Characterization and Prediction of Oxy-combustion Impacts in Existing Coal-fired Boilers

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ABSTRACT

Future use of coal for power generation in the US depends on technologies being made available to capture and store CO₂ emissions from power plants. A key candidate CO₂ capture technology is oxy-firing of coal. Application of oxy-firing to existing power plants presents unquantified challenges as the characteristics of oxy-firing compared to air-firing have not been fully determined.

This presentation will review the status of an on-going DOE-funded program to characterize and predict impacts of CO₂ flue gas recycle and burner feed design on flame characteristics (burnout, emissions and heat transfer), fouling, slagging, and corrosion, inherent in the retrofit of existing coal-fired boilers for oxy-coal combustion. Key deliverables from the project include:

- Multi-scale test data from 0.1 kW bench-scale, 100 kW laboratory-scale and 1.2 MW semi-industrial scale combustors that describe differences in flame characteristics, fouling, slagging and corrosion for coal combustion under air-firing and oxygen-firing conditions, including sensitivity to oxy-burner design and flue gas recycle composition.
- Validated mechanisms developed from multi-scale test data that describe fouling, slagging, waterwall corrosion, heat transfer, char burnout and sooting under coal oxy-combustion conditions.
- Principles to guide design of pilot-scale and full-scale coal oxy-firing systems and flue gas recycle configurations, such that boiler operational impacts from oxy-combustion retrofits are minimized.
- Assessment of oxy-combustion retrofit impacts in a full-scale coal-fired utility boiler using computational fluid dynamics (CFD) modeling of air-fired and oxygen-fired operation, together with the information generated in this research.

The experimental data, oxy-firing system principles and oxy-combustion process mechanisms provided by this work can be used by electric utilities, boiler OEMs, equipment suppliers, design firms, software vendors, consultants and government agencies to assess retrofit applications of oxy-combustion technologies to existing boilers or to guide development of new designs.
INTRODUCTION

The primary objective of this program is to develop tools to characterize and predict impacts of CO$_2$ flue gas recycle and burner feed design on flame characteristics (burnout, NO$_x$, SO$_x$, and fine particle emissions, heat transfer), fouling, slagging and corrosion, inherent in the retrofit of existing coal-fired boilers for oxy-coal combustion. This objective will be met by producing multi-scale experimental data focused on burner design, char oxidation, soot evolution, ash characterization and deposition and corrosion. Mechanisms capable of describing these phenomena under air and oxy-fired conditions will be developed using the data generated during experimentation and validated against all available data. The mechanisms will be implemented into CFD code and an existing coal-fired utility boiler will be modeled under air and oxy-fired conditions to identify the likely impacts of retrofit. Figure 1 provides an overview of the structure of this approach.

**Figure 1. Program Structure Overview**

Reaction Engineering International is the prime researcher on this Project and will be performing much of the mechanism development work and all of the CFD modeling. Subcontracts have been awarded to: University of Utah (who will perform lab and pilot-scale experimentation and some work on mechanism development), Siemens Energy (who will provide an oxy-coal research burner, and a full system firing system design for modeling), Sandia National Labs (who will perform bench-scale experimentation), Corrosion Management (who will provide corrosion monitoring hardware and will co-develop corrosion mechanisms), Praxair (who will provide gases for experimentation), Brigham Young University (who will perform soot measurements) and Vattenfall AB (who will be providing information concerning their current mechanisms for oxy-coal modeling). There is an advisory committee in place to help direct the focus of the research, comment on firing system design and to provide model inputs for a Retrofit Assessment. The members of the advisory committee consist of representatives from PacifiCorp, Southern Company, Vattenfall AB and Praxair.

PROGRAM METHODOLOGY

Multi-Scale Test Data

*Bench-Scale Optical Entrained Flow Reactor Experiments*

Bench scale experimentation will take place at Sandia National labs to further elucidate the behavior of char in an oxygen enriched flue gas recycle gas matrix. These experiments will be conducted in Sandia’s optical entrained flow reactor and associated particle-sizing pyrometry diagnostic detailed in Figure 2.
A flat-flame burner produces a high temperature (1000-2100K) and high velocity gas matrix with oxygen concentrations ranging from 0-60 vol-% in either a N₂ or CO₂ diluent. Moisture levels are tailored through judicious selection of fuel gases. Pulverized coal particles are fed at a rate of 0.5-1.0 g/hr and are heated to more than 1200 K in less than 10 ms residence time resulting in complete devolitalization within 15-20 ms.

A helium-quenched, water-cooled sampling probe with a micropore filter is used to sample partially reacted particles at well-defined residence times in the reactor for characterization of char burnout, char and ash chemistry, and char pore properties via ultimate/proximate/ash analysis, Hg porosimetry, BET, SEM, etc. Quartz plate walls contain the flow reactor gases and permit optical measurements to be performed on the entrained particles as they burn. The coded aperture in the optical diagnostic produces time traces of thermal emission from the burning particles with two peaks whose ratio is a function of char particle size. Measurement of the emission intensity through two spectral filters onto photomultiplier tubes allows the temperature of the particle to be determined, through calibrated two-color pyrometry. The time-of-flight of the particle across the coded aperture slits gives the particle velocity. Together, this information is used to solve for the instantaneous, surface-specific burning rate.

**100 kW Oxy-Fuel Combustor (OFC) Experiments**

Data related to ash characterization and deposition will be collected using experiments conducted in the University of Utah’s 100 kW OxyFuel Combustor (OFC), shown in Figure 3.
The furnace consists of an oxy-fuel combustion chamber and radiant zone in the vertical section, followed by a horizontal convective section where temperature profile is prescribed through adjustment of independently controlled cooling coils to simulate practical furnace temperature profiles. Pure CO₂ from a tank or FGR may be mixed at the burner to control adiabatic flame temperature. Optional flue gas equipment on the back end of this furnace includes a fabric filter, scrubber and condenser. Given this configuration, it is possible to operate the furnace to simulate recycling flue gas from different locations on the back end of the furnace and determine that effect on ash characterization. Data collected will include size segregated speciation of ash through Low Pressure Impactor (LPI) and Scanning Mobility Particle Sizer (SMPS) sampling and analysis. Deposition rate measurements and deposit characterization will also be performed. These experiments will be performed over the course of the three year program.

1.2 MW Pilot-Scale Furnace (L1500) Experiments

Pilot-scale experiments will be performed in the University of Utah’s 1.2 MW pulverized coal furnace (L1500). These experiments will be designed to investigate firing system configuration impact on flame stability and heat transfer, soot evolution and water wall and superheat material corrosion. A diagram of the L1500 is included as Figure 4.

The radiant section of the combustion chamber is 1.1 m x 1.1 m square and nearly 12.5 meters long. The walls are refractory lined with cooling tubes on the walls of the first four sections. This configuration allows for stable thermal and realistic radiation characteristics. The turbulent length scales of this furnace are appropriate for representing full-scale burner mixing and stability. The access ports are used for visual observations, fuel and/or air injection, and product sampling. This facility has been retrofit for oxy-combustion with flue gas recycle and includes a baghouse and a partial condenser on the recycle stream.
Four weeks of parametric testing will be performed in the L-1500 to develop practical guidelines that allow optimized operation of an actual burner. The test plan will be developed to meet these goals. Burner operating parameters of interest include:

1. Variable oxygen, FGR and coal distribution in the burner.
2. Variable flue gas recirculation ratio (molar ratio of FGR to oxygen). We will start with 3.2 (providing about 26% \( \text{O}_2 \) in the FGR/\( \text{O}_2 \) mixture) and reduce the FGR until near burner temperatures draw near to equipment limits
3. Variable burner stoichiometric ratio within the range of 0.7 to 1.2

REI and Corrosion Management will design and construct four probes for electrochemical noise measurement of corrosion rate. One of these four probes will be designed to measure corrosion under conditions (temperature and physical position with respect to the flue gas) expected for the waterwall in the lower furnace and the corrosion plates will be fabricated from low-carbon steel typical for this application. The remaining three probes will be fabricated to represent a superheater tube in cross flow. These probes will be constructed of high nickel alloys typical for superheaters (likely T22, T90, SS347). The deposition rate on the probe will also be measured by collecting and analyzing these deposits. Heat flux meters will be integrated into the corrosion or deposition probes to measure heat flux at the corrosion locations.

Six weeks of testing will be performed in the L1500 to characterize corrosion under both air and oxy-combustion conditions. Results from previous experiments will be used to identify regions in the furnace for installation of the corrosion probes where deposition, heat flux and flue gas compositions will be favorable for corrosion and relevant for full-scale utility boilers. The parameters of interest in the corrosion test plan will be:

1. Air and oxy-fired conditions
2. PRB and Bituminous Coals
3. Optimized oxygen, FGR and coal distribution in the burner, from previous experiments
4. Variable flue gas recirculation ratio, within the limits from previous experiments
5. Variable burner stoichiometric ratio, within the limits from previous experiments

Measurements that will be performed in the pilot-scale furnace include:
1. Flue gas composition (O₂, CO₂, CO, NOₓ and SO₂) using existing CEMs
2. Unburned carbon in ash, using loss on ignition analysis
3. Flame attachment, using ultraviolet sensors and cameras
4. Real-time corrosion measurements
5. Deposition rate and composition at the corrosion locations
6. Heat flux at the corrosion locations and other locations in the furnace
7. Local flue gas temperatures, using suction pyrometry
8. Soot volume fraction using the two-color extinction method

Firing Principles

Siemens Energy will provide for this project a versatile, oxy-coal research burner for the L1500. This burner will be based on an existing commercial burner. However the purpose here is not necessarily to optimize this particular burner for oxy-coal retrofit, but rather to develop general firing system principles relevant to most wall-fired systems.

The Siemens test burner to be used embodies the following design concept principles: It incorporates specifically designed components and flow pathways to create a wide range of mixing effects within the flame. Secondary and primary gas flows can be varied and mixed in three zones; inner secondary, outer secondary and primary streams. This level of flexibility provides the ability to vary the amount of CO₂ and oxygen to the different streams of the combustion process; thereby maximizing the burner’s flexibility for parametric testing. The effects and combustion patterns of variable mixing patterns in conventional air oxidized configuration have already been modeled with CFD software, with predictions that this new approach provides significantly improved combustion.

Burner performance will be assessed through a series of parametric tests. These tests will systematically map the performance of the burner. The tests will utilize the flexibility of the burner by varying the CO₂/O₂ ratio in each of the flow paths of the burner. A test matrix will be designed in such a way that performance maps will be developed from the test data. The optimum operating parameters will be determined to minimize flue gas recirculation and maximize the burner efficiency and mimic heat transfer characteristics in an air-fired system. This optimum operating condition will be determined from the burner performance maps created from the reduction of the test data.

Mechanism Development

Deposition

Changes in flue gas composition due to oxy-firing will cause differences in ash composition and particle size distribution and in turn impact particle deposition. The ability to understand and predict these changes will allow us to evaluate their impact on a boiler being retrofit for oxy-combustion.

The large increase in SO₂ and O₂ concentrations will result in increased SO₃ and acceleration of sulfation of the ash (verified by Okazaki and co-workers, 2003, 2008), and thereby change its fouling propensity (Figure 5). Increased capture of sulfur by ash under oxy-combustion conditions has been described by Okazaki and co-workers [Liu and Okazaki, 2003; Liu et al., 2001]. The ash loading in the gas phase under oxy-coal combustion conditions also shows a (3 to 5 fold) increase and even though larger
particles may have to be removed, the ultrafine (or sub-micron) fraction of the ash will remain to be available for making a surface sticky. Increasing the partial pressure of oxygen, however, might be advantageous for slagging by diminishing Fe₂O₃ versus FeO, the latter being a pro-slagging constituent. Indeed, oxy-coal might allow operation at lower stoichiometric ratios (SR) than under air without FeO formation (slagging) penalties. For slagging, inertial impaction of ash particles and ash viscosity control deposit initiation and growth on waterwall surfaces. Ash viscosity depends on composition (which can change under oxy-coal) and temperature. Although the bare waterwall tube surfaces are generally too cold (400 to 750°F or 200 to 400°C) to melt any solid ash particles, those that approach the tube surface are much hotter than the surface because of the flame environment, and some will have a sufficiently low viscosity such that they will stick to the tube surface. Once a deposit has started, the ash particles provide an insulating layer on the surface; thus the surface temperature will increase and the viscosity of the surface layer, decrease. Therefore, it will be more likely that ash particles impacting the surface will stick.

Traditional slagging indices, such as the base-to-acid ratio, are based on the bulk composition of the coal ash and do not take into account the specifics of the combustion system (in terms of the radiant environment and the fluid mechanics). Models that combine both advanced analyses of the coal minerals with boiler operating conditions have been developed and tested on full-scale power plants [Senior and Johnson, 1994; Helble et al., 1992]. Recently, advanced coal analyses have been combined with CFD models of to predict slagging as well as fouling in full-scale boilers [Ma et al., 2007].

Changes in three oxy-coal aspects of a firing system will have an impact on slagging (for a given coal composition):

- Particle composition and size distribution changes
- Changes in the radiant environment of the furnace caused by different heat-release profiles.
- Local changes in flue gas oxygen content, because ash viscosity can be sensitive to oxygen partial pressure, particularly for high-iron coals. The local partial pressure under oxy-coal combustion is likely to be higher than under air combustion, and this should lead to greater ash viscosities for high iron coals (which works against slagging).

Fouling refers to the formation of deposits in the convective section of the furnace not exposed directly to radiation. In high temperature fouling molten silicate ash particles can impact tube surfaces and bind ash particles together (see Figure 5). In low-temperature fouling sodium (and other alkali) sulfates condense on tube surfaces and form a sticky layer that can bind together ash particles. The amounts of sodium and sulfur in the gas phase affect the dew point of sodium sulfate, which can range from 1350°F to 1850°F. The initial deposit layer on convective pass tubes is high in sulfates of Na, Ca, and K. Mixtures of different sulfates can form eutectics that melt at lower temperatures <1600°F. Work by EERC on the role of calcium in fouling demonstrated the formation of hard, sulfated calcium deposits. Increases in the concentration of SO₂ in the flue gas were shown to correlate with an increase in compressive strength of deposits. Recycle of flue gas in an oxy-fired boiler will greatly increase the SOx concentration in the flue gas and the amount of submicron alkali-containing ash particles. Such fine alkali particles will vaporize at high temperatures in the flame. Both the sodium and sulfur contents of the flue gas are expected be higher in the convective
pass in an oxy-fuel system with flue gas recycle. This will increase the amount of fouling in the convective pass.

![Figure 5. Overview of Deposition Principles in a Pulverized Coal Combustor](image)

The mechanism describing deposition (slagging or fouling) will accept from the CFD code data describing flame-side conditions (gas composition, temperature and the rate that particles impact the surface). The deposition mechanism will then calculate the deposit growth, sintering and deposit properties and update the wall boundary conditions in the overall CFD simulation.

**Corrosion**

Fireside tube corrosion can occur in multiple locations within a coal-fired boiler and the use of oxygen and flue gas recycle has the potential to affect corrosion behavior in a number of different ways. Mechanisms are often specific to waterwalls, superheaters, and economizer regions and are affected by fuel properties, boiler design, and operating conditions. Waterwall corrosion mechanisms that have been identified include oxidation, chlorine-related reaction, gas-phase sulfur interactions, and the deposition of reduced sulfur forms. In the superheat/reheat area oxidation, active oxidation, molten chloride, molten sulfate, carburization, and sulfidation mechanisms have all been indicated as mechanisms for tube attack. Specific possibilities for corrosion effects of oxy-firing can be identified by examining specific correlations that have been developed to quantify corrosion rates. Table 1 shows examples of several of these correlations [Hong et al., 2006].

A simplistic consideration of oxy-coal combustion and the parameters illustrated here indicate a number of possible impacts on corrosion tendencies. The removal of nitrogen diluent and the potential concentration of minor species during flue gas recycle provide a potential means for increasing corrosion rates in a boiler. Again, one or more of the correlations above show a clear increase in corrosion as the concentration of corrosive sulfur or chlorine containing gas-phase constituents is increased. In addition to the thermal and concentration issues noted, deposit chemistry/stability is known to be negatively impacted by the existence of alternating oxidizing and reducing conditions. The potential existence of larger extremes in oxygen concentration during oxy-firing could further magnify any problems that occur.
### Table 1. Correlations for Corrosion Rates

<table>
<thead>
<tr>
<th>Waterwall Corrosion</th>
<th>Correlation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas phase H$_2$S Corrosion</td>
<td>$CR_{H2S} = a \cdot f_1(T_m) \cdot f_2(Cr%) \cdot f_3(H2S) + b$</td>
</tr>
<tr>
<td>Deposition of Unoxidized Material</td>
<td>$CR_{dep} = a \cdot f_1(dep) \cdot f_2(Stoichiometry) \cdot f_3(T_m) + b$</td>
</tr>
<tr>
<td>Chlorine-based Corrosion</td>
<td>$CR_{Cl} = a \cdot f_1(%Cl) \cdot f_2(Heat Flux) \cdot f_3(T_m) + b$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Superheat Corrosion</th>
<th>Correlation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Active Oxidation</td>
<td>$CR_{AO} = f_1(T_m) \cdot f_2(Cr%) \cdot f_3(T_g - T_m) \cdot f_4(Cl%)$</td>
</tr>
<tr>
<td>Molten Sulfate Corrosion</td>
<td>$CR_{MS} = a \cdot P \cdot HF \cdot f_1(T_m) \cdot f_2(Cr%) \cdot f_3(Fuel Corrosivity) \cdot f_4(dep) + b$</td>
</tr>
<tr>
<td>Sulfidation</td>
<td>$CR_{H2S} = a \cdot f_1(T_m) \cdot f_2(Cr%) \cdot f_3(H2S) + b$</td>
</tr>
</tbody>
</table>

REI has developed and implemented mechanisms for the prediction of corrosion in CFD codes for application to coal-fired power boiler. These correlations and their implementation have been refined repeatedly by comparison with field and pilot-scale testing [EPRI, 2001; Davis et al., 2004; Valentine et al., 2007]. Although this computational approach has proven very useful for conventional boilers, oxy-firing has the potential to create unusual conditions to which particular attention should be given. One such area involves heat flux. Local heat flux has been identified as perhaps the most important boiler design variable for quantifying chlorine-related corrosion [Hanson and Abbott, 2006]. In addition, the current understanding of the role of heat flux has been identified as an area needing further investigation for sulfur-related corrosion as well. As part of the proposed effort, the understanding of the role of heat flux will be further examined and its quantification within our corrosion mechanisms will be refined.

### Char Oxidation

Previous measurements of char oxidation rate in the Sandia facility have demonstrated that the high concentration of carbon dioxide (CO$_2$) under oxy-combustion conditions tends to reduce the char combustion rate, on account of the lower diffusivity of oxygen through the particle boundary layer when CO$_2$ is present. On the other hand, elevated concentrations of O$_2$, as are typically employed during oxy-combustion, tend to accelerate char combustion and can easily compensate for the inhibitory effect of the elevated CO$_2$ levels. Detailed modeling of single char particle combustion using the Surface Kinetics in Porous Particles (SKIPPY) computational program suggests that the single-film combustion model (i.e. unreactive boundary layer) currently employed for CFD computations of pc combustion fails for moderate size char particles (order of 100 μm) under some oxy-combustion conditions with elevated concentrations of O$_2$ (20% or higher, in the vicinity of the char particle). This finding has important ramifications regarding pc combustion modeling of char burnout during oxy-combustion.
The results from the char combustion experiments in this program will be interpreted with the aid of SKIPPY. A particular focus will be on determining the combustion conditions in which the single-film combustion model fails and on effectively capturing this combustion behavior in a computationally tractable char combustion model.

**Soot Formation**

Effects of Oxy-coal combustion on soot are also important. The change in oxygen and CO\textsubscript{2} present in oxy-combustion can influence the temperature history of the particles and thus change the amount of soot formed. The contribution of soot to flame emissivity relative to H\textsubscript{2}O and CO\textsubscript{2} effects needs to be characterized for oxy-fired coal flames. In a coal flame the soot is formed by the cracking of the tars evolved with the volatiles with an efficiency that is governed by the oxidation/temperature history. This differs from the more complex soot mechanisms for gaseous flames.

Theoretically if the oxygen/temperature history of a particle remains constant between air firing and oxy-firing, soot formation would also remain constant. However, in order to match the oxygen/temperature history, the H\textsubscript{2}O and CO\textsubscript{2} concentrations would need to change, thus changing the relative contributions of soot versus H\textsubscript{2}O and CO\textsubscript{2}. The spectral model RADCAL will be used to evaluate the relative contributions of soot and H\textsubscript{2}O and CO\textsubscript{2} to flame emissivity. REI’s existing soot model will be adjusted as needed to account for effects of H\textsubscript{2}O and CO\textsubscript{2} based on soot volume fraction data taken in the OFC. The overall radiative heat transfer model will be validated by comparison with heat flux data taken in the L1500 furnace.

**Full-Scale Firing System Development**

Siemens Energy and REI will use the developed firing system principles and operating conditions from the burner parametric testing in the L1500, determined to provide optimum firing system performance, to assess design and scale-up properties for a full-scale application. This full-scale firing system will include burner size and flow characteristics at the burner face (including fuel, O\textsubscript{2} and FGR mixtures) along with the specifics of additional oxygen and FGR that may be introduced downstream of the burner zone or “over-fire”. The remainder of the parametric test data will be analyzed to assess the flexibility and operating range of the firing system design. The system needs of a boiler retrofit for oxy-combustion and its auxiliaries will be evaluated. Upon completion of burner performance optimization, determination of the final requirements for a full-scale firing system will be made by Siemens. A conceptual design of full-size utility burner will be prepared which will address overall design concepts, parameters, sizing and requirements for application to a utility furnace.

**Retrofit Assessment**

With participation of utility Advisory Panel members, a suitable existing coal fired boiler will be selected to be simulated under both air and oxy-coal firing. The criteria for unit selection will be based on a number of items but will include at a minimum: 1) general representation of the coal fired generating capacity in the US, 2) access to geometrical and operating data for generation of the model, and 3) feasibility as a candidate for oxy-coal conversion. It is possible that the unit will be selected from REI’s large database of existing models. The unit will first be simulated and verified under air-coal operation,
followed by simulation under oxy-coal operation. The model will incorporate the design and operational modifications specified by the team as part of the burner scale-up task. Predicted differences in flame characteristics, heat transfer, emissions, fouling and slagging behavior, and waterwall corrosion will be compared for air-coal and oxy-coal operation.

Based on the simulation results for the first oxy-coal design, design and operational changes will be made to optimize firing system performance. Changes will include but will not be limited to burner and overfire air changes, and quantity and design of the FGR. The intent is to closely approach the unit heat balance that is achieved during air-coal firing, and to improve upon slagging and fouling and waterwall corrosion rates as compared to that seen and predicted under air-coal conditions. Insight gained in the simulations will be used in subsequent simulations to optimize the performance.

SUMMARY

This project has been tailored to both identify potential impacts of the oxy-combustion retrofit of existing coal-fired utility boilers (through multi-scale experiments) and to develop tools that will allow accurate prediction of these impacts (through mechanism development). Experiments will be performed on three different scales: (1) A bench-scale optical entrained flow reactor will be used to elucidate the impact of oxy-combustion flue gas composition on the rate of char oxidation; (2) a 100-kW lab-scale combustor will be used to characterize the effects of flue gas recycle on ash characteristics; and (3) a 1.2 MW pilot-scale combustor will be used to investigate burner and firing system principles, deposition, corrosion and radiative heat transfer, including soot evolution. The data from these experiments will be used to guide development of mechanisms that may be used to describe char oxidation, deposition (slagging and fouling), corrosion and soot evolution. The data generated will also be used to produce an overview of firing system principles for oxy-combustion that may help guide design of full-scale firing systems.

Based on incorporation of validated mechanisms in a CFD code and scale-up of oxy-firing principles, a retrofit assessment of a full-scale boiler will be conducted to identify potential operational improvements and challenges for oxy-combustion firing.

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