**Development of Standardized Air-Blown Coal Gasifier/
Gas Turbine Concepts for Future Electric Power Systems
Volume III
Appendix B: NO_x and Alkali Vapor Control Strategies**

Final Report

July 1990

Work Performed Under Contract No.: DE-AC21-89MC26291

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
CRS Serrine, Inc.
Power Division
Greenville, South Carolina

MASTER

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July 1990

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NO_x AND ALKALI VAPOR CONTROL STRATEGIES
FINAL REPORT

Prepared for:

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EXECUTIVE SUMMARY

CRS Serrine (CRSS) is currently evaluating a novel IGCC process in which gases exiting the gasifier are not cooled prior to being burned in a gas turbine combustion system for power production. The turbine exhaust gas is used to generate additional power in a conventional steam generator. This results in a significant increase in efficiency. However, the IGCC process requires development of novel approaches to control SO_2 and NO_x emissions and alkali vapors which can damage downstream turbine components.

Ammonia is produced from the reaction of coal-bound nitrogen with steam in the reducing zone of any fixed bed coal gasifier. This ammonia can be partially oxidized to NO_x when the product gas is oxidized in a gas turbine combustor.

There are no commercial high temperature NH_3 removal processes. Therefore, NO_x emissions must be controlled by combustion modification and post combustion reduction by reaction with ammonia.

Alkali metals found in the coal mineral matter and in the coal matrix vaporize in the high-temperature combustion zone of the gasifier. Vaporized alkali metals later condense in the process, when the temperatures become low, on the surface of small char or ash particles or on cooled metal surfaces. If these alkali-coated materials reach the gas turbine combustor the alkali will revaporize and condense on turbine blades. Alkali metals deposited on gas turbine blades cause rapid high temperature corrosion of the blades and efficiency reduction will result.

PSI Technology Company (PSIT) was contracted by CRSS to evaluate and recommend solutions for NO_x emissions and for alkali metals deposition. Various methods for NO_x emission control and the potential process and economic impacts were evaluated. This included estimates of process performance, heat and mass balances around the combustion and heat transfer units and a preliminary economic evaluation. The potential for

alkali metal vaporization and condensation at various points in the system was also estimated. Several control processes were proposed and evaluated, including an order of magnitude cost for the control process.

NO_x Control

NO_x control processes were evaluated for application to the gas turbine combustor, the heat recovery steam generator (HRSG), and the post combustion flue gas. These processes included:

- lean premixed turbine combustors
- rich-lean turbine combustors
- low NO_x burners
- staged combustion
- reburning
- selective catalytic reduction
- selective non-catalytic reduction.

The NO_x emission goal for the system was 0.1 lb/MBtu as NO₂.

The baseline system is a combustion gas turbine followed by a HRSG which produces steam at 1500 psia and 950°F. Power recovery from the steam generates about 40 percent of the power in the entire system. The heat rate for this baseline system is about 6,700 Btu/kWh. The ammonia content of the product gas from a Lurgi gasifier is expected to range between 1000 and 2000 ppm, depending on the steam required to prevent clinkering of the coal ash. The higher the steam injection rate, the higher the NH₃ content of the gas.

If this gas is burned in a conventional gas turbine combustor, data suggests that about 80 percent of the ammonia would be converted to NO_x, resulting in an emission of 1380 ppm at 15 percent O₂ (2.00 lb of NO₂/MBtu). The only way to reduce the conversion

of ammonia to NO_x is to utilize rich-lean combustion which could reduce NO_x to about 0.5 lb/MBtu. This was used as the baseline for all subsequent NO_x control strategies. An additional 80 percent reduction from this level would be required to meet the NO_x emission goal. SCR would be capable of achieving 80 percent reduction. NO_x control capital costs for SCR applied to this system would be about \$121/kW and operating costs are 1.5 mills/kWh.

Research from the early and middle 1970's showed that supplemental firing of the HRSG with gas or oil fuel should reduce the NO_x emissions for the plant on a lb/MBtu basis. Turbine exhaust gas can be mixed with the combustion air creating an environment similar to flue gas recirculation through the burners. This work showed that some of the NO in the gas turbine exhaust could actually be destroyed in the flames of the steam generator. This NO_x destruction was greatly enhanced if some of the burners were fired fuel-rich (staged combustion).

Such a phenomenon could also occur in gasification combined cycle system if the HRSG is fired by part of the gasifier product gas. A low- NO_x gas burner may be capable of reducing the total emission by about 20 percent, still requiring SCR to meet the NO_x emission goal. NO_x control capital cost would drop to \$74/kW while operating costs will be about 0.93 mills/kW-hr.

If the stoichiometric ratio in the burner zone is dropped below 1.0 by increasing the supplemental fuel flow, NO_x in the turbine exhaust gas can be reduced to N_2 as it passes through the flame in a process similar to reburning. At a burner air/fuel stoichiometric ratio of about 0.7, 60 percent NO_x reduction might be achieved, allowing the use of SNR to meet the NO_x emission goal. Increasing the heat released in the HRSG, while keeping it constant in the gas turbine, shifts power production to the steam cycle. If enough supplemental fuel is injected to use all the O_2 in the turbine exhaust gas the steam cycle would produce 89 percent of the power. Going to SNR reduces capital costs for the NO_x control system to \$7.6/kW and operating costs, dominated by the cost of ammonia, drop to 0.04 mills/kWh.

Staged combustion in the HRSG will allow the use of SNR rather than SCR to meet the NO_x emission goal. This will result in substantial savings both in capital costs by eliminating the reactor and in operating costs by eliminating catalyst replacement. Catalyst replacement accounts for 80 to 90 percent of the operating cost in a SCR system. NO_x reduction may be more readily achieved by supplemental firing with coal-derived gas than with non-nitrogen bearing fuels used previously. Ammonia in the fuel gas might react with the NO_x even at near stoichiometric conditions to form N_2 . However, due to the low oxygen content of the turbine exhaust gas (10 to 12 percent) it may be necessary to add air to the turbine exhaust gas to sustain stable combustion.

Supplemental firing of the HRSG will also increase steam flow and the proportion of power recovered from the steam cycle compared to the gas turbine. This will increase the heat rate for the power plant. One potential method to mitigate these effects is to reduce fresh air addition by designing a burner with an air rich core. Instead of increasing the total burner oxidant stream to 15 percent O_2 , a small part would be increased to 21 percent providing flame stability. Reducing fresh air flow to the burner will reduce the rate of supplemental firing required to achieve a substoichiometric burner.

There are, however, some positive effects to the steam cycle. Combustion in the HRSG results in gas temperatures which will support 1000°F steam temperatures and higher steam flow rates, and justify the use of higher steam pressures (2,400-2,700 psia) and a reheat cycle. These changes will mitigate the increases in the heat rate. Repowering of a coal fired steam generator with a combustion gas turbine and supplemental firing of the HRSG would also allow for better use of the existing steam generating equipment. On the down side, supplemental firing will increase the total fuel requirements, increasing capital and operating costs for the gasification-sulfur removal portion of the process.

Alkali Vaporization

Alkali metals (Na, K) in coal come from two primary sources, minerals associated with the coal and carboxylates chemically bound into the coal matrix. The concentration of various sodium compounds in the gas was predicted using both kinetic and equilibrium models developed at PSIT. Potassium was assumed behave in a manner similar to sodium.

The primary alkali containing minerals are clays such as illite or amorphite. Alkali in the minerals are not observed to vaporize in conventional coal combustion; instead they are transformed into glassy ash particles. Most of the vaporized sodium comes from the organically bound (acid soluble) sodium in the coal matrix. The fate of vaporized sodium was analyzed by performing equilibrium calculations at gas compositions, temperatures and pressures found at key points in the gasifier and combustion gas turbine systems. The points chosen were as follows:

- Oxidizing zone of the gasifier
- Reducing zone of the gasifier
- Gasifier exit
- Hot gas cleanup
- Gas turbine combustor exhaust
- Gas turbine exit

If bituminous coals are gasified at temperatures of (1400 to 2300°F), sodium will be present as a vapor at 1 to 3 ppm (weight). As the temperature drops to about 1300°F near the gasifier exit, about 18 percent of the sodium will exist as condensed NaCl (for initial sodium = 3 ppm). This material will tend to condense heterogeneously on small particles which comprise most of the particulate surface area or homogeneously as a vapor fume. As the gas cools further to about 1000°F during hot gas cleanup, 98 percent of the sodium will condense out as NaCl. If the sodium is not removed before the gas turbine combustor, it will revaporize as NaCl or NaOH. When the sodium reaches the cooler sections of the gas

turbine, it may condense on the gas turbine blades forming Na_2SO_4 and cause imbalance or high temperature corrosion of the gas turbine blades.

Two methods of sodium removal are available, adsorption of the NaCl from the gas or removal of particles on which sodium has condensed. Because the particles on which sodium may condense are small (0.1 to 1 microns), they may not be captured by inertial separation devices such as cyclones. Other solids separation approaches that can be used are filters, such as ceramic filters and candle filters being developed under the DOE heat engines programs, or hot electrostatic precipitators (ESPs). The disadvantage with filter devices is an associated a high pressure drop, but since the fuel gas pressure is 100 to 200 psi above the gas turbine combustor pressure this may not be a significant issue. Hot ESPs have a lower pressure drop but are more capital intensive and represent an additional parasitic power loss. None of these technologies are commercial yet.

Adsorption of the NaCl can occur in two places; within the gasifier or downstream of the existing cyclones. Downstream of the cyclones either bauxite or an aluminosilicate sorbent could be added to the zinc ferrite absorber. The resulting material would then have to be separated from the zinc ferrite and regenerated. However, because most of the sodium will be present as condensed material on small particulates, even the 10 to 15s in the zinc ferrite absorber would not be enough to achieve the required reductions.

The approach that has a higher probability of success is adsorption within the gasifier using a sorbent. Because the sorbent would be intimately mixed with the coal ash, it must be a throw away material. Also it must not melt or agglomerate within the gasifier. These factors make kaolin-based sorbents ideal candidates. A portion of the resulting coal ash could also be recycled to increase sorbent utilization and to take advantage of the natural sorbent potential of the coal ash. No added capital equipment would be required for this methodology, limiting the economic impact to increased operating cost for the sorbent. At \$100/ton for kaolinite the operating cost would be \$5 to 10 per ton of raw coal depending on the amount of sodium vaporized.

Recommendations

There are two areas of technical uncertainty in the demonstration of CRS SIRRINE's IGCC process. The first is the feasibility of alkali capture by aluminosilicates, the second is the design and effectiveness of supplemental firing systems in the HRSG. PSIT recommends that experimental programs be initiated to investigate these processes.

It may be possible to capture alkali metals vaporized during coal gasification by introducing aluminosilicate clay to the top of the gasifier. The level of capture which might be achieved in this manner is unknown. PSIT could perform bench-scale tests to determine the effects of sorbent type, concentration, grain size, gas composition, and temperature on the capture of sodium and potassium vapor compounds (metal, metal chloride, metal oxide). If the results of the bench-scale test are promising the work could be extended to pilot-scale.

NO_x reduction by supplemental firing in the HRSG is already developed to the point of pilot-scale testing. Tests conducted in the 1970s showed that supplemental firing in the HRSG could reduce NO_x. However, there are two significant differences between this situation and earlier work: lower O₂ concentration in the turbine exhaust gas, and ammonia in the supplemental fuel. PSIT could help direct pilot-scale combustion tests to develop the supplemental firing system. The pilot-scale tests should focus on ways to overcome the combustion stability limits of low Btu gas and low O₂ concentrations, and on the NO_x reduction potential of supplemental firing. Based on the results of these tests PSIT would design a prototype burner or recommend a commercial burner for prototype testing. Reliable predictions of NO_x emissions for the full-scale system could then be made.

Finally, PSIT has process modeling capabilities which we would like to make available to CRS SIRRINE. PSIT uses ASPEN to model component performance and conduct mass and energy balances on process flow schematics. ASPEN is a process independent, steady state, model developed for the DoE Morgantown Energy Technology Center. Models for process modules (heat exchangers, reactors, compressors, turbines, etc.) provided in

ASPEN are linked via a preprocessing program to model virtually any configuration and design specifications. ASPEN was used to conduct the process analysis described in this report PSIT could extend the modeling conducted under this project to include the balance of the process, allowing more complete assessment of the impact supplemental firing on process design and performance.

1. INTRODUCTION

Integrated Gasification Combined Cycle (IGCC) is a promising near term technology for environmentally acceptable power production from coal. Coal is gasified under pressure (10 to 15 atm) using air or oxygen and steam. The resulting gas is burned and expanded to about 1 atm in a gas turbine. Turbine exhaust gas (TEG) is still at a relatively high temperature (900 to 1200°F), allowing additional heat to be recovered in a conventional Brayton cycle steam generator/turbine system.

Unlike the clean fuels (No. 2 oil and natural gas) usually fired in combustion gas turbine (CGT) systems, coal derived gas contains H_2S , NH_3 , and vaporized alkali metals which are deleterious to downstream components, to the environment. Usual practice is to cool the gas to moderate temperatures ($<300^\circ F$) and remove the noxious species. However, this heat loss imposes a substantial penalty in thermodynamic efficiency.

CRS Serrine is evaluating an IGCC process with improved thermal efficiency, achieved by not cooling the coal derived gas. This requires a change in process emission control strategies. CRS Serrine has selected H_2S capture by zinc ferrite followed by oxidation and conversion of the H_2S/SO_2 to elemental sulfur as a sulfid emissions control strategy. Unfortunately, there are no commercial processes to remove ammonia and alkali vapors from coal derived gas. PSI Technology Company (PSIT) has evaluated alternative approaches for control of ammonia/ NO_x emissions and alkali vapors. Available control technologies, their effectiveness and cost impact are the discussed in the following sections.

2. AMMONIA/NO_x CONTROL

Previous research has shown that air blown gasification of bituminous coal in a Lurgi dry bottom gasifier produces a fuel gas containing about 2000 ppm of ammonia.¹ During combustion, a portion of this ammonia would be oxidized to NO and NO₂ (NO_x). To control the emission of NO_x from the power plant, technology exists to cool the gas and absorb the ammonia in a second gas stream for conversion to a useful by-product. However, cooling the gas to 200°F reduces thermal efficiency from 85 to 95 percent to 70 to 75 percent. To increase power production efficiency CRS Serrine is designing an IGCC system in which the coal derived gas is not cooled.

There are no commercially available hot gas cleanup technologies for the removal of ammonia. Therefore, this report focuses on the control of NO_x emissions which result from the oxidation of ammonia during the combustion process. NO_x control technologies fall into two basic categories: combustion modification and post combustion NO_x reduction or removal.

2.1 Combustion Modification

Water injection is the NO_x control method which has traditionally been employed in gas turbine combustors. Water is injected into the combustion chamber with the air. Evaporation of the water and the added mass flow act to absorb heat, depressing the average combustion temperature which tends to decrease the peak flame temperature. Hence, the formation of NO_x by thermal fixation of atmospheric nitrogen (thermal NO_x) is reduced. NO_x (referenced to 15 percent O₂) emissions while firing natural gas typically drop from 65 to 35 ppm.² This technique has the added advantage of increasing the work output from the gas turbine, though total plant efficiency decreases due the increased loss of energy to the stack gasses.

A relatively new technology, pre-mixed lean combustion avoids the increased stack gas losses. Mass flow to the primary combustion chamber is increased by premixing the fuel with large quantities of air. Though the turbine inlet temperatures are unchanged, premixing the fuel and air increases the mass flow in the vicinity of the burner, reducing the peak flame temperatures. Since thermal NO_x is only formed in large quantities when the temperature is above 2800°F , reducing peak flame temperatures can limit NO_x emissions to less than 10 ppm.³ This technique is limited by the ignition stability of the flame.

These NO_x control technologies deal with thermal fixation of nitrogen which is sufficient for the clean fuels (natural gas and light oils) for which gas turbine engines were developed. However, as previously mentioned, coal-derived gas contains significant amounts of ammonia which might be oxidized to NO_x . Furthermore, air blown coal derived gas contains significant amounts of inerts (i.e., N_2 , H_2O and CO_2) which will absorb energy during combustion. Depending on the heating value of the gas, the resulting flame temperatures may be lower than for light oil or natural gas, so thermal NO_x is not an issue.

Uncontrolled conversion of ammonia to NO_x is about 80 percent, therefore, any NO_x control techniques applied to IGCC systems must be effective in reducing the conversion rates of ammonia to NO_x . Rich-lean gas turbine combustors have been developed to control NO_x emissions when firing fuels which contain nitrogen. These combustors are an extension of the staged combustion technologies which were developed to control NO_x emissions from utility boiler furnaces firing coal or residual fuel oil. The primary combustor is fired fuel rich at a stoichiometric ratio of about 0.7. The remaining air for combustion and cooling is added in the second combustion chamber where temperatures are minimized to minimize NO_x formation. This technique has yielded NO_x reductions of 70 percent firing coal-derived liquid fuels and 50 percent firing simulated low Btu gas from coal.^{4,5} Tests conducted by Toshiba Company showed about a 50 percent conversion of NH_3 to NO_x at 2000 ppm NH_3 when the first stage air/fuel stoichiometric ratio is 0.7 and the overall stoichiometric ratio is 2.2 at atmospheric pressure. Increasing the pressure decreased NH_3 conversion to NO_x to 32 percent.⁵ For the range of coal gases specified by CRS Sitrine this translates to NO_x

emissions of about 0.5 to 0.62 lb/MBtu as NO₂. As these NO_x emissions of 0.5 lb/MBtu are well above the planned goal of 0.1 lb/MBtu, a supplemental method of NO_x control will be required.

2.2 Post Combustion NO_x Control

Commercially available post combustion NO_x control technologies are listed below.

- Selective Non-Catalytic Reduction (SNR)
- Selective Catalytic Reduction (SCR).

These systems are being applied to a limited number of utility boiler systems in the U.S. and may be applicable to IGCC systems.

In a SNR process ammonia or urea is injected into the post flame region (or turbine exhaust gas (TEG)) at 1400 to 2000°F.^{6,7,8} These nitrogen species react with NO formed in the flame to produce N₂. SNR processes can potentially provide greater than 50 percent NO_x reduction; however, they are temperature sensitive. The location in a utility boiler system where the optimum temperature occurs changes with changes in heat input or heat absorption. In the event that the optimum temperature window shifts from the point of injection, desired NO_x reductions might not be met, as shown in Figure 1 for a 110 MW oil and gas fired boiler.⁷ If temperatures are too low, unreacted ammonia may escape to the atmosphere stack, or if temperatures are too high NO_x emissions may increase due to the oxidation of the reductants.⁶

SCR overcomes these problems by the use of a catalyst to facilitate reaction. The temperature window is lowered and broadened to 600 to 900°F and ammonia slip is typically below 10 ppm. Shifting the temperature window puts the SCR downstream of the majority of the heat transfer surface in a boiler so that gas temperature is less affected by heat input. However, capital costs increase significantly with the addition of a reactor and catalyst. The

Figure 1. Effect of Heat Input on NO_x Reduction⁷ by Urea Injection into a 110 MW Oil and Gas Fired Boiler

reactor is designed to attain a desired residence time making the size dependent on the gas flow rate more than ammonia concentration. Operating costs increase due to the expense of the catalyst which must be replaced periodically. A SCR system can reliably reduce the NO_x concentration by 80 to 90 percent. Reducing the additive injection temperature also allows the additive injection system to be placed in the gas stream rather than at the wall, improving the rate and extent of mixing between additive and NO_x.

PSI Technology Company is developing a low temperature SNR system using a proprietary chemical.⁹ The desired reactions occur in the temperature range of 750 to 850°F. As with the SCR system the temperature window is such that its physical location in the system is less dependent on heat input. NO_x reductions of 60 to 80 percent have been achieved in the laboratory with this additive, as shown in Figure 2. In larger systems where

Figure 2. Laboratory NO_x Reduction Using PSIT's SNR Process

the mixing rates could be slower or temperature gradient larger we estimate 50 percent NO_x reduction at an additive to NO_x molar ratio of 0.9.

2.3 NO_x Control in the Steam Generator

If the exhaust from the gas turbine combustor is ducted to the windbox of a separately fired steam generator, additional NO_x reduction is possible. Based on experience gained by boiler suppliers and EPRI in the 1970s, it was established that under certain conditions the NO_x emitted by the gas turbine could be destroyed by "reburning" it to N₂ in a boiler flame.^{10,11} The key to NO_x reduction for these turbine exhaust-fired boilers is to establish low-NO_x combustion conditions (i.e., slow mixing of fuel and oxidant, local fuel-rich regions, low temperatures). During the tests at West Texas Utility's Lake Nasworthy Station and Oklahoma Public Service Co.'s Horseshoe Lake Station the NO_x formed in the gas turbine was reduced 10 to 20 percent during unstaged supplemental firing with natural gas and 17 percent O₂ turbine exhaust.^{10,11} Additional NO_x reductions up to 50 percent (based on lb/hr NO emitted by the gas turbine) could be achieved using staged combustion (burners

out of service) at a stoichiometric air/fuel ratio of about 0.8. Figure 3 shows the boiler design at Lake Nasworthy.

Figure 3. Lake Nasworthy Power Station, Unit No. 1,
West Texas Utilities Company, San Angelo, Texas

Research conducted by KVB, Inc. sponsored by the Electric Power Research Institute demonstrated that even greater NO_x reductions are possible.¹² In tests conducted with natural gas and #2 fuel oil in a 100,000 Btu/hr test facility, nearly 100 percent of the NO_x contained in the simulated turbine exhaust gas was destroyed when the burner was fired at an air/fuel stoichiometric ratio of 0.65, additional air to complete combustion injected downstream of the flame zone. Figure 4 shows their data for two conditions:

1. Turbine exhaust to the burner, fresh air to the staging air injectors (solid points)
2. Turbine exhaust to both the burners and staging air injectors (open points).

Only 80 percent NO_x reduction was achieved when the staging air contained 100 ppm NO_x from the gas turbine. However, these researchers may have been able to improve NO_x

Figure 4. Influence of NO in Bypass Air on NO Reduction Factor

reductions by optimizing the second-stage mixing and burnout zone temperature.¹³ The fuels in these tests contain no fuel nitrogen, if NH_3 in the fuel gas react with NO in the turbine exhaust gas it may be possible to reduce the level of staging required to meet the NO_x emission goal.

The gas turbine combustors specified for the subject system by CRS Serrine are far more efficient than those used previously for combined-cycle power generation. The turbine exhaust from a modern GE 7000 series machine would nominally contain 11 to 12 percent O_2 at a temperature of about 1000 to 1200°F. This gas stream may be too vitiated to support combustion. Unpublished test results¹⁵ indicated that the minimum O_2 concentration in simulated turbine exhaust gas required to stabilize a single oil or gas flame on a turbulent burner was about 13 percent. In a practical multi-burner system, it is felt that a minimum of 15 percent O_2 would be prudent for design purposes to allow a margin of safety. However, the elevated temperatures of the turbine exhaust gas and fuel stream may extend combustability limits. Therefore, a forced draft fan has been specified for this system to mix sufficient fresh air with the turbine exhaust to increase the O_2 concentration. The forced draft fan would supply both windbox air and fresh air to the overfire air injectors to maximize the NO_x reduction during staged combustion.

It is important to note that the CRS Serrine system is fired by coal-derived gas, not natural gas. Therefore, the combustion system must be designed to minimize conversion of NH_3 contained in the fuel to NO_x . Like the NO in the turbine exhaust, the NH_3 in the fuel gas can be converted to N_2 during combustion. Tests have been performed on a 1 MBtu/hr circular burner¹⁶ fired with actual coal-derived gas from a Wellman-Galusha atmospheric-pressure gasifier. The resulting product gas contained approximately 2000 ppm of ammonia at a heating value of 140 to 160 Btu/scf. When burned in a refractory-lined combustor, with a swirl stabilized burner, at about 3 percent excess O_2 , 25 percent of the ammonia was converted to NO_x . Although staged combustion was not attempted during these tests, we estimate that NH_3 conversion to NO_x would be limited to about 13 percent at a

stoichiometric air/fuel ratio of 0.7. This estimate should be substantiated by pilot-scale combustion tests using simulated Lurgi product gas.

2.4 Process Analysis

Heat and mass balances were performed on the CGT and steam generation portion of the system using the process simulator ASPEN. The purpose of this modeling was to assess the NO_x control strategies required to meet the emissions goal of less than 0.1 lb NO_x/MBtu as NO₂ and the effect on the balance of the IGCC plant. The system modelled included a combustion gas turbine system and a heat recovery steam generator (HRSG).

Three CGT systems were evaluated: LM/TG 5000PC, 7111EA and 7191F. Each CGT was evaluated with three HRSG configurations as listed below:

1. Unfired HRSG
2. Fired HRSG with unstaged combustion
3. Fired HRSG with staged combustion.

Because the 7191F CGT is the baseline system for CRS Serrine's analysis and the modeling results are similar for all the turbine sizes, only the 7191F results will be discussed in detail. Flowsheet and heat and mass balance results are included in Appendices A through F for the 7111EA and LM/TG 5000PC CGT systems.

2.4.1 Unfired Heat Recovery Steam Generator

Figure 5 shows the process flow diagram for the 7191F CGT with an unfired HRSG. Mass flow rates, temperatures and pressures are listed in Table 1 for all streams.

Fuel and air enter the first stage of the gas turbine combustor which is modelled as an adiabatic, isobaric reactor. The air/fuel stoichiometric ratio is 0.7 and the reactions are

Figure 5. Process Flow Sheet for the 7191F Combustion Gas Turbine with an Unfired Heat Recovery Steam Generator

allowed to go to 65 percent of completion. Gas exits the first stage of the combustor at 2983°F. Second stage air is mixed with the effluents from the first stage bringing the total stoichiometric ratio to 2.67. The mixture enters a second adiabatic, isobaric reactor and the reactions are allowed to go to completion. The temperature exiting the second stage of the combustor is 2299°F. About 0.5 lb/MBtu NO_x may be produced in the combustor.

Turbine cooling air is added to the gas stream and the gas enters the gas turbine. Cooling air would actually be added to the gas gradually as it is expanded, but this simplification adds little error and information for a more rigorous solution is not available. The gas turbine is assumed to have an isentropic efficiency of 80 percent and a mechanical/generator efficiency of 90 percent. The gas is expanded from 201 psia and 2299°F to 15 psia and 1171°F. Power recovered during the expansion is 901 MBtu/hr, but some of this power is used to compress the combustion air so that net power recovery is about 490 MBtu/hr (144 MWe).

**Table 1. Stream Properties for the 7191F Combustion Gas Turbine
with an Unfired Heat Recovery Steam Generator**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	644,108	1020	276
(2) First Stage Air	676,312	715	201
(3) First Stage Exit	1,320,237	2983	201
(4) Second Stage Air	1,899,839	715	201
(5) Second Stage Exit	3,220,091	2299	201
(6) Cooling Air	400,500	715	201
(7) Turbine Exhaust Gas	3,620,534	1171	15
(8) Superheater Exit Gas	3,620,534	994	15
(9) Boiler Exit Gas	3,620,534	554	15
(10) Economizer Exit Gas	3,620,534	245	15
(11) Boiler Feed Water	641,027	59	1500
(12) Economizer Exit Water	641,027	500	1500
(13) Saturated Steam	641,027	598	1500
(14) Superheated Steam	641,027	950	1500

The turbine exhaust gas (TEG) then enters the HRSG. Steam production is set to absorb heat removed from the gas in cooling it to 245°F. The first heat transfer surface is the superheater where the steam temperature is increased from 598 to 950°F. Gas temperature leaving the superheater is 994°F. Gas then enters a boiler bank where about 641,000 lb/hr of steam is generated and the gas is cooled to 554°F. Finally, the gas goes to an economizer where it is cooled to 245°F and boiler feed water is heated from 59 to 500°F. Using the information supplied by CRS Sirrinc, electricity generation should be about 0.16 MWe/Klb steam or 100 MWe. This is about 40 percent of the of the total power produced by the facility.

The NO_x emissions of 0.51 lb/MBtu will require 80 percent reduction to achieve the goal of 0.1 lb/MBtu NO_x, requiring the use of SCR. Even if ammonia SNR could achieve

the required level of reduction, the optimum injection temperature (1400 to 2000°F) exists only inside the gas turbine. SNR cannot be applied to a system with an unfired HRSG because turbine exhaust temperatures are too low.

The gas flow rate leaving the HRSG, 3,620,534 lb/hr, is equivalent to a 369 MWe coal fired power plant with a heat rate of about 10,000 Btu/KW-hr and 20 percent excess air (9800 lb gas/MWe). Figure 6 shows that in this size range an SCR system will cost about \$80/KWe, making the total capital cost \$29.6M including the catalyst.¹⁷

The dominant operating costs are replacement catalysts and ammonia to react with the NO_x . The gross heat input is 1635 MBtu/hr, making the NO_x throughput 834 lb/hr. At a NH_3/NO stoichiometric ratio of 1.0, 308 lb/hr of NH_3 are required. The ammonia cost is estimated at \$145/ton which translates to a annual cost of \$166,000 at a combined availability and capacity factor of 0.85. Catalyst costs are about \$27/KWe with an estimated life of 2 to 6 years. Annual costs are \$2,491,000 (\$9,963,000 average over 4 years). The operating cost exclusive of capital carrying charge are \$2,657,000/year.

Figure 6. Post Combustion NO_x Control Capital Cost

2.4.2 Fired Heat Recovery Steam Generator with Unstaged Combustion

Figure 7 shows the process flow diagram for the 7191F CGT with a fired HRSG and unstaged combustion. Mass flow rates, temperatures and pressures are listed in Table 2 for all streams.

Because the gas turbine is the same as in the previous section, the CGT heat and mass balances will be the same. However, the TEG is going to be used as an oxygen source for the combustion of additional fuel in the HRSG. The oxygen concentration must be increased from 11.3 to 15 percent to insure stable combustion. This will require the addition of 2,397,997 lb/hr of fresh air, and the addition of a forced draft fan.

This combined air and TEG stream is used to oxidize fuel in the furnace section of the HRSG. Fuel flow is set at 2,636,348 lb/hr coal derived gas to achieve a total

Figure 7. Process Flowshseet for the 7191F Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Unstaged Combustion

**Table 2. Stream Properties for the 7191F Combustion Gas Turbine with
a Fired Heat Recovery Steam Generator, Unstaged Combustion**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	644,108	1020	276
(2) First Stage Air	676,312	715	201
(3) First Stage Exit	1,320,237	2983	201
(4) Second Stage Air	1,899,839	715	201
(5) Second Stage Exit	3,220,091	2299	201
(6) Cooling Air	400,500	715	201
(7) Turbine Exhaust Gas	3,620,534	1171	15
(8) Fresh Air	2,397,997	59	15
(9) Burner Oxidant	6,018,531	758	15
(10) Burner Fuel	2,636,348	1020	276
(11) Furnace Exit Gas	8,654,878	1733	15
(12) Superheater Exit Gas	8,654,878	1193	15
(13) Economizer Exit Gas	8,654,878	245	15
(14) Boiler Feed Water	5,205,254	59	1500
(15) Economizer Exit Water	5,205,254	500	1500
(16) Saturated Steam	5,205,254	598	1500
(17) Superheated Steam	5,205,254	950	1500

stoichiometric ratio of 1.1. This fuel flow increases the total fuel to the system by a factor of 5.1. The coal gasification/H₂S absorption island will have to be increased in size to achieve this fuel supply rate. Compressed air and steam flows to the gasifier and pumping/compression power consumption will also increase in proportion to the fuel supply rate.

The first heat transfer surface in the fired HRSG is the water walls of the furnace, used to generate about 5,205,000 lb/hr steam. The furnace exit gas temperature is 1733°F. Gas then enters the superheater where the steam temperature is increased from 598 to 950°F. Gas temperature leaving the superheater is 1193°F. Finally, the gas goes to an economizer

where it is cooled to 245°F, and boiler feed water is heated from 59 to 500°F. Electricity production from the steam should be about 818 MWe, or 85 percent of the total production.

The NO_x emissions generated in the gas turbine are 0.51 lb/MBtu and 0.40 lb/MBtu are generated in the furnace. A weighted average based on the heat released in each location is 0.42 lb/MBtu NO_x and will require 76 percent reduction to achieve the goal of 0.1 lb/MBtu NO_x. Actual NO_x emissions may be lower than this since NH₃ in the coal-derived gas should react with NO_x at 1733°F in the TEG to form N₂.

The requirement of 76 percent NO_x reduction suggests the use of SCR, but SNR at 1400 to 2000°F could achieve a 50 percent level of reduction, which may be enough if used in conjunction with low-NO_x burners. The gas flow rate leaving the HRSG, 8,654,878 lb/hr, is equivalent to a 883 MWe coal fired power plant with a heat rate of about 10,000 Btu/KWe-hr and 20 percent excess air. Again an SCR system will cost about \$80/KWe, making the total capital cost \$70.7M including the catalyst.

The dominant operating costs are replacement catalysts and ammonia to react with the NO_x. The gross heat input is 8327 MBtu/hr, making the NO_x throughput 3497 lb/hr. At a molar ratio of 1.0, 1293 lb/hr of NH₃ are required. The ammonia cost is estimated at \$145/ton which translates to a annual cost of \$698,000 at a combined availability and capacity factor of 0.85. Catalyst costs are about \$27/KWe with an estimated life of 2 to 6 years. Annual costs are about \$5,960,000/yr (\$23,841,000 average over 4 years). The operating costs are \$6,658,000/yr exclusive of capital carrying charges.

2.4.3 Fired Heat Recovery Steam Generator with Staged Combustion

Figure 8 shows the process flow diagram for the 7191F CGT with a fired HRSG and staged combustion. Mass flow rates, temperatures and pressures are listed in Table 3 for all streams.

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Figure 8. Process Flowsheet for the 7191F Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Staged Combustion

Because the gas turbine is the same as for the unfired HRSG system, the CGT analysis will be the same. Again, the TEG is going to be used as an oxygen source for the combustion of additional fuel, and 2,397,997 lb/hr of fresh air must be mixed with the TEG to raise the oxygen concentration to 15 percent.

This combined air and TEG stream is used to oxidize fuel in the furnace section of the HRSG. Fuel flow is set at 44,140,864 lb/hr to achieve a stoichiometric ratio in the burner zone of 0.7. This fuel flow increases the total fuel to the system by a factor of 7.5 compared to the unfired HRSG. The coal gasification/H₂S absorption island will have to be increased in size to achieve this fuel supply rate. Compressed air to the gasifier, steam usage and pumping/compression power will also increase in proportion to the fuel supply rate. Finally, 2,506,407 lb/hr of overfire air is needed to complete burnout raising the total air/fuel stoichiometric ratio to 1.1, further increasing the forced draft fan size.

**Table 3. Stream Properties for the 7191F Combustion Gas Turbine with
a Fired Heat Recovery Steam Generator, Staged Combustion**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	644,108	1020	276
(2) First Stage Air	676,312	715	201
(3) First Stage Exit	1,320,237	2983	201
(4) Second Stage Air	1,899,839	715	201
(5) Second Stage Exit	3,220,091	2299	201
(6) Cooling Air	400,500	715	201
(7) Turbine Exhaust Gas	3,620,534	1171	15
(8) Fresh Air	4,904,404	59	15
(9) Burner Air	2,397,997	59	15
(10) Overfire Air	2,506,407	59	15
(11) Burner Oxidant	6,018,531	758	15
(12) Burner Fuel	4,140,864	1020	15
(13) Furnace Exit Gas	12,665,802	1762	15
(14) Superheater Exit Gas	12,665,802	1212	15
(15) Economizer Exit Gas	12,665,802	245	15
(16) Boiler Feed Water	7,772,752	59	1500
(17) Economizer Exit Water	7,772,752	500	1500
(18) Saturated Steam	7,772,752	598	1500
(19) Superheated Steam	7,772,752	950	1500

The first heat transfer surface in the fired HRSG is the water walls of the furnace, used to generate 7,773,000 lb/hr steam. The furnace exit gas exit temperature is 1762°F. Gas then enters the superheater where the steam temperature is increased from 598 to 950°F. Gas temperature leaving the superheater is 1212°F. Finally, the gas goes to an economizer where it is cooled to 245°F and boiler feed water is heated from 59 to 500°F. Electricity production should be about 1222 MWe, or 89 percent of total production.

The NO_x emissions generated in the gas turbine are 0.51 lb/MBtu, but these are reduced to 0 as the TEG passes through the fuel rich flame in the HRSG furnace. Substoichiometric combustion produces 0.21 lb/MBtu NO_x in the furnace. A weighted average based on the heat released in each location is 0.18 lb/MBtu and will require 46 percent reduction to achieve the goal of 0.1 lb/MBtu NO_x .

The requirement of 46 percent NO_x reduction will now allow the use of SNR at the furnace exit temperature of 1762°F. The gas flow rate leaving the HRSG, 12,665,802 lb/hr, is equivalent to a 1292 MWe coal fired power plant with a heat rate of about 10,000 Btu/MWe and 20 percent excess air. Figure 6 shows that an SNR system will cost about \$8/KWe, making the total capital cost \$10.4M.

The dominant operating cost is ammonia to react with the NO_x ; no catalyst is required. The gross heat input is 12,146 MBtu/hr, making the NO_x throughput 2,186 lb/hr. At a molar ratio of 1.0, 803 lb/hr of NH_3 are required. The ammonia cost is estimated at \$145/ton which translates to a annual cost of \$433,000 at a combined availability and capacity factor of 0.85.

2.5 Summary

Table 4 shows characteristic mass flows and Table 5 shows temperatures for all the configurations modelled. Table 6 summarizes the estimated NO_x emissions and reduction requirements, the post combustion NO_x control method selected and the capital and operating cost estimates as discussed in Section 2.4. As noted previously, trends are the same for all of the systems investigated. The only notable exception is that the TEG temperatures from the LM/TG 5000PC and 7111EA turbines will not allow the generation of 950°F steam without excessive heat transfer areas or a fired HRSG.

Disregarding the NO_x reduction requirements, none of the gas turbine systems create an environment in which SNR may be applied directly. The temperature entering the gas

Table 4. Characteristic Mass Flows in the CRS Sirrine IGCC Process

Gas Turbine	HRSB Configuration	Stack Gas Flow lb/hr	Total Air Flow lb/hr	Total Fuel Flow lb/hr	Steam Flow lb/hr
GE 7191F	Unfired	3,620,534	2,616,651	644,108	641,027
	Unstaged	8,653,878	5,014,648	3,280,456	5,205,254
	Staged	12,665,802	7,521,055	4,784,972	7,772,752
GE 7111EA	Unfired	2,589,385	2,102,040	487,345	381,089
	Unstaged	6,428,197	3,602,034	2,826,163	3,563,727
	Staged	9,493,926	5,333,548	4,160,387	5,465,390
GE LM/TG 5000 PC	Unfired	1,023,161	871,200	151,931	137,004
	Unstaged	2,068,150	1,284,102	784,048	1,236,694
	Staged	3,031,787	1,886,581	1,145,206	1,853,023

Table 5. Characteristic Gas Temperatures in the CRS Sirrine IGCC Process

Gas Turbine	System Configuration	Turbine Inlet Temperature and Pressure °F	Turbine Exit Temperature °F	HRSB Furnace Exit Gas Temperature °F	HRSB Superheater Exit Gas Temperature °F
GE 7191F	Unfired	2299/201	1171	994	554
	Unstaged	2299/201	1171	1782	1267
	Staged	2299/201	1171	1762	1212
GE 7111EA	Unfired	1951/194	1008	861	467
	Unstaged	1951/194	1008	1466	982
	Staged	1951/194	1008	1510	1010
GE LM/TG 5000PC	Unfired	2197/394	962	823	425
	Unstaged	2197/394	962	1520	971
	Staged	2197/394	962	1547	986

Table 6. Post Combustion NO_x Control for the CRS Sarrine ICGG Process

Gas Turbine	System Configuration	NO _x as lb NO ₂ MBtu	Required Reduction %	NO _x Control Method	Capital Cost \$M	Operating Cost \$M/yr
GE 7191F	Unfired	0.51	80	SCR	29.6	2.66
	Unstaged	0.42	76	SCR	70.7	6.66
	Staged	0.18	44	SNR	10.4	0.43
GE 7111EA	Unfired	0.63	84	SCR	21.2	2.01
	Unstaged	0.51	80	SCR	52.6	5.10
	Staged	0.22	54	SNR	7.9	0.43
GE LM TG 5000PC	Unfired	0.51	80	SCR	8.4	0.75
	Unstaged	0.42	76	SCR	16.9	1.61
	Staged	0.18	44	SNR	2.5	0.11

turbine is generally too high and the temperature exiting the gas turbine too low for ammonia or urea injection. Implementing supplemental firing of the HRSG provides a temperature region near the furnace exit which is optimal for SNR.

Despite the increased gas flow associated with the systems which allow the use of SNR, capital costs of post combustion NO_x control for these are substantially lower than for the SCR systems. These cost savings are even more apparent when considering constant heat input systems. The fired HRSG systems for the LM/TG 5000PC CGT have stack gas flows and heat inputs the same order of magnitude as the 7191F CGT unfired HRSG system, the capital cost of NO_x control drops by an order of magnitude. Cost estimates for ammonia, urea, and PSIT's SNR processes will vary in both capital cost and reductant costs. However, these variations are smaller than the accuracy of this estimate. More detailed and site specific engineering would be required to distinguish between these competing technologies.

There will also be a substantial savings in operating costs with SNR. Catalyst replacement accounts for about 80 percent of the operating costs for a SCR system. Comparing the fired HRSG with staged combustion to the unfired HRSG for the 7191F CGT shows savings of about \$1.5M in annual operating costs.

Power recovered by each CGT system was considered independent of supplemental firing, though power for compression will increase some as the coal gas demand increases. Net gas turbine power is the power generated by the gas turbine minus the power required to compress the air for the gas turbine combustor. Net power generation was 144 MWe (491 MBtu/hr), 80 MWe (273 MBtu/hr) and 35 MWe (119 MBtu/hr) for the 7191F, 7111EA and LM/TG 5000PC gas turbines respectively. It should be noted however that the gasifier for the LM/TG 5000PC system operates at about twice the pressure of the gasifier for the 7191F system. Power recovery from the steam turbine (ST) was calculated using the ratio of electricity production to steam flow provided by CRS Serrine in the flow sheet and heat and mass balances.

Table 7 summarizes power production for the systems modeled. Systems with an unfired HRSG have heat rates between 6700 and 7400 Btu/KW-hr, roughly 40 to 50 percent efficient. Heat rate decreases to about 9000 Btu/KW-hr for the fired HRSG systems (greater than 35 percent efficiency). Power recovery shifts from the gas turbine to the steam turbine in the fired HRSG systems. In the 7191F system with an unfired HRSG the gas turbine provides about 60 percent of the power while with a fired HRSG it produces between 15 and 10 percent. However, a fired HRSG would allow for steam temperatures of 1000°F and the higher steam flows would allow for the design of a high pressure reheat cycle. Both of these changes will improve heat rate in the steam cycle.

NO_x control costs will drop for both fired HRSG configurations compared to the unfired HRSG. Normalized NO_x control capital costs for the unstaged configuration are reduced by a factor of 2 and operating costs are about 1/3 lower compared to the unfired

**Table 7. Power Production and Normalized NO_x Control Costs
in the CRS Sirrine ICGG System**

Gas Turbine	System Configuration	CGT Power MWe	ST Power MWe	Gross Heat Rate Btu/KW-hr	NO _x Control	
					Capital \$/KW	Operating mils/KW-hr
GE 7191F	Unfired	144	100	6700	121	1.46
	Unstaged	144	818	8656	74	0.93
	Staged	144	1222	8897	7.6	0.04
GE 7111EA	Unfired	80	60	7412	151	1.93
	Unstaged	80	560	9403	82	1.07
	Staged	80	859	9434	8.4	0.06
GE LM/TG 5000PC	Unfired	35	22	6766	147	1.71
	Unstaged	35	195	8653	73	0.94
	Staged	35	291	8917	7.7	0.05

HRSG configuration. Normalized NO_x control capital and operating costs for the staged combustion in the HRSG are an order of magnitude lower than for the unfired HRSG.

Increasing the oxygen concentration of the TEG significantly increases the amount of fuel required to achieve fuel rich conditions for NO_x reduction. A burner design which provides a stable flame root could mitigate this by limiting the level of oxygen required to sustain combustion. One way to do this is to provide a local fresh air zone at or near the flame root to stabilize combustion. The Rodenhuis and Verloop TTL/MG burner does just this. The burner utilizes two air streams: a fresh primary air stream which contacts the fuel admission streams. This is surrounded by the major oxidant stream TEG or air. The primary air serves to stabilize the flame in low O₂ concentration environments. Rodenhuis and Verloop routinely builds facilities which burn natural gas and No. 6 oil utilizing TEG with 12 to 15 percent O₂. Currently they are building a unit which fires blast furnace gas utilizing 14 percent O₂ TEG.

If 20 percent of the oxygen at the burner comes from primary air the total fresh air requirements at the burner is about 600,000 lb/hr. This would reduce supplementary fuel requirements by about 50 percent. For conditions similar to those discussed in Sections 2.4.2 and 2.4.3 power production in the steam system would drop to 450 to 650 MWe making retrofit to a 500 MW boiler possible.

A second approach to provide a stable flame root is to use a class one continuous ignitor. This device is fired with a fuel such as natural gas and supplies up to 15 percent of the burner heat input. This type of system was explored in the 1970s by the oil and gas industry to burn low Btu gas. Researchers at the MIT Energy Laboratory have also postulated the characteristics of a low Btu gas burner with extended combustibility limits.¹⁸ These burners are refractory lined combustors which exit into the heat exchanger similar to a B&W cyclone burner or slagging combustor. Flame stability is achieved by external recirculation along the refractory wall and swirl induced internal recirculation.

3. FATE AND CONTROL OF ALKALI SPECIES IN IGCC SYSTEMS

Before IGCC systems can be implemented on a wide basis, appropriate handling and removal strategies for the alkali metal species contained in coal must be developed. Most inorganic constituents pose little or no problem during operation, as they are removed as dry ash at the gasifier bottom, or are captured from the fuel gas stream via cyclones. Alkali metals, however, may cause difficulties, as they behave differently than the bulk of the ash constituents. A small fraction of the volatile sodium and potassium contained in coal may escape the gasifier in the vapor phase. If these vapors make their way to the gas turbine, they will condense during the expansion stage as chlorides, hydroxides, or sulfates. Once condensation has occurred, rapid hot corrosion of metal surfaces will follow. Turbine surfaces are extremely sensitive to corrosive damage from even low levels of condensed alkali; based upon operating experience in oil-fired systems, turbine manufacturers have generally recommended upper operating limits of 50 to 100 ppb (wt) *total* alkali in the turbine combustor fuel.¹⁹ More recent analyses summarized by Mojtahedi and co-workers suggest levels as low as 24 ppb (v) may be necessary.²⁰

Minimization of alkali levels in the fuel gas is clearly a prerequisite to successful utilization of IGCC systems. This requires: 1) a clear understanding of the mechanisms of release of alkali species during the high temperatures of air/steam gasification, 2) an analysis of the resulting partitioning between condensed and vapor phases in the cooler fuel gas, and 3) a plan for minimizing alkali levels through use of sorbents or other cleanup strategies. These issues are addressed individually in the following pages of this report, with emphasis on conditions pertinent to air-blown Lurgi gasification systems.

3.1 Alkali Release and Partitioning During Gasification

Alkali metals typically represent 2 to 4 percent by weight of the ash contained in U.S. coals. In Eastern bituminous coals, potassium is the dominant alkali metal; potassium levels in the ash are typically 2 to 10 times greater than sodium concentrations. Although

most potassium is tied up in illitic clay minerals, a small fraction is bound as cations to the carboxylic acid groups in the coal structure.^{21,22} Sodium in bituminous coals is predominantly tied to the minerals illite and montmorillonite, with a small fraction bound to carboxylic acids as well. In lower-rank coals such as Western lignite and sub-bituminous coals, the reverse is generally true - sodium is the dominant alkali metal. In these coals, both sodium and potassium are primarily acid-extractable species, bound to the organic matrix via the carboxylic acid oxygen anion. In higher moisture coals, some sodium is present as hydrated anions within the coal-bound water.^{21,22} Mineral-bound levels of both potassium and sodium are low for these coals. In selected coals from Australia and the U.K., some sodium may be present as an ionic sodium chloride solution (often identified as crystalline NaCl which is formed by drying during analysis);^{21,23} this is generally not an issue for U.S. coals, as chlorine levels are usually less than 1000 ppm of the coal, low in comparison to alkali and total ash levels.

These trends can be seen more clearly in Table 8, which presents detailed coal and ash analyses for five U.S. coals as adapted from Reference 20. For the bituminous coals, ash levels are in the range 10 to 25 percent. Most of the ash mass is silica, alumina, and iron oxide, with total alkali metal oxide levels of less than 4 percent noted. As expected, potassium levels are greater than sodium levels for each of the bituminous coals. For the sub-bituminous coal listed in the last column, sodium concentrations exceed potassium concentrations. The observed ash composition in the first four coals is indicative of a mineralogy dominated by quartz, clays, and pyrite, typical of U.S. bituminous coals. As the data for Illinois #6 presented in Table 9 indicate, quartz, clays (silicates), and pyrite comprise over 85 percent of the minerals in this particular coal. Potassium levels in these minerals are much higher than sodium levels, with illite and miscellaneous clays containing the bulk of this metal. Carboxyl-bound alkali, shown in the acid-extractable section of Table 8, reflects a higher-than-average sodium concentration for this bituminous coal.

Table 8. Fuel and Ash Analysis for Selected U.S. Coals. Data From Ref. 21
PSI Technology Company As-Received for P.C. Grind Coals

	Illinois #6 (B)	Kentucky #9 (B)	Kentucky #11 (B)	Upper Freeport (B)	Eagle Butte (SB)
Proximate					
FC	42.2	42.9	42.0	52.3	36.2
VM	35.9	34.5	33.2	24.0	33.8
Ash	8.6	14.3	21.0	23.2	4.8
Moisture	13.3	8.3	3.8	0.5	25.2
Ultimate					
C	59.9	60.0	57.7	64.6	50.3
H	4.6	4.3	4.4	4.1	3.8
O	8.5	7.9	7.6	3.9	14.6
N	1.4	1.6	1.3	1.6	1.0
S	3.8	3.5	4.2	2.1	0.3
Ash Chemistry (wt%)					
SiO ₂	46.1	44.6	46.7	50.6	28.4
Al ₂ O ₃	18.0	20.6	19.4	24.6	15.5
TiO ₂	0.9	1.1	0.9	1.2	0.9
Fe ₂ O ₃	20.5	22.4	20.0	13.5	6.1
CaO	5.3	3.1	4.4	2.2	26.4
MgO	0.6	1.1	0.9	1.4	6.2
Na ₂ O	1.3	0.9	0.3	0.3	2.0
K ₂ O	2.1	2.6	2.2	3.1	0.2
SO ₃	4.4	4.7	5.0	2.4	13.7
P ₂ O ₅	0.1	0.1	0.2	0.1	0.7
Acid-Soluble (ppm)					
Na	2180	550	370	320	820
K	2380	2500	3520	2300	80
(B) bituminous; (SB) sub-bituminous					

Table 9. Mineralogy of Illinois #6 Coal. Data from Ref. 21

Mineral Species	Elemental Composition											wt%	ρ
	Na	Mg	Al	Si	S	Cl	P	K	Ca	Ti	Fe		
Quartz				96	1			1		1	1	20	2.65
Kaolinite			41	51	4	1		1		1	1	6	2.62
Illite			28	50	5			13	1	1	2	10	2.65
Chlorite		5	21	36	2			1			35	<1	3.1
Montmorillonite	2		16	72	5			1	2	1	1	<1	2.5
Misc. Silicates		1	21	61	2			10	2	2	4	18	2.65
Fe2+ Sulfate			1	2	48				1	1	46	<1	2.5
Pyrite			1	1	65						32	28	5.0
Fe-rich (siderite)			1	4	9						82	1	4.0
Ca-rich (calcite)			1	1	2		1	1	91			7	2.75
Ti-rich (rutile)			3	4	7	1				82		<1	4.2
Apatite (Ca,P)			1	1			28	1	66	2	1	<1	3.2
Sil/Sul	1	1	15	43	23	1		7		2	7	5+	2.5
Sil/Pyr			14	48	18	1		8	1	2	10	1+	3.5
Misc. Mixed	1	3	13	30	12	8	2	9	7	10	10	2	2.5

3.1.1 Alkali Vaporization

Understanding this division between mineral-bound and organically associated alkali in coal is an important factor in determining the release of alkali as a harmful vapor phase. Although traditionally, absolute levels of alkali metal have been used to assess the extent of alkali release (as implied by the use of total alkali in calculating fouling factors for utility boiler deposition²⁴), they do not provide an indication of the level of vaporization and emission which may occur. In fact, the absolute level of sodium or potassium in the coal shows little correlation with the amount of material released to the vapor phase.

In a 1981 summary report, Tamhankar and Wen noted that the total ash level in the coal must also be considered in determining the amount of alkali metal vaporized.²⁵ Peterson and co-workers have shown that it is the silica level, rather than total ash, which is important in determining vaporization of organically-bound alkali.^{21,26} The fraction of sodium vaporized was found to correlate positively with the level of organically dispersed

sodium in the coal, and inversely with the level of silica in the coal. Coals with high silica to sodium ratios demonstrated less than 1 percent alkali vaporization, while coals with low silica levels demonstrated up to 40 percent sodium vaporization.²⁶ Consistent with a previous Australian study,²⁷ the authors concluded that silicates can react with organically bound sodium, acting as a sponge to reduce the amount of alkali ultimately found in the vapor phase.

The ability of aluminosilicate minerals to absorb and retain organically bound alkali, thus reducing vaporization, can be seen in the composition diagrams of Figure 9. Figure 9a presents mineral compositions for clays and quartz in an Illinois #6 coal, as measured by computer-controlled scanning electron microscopy.²⁸ Figure 9b is the individual potassium aluminosilicate ash particle compositions, while Figure 9c is the individual sodium aluminosilicate ash particle compositions, resulting from combustion at 1750 K, 7 percent oxygen. In both cases, average alkali content of the ash particles was higher than that of the parent minerals. The scatter in Figures 9b and 9c indicates a range of ash particle compositions, with particles containing up to 40 percent K and 10 percent Na observed. As these particles were generated during the elevated particle temperatures of pulverized coal combustion, the increased alkali levels confirm the ability of the silicates to retain sodium and potassium throughout combustion, thus reducing vaporization of the organically-bound species.

Tamhankar and Wen have suggested that up to 10 percent of the mineral-bound alkali may be released during gasification;²⁵ recent studies under higher-temperature combustion conditions indicate significantly lower levels. Srinivasachar and co-workers²⁹ conducted combustion and vaporization experiments with 10 to 20 μm illite inclusions embedded in a synthetic coal matrix. Recall that illite is an aluminosilicate clay mineral typical of bituminous coals, generally containing 7 to 10 percent potassium. Under oxidizing conditions (1500 K, 3 percent oxygen), the authors found no evidence of potassium loss or vaporization. In a separate study using 1 to 3 μm illite inclusions also embedded in a synthetic carbon, researchers at MIT noted less than 2 percent vaporization of potassium at

Figure 9. K-Al-Si and Na-Al-Si Mineral and Ash Particle Composition. Ash Generated During Combustion at 1750 K, 7 percent O₂. Data from Ref. 19.
 (a) Mineral; (b) K-Al-Si; and (c) Na-Al-Si ash

particle temperatures of approximately 2000 K.²¹ Vaporization from these small minerals provides an upper bound for illitic potassium vaporization in coal combustion systems, as the amount vaporized will decrease with increasing mineral size.³⁰ As greater than 95 weight percent of illite inclusions are greater than 1 to 3 μm in bituminous coals,²¹ little vaporization of illitic potassium is expected. Gallagher et al.²⁶ suggest that a K (illite) - Na (organically bound) exchange may occur, liberating potassium from illite; this would have the effect of reducing sodium vaporization, however, resulting in no net increase in vaporizable alkali.

It can therefore be assumed that the dominant source of vaporized alkali results from the carboxyl-bound species, with the amount vaporized inversely proportional to the total silica level of the coal. No significant alkali vaporization is expected from mineral species such as illite, even under the high temperatures of pulverized coal combustion. As shown in Table 10, less than 1 percent of the sodium and 0.20 percent of the potassium vaporized for all bituminous coal considered in the Reference 20 study. Measurements in Table 10 are based upon atomic absorption analysis of the parent coal and size-fractionated ash samples. Three low-rank coals, presented for comparison, generated submicron fume rich in alkali metals as a result of increased sodium and potassium vaporization. This increase in vaporization was caused by an increase in the fraction of alkali present as inorganic species, with a corresponding reduction in the silica to alkali ratio of the ash.²¹

3.1.2 Predicted Alkali Release from Lurgi Gasifiers

For predictive purposes, alkali vapor levels generated during coal combustion can be used to estimate alkali vapor levels generated during gasification. In Lurgi dry-bottom gasifiers, char combustion occurs below ash fusion conditions, at temperatures of approximately 1250 to 1500 K, corresponding to 1800 to 2250°F.³¹ These temperatures are several hundred Kelvin lower than the flame temperatures in combustors burning bituminous coals. Because the extent of alkali vaporization will decrease with decreasing temperature²¹ and increasing pressure,²⁵ levels predicted under pulverized coal conditions provide a

Table 10. Partitioning of Alkali During Pulverized Coal Combustion*

Coal	Na as pct of Fume	K as pct of Fume	Percent Vaporized		SiO ₂ Mesh
			Na	K	
Illinois #6 (B)	3	1	0.6	0.08	46
Kentucky #9 (B)	2	1	1.0	0.20	45
Kentucky #11 (B)	5	3	0.6	0.07	47
Upper Freeport (B)	1	1	0.6	0.1	51
Eable Butte (SB)	1	1	2	2	28
Beulah (L)	68	3	6	7	22
Loy Yang (AB)	61	1	41	17	--
(B) bituminous; (SB) sub-bituminous; (L) lignite; (AB) brown, Australia					
*Data of Peterson, T.W., and Wendt, J.O.L., as presented in Ref. 21					

reasonable upper bound for Lurgi gasifier conditions. Note that it is the combustion temperatures in the gasifier which will determine alkali release in Lurgi systems.

Equilibrium calculations reporting Lurgi alkali vapor levels in the 50 ppb range have used the gasifier exit temperature as the basis for determining alkali release;³² these yield unrealistically low levels of alkali vaporization.

By assuming that combustion levels of vaporized alkali are representative of the gasifier combustion section, one can calculate maximum vapor concentrations of alkali species in the gasifier environment. The results of these calculations for several of the Table 10 coals are given in Table 11. Mass flow rates of coal, air, and steam provided by CRS Sirrine for the Lurgi system under consideration were used. Total initial alkali vapor levels of 1.5 to 5 ppm result from this analysis. Note that the highest bituminous coal emissions are for the Kentucky seam #9, with the corresponding highest flame temperature; it is reasonable to assume that these levels would be consistent with the other coals at a similar (lower) temperature. By decreasing temperature further to 1500 K

Table 11. Calculated Vapor Phase Levels in Gasification Zone, for Air-Blown Lurgi Gasifier at Conditions of this Report. Based upon data from Table 10; Temperatures from Ref. 21

Coal	Na (ppm, wt)	K (ppm, wt)	T (K)*
Illinois #6 (B)	1.2	0.4	1650
Kentucky #9 (B)	3.4	1.7	1750
Kentucky #11 (B)	1.0	0.9	1600
Upper Freeport (B)	1.1	1.9	1600
Eagle Butte (SB)	5.8	0.5	1680
*Peak flame temperature, air/fuel stoichiometric ratio = 1.2 (combustion)			

gasification/combustion conditions, a total initial alkali vapor level of 1 ppm appears to be a reasonable value. In the study of alkali evolution throughout the IGCC system presented in the following sections, 1.2 and 3 ppm were used as initial conditions, simulating the expected range of initial alkali levels.

If combustion/alkali vaporization data (e.g., Table 10) is not available for a particular coal, an estimate of expected alkali emissions can be made from analysis of the bulk ash composition. For coals with similar SiO_2/Na ratio, a reasonable rule of thumb is 0.75 percent total alkali vaporized during combustion, with 50 to 75 percent of that amount being sodium. Under Lurgi dry-bottom gasifier conditions, 50 percent of the value calculated for combustion conditions should provide a reasonable estimate. With this as a basis, equilibrium can then be used to predict species concentrations at various points within the system and determine whether alkali emissions will pose a threat to the gas turbine.

3.2 Evolution of Alkali Species

Using both 1.2 and 3.0 ppm (wt) sodium vapor levels as initial conditions, equilibrium calculations were performed at various locations within the IGCC system to

identify the alkali species and phases favored at each point. A modified version of the SOLGASMIX free-energy minimization equilibrium code developed by Eriksson³³ was used in these calculations. This code predicts the different phases - gaseous, liquid mixtures, and invariant condensed compounds - present at equilibrium, given the temperature, pressure, and initial quantities of each element in the system. Potential non-idealities in liquid systems are accounted for by addition of conformal ionic solution theory (CIS) "rules" to the code. CIS is used because of the ionic nature of mixed melts - chlorides, sulfates, etc. - which might exist in the condensed phases of this system. Details of CIS³⁵ and its application to SOLGASMIX^{36,37} are provided elsewhere.

Calculations were performed with an Illinois #6 coal as the feedstock, using the fuel analysis shown in Table 8. As stated previously, mass flows of coal, air, and steam from the overall gasifier mass balance discussed elsewhere in this report were used. Sodium was the only alkali species considered in these calculations, as it comprises 50 to 75 percent of the total alkali vapor generated during combustion of Illinois #6 and similar coals (cf. Table 10). Chlorine levels were set at 1000 ppm of the coal by weight, as measured for this Illinois #6 sample.²¹ Calculations corresponded to conditions at the gasification zone of the Lurgi gasifier, the gasifier exit, the cleaned (desulfurized) fuel gas stream, the turbine combustor outlet, and the turbine outlet; these are summarized in Figure 10 and Table 12.

In the previous section, analysis of existing vaporization data from coal combustion systems indicated that a reasonable starting level of vaporized alkali was 1 to 3 ppm within the gasifier. To determine the specific alkali species existing under these conditions, equilibrium calculations at 1500 K (2240°F), 18.8 atm pressure (276 psia) were run. A temperature of 1500 K was selected to represent the higher end of the temperature range attainable within the dry-bottom gasifier. Initial sodium concentrations were set at 1.2 ppm. Under these conditions, all of the alkali (sodium) was in the vapor phase. Sodium chloride was the dominant species, present at nearly 1.2 ppm levels; other vapor phase compounds contained less than 3 percent of the total sodium. Repeating the calculations at 1100 K (1520°F), representative of the combustion/gasification interface, yielded similar results.

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Figure 10. Schematic Showing Approximate Locations of Sodium Equilibrium Calculations

Table 12. Conditions of Sodium Equilibrium Calculations

Location	Figure 9 Notation	T (K)	P (psia)	Input Na Conc'n (ppm)	Comments
Gasifier	A	1500	276	1.2	Gasification zone conditions
	(A')	1100	276	3	Gasification/devolatilization zones
		1050	276	3	
		1050	276	1.2	Examine range of Na
Gasifier Exit	B	973	276	1.2	Max T at exit (1300°F)
Fuel Gas	C	822	276	1.2	Min T at exit; also post H ₂ S cleanup fuel gas
Combustor	D	1500	201	*	Oxidizing conditions
Turbine Exhaust	E	876	15	*	Identify condensed phases at exit
* 1 to 2 ppm gasifier exit with combustion air added					

NaCl vapor was the dominant species, representing 93 percent of the total sodium. The sodium chloride dimer, Na_2Cl_2 , constituted most of the remainder at both temperatures. No condensed sodium phases were formed at either temperature. Note that kinetic calculations have shown that formation of the chlorides will occur rapidly under these conditions indicating that equilibrium is a reasonable approach.²¹

Similar results were also obtained at a temperature of 1050 K (1430°F) and a pressure of 18.8 atm. Under these conditions, no condensed phases were formed. Over 91 percent of the vapor was NaCl, resulting in an NaCl concentration of nearly 1.2 ppm. Thus, no alkali species are expected to condense within the coal bed at temperatures typical of steady-state Lurgi gasifier operation.

To explore the effects of extreme pressure changes on sodium equilibrium at this temperature, these calculations were repeated at 1 atm total pressure. Although the Lurgi system will not be run under these conditions, they provide a means for determining whether extreme shifts in equilibrium would result from a pressure decrease. Despite large changes in product distribution for the bulk fuel gas (eg., at 1050 K, more hydrogen, less methane at atmospheric pressure), little difference was noted in alkali species distribution, showing an insensitivity to system pressure at this temperature. Sodium chloride remained dominant (99 percent); as expected, no condensed phases were formed. Initial sodium level effects were also explored in these calculations by assuming an input vapor concentration of 3 ppm sodium. Again, sodium chloride was dominant and present at levels of 2.6 ppm, with slightly larger relative concentrations of the sodium chloride dimer were higher (17 percent of the total sodium under these conditions).

As the gases flow upward through the gasifier, gas temperatures decrease to 1000 to 1300°F, corresponding to 811 to 973 K. Calculations at these conditions can therefore assess any changes in the dominant species with decreasing temperature, and can also point to the onset of condensation for alkali species. At 973 K, a pressure of 18.8 atm, and an initial sodium level of 3 ppm, sodium chloride remained the dominant vapor phase species.

A significant condensed phase, however, existed under these conditions. At 973 K, fully 18 percent of the total sodium was present as condensed crystalline NaCl. Decreasing the temperature further to 822 K, corresponding to the gasifier exit condition of 1020°F, caused 98 percent of the sodium to be present as condensed phase NaCl (using 1.2 ppm as the initial condition; raising the starting level to 3 ppm would only increase the amount of condensed NaCl present). Re-evaluation of this result in the absence of hydrogen sulfide (post-gas cleanup) yielded an identical result.

The condensation of NaCl at conditions approaching the gasifier exit serves to reduce the vapor phase alkali concentrations to < 25 ppb (wt). Although this level is within acceptable turbine operating limits, the remainder of the sodium present as a condensed phase must be considered in the system analysis. As the calculations have shown, condensation of volatile sodium occurs in the upper, cooler portions of the gasifier. At this point, most of the silicates contained in the ash is collected at the bottom of the gasifier and is unavailable for reaction with the alkali. Condensation will therefore occur heterogeneously on the surfaces of small entrained ash and carbon particles. Although sodium concentrations are sufficiently low that a homogeneous sodium fume is unlikely, experience in coal combustion systems suggests that the bulk of this condensation will be on particles that are submicron in size since these particles would have a very high surface area.^{21,38} Experience in pulverized coal has shown that these particles have a stratified composition, with cores rich in Mg, Ca, Fe, and Si and surfaces rich in alkali.^{20,43} Because alkali levels are much greater than silica levels in these particles minimal silicate formation is observed in combustion systems. Therefore silicate formation in submicron particles in gasification systems is not expected. These particles are difficult to remove from gas streams with cyclones, and may therefore propagate downstream to the turbine combustor.

It is also possible that alkali could condense on larger, cold coal particles injected into the gasifier. If this occurs, the condensed alkali will move deeper into the coal bed until temperatures are high enough to revaporize it. Such an internal alkali "reflux" loop would have minimal impact on the alkali content of the gasifier product gas.

As the results summary in Table 13 shows, this situation will likely give rise to deposition of corrosive species on turbine surfaces. At the high temperatures of the combustor all of the condensed NaCl will revaporize; no condensed alkali phases will exist through the combustor. Both NaCl and NaOH will be present in the vapor phase under these oxidizing conditions. Total vapor phase alkali levels are of order 300 ppb, a factor of 3 to 10 above the range recommended for turbine inlet conditions. As the gases expand through the turbine portion of the system, condensation will occur, leading to formation of the condensed, corrosive sulfate phase as indicated in Table 6. Although kinetic limitations likely preclude formation of sulfate the vapor phase sulfate, it will form condensed sodium species which will react rapidly to form Na_2SO_4 under these conditions.³⁹

Table 13. Summary of Sodium Equilibrium Calculations
(complete list of conditions, Table 12)
Na input levels = 1.2 ppm (wt) in vapor phase

Location	T (K)	NaCl (g) ppm (wt)	Total Na Vapor Species ppm (wt)	Condensed Phases
Gasifier	1500	1.2	1.2	0%
	1050	1.2	1.2	0%
Gas Exit*	973	1.6	2.2	18% of Na (NaCl)
Fuel Gas	822	0.02	0.03	98% of Na (NaCl)
Combustor	1500	0.23	0.30	0%
Turbine Exhaust	876	0.002	0.002	> 99.9% (Na_2SO_4)
*input level 3 ppm for this condition only				

3.3 Strategies for Alkali Removal

Equilibrium calculations performed for the products of Illinois #6 coal gasification in an air-blown Lurgi gasifier/IGCC system suggest that NaCl vapor will be the dominant sodium phase within most of the gasifier. Previous results¹⁹ as well as the similarity of the alkali metals suggest that vapor phase KCl will be the dominant potassium species under

these conditions. As the gases approach the gasifier exit, however, condensation of the volatile chlorides will occur preferentially on the smallest ash and carbon particles within the system. As these particles are not effectively removed by cyclone collection devices, they will propagate downstream to the turbine combustor, where re-vaporization will occur. This will be followed by condensation and formation of alkali sulfate within the turbine expander portion of the system, leading to hot corrosion and degradation of system metal components.

To avoid this scenario, steps must be taken to remove the alkali vapor from the process gas stream before it can condense within the gas turbine. Capture of alkali metals with appropriate sorbents is the most promising method; a fair amount of literature is available on this subject.²⁴ Based upon this review as well as recent literature, several promising candidate materials can be identified. These are summarized in Table 14.

3.3.1 Discussion of Potential Sorbent Materials

Kaolinite and emathlite are effective getters for alkali species at high temperatures. Upon reacting with sodium chloride, both materials have been shown to form stable sodium aluminosilicates which do not lose sodium even at the elevated temperatures of combustion.^{32,40} Studies in pulverized coal combustion systems have shown that kaolinite in particular is one of the most effective alkali getters, significantly reducing alkali-driven deposit strength when added to the pulverized coal feed at 3 percent levels.⁴⁰ Emathlite has also been shown to be an effective sink for alkali chlorides in the vapor phase, showing significantly higher alkali sorption capacity at 1123 K and 11 atm than many sorbents.³² Although a bench scale kinetic study showed it to be nearly as effective as kaolinite, the formation of a relatively low melting albite phase makes this material unsuitable for applications above 1300 K.⁴¹

Kaolinite and emathlite appear to be the most promising aluminosilicate sorbent materials, although studies with other compounds are continuing. Some researchers have suggested that diatomaceous earth may also be effective, but it is more expensive than

Table 14. Summary of Potential Alkali Sorbents

Location	References	Comments
Kaolinite	23, 38, 39	Proposed for deposit remediation in coal-fired utility boilers Forms chemically stable phase
Emathlite	17, 30, 39	Forms chemically stable phase
Diatomaceous Earth	23, 38	
Silica	17, 23, 38	High capacity, expensive
Bauxite	17, 23, 40	Potentially high capacity; may work at high T Regenerable (reversible physical adsorption)

emathlite or kaolinite (see Table 15). High surface area silica gels are also effective getters, but these soften at combustion temperatures, and more importantly, are extremely costly.

Activated bauxite, the lone non-siliceous sorbent in Table 14, is also an effective alkali chloride getter material. Initial removal rates are comparable to those of kaolinite,⁴¹ while sorption capacities are relatively high. Research at Westinghouse has suggested that emathlite, kaolinite, and diatomaceous earth are all more effective getters,³² although contrasting results obtained at Argonne suggest that activated bauxite may be superior.

Table 15. Estimated Alkali Sorbent Costs (as-delivered);
Selected Sulfur Sorbents Included for Comparison
Data from Reference 42 or as noted

Material	Cost (\$/ton)
Lime	75
Limestone	30
Kaolin*	100
Diatomaceous Earth	320
Trona	130
*Estimated, based upon data of Ref. 23	

Calculations by Scandrett and Clift also suggest the superior gettering ability of aluminosilicates relative to bauxite, based upon equilibrium analysis of the resulting stable chemical phases.¹⁹ This analysis does not consider the finding that activated bauxite collects alkali primarily by physical adsorption. Removal efficiencies of over 90 percent NaCl from simulated pressurized fluidized bed combustion (PFBC) flue gases have been reported;²⁵ research at Argonne has shown that 99.8 percent can be achieved if the sorbent is first heat treated at 1173 K to remove any inherent alkali.⁴² Because it collects alkali by physical adsorption, activated bauxite is relatively easy to regenerate with a water wash. Additional facilities, equipment, and costs associated with sorbent regeneration and wash-water treatment may, however, inhibit wide-scale application to IGCC systems. Research is continuing in this area.

3.3.2 Application of Sorbents to IGCC System

In considering application of these materials to an IGCC system utilizing an air-blown Lurgi gasifier, the following criteria were employed: 1) No additional process equipment should be required. Sorbent addition should be at an existing location within the system if possible; 2) No cooling of the process gas stream shall be permitted; 3) Sorbent costs, taking into account the sorption capacity of the material, shall be as low as possible.

With these system constraints, two options for sorbent addition are available. The first, addition with the coal feed to the top of the gasifier, requires a material which reacts with alkali at gasification temperatures, and retains the alkali in an inert form at higher combustion temperatures existing in the lower portions of the gasifier. The sorbent must not form a molten phase at low temperatures, or clinkering within the gasifier will occur. A throwaway sorbent is essential, given the difficulty of separating ash from sorbent at the gasifier exit. The second possible location for sorbent addition is the H₂S absorption train downstream of the first cyclone. The packed bed reactor at this location is ideal for addition of a sodium sorbent, provided it does not interfere with the zinc ferrite already present. Both bauxite and the aluminosilicates could be added at this location if a mechanism for

separating the zinc ferrite sorbent from the alkali sorbent (e.g., smaller alkali sorbent particles, facilitating aerodynamic separation with cyclones) could be devised. Temperature may provide the largest detriment to alkali sorption, however, as equilibrium calculations (Section 2) have shown that a condensed sodium phase will exist at the 850 K temperature of the absorption train. If condensation has occurred on high surface area submicron ash particles as expected, it will be difficult to capture with sorbents in the 10 to 15s gas residence time available.

For these reasons, addition of kaolin-based sorbents to the gasifier is the most promising option. Minerals such as kaolinite are inexpensive, can function at the high temperatures of gasification and combustion where alkali-aluminosilicate reaction rates will be highest, and will form stable phases. Materials treatment and handling problems are therefore avoided, as the material becomes part of the gasifier bulk ash.

At the present time, the exact amount of kaolinite required to reduce the alkali concentration to acceptable limits is uncertain. Although some kinetic data for simulated flue gas conditions exist, more information is needed on the performance of this and other additives under air-blown Lurgi gasifier conditions. Approximate costs can, however, be estimated as follows: Based upon experience in coal-fuel boilers,⁴⁰ kaolin addition at 3 percent of the coal feed was effective in minimizing alkali-initiated fouling, while laboratory experiments have suggested that kaolin/NaCl ratios of 10 cause significant reductions in alkali volatilization.²⁵ Assuming that the appropriate kaolin loading is therefore 5 to 10 percent of the coal fed, a kaolin cost of \$100/ton would equal \$5 to 10 per ton of coal gasified. Process equipment and ash handling costs are assumed to be unchanged from the base plant in this analysis.

To reduce these costs further, it may be possible to substitute the recycled ash for a portion of the kaolin in the feed. As was discussed previously, U.S. bituminous coals are rich in kaolinite and other silicates. This material already reacts with the bulk of the alkali in coals during combustion.^{21,26} One reason alkali retention by the ash is less than

100 percent is the inability of the minerals within the coal particles to contact all of the vaporizing alkali. By recycling the ash, this problem would be eliminated. Using it as a kaolin supplement would therefore reduce raw materials costs, and decrease slightly the amount of ash to be disposed of. Recycling illite-derived ash particles (potassium-containing; cf. Table 9) through the reactor should not pose problems, as these are inert to vaporization due to the extremely low activity of alkali in an aluminosilicate melt.²¹ Kinetic information regarding the reactivity of these ash particles in gasification atmospheres must first be generated before this option can be considered further.

3.3.3 Other Alkali Removal Strategies

Clearly, removal of alkali vapors with well-characterized sorbents in existing process equipment is the best-established route for minimization of this problem. If this is not possible, other approaches should be considered. Construction of a packed sorbent bed, for example, has been suggested for alkali filtration in PFBC systems to remove alkali vapors. This approach, however, is likely inapplicable for an air-blown Lurgi IGCC system. Low process temperatures from the gasifier exit onwards will cause a large fraction of the alkali to exist in a condensed phase, most probably as a coating on submicron ash particles.^{21,43} These particles are extremely difficult to remove without filtration. A packed bed to accomplish this may produce an unacceptably high pressure drop, making this option impractical. Electrostatic precipitation (ESP), as used in utility boilers, may provide a means for capturing the alkali-coated particles without the need for a high pressure drop filter or packed bed. If the condensed particles could be removed, remaining alkali vapor levels would be within turbine tolerances. An additional advantage is the need to treat only the fuel gas entering the turbine combustor - gas entering the HRSG would not require treatment, thus reducing the size of the precipitator needed. Disadvantages include high capital cost of ESP's as well as the difficulty of capturing 0.1 to 1 μm particles by electrostatic precipitation due to a decrease in collection efficiency for 0.1 to 1 μm particles. Data describing ash particle sizes and alkali surface enrichment would first be required to insure that the particles can be captured by ESP's.

Two other options for alkali control have been discussed briefly in the literature and bear mentioning. In a study at General Electric,⁴⁵ a surface ionization technique which selectively ionizes alkali species and then collects them on cooled electrodes has been proposed as a potential means of alkali collection. This technology is in the research stages; effectiveness under Lurgi/IGCC conditions needs to be determined. Also at General Electric, addition of a submicron fume in the turbine combustor portion of the system has been suggested as a means of avoiding condensation on downstream turbine surfaces. Fumed silica, titania, or other smokes would be generated by combustion of the appropriate metal chloride, generating high surface area 100Å particles. These fine particles would then serve as condensation sites for the volatile alkali species during expansion in the turbine, thus avoiding deposition on blade surfaces. Questions of amounts required, costs of setting up the combustion system, prevention of erosion (via prevention of agglomeration), effectiveness, and downstream cleanup remain to be answered in this proposed technology.

4. CONCLUSIONS AND RECOMMENDATIONS

NO_x emissions from the CGT of an IGCC system can be cut by about 50 percent using a rich-lean combustor on the gas turbine. Resulting NO_x emissions are 0.5 to 0.6 lb/MBtu. Additional reductions can be achieved in supplemental firing of the HRSG using coal gas as the fuel and the NO_x bearing TEG as an oxygen source, resulting in a type of reburning. Additional air will be required via a separate forced draft fan to provide an environment for stable combustion. Unstaged supplemental firing results in a 20 percent NO_x reduction primarily due to lower conversion of ammonia to NO_x in the second flame (0.4 to 0.5 lb/MBtu). NO_x reductions could be greater due to interaction of the NH_3 and NO_x . Substoichiometric supplemental firing results in a 65 percent reduction in NO_x emissions (0.2 lb/MBtu).

In order to achieve the goal of 0.1 lb/MBtu NO_x emissions, post combustion NO_x control will be required. SCR must be used if there is no supplemental firing or unstaged supplemental firing. If staged supplemental firing is employed then SNR can be used, resulting in an order of magnitude decrease in capital costs. However, the cost of other subsystems such as the HRSG, and gasifier, if the CGT system size is held constant, will increase.

The use of supplemental firing could decrease the gross heat rate, increasing power production cost. It also shifts power production from the CGT to the steam turbine. However, the supplemental firing of the HRSG will also allow higher steam temperatures and more sophisticated steam cycles which would mitigate reductions in heat rate. To limit use of supplementary fuel innovative burner designs will be required which provide stable combustion with O_2 concentration below 13 percent.

Analysis of literature and recent experimental results in the pulverized coal combustion field has provided guidelines for the amount of alkali vaporization expected in an air-blown Lurgi gasifier. Alkali vaporization from the coals being considered - U.S.

bituminous - will depend upon the amount of organically bound alkali in relation to the amount of total silicate minerals in the coal. For these coals, a reasonable estimate is that 0.25 to 0.50 percent of the total alkali in the coal will vaporize during gasification, producing total alkali vapor levels of 1 ppm (wt) in the product gas.

Once the alkali has entered the vapor phase, it will be converted to the chloride form, as shown by equilibrium calculations for the sodium system. Over 90 percent of the total sodium released from the coal will remain as chloride vapor throughout the gasifier. As temperatures decrease in the upper portions of the gasifier, condensation of sodium chloride will occur. If small, high surface area ash particles are present, the alkali chloride will condense to form a coating on these. In the absence of these ash particles, the majority of the sodium chloride will condense homogeneously to form a submicron fume. In both cases, the particles will not be removed by the cyclones, and will instead escape downstream to the turbine combustor. Vaporization will then occur, followed by recondensation and corrosive sodium sulfate formation on turbine surfaces.

To avoid this corrosion problem, alkali must be removed prior to entering the turbine combustor. Based upon analysis of current alkali control strategies, the most promising technique is removal of the vapor *within the gasifier* with an aluminosilicate sorbent such as kaolinite. Once the vapors have escaped the gasifier, the difficult-to-capture condensed alkali phases will dominate. Further evaluation is needed, however, prior to implementation of a full-scale alkali control strategy. Kinetic rates for alkali capture within the gasifier environment must be ascertained. Optimum particle sizes and sorbent capacities must also be determined. As a cost-reduction mechanism, the possibility of supplementing the kaolinite sorbent with recovered ash needs to be addressed. Ash from gasification of bituminous coals will contain a high aluminosilicate content, making it an ideal choice for alkali capture and removal once the appropriate kinetic parameters have been determined for this material as well.

PSIT recommends that future work on this process includes the following tasks:

- Pilot-scale combustion testing of simulated coal gas containing NH_3 using simulated TEG containing NO_x to more accurately assess the NO_x reduction potential of supplemental firing of the HRSG and flame stability limits of low Btu gas in TEG. These tests should be carried out at 1 to 10 MBtu/hr so that mixing rates in the pilot system will be similar to full scale. The TEG could be simulated by the combustion of oil with air added to adjust the oxygen and NO_x concentrations. Coal derived gas could be simulated by mixing bottled gases.
- Process modelling of the entire IGCC system including air compression, coal gasification, and steam generation and expansion systems to better assess the economic and efficiency aspects of supplemental firing of the HRSG. This would allow CRS Sirrine to assess the effects of gasifier, pressure and temperature, CGT inlet pressure, steam pressure, temperature and cycle design in a cost effective manner.
- Limited laboratory kinetic evaluation of alkali sorbents, followed by pilot-scale testing of alkali metal sorbents to determine the practical limits of reduction. The practicality of achieving 25 to 100 ppb total alkali vapor concentrations in the gas turbine must be assessed. Key variables are sorbent feed size, and ash recycle rate. These experiments could be carried out in a small gasifier similar to the one described in reference 16. A pilot-scale gasifier could also replace the mixing of bottled gases in the pilot-scale combustion tests.

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APPENDIX A

**7111E COMBUSTION GAS TURBINE WITH AN UNFIRED
HEAT RECOVERY STEAM GENERATOR**

Figure A-1. Process Flowsheet for the 7111EA Combustion Gas Turbine with an Unfired Heat Recovery Steam Generator

**Table A-1. Stream Properties for the 7111EA Combustion Gas Turbine
with an Unfired Heat Recovery Steam Generator**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	487,345	1130	279
(2) First Stage Air	401,500	707	194
(3) First Stage Exit	888,845	2639	194
(4) Second Stage Air	1,530,692	707	194
(5) Second Stage Exit	2,419,537	1951	194
(6) Cooling Air	169,848	707	194
(7) Turbine Exhaust Gas	2,589,385	1608	15
(8) Superheater Exit Gas	2,589,385	861	15
(9) Boiler Exit Gas	2,589,385	467	15
(10) Economizer Exit Gas	2,589,385	245	15
(11) Boiler Feed Water	381,089	59	1500
(12) Economizer Exit Water	381,089	450	1500
(13) Saturated Steam	381,089	598	1500
(14) Superheated Steam	381,089	950	1500

APPENDIX B

**7111E COMBUSTION GAS TURBINE WITH A FIRED
HEAT RECOVERY STEAM GENERATOR, UNSTAGED COMBUSTION**

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Figure B-1. Process Flowsheet for the 7111EA Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Unstaged Combustion

**Table B-1. Stream Properties for the 7111EA Combustion Gas Turbine with
a Fired Heat Recovery Steam Generator, Unstaged Combustion**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	487,345	1130	269
(2) First Stage Air	401,500	707	194
(3) First Stage Exit	888,845	2639	194
(4) Second Stage Air	1,530,692	707	194
(5) Second Stage Exit	2,419,537	1951	194
(6) Cooling Air	169,848	707	194
(7) Turbine Exhaust Gas	2,589,385	1008	15
(8) Fresh Air	1,499,994	59	15
(9) Burner Oxidant	4,089,379	687	15
(10) Burner Fuel	2,338,818	1130	15
(11) Furnace Exit Gas	6,428,197	1466	15
(12) Superheater Exit Gas	6,428,197	982	15
(13) Economizer Exit Gas	6,428,197	245	15
(14) Boiler Feed Water	3,563,727	59	1500
(15) Economizer Exit Water	3,563,727	450	1500
(16) Saturated Steam	3,563,727	598	1500
(17) Superheated Steam	3,563,727	950	1500

APPENDIX C

**7111E COMBUSTION GAS TURBINE WITH A FIRED
HEAT RECOVERY STEAM GENERATOR, STAGED COMBUSTION**

B-5189

Figure C-1. Process Flowsheet for the 7111EA Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Staged Combustion

Table C-1. Stream Properties for the 7111EA Combustion Gas Turbine with
a Fired Heat Recovery Steam Generator, Staged Combustion

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	487,345	1130	269
(2) First Stage Air	401,500	707	194
(3) First Stage Exit	888,845	2639	194
(4) Second Stage Air	1,530,692	707	194
(5) Second Stage Exit	2,419,535	1951	194
(6) Cooling Air	169,848	707	194
(7) Turbine Exhaust Gas	2,589,385	1008	15
(8) Fresh Air	3,231,508	59	15
(9) Burner Air	1,499,944	59	15
(10) Overfire Air	1,713,514	59	15
(11) Burner Oxidant	4,089,379	687	15
(12) Burner Fuel	3,673,033	1130	269
(13) Furnace Exit Gas	9,493,926	1510	15
(14) Superheater Exit Gas	9,493,926	1010	15
(15) Economizer Exit Gas	9,493,926	245	15
(16) Boiler Feed Water	5,465,390	59	1500
(17) Economizer Exit Water	5,465,390	450	1500
(18) Saturated Steam	5,465,390	598	1500
(19) Superheated Steam	5,465,390	950	1500

APPENDIX D

**LM/TG 5000PC COMBUSTION GAS TURBINE WITH AN UNFIRED
HEAT RECOVERY STEAM GENERATOR**

Figure D-1. Process Flowsheet for the LM/TG 5000PC Combustion Gas Turbine with an Unfired Heat Recovery Steam Generator

**Table D-1. Stream Properties for the LM/TG 5000 PC Combustion Gas Turbine
with an Unfired Heat Recovery Steam Generator**

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	151,931	1020	469
(2) First Stage Air	158,520	968	394
(3) First Stage Exit	310,451	3082	394
(4) Second Stage Air	637,451	968	394
(5) Second Stage Exit	947,531	2197	394
(6) Cooling Air	75,600	968	394
(7) Turbine Exhaust Gas	1,023,131	962	15
(8) Superheater Exit Gas	1,023,131	823	15
(9) Boiler Exit Gas	1,023,131	425	15
(10) Economizer Exit Gas	1,023,131	245	15
(11) Boiler Feed Water	137,004	59	1500
(12) Economizer Exit Water	137,004	400	1500
(13) Saturated Steam	137,004	598	1500
(14) Superheated Steam	137,004	950	1500

APPENDIX E

**LM/TG 5000PC COMBUSTION GAS TURBINE WITH A FIRED
HEAT RECOVERY STEAM GENERATOR, UNSTAGED COMBUSTION**

8-5188

Figure E-1. Process Flowsheet for the LM/TG 5000PC Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Unstaged Combustion

Table E-1. Stream Properties for the LM/TG 5000PC Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Unstaged Combustion

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	151,931	1020	469
(2) First Stage Air	158,520	968	394
(3) First Stage Exit	310,451	3082	394
(4) Second Stage Air	637,080	968	394
(5) Second Stage Exit	947,531	2197	394
(6) Cooling Air	75,600	968	394
(7) Turbine Exhaust Gas	1,023,131	962	15
(8) Fresh Air	412,902	59	15
(9) Burner Oxidant	1,436,033	719	15
(10) Burner Fuel	632,117	1020	15
(11) Furnace Exit Gas	2,068,150	1520	15
(12) Superheater Exit Gas	2,069,150	971	15
(13) Economizer Exit Gas	2,068,150	245	15
(14) Boiler Feed Water	1,236,694	59	1500
(15) Economizer Exit Water	1,236,694	400	1500
(16) Saturated Steam	1,236,694	598	1500
(17) Superheated Steam	1,263,694	950	1500

APPENDIX F

**LM/TG 5000PC COMBUSTION GAS TURBINE WITH A FIRED
HEAT RECOVERY STEAM GENERATOR, STAGED COMBUSTION**

B-5189

Figure F-1. Process Flowsheet for the LM/TG 5000PC Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Staged Combustion

Table F-1. Stream Properties for the LM/TG 5000PC Combustion Gas Turbine with a Fired Heat Recovery Steam Generator, Staged Combustion

Stream (No.) Name	Flow lb/hr	Temperature °F	Pressure psia
(1) Combustor Fuel	151,931	1020	469
(2) First Stage Air	158,520	968	394
(3) First Stage Exit	310,451	3082	394
(4) Second Stage Air	637,080	968	394
(5) Second Stage Exit	947,534	2197	394
(6) Cooling Air	75,600	968	394
(7) Turbine Exhaust Gas	1,023,131	962	15
(8) Fresh Air	1,015,381	59	15
(9) Burner Air	412,902	59	15
(10) Overfire Air	602,479	59	15
(11) Burner Oxidant	1,436,033	719	15
(12) Burner Fuel	993,275	1020	15
(13) Furnace Exit Gas	3,031,787	1547	15
(14) Superheater Exit Gas	3,031,787	986	15
(15) Economizer Exit Gas	3,031,787	245	15
(16) Boiler Feed Water	1,853,023	59	1500
(17) Economizer Exit Water	1,853,023	400	1500
(18) Saturated Steam	1,853,023	598	1500
(19) Superheated Steam	1,853,023	950	1500

Development of Standardized Air-Blown Coal Gasifier/
Gas Turbine Concepts for Future Electric Power Systems
Volume III
Appendix B: NO_x and Alkali Vapor Control Strategies

END

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