangles. Electronic components are illustrated with rounded corners.

After exiting the furnace, the beam is directed through the test section of the Multifuel Combustor (MFC). Significant beam steering is induced by index of refraction gradients in both the furnace and the MFC test section. The MFC has added complexities of particle interference and turbulent fluctuations in gas temperature and composition. Measures have been taken to minimize the impacts of these issues by using modulated signals and lock-in amplifier detection, averaging scans both in real time and by repeated experiments, and shielding the detector from

stray radiation.

The laser beam terminates on a copper-doped germanium detector. This detector operates at liquid helium temperature and is contained in a dewar system. Its low operating temperature ($\approx$4 K) is not trivial to maintain, and its sensitivity to temperature variation is strong. Cold, broad bandpass filters in front of the detector are used to further reduce contributions from stray radiation sources.

The laser beam to help discriminate against stray sources of radiation and to accommodate the AC-coupled detector. The detector signal is filtered with a lock-in amplifier. The laser is scanned at a rate of about 0.01 Hz over a range of approximately 200 mA (current, rather than temperature is used to tune the laser). The current can be increased, decreased, or alternately increased and decreased from its set point.
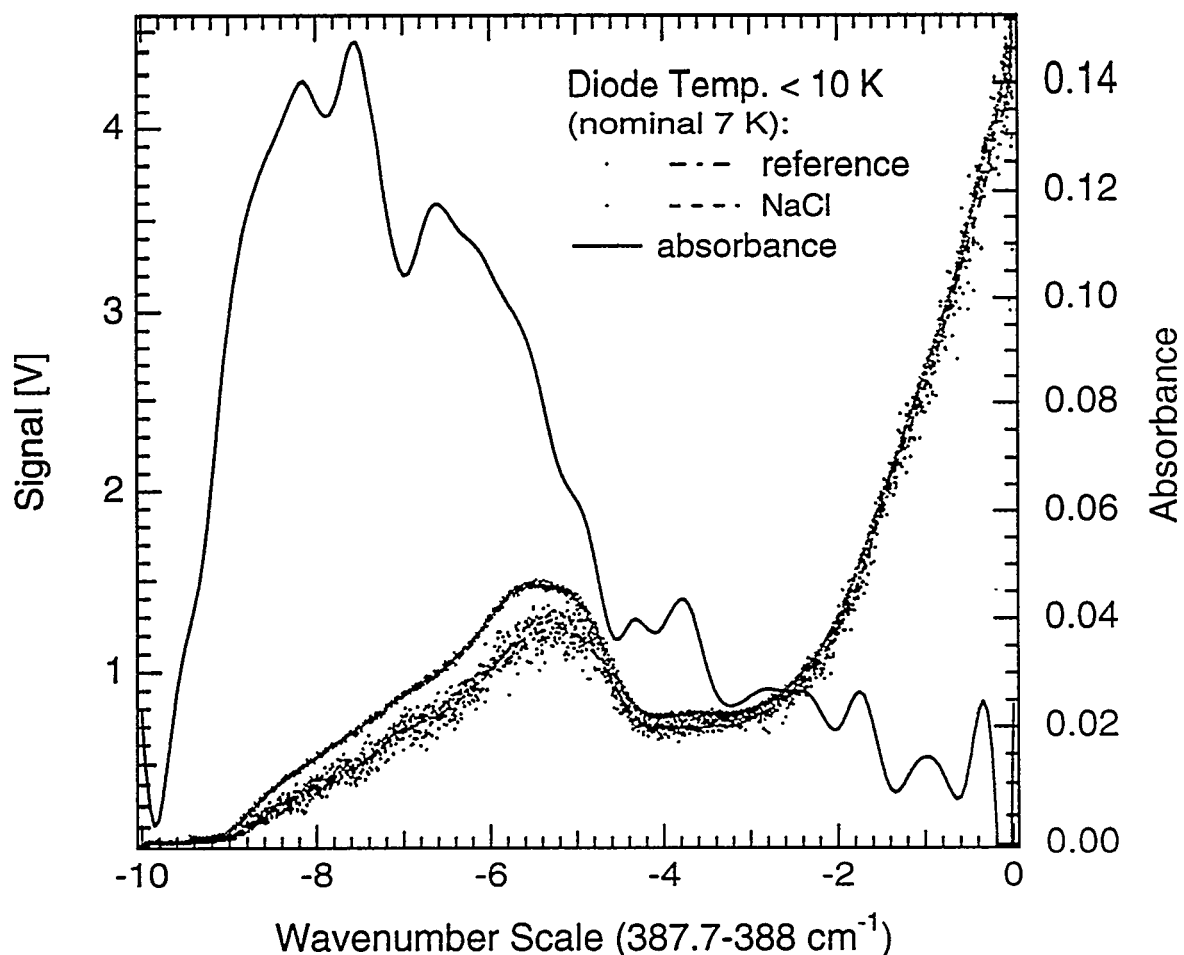

Figure 2   Reference and probe signals, together with absorbance spectrum, for a portion of the NaCl spectrum between approximately 387.7 and 388 wavenumbers. The rise in absorbance is an indicator of the NaCl R-branch bandhead located at 387.8 cm^{-1}.

Figure 2 illustrates the spectra collected recently near the bandhead of the R-branch of NaCl (NaCl spectrum illustrated in Fig. 3). The abscissa is approximately linearly proportional to wavenumbers over the approximate range from 387.7 to 388 cm^{-1}, but the wavenumber calibration has not yet been performed. The solid line, referenced to the right, represents the absorbance spectrum of sodium chloride. The intensity of the laser varies in complex ways with wavenumber, as represented by the reference beam. The wavenumber-resolved laser intensity when passing through NaCl vapor is also indicated. The data are reasonably dense and tend to obscure the lines designating the reference beam and the NaCl absorption beam. In both figures, the reference beam intensities are generally higher and less scattered than the NaCl absorption data. The absorbance spectrum is calculated from the logarithm of the ratio of reference to NaCl results.

The laser is being forced to its low-temperature limit to collect these data because the laser mode we are using barely overlaps the bandhead we are trying to measure. A more ideally located mode would allow us to perform the experiment without pushing the equipment quite as close to its performance limit. Diode lasers that operate in this region are rare – we know of only one supplier. Even for this supplier (Laser Photonics), these diodes are difficult to produce. We are working with them to obtain additional lasers with more convenient modes.
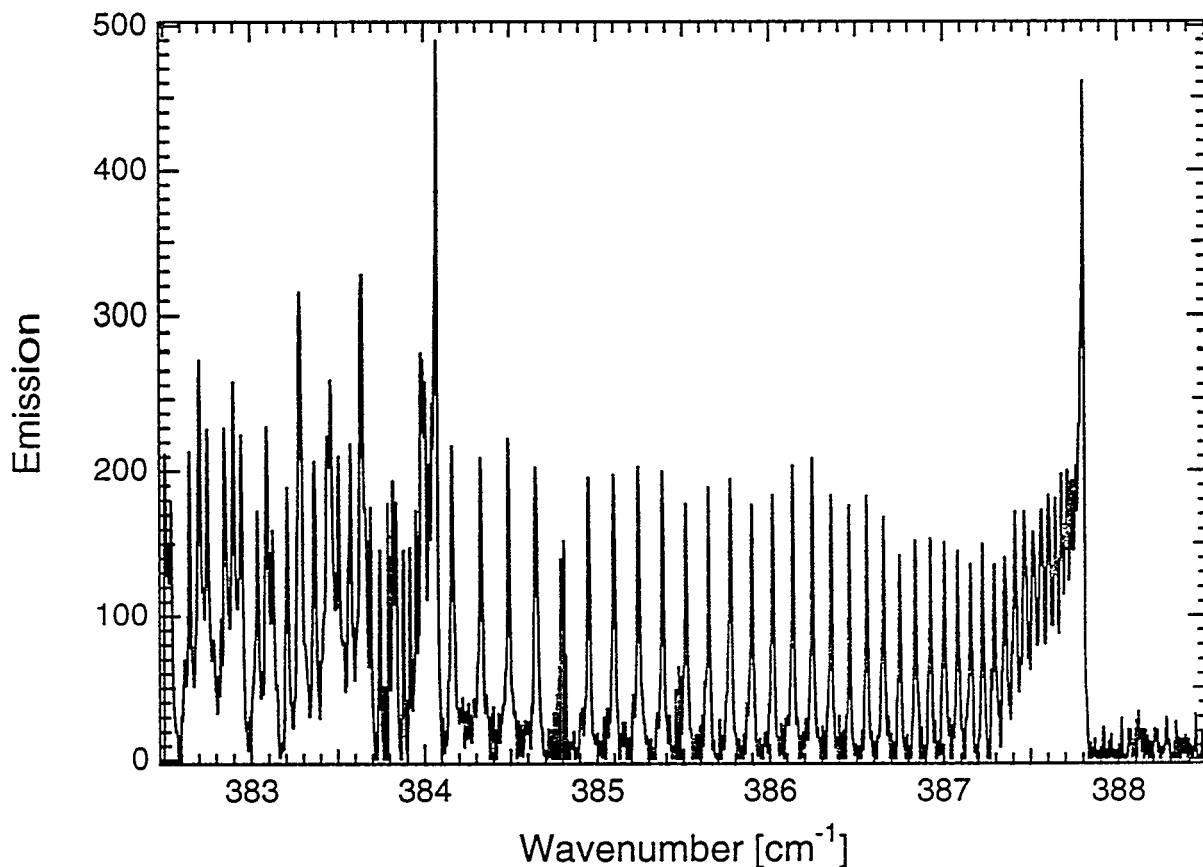

Figure 3   Emission spectra of NaCl at 20 Torr collected by collaborator Peter Bernath using high resolution FTIR spectroscopy and a heated test chamber. The data in Fig. 2 indicate an attempt to resolve the bandhead located at ≈ 388 cm^{-1} at 1 atm.

The data are encouraging. The absorbance indicates the expected dependence on wavenumber and has nearly identical peak values over this wavenumber range even if the peak laser power increases by 50%. The fine structure in the absorbance data is indistinguishable from noise, primarily because of beam steering observed during collection of the NaCl data through a hot cell. The decline of absorbance at the lowest wavenumbers (left side of the abscissa) is attributable to a loss of laser power, not a loss of absorption. The decline in absorbance at high wavenumbers is indicative of the bandhead (compare with Fig. 3).

While the data are encouraging, they are not definitive. The potential for thermal feedback from the hot furnace to the cryogenically cooled laser and detector could cause biases in laser or detector performance that, in turn, could be misinterpreted as absorption. The impact on the detector should be slight since the beams that arrive there must first pass through optical filters and a monochromator. The additional energy within the resolution of the monochromator (about 0.7 cm^{-1} in this region) is likely insufficient to change the detector temperature or otherwise bias its performance. The laser, on the other hand, is closer to the furnace. The only optical components between the laser and the furnace are choppers, baffles, mirrors, and apertures. The principal problem is that we are at the extreme limit of the laser operating window (low temperature and low current) for this laser mode to collect these data. There are other modes that operate at still lower frequency ranges, but the bandhead is the most definitive and forgiving feature of the NaCl spectrum to work with. While we continue to optimize this technique with this laser, we are also pursuing a different laser with a more conveniently available (say 387-388 cm^{-1}) tunable mode.

PRESENTATIONS AND PUBLICATIONS

The following publications were submitted in the last year based in part or in whole on PETC-sponsored work.

Baxter, L. L., Mitchell, R. E., and Fletcher, T. H. (1995 (to appear)). "Release of Inorganic Material During Coal Devolatilization." *Combustion and Flame, to appear*.

Richards, G. H., Harb, J. N., Baxter, L. L., Bhattacharya, S., Bupta, R. P., and Wall, T. F. (1994). "Radiative Heat Transfer in PC-Fired Boilers — Development of the Absorptive/Reflective Character of Initial Ash Deposits on Walls". In *Twenty-Fifth Symposium (International) on Combustion*, . Irvine, CA: The Combustion Institute.

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