# LAB-SCALE STUDIES OF OXYFUEL COMBUSTION

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Grant Number: DE-FG26-05NT4253
Performance Period: 8/1/05-2/28/07

## OBJECTIVES

As part of the Global Climate Change Initiative and the need to manage green house gases, there is strong interest in developing a portfolio of technologies for minimizing emissions of carbon dioxide from coal-fired systems. The low concentration of carbon dioxide in the flue gas of conventional coal-fired plants makes it difficult to develop cost effective means of capturing and sequestering carbon dioxide. An operational scheme based on flue-gas recycling and pure oxygen can result in greater than 95% carbon dioxide in the coal combustor exhaust. This will improve carbon dioxide recovery and also potentially minimize NOx, SOx, and trace species emissions. Past studies have focused on replacing the nitrogen in the air with $CO_2$ from the exhaust gas. While this approach is effective at increasing the $CO_2$ concentration in the flue gas, the potential for even greater improvements in the overall cycle is possible. This can be appreciated by recognizing that since there are actually three streams (oxygen, $CO_2$, and coal), there are an unlimited number of ways these three streams can be mixed, and the resulting products can vary dramatically.

The goal of this program is determining how to best implement oxy-fuel combustion with flue gas recirculation and sorbent processes to minimize emissions and maximize combustion efficiency and the concentration of $CO_2$ in the flue gas. The optimum approach to mixing of the coal, oxygen and $CO_2$ and sorbents will be evaluated and the effect of this mixing and the enriched carbon dioxide and SOx environment on emission of NOx, SOx, PM 2.5 and mercury will be evaluated as well. The effect of the stream temperature will also be evaluated to identify the optimum combination of mixing, composition and preheating to minimize emissions.

Initial experiments were performed in a drop tube furnace to understand the role of $CO_2$ without considering mixing. A laboratory scale coal-fired burner is under construction to test mixing concepts for a wide range of feed conditions and temperatures.

## ACCOMPLISHMENTS TO DATE

Primary conclusions of results to date are summarized below. Detailed information can be obtained from the references cited.

1) Replacing nitrogen with carbon dioxide can lead to a smaller size distribution of submicron particles and lower number concentration. The large specific heat of carbon dioxide lowers flame temperature and this slows the vaporization rate, leading to a delay in the onset of nucleation and less particles. This also implies less time for coagulation and condensation, which results in a smaller size. When the oxygen concentration was increased to yield a similar flame temperature to that of air-fired combustion, the size distribution was similar to that for air-fired combustion.

2) A study of mercury emission was conducted for 20% $O_2$/80% $CO_2$ and 25% $O_2$/75% $CO_2$. The results revealed that the total gaseous mercury concentration was similar to that of air fired combustion, as were the ratio of oxidized to elementary mercury. The results suggest that carbon dioxide does not have a strong impact on mercury speciation.

3) Mercury was found to exist only in the elemental form when the carrier gas is either air or a mixture of $O_2$-$CO_2$. However, in the presence of chlorine species, oxidized mercury was found

in higher concentrations in the $O_2$-$CO_2$ system than in the conventional (air) system. Water vapor did not affect mercury speciation in either $O_2$-$CO_2$ or conventional systems. However, it did play an important role on oxidation of mercury when chlorine species were present.

4) Submicrometer and ultrafine particles produced by coal combustion were found to have a bipolar distribution at the outlet of the combustor, which is slightly skewed towards positive charge. This appears to be relatively independent of combustion gas composition. In addition, electrostatic precipitators required higher energy inputs for $O_2$-$CO_2$ systems to produce the same number concentration of ions as $O_2$-$N_2$ systems. Furthermore, the penetration of submicrometer and ultrafine particles was an order of magnitude higher in $O_2$-$CO_2$ systems as compared to $O_2$-$N_2$ systems. These findings should be taken into consideration during design of electrostatic precipitator systems used with $O_2$-$CO_2$ coal combustion.

5) A model of soot inception limits under oxy-fuel combustion conditions was developed and validated with gaseous fuels. These results indicate that the primary parameters controlling soot inception are local temperature, local C/O ratio and residence time.

PAPERS PUBLISHED, CONFERENCE PRESENTATIONS, STUDENTS SUPPORTED

Papers and Presentations:


1. Suriyawong, A., Gamble, M., Lee, M.-H., Axelbaum, R.L. and Biswas, P., "Submicron Particle Formation and Mercury Speciation under Oxygen-Carbon Dioxide Coal Combustion," Twenty-Second Annual International Pittsburgh Coal Conference, Pittsburgh, PA, Sept. 12-15, 2005.
2. Kumfer, B.M., Biswas, P. and Axelbaum, R.L., "Oxy-Combustion: Novel Strategies for Improving Combustion and Multi-Pollutant Control," Twenty-Second Annual International Pittsburgh Coal Conference, Pittsburgh, PA, Sept. 12-15, 2005.
3. Suriyawong, A., Smallwood, M., Noel, J.D., Lee, M.H., Giammar, D.E., and Biswas, P. A Strategic Approach for Optimizing the Use of Sorbents for Mercury Removal from Coal-burning Utilities. The Air and Waste Management Association's 98th Annual Conference and Exhibition, Minneapolis, MN, 2005.
4. Suriyawong, A., Lee, M.H., and Biswas P. Submicrometer Particle Formation and Mercury Emission under Enriched Oxygen Coal Combustion. Student Paper and Poster Competition, the Air and Waste Management Association's 98th Annual Conference and Exhibition, Minneapolis, MN, 2005.
5. Biswas, P., Suriyawong, A., Smallwood, M., Noel, J.D., Lee, M.H., Giammar, D.E., and Biswas, P. Nanostructured-sorbents for Heavy Metals Emissions Control-A Review. American Chemical Society (ACS) 2005 Annual Conference, San Diego, CA, 2005.
6. Giammar, D.E., Noel, J.D., Smallwood, M., Suriyawong, A., Lee, M.H., and Biswas, P. Nanostructured Sorbents for Control of Mercury Emissions from Coal Combustion. The Association of Environmental Engineering and Science Professors (AEESP), Syracuse, NY, 2005.
7. Suriyawong, A.; Gamble, M. D.; Lee, M.; Axelbaum, R. L.; Biswas, P., Submicrometer Particle Formation and Mercury Speciation under O2-CO2 Coal Combustion. *Energy & Fuels* 20, 2357-2363 (2006).
8. Suriyawong, A.; Hogan, C. J. J.; Biswas, P., Charged Fraction and Electrostatic Capture of Ultrafine and Submicrometer Particles Formed under O2-CO2 Coal Combustion. *Fuels*, Submitted.
9. Skeen, S.A., Kumfer, B.M., and Axelbaum, R.L., An Experimental and Theoretical Approach to Soot Particle Inception in Laminar Diffusion Flames, 7th International Aerosol Conference, September 10-15, 2006, St. Paul, MN.
10. Axelbaum, R.L. (Invited), Oxy-Fuel Combustion: New opportunities for non-premixed combustion brought about by global warming, 22nd Annual Symposium of the Israeli Section of the Combustion Institute. Tel Aviv, Israel December 21, 2006.


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