

APPENDIX B: CARBON DIOXIDE CAPTURE TECHNOLOGY SHEETS

# OXY-COMBUSTION



# OXYGEN TRANSPORT MEMBRANES FOR INDUSTRIAL APPLICATIONS

B-425

OXY-COMBUSTION

## primary project goals

Praxair is optimizing oxygen transport membrane (OTM) performance, materials, and process configurations leading to subsequent development-scale testing of OTM technology for synthesis gas (syngas) production applications, providing valuable experience needed to develop commercial OTM technology in industrial applications and future utility-scale power generation applications.

## technical goals

### Phase I:

- Develop more detailed OTM cost and performance estimates based on experiments.
- Develop a preliminary conceptual design and cost models for a pilot plant utilizing OTM technology.
- Identify the rate-limiting steps for oxygen (O<sub>2</sub>) separation through the OTM and address kinetic or mass transport limitations by appropriate materials selection and membrane architecture.
- Develop procedures to manufacture one-third pilot-size OTM tubes; test them for O<sub>2</sub> flux and durability in carbon monoxide (CO), hydrogen (H<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), and water (H<sub>2</sub>O) fuel streams with the presence of sulfur impurities.
- Test OTMs in a coal gas OTM reactor.

### Phase II:

- Demonstrate the ability to produce OTM tubes with the appropriate dimensions and manufacturing yield required to proceed with pilot demonstration.
- Deliver preliminary engineering cost estimate for OTM pilot plant system (OTM partial oxide reactor [POx] and Boiler).

### Phase III:

- Develop technology to support the conversion of natural gas to syngas utilizing oxygen from a ceramic membrane in a development-scale syngas system.
- Develop technology to support the oxy-combustion of a syngas fuel utilizing oxygen from a ceramic membrane with a portion of the heat of combustion utilized to raise saturated steam and with the primary flue gas products being comprised of H<sub>2</sub>O and CO<sub>2</sub>.
- Further develop ceramic materials and processes to support high-volume, low-cost manufacturing and stable performance over an anticipated commercial operation lifetime.

technology maturity:

Laboratory Scale

project focus:

Oxygen Transport Membrane-Based Syngas Production

participant:

Praxair

project number:

FC26-07NT43088

NETL project manager:

Jose Figueroa

jose.figueroa@netl.doe.gov

principal investigator:

Sean M. Kelly P.E.  
Praxair, Inc.

sean\_kelly@praxair.com

partners:

ENrG Inc.  
The Shaw Group  
The University of Utah

performance period:

4/30/07 – 9/30/15

B-426

As oxy-combustion is currently practiced, a pure stream of O<sub>2</sub> is separated in a cryogenic air separation unit (ASU) and then delivered to a boiler for combustion. OTM technology integrates O<sub>2</sub> separation and combustion in one unit. An OTM consists of an inert porous support coated with a dense gas separation layer, as illustrated in Figure 1. Air contacts the separation layer where molecular O<sub>2</sub> reacts with O<sub>2</sub> vacancies and electrons on the membrane surface to form O<sub>2</sub> ions, which are transported through O<sub>2</sub> vacancies in the separation layer using a chemical potential difference as the driving force. Fuel species (CO, H<sub>2</sub>, methane [CH<sub>4</sub>], etc.) located on the porous support side diffuse through the support and react with O<sub>2</sub> ions at the membrane surface to form oxidation products (H<sub>2</sub>O, CO<sub>2</sub>) and release electrons, which are transported back through the separation layer.

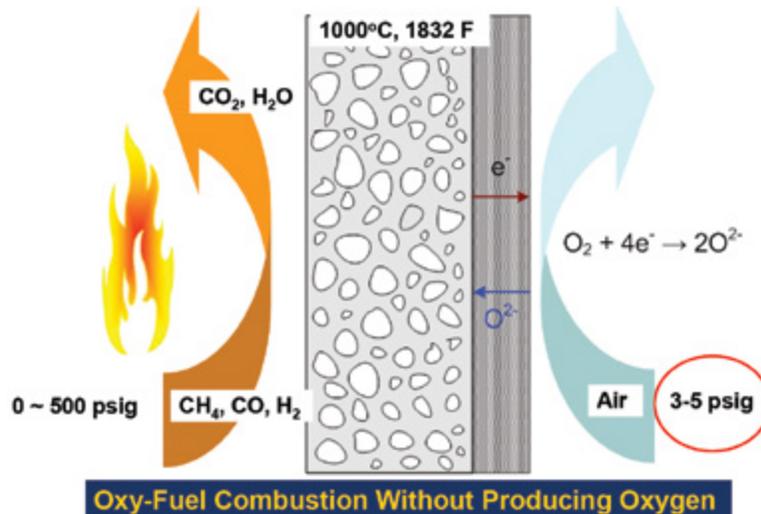


Figure 1: Schematic of Ceramic OTM

Several process concepts incorporating ceramic OTM are being explored to understand their impact on process economics. One process concept under development is shown in Figure 2. In this process, coal is first gasified in an O<sub>2</sub>-blown gasifier to generate syngas. The syngas is optionally reacted in an OTM POx to raise its temperature. The hot syngas is expanded to recover power. After the syngas is expanded to ambient pressure, it is sent to the OTM boiler. Within the OTM boiler, syngas is first passed over an array of OTM tubes. Air is preheated by heat exchange with the O<sub>2</sub>-depleted air and then passed on to the feed side of the OTM tubes. Oxygen from the air transports across the membrane and reacts with the syngas. Since the rate of O<sub>2</sub> transport is limited by the availability of the membrane area, the oxidation of syngas will take place over a large area (the OTM zone) within the boiler. As the syngas gets oxidized, the driving force for O<sub>2</sub> transport will decrease and the required membrane area will increase. For practical reasons, the OTM will be used to supply O<sub>2</sub> to the fuel side until 80 to 90 percent fuel utilization is achieved. The remainder of fuel will be combusted using O<sub>2</sub> supplied from the cryogenic ASU.

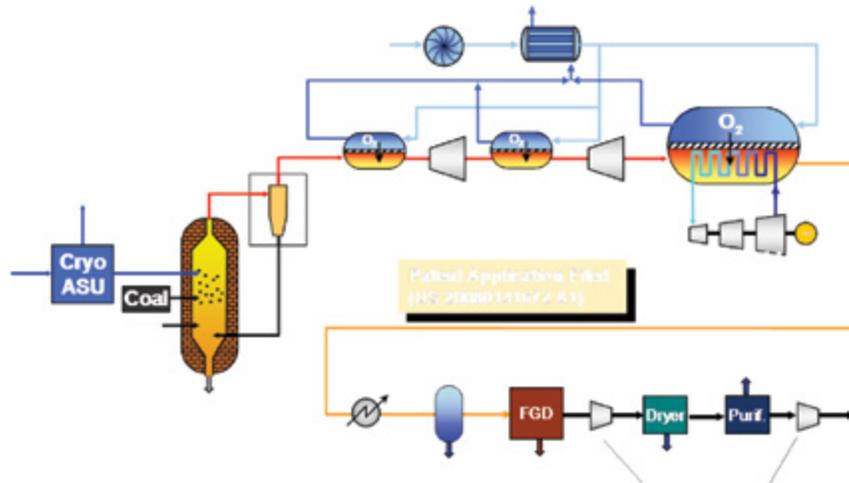


Figure 2: OTM-Based Process for Power Generation with CO<sub>2</sub> Capture

The thermal energy released within the boiler is used for steam generation. In the OTM zone, steam tubes will be interspersed with the OTM tubes such that the temperature is maintained at the optimum level for membrane performance. After the fuel is completely oxidized with externally supplied O<sub>2</sub>, the flue gas will pass through a convective section of the boiler for further steam generation and boiler feed water preheating. The flue gas exiting the boiler is processed according to a purification process proposed for a conventional oxy-fuel technology.

A number of OTM-based power cycles have been simulated for comparison against the U.S. Department of Energy (DOE) cost of electricity (COE) targets in 2008 dollars. Table 1 illustrates OTM cases with ultra-supercritical steam cycles and a warm gas clean-up (WGPU) desulfurization system. The base case for comparison is a DOE pulverized coal (PC) air-based combustion power cycle (without CO<sub>2</sub> capture) using a supercritical steam cycle. The table includes data regarding the net efficiency, plant cost, and COE for three different coal prices. The COE increase over the DOE base case is calculated, and those cases meeting the DOE target of <35 percent increase in COE are highlighted in green. The high net efficiency (>36 percent higher heating value [HHV]) seen in the OTM cases is a major contributing factor leading to achievement of the COE increase target.

TABLE 1: COE COMPARISON OF A STANDARD DOE AIR-COAL POWER CYCLE WITH OTM POWER CYCLES

Case	Current OTM Cases			Air-PC Case
	1 Main	2 3 Expanders	3 1 POx	Praxair/DOE No CCS SC
Net Efficiency	38.2	38	37.9	39.7
Cost Basis (year)	3/2008	3/2008	3/2008	3/2008
Plant Cost (\$/kW)	\$3,712	\$3,732	\$3,706	\$1,908
	Coal Price (\$/MMbtu)			
Increase in COE over Reference	1.8	42.1%	42.8%	42.1%
	3	36.4%	37.2%	36.6%
	4	32.8%	33.6%	33.0%

One part within Phase III of the project will focus on developing the technology to support the conversion of natural gas to syngas utilizing oxygen from a ceramic membrane in a development-scale syngas system (Figure 3). It is anticipated that this system will incorporate a second-generation OTM module that improves on the performance and cost of the proof-of-concept modules. A second part within Phase III of the project will focus on developing the technology to support the oxy-combustion of a syngas fuel utilizing oxygen from a ceramic membrane with a portion of the heat of combustion utilized to raise saturated steam and with the primary flue gas products being comprised of  $H_2O$  and  $CO_2$ . The unit will be designed to demonstrate heat transfer from the OTM modules to a process stream.

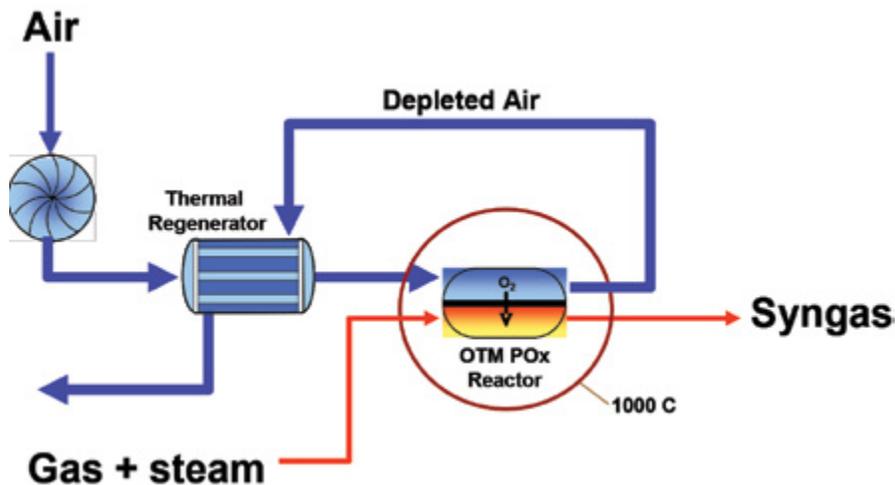


Figure 3: Phase III OTM Syngas System Concept

A third part within Phase III of the project will focus on the further development of ceramic materials and processes to support high-volume, low-cost manufacturing, as well as stable performance over an anticipated commercial operation lifetime. The main areas of focus will be on flux degradation, membrane creep life, and seal reliability.

### technology advantages

- The OTM oxy-combustion system can provide a highly concentrated, sequestration-ready stream of  $CO_2$  without costly cryogenic oxygen production or  $CO_2$  separation processes.
- The use of reactively driven OTMs is expected to reduce the power associated with oxygen production by 70 to 80 percent. This represents a step change in the cost and related  $CO_2$  emissions, and will enable a variety of oxy-combustion technologies, as well as other combustion applications, where  $CO_2$  capture may be required.
- The development of OTMs will also benefit industrial processes used to produce syngas for subsequent processing into a variety of chemical and/or petrochemical end products by dramatically reducing the power requirements and capital costs to supply oxygen to the process.

### R&D challenges

- Reliability and stability of the OTM tubes at high pressure and temperature.
- Achieving membrane cost and performance targets.
- Engineering design of OTM equipment, including OTM syngas reactor and boiler.

## results to date/accomplishments

- Demonstrated stable OTM performance with sulfur impurities in simulated coal-derived fuel gas.
- Developed pilot plant specifications for OTM oxygen flux and fuel utilization.
- Continued OTM performance improvement through characterization and manufacture of OTM tubes, as well as preparations for scale-up.
- Demonstrated achievement of flux and performance targets with advanced OTM materials.
- Developed manufacturing protocol for pilot-sized OTM tubes.
- Completed design and construction of the OTM multi-tube reactor at the University of Utah.
- Designed a hot oxygen burner (HOB) coal gasifier to achieve required testing parameters for the OTM system. The HOB was tested at Praxair's facilities and then sent to the OTM reactor at the University of Utah for integration. Testing on coal-derived syngas has been initiated.
- Completed testing on coal-derived syngas at the University of Utah.
- Developed conceptual design of prototype OTM module and methodology for reactor integration.
- Completed a basic design and cost estimate for an OTM boiler and POx unit supporting the OTM enabled Advanced Power Cycle with Shaw Energy and Chemicals.
- Completed detailed process models of an OTM syngas unit that have been used for development of equipment specifications and a design for a 160,000-standard cubic feet per day (scfd) development system.
- Completed construction of the 160,000-scfm OTM syngas development system.
- Demonstrated conversion of natural gas to syngas with oxygen supplied through OTM with initial proof-of-concept, multi-membrane modules.

## next steps

- Perform testing on the development-scale syngas system (160,000 scfd), which will validate all required startup and shut-down processes, as well as the overall performance of a fully integrated system.
- Execute design of second-generation OTM membrane and syngas module and demonstrate improvements in performance, stability, and economics.

## available reports/technical papers/presentations

Christie, M., et.al, "Oxygen Transport Membrane Based OxyCombustion for CO<sub>2</sub> Capture from Coal Power Plants," presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, September 2010. <http://www.netl.doe.gov/publications/proceedings/10/co2capture/presentations/tuesday/Maxwell%20Christie%20-%20NT43088.pdf>.

Adams, J.; Eddings, E.; Wilson, J.; and Li, J., "Development of a Coal-fired Oxygen Transport Membrane (OTM) Combustor," presented at the 2009 American Institute for Chemical Engineers Annual Meeting, Nashville, Tennessee, November 2009. <http://netl.doe.gov/File%20Library/Research/Coal/ewr/co2/43088-Praxair-OTM-AICHe-mtg-nov09.pdf>.

Wilson, J., et.al, "Oxygen Transport Membrane Based OxyCombustion for CO<sub>2</sub> Capture from Coal Power Plants," Technical paper from the Proceedings of the 34<sup>th</sup> International Technical Conference on Coal Utilization & Fuel Systems, Jun 2009, <http://netl.doe.gov/File%20Library/Research/Coal/ewr/co2/43088-Praxair-OTM-paper-Clearwater-jun09.pdf>.

Christie, M.; Wilson, J.; Degenstein, N.; and Shah, M., "Oxygen Transport Membrane Based OxyCombustion for CO<sub>2</sub> Capture from Coal Power Plants," presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D Meeting in Pittsburgh, Pennsylvania, March 2009, <http://www.netl.doe.gov/publications/proceedings/09/CO2/pdfs/43088%20Praxair%20oxy-combustion%20OTM%20%28Christie%29%20mar09.pdf>.

# OXY-COMBUSTION TECHNOLOGY DEVELOPMENT FOR INDUSTRIAL SCALE BOILER APPLICATIONS

B-430

OXY-COMBUSTION

## primary project goals

Alstom is developing and testing oxy-combustion technology for tangentially fired (T-fired) boilers in retrofit and new power plant applications.

## technical goals

- Design and develop an innovative oxy-combustion firing system for existing T-fired boilers that minimizes overall capital investment and operating costs.
- Evaluate the performance of oxy-combustion T-fired boilers in pilot-scale tests at Alstom's 15-MWth Boiler Simulation Facility (BSF).
- Determine the boiler design and performance impacts for oxy-combustion.
- Evaluate and improve engineering and computational fluid dynamic (CFD) modeling tools for oxy-combustion.
- Develop the design, performance, and costs for a demonstration-scale oxy-combustion boiler and auxiliary systems.
- Develop the design and costs for both industrial and utility commercial-scale reference oxy-combustion boilers and auxiliary systems, which are optimized for overall plant performance and cost.

## technical content

Initial screening studies were conducted to assess the impacts of a broad range of process variables and boiler design parameters on oxy-combustion boiler design, performance, and cost. This information was used to refine the test plan and establish design requirements for the 15-MWth testing.

### technology maturity:

15-MWth Pilot-Scale  
Oxy-Fired Boiler

### project focus:

Tangentially Fired Oxy-  
Combustion Boilers

### participant:

Alstom Power

### project number:

NT0005290

### NETL project manager:

Steven Mascaro  
steven.mascaro@netl.doe.gov

### principal investigator:

Armand A Levasseur  
Alstom Power  
armand.a.levasseur  
@power.alstom.com

### partners:

Illinois Clean Coal Institute  
North Dakota Industrial  
Commission  
Utility Advisory Group (10 utility  
companies)

### performance period:

9/30/08 – 4/30/14

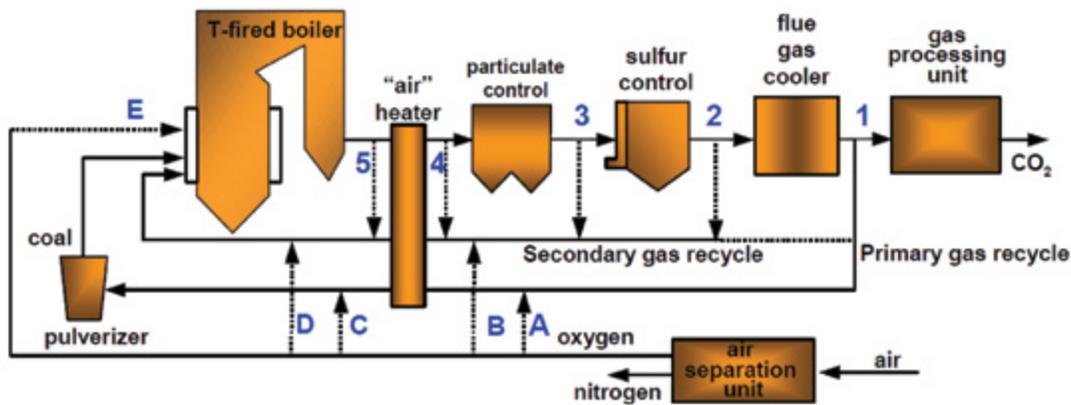


Figure 1: Simplified Oxy-Combustion Process Diagram

Figure 1 is a schematic representation of an oxy-combustion process showing the location of a T-fired boiler and several possible take-off locations for flue gas recirculation (1-5) and locations for oxygen injection (A-E). The different take-off locations will affect variables in the recirculation stream (i.e., water, particulate, and sulfur content) and impact the size of equipment.

The screening studies included evaluation of the following variables: gas recycle take-off location, gas recycle composition, gas recycle ratio, oxygen injection concentration and distribution, windbox design, and separate over-fire air design. Both process modeling and CFD analysis were applied.

CFD simulations of the BSF and an 850-MWe supercritical T-fired boiler were developed and used to evaluate various oxy-combustion design options. The BSF models were updated using boundary conditions and data from the BSF test runs to refine simulations and validate predictions.

Alstom Power has conducted a series of pilot tests in its 15-MWth BSF facility, shown in Figure 2. The BSF replicates the T-firing conditions in utility boilers. The BSF was modified for oxy-combustion operation and to provide flexibility to test over a broad range of conditions. The primary oxy-combustion test parameters evaluated include:

- Gas recycle rate and recycle sulfur capture rate.
- Distribution of gas recycle into the furnace.
- Excess oxygen.
- Oxygen injection location, distribution, and concentration.

Key aspects of boiler operation that were investigated under this project during the 15-MWth pilot testing and during design studies are shown in Figure 3.



Figure 2: Alstom 15 MWth Boiler Simulation Facility

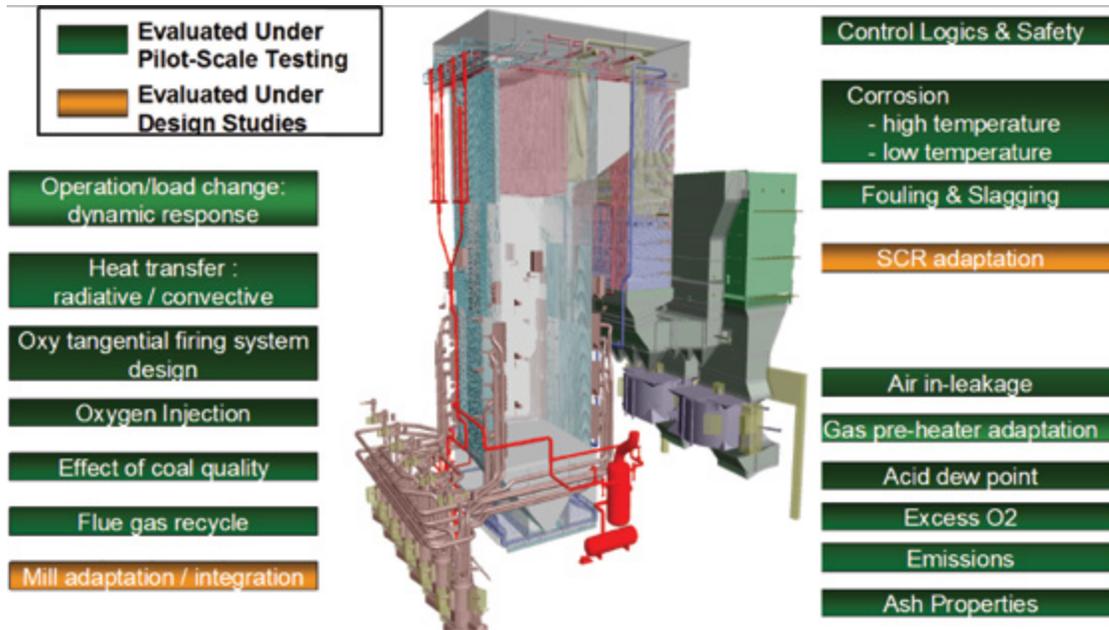


Figure 3: Aspects of Boiler Operation Requiring Assessment

The testing focused on control of furnace heat release rates and heat transfer for boiler thermal performance during oxy-combustion, while obtaining good fuel burnout and control of emissions. Measurements are conducted to assess ash deposition and fireside corrosion, as well as sulfur trioxide (SO<sub>3</sub>) formation and behavior of trace metals such as mercury.

Detailed furnace mapping measurements were also performed to better understand behavior during oxy-combustion, as well as to provide comprehensive data sets for model refinement and validation. Furnace and convection pass temperatures were measured using suction pyrometers. Heated gas extraction probes were used with a dedicated gas analyzer system to measure in-furnace gas compositions. Incident heat fluxes to the furnace walls were measured using total heat flux probes and radiant heat fluxes measured by ellipsoidal radiometer probes. Typical measurement planes are shown in Figure 4.

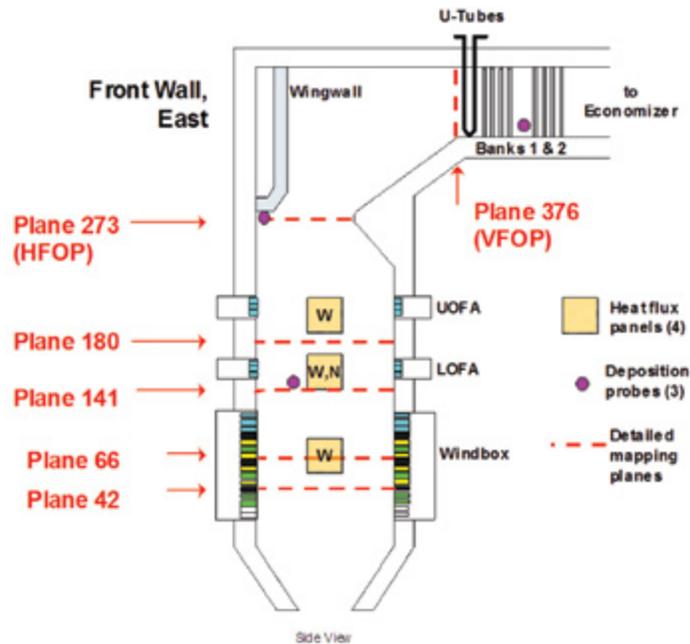


Figure 4: BSF Furnace Probe Measurement Planes

Results from the 15-MWth testing, along with the refined modeling tools (CFD and dynamic simulations), are being applied to develop a basic design for a full-scale oxy boiler, which can be rapidly applied for demonstration. This effort includes the oxy-boiler design with advanced oxy-firing system and operational controls, as well performance and cost data. Additionally, large, commercial-scale oxy-boiler reference designs are being developed for industrial and utility applications, which are integrated and optimized for overall oxy-plant performance and costs, and to provide a vision for the final commercial product.

### technology advantages

Oxy-combustion is a cost-competitive, near-term solution for carbon dioxide (CO<sub>2</sub>) capture that offers a relatively low technical risk due to use of conventional components. It can be used in new and retrofit applications, and has the potential for greater than 90 percent CO<sub>2</sub> capture. Oxy-combustion technology can be configured to provide a high degree of operating flexibility, which is increasingly important for electric grid stability as the portion of renewable power generation grows.

### R&D challenges

- Understanding difference from air-firing in pollutant formation, ash deposition, fireside corrosion, and heat transfer rates in an oxy-combustion boiler, as well as the control of air in-leakage.
- Application of this knowledge to reduce design risks and optimize designs for performance and costs.
- Overall integration to provide commercially attractive CO<sub>2</sub> reduction solutions.

### results to date/accomplishments

- Completed CFD and process screening evaluations.
- Completed modifications to the BSF for oxy-combustion operation to permit the firing under both air and oxygen, as well as with several flue gas recycle configurations and oxygen injection methods.
- Completed 15-MWth pilot-scale testing over a range of coal types, including sub-bituminous, low-sulfur bituminous, high-sulfur bituminous, and North Dakota lignite. Additionally, the test fuel burned at the 30-MWth Oxy-Pilot Plant at Vattenfall's Schwarze Pumpe station was also tested, providing a link to that comprehensive development program.
- These tests included both oxy- and air-firing tests, which examined the impacts of combustion and oxy-process parameters on boiler design and operation for each of the coals. Test parameters included re-circulated flue gas ratio, effect of oxygen concentration and oxygen distribution into the furnace, re-circulated flue gas take-off location, total excess oxygen, furnace combustion staging, air in-leakage rates, and reduced load operation.
- The following is a brief overview of the 15-MWth test results in some key areas examined:
  - General Oxy-Fired Operation – Stable operation over a broad range of test conditions; able to produce flue gas with greater than 90 percent CO<sub>2</sub> concentration (dry basis).
  - Combustion – Good performance during both air- and oxy-combustion testing for all coal types; able to operate at low-excess oxygen (less than 2 percent oxygen [O<sub>2</sub>] at the economizer outlet), low (near-zero) carbon monoxide (CO) emissions, and low carbon in fly ash.
  - Nitrogen Oxide (NO<sub>x</sub>) Emissions – Lower NO<sub>x</sub> emissions with oxy-combustion than air-firing; NO<sub>x</sub> emissions were generally more than 50 percent lower for oxy-fired than air-firing under similar staged combustion conditions.
  - Heat Transfer – Able to control furnace heat flux and temperature profiles during oxy-firing to be similar to those for air-firing.
  - SO<sub>3</sub> Formation – Sulfur dioxide (SO<sub>2</sub>) to SO<sub>3</sub> conversion appears to be similar to air-firing; however, SO<sub>3</sub> concentrations could be much higher for oxy-combustion depending on recycle scheme.
  - Ash Deposition – Appeared generally similar to air-firing in terms of deposits physical characteristics and in composition.
  - Waterwall Corrosion – Appeared generally similar to air-firing.

- Successfully completed 15-MWth BSF testing to evaluate second generation oxy-boiler concepts.
  - Firing system conditions were adjusted to optimize furnace heat flux distribution (lower peak heat flux, more uniform heat flux vertically and laterally), enabling lower gas recycle rates and high furnace heat absorption while maintaining material requirements.
  - Demonstrated close-coupled gas recycle concept, which has the potential to significantly reduce equipment cost and improve efficiency.
  - Demonstrated the use of eductors to drive flue gas recycle using oxygen as the motive gas.
- Assessed the overall integration of pollutant removal from the boiler to Gas Processing Unit, and evaluated the performance of different methods of controlling mercury (Hg) and acid gases during 15-MWth testing.
- Upgraded FLUENT CFD submodels for oxy-firing and applied detailed experimental data from the 15-MWth testing to refine CFD and validate simulations.
- Developed a detailed oxy-boiler model in Aspen Dynamics and integrated this model into an overall oxy-fired CO<sub>2</sub> capture plant dynamic model to support development of advanced controls and assess various transient operating scenarios.
- Developed the design of a nominal 400 MWe-gross, ultra supercritical steam boiler and auxiliary equipment that could be applied for oxy-combustion demonstration projects. The design allows dual air/oxy-fired operation and applies advanced controls.
- Developed the design of a nominal 900 MWe-gross, oxy-fired ultra supercritical steam boiler and auxiliary equipment optimized for overall power plant performance (cost of electricity).

#### next steps

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- Complete 15-MWth test data evaluation and documentation. Complete CFD model validation and uncertainty analysis for oxy-fired applications.
- Complete documentation of oxy-boiler design guidelines.
- Refine the demonstration-scale oxy-boiler design, performance and emission predictions, and costing information.
- Refine the design, performance, and costing for the large utility reference oxy-boiler and develop overall plant economics.

#### available reports/technical papers/presentations

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Levasseur, A., “Recovery Act: Oxy-Combustion Technology Development For Industrial-Scale Boiler,” presented at the 2012 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, July 2012. <http://www.netl.doe.gov/File%20Library/Events/2012/CO2%20Capture%20Meeting/A-Levasseur-Alstom-Oxy-combustion-Industrial.pdf>.

Kluger, F.; Wild, T.; Mönckert, P.; Levasseur, A.; Turek, D.; and Kenney, J., “15 MWth and 30 MWth Oxy-Combustion Pilot Testing of Lignite from Vattenfall’s Lusatia Open Cast Mine” presented at the 37<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems, Clearwater, Florida, June 2012.

Levasseur, A., et. al, “Oxy-combustion: an economic and robust clean power solution” presented at the 11<sup>th</sup> Annual Carbon Capture & Sequestration Conference, Pittsburgh, Pennsylvania, April 30 - May 3, 2012.

Levasseur, A.; Turek, D.; Kenney, J.; Edberg, C.; and Kang, S., “Oxy-Fired Tangential Boiler Development and Large-Scale (15 MWth) Validation,” presented at the IEA 2<sup>nd</sup> International Oxy-Combustion Conference, Yeppoon, Queensland, Australia, September 2011.

Levasseur, A.; Darling, S.; Kluger, F.; Mönckert, P.; Heinz G.; and Prodhomme, B., “Oxy-Combustion: A Promising CCS Technology,” presented at the CoalGen Conference & Exhibition, Columbus, Ohio, August 2011.

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Prodhomme, B. ; Pourchot, T. ; Heinz, G.; Mönckert, P.; Kluger, F.; and Levasseur, A., “Oxy-combustion: toward a successful CO<sub>2</sub> capture technology,” presented at the Power Gen Europe, Milano, June 2011.

Darling, S., et.al, “Oxy-Combustion: A Sound CCS Solution Built From Pilot Operation.” Power Gen International – 2010, Orlando, Florida, December 2010.

Clark, M.; Kenney, J.; Levasseur, A.; and Kang, S., “SO<sub>3</sub> Emissions From a Tangentially-Fired Pilot Scale Boiler Operating SO<sub>3</sub> Emissions From a Tangentially-Fired Pilot Scale Boiler Operating Under Oxy-Combustion Conditions,” 2010 AIChE Annual Meeting, Salt Lake City, Utah, November 2010.

Levasseur, A., “Oxy-Combustion Boiler Development for Tangential Firing,” presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, September 2010. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/Armand-Levasseur---NT0005290.pdf>.

Kluger, F., et.al, “Alstom’s Oxy-Combustion Technology Development – Update on Pilot Plants Operation,” presented at the 35<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems, Clearwater, Florida June 2010.

Levasseur, A., et.al, “Assessment of Oxy-Combustion Impacts on Boiler Design and Performance During 15 MWth Pilot-scale Testing,” presented at the 9<sup>th</sup> Annual Carbon Capture & Sequestration Conference, Pittsburgh, Pennsylvania, May 2010.

Levasseur, A., et.al, “Update on Alstom’s Oxy-Combustion Technology Development,” presented at the 12<sup>th</sup> Annual ELECTRIC POWER Conference & Exhibition, Baltimore, Maryland, May 2010.

Levasseur, A., et.al, “Alstom’s Oxy-Firing Technology Development and Demonstration-Near Term CO<sub>2</sub> Solutions,” presented at the 34<sup>th</sup> International Technical Conference on Coal Utilization and Fuel Systems. June 2009.

Andrus, H., “Oxy-Combustion Boiler Development for Tangential Firing,” presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D meeting, Pittsburgh, Pennsylvania, March 2009.

Levasseur, A.; Nsakala, N.; and Kluger, F., “Oxyfuel PC and CFB Solutions - A Promising Option for CO<sub>2</sub> Capture,” presented at 8<sup>th</sup> Annual Conference Carbon Capture & Sequestration, Pittsburgh, Pennsylvania, May 2009.

“Oxy-Firing Technology – Pilot Testing Leading to Large-Scale Demonstration,” presented at the 11<sup>th</sup> Annual Electric Power Conference, Chicago, Illinois, May 2009.

# FUTURE OF CCS TECHNOLOGY ADOPTION AT EXISTING PC PLANTS

B-436

OXY-COMBUSTION

## primary project goals

Argonne National Laboratory (ANL) is constructing scenarios that affect carbon capture and storage (CCS) adoption as combinations of cases for the following dimensions: electricity demand, nuclear growth, renewable energy growth, higher or lower gas price factors, and alternative policies.

## technical goals

- Extension of ANL's previous work in project FWP49539, "Evaluation of CO<sub>2</sub> Capture/Utilization/Disposal Options."
- Simulate oxy-combustion and amine-based processes using ASPEN.
- Expand the scenario analyses to focus on the value of coal-based CCS for existing pulverized coal (PC) plants and for other technologies, such as coal-to-liquids with CCS.
- ANL will examine pathways that expedite CCS adoption, such as accelerated research and development (R&D) and carbon dioxide (CO<sub>2</sub>) utilization for enhanced oil recovery (EOR).
- ANL will examine opportunities for R&D related to shale gas, such as developing CCS specifically for natural gas combined cycle (NGCC) units.

## technical content

In a previous project (FWP49539), ANL conducted engineering assessments and economic evaluations on retrofitting PC boilers with oxy-combustion, and then eventually repowering the site with integrated gasification combined cycle (IGCC). The engineering assessment for oxy-combustion was conducted with the ASPEN process model and the economic evaluations with the AMIGA macroeconomic model. The assessment investigated the entire life cycle of the plant, which included the mining of the coal, coal transportation, coal preparation, power generation, environmental controls, water use, pipeline CO<sub>2</sub> conditioning, and pipeline transport of CO<sub>2</sub> for sequestration.

ANL also conducted ASPEN modeling for 18 different oxy-combustion and air-fired cases. Three different power production ratings (150 MW, 300 MW, and 450 MW) were investigated. The model included a selective catalytic reduction (SCR) system and a flue gas desulfurization (FGD) system for flue gas clean-up.

The analysis in the current project will demonstrate and, to the extent possible, quantify the role and benefit of R&D related to the utilization and environmental control of fossil fuels. The impacts of R&D will be shown by comparing model results such as deployment rates, emissions reductions, and electricity costs across various scenarios. The scenarios will capture a number of CO<sub>2</sub> control regimes, R&D programs, and economic conditions in order to fully understand the role that R&D plays in each. With widespread deployment of CCS under a CO<sub>2</sub> reduction target, R&D that lowers cost of CO<sub>2</sub> capture and increases efficiency will be shown to have a high economic payoff.

Other topics to be analyzed with the ANL model are as follows: the value of coal-based CCS in high natural gas price scenarios; opportunities and obstacles for R&D, specifically

technology maturity:

Systems Analysis and  
Macroeconomic Modeling

project focus:

Analysis of CCS Technology  
Adoption

participant:

Argonne National Laboratory

project number:

FWP49806

*continued from FWP49539*

NETL project manager:

José Figueroa

jose.figueroa@netl.doe.gov

principal investigator:

Donald Hanson  
ANL

dhanson@anl.gov

partners:

None

performance period:

2/1/11 – 1/31/14

on NGCC with CCS, including cost and performance parameters; the conditions under which CO<sub>2</sub>-EOR enables faster deployment of CCS systems; the market opportunity for coal and biomass to liquid fuels and power co-production with CCS; and impacts on PC units, especially those retrofitted with CCS, from cycling due to intermittent grid generation from renewables.

### technology advantages

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The ANL model is especially designed to analyze the issues and scenarios described above.

### R&D challenges

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Capturing the impacts and costs of high intermittent renewable generation as it affects dispatchable coal generators, especially those that have adopted CCS.

### results to date/accomplishments

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Simulated the benefit of higher utilization (i.e., capacity factor) for PC plants that retrofit CCS because of rising up the loading order (i.e., dispatch order).

### next steps

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- Analysis of the value of coal-based CCS in high natural gas price scenarios.
- Analysis of opportunities and obstacles for R&D, specifically on NGCC with CCS, including cost and performance parameters.
- Analysis of market opportunity for coal and biomass to liquid fuels and power co-production with CCS.
- Analyze impacts on PC units, especially those retrofitted with CCS, from cycling due to intermittent grid generation from renewable.

### available reports/technical papers/presentations

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Hanson, D., and Schmalzer, D., “CCS Adoption Under Alternative Market Conditions,” presented at U.S. Association for Energy Economics Conference, Austin, TX, November 2012.

Hanson, D., “Economics and Adoption of CO<sub>2</sub> Capture for Existing PC Plants in a Power System Context,” Eleventh Annual Conference on Carbon Capture, Utilization & Sequestration, Pittsburgh PA, May 2012.

Hanson, D., “Future of CCS Technology Adoption at Existing PC Plants,” presented at CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, July 2012.

Hanson, D.; Marano, J.; and Fout, T., “Economic Analysis of Existing Coal Plant Retrofits with CCS,” Energy, Utility, & Environmental Conference, Phoenix AZ, January 2012.

Hanson, D., “A Market Scenario Approach to Managing Existing Power Plant Assets,” 13<sup>th</sup> Annual Electric Power Conference and Exhibition, Rosemont, IL, May 2011.

Hanson, D., and Doctor, R., “Future of CCS Technology Adoption at Existing PC Plants,” presented at CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, August 2011.

“ANNUAL REPORT 2009: Evaluation of CO<sub>2</sub> Capture and Sequestration Using Oxyfuels with AMIGA Economic Modeling,” November 23, 2009.

Doctor, R.; Hanson, D. A.; and Molburg, J. C., “Evaluation of CO<sub>2</sub> Capture and Sequestration Using Oxyfuels with AMIGA Economic Modeling,” presented at 2009 NETL Capture Technology Meeting, March 2009.

# OXY-FIRED PRESSURIZED FLUIDIZED BED COMBUSTOR DEVELOPMENT AND SCALE-UP FOR NEW AND RETROFIT COAL-FIRED POWER PLANTS

## primary project goals

This project will evaluate a novel process for pressurized oxy-combustion in a fluidized bed reactor. The pressurized combustion in oxygen and the recycle of carbon dioxide (CO<sub>2</sub>) gas eliminates the presence of nitrogen and other constituents of air, minimizing the generation of pollutants and enabling the economic capture of CO<sub>2</sub> gas.

## technical goals

- Validate the performance of the Oxy-Pressurized Fluidized Bed Combustor (PFBC) system using standardized National Energy Technology Laboratory (NETL) tools and to identify technology gaps that need to be closed.
- The primary project goal is to validate the Oxy-PFBC technology's ability to exceed the U.S. Department of Energy (DOE)/NETL carbon capture goal of achieving greater than 90 percent CO<sub>2</sub> removal with no more than a 35 percent increase in cost of electricity (COE).

## technical content

Phase I – Conducted risk reduction of the PFBC concept through process and economic models, bench-scale chemistry testing, and cold flow fluidized bed testing to validate heat transfer, particle models, and reaction rates.

Phase II – Validate the oxy-fired PFBC process with component development, pilot plant testing, and updated specific process performance and economic models. Improve the quality of the model predictions through targeted research testing the validity of assumptions.

Phase III – Commercialize the technology in retrofit project at a working municipal power utility.

technology maturity:

Bench-Scale

project focus:

Pressurized Fluidized Bed Combustor Development

participant:

Aerojet Rocketdyne

project number:

FE0009448

NETL project manager:

Bruce Lani

bruce.lani@netl.doe.gov

principal investigator:

Mark Fitzsimmons  
Aerojet Rocketdyne

mark.fitzsimmons@pwr.utc.com

partners:

Linde LLC

Electric Power Research Institute  
- EPRI

Pennsylvania State University  
Jamestown Board of Public  
Utilities

performance period:

10/1/12 – 9/30/13

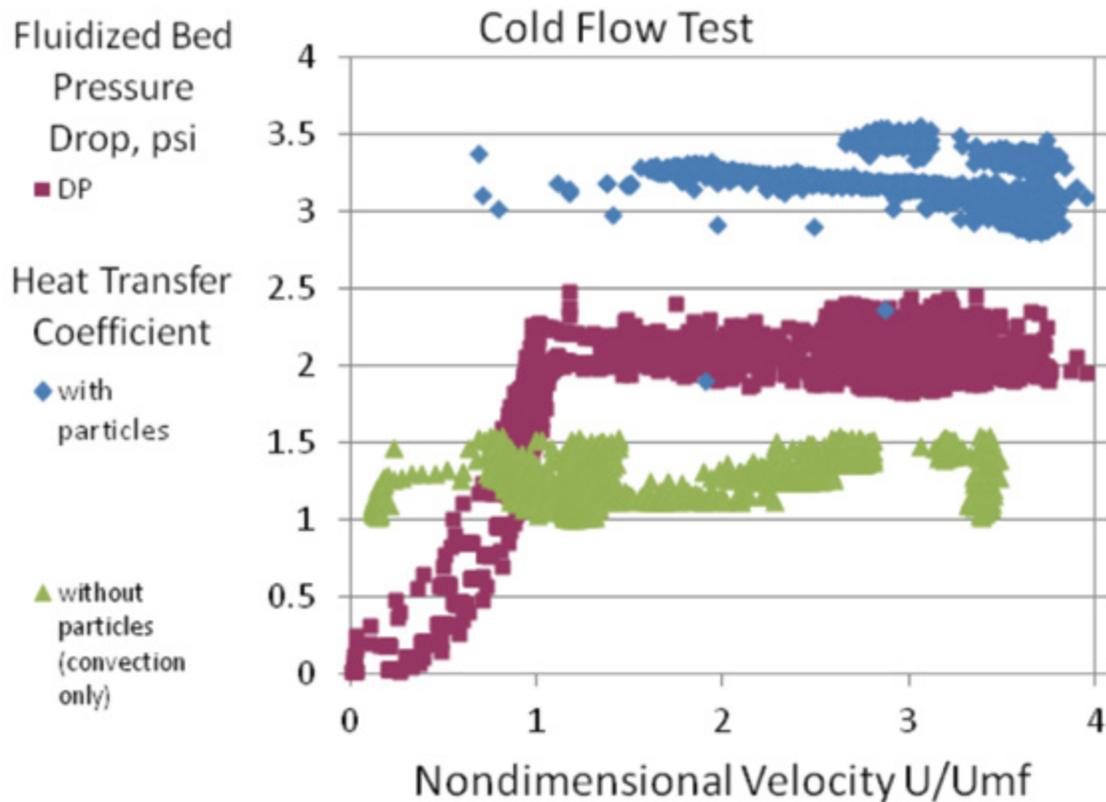


Figure 1: Cold Flow Test

Cold flow tests have demonstrated predictable (flat) behavior over a very wide range of flow rates. Historically, fluidized bed combustors also demonstrate constant temperatures throughout the bed, and these tests are also confirming this result, even in a situation where one might predict thermal transients due to uneven heat input across one or more dimensions of the bed.

Particle residence times have been measured on the order of minutes, validating the prediction that the high aspect ratio pressurized fluidized bed under development will combine the best features of a circulating fluidized bed (high carbon utilization) and a bubbling fluidized bed (low velocities), and put it all in a smaller package than either of these atmospheric pressure devices

### technology advantages

- Production of electricity from coal with near-zero emissions.
- Captured  $\text{CO}_2$  may be utilized for enhanced oil recovery or sequestered.

### R&D challenges

- Prediction of particle temperatures.
- Identification of common assumptions for equipment that may be re-used versus equipment that must be new for the proposed PFBC technology.

results to date/accomplishments

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B-440

OXY-COMBUSTION

- Bench-scale chemistry (kinetics) testing of sulfation at pressurized oxy-combustion conditions.
- Cold flow testing of full-scale tubes and particles in fluidized beds with similar non-dimensional parameters has validated heat transfer coefficients and correlations for predicting in the pressurized fluidized bed.
- Particle elutriation rates and residence times measured in cold flow.
- Bed stability tests in cold flow validate the method of bubble control.
- Particle model that balances fluidization, heat release, and steam production combined into a finite difference model using the method of characteristics to compute fluidized bed design fitness.
- Cost performance validated with revised and higher fidelity process and economic model.
- Cost and performance model tied to detailed design parameters in integrated system optimization architecture to trace component design changes and how they cascade through the system to alter COE, fluidization, thermals, carbon utilization, and other top-level constraints.

next steps

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- Combine kinetics, fluidization, and heat removal in pilot test to validate integrated combustion and steam production analyses.
- Develop and anchor particle models with test data for inclusion in computational fluid dynamic simulations.
- Scale-up technology to the demonstration plant-scale for an existing retrofit plant.

available reports/technical papers/presentations

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N/A.

# HIGH EFFICIENCY MOLTEN-BED OXY-COAL COMBUSTION WITH LOW FLUE GAS RECIRCULATION

B-441

OXY-COMBUSTION

## primary project goals

Gas Technology Institute (GTI) is developing a novel pressurized, oxy-coal combustion process to enable efficient carbon capture from coal-fired power plants. The project objective is to assess the performance and cost of a fully integrated GTI pressurized oxy-combustion supercritical pulverized coal (SCPC) power plant with carbon dioxide (CO<sub>2</sub>) recovery, burning Illinois #6 bituminous coal, for comparison to a reference 550-MW air-fired atmospheric SCPC power plant without CO<sub>2</sub> recovery.

## technical goals

- Complete engineering design and economic analysis of a pressurized molten-bed oxy-coal boiler.
- Complete thermodynamic energy and exergy analyses, oxy-coal burner testing, and corrosion assessment.

## technical content

The molten-bed combustor concept is derived from proven GTI engineering of a submerged combustion melting process. It is designed to maximize efficiency and minimize the cost of pressurized oxy-combustion by greatly reducing flue gas recirculation (FGR) while operating at elevated pressure. The unique combustion and heat transfer design employs a smaller and less expensive combustor and reduced gas-phase heat exchanger surface area. Decreasing the amount of FGR by up to 90 percent reduces capital cost and requires less maintenance.

A small amount of flue gas is used for coal crushing and transportation to the burners. Exhaust gas handling and cleaning equipment will be reduced in size and cost. Cost savings are also realized from ash recovery as frit rather than micron-sized fines, which can be difficult to handle, and the potential exists to capture a large portion of the sulfur and mercury in stable forms in the molten slag.

The design pulverized coal power plant in this study is a pressurized, oxy-combustion supercritical steam-electric generating power plant with 90 percent carbon capture and generating a nominal 550 MWe. The Combustion Block contains the following major systems that are directly associated with pressurized oxy-combustion of coal:

- Pulverized Coal Pressurized Storage and Transport/Feed System.
- Pressurized Submerged Combustion Molten-Bed Furnace/Boiler.
- Pressurized Convective Superheater, Reheater, Economizer, and Condenser.
- CO<sub>2</sub>-Rich Flue Gas Treating and Conditioning Facilities.
- Product CO<sub>2</sub> Recovery, Purification, and Compression Facilities.

In Phase I of this project, work will be focused on technical and economic analysis of a pressurized oxy-combustion power plant design based on the molten-bed combustor and following National Energy Technology Laboratory (NETL) protocols with comparisons to a

technology maturity:

Bench-Scale

project focus:

High Efficiency Molten-Bed Oxy-Combustion

participant:

Gas Technology Institute (GTI)

project number:

FE0009686

NETL project manager:

Steven Mascaro

steven.mascaro@netl.doe.gov

principal investigator:

David Rue

Gas Technology Institute

david.rue@gastechnology.org

partners:

Nexant

Brigham Young University (BYU)

Reaction Engineering

International (REI)

performance period:

10/1/12 – 9/30/13

baseline supercritical steam power plant burning Illinois #6 bituminous coal.

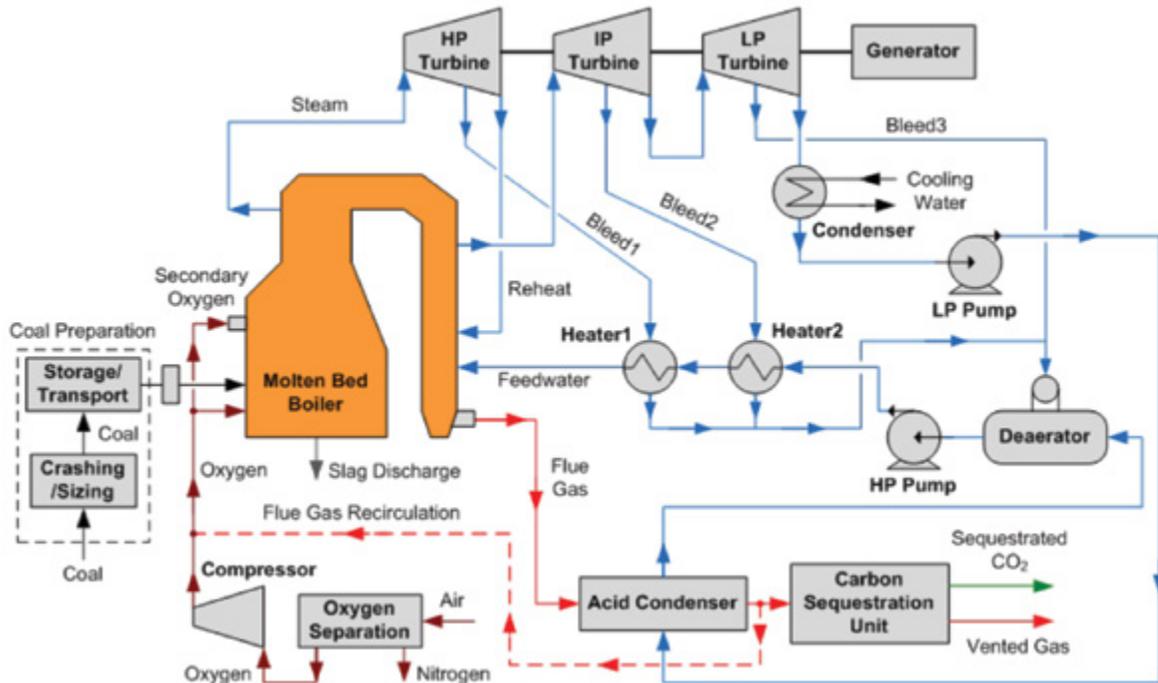


Figure 1: Schematic Molten Bed Boiler

### technology advantages

- Boiler is fuel-flexible with no modification, using coal and natural gas.
- Higher efficiency than other known oxy-coal combustors by greatly reducing the FGR and by operating at elevated pressure.
- Smaller, less expensive combustor and reduced gas phase heat exchanger surface area.

### R&D challenges

- Chance of elutriation of slag into the convective pass.

### results to date/accomplishments

- Completion of basic engineering design with ASPEN Plus process model.
- Completion of techno-economic study design basis and methodology report.

### next steps

- Oxy-coal burner testing and thermodynamic analysis.
- Corrosion testing and set-up for preliminary computational fluid dynamic model.

### available reports/technical papers/presentations

Kickoff meeting presentation, "High Efficiency Molten-bed Oxy-Coal Combustion with Low Flue Gas Recirculation," Pittsburgh, Pennsylvania, October 2012. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/gti-kick-off.pdf>.

# CHARACTERIZATION OF OXY-COMBUSTION IMPACTS IN EXISTING COAL-FIRED BOILERS

## primary project goals

Reaction Engineering International (REI) is developing mechanistic descriptions and analytical tools to characterize and predict impacts of carbon dioxide (CO<sub>2</sub>) flue gas recycle (FGR) and burner feed design on flame characteristics, fouling, slagging, and corrosion, inherent in the retrofit of existing coal-fired boilers for oxy-combustion. Investigations include laboratory-, bench-, and pilot-scale tests, as well as computer simulations of pilot-scale and full-scale systems.

## technical goals

- Utilize multi-scale testing and theory to develop:
  - Fundamental data that describe flame characteristics, corrosion rates, and ash properties during oxy-combustion coal firing.
  - Validated mechanisms that describe oxy-combustion processes.
  - Firing system principles that guide oxy-burner design and FGR properties.
- Incorporate validated mechanisms into a computational fluid dynamics (CFD) model to evaluate full-scale oxy-combustion retrofit designs:
  - Predict flame characteristics and surface impacts for different full-scale oxy-firing designs and FGR properties.

## technical content

REI is characterizing and predicting the performance and operational impacts of oxy-combustion retrofit designs on existing coal-fired boilers. The project focus is to develop tools to characterize and predict impacts of FGR and burner feed design on flame characteristics (burnout, nitrogen oxide [NO<sub>x</sub>], sulfur oxide [SO<sub>x</sub>] and fine particle emissions, heat transfer), fouling, slagging, and corrosion. Testing includes production of 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-combustion conditions are being developed and validated using the data generated during experimentation and from the literature. The mechanisms are being implemented into a CFD code and an existing coal-fired utility boiler is modeled under air- and oxy-combustion conditions to identify the likely impacts of retrofit.

This project is 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 allow accurate prediction of these impacts (through mechanism development). Experiments are performed on three different scales: (1) a bench-scale optical entrained flow reactor is used to elucidate the impact of oxy-combustion flue gas composition on the rate of char oxidation; (2) a 100-kW lab-scale combustor is used to characterize the effects of FGR on ash characteristics; and (3) a 1.2-MW pilot-scale combustor is used to investigate burner and firing system principles, deposition, corrosion, and radiative heat transfer, including soot evolution. The data from these experiments are used to guide development of mechanisms that may be used to describe char oxidation, deposition (slagging and fouling), corro-

### technology maturity:

Laboratory-, Bench-, and Pilot-Scale; Simulated and Actual Flue Gas; 0.3-8 Tonnes of CO<sub>2</sub> Per Day

### project focus:

Characterization of Oxy-Combustion Impacts

### participant:

Reaction Engineering International

### project number:

NT0005288

### NETL project manager:

Andrew Jones  
andrew.jones@netl.doe.gov

### principal investigator:

Bradley R Adams  
Reaction Engineering International  
adams@reaction-eng.com

### partners:

Brigham Young University  
Corrosion Management LTD  
PacifiCorp  
Praxair, Inc.  
Siemens Energy  
University of Utah  
Vattenfall AB

### performance period:

10/1/08 – 9/30/13

sion, and soot evolution. The data generated are used to produce an overview of firing system principles for oxy-combustion that should help guide design of full-scale firing systems.

B-444

### Bench-Scale Optical Entrained Flow Reactor Experiments

Bench-scale experimentation is conducted at Sandia National Laboratories to further clarify the behavior of char in an oxygen ( $O_2$ )-enriched FGR gas matrix. These experiments are conducted in Sandia's optical entrained flow reactor and associated particle-sizing pyrometry diagnostics.

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

Data related to ash characterization and deposition are collected using experiments conducted in the University of Utah's 100-kW Oxy-Fuel Combustor (OFC), shown in Figure 1. The furnace consists of an OFC 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.

These air- and oxy-fired experiments focus primarily on effects of FGR on ash chemistry, under practical time/temperature/particle composition conditions. Multiple modes of gas recycle are tested to investigate FGR conditions ranging from hot, moist, and particle-laden gases to clean  $CO_2$ . This information provides insight into how slagging and fouling could be impacted under oxy-firing conditions. Measurements include sampling of ash aerosol using low-pressure impactors and isokinetic dilution probes. In addition, mobility particle sizers determine ultrafine (sub-micron) particle concentrations and particle size distributions, and energy-dispersive X-ray spectroscopy (EDS) and computer-controlled scanning electron microscopy (CCSEM) techniques are used to identify ash compositions.

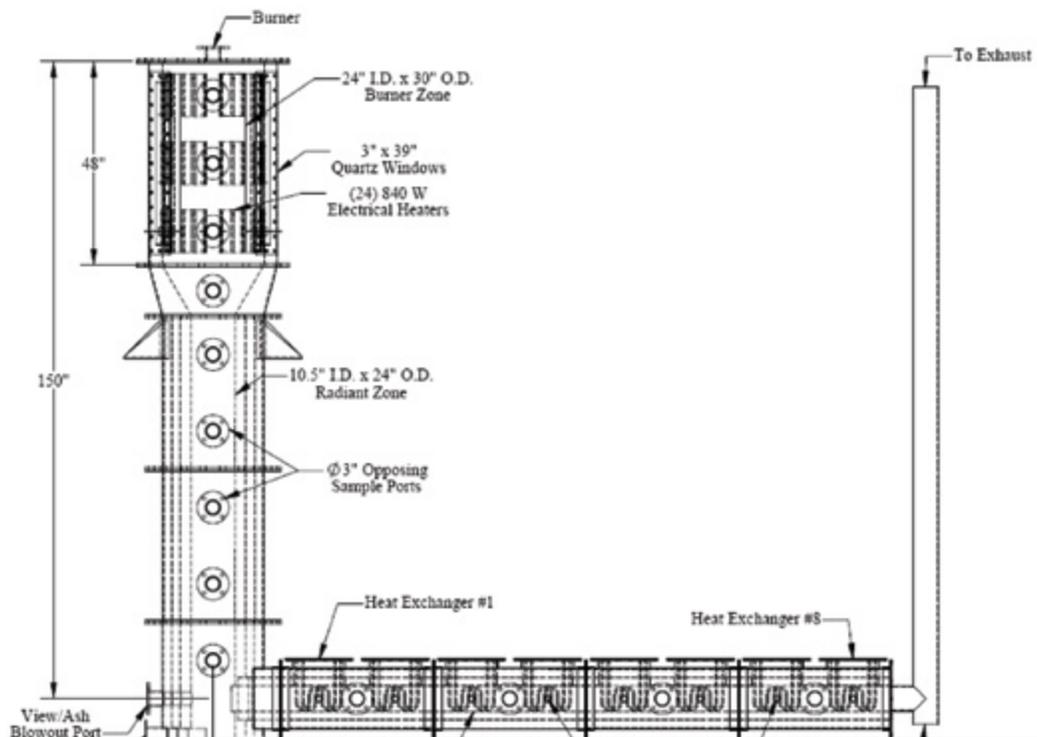


Figure 1: Diagram of 100-kW Oxy-Fuel Combustor



Figure 2: Pilot-Scale Combustion Facility (L1500)

### 1.2 MW Pilot-Scale Furnace (L1500) Experiments

Pilot-scale experiments are performed in a 1.2-MW pulverized coal furnace (L1500). These experiments are designed to investigate firing system configuration impact on flame stability and heat transfer, soot evolution and water wall, and superheat material corrosion. A picture of the L1500 is shown in Figure 2. Four weeks of parametric testing were performed in the L1500 to develop practical guidelines that allow optimized operation of an actual burner under oxy-combustion conditions with FGR. Burner operating parameters of interest include:

1. Variable  $O_2$ , FGR, and coal distribution in the burner.
2. Variable FGR ratios that produce 27 and 32%  $O_2$  in the  $O_2$ /FGR mixture.
3. Variable burner stoichiometric ratio within the range of 0.8 to 1.2.
4. Targeted  $O_2$  injection.
5. Furnace staging of FGR independent of burner conditions.

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

1. Air- and oxy-fired conditions.
2. Powder River Basin (PRB) and bituminous coals.
3. Optimized  $O_2$ , 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 performed in the pilot-scale furnace include:

1. Flue gas composition ( $O_2$ ,  $CO_2$ , carbon monoxide [CO],  $NO_x$ , mercury [Hg], and sulfur dioxide [ $SO_2$ ]) using existing measurement equipment.
2. Unburned carbon in ash, using loss on ignition (LOI) 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.

Follow-on testing was performed to characterize performance of mercury control technologies under air- and oxy-firing conditions. Bromine corrosion impacts were also measured.

### technology advantages

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- Enable the development of validated combustion mechanisms specifically designed for oxy-combustion with FGR for retrofitting existing coal-fired boilers.
- Identify firing system principles that guide oxy-burner design and potential retrofit strategies.
- Quantify impacts of FGR properties on ash deposition.
- Quantify impacts of oxy-combustion firing on corrosion of typical boiler waterwall and superheat materials.
- Develop the capability to assess the performance and optimize the retrofit of oxy-combustion to a full-scale existing boiler to minimize retrofit cost and maximize operability.

### R&D challenges

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- Ability to control and quantify how much air-in leakage occurs inside the OFC and L1500 furnace during the experiments.
- Development and operation of a coal-fired research burner specifically designed to span a range of coal/O<sub>2</sub>/FGR injection strategies and operating conditions.
- Correlation of experimental data with mechanisms and model predictions in order to provide adequate mechanism/model validation.

### results to date/accomplishments

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- Parametric burner test completed.
- Burner primary retrofit results:
  - Matching burner primary gas/fuel ratio or momentum ratio with air-fired operation produced a stable attached flame (good retrofit strategies).
  - Matching primary velocity with air-fired operation did not provide a stable attached flame (may depend on burner flexibility).
  - There is a fundamental difference in devolatilization rates and ignition between air and oxygen/FGR firing.
  - A stable flame can be achieved with no oxygen enrichment in the primary.
- Oxygen injection results:
  - Targeted oxygen injection at the burner primary-inner secondary boundary produced the most effective flame stabilization and ignition. Axial injection produced a permanently detached flame and radial injection produced a wider flame, but did not improve flame attachment.
  - Axial injection in the primary produced a more intensely radiating flame downstream of burner; secondary injection produced a less intense flame near the burner.
  - For maximum pre-mixed primary O<sub>2</sub>, adding small amounts of oxygen enrichment radially through the bluff body did not improve flame attachment.
- Soot volume fraction  $\approx$ 40 percent lower for oxy-combustion cases than air-fired cases for the Utah coal, and 10 to 20 percent lower for PRB coal.
- NO<sub>x</sub> emissions were up to 75 percent lower with oxy-combustion, depending on level of staging.

- Corrosion testing results:
  - Waterwall (SA210) corrosion rates decreased when converting from air- to oxy-firing for all coals.
  - Superheater (T22, P91, and 347H) rates generally increased when converting from air- to oxy-firing.
  - 347H corrosion rates increased dramatically for  $\text{SO}_2 > \approx 3,000$  parts per million (ppm) and  $T_{\text{probe}} < \approx 1,150^\circ\text{F}$ .
  - Corrosion for lower alloyed materials (T22, SA210) increased when changing between oxidizing-reducing. Likely to contribute to in-plant corrosion in near-burner and near-overfire air (OFA) port regions. Transient effects cannot be resolved using coupon tests.
- Mercury testing results:
  - Native mercury removal was higher for both OFC and L1500 cases.
  - Similar speciation for air- and oxy-firing; slightly lower total gas mercury under oxy conditions.
- Aerosol/Fine ash testing results:
  - LOI or carbon in the ash is nearly always reduced under oxy-fired conditions of 27 to 32 percent  $\text{O}_2$  as a result of increased oxygen and residence time to facilitate greater char oxidation when compared to air-fired combustion.
  - Black carbon or soot is also nearly always reduced when switching from air- to oxy-fired conditions in the 27 to 32 percent  $\text{O}_2$  range.
  - Overall particle size distributions for a given coal are not fundamentally different between air- and oxy-fired conditions of similar temperatures (i.e., 27 to 32 percent  $\text{O}_2$  range).
  - Black carbon or soot is a significant contributor to the ultrafine particle mode.
  - Particle size distributions are affected by stoichiometric ratio due to differences in soot and sulfate production.
  - Differences between air- and oxy-fired partitioning of elements in the ash are subtle and do not indicate the need for fundamental changes in aerosol handling or potential aerosol toxicity.
  - Coal rank and chemistry dominates the partitioning of ash particles and elements and combustion atmosphere has a comparatively limited mechanistic influence.
  - No aerosol data were discovered across a wide range of coal compositions and oxy-fired combustion conditions that would indicate a significant problem or hurdle for retrofitting an existing boiler with oxy-fired technology.
- Retrofit modeling results (PacifiCorp Hunter Unit 3):
  - REI's Glacier combustion simulation (CFD) tool was used to simulate air- and oxy-firing in Hunter Unit 3, based on an oxy-retrofit design by Siemens. Modeling included improved char oxidation, soot, slagging, and corrosion models.
  - The new, enhanced char oxidation sub-model predicted reasonable unburned carbon (UBC) concentrations (3.8 percent for air-firing compared with the 2 to 3 percent UBC observed at the Hunter plant) and decreased UBC for oxy-combustion firing. This decrease in UBC level from air- to oxy-firing was consistent with test results from the L1500 pilot-scale tests.
  - A 26 percent  $\text{O}_2$  mixture of oxygen and flue gas recycle was able to match the air-fired case heat transfer fairly well. For the 26 percent  $\text{O}_2$  condition, unburned carbon was lower in the oxy-fired case (2.1 percent versus 3.6 percent in the air-fired case). Total corrosion potential was also higher for all surfaces in the oxy-fired case. However, peak corrosion was lower for the waterwalls in the oxy-fired case and higher for the other surfaces. These modeling results suggest that selection of flue gas composition with a 26 percent  $\text{O}_2$  mixture of  $\text{O}_2$  and flue gas recycle can reasonably match heat transfer in an existing coal boiler and that there are not any catastrophic differences in an oxy-fired system compared to air-firing.

### next steps

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- Complete pilot-scale mercury testing.
- Complete pilot-scale corrosion testing (to study bromine-based corrosion).
- Complete boiler mercury case study.

B-448

Fry, A.; Adams, B.; and Fout, T., "Carbon, Sulfur and Nitrogen Behavior during Pilot-Scale Testing of Oxy-Coal Combustion," Paper #49, Power Plant Air Pollutant Control "MEGA" Symposium, Baltimore, MD, August 2012.

Fry, A.; Adams, B.; Davis, K.; Swensen, D.; and Cox, W., "Fire-Side Corrosion of Heat Transfer Surface Materials with Air- and Oxy-coal Combustion," Paper #51, Power Plant Air Pollutant Control "MEGA" Symposium, Baltimore, MD, August 2012.

Fry, A., et.al, "Fireside Corrosion of Heat Transfer Surface Materials for Air- and Oxy-coal Combustion," presented at the IEA 2<sup>nd</sup> Oxyfuel Combustion Conference, Yeppoon, Australia, September 2011.

Fry, A., and Adams, B., "Assessment of Full-scale Boiler Oxy-combustion Retrofit Using CFD Modeling," presented at the IEA 2<sup>nd</sup> Oxyfuel Combustion Conference, Yeppoon, Australia, September 2011.

Fry, A., and Adams, B., "Oxy-burner Retrofit Principles for Existing Coal-fired Utility Boilers," Poster at IEA 2<sup>nd</sup> Oxyfuel Combustion Conference, Yeppoon, Australia, September 2011.

Eddings, E.; Ahn, J.; Wang, L.; Okerlund, R.; and Fry, A., "Transformations of Fuel Sulfur During Oxy-Coal Combustion," presented at the IEA 2<sup>nd</sup> Oxyfuel Combustion Conference, Yeppoon, Australia, September 2011.

Adams, B., and Fry, A., "Characterization of Oxy-combustion Impacts in Existing Coal-fired Boilers," presented at the 2011 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, August 2011.

Fry, A., et.al, "Fire-Side Corrosion of Heat Transfer Surface Materials for Air- and Oxy-coal Combustion," presented at The 36<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems (Clearwater Clean Coal Conference), Clearwater, Florida, June 2011.

Fry, A.; Van Otten, B.; Adams, B.; and Bool, L., "Mercury Speciation and Emission from Pilot-scale PC Furnaces under Air- and Oxy-fired Conditions," presented at The 36<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems (Clearwater Clean Coal Conference), Clearwater, Florida, June 2011.

Adams, B.; Fry, A.; Shig-Shim, H.; and Van Otten, B., "CFD Modeling of Pilot-Scale Oxy-combustion Experiments," presented at The 36<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems (Clearwater Clean Coal Conference), Clearwater, Florida, June 2011.

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Eddings, E.G.; Ahn, J.; Wang, L.; Okerlund, R.; and Fry, A., "Transformations of Fuel Sulfur During Oxy-Coal Combustion," presented at The 36<sup>th</sup> International Technical Conference on Clean Coal & Fuel Systems (Clearwater Clean Coal Conference), Clearwater, Florida, June 2011.

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# NOVEL SUPERCRITICAL CARBON DIOXIDE POWER CYCLE UTILIZING PRESSURIZED OXY-COMBUSTION IN CONJUNCTION WITH CRYOGENIC COMPRESSION

## primary project goals

Southwest Research Institute will demonstrate a novel supercritical carbon dioxide (CO<sub>2</sub>) power cycle utilizing pressurized oxy-combustion through system process and economic modeling to evaluate overall plant performance and economic cost.

## technical goals

- Achieve high overall plant efficiencies with 90 percent CO<sub>2</sub> capture and compression to 2,200 pounds per square inch (psi).
- Identify a favorable pressurized supercritical CO<sub>2</sub> cycle configuration from several candidate cycles.
- Identify critical enabling technologies that require further development to realize the supercritical oxy-combustion power cycle.

## technical content

Conventional oxy-combustion with carbon capture includes two sub-systems that require significant power consumption that have a significant impact on overall plant efficiencies: the power required to generate O<sub>2</sub> through cryogenic air separation prior to combustion, and the power required to compress the captured CO<sub>2</sub> to pipeline pressures. Supercritical oxy-combustion addresses these two areas by combusting coal in a supercritical oxy-combustor where the recycled CO<sub>2</sub> is above the critical pressure and is in the dense phase region.

It is anticipated that the supercritical oxy-combustor will achieve high combustion efficiencies, decreasing fuel and oxygen consumption for a fixed power output, and will require less power for CO<sub>2</sub> compression because the captured CO<sub>2</sub> is already in the dense phase region where pumping can be used to reach pipeline pressure as opposed to gaseous phase compression.

technology maturity:

**Bench-Scale**

project focus:

**Supercritical CO<sub>2</sub> Power Cycle**

participant:

**Southwest Research Institute**

project number:

**FE0009395**

NETL project manager:

**Bruce Lani**

[bruce.lani@netl.doe.gov](mailto:bruce.lani@netl.doe.gov)

principal investigator:

**Klaus Brun**

**Southwest Research Institute**

[klaus.brun@swri.org](mailto:klaus.brun@swri.org)

partners:

**Thar Energy LLC**

performance period:

**10/1/12 – 9/30/13**

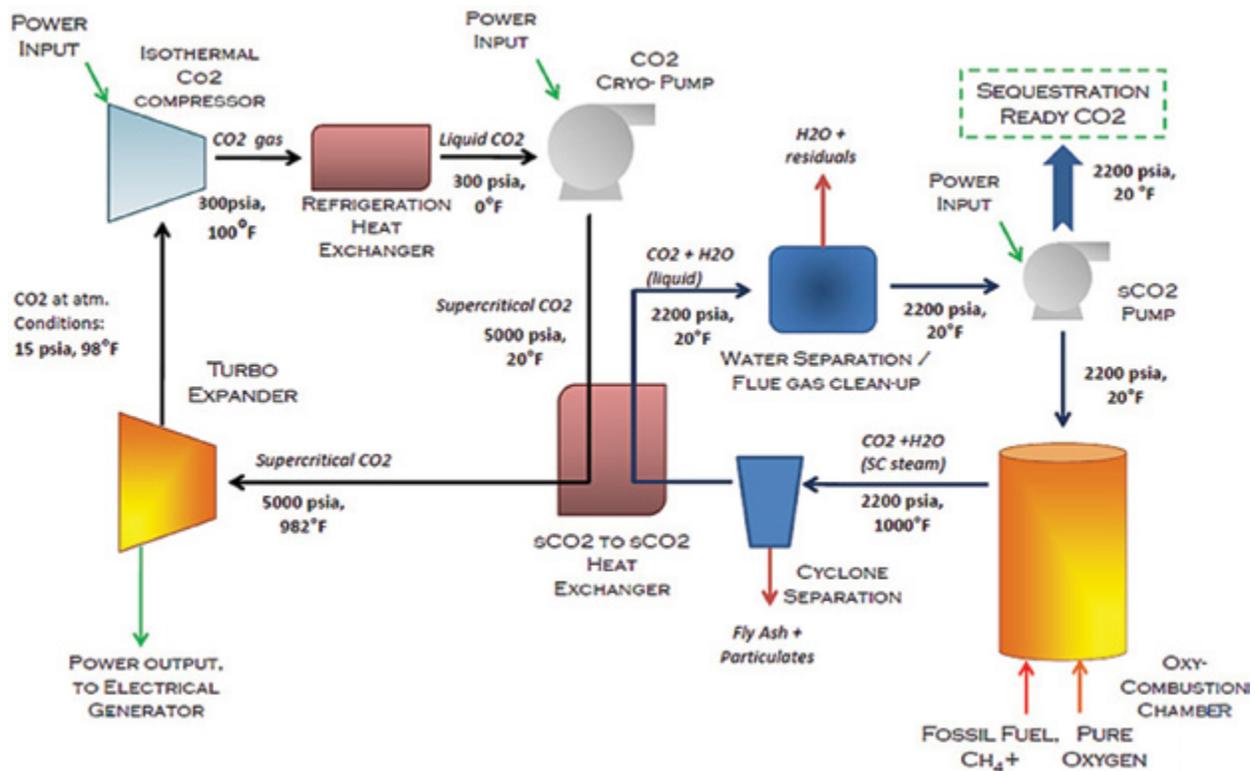


Figure 1: Dual Loop Cryogenic Pressurized Oxy-Combustion Cycle Concept

## technology advantages

- High plant efficiencies with inherent CO<sub>2</sub> capture.
- Increased combustion efficiencies.
- Reduced power consumption for CO<sub>2</sub> compression.
- Reduced plant footprint due to compact cycle components and turbomachinery.

## R&D challenges

- Limited data for coal combustion at supercritical CO<sub>2</sub> pressures.
- CO<sub>2</sub> turbomachinery still in development.
- Flue gas cleanup must be performed at pressure.

## results to date/accomplishments

- Evaluation of candidate cycles and selection of an indirect-fired supercritical CO<sub>2</sub> recompression cycle with a supercritical oxy-combustion loop.
- Optimization of the recompression supercritical CO<sub>2</sub> cycle to achieve high cycle efficiencies and the supercritical combustion loop to minimize thermal and power losses.
- Laboratory testing of coal combustion properties in supercritical CO<sub>2</sub>.
- Development of a novel supercritical oxy-combustor.

next steps

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B-452

- Demonstrate and document the performance of the supercritical oxy-combustor.
- Evaluate the impact of particle loading on heat exchanger performance.
- Identify materials enabling temperatures above 650°C to take advantage of the increase in supercritical CO<sub>2</sub> closed Brayton cycle efficiencies with increasing temperature.

available reports/technical papers/presentations

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Brun, K., “Novel Supercritical Carbon Dioxide Power Cycle Utilizing Pressurized Oxy-Combustion in Conjunction with Cryogenic Compression,” Kickoff presentation, Pittsburgh, Pennsylvania, October 2012. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/DE-FE0009395-swri-Kickoff.pdf>.

# OPTIMIZATION OF PRESSURIZED OXY-COMBUSTION WITH FLAMELESS REACTOR

## primary project goals

Unity Power Alliance (UPA) will examine a novel pressurized oxy-combustion technology with potential for increasing process efficiency and reducing emissions and costs associated with carbon dioxide (CO<sub>2</sub>) capture and compression.

## technical goals

- Identify the optimum operating pressure, flue gas recycling method, and oxygen purity level for flameless pressurized oxy-combustion to improve overall system efficiency.
- Confirm the fate of sulfur, nitrogen oxides (NO<sub>x</sub>), mercury (Hg), particulate matter, and other toxic pollutants with the goal of substantially reducing or eliminating emissions.
- Confirm that the system design can achieve net water positive conditions and near-zero level discharge.

## technical content

Phase I of the project will encompass two independent research efforts: (1) computer modeling and optimization of pressurized oxy-combustion with a flameless reactor; and (2) design, construction, and testing of a bench-scale flameless reactor.

At the outset of Phase I, upon receiving a basis of modeling from ITEA, the Massachusetts Institute of Technology (MIT) will construct a techno-economic computer model of the proposed pressurized oxy-combustion system. The model will be capable of simulating a range of operating pressures, power cycle configurations, and other process conditions, and will utilize empirical data from a 5-MWth pilot facility operating at 4 bar. MIT will use the model to simulate the performance of the proposed system over a range of operating pressures (up to 70 bar). A high operating temperature will also be assumed, in order to take advantage of an advanced heat recovery steam generator design developed by ENEL/ITEA.

The use of high pressure is expected to reduce the cost of electricity by improving generation efficiency through higher heat transfer rates, recycling of high-temperature condensate, and by reducing capital and operating expenses associated with pressurizing the emitted CO<sub>2</sub>. In addition to the efficiency benefits, the capture of CO<sub>2</sub> at higher pressure would enable nearly 100 percent CO<sub>2</sub> capture and would cause the power cycle to generate more water than it consumes (the system would be “net water positive”). Condensate could be recycled for use in preparation of the coal-slurry feedstock, significantly reducing the water demands of the system.

In contrast to a conventional coal combustion furnace, flameless combustion approximates the conditions of an isothermal chemical reactor in which temperatures remain uniform and constant throughout the reaction process. This highly efficient, carefully controlled reaction promotes complete combustion of coal at high temperatures with negligible generation of airborne particulates or hazardous pollutants. The majority of the pollutants that are normally released in flue gas are instead permanently entrained in a vitrified or glass-like slag.

technology maturity:

Bench-Scale

project focus:

Pressurized Oxy-Combustion with Flameless Reactor

participant:

Unity Power Alliance

project number:

FE0009478

NETL project manager:

Steve Mascaro

steve.mascaro@netl.doe.gov

principal investigator:

August Arrigo

Unity Power Alliance

august.arrigo@unitypoweralliance.com

partners:

MIT

GA Tech

ThermoEnergy

ITEA SpA

performance period:

10/1/12 – 9/30/13

As a result, the use of a flameless reactor mitigates the need for costly and energy-intensive flue gas treatment.

In the United States, UPA will provide construction of a bench-scale flameless reactor unit based on ITEA's engineering specifications. This bench-scale system will be installed and tested at Georgia Tech's Carbon Neutral Energy Solutions (CNES) Laboratory, with the objective of confirming that flameless conditions can be sustained at pressures up to 70 bar using various coal feedstocks. The composition of the flue gas will also be monitored to verify that all major pollutants are entrained in a glass-like byproduct. The combined results of the techno-economic modeling and the bench-scale test will enable UPA to proceed with the construction of a 50-MWth demonstration facility for Phase II.

### technology advantages

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- Proven ability to utilize low-rank coals.
- High CO<sub>2</sub> concentration in flue gas minimizes need for treatment.
  - Vitrified slag eliminates leaching issues leading to a lower cost of disposal when compared to conventional coal combustible residuals.
  - Dramatic reduction in water usage.

### R&D challenges

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- Vitrification process may not completely entrain all pollutant emissions.

### results to date/accomplishments

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- Completion of the final design package of the bench-scale reactor, including process flow diagrams, logic control, and engineering and qualified vendor lists such that procurement of the equipment required for testing the bench-scale reactor.
- Bench-scale reactor fabrication initiated.
- Procurement of long lead components and the initiation and procurement of all other bench-scale reactor components.

### next steps

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- Complete procurement and fabrication of the bench-scale reactor unit.
- Prepare the Georgia Tech facility to accept the pilot and ship all supporting equipment.
- Ship the bench-scale reactor and have it rigged into place, connecting all utilities and appurtenances, and begin preliminary testing and reporting.
- Perform pilot testing at the Georgia Tech facility.

### available reports/technical papers/presentations

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N/A.

# STAGED, HIGH-PRESSURE OXY-COMBUSTION TECHNOLOGY: DEVELOPMENT AND SCALE-UP

B-455

OXY-COMBUSTION

## primary project goals

Washington University proposes to evaluate the technical feasibility and improved economics of a unique pressurized oxy-combustion system that incorporates a fuel-staged combustion approach. By staging the combustion, the temperature and heat transfer can be controlled while minimizing recycle.

## technical goals

- The goal of this one-year project is to perform a techno-economic analysis of a staged, high-pressure oxy-combustion process for producing steam for power plants designed for carbon capture, utilization, and storage (CCUS).
- The project is expected to demonstrate that staged pressurized oxy-combustion is a viable method of capturing, utilizing, and sequestering carbon dioxide (CO<sub>2</sub>) and will achieve at least 90 percent CO<sub>2</sub> removal at no more than a 35 percent increase in cost of electricity (COE).

## technical content

Staged combustion is an advanced oxy-combustion approach that holds promise to significantly reduce the cost of CO<sub>2</sub> capture from coal-fired power plants. By staging the combustion, the temperature and heat transfer can be controlled. This allows for the elimination of other temperature control processes, such as flue gas recycle or water/steam injection. The potential benefits of this process are higher efficiency, reduced process gas volume, increased radiative heat transfer, reduced oxygen demands, reduced capital equipment costs, increased CO<sub>2</sub> purity entering the carbon compression and purification unit, and reduced auxiliary power demands.

## technology advantages

- Increased efficiency.
- Reduced oxygen demands.
- Increased CO<sub>2</sub> purity entering the carbon compression and purification unit.
- Reduced equipment and operation and maintenance costs.

## R&D challenges

- Some phenomena, such as radiant heat transfer, nitrogen oxide (NO<sub>x</sub>) formation, sulfur partitioning, acid condensation, and metal corrosion rates, have not been measured in high-pressure, low recycle environments.
- The accuracy of the computational fluid dynamics (CFD) and ASPEN Plus models are limited, leading to uncertainties.

technology maturity:

Bench-Scale

project focus:

Staged, High-Pressure Oxy-Combustion

participant:

Washington University

project number:

FE0009702

NETL project manager:

Bruce Lani

bruce.lani@netl.doe.gov

principal investigator:

Richard Axelbaum

Washington University

axelbaum@wustl.edu

partners:

Burns & McDonnell Engineering Company, Inc  
EPRI

performance period:

10/1/12 – 9/30/13

results to date/accomplishments

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B-456

- Completed ASPEN Plus process simulation of a 500 MWe power plant with staged, pressurized oxy-combustion technology.
- Completed conceptual design of module radiant boilers for staged combustion process using CFD modeling.

next steps

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- Complete the economic analysis and determine the COE for the base system (supercritical).
- Determine optimal system pressure.
- Complete the economic analysis when incorporating the advanced, ultra-supercritical steam cycle.

available reports/technical papers/presentations

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Axelbaum, R.; Kumfer, B.; and Phillips, J., “Staged, Pressurized Oxy Combustion (SPOC),” presented at NETL Kickoff Meeting, Morgantown, West Virginia, October 2012. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/project9702-kickoff-10-24-12.pdf>.

# CLEAN AND SECURE ENERGY FROM COAL

B-457

OXY-COMBUSTION

## primary project goals

Through the tightly coupled simulation and experimental designs, the University of Utah will develop computer simulation tools for oxy-coal combustion and chemical looping combustion (CLC), which predict performance with quantified uncertainty.

## technical goals

- **Oxy-Coal Combustion:** Produce predictive capability with quantified uncertainty bounds for pilot-scale, single-burner, oxy-coal operation. The predictive tool developed under this effort will form the basis for application to full-scale, industrial burner operation.
- **Chemical Looping Combustion:** To develop a new carbon capture technology for coal through CLC and to transfer this technology to industry through a numerical simulation tool with quantified uncertainty bounds.

## technical content

The University of Utah, via their Institute for Clean and Secure Energy (ICSE), is pursuing research to utilize the vast energy stored in domestic coal resources, doing so in a manner that will capture carbon dioxide (CO<sub>2</sub>) from combustion from stationary power generation. The research is organized around the theme of validation and uncertainty quantification through tightly coupled simulation and experimental designs. The results of the research will be embodied in the computer simulation tools that predict performance with quantified uncertainty; thus transferring the results of the research to practitioners to predict the effect of energy alternatives using these technologies for their specific future application. Over-arching project objectives are focused in three research areas and include clean coal utilization for power generation “retrofit,” secure fuel production by in-situ substitute natural gas production from deep coal seams; and environmental, legal, and policy issues.

Two aspects of the power generation “retrofit” areas are:

**Oxy-Coal Combustion** – The ultimate objective of this task is to produce predictive capability with quantified uncertainty bounds for pilot-scale, single-burner, oxy-coal operation and for a commercial-scale, tangentially fired, oxy-coal boiler. This validation research brings together multi-scale experimental measurements and computer simulations. The predictive tool forms the basis for application to full-scale, industrial burner operations. Particular attention is focused on heat flux, temperature, ignition, coal flame stability, and ash partitioning and ash deposition under pulverized oxy-coal conditions, with a smaller companion effort on application of circulating fluidized beds (CFBs) to oxy-coal combustion conditions. Additional detail on the work being performed follows:

*Oxy-Coal Combustion Large Eddy Simulations* – The University of Utah is expanding its high-performance simulation tools coupled with direct quadrature method of moments (DQMOM) to quantitatively predict oxy-coal conditions. This work is being performed using experimental data from the University of Utah’s 100-kW oxy-fuel combustor (OFC) and Alstom’s Boiler Simulation Facility (BSF) tangentially fired boiler. The OFC studies have focused on identifying mechanisms important to flame stability, and the OFC and BSF studies will focus on predicting temperature and heat flux.

technology maturity:

Laboratory-Scale

project focus:

Simulation and Modeling for Oxy-Combustion and Chemical Looping

participant:

University of Utah

project number:

FE0005015

NETL project manager:

David Lang  
david.lang@netl.doe.gov

principal investigator:

Phillip Smith  
University of Utah  
philip.smith@utah.edu

partners:

Brigham Young University

performance period:

9/10/08 – 8/31/13

*Near-Field Aerodynamics, Temperature, and Heat Flux in Oxy-Coal Flames* – Under this project, the University of Utah has built a 100-kW OFC and has studied the effect of various operating parameters on flame stability under oxy-combustion conditions. The reactor is equipped with quartz and sapphire windows optical access, heated or cooled walls, flue gas recycle, and various burners for studying coaxial, zero-swirl conditions and directed oxygen. The entire system is controlled by Opto22 commercial software with automated data logging of temperature at multiple locations, pressure, and gas concentrations. Using the OFC, the University of Utah is applying advanced diagnostics to investigate temperature and heat flux, both in the burner near field and in the far field, under oxy-fired conditions. The investigators are continuing to work closely with the simulation and advanced diagnostics group to develop an understanding of the most important mechanisms governing both flame stability and resultant heat flux.

*Advanced Diagnostics for Oxy-Coal Combustion* – The advanced diagnostics team developed the capability for applying particle image velocimetry, particle shadow velocimetry, and high-speed visible-range and infrared photo imaging to laboratory coal burners and is applying these to the 100-kW OFC. They have also developed accompanying image-processing techniques to provide simultaneous 2D velocity maps with corresponding temperature and heat flux information based on emissions in the visible and infrared ranges for oxy-coal flames under various combustion conditions.

*Oxy-Coal Combustion in Circulating Fluidized Beds* – This work is aimed at developing an understanding of the process dynamics and the impact of key process variables on bed temperature, bed agglomeration, solids recycle rate, and sulfur capture. The recently modified, oxy-fired, pilot-scale CFB is being used to study operational impacts of variations in oxygen concentration, in-bed heat removal, and external heat removal (from the solids recycle stream). In addition, the formation of  $\text{SO}_3$  in the high  $\text{CO}_2$  and  $\text{O}_2$  environment of the CFB is being evaluated to develop an understanding of its potential for sulfuric acid condensation and corrosion.

*Single-Particle Oxy- $\text{CO}_2$  Combustion* – This subtask will focus on both pulverized coal and fluidized-bed systems, with two objectives: (1) for pulverized coal systems, single-particle kinetics for char oxidation under oxy- $\text{CO}_2$  combustion conditions are being developed using literature data in conjunction with experiments performed at Sandia National Laboratories; (2) for fluidized bed conditions, the impact of an  $\text{O}_2/\text{CO}_2$  environment on carbon, nitrogen, and sulfur release from coal and coal char is being explored in a bench-scale, single-particle, fluidized-bed reactor. These experimental results will help refine the first-generation, single-particle reaction developed in the previous project phase. Measurements focus on both rate determination with detailed uncertainty quantification for use in model development, as well as the identification of the influence on major operating variables such as oxygen concentration and bed temperature on the release of nitrogen and sulfur impurities.

*Ash Partitioning Mechanisms for Oxy-Coal Combustion* – The University of Utah is focused on understanding ash partitioning under oxy-coal combustion conditions by considering experiments on two scales: (1) at the bench-scale using a drop tube reactor with coal flow rates of  $\approx 1\text{--}4$  g/h, to determine fundamental ash partitioning mechanisms using simulated flue gases; (2) at 100 kW, self-sustained combustor scale (coal flow rates of  $\approx 10$  kg/h) where the focus is on how the amount of recycled flue gas affects ash partitioning mechanisms. In both experiments, ash aerosol particle size distributions and size-segregated ash compositions are obtained using aerosol mobility and impaction techniques. To date, investigators have studied effects of coal type and a wide range of oxygen concentrations and flue gas recirculation rates on ash partitioning behavior.

**Chemical Looping Combustion** – The ultimate objectives of this task are to develop a new, low-cost carbon capture technology for coal through CLC and to transfer this technology to industry through a numerical simulation tool with quantified uncertainty bounds. The research will primarily focus on  $\text{CuO}/\text{Cu}_2\text{O}$ , but also include iron-based carriers for some of the modeling studies using data from the literature for kinetics and verification. It will also include chemical looping with gasification products. The specific research targets for these tasks are to quantitatively identify reaction mechanisms and rates, explore operating options with a laboratory-scale bubbling bed reactor, identify process-modeling economics, and demonstrate and validate simulation tools for a pilot-scale fluidized bed as described below.

*CLC Kinetics* – The CLC team performed thermogravimetric analysis (TGA) experiments to refine the chemical kinetics of the copper metal/copper oxides CLC oxygen carrier system developed in Phase II. The goal is to extract reaction kinetics for the process modeling studies. The investigators also developed supported copper materials for evaluation in the laboratory-scale studies.

*Laboratory-Scale CLC Studies* – The CLC team is studying the performance of oxygen carriers in a TGA and fluidized bed-based system, focusing specifically on the high-temperature regime where oxygen uncoupling (CLOU) occurs. The studies range from the characterization of fundamental chemical behavior of copper carriers to more industrially relevant aspects, including long-term performance (capacity, attrition, kinetics and avoidance of agglomeration) over many oxidation and reduction cycles. The investigators also evaluated various supports, including titania and alumina, and CuO loadings to 50 wt%. Based on experience with the single-bed, lab-scale reactor, a design for a larger 10-kWth, dual-bed, continuous flow bench-scale CLOU CLC system process development unit is being developed.

*High-Performance Computing (HPC), Computational Fluid Dynamics (CFD) Simulation of a Pilot-Scale Fluidized Bed* – The University of Utah is expanding their models and high-performance simulation tools to dense-particle regimes. Their current work is focused on fluidized-bed CLC process applications with no reaction using literature data. Once the fluidized-bed simulation capabilities are developed, the investigators will perform a residence-time study and a sensitivity analysis of the numerical parameters that feed the HPC CFD simulations, including different turbulence models, and model parameters, including different particle size distributions.

*Process Modeling and Economics* – The investigators are developing process models of the CLOU process, using the ASPEN models developed in Phase II, as well as the kinetics developed in the thermogravimetric studies and laboratory-scale tests. The preliminary mass and energy balance ASPEN modules developed in Phase II have been replaced by more sophisticated expressions of kinetics for given residence times and reactor sizes. The ultimate objective of this subtask is to develop the models such that reactor size, various carrier options, material use, and other economic factors can be minimized, and CLOU can be compared to CLC. For the CLC case, existing kinetic data will be used for iron-based carriers.

### technology advantages

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- The entire project will provide experimental results and simulation tools to aid in the development, production, and utilization of coal for power generation in a carbon-constrained world. The strength of the tool-set is the inclusion and quantification of uncertainty associated with the individual options in order to better enable informed energy futures decisions.
- If the oxy-coal project is successful, it will provide enabling technologies that promote the deployment of oxy-coal combustion as a cost-effective carbon capture technology for retrofit power applications. The predictive tool will form the basis for application to full-scale, industrial-burner operations. The experimental and simulation work will improve the understanding of critical operating parameters that effect performance, safety, and environmental impacts.
- If successful, the CLC project will lead to the development of a new, low-cost, carbon capture technology and accelerate industry adoption of the technology through a numerical simulation tool with quantified uncertainty bounds. This project will contribute to the knowledge base for CLC and facilitate its deployment.

### R&D challenges

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- Quantity and quality of available data may inhibit the thoroughness of the analysis.
- Failure of submodels and larger simulations to represent the complex physical and chemical processes associated with energy generation.
- Extensive computation and storage resources necessary to complete the verification and validation quantification analysis.

## results to date/accomplishments

B-460

OXY-COMBUSTION

- Completed a V/UQ study of flame standoff distance in an oxy-coal furnace and identified important factors determining flame stability.
- Demonstrated high-speed camera and particle-shadow velocimetry techniques for identifying particle shape, size, and velocity in a 100-kW, oxy-coal flame.
- Identified the importance of gasification reactions on coal char burning rates under oxy-fired conditions.
- Developed a submodel for gasification and oxidation of coal under oxy-fuel conditions.
- Developed a better understanding of the effect of oxy-combustion and consequently high CO<sub>2</sub> concentrations in flue gas on the formation of ultrafine (below 0.1 μm) particulate matter.
- Identified that the extent of flue gas recycle under oxy-coal conditions can significantly reduce soot mass emissions.
- Completed a copper attrition study in a laboratory-scale reactor.
- Completed an evaluation of CLOU vs. CLC.
- Developed and tested supported copper oxide materials.

## next steps

- Data analysis and integration.
- Preparation of topical reports.
- Preparation of peer-reviewed publications.
- Perform baseline simulations of the Alstom BSF on the U.S. Department of Energy's Titan supercomputer.
- Perform a consistency analysis for the OFC and BSF simulation and experimental results.

## available reports/technical papers/presentations

Morris, W.J.; Yu, D.; and Wendt, J.O.L., "A comparison of soot, fine particle and sodium emissions for air-and oxy-coal flames, with recycled flue gases of various compositions," *Proceedings of the Combustion Institute*. Volume 34, Issue 2, 2013, Pages 3453–3461. <http://www.sciencedirect.com/science/article/pii/S1540748912002283>.

Pedel, J.; Thornock, J. N.; and Smith, P.J., "Ignition of co-axial turbulent diffusion oxy-coal jet flames: experiments and simulations collaboration," *Journal of Computational Physics*, Volume 160, June 2013, 1112-1128.

Lighty, J.S., et al, "Chemical Looping Combustion and Chemical Looping with Oxygen Uncoupling for Solid Fuels," presented at the 245<sup>th</sup> ACS National Meeting, Symposium on CO<sub>2</sub> Capture, Sequestration, Conversion and Utilization, New Orleans LA, April 2013.

Sahir, A., et.al, "Rate analysis relevant to chemical-looping with oxygen uncoupling (CLOU) for solid fuels," presented at the 2012 AIChE Annual Meeting, Pittsburgh, Pennsylvania, November 2012.

Sahir, A.; Lighty, J. S.; and Bigelow, L., "The role of open innovation in development of futuristic technologies for carbon capture in coal-fired power plants: an academic perspective," presented at the 2012 AIChE Annual Meeting, Pittsburgh, Pennsylvania, November 2012.

Hecht, E. S., et.al, "Effect of CO<sub>2</sub> and steam gasification reactions on the oxy-combustion of pulverized coal char," Volume 159, Issue 11, November 2012, Pages 3437–3447. <http://repository.icse.utah.edu/dspace/handle/123456789/11236>.

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# OXY-FUEL TURBOMACHINERY DEVELOPMENT FOR ENERGY-INTENSIVE INDUSTRIAL APPLICATIONS

B-462

OXY-COMBUSTION

## primary project goals

Clean Energy Systems (CES) set out to design and develop a pre-commercial oxy-fuel (O-F) combustor, able to utilize synthesis gas (syngas), for use in a power generation cycle able to achieve high thermal efficiency with near-zero atmospheric emissions, including carbon dioxide (CO<sub>2</sub>). The technology is a high-pressure O-F combustor that produces a steam/CO<sub>2</sub> working fluid for expansion in a turbine. The high pressure O-F combustor utilizes syngas and pure oxygen to produce a working fluid comprised of steam and CO<sub>2</sub> that is used to power turbo generators.

CES also set out to design and develop a commercial-scale O-F turbine (OFT) for deployment in industrial O-F plants that capture >99 percent of the produced CO<sub>2</sub>, at competitive cycle efficiency and cost-of-electricity (COE), using diverse fuels, including gasified petcoke/coal, gasified or liquefied renewable fuels, and natural gas (NG).

## technical goals

- Develop a detailed design of a 100 megawatt thermal (MWth) oxy-syngas combustor.
- Integrate, commission, and test a first-generation O-F intermediate pressure turbine (IPT) powered by an O-F combustor using NG.
- Design, manufacture, and test an O-F reheat (RH) combustor in a dedicated test rig.
- Develop a detailed design to modify an existing gas turbine to a commercial-scale, second-generation OFT.
- Identify, purchase, disassemble, and inspect a candidate gas turbine for conversion to an OFT.
- Repair/refurbish, manufacture, and assemble new OFT.
- Install, commission, and test commercial-scale OFT.

## technical content

The CES cycle involves burning high purity oxygen (O<sub>2</sub>) with a gaseous carbonaceous fuel (NG, coal syngas, gasified biomass, etc.) in the presence of water to generate a high-pressure, high-temperature drive gas comprised of approximately 90%v steam and 10%v CO<sub>2</sub> when using NG (75%v steam and 25%v CO<sub>2</sub> when using syngas). The drive gas is controlled to temperatures ranging from 315 to 1,760°C (600 to 3,200°F) and expanded through steam or aero-derivative turbines to produce electricity. In order to achieve high plant efficiencies, the OFT is equipped with a set of RH combustors used to attain greater turbine inlet temperatures (TITs).

CES has prepared the detailed design for a pre-commercial oxy-syngas combustor (100 MWth). The heart of the combustor is the main injector where intricate channels control precise quantities of O<sub>2</sub>, fuel, and water entering the main combustion chamber. After stoichiometric combustion, water is introduced in stages to cool the working fluid to the

### technology maturity:

100-MWth Commercial-Scale  
Oxy-Fuel Combustor and Turbine

### project focus:

Oxy-Syngas Combustor and  
Turbine

### participant:

Clean Energy Systems

### project number:

FC26-05NT42645

### NETL project manager:

Travis Shultz  
travis.shultz@netl.doe.gov

### principal investigator:

Rebecca Hollis  
Clean Energy Systems, Inc.  
rhollis@cleanenergysystems.com

### partners:

Florida Turbine Technologies  
Siemens Energy

### performance period:

10/1/05 – 3/31/13

desired TIT. Figure 1 is a photograph of an oxy-NG combustor (similar to an oxy-syngas combustor). The main injector assembly is shown on the left-hand side, followed by staged water injection, or cooling.



Figure 1: CES OF Combustor Assembly

Cycle studies have shown that efficiencies increase with increased turbine operating temperatures. To accelerate the development and deployment of advanced, high-temperature OFTs, CES proposed modifying existing commercial gas turbines, as these routinely operate with higher TITs than traditional steam turbines. The concept was proven when CES successfully modified and demonstrated an O-F J79 turbine at the Kimberlina test facility (Figure 2). Several of the traditional gas turbine components had to be modified or removed completely to convert the engine to a functional OFT. For example, because the O-F combustor can deliver a high-temperature, high-pressure drive gas, both the compressor and combustor sections of the J79 were removed and replaced with a new steam/CO<sub>2</sub> inlet.



Figure 2: OFTJ79 and Electric Generator

To further plant efficiencies, CES developed an RH combustion system to boost TITs to the maximum allowable by the gas turbine's sophisticated materials and cooling schemes. A first-generation O-F RH was designed with development partners Florida Turbine Technologies (FTT) to replace the existing combustor cans of the J79 engine. A test article was manufactured and demonstrated in a dedicated rig at CES's Kimberlina test facility in Bakersfield, California. Although the unit met or exceeded criteria such as exhaust temperature, pressure drop, and flame out resistance, the unit fell short in areas related to uniform mixing.

The next phase of the program focused on the development of an advanced, second-generation, commercial-scale OFT that could be deployed in industrial O-F plants capable of capturing greater than 99 percent of all produced emissions, including CO<sub>2</sub>. Again, the team chose to utilize an existing turbine, already capable of elevated operating temperatures, to modify for the steam/CO<sub>2</sub> working environment, as this approach is much faster and much less expensive than building a new design from scratch. The Siemens SGT-900 engine was selected as it most closely resembled the desired operating parameters, such as temperature, pressure, and power output. Along with FTT, Siemens Energy was a development partner on the program, offering unmatched expertise and insight into the engine.

A used SGT-900 B12 Econopac was located and purchased by CES and delivered to a Siemens team in Houston, Texas, for disassembly and inspection. During its lifetime, the unit had been routinely serviced and was found to be in good condition. Reused components were repaired, refurbished, or replaced in preparation for reassembly into the new high-power OFT.

The detailed engineering design to convert the SGT-900 into an OFT was completed by FTT in early 2011. Again, components such as the engine's compressor were to be removed and replaced with a unique thrust balancing system. Siemens led the effort to manufacture any necessary new components, as well as work to reassemble the first-of-its-kind engine.

The new, fully assembled OFT-900 was delivered to the CES test facility in the fall of 2012. A considerable effort was devoted to upgrade the test facility to support commissioning and test of the large-scale engine. This involved increasing the capacities of many of the site's balance of plant (BOP) systems, such as high-pressure NG and O<sub>2</sub> supplies, feed and cooling water systems, and electrical energy dissipation. By the close of the year, the engine was set, anchored, and connected to all BOP equipment and systems. Figure 3 is a picture of the OFT-900 during installation at the Kimberlina test facility.



Figure 3: OFT-900 and Generator During Installation at Kimberlina Test Facility

The successful demonstration of the OFT-900 was completed in March 2013. Testing consisted of both main and engine sub-system checkouts. Critical turbine support systems, such as lube oil, steam feed, and steam cooling, were commissioned prior to powering the OFT-900 with steam/CO<sub>2</sub> from the CES O-F combustor. Numerous tests of the industrial-scale OFT included turbine ramp, deceleration, and synchronization, as well as verification of normal and emergency shutdowns.

On a parallel path, CES designed, manufactured, and tested an RH combustion system to be integrated into the OFT-900. A single-can combustor (one of eight to be installed in the engine) was fabricated and tested in a rig designed to simulate the flow conditions of the OFT-900. Building upon knowledge gained from the first-generation RH development, the second-generation

RH utilized a fuel and O<sub>2</sub> injector, similar to that of the main O-F combustor. Rig tests ramped the unit from low- to full-power output while maintaining desired exhaust temperatures and low-pressure losses.

### technology advantages

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*O-F Combustor:* Can burn a variety of fossil and renewable fuels to produce a drive gas consisting of steam and readily separable CO<sub>2</sub>. This process (the CES cycle) yields a base-load power cycle that captures virtually 100 percent the CO<sub>2</sub> emissions through condensation of the steam from the effluent. In addition, the high-temperature drive gas produced by the O-F combustor enables higher-efficiency power cycles when coupled with high-temperature turbines.

*High-Temperature Turbine:* The capabilities of advanced, high-temperature turbines (up to 1,204°C [2,200°F]) permits plant heat rate improvements of 25 percent compared to conventional steam turbines. By utilizing modified gas turbines as O-F IPTs, development schedules and costs are significantly reduced, allowing accelerated deployment of high-temperature, O-F compatible steam turbo-generators to the market.

### R&D challenges

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*High Temperatures:* Elevated temperatures subject turbine equipment to increased thermal stress, potentially increasing maintenance/reducing life cycles.

*Steam/CO<sub>2</sub> Drive Gas:* Condensate from steam/CO<sub>2</sub> drive gases is acidic (carbonic acid), requiring the use of costlier, corrosion-resistant materials. Also, steam/CO<sub>2</sub> transfers heat to metal surfaces more readily than the drive gases from air-fuel combustion, requiring provision for more effective cooling. Use of steam and/or CO<sub>2</sub> as a cooling medium instead of air may be cost effective.

### results to date/accomplishments

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- Completed the detailed design of a 100-MWth oxy-syngas combustor and its enclosure.
- Completed integrated testing of a 33-megawatt electric (MWe) OFT (modified J79 engine) and oxy-NG combustor.
- Designed, fabricated, and demonstrated a first-generation RH combustor, suitable for integration into an OFT-J79, in a dedicated test rig.
- Completed the detailed engineering design to convert an SGT-900 gas turbine into a 150-MWe OFT.
- Located, purchased, disassembled, and inspected a used SGT-900 B12 power plant.
- Refurbished, repaired, and replaced existing engine components to be reinstalled into a new OFT-900 engine. Manufactured new components required to convert an existing SGT-900 to an industrial-scale OFT. Assembled new OFT-900 engine and delivered to CES test facility.
- Completed the install, commissioning, and test of a commercial-scale OFT-900.
- Upgraded test facility to support commercial-scale OFT demonstration testing.
- Designed, fabricated, and demonstrated a second-generation RH combustor, suitable for integration into an OFT-900, in a dedicated test rig.

### next steps

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This project ended on March 31, 2013.

B-466

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# JUPITER OXY-COMBUSTION AND INTEGRATED POLLUTANT REMOVAL RESEARCH AND DEVELOPMENT TEST FACILITY

## primary project goals

Jupiter Oxygen set out to design, construct, and operate a 5-MWe, high flame temperature, oxy-combustion test facility with a 20-kWe integrated pollutant removal (IPR) bench-scale system to demonstrate carbon dioxide (CO<sub>2</sub>) capture from a high flame temperature oxy-combustion process.

## technical goals

- Develop high flame temperature oxy-fuel burners.
- Retrofit an existing boiler with high flame temperature oxy-combustion, and operate with coal and natural gas without altering interior boiler materials.
- Collect data on burner performance and boiler heat transfer.
- Conduct a study of the ash and slagging characteristics of the process and its impact on boiler materials.
- Capture CO<sub>2</sub> and collect data on impurity removal using the Jupiter Oxygen combustion process, along with IPR technology developed by NETL.
- Evaluate the high flame temperature approach with respect to capital and operating costs.
- Design, build, and test a new 1-MWth module boiler design for high flame temperature oxy-combustion.

## technical content

There are two different approaches to oxy-combustion. Jupiter's approach was to use a high-temperature flame that is minimally tempered with CO<sub>2</sub> (the only tempering occurs as a result of flue gas recycle that is used to motivate coal). High flame temperature oxy-combustion results in improved heat transfer in the boiler's radiant zone. Other oxy-combustion facilities use a low flame temperature approach, which uses large amounts of CO<sub>2</sub>/H<sub>2</sub>O recycled through or at the burner to cool the flame to a temperature similar to air firing. The unique combination of the high-temperature approach coupled with the IPR system allowed the evaluation of the impact of using high- and low-temperature approaches and energy recovery on a variety of aspects of power plant operations.

Coal analyses, such as heating value, mineralogy, and trace element content; proximate; and ultimate analyses were determined using ASTM procedures. This information was used to determine the effect of the coal characteristics on oxy-combustion performance and the effectiveness of emissions capture. Other performance measurements for the test facility included water tube and web temperature, heat transfer rate, flue gas emissions (nitrogen oxides [NO<sub>x</sub>], carbon monoxide [CO], CO<sub>2</sub>, sulfur dioxide [SO<sub>2</sub>], and trace metals), and loss on ignition (LOI) of the ash. The facility incorporated the following approaches to conduct measurements:

technology maturity:

Pilot-Scale

project focus:

Oxy-Combustion and IPR

participant:

Jupiter Oxygen Corporation

project number:

FC26-06NT42811

NETL project manager:

Timothy Fout

timothy.fout@netl.doe.gov

principal investigator:

Mark Schoenfield

Jupiter Oxygen Corporation

m\_schoenfield@jupiteroxygen.com

partners:

Reaction Engineering

Maxon Corporation

Michigan State University

Purdue University

University of Wyoming

performance period:

9/28/06 – 9/30/12

- Flue gas species concentrations measured by Fourier Transform Infrared Spectroscopy (FTIR).
- Ash LOI measured by laboratory testing.
- Heat transfer in the radiant zone determined by computational fluid dynamics (CFD) modeling, spectral flame mapping, furnace gas temperature measurement (at the screen wall and boiler exit), temperature measurements of the flux through the boiler tubes, and optical measurements of the total radiant heat flux from the flame.
- Flame shape and transient behavior were to be evaluated by high-speed video.
- Net heat output from the burner and heat absorbed by the boiler calculated based on combustion and steam-side energy balances.
- Combustion-side mass balances calculated by combining species measurements with mass flows.
- Corrosion monitoring probes used.
- Gas-phase and particulate-phase trace elements, including mercury (Hg), measured in samples from select runs.
- IPR contaminant removal measured by laboratory analyses and FTIR.

The IPR system was added to the pilot facility to remove pollutants from the oxy-combustion flue gas re-circulated stream. The device was used to process 45 kg/hr (100 lb/hr) of flue gas from the facility. The IPR system captured, separated, and produced a dry, supercritical stream of CO<sub>2</sub>; a stream of captured pollutants; and a stream of condensed water from the flue gas. Figure 1 shows a representation of major components of a typical IPR system.

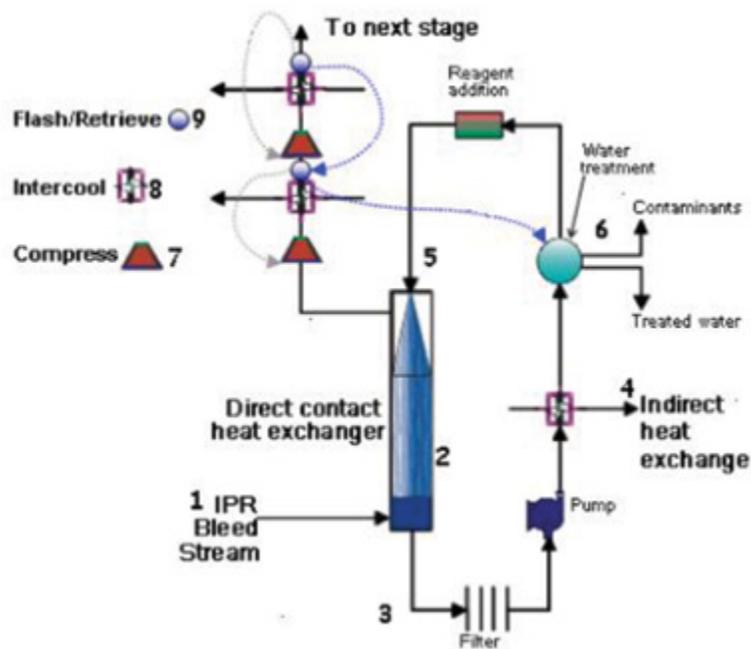


Figure 1: Representation of IPR System Integrated at a Pilot-Scale Facility

The generalized IPR process flow diagram is presented above: a flue gas bleed stream (1) enters the IPR system into a spray tower (2) in which sulfur oxide (SO<sub>x</sub>) is removed by a spray stream (reagent addition) as the gas rises. The spray water and combustion condensed water (3) are cooled (4) and partially re-circulated back into the spray tower (5). The water that is not re-circulated back in to the spray tower is treated outside the IPR system (6). As the scrubbed gas leaves the tower, it enters a two-stage reciprocal compressor (7) and a water-cooled, counter flow heat exchanger (8). During the compression stage, separated water can be collected in the collection vessel (9).

## technology advantages

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- The higher flame temperature improves heat transfer in the radiant zone, which increases boiler efficiency. Oxy-fuel combustion lowers the quantity of flue gas, concentrates CO<sub>2</sub> in the flue gas, and significantly reduces NO<sub>x</sub> emissions.
- For retrofit applications, this technology maintains actual water wall and steam temperatures without altering the boiler design or size.
- For new construction, this technology can use a smaller boiler, which provides the same thermal output as larger, existing power plant boilers.

## R&D challenges

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Design, build, and test a new, 1-MWth module boiler design for high flame temperature oxy-combustion.

## results to date/accomplishments

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- Completed high flame temperature oxy-combustion burner test runs.
- Completed air-coal burner test runs.
- Retrofitted and operated a 5-MWe equivalent air-fired boiler as an oxy-coal combustion test facility without major boiler modifications. The test facility was operated along with ancillary systems, including oxygen production and generation of appropriate steam.
- Performed a series of high flame temperature oxy-coal burner development tests that resulted in modified generation burners.
- Performed parametric studies with the modified generation oxy-coal burners.
- No increased fouling, slagging, or damage to boiler materials indicated.
- Developed a CFD model of the modified generation burners.
- Designed, constructed, and operated a 20-kWe equivalent IPR facility.
  - Demonstrated CO<sub>2</sub> capture at 95 to 100 percent.
  - 95 percent NO<sub>x</sub>, SO<sub>x</sub>, and particulate removal; 60 to 90 percent Hg removal.
  - FeCl<sub>3</sub>/polymer pairing found to be effective flue gas condensate flocculent.
- Full-scale model of a power plant retrofitted with high-temperature oxy-combustion and an IPR system was developed with an economic evaluation.

## next steps

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This project ended on September 30, 2012.

B-470

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# NEAR-ZERO EMISSIONS OXY-COMBUSTION FLUE GAS PURIFICATION

B-471

OXY-COMBUSTION

## primary project goals

Praxair set out to develop a near-zero emission flue gas purification technology for a retrofitted existing coal-fired power plant with oxy-combustion technology

## technical goals

- Design a contaminant-removal system that will produce saleable sulfuric acid and nitric acid without the need for flue gas desulfurization (FGD) or selective catalytic reduction (SCR) units.
- Design a second contaminant removal system that will produce gypsum.
- Achieve greater than 95 percent carbon dioxide (CO<sub>2</sub>) capture by incorporating a vacuum pressure swing adsorption (VPSA) unit in an existing plant with a high air ingress, and reduce sulfur oxide (SO<sub>x</sub>) and mercury (Hg) emissions by more than 99 percent and nitrogen oxide (NO<sub>x</sub>) emissions by more than 90 percent (high- and low-sulfur coal).
- Perform a techno-economic study and an operability and integration evaluation to assess the commercial viability of retrofitting an existing power plant with the proposed technology.

## technical content

Two approaches for SO<sub>x</sub>/NO<sub>x</sub>/Hg removal depend on the SO<sub>x</sub> levels in the flue gas. By carrying out these unit operations at high pressure, it is envisioned that capital costs would be reduced while achieving low levels of SO<sub>x</sub> and NO<sub>x</sub> in the CO<sub>2</sub> stream. For plants with existing FGD and SCR, operating cost savings could be realized by shutting down those units while operating the proposed SO<sub>x</sub>/NO<sub>x</sub> removal process. For plants burning low-sulfur coal, there is no need for investment in separate FGD and SCR equipment for producing high-purity CO<sub>2</sub>.

High air ingress in existing plants limits the amount of CO<sub>2</sub> that can be recovered from oxy-combustion flue gas using a cold box alone to <65 percent. The CO<sub>2</sub>-recovery limitation is overcome by using a hybrid process that combines a cold box and VPSA (Figure 1). In the proposed hybrid process, up to 90 percent of CO<sub>2</sub> in the cold box vent stream is recovered by CO<sub>2</sub> VPSA and then recycled and mixed with the flue gas stream upstream of the compressor. The recovery from the process will be >95 percent.

### Pollutant Removal

The high-sulfur coal tests were bench-scale and utilized a single gas/liquid contact column that operates at up to 17 atm (250 pounds per square inch absolute [psia]) and 150°C (300°F) for testing multiple reactions. Nitric oxide (NO) in the flue gas is converted to nitrogen dioxide (NO<sub>2</sub>), which catalyzes sulfur dioxide (SO<sub>2</sub>) oxidation to sulfur trioxide (SO<sub>3</sub>). The hydrolysis of SO<sub>3</sub> and NO<sub>2</sub> forms sulfuric and nitric acids.

### technology maturity:

Bench-Scale, 5 kg of CO<sub>2</sub>/hr

### project focus:

Near-Zero Emission Flue Gas Purification

### participant:

Praxair, Inc. – Tonawanda, NY

### project number:

FC26-08NT0005341

### NETL project manager:

Morgan Mosser  
morgan.mosser@netl.doe.gov

### principal investigator:

Minish Shah  
Praxair, Inc. – Tonawanda, NY  
minish\_shah@praxair.com

### partners:

AES  
Foster Wheeler  
WorleyParsons Canada

### performance period:

10/1/08 – 6/30/12

The low-sulfur coal experiments used a single-column unit (2.5 cm [1 inch] diameter, 3.8 cm [1.5 inch] long), and operate up to 17 atm (250 psia) and 93°C (200°F). Activated carbon is used as an adsorbent/catalyst for the capture of SO<sub>x</sub> and NO<sub>x</sub> from the flue gas. The activated carbon oxidizes the SO<sub>2</sub> to SO<sub>3</sub>, NO to NO<sub>2</sub>, and a periodic water wash will be used to remove the acids formed.

B-472

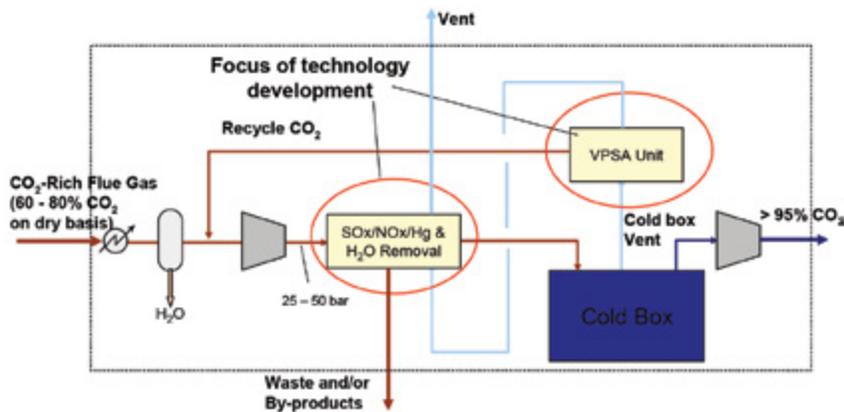


Figure 1: Technology Concept

The chemical reactions for the high- and low-sulfur coal pollutant removal system are summarized below.

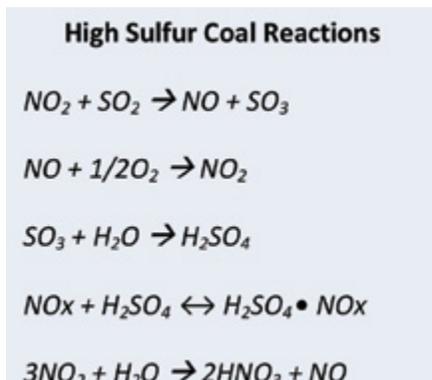


Figure 2: High-Sulfur Coal Pollutant Removal

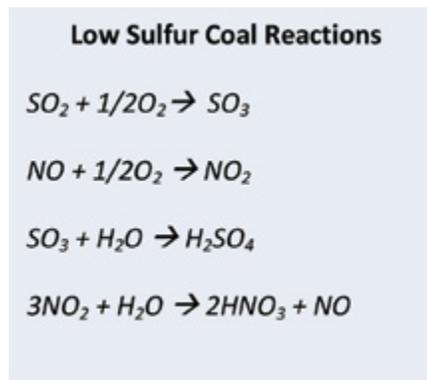


Figure 3: Low-Sulfur Coal Pollutant Removal System

### High CO<sub>2</sub> Recovery Using VPSA

The VPSA unit is a multi-bed unit that performs multiple depressurization/re-pressurizations steps. Oxy-combustion flue gas will enter the CO<sub>2</sub> VPSA from the “cold box” (25 to 35 atm and ambient temperature) that will recover additional CO<sub>2</sub> (produce 80 to 95% CO<sub>2</sub> concentration) and recycle the CO<sub>2</sub> back into the CO<sub>2</sub>-rich flue gas stream at ambient pressure. The flue gas stream not recycled from the VPSA contains mainly oxygen (O<sub>2</sub>), nitrogen (N<sub>2</sub>), and argon (Ar) that will be vented to the atmosphere.

### technology advantages

- Cold box-VPSA hybrid technology achieves >95 percent CO<sub>2</sub> recovery, even for plants with high air ingress.
- The flue gas purification process for high-sulfur coal has lower capital and operating costs than FGD and SCR and it allows for revenue from sale of acids.
- The flue gas purification process for low-sulfur coal will not need investment in expensive FGD and SCR units.

## R&D challenges

- Sulfuric acid process for SO<sub>x</sub>/NO<sub>x</sub>/Hg removal from high-sulfur coal:
  - Reactor materials must be able to withstand the operating conditions in the process.
  - Determine an effective NO<sub>x</sub> catalyst for producing saleable sulfuric acid.
- Activated carbon process for SO<sub>x</sub>/NO<sub>x</sub>/Hg removal from low-sulfur coal:
  - Find activated carbon materials that are effective for regeneration based on sorption capacity and ability to maintain performance.
- Identify adsorbents with a tolerance to residual SO<sub>x</sub>/NO<sub>x</sub> to be used in the VPSA process.
- Determine VPSA cost benefit for recovering additional CO<sub>2</sub>.
- Establish proper modifications required for retrofitting existing plants.

## results to date/accomplishments

Bench-scale experimental test systems have been built and commissioned for all three experimental programs.

- Sulfuric Acid Process:
  - Gas phase NO oxidation kinetics confirmed.
  - Greater than 98 percent NO<sub>x</sub> absorption in one stage.
  - Overall process projected to achieve high SO<sub>x</sub> (>99%) and NO<sub>x</sub> (>90%) removal efficiencies.
  - Sulfuric acid and nitric acid could not be produced to meet commercial grades.
- Activated Carbon Process:
  - Simultaneous SO<sub>x</sub>/NO<sub>x</sub> removal demonstrated – SO<sub>2</sub> >99%, NO<sub>x</sub> >98%.
  - Process was found to be suitable for power plants burning both low- and high-sulfur coals.
  - A 40-day long continuous operation test confirmed the excellent SO<sub>x</sub>/NO<sub>x</sub>-removal efficiencies.
  - Further efforts are needed for maintaining long-term activity of activated carbon.
  - Technology is ready for scale up to 10 to 50 tpd CO<sub>2</sub> range.
- High CO<sub>2</sub> Recovery Using VPSA:
  - Three adsorbents selected based on cost, CO<sub>2</sub> recovery, CO<sub>2</sub> purity, and vacuum pump.
  - Process tested in a pilot unit with 12 beds.
  - Process exceeded CO<sub>2</sub> recovery and purity targets.
  - Overall CO<sub>2</sub> recovery from the cold box-VPSA hybrid process was projected to be >99% for plants with low air ingress (2%) and >97% for plants with high air ingress (10%).
- Economic analysis was performed to assess value of the near-zero emissions CO<sub>2</sub> processing unit (CPU).
  - For greenfield plants, cost of avoided CO<sub>2</sub> and cost of captured CO<sub>2</sub> are generally about 11 to 14% lower using the near-zero emissions CPU compared to using a conventional CPU.
  - For older plants with high air intrusion, the cost of avoided CO<sub>2</sub> and capture CO<sub>2</sub> are about 18 to 24% lower using the near-zero emissions CPU.
  - Lower capture costs for near-zero emissions CPU are due to lower capital investment in FGD/SCR and higher CO<sub>2</sub> capture efficiency.
- Near-zero emissions CPU technology developed based on the activated carbon process and cold box-VPSA hybrid process.
  - Technology is projected to work for both low- and high-sulfur coal plants.
  - The near-zero emissions CPU technology is projected to achieve near-zero stack emissions, produce high-purity CO<sub>2</sub> relatively free of trace impurities, and achieve ≈99% CO<sub>2</sub> capture rate while lowering the CO<sub>2</sub> capture costs.

B-474 This project ended on June 30, 2012.

available reports/technical papers/presentations

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Shah, M. M. et al., “Purification of oxy-combustion flue gas for SO<sub>x</sub>/NO<sub>x</sub> removal and high CO<sub>2</sub> recovery,” presented at the 2<sup>nd</sup> Oxyfuel Combustion Conference, Yeppoon, Australia, September 2011. [http://www.ieaghg.org/docs/General\\_Docs/OCC2/Presentations/OCC2%20-%20Near%20Zero%20Emissions%20CPU%20-%20Sept%202011%20-%20no%20animation.pdf](http://www.ieaghg.org/docs/General_Docs/OCC2/Presentations/OCC2%20-%20Near%20Zero%20Emissions%20CPU%20-%20Sept%202011%20-%20no%20animation.pdf).

Shah, M. M., et.al, “Near-Zero Emissions Oxy-Combustion Flue Gas Purification,” presented at the 2011 NETL CO<sub>2</sub> Capture Technology Meeting, August 2011. <http://www.netl.doe.gov/publications/proceedings/11/co2capture/presentations/3-Wednesday/24Aug11-Shah-Praxair-Near%20Zero%20Emissions%20OxyComb.pdf>.

Shah, M. M., et.al, “Near-Zero Emissions Oxy-Combustion Flue Gas Purification,” presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, September 2010. <http://www.netl.doe.gov/publications/proceedings/10/co2capture/presentations/tuesday/Minish%20Shah%20-%20NT0005341.pdf>.

# OXY-COMBUSTION BOILER MATERIAL DEVELOPMENT

B-475

OXY-COMBUSTION

## primary project goals

Foster Wheeler set out to conduct a laboratory test program to assess the corrosion characteristics of oxy-combustion relative to air-fired combustion for pulverized coal (PC)-fired boilers; identify the corrosion mechanisms involved; and determine oxy-combustion's effects on conventional boiler tube materials, conventional protective coatings, and alternative materials and coatings when operating with high- to low-sulfur coals.

## technical goals

- Conduct computational fluid dynamic (CFD) modeling of air- and oxy-fired PC boilers operating with high-, medium-, and low-sulfur coals to determine the flue gas compositions that will exist throughout these units and especially along the furnace waterwalls where highly corrosive microclimates can exist.
- Conduct corrosion tests using coupons of conventional and advanced boiler tube materials that are coated with deposits representative of low- to high-sulfur coals and exposed to the CFD-predicted oxy-combustion and air-fired flue gases for up to 1,000 hours in electric furnaces using synthesized gases from pressurized cylinders.
- Conduct post-test macroscopic and microscopic analyses of the material coupons to identify corrosion mechanisms, evaluate the corrosiveness of oxy-fired flue gas relative to air-fired flue gas, and identify materials suitable for oxy-combustion.

## technical content

An oxy-combustion boiler retrofit will utilize flue gas recycle to maintain the heat absorption of the original air-fired boiler and limit the combustion temperature. With air nitrogen (N<sub>2</sub>) eliminated, and with the recycle consisting of carbon dioxide (CO<sub>2</sub>) and water vapor (H<sub>2</sub>O), along with corrosive products of combustion, the level of reducing and corrosive gases in the boiler (e.g., carbon monoxide [CO], sulfur dioxide [SO<sub>2</sub>], sulfur trioxide [SO<sub>3</sub>], hydrogen sulfide [H<sub>2</sub>S], and hydrogen chloride [HCl]) will increase and could cause increased corrosion. To assess the corrosiveness of oxy-combustion flue gas, coupons of conventional and advanced waterwall and superheater/reheater materials were exposed to oxy- and air-fired flue gases in electric tube furnaces. Rectangular-shaped coupons, typically 19 mm (3/4 inch) wide by 25 mm (1 inch) high by 3 mm (1/8 inch) thick, will be used to investigate tube materials, tube welds, and tube weld overlays; bullet-shaped coupons, typically 19 mm (3/4 inch) in diameter by 38 mm (1-1/2 inch) high, will investigate thermal spray coatings. The conventional and advanced materials to be tested are listed in Table 1 and Table 2.

technology maturity:

Laboratory-Scale

project focus:

Boiler Materials for Oxy-Combustion

participant:

Foster Wheeler

project number:

NT0005262

NETL project manager:

Timothy Fout  
timothy.fout@netl.doe.gov

principal investigator:

Horst Hack  
Foster Wheeler  
horst\_hack@fwc.com

partners:

None

performance period:

10/1/08 – 1/31/12

TABLE 1: WATERWALL MATERIALS

	Material	Description	Boiler Use	Nominal Composition
1	Tube	SA210-A1	Conventional	0.27% Carbon
2	Tube	SA213-T2	Conventional	1/2 Cr - 1/2 Mo
3	Tube	SA213-T11	Conventional	1-1/4 Cr - 1/2 Mo
4	Weld	T11 to T11	Conventional	1-1/4 Cr - 1/2 Mo
5	Weld Overlay	309L StnStl	Conventional	24 Cr
6	Weld Overlay	Inconel 622	Conventional	21 Cr - 55 Ni
7	Weld Overlay	VDM Alloy 33	Conventional	33 Cr - 31 Ni
8	Thermal Spray	IGS UTEx 5-450	Relatively New	40 Cr - 55 Ni
9	Thermal Spray	IGS UTEx 5-480	Relatively New	25 Cr - 60 Ni
10	Thermal Spray	IGS UTEx 5-500	Relatively New	15 Cr - 80 Fe

TABLE 2: SUPERHEATER/REHEATER MATERIALS

	Material	Description	Boiler Use	Nominal Composition
1	Tube	T22	Conventional	2-1/4 Cr - 1 Mo
2	Tube	304H StnStl	Conventional	18 Cr - 8 Ni
3	Tube	347H StnStl	Conventional	18 Cr - 9 Ni
4	Tube	T91/T92	Newer Boilers	9 Cr
5	Tube	NF709	Newer Boilers	20 Cr - 25 Ni
6	Tube	HR <sub>3</sub> C	Newer Boilers	25 Cr - 20 Ni
7	Weld Overlay	Inconel 622	Conventional	21 Cr - 55 Ni
8	Weld Overlay	VDM Alloy 33	Conventional	33 Cr - 31 Ni
9	Weld Overlay	Inconel 72	Conventional	44 Cr - 55 Ni
10	Welded Coupon	T22 to 304H	Conventional	1-1/4 Cr - 18 Cr

The coupons, mounted in racks and inserted in electric tube test furnaces (see Figure 1), were exposed for 1,000 hours to synthesized air- and oxy-fired flue gases. Foster Wheeler set out to conduct CFD analyses of nominal 500-megawatt electric (MWe) air- and oxy-fired boilers (see Figure 2) to determine the range of flue gas compositions that existed throughout these boilers and especially along their furnace walls where highly corrosive microclimates can exist; based on those ranges, gas compositions were to be selected for the corrosion tests. The test gases were synthesized/blended from gas cylinders and consist of varying concentrations of CO<sub>2</sub>, H<sub>2</sub>O, N<sub>2</sub>, CO, SO<sub>2</sub>, H<sub>2</sub>S, and HCl.

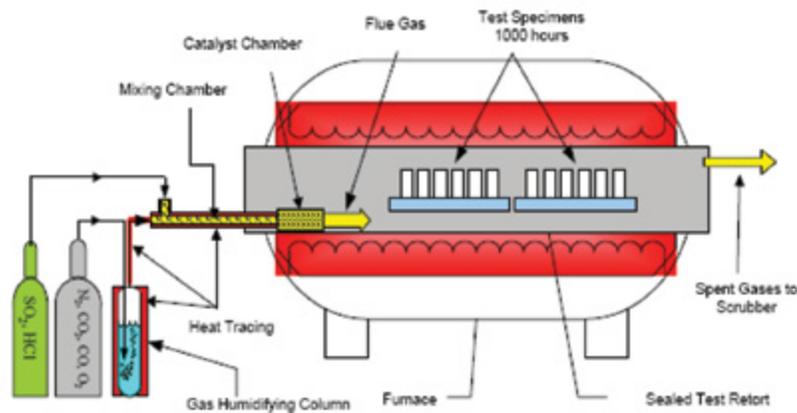


Figure 1: Coupons Mounted in Racks and Inserted in Electric Tube Test Furnaces

The material coupons were coated with three types of synthetic ash deposits representative of high-, medium-, and low-sulfur coals. The deposits were produced from reagent grade powders that are mixed and applied to the coupons as a paste. The water-wall materials were tested at three temperatures: 399, 468, and 538 °C (750, 875, and 1,000 °F); the superheater/reheater materials will be tested at: 538, 593, and 649 °C (1,000, 1,100, and 1,200 °F) – temperatures that span the range of boilers operating at subcritical and supercritical pressure. Upon completion of exposure testing, the condition of the coupons was evaluated macroscopically and microscopically and the materials were assessed for their suitability for oxy-combustion.

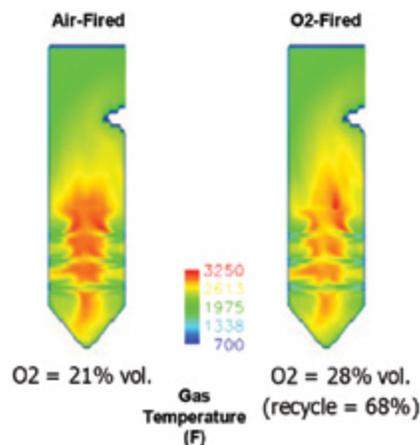


Figure 2: Example of Foster Wheeler CFD Analyses

## technology advantages

This project will identify the corrosion mechanisms that occur under oxy-combustion and assess the suitability of conventional and advanced boiler materials for this new combustion environment.

## R&D challenges

Flue gas recycle used for the oxy-combustion process could increase corrosion of boiler materials.

## results to date/accomplishments

- CFD analyses of nominal 500-MWe PC boilers (wall- and tangential-fired) have identified the bulk flue gas compositions and waterwall microclimates that will exist in these units under air- and oxy-firing.
- The most corrosive gas compositions together with intermediate levels of corrosiveness for each combustion mode were selected for use in laboratory electric furnace corrosion studies.

- The boiler materials coupon testing included five 1,000-hour test series; four test series investigated furnace waterwall microclimates that range from highly reducing (20 percent CO) to mildly oxidizing (1 percent O<sub>2</sub>) conditions, and one test series investigated the oxidizing (nominally 3 percent O<sub>2</sub>) superheater/reheater condition. The superheater/reheater and three furnace wall test series have been completed and the fifth and final test series (waterwall with 5 percent CO) began in January 2011. An example of some preliminary corrosion test results are shown below in Figure 3 for waterwall materials exposed to 2 percent CO air- and oxy-fired microclimates at 875°F for 1,000 hours with three levels of iron sulfide (FeS) deposits.

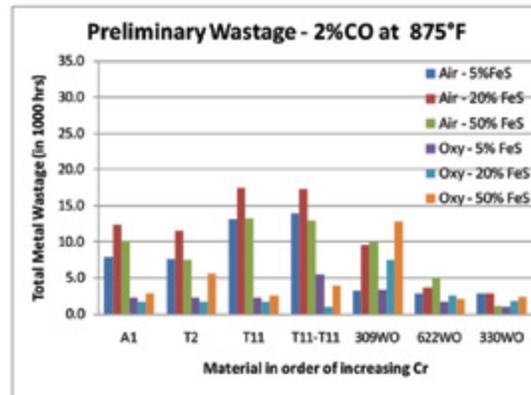


Figure 3: Example of Preliminary Corrosion Results for Waterwall Materials

- The preliminary 1,000-hour results show that under oxy-combustion:
  - The effect of oxy-combustion on test coupon corrosion varies with the type of material, deposit, and temperature.
  - Superheater/reheater and waterwall wastages are generally less than under air-firing.
  - Superheater/reheater and waterwall wastage increases with increasing temperature and tends to decrease with increasing chromium concentrations.
  - Waterwall wastages at 2 percent CO were higher than at 1 percent O<sub>2</sub>.

#### next steps

This project ended on January 31, 2012.

#### available reports/technical papers/presentations

Robertson, A.; Agarwal, H.; Gagliano, M.; Seltzer, A.; and Wang, L., "Oxy-Combustion Boiler Material Development," presented at the 2011 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, August 2011, <http://www.netl.doe.gov/publications/proceedings/11/co2capture/presentations/3-Wednesday/24Aug11-Robertson-FW-Oxy-Comb%20Boiler%20Matl.pdf>.

Robertson, A.; Agarwal, H.; Gagliano, M.; and Seltzer, A., "Oxy-Combustion Boiler Material Development," presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, September 2010. <http://www.netl.doe.gov/publications/proceedings/10/co2capture/presentations/tuesday/Archie%20Robertson-NT0005262.pdf>.

Robertson, A., "Oxy-Combustion Boiler Material Development," presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D Meeting, Pittsburgh, Pennsylvania, March 2009. <http://www.netl.doe.gov/publications/proceedings/09/CO2/pdfs/5262%20Foster%20Wheeler%20oxy-combustion%20materials%20%28Robertson%29%20mar.pdf>.

# DEVELOPMENT OF COST-EFFECTIVE OXY-COMBUSTION TECHNOLOGY FOR RETROFITTING COAL-FIRED BOILERS

B-479

OXY-COMBUSTION

## primary project goals

Babcock & Wilcox Power Generation Group, Inc. (B&W) set out to develop oxy-combustion technology for application to new and existing Cyclone and wall-fired boilers. This two-phase research project was aimed at further advancing the technology through pilot-scale testing and full-scale engineering and economic analyses of a reference coal-fired plant.

## technical goals

- Conduct pilot-scale testing to evaluate the effect of coal rank (i.e., bituminous, subbituminous, and lignite) on oxy-combustion boiler operation.
- Determine the equipment requirements for the boiler island, flue gas purification, carbon dioxide (CO<sub>2</sub>) compression, CO<sub>2</sub> transportation, and CO<sub>2</sub> sequestration for different coal ranks and boiler designs.
- Investigate the potential for multi-pollutant (nitrogen oxides [NO<sub>x</sub>], sulfur dioxide [SO<sub>2</sub>], and particulate) emissions control.
- Validate an existing three-dimensional computational flow, heat transfer, and combustion model for oxy-combustion scale-up to a commercial-size boiler.
- Conduct an engineering and economic assessment of the technology for commercial-scale retrofit and green field application for Cyclone and wall-fired boilers.
- Assess CO<sub>2</sub> capture cost reductions via energy integration of the air separation unit (ASU), flue gas purification, and CO<sub>2</sub> compression systems.
- Evaluate the impact of oxy-combustion implementation on net power production and cost of electricity (COE) for Cyclone and wall-fired boilers.

## technical content

B&W had previously conducted pilot-scale oxy-combustion testing for wall-firing at 1.8 and 30 MWth. In this project, a pilot-scale evaluation – 14 GJ/hr (6 million Btu/hr) – was conducted for three coals using a Cyclone boiler configuration at its Barberton, Ohio, test facility. An illustration of the oxy-combustion pilot-scale test facility is shown below (Figure 1). The three types of coal tested were North Dakota lignite, Western subbituminous, and Eastern bituminous. Each of the oxy-combustion tests were run for 100 continuous hours to assess the slagging, fouling, heat transfer, and overall operability characteristics. Data from the pilot-scale testing was used to validate a computational fluid dynamic (CFD) model of the oxy-combustion process. From the test data, equipment required for flue gas purification, compression, transportation, and sequestration was determined for the engineering and economic assessment.

### technology maturity:

Pilot-Scale; Equivalent to 13 tons of CO<sub>2</sub>/day

### project focus:

Oxy-Combustion Design for Retrofits

### participant:

Babcock & Wilcox Power Generation Group

### project number:

FC26-06NT42747

### NETL project manager:

José Figueroa  
jose.figueroa@netl.doe.gov

### principal investigator:

Hamid Farzan  
Babcock & Wilcox PGG  
hfarzan@babcock.com

### partners:

Air Liquide  
Battelle

### performance period:

4/1/06 – 3/31/11

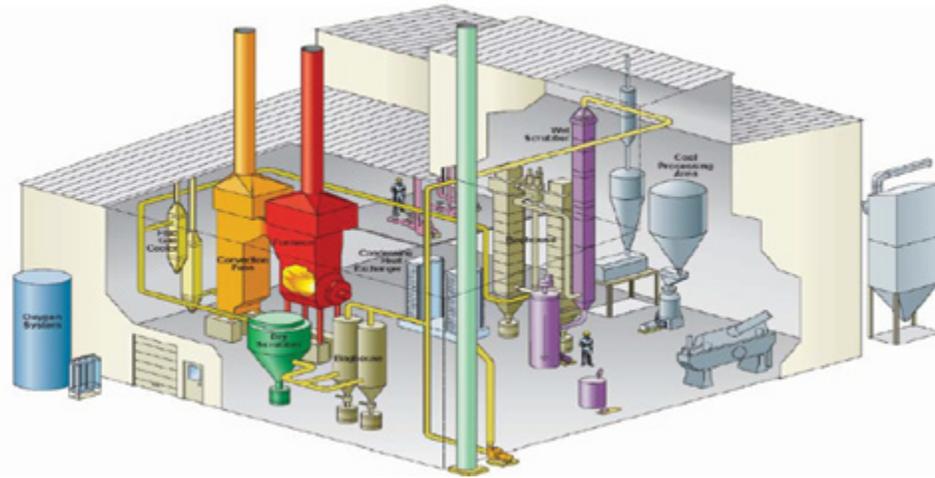


Figure 1: Illustration of B&W's Oxy-Combustion Pilot-Scale Test Facility in Barberton, Ohio

A modeling assessment was also conducted to compare three CO<sub>2</sub> capture purification processes: (1) no purification – only drying to Kinder Morgan pipeline specifications with water (H<sub>2</sub>O) at 600 parts per million volume (ppmv); (2) partial condensation at cryogenic conditions (cold box) – 95 percent CO<sub>2</sub> purity target; and (3) cold box including distillation – 1 ppm oxygen (O<sub>2</sub>) target. The purification assessment included investigation of operating costs, energy requirements, and effects of air infiltration. The following graph (Figure 2) represents a model analysis showing the effect of purification process on CO<sub>2</sub> recovery, purity, and specific energy.

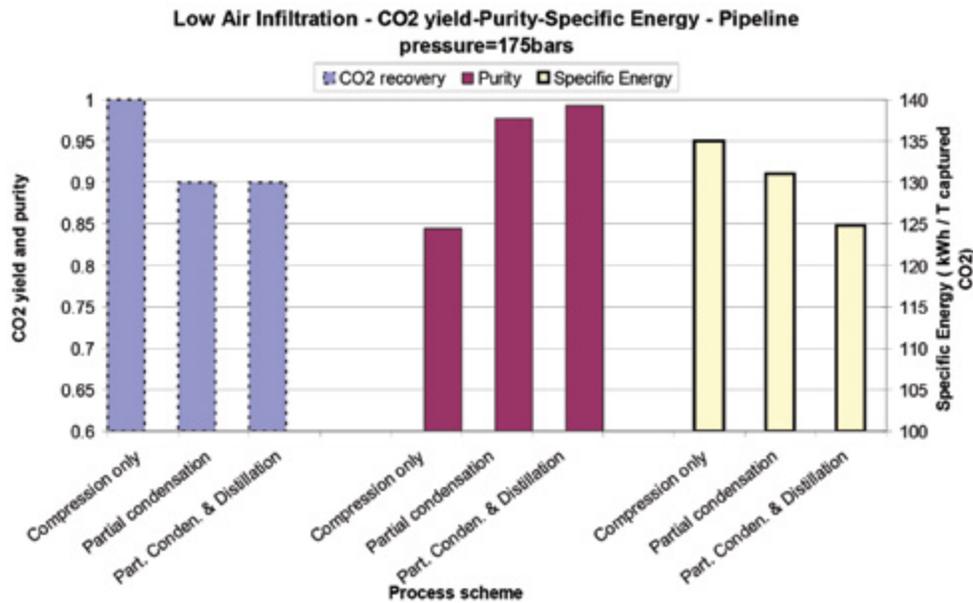


Figure 2: Model Analysis Showing Effect of Purification Process on CO<sub>2</sub> Recovery, Purity, and Specific Energy

## technology advantages

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Oxy-combustion has the potential to offer a lower-cost solution for CO<sub>2</sub> capture compared to post-combustion CO<sub>2</sub> capture technologies.

## R&D challenges

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The necessary level of flue gas purification remains an issue regarding potential adverse impacts on CO<sub>2</sub> transportation and storage:

- Potential precipitation problems with SO<sub>2</sub> forming sulfate minerals (e.g., anhydrite) if high-sulfur coal is used without scrubbing.
- Non-condensable gases, such as nitrogen (N<sub>2</sub>) and O<sub>2</sub>, could affect subsurface processes that might require more purification. For example, non-condensable gases could create multi-phase flow, which can reduce injectivity or the capacity of the storage site.

## results to date/accomplishments

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Completed oxy-combustion pilot-scale testing with the following general results:

- Oxy-combustion is a technically feasible technology for wall-fired and Cyclone boilers.
- The flame stability and Cyclone slagging characteristics was not negatively impacted by oxy-combustion.
- Boiler emissions (NO<sub>x</sub>, carbon monoxide [CO], and unburned combustibles) to the atmosphere are lower for oxy-combustion than air-firing.
- Radiant boiler and convection pass heat absorptions under optimum oxy-combustion conditions was similar to air-firing.
- Higher CO<sub>2</sub> (up to 90%) levels were achieved when air leakage was minimized by reducing pressure drop in the boiler back-end equipment.
- Sulfur dioxide must be scrubbed from recycle gas for high-sulfur coal applications to maintain acceptable boiler corrosion.
- Higher sulfur trioxide (SO<sub>3</sub>) concentrations occur with oxy-combustion than air-firing at the convection pass exit that could increase corrosion if the flue gas temperature goes below the acid dew point.

Completed engineering feasibility and economic analysis with the following general results:

- Co-sequestration of CO<sub>2</sub> and SO<sub>2</sub> might be feasible in deep geological reservoirs. It was concluded that the pipeline transportation corrosion by acid gas can be minimized by removing the moisture from the flue gas.
- Modeling of the compression and purification unit (CPU) demonstrated that the overall energy requirement is lower if flue gas inerts are removed in the CPU rather than compressing the entire flue gas for pipeline transport.
- Oxy-combustion is an economically viable technology. The incremental cost of oxy-combustion for existing boilers varied between \$.05 to \$.07/kWh, which is competitive with other technologies.
- COE for green field boilers using oxy-combustion coupled with ultra-supercritical boilers was 25 percent higher than COE for a supercritical boiler under air-firing without CO<sub>2</sub> capture, which is below the U.S. Department of Energy/National Energy Technology Laboratory (NETL) target of 35 percent.
- Oxy-combustion can be applied to the majority of the existing wall-fired and Cyclone boilers depending on space and existing equipment. The site requirements are similar to those for post-combustion capture.

## next steps

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This project ended on March 31, 2011.

- B-482 Farzan, H., et.al, “Development of Cost Effective Oxy-Combustion for Retrofitting Coal-Fired Boilers,” presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, Pittsburgh, Pennsylvania, September 2010. <http://www.netl.doe.gov/publications/proceedings/10/co2capture/presentations/tuesday/Hamid%20Farzan-NT42747.pdf>.
- Farzan, H., et.al, “Development of Cost Effective Oxy-Combustion for Retrofitting Coal-Fired Boilers,” presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D Meeting, Pittsburgh, Pennsylvania, March 2009. [http://www.netl.doe.gov/publications/proceedings/09/CO<sub>2</sub>/pdfs/42747%20B%26W%20oxy-combustion%20%28Farzan%29%20mar09.pdf](http://www.netl.doe.gov/publications/proceedings/09/CO2/pdfs/42747%20B%26W%20oxy-combustion%20%28Farzan%29%20mar09.pdf).
- Sass, B.M., et.al, “Considerations for Treating Impurities in Oxy-Combustion Flue Gas Prior to Sequestration,” presented at the 9<sup>th</sup> International Conference on Greenhouse Gas Control Technologies, Washington, DC, November 2008.
- Farzan, H., et.al, “Developing Oxy-combustion for Retrofitting Coal-fired Boilers,” presented at the 7<sup>th</sup> Annual Conference on Carbon Capture and Sequestration, Pittsburgh, Pennsylvania, May 2008. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/42747-B-W-oxy-combustion-7th-CCS-conference-2008.pdf>.

# FLUE GAS PURIFICATION UTILIZING SO<sub>x</sub>/NO<sub>x</sub> REACTIONS DURING COMPRESSION OF CO<sub>2</sub> DERIVED FROM OXYFUEL COMBUSTION

## technology maturity:

Pilot-Scale, 2.45 Tonnes CO<sub>2</sub>/Day

## project focus:

Flue Gas Purification

## participant:

Air Products and Chemicals

## project number:

FC26-08NT0005309

## NETL project manager:

Timothy Fout  
timothy.fout@netl.doe.gov

## principal investigator:

Kevin Fogash  
Air Products and Chemicals  
fogashkb@airproducts.com

## partners:

N/A

## performance period:

10/1/08 – 9/30/10

## primary project goals

Air Products and Chemicals set out to design and develop a system for purifying an oxy-combustion-derived flue gas by utilizing the reactions of sulfur oxides (SO<sub>x</sub>) and nitrogen oxides (NO<sub>x</sub>) that occur during compression, leaving behind a pressurized, pure stream of carbon dioxide (CO<sub>2</sub>).

## technical goals

- Design and construct a 15-atm flue gas pilot development unit (PDU) for the removal of SO<sub>x</sub> and NO<sub>x</sub> from actual oxy-combustion-derived CO<sub>2</sub>-rich flue gas.
- Evaluate PDU performance based on effluents at different pressures and water recycle rates.
- Characterize the PDU effluents to assess any change in performance.
- Develop an engineering model to describe the 15-atm PDU performance.

## technical content

Acidic gases must be removed from a CO<sub>2</sub> stream prior to pipeline transportation to avoid corrosion and to comply with purity requirements for applications such as enhanced oil recovery (EOR) and geological storage. In order to address this requirement, Air Products and Chemicals worked on developing a novel approach to remove SO<sub>x</sub> and NO<sub>x</sub> from the flue gas by converting them to sulfuric acid and nitric acid (HNO<sub>3</sub>).

In order to determine the effect of pressure on sulfur dioxide (SO<sub>2</sub>) and nitric oxide (NO) conversion, previous experiments were performed where one standard liter per minute (sl/min) of gas was supplied at both 8 and 15 atm. The results are shown in the Table 1.

TABLE 1: PRESSURE VS. CONVERSION

	15 atm			8 atm		
	Inlet	After Compressor and Receiver	Conversion	Inlet	After Compressor and Receiver	Conversion
ppm SO <sub>2</sub>	900	20	98%	950	150	84%
ppm NO <sub>x</sub>	500	50	90%	390	120	68%

The conversion rate clearly increases significantly with pressure. Therefore, it is logical to assume that these contaminants can be removed during the compression of CO<sub>2</sub>.

The PDU developed for this project includes three main units, as indicated below: the scrubber/condenser, the compressor, and the reactor. Fine particulate ash and acid mist in the flue gas are removed prior to compression to avoid damage to the compressor.

The compressor increases the pressure of the gas from near-atmospheric to approximately 15 atm in a multistage adiabatic compressor unit. After the initial compression, the flue gas is cooled prior to entering the reactor. In the reactor, the flue gas is contacted with water to obtain complete conversion of SO<sub>2</sub> to sulfuric acid and high conversion of NO<sub>x</sub> to HNO<sub>3</sub>.

Multiple tests were conducted within two campaigns of the project. The host site was Alstom Power's 15-MWth oxy-combustion pilot unit. The 15-bar reactor system received a slipstream of 0.25 to 0.33 MWth equivalent flow rate from the Alstom unit for several days. A variety of process conditions were tested, including changes in SO<sub>x</sub> and NO<sub>x</sub> feed levels, to enable a broad understanding of the technology. The flue gas PDU is shown in Figure 2. In the reactor, the flue gas was contacted with water to obtain up to complete conversion of SO<sub>2</sub> to sulfuric acid and high conversion of the NO<sub>x</sub> to HNO<sub>3</sub>. Figure 3 shows an example of the results obtained.

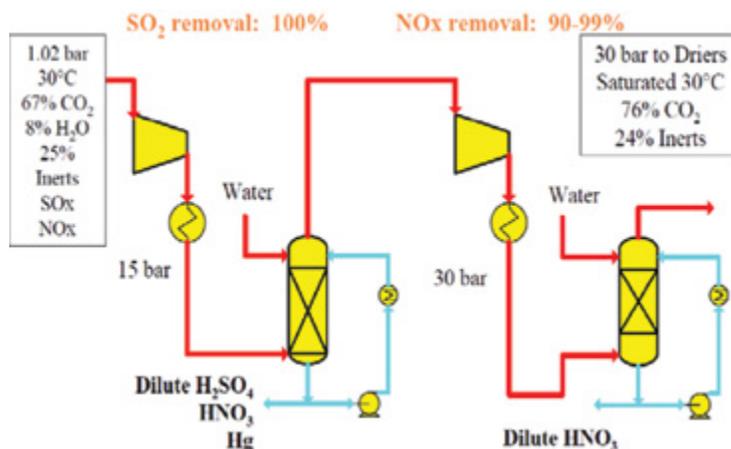


Figure 1: Flue Gas Pilot Development Unit (PDU)



Figure 2: PDU at the Host Site

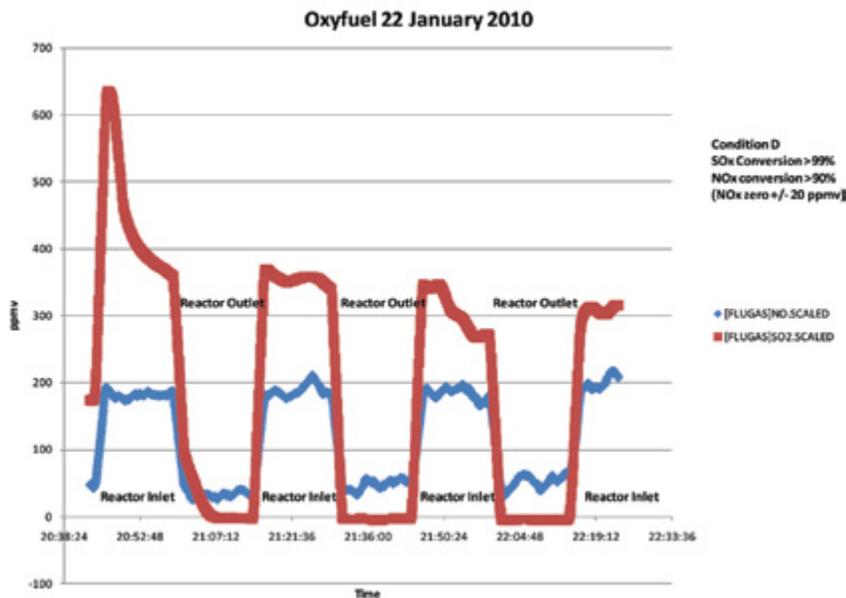


Figure 3: Example Results from the Oxyfuel Test

## technology advantages

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By incorporating an efficient NO<sub>x</sub> and SO<sub>x</sub> removal system with a compressor, the need for low NO<sub>x</sub> burners, flue gas desulfurization (FGD), and post-combustion NO<sub>x</sub> control systems are greatly reduced or eliminated for oxy-combustion plants. In particular, this system allows for a degree of freedom to optimize any upstream purification needs and target reduction in size or removal of upstream equipment for the retrofit of an existing plant or the construction of a new plant.

## R&D challenges

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- Obtaining sufficient data for engineering design and to further develop the understanding of the effect of residence time, pressure, and temperature on the unit performance.
- The prolonged presence of acid gases in the system and the presence of such gases at high pressures in the compressor may lead to the requirement of more advanced materials of construction.

## results to date/accomplishments

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- Developed a simulation of the flue gas PDU and modeled the reactions occurring within the reactor.
- Conducted simulations to understand the influence of liquid to vapor flow rates on SO<sub>2</sub> and nitrogen dioxide (NO<sub>2</sub>) conversions, as well as overall residence time in the reactor.
- Developed a design specification for the PDU and auxiliary equipment.
- Completed construction and installed the PDU at host site.
- For the overall process, total SO<sub>2</sub> removal was 40 to 100 percent (based on gas compositions).
- For the overall process, total NO<sub>x</sub> removal was 60 to 90 percent (based on gas compositions).
- The effects of variations in the SO<sub>2</sub>/NO<sub>x</sub> feed ratio, column pressure, gas flow rate, and liquid recirculation on the reactor performance were elucidated. Process performance was most sensitive to SO<sub>2</sub>/NO<sub>x</sub> feed ratio over the range of parameter values investigated.
- SO<sub>2</sub> was removed from the flue gas through both sulfite and sulfate mechanisms.
- No evidence of NO<sub>x</sub> removal was observed prior to compression, confirming that elevated pressure was required to accelerate the oxidation reaction of NO to NO<sub>2</sub> to a rate at which appreciable NO<sub>x</sub> removal (as HNO<sub>3</sub>) could be achieved.

## next steps

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This project ended on September 30, 2010.

## available reports/technical papers/presentations

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Fogash, K., "Flue Gas Purification Utilizing SO<sub>x</sub>/NO<sub>x</sub> Reactions During Compression of CO<sub>2</sub> Derived from Oxyfuel Combustion," presented at the 2010 NETL CO<sub>2</sub> Capture Technology Meeting, September 2010. <http://www.netl.doe.gov/publications/proceedings/10/co2capture/presentations/tuesday/Kevin%20Fogash%20-%20NT0005309.pdf>.

Fogash, K., "Flue Gas Purification Utilizing SO<sub>x</sub>/NO<sub>x</sub> Reactions During Compression of CO<sub>2</sub> Derived from Oxyfuel Combustion," presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D Meeting, March 2009. <http://www.netl.doe.gov/publications/proceedings/09/CO2/pdfs/5309%20Air%20Products%20oxy-combustion%20purification%20%28Fogash%29%20mar09.pdf>.

White, V.; Allam, R.; and Miller, E., "Purification of Oxyfuel-Derived CO<sub>2</sub> for Sequestration or EOR," Technical paper presented at the 8<sup>th</sup> International Conference on Greenhouse Gas Control Technologies in Trondheim, Norway, 2006. <http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/5309-Air-Products-oxy-combustion-GHGT-8-paper.pdf>.

# OXYGEN-FIRED CO<sub>2</sub> RECYCLE FOR APPLICATION TO DIRECT CO<sub>2</sub> CAPTURE FROM COAL-FIRED POWER PLANTS

## primary project goals

Southern Research Institute (SRI) set out to design and develop carbon dioxide (CO<sub>2</sub>) recycle technology for oxy-combustion retrofit applications.

## technical goals

- Modify the 1-MWth pilot-scale Combustion Research Facility (CRF) to allow oxy-combustion and CO<sub>2</sub>-recycle operations.
- Design, manufacture, and install an oxy-combustion burner specifically for the CRF.
- Collect data on furnace temperatures, unburned carbon (UBC), gas composition, and flow rates into and out of the furnace.
- Evaluate the effect of various parameters, including firing configuration, oxygen (O<sub>2</sub>) purity, CO<sub>2</sub> recycle rate, O<sub>2</sub> concentration, and coal type.

## technical content

SRI developed flue gas recycle for oxy-combustion retrofit application to coal-fired utility boilers in order to avoid the excessive flame temperatures that are associated with oxy-combustion and to maintain flow and heat-transfer requirements in the furnace and convective sections.

SRI conducted the pilot-scale, oxy-combustion experiments using a modified CRF. Figure 1 contains a diagram that represents the CRF with the necessary modifications. These modifications include the addition of an O<sub>2</sub> storage tank and concrete pad; O<sub>2</sub> skid, control, and safety systems; a new burner by Maxon that is designed for O<sub>2</sub> combustion and CO<sub>2</sub> recycle; the additional ducting to allow flue gas recirculation to the burner; and the decrease in ducting size to account for smaller flue gas flow rate.

### technology maturity:

Pilot-Scale Using Actual Flue Gas, 25 tonnes of CO<sub>2</sub>/day

### project focus:

Evaluation of Gas Recycle for Oxy-Combustion

### participant:

Southern Research Institute

### project number:

FC26-05NT42430

### NETL project manager:

Timothy Fout  
timothy.fout@netl.doe.gov

### principal investigator:

Bob Dahlin  
Southern Research Institute  
dahlin@southernresearch.org

### partners:

CORR Systems  
Doosan Power Systems  
Linde Gas  
Maxon Corporation  
Reaction Engineering International  
Southern Company

### performance period:

9/27/05 – 9/25/10

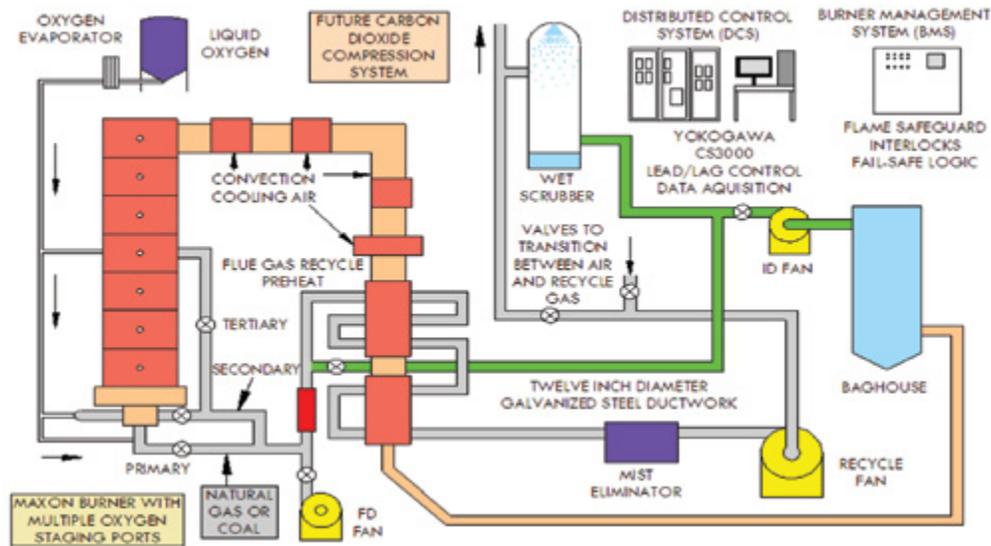


Figure 1: Diagram of the Modified CRF

The CRF was used to investigate the effects of coal type, firing staging, percentage of flue gas recycled, and  $O_2$  purity on oxy-combustion.

In addition to flue gas recirculation, an advanced  $O_2$  burner developed by Maxon was used to allow the flame shape and heat released to be controlled and to provide a stable attached flame. Figure 2 is an image of the flame produced by the oxy-combustion burner.

A preliminary test was performed on the Maxon burner using Illinois Bituminous coal. The test chamber was heated to  $2,400^\circ\text{F}$  with 3 percent excess  $O_2$ . Performance of the burner under air-fired conditions resulted in a nitrogen oxides ( $NO_x$ ) emission rate of 0.3 to 0.4 lbs/MMBtu. However, oxy-combustion testing resulted in a lower emission rate of 0.16 to 0.18 lbs  $NO_x$ /MMBtu.

Reaction Engineering International (REI) updated a computational fluid dynamics (CFD) model of the CRF facility to predict oxy-combustion burner performance. The model describes temperatures, reaction rates, char burnout, and  $NO_x$  formation and/or destruction as a function of  $O_2$  purity, stoichiometry, coal type, staging, furnace exit  $O_2$  (FEO), and fuel processing. The model was validated by the CRF oxy-combustion experiments and can be utilized for any follow-on preliminary designs for project scale-up.

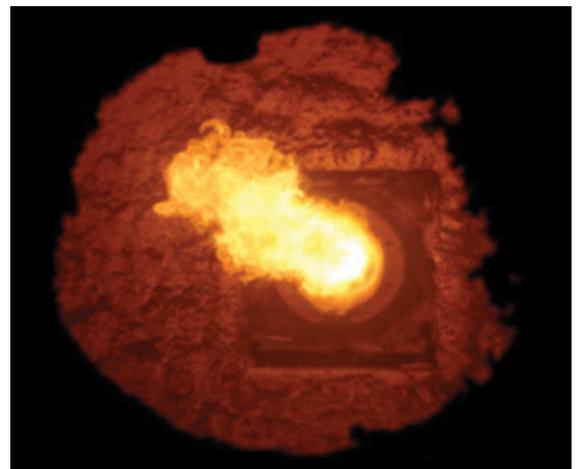


Figure 2: Image of Maxon Oxygen Burner Flame

## technology advantages

- The oxy-combustion burner is able to maintain a stable attached flame and can light off without natural-gas assist.
- The reduced volume of flue gas produced under oxy-combustion conditions should be less expensive to purify and compress for  $CO_2$  sequestration than flue gas of a conventional air-fired, pulverized coal (PC) plant.
- For plants burning Powder River Basin (PRB) coal, recycling dry flue gas through the coal pulverizers should minimize concern of pulverizer fires.
- Oxy-combustion burners and flue gas recycle rate can be tuned to achieve low-cost operation (i.e., minimize recycle) and maximum heat transfer for a given boiler type and plant configuration.
- Advanced thermodynamic cycles can recover some of the energy penalty associated with air separation for new power plant applications.

## R&amp;D challenges

B-488

- Reduce cost of oxy-combustion retrofit for existing plants.
- Reduce energy penalty for O<sub>2</sub> production, which is approximately 25 percent for conventional cryogenic process.
- Address potential corrosion of low-temperature ductwork and equipment due to flue gas recycle.

## results to date/accomplishments

- Completed the oxy-combustion and flue gas recycle retrofit of the CRF.
- Completed the update of the CFD model for oxy-combustion and flue gas recycle.
  - The CFD model was used to predict the flame geometry and temperatures in the CRF and make a comparison with the air-fired case.
  - The CFD model predictions were consistent with the experimental data in showing that the O<sub>2</sub>-fired Maxon burner produced lower flame temperatures than the air-fired burner.
- Conducted oxy-combustion and flue gas recycle testing using the CRF.
  - The Maxon staged oxy-combustion burner produced a stable flame over a significant range of firing turndown, staging, and while firing five different U.S. coal types, including three eastern bituminous coals, a PRB sub-bituminous coal, and a western bituminous coal.
  - The parametric testing included extent of recycle and recycle turndown, FEO percentage, O<sub>2</sub> staging in the burner cup, burner quarl, secondary recycle, and overfire recycle. The load was also varied with and without changes in the amount of recycle flow.
  - The oxy-combustion burner produced lower flame temperatures than for air firing, which should enable safe operation, reduction of recycle flow without concern about furnace flame temperatures, and could be effective at reducing slagging and fouling in the boiler and super heater for full-scale applications.
  - The temperature/time profile was affected by four main factors: (1) the difference in load, (2) the difference in recycle flow back to the furnace and the absence of nitrogen (N<sub>2</sub>) flow, (3) the difference in the Maxon oxy-burner design and an air-burner design, and (4) the difference in diffusivity of CO<sub>2</sub> and N<sub>2</sub>.
  - Hydrochloric (HCl) acid concentration in the flue gas was consistent with the coal chlorine content and no buildup of HCl was observed via flue gas recirculation.
  - The sulfur dioxide (SO<sub>2</sub>) concentration in the flue gas was consistent with the coal sulfur content and no buildup was observed via flue gas recirculation.
  - The carbon monoxide (CO) levels were also consistent with that of air firing, and the CO concentration had a significant correlation with only one parameter, that of FEO. Above 1.5 percent FEO, the CO concentration was around 50 parts per million by volume (ppmv); below 0.5 percent FEO, the CO concentration could reach hundreds of ppmv.
  - The relation of UBC in the ash with FEO was consistent with that of CO. The UBC in the ash increased with increased furnace staging similar to air firing.
  - NO<sub>x</sub> emissions were high under baseline oxy-combustion conditions. However, NO<sub>x</sub> emissions were dramatically reduced with staging of the O<sub>2</sub> in the secondary recycle gas and overfire-recycle flue gas.
  - The control system and retrofit concept allowed safe and controlled startup, changing of conditions, continuous operation without buildup of moisture, acid gases, pollutants, or other problems, and yielded efficient combustion of the coal and associated volatile gases.

## next steps

The project ended on September 25, 2010.

[available reports/technical papers/presentations](#)

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Gale, T. K., "Oxy-Fired CO<sub>2</sub> Recycle for Application to Direct CO<sub>2</sub> Capture from Coal-Fired Power Plants," presented at the 34<sup>th</sup> International Technical Conference on Coal Utilization & Fuel Systems, Clearwater, Florida, June 2009.

Gale, T. K., "Oxy-Fired CO<sub>2</sub> Recycle for Application to Direct CO<sub>2</sub> Capture from Coal-Fired Power Plants," presented at the Annual NETL CO<sub>2</sub> Capture Technology for Existing Plants R&D Meeting, Pittsburgh, Pennsylvania, March 2009. <http://www.netl.doe.gov/publications/proceedings/09/CO2/pdfs/42430%20SRI%20oxy-combustion%20%28Gale%29%20mar09.pdf>.

Gale, T. K., "Oxy-Fired Flue-Gas Recycle Pilot-Plant Demonstration," presented at the 14<sup>th</sup> Annual EUEC Conference, Phoenix, Arizona, February 2011. [http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/F8\\_4-Gale\\_EUEC-2011\\_Presentation.pdf](http://www.netl.doe.gov/File%20Library/Research/Coal/ewr/co2/F8_4-Gale_EUEC-2011_Presentation.pdf).

# CANMETENERGY CO<sub>2</sub> R&D CONSORTIUM

B-490

OXY-COMBUSTION

## primary project goals

The CanmetENERGY CO<sub>2</sub> R&D Consortium (Consortium) set out to conduct oxy-fuel combustion research and development (R&D) using a 0.3 MWth (1 million Btu/h) modular pilot-scale facility. The Consortium has completed nine successive phases of R&D that include oxy-fuel combustion, advanced power cycles, integrated multi-pollutant control, and carbon dioxide (CO<sub>2</sub>) capture and compression technologies.

## technical goals

The technical goal of the Consortium is to develop advanced energy conversion technologies with near-zero emissions for improved efficiency and commercial competitiveness for capture of CO<sub>2</sub> and air pollutants resulting from combustion of fossil fuels. One emphasis of the Consortium research program is the oxy-fuel combustion technology. Since combustion takes place in an oxygen (O<sub>2</sub>)-enriched environment, the flue gas comprises mainly CO<sub>2</sub>, water, and minor impurities. This CO<sub>2</sub>-rich flue gas stream can then be purified, dried, and compressed for pipeline transport and use or permanent storage in geological formations. Oxy-fuel combustion also results in efficiency advances of high flame temperatures and reduced equipment sizes due to lower gas volume.

## technical content

The Consortium activities in the past have included experimental investigations using coal; coal slurry; bitumen and natural gas to study the characteristics of oxy-fuel combustion; advanced near-zero emissions Brayton and Rankine cycles; solid oxide fuel cell modeling; multi-pollutant capture research for integrated removal of fine particulates, nitrogen oxide (NO<sub>x</sub>), sulfur oxide (SO<sub>x</sub>), and mercury (Hg); advanced oxy-fuel combustion processes and co-firing with opportunity fuels such as petroleum coke; system components and prototype design and pilot-scale testing; and modeling and development of new CO<sub>2</sub> capture and compression processes.

The latest completed Phase 9 of the Consortium's program included the development of a CO<sub>2</sub> capture and compression unit (CO<sub>2</sub>CCU). This unit is capable of separating and compressing CO<sub>2</sub> from combustion flue gas streams for pipeline transport and storage. Part of this work involved the development of a CO<sub>2</sub> high-pressure test cell for studying CO<sub>2</sub> phase change, generating vapor-liquid equilibrium (VLE) data, and studying the impact of impurities in the flue gas stream on the capture processes. This has important practical applications relating to the CO<sub>2</sub> pipeline, material selection, and commercial design of these systems. Other ongoing R&D activities include the modeling of advanced supercritical oxy-coal plants with CO<sub>2</sub> capture; cost analysis; the development and testing of multi-pollutant control strategies, as well as testing in oxy-steam mode; and optimization of a novel multi-function oxy-fuel/steam burner.

Figure 1 shows the major process components comprising the 0.3 MWth oxy-fuel Vertical Combustor Research Facility (VCRF) integrated with the CO<sub>2</sub>CCU. The overall pilot-scale research facility is used to develop pollutant control technologies that incorporate a fabric filter or electrostatic precipitator (ESP) for particulate capture, condensing heat exchangers and/or SO<sub>x</sub> scrubbing to remove acid gases and oxidized Hg from the flue gas combustion stream.

### technology maturity:

Pilot-Scale Research

### project focus:

Engineering Assessment of Oxy-Combustion

### participant:

Natural Resources Canada-CanmetENERGY

### project number:

IEA-CANMET-CO<sub>2</sub>

### NETL project manager:

Timothy Fout  
timothy.fout@netl.doe.gov

### principal investigator:

Dr. Kourosh Zanganeh  
Natural Resources Canada-CanmetENERGY  
kourosh.zanganeh@nrcan.gc.ca

### partners:

**Phase 9 Consortium Members:**  
Ontario Power Generation, SaskPower, Governments of Canada and Alberta, Babcock and Wilcox, U.S. Dept. of Energy, and the CO<sub>2</sub> Capture Project (CCP2)

### performance period:

9/30/99 – 12/31/09

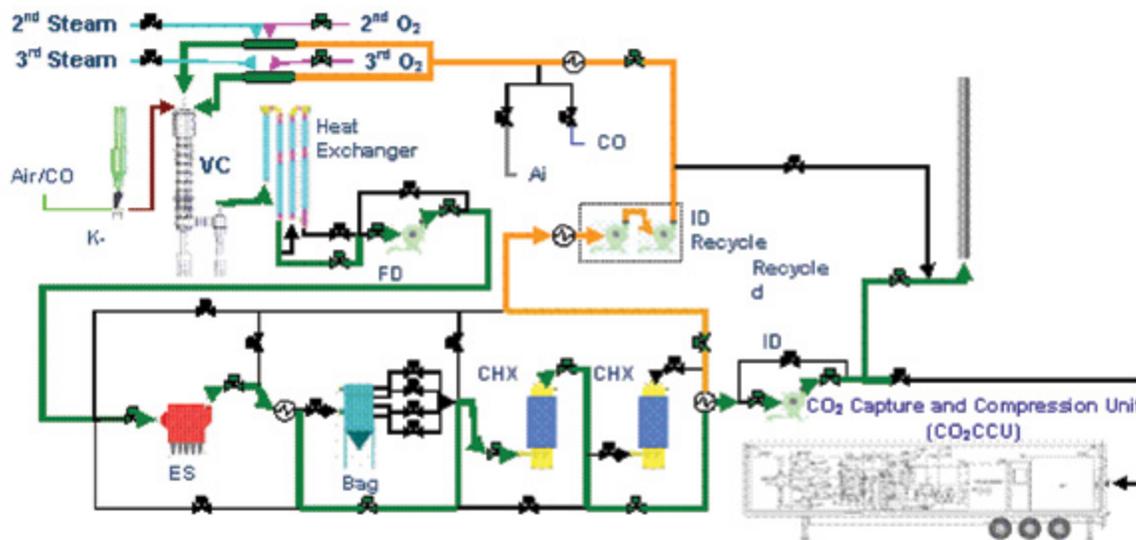


Figure 1: Schematic of the CanmetENERGY's Integrated Oxy-Fuel Vertical Combustor Research Facility

Figure 2 displays the pilot-scale CO<sub>2</sub>CCU. The CO<sub>2</sub>CCU is capable of processing CO<sub>2</sub> flue gas streams at a maximum rate of 160 Kg/hr with CO<sub>2</sub> concentrations of 50 percent or higher (in dry volume) to produce a CO<sub>2</sub> product stream with more than 95 percent purity.



Figure 2: CanmetENERGY's CO<sub>2</sub> Capture and Compression Unit

The high-pressure CO<sub>2</sub> test cell and its high-pressure viewing chamber (HPVC) shown in Figure 3 were used to create supercritical CO<sub>2</sub> and study the CO<sub>2</sub> phase change in a controlled environment. The chamber can handle a maximum pressure of 200 atm and a temperature range of -60 to 150°C. There are two gas and liquid sample ports located at different heights and optical windows with a camera for observations inside the chamber to study the CO<sub>2</sub> liquid-gas interface.



Figure 3: CanmetENERGY's High-Pressure CO<sub>2</sub> Test Cell and Bench-Scale Facility

### technology advantages

The program allows research to be carried out at a pilot scale small enough to reduce the overall R&D cost, while the experiments scale is sufficiently large enough to provide proof-of-concept before proceeding to a larger and more costly medium-scale pilot technology demonstration.

### R&D challenges

- Integration and cycle development for O<sub>2</sub>/flue gas recirculation (FGR), O<sub>2</sub> combustion, and hydroxy-fuel combustion of fossil fuels in different advanced cycles.
- Improving the understanding of combustion, heat transfer, and emissions in oxy-fuel combustion.
- Development of environmental multi-pollutant controls for NO<sub>x</sub>, SO<sub>x</sub>, Hg, and particulates.
- Minimizing energy demand for O<sub>2</sub> production while keeping the O<sub>2</sub> purity high.
- Decreasing energy consumption for capture and compression of CO<sub>2</sub>.

### results to date/accomplishments

- Developed new, ultra-low NO<sub>x</sub> oxy-combustion burner and tested the prototype burners in VCRF with sub-bituminous and lignite coals.
- Determined that FE<sup>3+</sup> salts were capable of oxidizing Hg and achieved a 75 percent Hg oxidation with an optimal pH between 1 and 3 on bench-scale tests.
- Increased the computational fluid dynamic (CFD) tools for model simulation of oxy-combustion flame characteristics.
- Created a CO<sub>2</sub> capture and compression process simulator and implemented a pilot-scale CO<sub>2</sub> capture research facility that has enhanced the program's CO<sub>2</sub> research capabilities.
- Developed new advanced gas turbine and high-efficiency fuel cell-based power generation cycles.
- Developed models of advanced supercritical oxy-coal plants with CO<sub>2</sub> capture and cost models for economic analysis.
- Developed and tested multi-pollutant control strategies and processes.
- Conducted testing in oxy-steam mode for pulverized coal and performed optimization of a novel multi-function oxy-fuel/steam burner.

[next steps](#)

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This project ended on December 31, 2009.

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[available reports/technical papers/presentations](#)

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“Novel Oxy-Steam Burner for Zero-Emission Power Plants,” 1<sup>st</sup> International Oxy-Fuel Combustion Conference, Cottbus, Germany, September 2009.

“Performance of an Advanced Pilot-Scale CO<sub>2</sub> Capture and Compression Unit,” 1<sup>st</sup> International Oxyfuel Combustion Conference, Cottbus, Germany, September 2009.

“An Integrated Approach for Oxy-fuel Combustion with CO<sub>2</sub> Capture and Compression,” 7<sup>th</sup> Annual Conference on CCS – May 5-8, 2008.

# MULTI-POLLUTANT CONTROL THROUGH NOVEL APPROACHES TO OXYGEN-ENHANCED COMBUSTION

## primary project goals

Washington University set out to perform experiments and modeling to develop best practices for implementing oxy-fuel combustion with flue gas recirculation and sorbent processes to minimize nitrogen oxides (NO<sub>x</sub>), particulate matter (PM<sub>2.5</sub>), and mercury (Hg) emissions, while maximizing combustion efficiency.

## technical goals

- Determine the influence of gas composition in the primary and secondary oxidizer streams on combustion temperature, flame stability, the formation of NO<sub>x</sub>, and submicron particle formation during oxy-fuel combustion.
- Experimentally determine the optimum approach to mixing coal, oxygen (O<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), and sorbents.
- Determine the effect of flue gas composition on electrostatic precipitator (ESP) performance for oxy-combustion.
- Evaluate the performance of non-carbon-based sorbents (nanostructured titanium dioxide [TiO<sub>2</sub>] and potassium iodide) for mercury capture in coal combustion systems.

## technical content

Oxy-combustion offers several advantages over traditional air-combustion technologies, including higher temperatures and smaller flue gas volume. However, in order to maintain the combustion temperatures at appropriate levels for existing materials, the reactants (fuel or O<sub>2</sub>) must be diluted with recirculated flue gas. The flue gas can be added to the primary oxidizer (PO; i.e., the fuel stream), the secondary oxidizer (SO; i.e., the oxidizer stream), or some combination of both. Washington University researched the impact of flue gas placement on critical parameters such as flame temperature, NO<sub>x</sub> formation, and particulate formation. Washington University also performed flame stability studies for oxy-combustion. The experiments used a 10- to 25-kW down-fired coal combustor consisting of a non-swirling PO and a swirling SO. The system was run under slight negative pressure and the PO preheat temperature was maintained between 300 and 350 °C.

Results from these studies showed that 30 mole percent O<sub>2</sub> is required when using CO<sub>2</sub> as the inert gas to obtain stability results similar to conventional coal air-combustion. Also, “inert exchange” flames (i.e., flames where the O<sub>2</sub> concentration was reduced in the PO stream and enriched in the SO stream) were shown to have improved flame stability when compared with conventional coal air-combustion despite the O<sub>2</sub> concentration in the PO being substantially reduced.

### technology maturity:

Laboratory-Scale, 1.2 tonnes CO<sub>2</sub>/day

### project focus:

Multi-Pollutant Control

### participant:

Washington University

### project number:

FG26-05NT42531

### NETL project manager:

Arun Bose  
arun.bose@netl.doe.gov

### principal investigator:

Richard L. Axelbaum  
Washington University  
axelbaum@wustl.edu

### partners:

N/A

### performance period:

8/29/05 – 2/28/09

## technology advantages

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Oxy-combustion can offer increased temperature, increased thermal efficiency, reduced pollutant emissions, reduced fuel consumption, and improved flame stability. Oxy-combustion, when coupled with appropriate burner designs, can reduce NO<sub>x</sub> emissions beyond levels achieved by using overfire air and low-NO<sub>x</sub> burners. Also, oxy-combustion with flue gas recirculation concentrates CO<sub>2</sub> levels, helping to reduce the cost of capture; concentrations up to 95 percent can be achieved.

## R&D challenges

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The flame structure of non-premixed systems is changed dramatically when oxy-combustion is used and depends on where the flue gas is added to control flame temperature.

## results to date/accomplishments

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- Developed an understanding of flame geometry and flame stability under oxy-fuel combustion.
- Developed a simple model that explains the presence of appreciable molecular O<sub>2</sub> at the location of peak temperature in oxy-fuel combustion.
- Obtained NO<sub>x</sub> measurements as a function of gas composition in the PO and SO streams.
- Tested the performance of TiO<sub>2</sub> with UV irradiation for Hg capture in a bench- and pilot-scale system.
- Quantified flame stability in a Type I laboratory-scale pulverized coal combustor as a function of inert gas type and O<sub>2</sub> concentration in both the PO and SO streams.
- Developed a model of soot inception limits under oxy-fuel combustion conditions and validated with gaseous fuels.
- Evaluated the performance of an ESP under flue gas conditions associated with oxy-combustion.
- Completed a study of blow-off limits in oxy-coal combustion.
- Demonstrated that the approach used to mixing the oxy-coal flame can lead to stronger flames even with reduced O<sub>2</sub> in the primary region.

## next steps

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This project ended on February 28, 2009.

## available reports/technical papers/presentations

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Axelbaum, R.L., "Multi-pollutant Control Through Novel Approaches to Oxygen Enhanced Combustion," Final Technical Report, December 2009.

Skeen, S.A.; Kumfer, B.M.; and Axelbaum, R.L. "Nitric oxide emissions during coal and coal/biomass combustion under air-fired and oxy-fuel conditions," *Energy & Fuels*, 24(8), 4144-4152, 2010.

Suriyawong, A.; Chen, X.; and Biswas, P. "Nano-Structured Sorbent Injection Strategies for Heavy Metal Capture in Combustion Exhausts," *Aerosol Science and Technology*, 44(8): 676-691 (2010).

Skeen, S.A.; Yablonsky, G.; and Axelbaum, R.L. "Characteristics of non-premixed oxygen-enhanced combustion: II. Flame structure effects on soot precursor kinetics resulting in soot-free flames," *Combustion and Flame*, 157(9), 1745-1752, 2010.

Skeen, S.A.; Yablonsky, G.; and Axelbaum, R.L. "Characteristics of non-premixed oxygen-enhanced combustion: I. The presence of appreciable oxygen at the location of maximum temperature," *Combustion and Flame*, 156(11), 2145-2152, 2009.

Li, Y.; Suriyawong, A.; Daukoru, M.; Zhuang, Y.; and Biswas, P. "Measurement and Capture of Fine and Ultrafine Particles from a Pilot-Scale Pulverized Coal Combustor with an Electrostatic Precipitator," *Journal of the Air & Waste Management Association*, 59(5): 553-559 (2009).

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Kumfer, B.M.; Skeen, S.A.; and Axelbaum, R.L. "Soot inception limits in laminar diffusion flames with application to oxy-fuel combustion," *Combustion and Flame*, 154(3), 546-556, 2008.

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# A MECHANISTIC INVESTIGATION OF NITROGEN EVOLUTION AND CORROSION WITH OXY-COMBUSTION

technology maturity:

Pilot-Scale Using Simulated Flue Gas, 0/05 Tonnes CO<sub>2</sub>/day

project focus:

Nitrogen behavior in Oxy-Combustion

participant:

Brigham Young University

project number:

FG26-05NT42530

NETL project manager:

Arun C. Bose  
arun.bose@netl.doe.gov

principal investigator:

Dale R. Tree  
Brigham Young University  
treed@byu.edu

partners:

Air Liquide

performance period:

8/4/05 – 12/31/08

## primary project goals

Brigham Young University (BYU) set out to investigate the evolution of nitrogen from its origin in coal to emissions in both air- and oxy-combustion. In addition, BYU also set out to develop a model of detailed kinetics, devolatilization, and char oxidation in a simple plug flow to interpret the data.

## technical goals

- Modify the flat flame burner (FFB) to run using simulated oxy-fuel combustion.
- Modify the multi-fuel flow reactor (MFR) to run with simulated oxy-fuel combustion.
- Gather data through experimental measurements of gas species in air-fired and oxy-fuel pulverized coal flames.
- Develop a computational model of the combustion process, including fuel devolatilization, gas phase kinetic mechanisms, and char oxidation.

## technical content

The experiments were performed at two facilities designed to investigate the evolution of coal nitrogen species, fuel nitrogen oxide (NO<sub>x</sub>) formation, and emissions during combustion processes in air and oxygen (O<sub>2</sub>)/carbon dioxide (CO<sub>2</sub>) mixtures. A model of detailed kinetics, devolatilization, and char oxidation in a plug flow operation was developed and used to interpret the data collected.

In the experiments performed by BYU, the flue gas was not recycled. Rather, bottled CO<sub>2</sub> was used to simulate dry recycled flue gas. While the results are applicable to entrained-flow pulverized coal combustion in general, the absence of turbulence in the laminar flow experiment is a notable difference from any practical combustor.

Pulverized coal was burned in a refractory-lined, laminar flow reactor referred to as the MFR, shown in Figure 1.

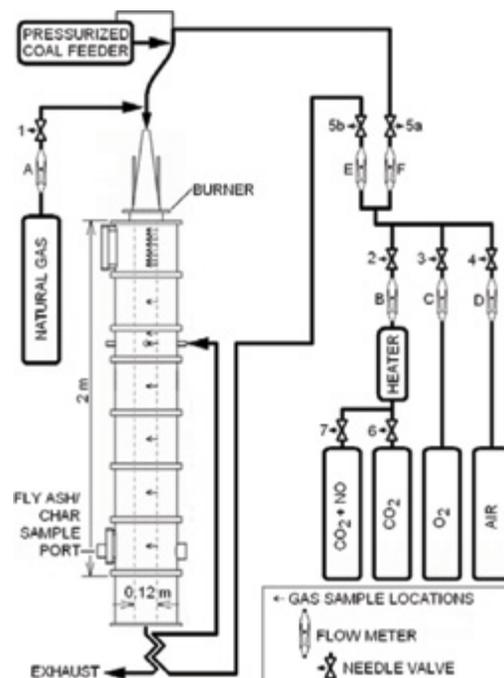


Figure 1: Schematic of the Multi-Fuel Flow Reactor

The unit at the first facility was a premixed, down-fired staged reactor where the first stage was fuel-rich followed by a burnout oxidizer stage. The oxidizer was varied from air (23% O<sub>2</sub> by mass in nitrogen [N<sub>2</sub>]) to two different mixtures of O<sub>2</sub> and CO<sub>2</sub> (25 and 30% O<sub>2</sub> by mass). The coals used were Pittsburgh #8, Illinois #6, and Powder River Basin (PRB). Selected coal properties are shown in Figure 2.

	Sub-bituminous	Illinois #6	Pittsburgh #8
<b>Proximate Analysis</b>			
	DAF wt%	DAF wt%	DAF wt%
Volatile Matter	49.72	44.17	41.96
Fixed Carbon	50.28	55.83	58.04
Ash (wt%, dry)	6.42	9.31	10.67
Higher Heating Value (Btu/lb, DAF)	11981	14226	14785
ASTM Rank	Sub-bituminous A	High-volatile C bituminous	High-volatile A bituminous
<b>Ultimate Analysis</b>			
	DAF wt%	DAF wt%	DAF wt%
C	70.56	81.88	85.19
H	4.18	4.37	4.87
O	23.63	7.83	4.70
N	1.04	1.27	1.38
S	0.59	4.64	3.86
	100	100	100

Figure 2: Selected Properties of the Coals

Both the air and mixture cases produced a rapid initial formation of nitric oxide (NO), with a similar amount of total fuel nitrogen converted to NO. In air combustion, NO can be either formed or reduced by thermal equilibrium forces dependent on the local equivalence ratio. At an initial or primary zone stoichiometric ratio (SR) of 0.82, air combustion appeared to produce thermal NO; while at an SR of 0.65, no evidence of thermal NO is seen. In oxy-combustion, initial NO formation produced concentrations above equilibrium, creating a situation where NO was being destroyed by thermal processes at all measured SR.

There is competition between the NO destruction in the fuel-rich region and the NO formation at tertiary air injection, which creates an effluent out of NO minimum for each oxidizer. The magnitude of the minimum was similar for air- and oxy-fuel combustion; however, the SR at the minimum was higher for oxy-fuel combustion, suggesting that oxy-fuel combustion does not require as deep of a staging environment to achieve NO<sub>x</sub> reduction and can therefore achieve higher burnout.

The second facility included an FFB with particle and gas sampling. Char particles were sampled after passing through either air- or oxy-flames. In oxy-flames, the normal diluents of N<sub>2</sub> were replaced with CO<sub>2</sub>. The ratio of O<sub>2</sub>/CO<sub>2</sub> was varied in order to produce different flame temperatures. There was little difference observed between air- and oxy-fuel pyrolysis of coals.

The probe, shown in Figure 3, was used to sample gas in the reactor.

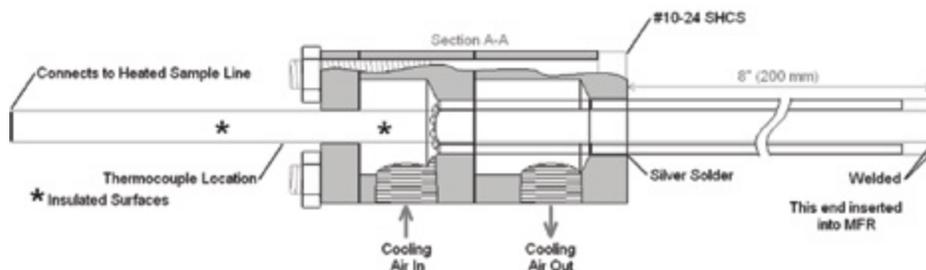


Figure 3: Diagram of the Air-Cooled Gas Sampling Probe

In unstaged, premixed combustion, air- and oxy-fuel combustion produced similar levels of fuel nitrogen conversion to NO<sub>x</sub>. Low-NO<sub>x</sub> emissions from oxy-fuel combustion are therefore not achieved without staged mixing of the oxidizer and fuel, as is the case for conventional air combustion.

## technology advantages

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The research will supply information that will significantly aid in the development of oxy-combustion technology.

## R&D challenges

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Due to the use of bottled CO<sub>2</sub> to simulate dry recycled flue gas, there was an absence of turbulence in the laminar flow experiment, which creates a notable difference from any practical combustor. This could possibly lead to difficulties when scaling to a commercial plant.

## results to date/accomplishments

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- Modified the FFB to run using simulated oxy-fuel combustion.
- Modified the MFR to run with simulated oxy-fuel combustion.
- Completed air- and oxy-combustion NO<sub>x</sub> profiles with 500 parts per million (ppm) NO added to the reactants to determine the extent of reburning in oxy-combustion.
- Completed a staged combustion experiment of NO<sub>x</sub> and major gas species profiles in the MFR.
- Produced a full kinetic mechanism model of oxy-fuel combustion.

## next steps

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This project ended on December 31, 2008.

## available reports/technical papers/presentations

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