DOE/NETL's NOx Emissions Control R&D Program – Bringing Advanced Technology to the Marketplace

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Executive Summary
Advanced nitrogen oxide (NOx) emissions control technology research was an important component of the Innovations for Existing Plants (IEP) Program conducted by the U.S. Department of Energy's National Energy Technology Laboratory (DOE/NETL). The short-term research goal was to develop advanced in-furnace technologies for coal-fired power plants capable of controlling NOx emissions to a level of 0.15 pounds per million Btu heat input (lb/MMBtu) by 2007 and 0.10 lb/MMBtu by 2010, while achieving a levelized cost savings of at least 25% compared to state-of-the-art selective catalytic reduction (SCR) control technology. The program's long-term goal was to further develop a combination of advanced in-furnace and SCR control technologies that could achieve a NOx emission rate of 0.01 lb/MMBtu by 2020. The Advanced NOx Research and Development (R&D) Program employed a holistic approach to pollutant control by not only addressing NOx emissions control technologies, but also important balance-of-plant issues associated with technologies such as unburned carbon, ammonia slip, catalyst management, and sulfuric acid emissions. Due to shifts in program priorities, the IEP NOx R&D activity ended in 2007.

This paper provides an overview of DOE/NETL's advanced in-furnace NOx control R&D efforts conducted from 1999 to 2007. Currently, commercially available low-NOx burners (LNB) and SCR NOx control technologies are enabling industry compliance with today's regulatory requirements. In addition, the NOx control technologies developed by DOE/NETL will provide more cost-effective options for coal-fired power plants to comply with the ever more stringent environmental regulatory and legislative requirements expected of tomorrow. NOx emission control costs are significant and can exceed 20% of the total cost for environmental controls in today's coal-fired power plants. The capital and operating costs of SCR controls are relatively high and may not be cost-effective for older, smaller coal-fired power plants and some larger base load plants. In particular, the high capital cost for SCR can result in a levelized cost of control significantly greater than today's NOx allowance price for units with low capacity factors. Potentially more stringent state, regional, and/or federal regulations in the near future will require the retrofit of NOx controls on a greater proportion of existing coal-fired power plants. As a result, a portion of these plants could be at-risk for early retirement if more cost-effective control technologies than today's SCR are not developed. Additionally, future coal-fired plants can take advantage of these technology developments to reduce both their capital and operating cost of NOx compliance. DOE/NETL's IEP NOx R&D focused on the development of advanced in-furnace NOx controls that can approach the performance of SCR, but at significantly lower cost. The development of these advanced in-furnace technologies – such as

ultra-low NOx burner systems, oxygen-enhanced combustion, and rich reagent injection – has led to several successful commercial demonstrations.

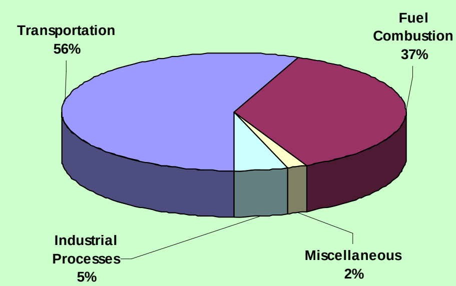
Introduction

The environmental performance of the U.S. fleet of coal-fired boilers has steadily improved since passage of the 1990 Clean Air Act Amendments (CAAA). Emissions of sulfur dioxide (SO_2), NOx, and particulate matter (PM) have been significantly reduced. However, in response to issues such as mercury, acid rain, ground-level ozone, nitrification of aquatic ecosystems, ambient fine particulate matter, and visibility impairment (regional haze), further restrictions on emissions from power plants have been implemented. Due to these environmental challenges, DOE/NETL has been carrying out a comprehensive, integrated R&D effort under its IEP Program. The overall goal of the IEP Program is to enhance the efficiency and environmental performance of the existing fleet of fossil fuel-fired power systems, which represents more than 320 gigawatts of generating capacity and applies these concepts to advanced power systems. As such, conducting R&D of advanced NOx control technologies has been an important component of the program.

Commercially available technologies for reducing NOx emissions from coal-fired boilers are enabling industrial compliance with today's regulatory requirements. For example, LNB and SCR technologies have enabled hundreds of power plants to reach mandated emission levels. However, for a number of plants these technologies cannot meet the tighter limits at acceptable cost. Without compliance options that are more cost-effective than today's SCR controls, a portion of the existing fleet of coal-fired power plants could be at-risk for early retirement or disrupting the cost and/or reliability of electricity supply in the United States.

What is NOx?

Nitrogen oxide is a general term used to describe a group of gases that contain varying amounts of nitrogen and oxygen. NOx forms at high temperatures during fossil fuel combustion. The primary sources of NOx emissions in the United States are motor vehicles, power plants, and other commercial, industrial, and residential sources that burn fossil fuels. Direct NOx emissions from these sources include nitrogen dioxide (NO_2), nitrous oxide (N_2O), and nitric oxide (NO). NOx emissions can also react with other compounds in the atmosphere to form secondary products that includes ozone (O_3), nitric acid (HNO_3), and nitrate particles. As shown in the figure below, fuel combustion sources contributed 37% of approximately 22 million tons of U.S. NOx emissions in 2002.



U.S. NOx Emissions by Source Category in 2002

This paper highlights the DOE/NETL advanced NOx emissions control R&D efforts to provide more cost-effective options for coal-fired power plants to comply with ever more stringent emission limits. Regulatory and legislative requirements have predominantly driven the need to develop NOx control technologies for existing coal-fired power plants. Beginning in the mid-1990s, first generation LNB designs enabled most power plants to adhere with the CAAA Title IV NOx emission rates that range from 0.40 to 0.86 lb/MMBtu depending on the type of boiler. The capital cost for LNB is relatively low – typically less than $20/kW. The inability of states to

> ***NOx Emissions from Coal-Fired Power Plants***
>
> Coal consists of both organic and inorganic matter. In the high-temperature environment during and after the combustion process, the components of coal and air are physically and chemically transformed into a suite of products. One of the products is NOx, which can be formed by: (1) nitrogen and oxygen in the combustion air reacting at high temperature to produce "*thermal NOx*" and (2) fuel-bound nitrogen reacting with the combustion air to produce "*fuel NOx.*" The amount of NOx formed depends on numerous factors including flame temperature, nitrogen content of the coal, combustion excess air, residence time, and degree of mixing. For most coal-fired units, thermal NOx typically represents about 20% and fuel NOx about 80% of the total NOx formed. However, for cyclones and other boilers that operate at very high temperatures, thermal NOx can be considerably higher than fuel NOx. In addition, minor amounts of NOx are formed early in the combustion process through complex interactions of molecular nitrogen with hydrocarbon free radicals to form reduced nitrogen species that are later oxidized to NOx, referred to as "*prompt NOx.*" The quantity of thermal NOx formed depends primarily on the "three T's" of combustion: temperature, time, and turbulence. Thus flame temperature, the residence time at temperature, and the degree of fuel/air mixing, along with the nitrogen content of the coal and the quantity of excess air used for combustion, determine NOx levels in the flue gas.

achieve attainment with the one-hour average ozone National Ambient Air Quality Standards (NAAQS) led EPA to further regulate NOx emissions from power plants in the Northeast, Midwest, and Southeast United States through the NOx state implementation plan rule (known as the NOx SIP call rule) beginning in 2003/04. Under the NOx SIP call rule, states have implemented power plant NOx reductions through a market-based NOx Allowance Cap-and-Trade Program that is equivalent to an overall average emission rate of 0.15 lb/MMBtu during the five-month summer ozone season. However, the first generation LNB was not capable of meeting this level of performance. As a result, many of the larger, base-loaded power plants had to install SCR to meet the NOx emission caps. The capital cost for SCR is relatively high – typically greater than $100/kW – and would be prohibitive for smaller, intermittent-loaded power plants. The most recent regulatory driver is the Clean Air Interstate Rule (CAIR), which impacts 28 eastern states and the District of Columbia thru a two-phase cap-and-trade program. Beginning in 2009, the CAIR NOx emission caps are based on an equivalent emission rate of 0.15 lb/MMBtu for Phase I, but beginning in 2015, the cap decreases to 0.125 lb/MMBtu for Phase II. Another regulatory driver, the Clean Air Visibility Rule (CAVR), could impose additional NOx controls on plants in the Western United States under Best Available Retrofit Technology (BART) requirements. The continued ratcheting down of NOx emissions by new regulations will require additional power plants to reduce their emission rates below 0.15 lb/MMBtu. To meet these requirements, power producers will need to retrofit existing boilers with additional NOx control technologies, some of which will adversely impact plant efficiency and performance. The challenge was to develop cost-effective NOx control technologies for the smaller, older, less efficient facilities that are not candidates for the current state-of-the-art SCR controls, because of space constraints or the reluctance of owners to invest significant capital in aging plants. These facilities, with a generating capacity of 300 MW or less, comprise 65% of the boilers – representing 27% of total U.S. generation capacity – and have an average age of 46 years, in comparison to the remaining fleet's average age of 30 years.[1] For these reasons, the IEP NOx R&D activity focused on the development of next generation, advanced LNB and other in-furnace control technologies that can achieve a NOx emission rate of less than 0.15 lb/MMBtu at a capital cost comparable to first generation LNB.

Advanced NOx Control Technology R&D Activities

DOE/NETL has been at the forefront of conducting NOx control technology R&D for coal-fired power plants. The success of the Title IV Acid Rain Program NOx reductions in the 1990s can be largely attributed to the utility industry's adoption of LNB technology. The LNB that are currently installed in 75% of the nation's coal-fired power plants are a direct result of the DOE/NETL Clean Coal Technology Program's government–industry partnerships.[2] From 1999 to 2007, advanced NOx emissions control technology R&D was an important component of the IEP Program.[3] The benefits of this program will be realized by both the existing fleet and new capacity as the targeted NOx control technologies are adopted. The IEP R&D effort focused primarily on developing systems capable of controlling NOx emissions at a level of 0.15 lb/MMBtu by 2007 and 0.10 lb/MMBtu by 2010, while achieving a levelized cost savings of at least 25% compared to state-of-the-art SCR control technology. The program's long-range goal was to further develop a combination of advanced in-furnace and SCR control technologies to achieve a NOx emission rate of 0.01 lb/MMBtu by 2020. The development of these advanced NOx control technologies was necessary to ensure the U.S. power generation industry can continue to comply with the Federal NOx emission tonnage caps as the size of the coal-fired power plant fleet continues to grow. According to the latest projections from the Energy Information Administration (EIA), coal-fired power plant capacity is expected to increase over 340 GW by 2020 and 450 GW by 2030.[4] The technologies were developed to negate balance-of-plant impacts, apply to a wide range of boiler types and configurations, and maintain performance over a wide range of feed coals and operating conditions. The research also provided an improved understanding of the advanced technologies' impact on related issues such as unburned carbon, waterwall wastage, and mercury speciation and capture. The IEP portfolio of NOx control technology R&D projects encompassed laboratory studies, modeling, and pre-commercial full-scale testing. The success of the projects was intimately tied to key collaborations and partnerships between industry; federal, state, and local agencies; and the academic and research communities.

The IEP Program's advanced NOx control technology R&D projects can be grouped into five research areas:

- Next Generation Low NOx Burners
- Rich Reagent Injection
- Oxygen-Enhanced Combustion
- Novel Enhanced Combustion
- SCR Optimization

The following sections include brief summaries of the R&D completed in each of these areas.

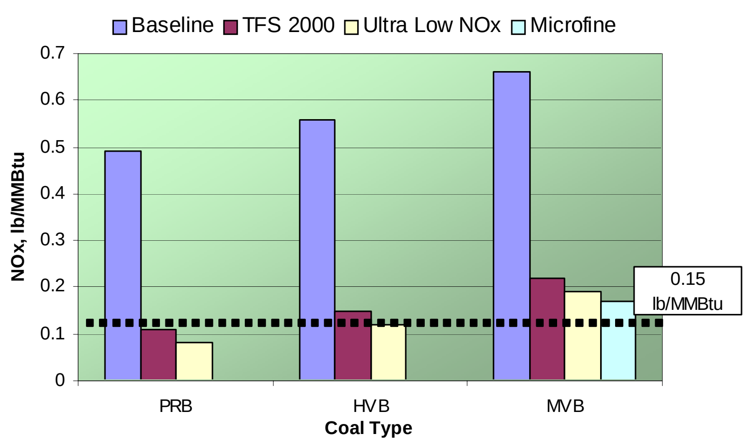
Next Generation Low NOx Burners

As aforementioned, first generation LNB enabled most coal-fired power plants to comply with the CAAA Title IV requiring NOx emission rates to range from 0.40 lb/MMBtu to 0.86 lb/MMBtu depending on the type of boiler. NETL research has since focused on development of the next generation of advanced LNB that are based on the second and third generation of commercially-available LNB, but enhanced to achieve a NOx emission rate of less than 0.15 lb/MMBtu.

Low NOx Firing System for Tangential Boilers

From 2000 to 2002, ALSTOM Power, Inc. conducted a comprehensive pilot-scale study to improve the performance of their TFS 2000™ low NOx firing system used for tangential-fired boilers.[5,6] At the time, typical NOx emission rates for tangential-fired boilers ranged from 0.18 lb/MMBtu to 0.24 lb/MMBtu for bituminous coals and 0.10 lb/MMBtu to 0.13 lb/MMBtu for subbituminous coals. The project's goal was to achieve a NOx emission rate of less than 0.15 lb/MMBtu when firing bituminous coal and 0.10 lb/MMBtu for subbituminous coal at a cost at least 25% less than SCR technology. Pilot-scale testing was conducted using three coal types, including a very reactive subbituminous Powder River Basin coal (PRB); a moderately reactive, high volatile Midwestern bituminous coal (HVB); and a less reactive, medium volatile eastern bituminous coal (MVB). The technologies evaluated during the pilot-scale testing consisted of finer coal grinding, oxidative pyrolysis burners, windbox auxiliary air optimization, and various burner zone firing arrangements in concert with overfire air (OFA). Figure 1 shows the most reactive coal, PRB, resulted in the lowest NOx emission rate, followed by the moderate and less reactive HVB and MVB coals, respectively. The combination of firing system modifications resulting in the lowest NOx emissions is referred to as the Ultra Low NOx (ULN) integrated system. ALSTOM has since applied many of the firing system components developed in this project to the TFS 2000 firing system, resulting in improved NOx emissions without significantly affecting the unburned carbon levels.

Figure 1 - ALSTOM Pilot-Scale Test Results



From 2005 to 2006, ALSTOM conducted a follow-up project to further develop an enhanced low NOx coal nozzle tip for the TFS 2000 firing system.[7] The objective was to optimize combustion via control of near-burner time/temperature history, turbulence, and stoichiometry. The project included CFD modeling and large pilot-scale testing. An illustration of the baseline low-NOx concentric firing system (LNCFS) P2 coal nozzle tip is shown in Figure 2. ALSTOM conducted pilot-scale testing of improved coal nozzle tips firing an Illinois No. 6 high volatile bituminous coal. Figure 3 presents the NOx reduction achieved for each of the tested coal nozzle tips relative to the baseline LNCFS P2 tip. It is apparent that the coal nozzle tip design has a significant impact because some of the new tip designs achieved NOx emissions 40% lower than those achieved with the baseline P2 tip in the pilot-scale tests. A final series of pilot-scale combustion performance evaluations showed similar NOx reductions on a subbituminous fuel, and slightly lower percentage reductions on a western bituminous fuel.

Figure 2 – ALSTOM LNCFS P2 Coal Nozzle Tip

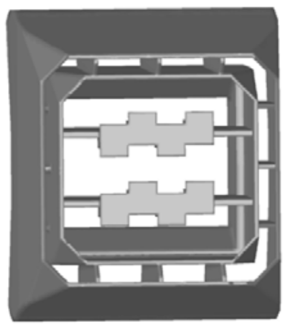
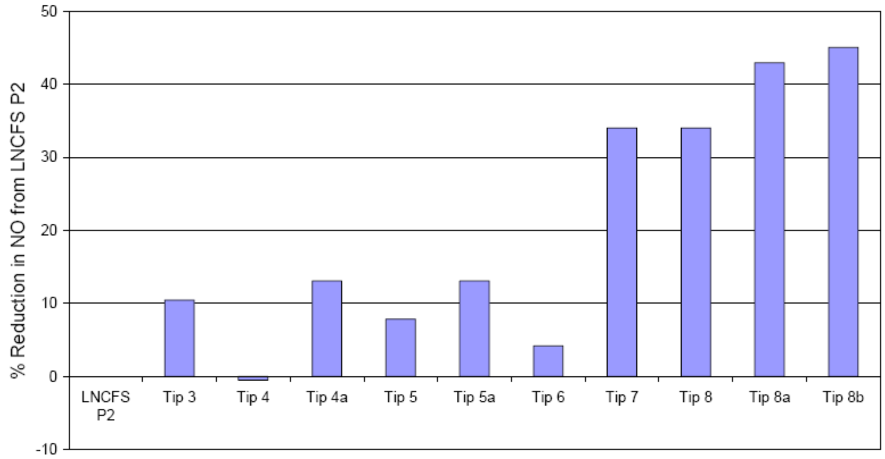


Figure 3 – ALSTOM Pilot-Scale Low NOx Burner Tip Performance Results

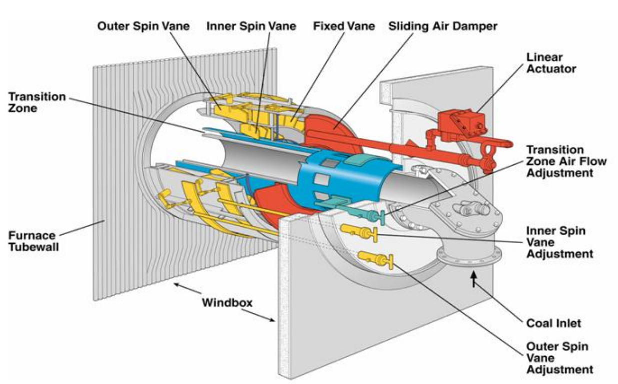


Integrated Low NOx Burners and SNCR for Wall-Fired Boilers

From 2000 to 2005, McDermott Technology, Inc. (MTI), the Babcock & Wilcox Company (B&W), and Fuel Tech teamed together to investigate an integrated solution for NOx control.[8,9,10] The system was comprised of B&W's DRB-4Z™ LNB technology (Figure 4) and Fuel Tech's NOxOUT®, a urea-based SNCR technology. Large pilot-scale testing was conducted on a wide range of coals including: PRB, a subbituminous coal; Pittsburgh No. 8, a high volatile bituminous coal; and Middle Kittanning, a medium volatile bituminous coal. As shown in Figure 5, the DRB-4Z™ burner alone without air staging achieved NOx emissions of 0.26 lb/MMBtu, 0.30 lb/MMBtu, and 0.40 lb/MMBtu for the PRB coal, Pittsburgh No. 8, and Middle Kittanning coal, respectively.

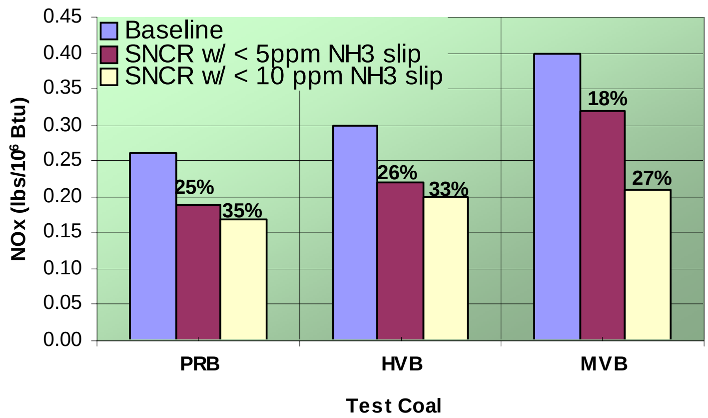
At full load, the baseline DRB-4Z™ NOx levels were reduced by the SNCR system (configured with wall injectors only) to 0.19 lb/MMBtu (25% reduction) for PRB, 0.22 lb/MMBtu (26% reduction) for Pittsburgh No. 8, and 0.32 lb/MMBtu (18% reduction) for Middle Kittanning coal. These data indicate that an additional 25% NOx reduction is feasible using SNCR while firing PRB and high volatile, eastern coals with a baseline NOx of 0.2 to 0.3 lb/MMBtu when the NH_3 slip is limited to less than 5 ppm. Higher NOx reductions were possible when the ammonia slip ranged from 5 ppm to 10 ppm. For units firing coals with lower volatile content, such as Middle Kittanning, the higher boiler gas temperatures could limit the SNCR NOx reduction from 15% to 20%.

Figure 4 – B&W's DRB-4Z Low NOx Burner



In 2004, additional testing was conducted using a PRB coal to further investigate more favorable conditions of the layered technology, including the use of the DRB-4Z burner, staged combustion with OFA, and SNCR with a convective pass multiple nozzle lance (MNL) in front of the superheater tubes. Urea injection at this location offered the following advantages: 1) lower injection temperature; 2) improved urea/flue gas mixing; and 3) optimum urea particle size. Using this arrangement, the baseline NOx emission rate (0.094 lb/MMBtu to 0.162 lb/MMBtu depending on burner stoichiometry) with the DRB-4Z burner and OFA was reduced approximately 25% (0.071 lb/MMBtu to 0.124 lb/MMBtu) using the SNCR with MNL and an ammonia slip of 6 ppm.

Figure 5 - B&W DRB-4Z and SNCR Full-Load Test Results



NOx Control: Combustion Modification Technologies

NOx reduction technologies can be grouped into two broad categories: combustion modifications and post-combustion processes. Some of the more important combustion modification technologies are briefly discussed below.

Low-NOx Burners — LNB are designed to control the mixing of fuel and air to achieve what amounts to staged combustion. This staged combustion reduces both flame temperature and oxygen concentration during some phases of combustion, which reduces both thermal NOx and fuel NOx production.

Overfire Air — OFA is air that is injected into the furnace above the normal combustion zone. Generally when OFA is employed, the burners are operated at a lower than normal air-to-fuel ratio, which reduces NOx formation. OFA, which is frequently used in conjunction with LNB, completes the combustion process at a lower temperature.

Reburning — In the reburning process, part of the boiler fuel input (typically 10-25%) is added in a separate reburn zone. The fuel rich reducing conditions in this zone lead to the reduction of NOx formed in the normal combustion zone. OFA is injected above the reburn zone to complete combustion. Thus, there are three zones in respect to reburn in the furnace: (1) a combustion zone with an approximately normal air-to-fuel ratio; (2) a reburn zone, where added fuel results in a fuel-rich condition; and (3) a burnout zone, where OFA completes the combustion. Coal, oil, or gas can be used as the reburn fuel.

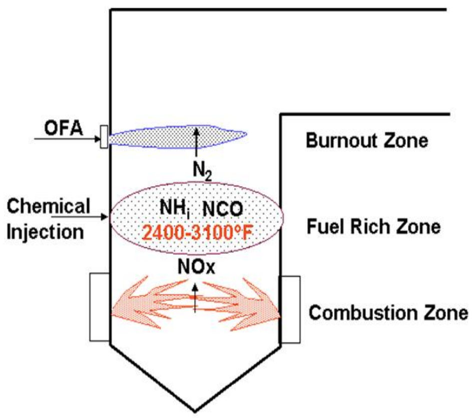
Rich Reagent Injection/Advanced Layered Technology Approach

Developed by Reaction Engineering International (REI) and the Electric Power Research Institute, Rich Reagent Injection (RRI) uses a nitrogen-containing additive, such as ammonia or urea, to non-catalytically reduce NOx in the lower furnace – similar to SNCR. A schematic of the RRI process is shown in Figure 6. Combining RRI with OFA and SNCR is known as the Advanced Layered Technology Approach, or "ALTA."

RRI for Cyclone Burners

From 2000 to 2006, Reaction Engineering International (REI) conducted optimization studies of the RRI process for NOx reduction on cyclone burners.[11,12] Cyclone burners create an intense flame that melts the ash to form slag. The high temperature generated by this burner results in relatively high, uncontrolled NOx emissions, typically exceeding 1.2 lb/MMBtu. Initial full-scale field-testing of RRI was successfully completed at Conectiv's 138 MW B.L. England Unit 1 and AmerenUE's 500 MW Sioux Unit 1 in 2000 and 2002, respectively.

Figure 6 - Rich Reagent Injection Process Schematic



At Conectiv's B.L. England Unit 1, prior installation of OFA and SNCR had reduced uncontrolled NOx emissions from 1.2 lb/MMBtu to 0.35 lb/MMBtu. REI utilized CFD combustion simulation software to design the RRI system. A summary of the RRI test results at B.L. England Unit 1 are shown in Figure 7. Field-testing confirmed modeling predictions and demonstrated that the RRI system alone could achieve a 25% to 30% NOx reduction beyond OFA levels with less than 1 ppm ammonia slip. Also, the inclusion of SNCR could achieve an additional 35% NOx reduction to 0.25 lb/MMBtu with less than 5 ppm NH_3 slip.

The objective of the testing at AmerenUE's Sioux Unit 1 was to determine whether similar performance could be obtained with RRI in a significantly larger unit. A summary of the RRI test results at Sioux Unit 1 are shown in Figure 8. The field test results were found to be consistent with the CFD model predictions. Both showed that NOx reductions of 30% from full load baseline emissions of 0.38 lb/MMBtu with OFA to 0.27 lb/MMBtu were achievable with RRI. These reductions were achieved with no predicted or measurable ammonia slip. During this 2002 test program SNCR testing was not successfully carried out in combination with RRI and OFA. However, it prompted the question of what NOx emissions might be achievable in this unit when optimizing the three technologies. Although the target emissions of 0.15 lb/MMBtu were ambitious for this style of burner, these results are significant when compared to the Title IV NOx limit of 0.86 lb/MMBtu for cyclone-fired boilers.

In 2005, REI conducted additional CFD modeling and full-scale field testing to optimize the ALTA technology at the Sioux Station.[13,14] Sioux Station typically burns an 80/20 blend of Powder River Basin subbituminous coal and Illinois No. 6 bituminous coal. Parametric testing was also conducted with 60/40 and 0/100 blends. The testing also evaluated process impacts on balance-of-plant issues, such as the amount of unburned carbon in the ash, slag tapping, waterwall corrosion, ammonia slip, and heat distribution.

Prior to the 2005 field testing, REI used CFD modeling to optimize the number and location of the urea reagent injection locations for both the RRI and SNCR systems. Previous RRI testing in 2002 used 20 RRI injectors. Based on the CFD modeling results, eight additional RRI injectors and 14 SNCR injectors were installed for the 2005 field testing. The majority of field testing was conducted using the 80/20 coal blend at 480 MW under two levels of air staging with OFA, resulting in baseline NOx emission rates of 0.25 lb/MMBtu with normal air staging and 0.20 lb/MMBtu with deep staging.

Figure 7 - RRI Test Results at B.L. England Unit 1 in 2000

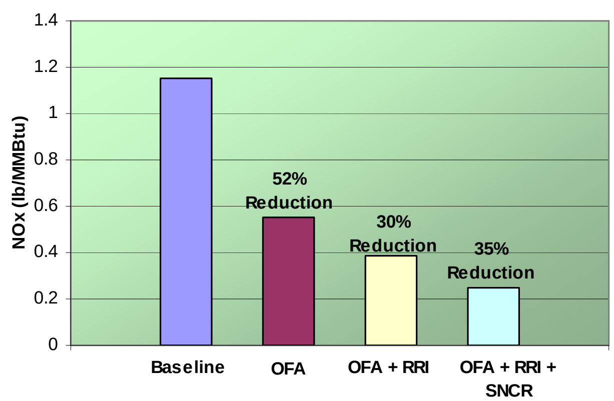
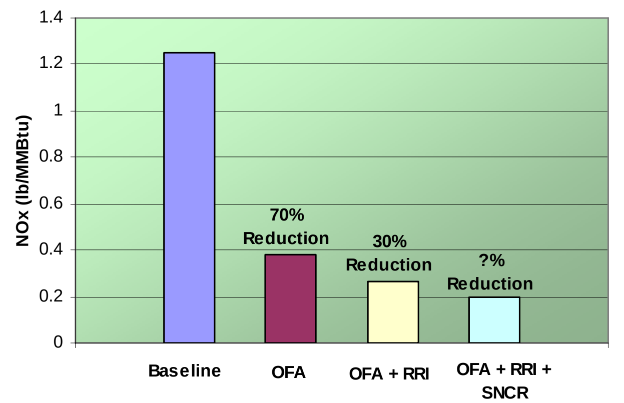
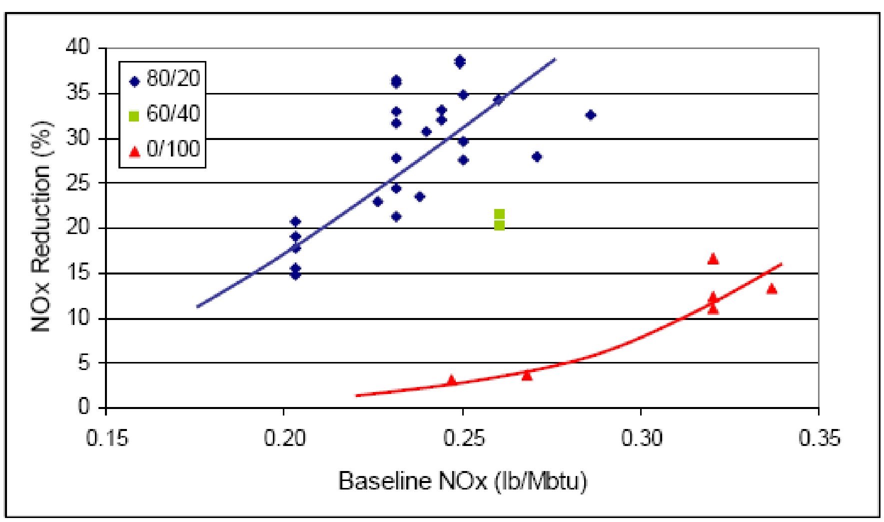


Figure 8 - RRI Test Results at Sioux Unit 1 in 2002



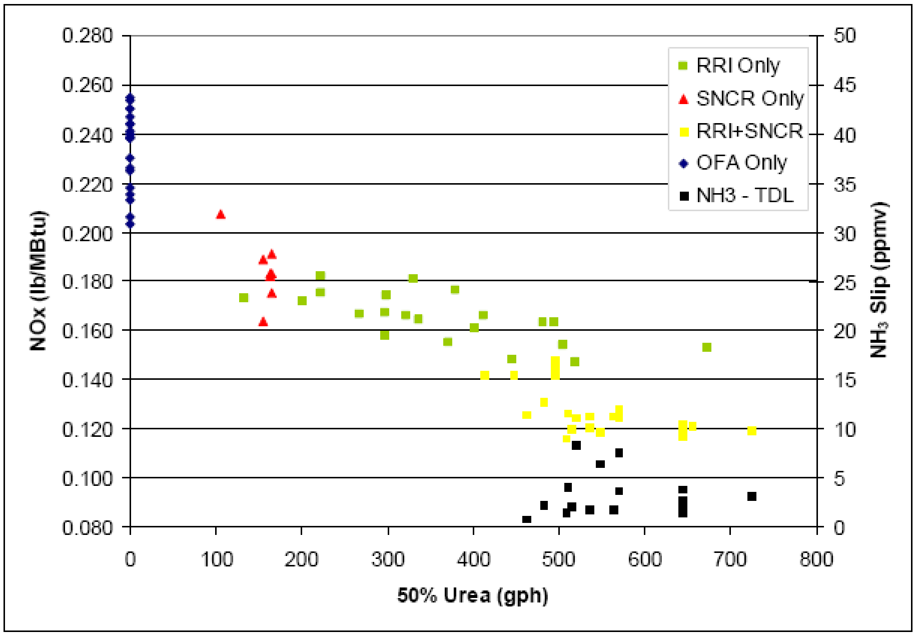
REI conducted parametric testing for the complete ALTA configuration and with RRI and SNCR separated. Using the 80/20 coal blend, the NOx emission rate achieved with ALTA averaged 0.12 lb/MMBtu with an ammonia slip less than 5 ppm. For the 60/40 and 0/100 coal blends, the ALTA configuration achieved NOx emission rates of 0.15 lb/MMBtu and 0.165 lb/MMBtu, respectively. RRI alone reduced NOx emissions to between 0.18 lb/MMBtu and 0.15 lb/MMBtu with less than 1 ppm ammonia slip when firing the typical 80/20 coal blend. The additional NOx reduction during RRI parametric testing for the three coal blends and varying levels of baseline NOx is shown in Figure 9.

Figure 9 - REI Parametric Test Results with RRI at Sioux Unit 1 in 2005



Parametric testing with SNCR alone demonstrated average NOx emissions of 0.156 and 0.165 lb/MMBtu for the 80/20 and 0/100 coal blends, respectively, at less than 5 ppm ammonia slip under most test conditions. Figure 10 shows the NOx emission rate for various test conditions using the 80/20 coal blend: baseline, using only OFA; OFA with RRI; OFA with SNCR; and the complete ALTA configuration using OFA, RRI, and SNCR. Also, Figure 10 includes the measured levels of ammonia slip at various urea feed rates.

Figure 10 – REI Parametric Test Results with OFA, RRI, SNCR, and ALTA at Sioux Unit 1 in 2005



Following the 2005 ALTA tests at Sioux Unit 1, Ameren chose to discontinue engineering efforts towards planned SCR deployment at the two Sioux units. Rather, commercial ALTA systems were installed on both units during winter 2006/2007. The ALTA systems were operational for full-time use on both units prior to the 2007 ozone season.[15]

<u>Pilot-Scale Testing of ALTA NOx Control for Pulverized Coal-Fired Boilers</u>

From 2005 to 2006, REI began developing and verifying performance of the ALTA NOx control technology for pulverized coal-fired (PC) boiler applications in order to achieve an emission rate of less than 0.15 lb/MMBtu.[16] The key challenge in implementing ALTA for PC boilers was the lack of a homogeneous, fuel-rich environment in the lower furnace due to the stratified combustion common with traditional LNB. To overcome this demanding condition, new LNB were designed for enhanced near-burner combustion, which generated greater homogeneity of the combustion products in the furnace. Not only does this create optimal conditions for combustion-related NOx control, it also results in a stoichiometry and temperature distribution above the burners that is ideal for the chemistry involved in RRI.

REI conducted CFD modeling and pilot-scale testing to optimize the near-burner combustion system and reagent injection. The pilot-scale testing was conducted by the University of Utah using a western bituminous coal on a four million Btu/hr coal combustion furnace. Test results showed that an ALTA burner was able to produce a fuel-rich homogeneous environment favorable to RRI near the burner, whereas the fuel and air from a standard LNB remained stratified for some distance downstream of the burner.

Test results suggested the best use of the ALTA burner with RRI for NOx control appeared to be in applications with relatively short residence times and with burner stoichiometry ratios (BSR) less than 0.85. For long residence times and staged operation, the standard LNB produced NOx similar to or lower than from RRI combined with the ALTA burner. However, for shorter times typical of burner to OFA residence times in utility boilers, RRI with the ALTA burner showed lower NOx levels for a BSR less than 0.85.

For example, at a BSR of 0.75 and RRI stoichiometry ratio (SR) of 2.0, NO_x concentrations were 0.09 lb/MMBtu with the ALTA burner (a 30% reduction from the no-RRI baseline) and 0.107 lb/MMBtu with the standard LNB (a 2% reduction from the no-RRI baseline). For short residence times and BSR greater than 0.85, the standard LNB consistently produced lower NOx than the ALTA burner with RRI due to the significant increase in NOx from the ALTA burner before reagent injection.

CFD simulations were performed to predict the effectiveness of deep staging using the ALTA burner combined with RRI on a 180 MW, front-wall fired, PC boiler. This furnace had been previously modeled to evaluate the effectiveness of RRI for NO_x control. The results of that past effort suggested no more than 10% reduction in NOx emissions. For this investigation, the existing, dual-register, LNB were replaced by ALTA burners, the OFA ports were modified, and the furnace was staged more deeply at a BSR of 0.80. Modeling results showed the ALTA burner was effective in improving the homogeneous, fuel-rich environment in the lower furnace. When four levels of RRI injection were used with a total SR of 2.0, modeling predicted an average NOx concentration of 0.185 lb/MMBtu, a 37% reduction in NO_x from baseline. This included a 7% increase in baseline NOx with the ALTA burners (no RRI) followed by a 41% NOx reduction from the ALTA burner baseline with RRI. Combining this performance with the 25%-35% NOx reduction expected from commercially-available SNCR systems suggests that overall NOx emissions with the full ALTA system could be in the 0.12-0.14 lb/MMBtu range for this type of boiler. These pilot-scale tests and full-scale model predictions indicate that ALTA could be an effective NOx control technology for PC-fired boilers, as well as cyclone-fired boilers.

Oxygen-Enhanced Combustion

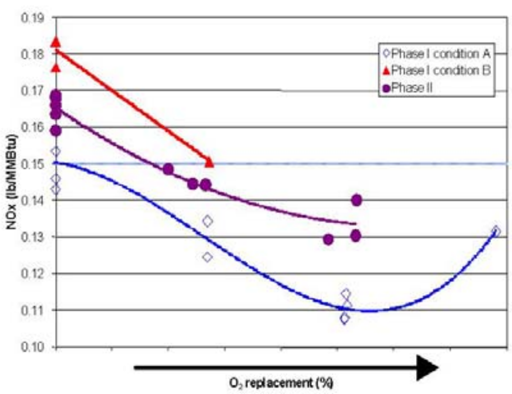
Replacing a small fraction – less than 10% – of combustion air with pure oxygen can enhance the NOx reduction achievable with LNB and OFA, while minimizing the production of unburned carbon. The oxygen-enhanced combustion reduces NOx formation due to the creation of a more fuel-rich condition and increased flame temperature at the burner, which drives the combustion reactions toward formation of molecular nitrogen (N_2), rather than NOx. The increased flame temperature and higher oxygen concentration also increases coal devolatilization rates, resulting in reduced unburned carbon.

Oxygen-Enhanced Combustion for PC-Fired Boilers

From 2000 to 2004, Praxair, Inc. and its partners tested oxygen-enhanced combustion for PC-fired boilers.[17,18] The technology was recognized as a finalist for Chemical Engineering Magazine's Kirkpatrick Award for Chemical Engineering Achievement in 2005. Testing was conducted using ALSTOM Power's large pilot-scale combustion facility. The experiments were designed to demonstrate that the concept of oxygen-enhanced low NOx combustion could meet the emissions target of 0.15 lb/MMBtu with minimal impact on carbon monoxide (CO) emissions and furnace performance. A commercially available ALSTOM wall-fired LNB was fired at 24 MMBtu/hr for these tests.

Figure 11 presents the results from a three-phase test of different coals at baseline conditions and varying levels of oxygen addition. Illinois No. 6 bituminous coal was used during the initial Phase I-A tests, during which the baseline emission rate of approximately 0.15 lb/MMBtu was reduced with oxygen addition to approximately 0.11 lb/MMBtu. These tests were performed to shake down the furnace and obtain baseline NOx data for this facility, burner, and coal combination. A series of experiments were then performed to evaluate the effect of oxygen addition on NOx emissions. In Phase I-B, an eastern bituminous coal, Mingo Logan, was used to evaluate the effects of a lower volatile coal and oxygen addition method. During Phase II, the selected experiments were repeated with the Illinois No. 6 coal. As shown in Figure 11, data from the Illinois No. 6 experiments show that even when the baseline emissions are very low, oxygen addition can drive the NOx emissions even lower. The overall data further show that the reductions are relatively independent of the initial NOx concentration.

Figure 11 - Praxair Oxygen-Enhanced Combustion Test Results



Subsequent full-scale testing of oxygen-enhanced combustion at two utility boilers, City Utilities' James River Power Station and Northeast Utilities' Mount Tom Generating Station, has demonstrated the viability of this technology. At the 44 MW James River Unit 3, a NOx reduction of approximately 40% was achieved burning a blend of PRB and bituminous coal; while testing conducted at the 146 MW bituminous coal-fired Mount Tom Unit 1 lowered the NOx emission rate to 0.20 lb/MMBtu from 0.26 lb/MMBtu – equivalent to a 32% to 47% reduction from the 0.38 lb/MMBtu baseline.

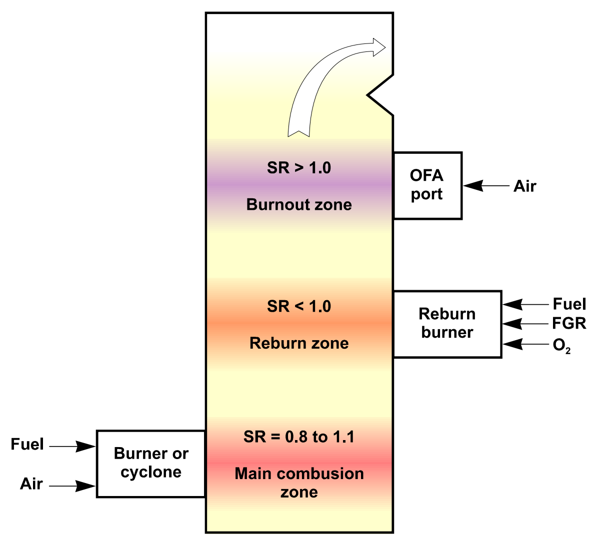
Advanced In-Furnace NOx Control for Wall- and Cyclone-Fired Boilers

From 2005 to 2007, B&W began development and demonstration of an advanced NOx control technology capable of achieving an emission rate of 0.10 lb/MMBtu while burning high volatile bituminous coal for both wall- and cyclone-fired boilers. The technology is based on a "layered" strategy that combines deep air staging using OFA, continuous corrosion monitoring, advanced combustion control enhancements, and a proprietary oxygen injection-based combustion technique. Figure 12 shows a boiler schematic of the layered process, which includes three zones: main combustion, reburn, and burn-out. The SR in the main combustion zone is varied from 0.7 to 1.1. The reburn zone features oxygen-enhanced combustion of the re-burn fuel and flue gas recirculation (FGR), at a SR of less than 1. The burnout zone utilizes OFA to achieve complete combustion, at a SR greater than 1.

At B&W's test facility, wall- and cyclone-fired pilot-scale testing was conducted at a firing rate of five million Btu/hr to evaluate the oxygen injection process. Oxygen was injected at various locations in the combustion zone and data gathered by optical sensors were used to optimize the combustion process. Corrosion monitoring sensors assessed potential waterwall wastage that could occur under severe operating conditions. The testing also evaluated the effects of oxidizer composition, the extent of deep air staging, and the overall excess oxygen level on combustion behavior, heat transfer characteristics, and emissions levels.

A scaled down version of B&W's DRB-4Z low NOx burner coupled with OFA was used for the wall-fired boiler pilot-scale testing with a baseline NOx emission rate of 0.324 lb/MMBtu while burning a high volatile bituminous coal. The NOx emission rate was reduced to 0.19 lb/MMBtu (41% reduction) with the use of OFA, a reburn burner, and 10% oxygen injection.

Figure 12 – Schematic of B&W's Layered NOx Combustion Control Concept



B&W also conducted pilot-scale testing of a cyclone-fired boiler configuration burning a subbituminous PRB coal and a high volatile Eastern bituminous coal. The baseline NOx emission rate of 1.04 lb/MMBtu was reduced to approximately 0.15 lb/MMBtu (85% reduction) with the use of OFA and reduced even further to 0.12 lb/MMBtu (additional 20% reduction) with the use of OFA and 5% oxygen injection. In the case of firing the Eastern bituminous coal in the optimum cyclone arrangement, a NOx level of 0.146 lb/MMBtu was achieved, a reduction of 88% from the baseline value of 1.24 lb/MMBtu.

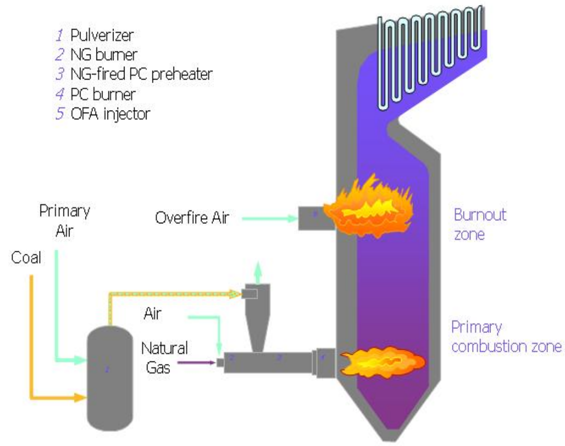
Novel Enhanced Combustion

DOE/NETL's Advanced NOx Control R&D Program also included pilot-scale testing of two novel NOx control technologies – METHANE De-NOx and Dense Phase Reburn.

Methane De-NOx

From 2000 to 2005, the Gas Technology Institute (GTI) conducted pilot-scale testing of the METHANE de-NOX® NOx reduction process.[19,20] The METHANE de-NOx technology combines several NOx reduction strategies into an integrated system, including a novel burner design using natural gas-fired coal preheating and internal and external combustion staging in the primary and secondary combustion zones. Figure 13 provides a schematic of the METHANE de-NOx process. A natural gas-fired coal preheat combustor preheats the coal to elevated temperatures (up to 1500°F). The preheating releases coal volatiles, including fuel bound nitrogen compounds, into a controlled reducing environment, which reduces the coal-derived nitrogen compounds to molecular N_2. The preheated coal is converted to a mixture of char and gaseous volatile matter, which is then fired through the main burner into the boiler furnace. The quantity of natural gas fuel required for coal preheating is in the range of 3% to 5% of the total burner heat input.

Figure 13 - GTI METHANE de-NOx Process Schematic



Using a PRB coal, initial pilot-scale testing demonstrated that the coal preheating process has a significant effect on final NOx formation in the coal burner. Based on CFD modeling, modifications to both the pilot-scale gas-fired combustor and the coal burner led to NOx levels below 100 ppmv with CO in the range of 35-112 ppm_v and without any furnace air staging.

Initial pilot-scale testing with a bituminous coal resulted in deposition and plugging by caked material on the inside of the gas combustor. A series of modifications to the combustor configuration and operation were subsequently tested. One of these approaches was successful in sustaining operation up to 85% of the targeted fuel input with the bituminous coal, although some deposition and unburned carbon issues remained. NOx levels were approximately 100 ppm_v with 6% oxygen in the flue gas at the furnace exit.

Dense Phase Reburn Combustion System

From 2002 to 2003, Wiley & Associates conducted full-scale field-testing of a Dense Phase Reburn Combustion System (DPRCS).[21] Using dense phase injection of micronized coal at a particle size of 80% minus 325 mesh, DPRCS controls combustion stoichiometry from the bottom to the top of the furnace. The testing was conducted at CP&L's 150 MW Cape Fear Unit No. 5 that burns a low sulfur bituminous coal in a tangentially-fired furnace. The DPRCS was used in conjunction with Mobotec's rotating OFA (ROFA™) system. Baseline NOx emission rates with the ROFA system ranged from 0.17 lb/MMBtu to 0.26 lb/MMBtu. During DPRCS testing, the micronized coal feed rate averaged 13% of total fuel input and the unit operated at approximately 75% capacity. Tests were conducted at four elevations with micronized coal injection, ranging from the lower furnace to below the ROFA injection level in the upper furnace. Depending on injection location, NOx control performance during the DPRCS testing was mixed. Injection below the ROFA resulted in a 14% increase in the NOx emission rate; while various combinations of DPRCS injection points from the bottom to top coal burner elevations resulted in NOx emission rate reductions ranging from 10% to 30%.

NOx Control: Post-Combustion Technologies

NOx reduction technologies can be grouped into two broad categories: combustion modifications and post-combustion processes. Some of the more important post-combustion technologies are briefly discussed below.

Selective Catalytic Reduction — In SCR, a catalyst vessel is installed downstream of the furnace. Ammonia (NH_3) is injected into the flue gas before it passes over the fixed-bed catalyst. The catalyst promotes a reaction between NOx and NH_3 to form nitrogen and water vapor. NOx reductions as high as 90% are achievable, but careful design and operation, such as control of the reagent dosage and assuring good mixing, are necessary to keep NH_3 emissions (referred to as NH_3 slip) to a concentration of a few ppm.

Selective Noncatalytic Reduction — In SNCR, a reducing agent (typically NH_3 or urea) is injected into the furnace above the combustion zone, where it reacts with NOx as in the case of SCR. Two critical factors that apply to SNCR are sufficient residence time in the appropriate temperature range and uniform distribution and mixing of the reducing agent across the full furnace cross section.

SCR Optimization

In addition to the advanced in-furnace NOx control technology projects discussed above, DOE/NETL also initiated an R&D effort to optimize the performance of SCR controls to support the program's long-range goal to achieve a NOx emission rate of 0.01 lb/MMBtu by 2020 through the combination of advanced in-furnace and advanced SCR concepts.

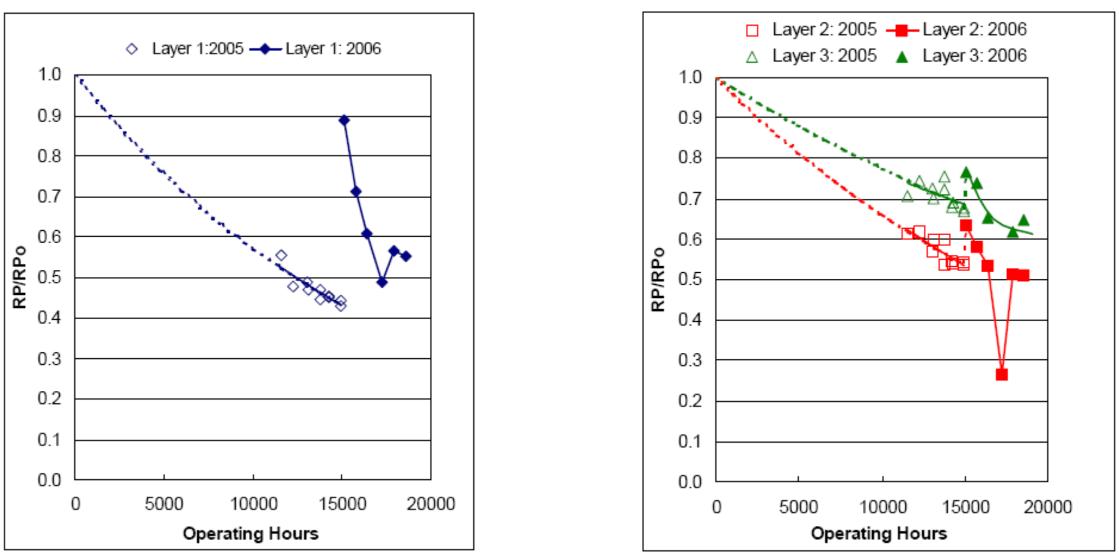
SCR Catalyst Performance Monitoring

While SCR has proven to be an effective NOx control technology, catalyst deactivation and blockage requires a comprehensive catalyst management program to guide the addition and replacement of catalyst. This, in turn, requires access to the SCR reactor to obtain physical catalyst samples for laboratory testing of activity. Access to the reactor to obtain physical samples will become more difficult as the SCR systems move from ozone season operation to year-round operation. To facilitate the catalyst management process, Fossil Energy Research Corporation (FERCo) developed KNOxcheck™ – an *in situ* SCR catalyst deactivation measurement system.[22,23] The KNOxcheck™ system collects real-time SCR activity data that can be incorporated into catalyst management software. With this real-time data, power plants can evaluate the impacts of fuel and operating parameter changes on the SCR and better plan for catalyst additions and replacements.

The KNOxcheck™ system in effect converts small sections of the actual catalyst layers to "laboratory" sections, at which the *in situ* activity is measured. Supplemental ammonia is injected and the NOx reduction across this small section of catalyst measured. SCR reactor performance is determined by a parameter called reactor potential. KNOxcheck™ provides a direct measurement of the reactor potential, which is the product of the catalyst activity times the total amount of catalyst surface area per unit flue gas flow.

FERCo conducted KNOxcheck™ tests during the 2005 and 2006 ozone seasons on the SCR installed at Southern Company's 700 MW, tangentially-fired Gorgas Unit 10 that burns bituminous coal. The Gorgas Unit 10 SCR began operation in 2002 and is designed with two parallel reactors that were originally equipped with three extruded honeycomb catalyst layers and a spare layer. Prior to the start of the 2006 ozone season, the top honeycomb catalyst layer in each reactor was relocated to the bottom spare position of the opposite reactor and was replaced with a new layer of plate catalyst. FERCo installed a single KNOxcheck™ measurement location on each of the catalyst layers in the B reactor for the 2005 ozone season. Additional measurement locations were added to the B reactor for the 2006 ozone season. Catalyst activity measurements were performed throughout the 2005 and 2006 ozone seasons, which are shown in Figure 14 - the left graph is for the top catalyst layer and the one on the right is for the second and third layers.

Figure 14 – FERCo In Situ SCR Reactor Potential Measurements for 2005-06



The data for the 2005 ozone season (open symbols) shows that the top layer deactivates at a higher rate than the second and third layers (as might be expected for a bituminous coal where arsenic is the primary catalyst poison). The large increase in reactor potential for layer 1 between the end of the 2005 and start of the 2006 ozone season is due to the replacement of the extruded honeycomb catalyst with new plate catalyst. The large decrease in reactor potential for all layers at the start of the 2006 ozone season was due to a sootblower malfunction resulting in blockage and loss of active catalyst surface (not necessarily catalyst deactivation). In fact, part way through the 2006 ozone season, the reactor potential increases on layers 1 and 2 after the sootblowers were repaired.

The *in situ* KNOxcheck™ measurements were in good agreement with *ex situ* laboratory measurements from catalyst samples removed from the reactor at the end of the 2005 and 2006 ozone season. Southern Company extended the use of the KNOxcheck™ system at Gorgas through the 2007 ozone season and plans to use it again during the 2008 ozone season. As a result of this successful demonstration, FERCo has since begun commercial sales of the KNOxcheck™ system.

From R&D to Commercial Application

As the following examples illustrate, the market potential for DOE/NETL's advanced NOx control technology R&D projects has begun to be realized:

- Ameren installed REI's ALTA NOx control technology on both 500 MW PRB-fired cyclone units at the Sioux Station in 2007. Initial performance testing on Unit 2 showed NOx emission rates of approximately 0.13 lb/MMBtu at 470 MW and 0.09 lb/MMBtu at 330 MW.
- In September 2007, Fuel Tech, Inc. announced it had received an order to supply SNCR and RRI for two small coal-fired units located in the Midwest.
- Many of the components developed in ALSTOM's TFS 2000™ low NOx firing system project have been applied to its commercially available equipment, resulting in improved NOx emissions without significantly affecting the unburned carbon levels. There are now numerous PRB coal-fired boilers that utilize aspects of the technologies demonstrated during the DOE/NETL testing and are achieving NOx emissions at or below 0.15 lb/MMBtu.
- Subsequent testing of Praxair's oxygen-enhanced combustion technology at two utility boilers, City Utilities' James River Unit 3 and Northeast Utilities' Mt. Tom Generating Station, has demonstrated the benefits of the technology. Oxygen-enhanced combustion has also been installed on two coal-fired boilers at the P.H. Glatfelter pulp and paper mill in Spring Grove, Pennsylvania.
- FERCo's KNOxcheck™ online SCR catalyst activity measurement system became commercially available in 2007.

Conclusion

With the advent of increasingly stringent regulation of NOx emissions from the electric power sector, it is critical that technology continues to improve in terms of both performance efficiency and cost. This has become even more important in light of the potential for climate change regulations that will place an even greater premium on efficient plant operations. DOE/NETL's partnership with industry and other key stakeholders in carrying out a comprehensive Innovations for Existing Plants Program has helped bring to the marketplace advanced NOx control technologies. These technologies, coupled with developments made in the areas of advanced mercury and CO_2 capture, will allow coal-fired power plants to continue to play a critical role in the Nation's energy security.

Additional information on DOE/NETL's advanced NOx emissions control activities can be found at:
http://www.netl.doe.gov/technologies/coalpower/ewr/nox/index.html.

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