TITLE: **A MECHANISTIC INVESTIGATION OF NITROGEN EVOLUTION AND CORROSION WITH OXYCOMBUSTION**

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1. ABSTRACT

Objectives

In pilot-scale testing, pulverized coal oxy-fuel combustion has shown substantially reduced NO_X emissions relative to air combustion. The mechanisms behind this reduction are not yet understood. In addition, altered temperature profiles and minor species concentrations may affect the corrosion tendencies of a combustion system switched from air to oxy-fuel combustion. The objectives of this research are:

- Determine the mechanisms by which the NO_X reduction is obtained in oxy-fuel combustion.
- Measure deposited species on simulated superheater tubes which indicate the corrosion tendencies of oxy-fuel combustion.

Accomplishments to date

Pulverized subbituminous and bituminous coals have been burned in an unheated, down-fired, laminar flow combustor while NO_X, CO and O_2 were measured as a function of distance from the burner. For the oxy-fuel cases recycled flue gas was simulated with bottle CO_2. Both lean premixed and oxidizer-staged premixed configurations have been used leading to the conclusion that the NO_X reduction potential of oxy-fuel combustion is associated with oxidizer staging. Under oxidizer-staged conditions oxy-fuel exhibits higher CO concentrations in the fuel-rich zone relative to comparable air cases. In these fuel-rich regions NO_X is reduced to lower levels (on a lb/MMBtu basis) in the oxy-fuel cases. This observation is independent of destruction of recycled NO_X. For the bituminous coals, initial nitrogen release and NO_X formation from the coal volatiles is comparable between air and oxy-fuel in these and flat flame burner experiments pointing to more effective NO_X destruction in the oxy-fuel cases. For the subbituminous coal

tested, the amount of NO_X formed was lower and the amount of NO_X reduced was comparable in oxy-fuel combustion, with an end result of lower effluent NO_X.

One experiment was performed where the incoming CO_2 was doped with NO to determine the effects of NO recycling. Consistent with findings reported in the literature it was observed that initial NO_X formation was somewhat suppressed by the already high NO_X concentrations. Faster NO_X destruction rates were also observed, consistent with the rate of NO_X destruction a function of the NO_X concentration.

A computational model was programmed using the CPD-NLG coal devolatilization model, a char oxidation and gasification model, and the GRI-Mech 3.0 mechanism with advanced-reburning reactions added. Much insight has been gained from this model. Many features of the experimental data are predicted by the model including the trends observed in the NO recycling experiment. It appears from the kinetic model predictions that the high CO concentrations in the oxy-fuel reducing zones are a key factor in the NO_X reduction potential of oxy-fuel combustion. Thermal dissociation of the CO_2 was determined to be the source of this CO. The model suggests gasification of the char by the higher levels of CO_2 in oxy-fuel combustion is insignificant and does not contribute to the high CO levels. The low equilibrium levels of NO_X in oxy-fuel seem to play a role in increasing the range of stoichiometry under which NO_X can be reduced relative to an air-coal flame.

Some simulated superheater deposits have been collected and analyzed but there is as yet no indication from these samples that oxy-fuel combustion increases the corrosion potential of the deposit. Mineral ash analysis of subbituminous ash from air and oxy-fuel combustion did show some differences, notably increased sulfur and calcium content in the oxy-fuel ash.

Future work

The work planned for the remaining months of the award period is as follows:

- Measure NH_3 and HCN concentrations under the oxidizer-staged conditions already studied
- Further refine the computational model
- Collect and analyze additional simulated superheater fly-ash deposit samples
- Publish the outcomes of the investigation

2. LIST OF CONFERENCE PRESENTATIONS AND STUDENTS RECEIVING SUPPORT FROM THE GRANT

Conference presentations

- An Investigation of Nitrogen Evolution in Oxy-fuel Combustion, A. J. Mackrory, S. Lokare, L. L. Baxter, D. R. Tree, 32nd International Technical Conference on Coal Utilization & Fuel Systems, June 10 – 15, 2007, Clearwater, FL
- Measurements and Modeling of Nitrogen Evolution in Staged Oxy-fuel Combustion, A. J. Mackrory and D. R. Tree, AIChE Annual Meeting, November 4 – 9, 2007, Salt Lake City, UT
- Investigation of NO_X Destruction Catalyzed by Char Oxidation in Oxy-fuel Combustion D. R. Tree, A. J. Mackrory, Spring 2008 ACS National Meeting & Exposition, April 6 – 10, 2008, New Orleans, LA
- NO_X Destruction Experiments and Modeling in Oxy-fuel Combustion, A. J. Mackrory, D. R. Tree, 33rd International Technical Conference on Coal Utilization and Fuel Systems, June 1 – 5, 2008, Clearwater, FL

Students supported under this grant

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