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The U.S. Department of Energy's NO_x control technology R&D programme for existing power plants

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Abstract: The U.S. Department of Energy (DOE) has established a set of national priorities through its Strategic Plan that includes the goal to promote secure, competitive, and environmentally responsible energy systems that serve the needs of the public. The Innovations for Existing Plants (formerly the Advanced Research and Environmental Technologies) programme, managed by the National Energy Technology Laboratory (NETL), develops advanced environmental control technologies for coal-based power systems. The programme also provides high-quality scientific information on present and emerging environmental issues for use in regulatory and policy decision-making.

An important component of the programme is the research and development of advanced nitrogen oxide (NO_x) control technologies. This effort is focused primarily on systems capable of controlling NO_x emissions to a level at or below 0.15 lb/million Btu at a cost significantly lower than state-of-the-art technology. The programme will also provide an improved understanding of the impact of these advanced technologies on related issues such as unburned carbon, waterwall wastage, and mercury speciation and capture. The research is driven by continuing pressure for further reductions in NO_x emissions from coal-fired utility boilers to address ground-level ozone and other environmental considerations including ambient fine particulates, visibility, eutrophication, and climate change. NETL is currently managing a portfolio of NO_x control technology R&D projects ranging from laboratory studies to modelling to full-scale demonstration. This paper will provide an update on the status of these projects covering ultra-low-NO_x burners, advanced reburning, selective non-catalytic reduction (SNCR), selective catalytic reduction (SCR), methane de-NO_x, and enhanced-oxygen combustion.

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1 Background

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 nitrogen oxides (NO_x), which can be formed by (1) nitrogen and oxygen in the combustion air reacting at high temperature to produce 'thermal NO_x' and (2) fuel-bound nitrogen reacting with the combustion air to produce 'fuel NO_x.' The amount of NO_x formed depends on numerous factors including flame temperature, nitrogen content of the coal, combustion excess air, residence time, and degree of mixing.

The emission of NO_x to the atmosphere can contribute to a number of environmental concerns. NO_x can react with volatile organic compounds in the presence of sunlight to form ozone. NO_x is also a precursor to secondary fine particulate matter that may impact human health or contribute to regional haze and can form acidic compounds (acid rain) through reactions with water, oxygen, and oxidants in the atmosphere. The deposition of nitrogen compounds in and around bodies of water has been linked to 'eutrophication' – an over-enrichment of nutrients that can deplete the oxygen content of lakes and rivers. Atmospheric deposition has been identified as a primary source of nitrogen in the Chesapeake Bay. The U.S. Environmental Protection Agency (EPA) has recently proposed using the Clean Water Act as a mechanism to further reduce NO_x emissions near sensitive waters. Finally, one compound of NO_x, nitrous oxide (N_2O), is a greenhouse gas.

Since passage of the 1990 Clean Air Act Amendments (CAAA), the U.S. electric-utility industry has taken considerable action to control emissions of NO_x. Title IV (the 'acid rain' provision) of the CAAA is a two-phase strategy to reduce NO_x emissions from electric-utility boilers. For Phase 1, the EPA promulgated emission limitations for two boiler types – dry-bottom, wall-fired boilers and tangentially fired boilers (known as Group 1 boilers). Starting January 1, 1996, dry-bottom boilers were required to meet a 0.50 lb NO_x/million Btu emission limit, while T-fired boilers were limited to 0.45 lb NO_x/million Btu. Under Phase II of the programme, limits for the Group 1 boilers have been tightened, while new standards were set for Group 2 boilers – wet-bottom boilers, cyclones, cell burners, and vertically fired boilers.

Under Phase II of the programme, limits for the Group 1 boilers have been tightened, while new standards were set for Group 2 boilers – wet-bottom boilers, cyclones, cell burners, and vertically fired boilers. The Phase II emission limits are shown in Table 1.

The first stage of Title IV will reduce Group 1 boiler annual NO_x emissions by over 400,000 tons per year between 1996 and 1999, and by approximately 1.17 million tons per year beginning in the year 2000 (Phase II). The Phase II, Group 2 boiler restrictions will achieve an additional reduction of 890,000 tons of NO_x

Table 1 Phase II NO_x emission limits under Title IV of the 1990 Clean Air Act Amendments.

	NO_x emissions limit, lb/MMBtu
Group 1 Boilers	
Dry bottom wall fired	0.46
T-fired	0.40
Group 2 Boilers	
Cell burners	0.68
Cyclones	0.86
Wet bottoms	0.84
Vertically fired	0.80

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annually by the year 2000. In 1980, coal-based electric utilities emitted about 6.1 million tons of NO_x. The overall goal of Title IV is to reduce NO_x emissions from coal-fired power stations by 2 million tons below 1980 levels.

Coal-based power production will also be affected by Title I of the 1990 CAAA. Title I addresses six priority pollutants including ozone. In September 1998, EPA announced a final rule for reducing regional transport of ground-level ozone. The final rule requires 22 states and the District of Columbia to submit State Implementation Plans (SIP) to address ozone transport through reductions in NO_x emissions was issued in September 1998. Under what is commonly referred to as the NO_x SIP Call, states will have the flexibility to choose which sources to regulate. However, it is most likely that fossil-fuel-fired electric utilities will be targeted. In fact, EPA established State NO_x budgets based on a NO_x emissions rate for electric power plants of 0.15 lb/million Btu during the five-month (May through September) 'summer' ozone season.

In March 2000, the U.S. Court of Appeals for the District of Columbia Circuit upheld EPA's authority to issue the NO_x SIP Call and to establish state 'NO_x budgets'. Under Phase 1 of the SIP Call, states will be required to submit their implementation plans by October 2000 and have NO_x control technology in service by May 2004. EPA estimates that implementation of the first phase of the SIP Call, which will effect 19 states and the District of Columbia, will reduce annual NO_x emissions by an additional 860,000 tons.

2 DOE-NETL'S NO_x R&D programme

Building upon the success of the Clean Coal Demonstration Programme, DOE-NETL continues the research, development, and demonstration of advanced NO_x control technology through the Innovations for Existing Plants programme [1]. This programme is in direct response to various calls for further reductions in atmospheric emissions from coal-fired boilers. A key to the success of the DOE-NETL NO_x technology research is close coordination and cooperation with industry. As a result, the programme has had a strong history of assisting in the development of useful commercial products, such as low-NO_x burner (LNB) technology. Figure 1 shows how the introduction of advanced NO_x control technology, such as LNBs, has helped to significantly reduce the emission of NO_x from coal-fired power plants on a pound per million Btu basis.

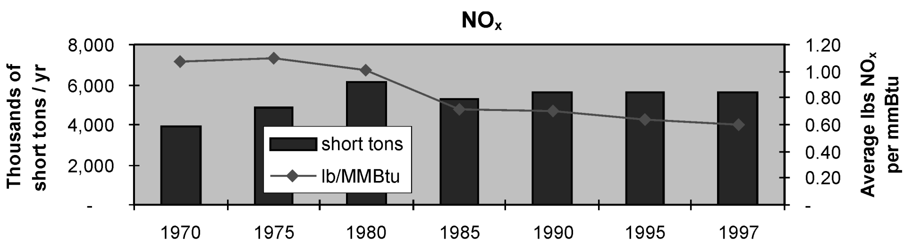

Figure 1 NO_x emissions from coal-fired power plants in the U.S.

Partnership and collaboration with industry, the research community, and academia continues to be key to the success of the DOE-NETL NO_x control technology R&D programme. American Electric Power, the Tennessee Valley Authority, FirstEnergy, and Southern Company Services are a few of the electric-utility companies working with DOE-NETL in the development of new technology. In addition, the programme has strong representation by equipment developers, manufacturers, and vendors including McDermott Technologies, ABB-Alstom Power, the Institute of Gas Technology, and GE-EERC. EPRI and the Gas Research Institute also play an active role in the research being sponsored by DOE-NETL.

The current portfolio of NO_x control technology projects within the Innovations for Existing Plants programme is primarily an outgrowth of two R&D initiatives. In October 1998, phase II extensions were granted for NO_x technology development as part of DOE-NETL's 1995 *Advanced Environmental Control Technologies for Coal-Based Power Systems* initiative. Under this programme, DOE-NETL was interested in (1) basic research to improve the performance of 'in-furnace' NO_x control processes such as low-NO_x burners, staged combustion, reburning, and selective non-catalytic reduction, and (2) unique concepts for combustion and post-combustion NO_x controls. Reaction Engineering and GE-Energy and Environmental Research Corporation are carrying out separate projects in this area.

Recognizing the need for lower-cost emission control systems for existing power plants in the face of ever present environmental pressures, DOE-NETL issued a follow-on competitive solicitation in 1999 entitled Emission Control Technology for Fine Particulate Matter ($PM_{2.5}$), Ozone, and Related Environmental Issues. The NO_x component of the solicitation was specifically driven by the NO_x SIP call and addressed the need for strategic research, development, and testing of efficient, cost-effective NO_x control technologies, processes and concepts that could be retrofitted to existing coal-fired electric utility boilers. These advanced systems were to be capable of meeting a target NO_x emission limit of 0.15 lb/million Btu or lower while achieving a levelized cost savings on a dollar-per-ton of NO_x removed of at least 25% over SCR.

Moreover, the technologies are to be ready for commercial deployment by 2002–2004 in order to parallel as closely as possible the implementation schedule of the NO_x SIP call. Under this deployment schedule, the technologies will also be available to address potential future regulatory action related to the contribution of utility NO_x to ambient fine particulates, regional haze, atmospheric nitrogen deposition, and climate change. Further, the technologies were to have (1) negligible impact on balance-of-plant issues, (2) applicability to a wide range of boiler types and configurations, and (3) reliability over a wide range of feed coals and operating conditions. Five NO_x projects were selected under the Emission Control Technology for Fine Particulate Matter ($PM_{2.5}$), Ozone, and Related Environmental Issues initiative. The projects are with Reaction Engineering, ABB-Alstrom Power, McDermott Technology, Praxair, and IGT.

The DOE-NETL programme also includes the full-scale demonstration of SNCR under a cooperative agreement with American Electric Power. In addition, the University of North Dakota Energy and Environmental Research Centre is investigating the effect of SCR catalysts on the oxidation and subsequent capture of mercury.

3 Project descriptions

3.1 Advanced NO_x control technology for coal-based power systems

An objective of the 1995 Advanced Environmental Control Technologies for Coal-Based Power Systems initiative was to carry out basic research to improve the performance of 'in-furnace' NO_x control processes, and to develop unique concepts for combustion and post-combustion NO_x controls. Two projects in this area were selected in 1997 to proceed with the second phase of this two-phase programme.

Reaction Engineering is carrying out fundamental research and engineering development of LNBs and reburning fuel injectors. The team of REI, the University of Utah, Brown University, and DB Riley, Inc., will develop combustion systems that will minimize NO_x emissions and reduce carbon in the fly ash. The five-year project is focused on 'in-furnace' NO_x control for pulverized-coal (PC) boilers including staged LNBs, reburning, SNCR, and hybrid approaches (e.g. reburning with SNCR). The research involves: (1) fundamental studies at laboratory and bench scale to define NO_x reduction mechanism in flames and reburning jets, (2) laboratory experiments and computer modelling to improve two-phase mixing predictive capability, (3) evaluation of commercial LNB fuel injectors to develop improved designs, and (4) demonstration of coal-injector reburning and LNBs at commercial scale.

During Phase I, REI's efforts concentrated on an investigation of fuel-injection systems and on the evaluation of advanced burnout models for predicting loss-on-ignition (LOI) from LNB firing systems [2]. A new Carbon Burnout Kinetic (CBK) model has been added to REI's GLACIER reacting code to evaluate and predict carbon burnout and fly-ash-carbon content. The lower furnaces of two commercial coal-fired boilers were modelled using the updated GLACIER. Observations from the two plants were consistent with model runs that suggested high corrosion rates in excess of 50 mils/year were occurring in regions of the water wall where there are high gradients in gas concentrations.

To quantify the relationship between NO_x and unburned carbon, REI has also performed a series of experiments in their 100,000 Btu/hr 'U' furnace with a Utah bituminous coal. REI's results suggest that to simultaneously achieve good carbon burnout and low NO_x emissions it will be necessary to provide sufficient oxygen early in the combustion process to ensure that the largest particles are completely burned. This will inevitably result in the production of significant NO_x that must subsequently be destroyed. To address this need, REI investigated the effect of coal reburning on NO_x formation and destruction. The results showed rather conclusively that there exists an important reaction mechanism between CO and NO_x that, if properly utilized, can help decrease NO_x from large coal particles in the early part of the flame.

As part of Phase I, REI identified four possible paths to achieve low NO_x emissions (< 0.2 lb NO_x/million Btu), while simultaneously minimizing carbon loss:

- Reduction in coal particle size in conjunction with optimized LNBs
- Conventional natural gas reburning downstream of the primary combustion zone.
- Coal reburning plus downstream chemical addition for selective reduction.

- Improved LNBs optimized for both minimum NO_x and acceptable LOI.

Unfortunately, the first three techniques can be expensive if additional capital and operating expenses are fully considered. Therefore, REI focused the last portion of Phase I on two approaches to improving LNB firing systems:

- Particle size and stoichiometry segmentation
- Gas stabilized axial flames

In segmented firing, a segregated premixed pulverized coal (SPPC) burner was created. The burner consisted of inner and outer premixing chambers, each with separate primary coal and secondary air injection. A range of experiments was conducted varying overall excess air, inner stoichiometry (SR_i), outer stoichiometry (SR_o), inner/outer velocity ratio and inner/outer coal split. Table 2 below summarizes the results of the SPPC burner studies at an overall stoichiometry of 1.05. As shown, in each inner/outer feed-coal split the NO_x level was reduced compared against a baseline NO_x concentration of 650 p.p.m. (0.90 lb/million Btu).

Previous work by REI indicated that a small amount of natural gas added around the periphery of the axial coal jet could further reduce NO_x emissions. In the Phase II programme, tests were run to determine if additional reductions could be achieved by adding the natural gas on the axis of the pulverized fuel jet. NO_x emissions of only 132 p.p.m. were achieved with 4% natural gas and a coal jet stoichiometry of 0.9. In Phase II of the REI project, which will be completed in November 2000, an optimized injector design will be tested at the 100-million Btu/hr DB Riley Coal Burner Test Facility. It is anticipated that this work will provide improved hardware designs and computer simulation models for reduced NO_x emissions and minimized carbon loss.

The second project is being carried out by GE-Energy and Environmental Research Corporation (GE-EER) to develop a family of novel NO_x control technologies called Second Generation Advanced Reburning (SGAR) capable of achieving 90% or greater reduction in NO_x at a cost significantly lower than selective catalytic reduction (SCR). Second generation advanced reburning is an integration of basic reburning and injection of a nitrogen agent (N-agent).

In the SGAR system, the N-agent (typically ammonia or urea) is injected into the reburning zone, along with overfire air, or downstream in the burnout zone. Several advanced reburning (AR) configurations are being tested by GE-EER. In AR-Lean, the N-agent is injected along with overfire air (OFA) at relatively low temperatures (1800–2100°F) in comparison to basic reburning. In AR-Rich, the N-agent is injected into the reburning zone. Another approach is injection of the N-agent in the reburning zone and with the OFA called Multiple Injection Advanced Reburning

Table 2 The effect of segmented firing on NO_x levels.

Inner/outer split, %	SR_i	SR_o	NO_x, p.p.m.
75/25	0.5	2.6	480
50/50	0.8	1.3	460
25/75	1.6	0.9	425

(MIAR). The N-agent can also be injected in a normal SNCR configuration downstream of AR-Rich or AR-Lean. Finally, N-agent injection can be accompanied by the addition of soluble promoters that further enhance NO_x reduction. A schematic of the several variations in SGAR is shown in Figure 2.

Recent project activities have included proof-of-concept combustion tests at the GE-EER Tower Furnace (TF) in Irvine, California. The TF (Figure 3) is a down-fired combustor with a nominal-firing rate of 10 million Btu/hr. The facility is designed simulate the temperatures, gas compositions, and flow-field characteristics of a coal-fired boiler, thus providing a means of directly applying results to full-scale systems.

The objective of the TF tests was to provide data that could be used to project full-scale SGAR performance. A high-sulphur, bituminous Illinois coal was used as the main fuel for all tests. An air-staging system was applied to the primary burner to simulate a commercial LNB, thus providing initial NO_x concentrations similar to those obtained in full-scale boilers. The N-agent and promoter consisted of urea and sodium carbonate, respectively.

Figure 4 shows overall NO_x reductions for each process, including the burner air staging system, which simulates a low-NO_x burner. Staging provided 57% NO_x control. Basic reburning increased this to 70% (only 6% natural gas was used).

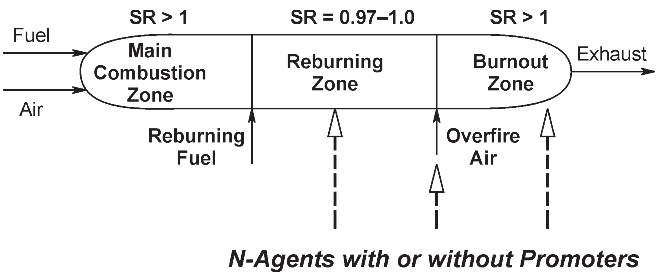

Figure 2 SGAR variants.

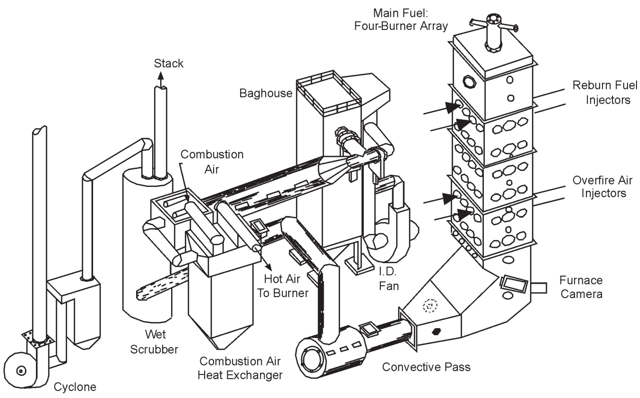

Figure 3 Schematic of 10 million Btu/hr tower furnace.

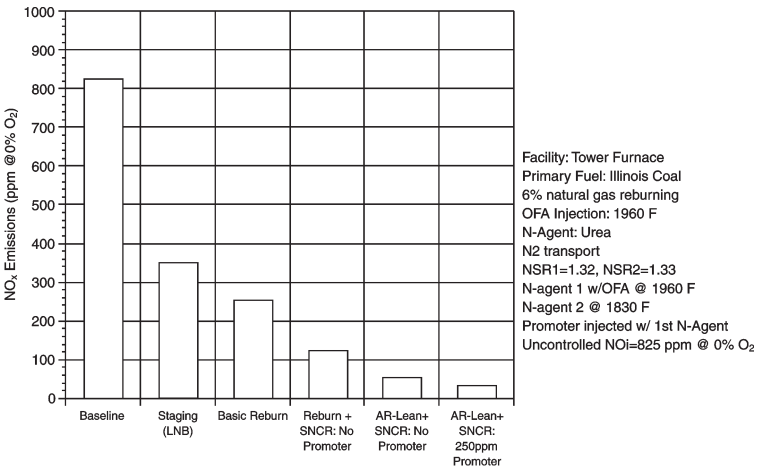

Figure 4 Results of tower furnace tests.

Adding SNCR increased overall NO_x reduction to 85%. Adding AR-Lean (urea injection with OFA) increased performance to 94%. Overall NO_x reduction for the complete MIAR process, including sodium promoter, was 96%. The complete process caused NO_x emissions to decrease from a baseline concentration of 825 p.p.m. to 32 p.p.m.

During the remainder of the project, GE-EER will continue optimization of SGAR in the 10 million Btu/hr TF to provide the data necessary for full-scale commercial plant designs.

3.2 Advanced technology for meeting the NO_x SIP call

Five NO_x control technology projects were selected under the 1999 Emission Control Technology for Fine Particulate Matter ($PM_{2.5}$), Ozone, and Related Environmental Issues initiative. As noted above, these five projects are focused on the development of technology capable of achieving 0.15 lb NO_x/million Btu while achieving a levelized cost savings of at least 25% over SCR. Moreover, the technologies are to be commercially available by 2004.

Alstom Power is carrying out a 21-month effort to develop an ultra low NO_x integrated system for coal-fired power plants to address present and anticipated NO_x emissions control legislation. The proposed system will build on Alstom Power's field-proven TFS 2000^{TM} low NO_x firing system to achieve furnace outlet NO_x emissions at or below 0.15 lb/million Btu for existing tangentially-fired boilers, firing a wide range of coals. Field retrofit TFS 2000^{TM} systems have demonstrated NO_x emissions at or below 0.2 lb/million Btu when firing an eastern bituminous coal, and at or below 0.15 lb/million Btu when firing a western sub-bituminous coals. The target NO_x emissions will be obtained without increasing the level of unburned carbon in the fly ash through advances in combustion process modification and a post-combustion carbon burnout technology. In addition, an advanced, neural control system will be employed to maintain target NO_x emissions over the range of boiler operation and load.

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The cost of the Alstom Power system is anticipated to be less than half that of SCR systems. The system will be available for market application in 2001. The target market is tangentially fired coal boilers that represent 10,000 MW, or 40%, of the current SIP Call region boilers. The project team includes ABB C-E Services, ABB Power Plant Controls, ABB Environmental Systems, and Progress Materials, Inc. Testing will be conducted at Alstom Power's 15 MW Boiler Simulation Facility located in Windsor, Connecticut.

In a second project, McDermott Technology, Babcock & Wilcox, and Fuel Tech are teaming on a two-year effort to provide an integrated solution for NO_x control comprised of an ultra Low-NO_x (ULNB) pulverized coal burner technology (B&W's DRB-4Z^{TM}) plus urea-based, selective non-catalytic reduction system (Fuel Tech's NO_xOUT). The market niche for the ULNB/SNCR technology are front and opposed wall-fired boilers within the NO_x Sip Call region, with cell-fired, roof-fired, and arch-fired boilers also among candidate boilers. An estimated 86,000 MW (about 75% of the 115,000 MW wall-fired units in the SIP region) is targeted.

The goal of this project is to develop a cost-effective NO_x control system that will be deployable in coal-burning power plants by the year 2002, and achieve a NO_x level below 0.15 lb/million Btu for a wide range of coals. The project is scheduled to be completed in September 2001. Since the first retrofit of a double-register burner (DRB) in 1973 produced a 50% reduction in NO_x, these burners have been installed in 70 boilers with a combined capacity exceeding 40,000 MW. The next major advance, the DRB-XCL^{TM}, yielded reductions of NO_x in the 50–70% range without over-fire air (OFA); to date, the XCL^{TM} has been installed in 86 boilers, some with and some without OFA, for more than 23,000 MW total. The most recent burner, the ultra low-NO_x DRB-4Z^{TM}, was developed during the period 1993–1997, in DOE's Low Emissions Boiler System programme. NO_x levels below 0.20 lb were recorded with Illinois No. 6 coal. To simplify retrofits, MTI recently tested an equally successful, small throat (plug-in) version of the DRB-4Z^{TM}, in a project supported by DOE and the Ohio Coal Development Office.

The current project will optimize and test the plug-in burner with SNCR in the 100 million Btu/hr Clean Environment Development Facility at Alliance, Ohio. This will provide data for a single burner in the capacity range of utility applications under well-controlled and commercially representative conditions. The effects of coal rank will be examined by firing three coals: a Powder River Basin subbituminous (Spring Creek), a high-volatile bituminous (Illinois No. 6) and a medium-volatile bituminous (Pennsylvania Middle Kittanning). No staging will be employed; a stoichiometric ratio range from 1.10 to 1.28 will be tested, with 17% excess air being the target at full load. Coal will be pulverized as common boiler grind, 70 to 75% through 200 mesh.

MTI recognizes that commercial boilers could produce higher levels of NO_x than indicated for similar conditions in the CEDF, owing to burner flame interactions, coal property variations, etc. Therefore, the target in the CEDF has been set at 0.125 lb/million Btu, in order to provide confidence for the 0.15 lb/million Btu figure in commercial installations. In addition to the NO_x work, MTI will perform some tests to investigate the influence of the ULNB on the fate of mercury during coal combustion. Mercury has been a subject of intense research for a decade, with reference to its partitioning into gas and particulate phases and speciation in the gas phase. The ULNB introduces a new factor, however, since attempts to reach very low levels of NO_x have been associated with an increase in unburned carbon. This carbon generally is weighted to the smaller size fraction of the fly ash, providing active sites for reaction with mercury.

A third project was awarded to the Institute of Gas Technology (IGT). IGT is teaming with the All-Russian Thermal Engineering Institute, DB Riley, and the Gas Research Institute to develop a pulverized-coal (PC)-combustion system that is an extension of IGT's METHANE de-$NO_x^{®}$ technology. Specifically, the technology is composed of a novel PC burner design using natural gas fired coal preheating developed and demonstrated in Russia, LNBs with internal combustion staging, and additional natural gas injection with overfire air. The coal is preheated at elevated temperatures (up to 1500°F) in oxygen deficient conditions prior to combustion. Coal preheat releases fuel-bound nitrogen together with volatiles present in the coal. These conditions promote the conversion of fuel-bound nitrogen to molecular nitrogen rather than to NO_x. A simplified schematic diagram of the technology is shown in Figure 5.

During this 31-month effort, a pilot-scale preheated PC burner system will be installed and tested at DB Riley's 4-million Btu/hr combustor located in Worchester, Massachusetts. Testing will be performed on a Western low-sulphur subbituminous coal and a high-volatile Midwestern bituminous coal. Key parameters to be evaluated include coal throughput, excess air, coal preheat temperature, primary zone air-to-fuel ratio, gas-to-secondary-air zone, and combustion air staging. Based on the pilot-scale results, development of a commercial prototype NO_x reduction system will be carried out using DB Riley's 100-million Btu/hr combustion facilities. The goal of this task will be to validate system performance in order to move to commercial field demonstration. IGT estimates the market for the technology to include more than 21,000 burners (over 260,600 MW) in the 37 eastern states encompassing wall-fired (wet and dry bottom), T-fired, roof-fired, and cell burners.

A fourth project was awarded to Reaction Engineering International (REI) to optimize the performance of, and reduce the technical risks associated with, the combined application of low-NO_x firing systems (LNFS) and post combustion controls through modelling, bench-scale testing, and field verification. Teaming with REI are the University of Utah and Brown University. During this two-year effort, REI will assess real-time monitoring equipment to evaluate water-wall wastage, soot formation, and burner stoichiometry, demonstrating analysis techniques to improve LNFS in combination with reburning/SNCR, assessing selective catalytic reduction catalyst life, and developing UBC/fly ash separation processes.

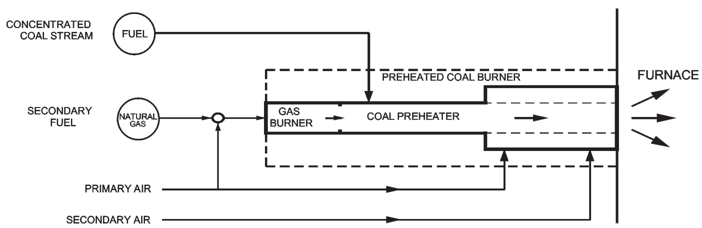

Figure 5 Simplified schematic of PC methane de-$NO_x^{®}$ technology.

The REI programme will be applicable to coal-fired boilers currently in use in the United States, including corner-, wall-, turbo-, and cyclone-fired units. However, the primary target of the research will be cyclone boilers, which are high NO_x producing units and represent about 20% of the U.S. generating capacity. The results will also be applicable to all U.S. coals. The research will be divided into four key components:

- *Lower furnace*: Work will be performed to demonstrate the value of real-time monitoring equipment to evaluate waterwall wastage and burner stoichiometry. In addition, REI will study the formation of carbonaceous particulate (soot) from coal volatiles.
- *Upper furnace/convective section*: REI will investigate analysis techniques that will help optimize the performance of LNFS in combination with reburning and/or SNCR.
- *Catalyst bed*: REI will assess the useful life of catalyst exposed to flue gases produced from the combustion of U.S. coals.
- *Fly ash disposal/use*: Studies will be carried out on the adsorptive properties of fly ash to help develop cost-effective processes to remove ammonia.

The fifth project under the Emission Control Technology for Fine Particulate Matter ($PM_{2.5}$), Ozone, and Related Environmental Issues was awarded to Praxair to develop and demonstrate oxygen-enhanced combustion and oxygen-enhanced secondary control technologies for controlling NO_x, as well as a novel oxygen separation process. Oxygen enhanced combustion can be used to control both thermal and fuel NO_x. The concept of adding oxygen to the combustion process appears counter intuitive due to the higher flame temperatures typically associated with oxy-fuel firing. For example, when small amounts of combustion air are replaced with oxygen, a significant increase in flame temperature can be realized. The key to this project is the use of controlled conditions to take advantage of the combustion benefits of oxy-fuel firing and reduce NO_x emissions below the 0.15 lb/million Btu goal.

Oxygen-fired combustion has been utilized in industrial furnaces to improve energy efficiency and reduce emissions. NO_x emissions reductions of as much as 80–90% have been demonstrated at commercial glass melting furnaces that have been converted to oxygen-fuel firing. One of the required keys to successfully implementing oxygen-fired or enhanced combustion is an economical source of oxygen, which also can benefit numerous other technologies. Praxair is developing a novel oxygen separation technology at its Tonawanda, New York facility, using an oxygen transport membrane (OTM). Ceramic membranes are attractive since they can have virtually infinite selectivity for oxygen, thereby allowing only the oxygen to pass through. There are two basic types of ceramic membranes, pressure and electrically driven, as shown in Figure 6. This project will use pressure as the driving force for separation.

Ultimately, the technology is looking to reduce the cost of NO_x emissions reductions. Preliminary calculations show a potential for savings of 25–75% versus SCR. Teaming with Praxair are the University of Utah, the University of Arizona, Reaction Engineering, and Alstom Power. Modelling and laboratory testing will be

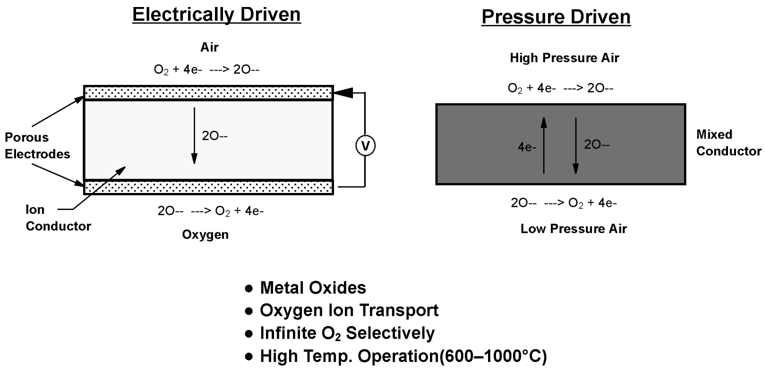

Figure 6 Oxygen transport membrane (OTM) technology.

conducted at REI, the University of Utah, and the University of Arizona, while full-scale testing of an oxygen-enhanced LNB will be carried out at Power Plant Laboratories in Windsor, Connecticut.

3.3 SNCR field demonstration

In addition to the development of advance primary NO_x control systems, DOE-NETL recently participated in a government-industry collaboration involving the full-scale testing and evaluation of SNCR technology [3]. This $6.5 million effort was completed in April 2000 in partnership with American Electric Power (AEP), the Ohio Coal Development Office, and EPRI. A consortium of electric utilities including GPU GENCO, Allegheny Energy, Illinova, Ameren, Louisville Gas and Electric Company, Baltimore Gas and Electric, New England Electric System, Buckeye Power, Southern Company Services, Cinergy, TVA, East Kentucky Power Cooperative, WEPCO, and FirstEnergy, also participated in the programme.

As it is likely that any new NO_x rule will allow for system-wide averaging of emissions, AEP was interested in evaluating the maximum NO_x reduction that could be achieved by coupling SNCR with combustion controls. SNCR involves the injection of a solution of ammonia (NH_3) or urea into the furnace in a temperature window between 1800°F and 2200°F to react with and remove NO_x. Conceptually, SNCR is a simple process. The nitrogen-based reagent reacts selectively in the presence of oxygen to reduce the NO_x to molecular nitrogen (N_2) and water (H_2O).

The test programme was carried out at the AEP Cardinal Plant Unit 1, a 600 MW_eopposed-wall, cell-fired, dry-bottom, pulverized coal-fired boiler located in Brilliant, Jefferson County, Ohio (Ref. 2). Equipped with LNBs, Unit 1 was in compliance with the Title IV emission limit of 0.68 lb/million Btu. The specific objective of the SNCR project was to reduce NO_x by an additional 30%, while maintaining ammonia concentrations in the flue gas, known as 'slip', at or below 5 p.p.m. This level of control, when combined with the reduction from the LNBs, would achieve an overall reduction from the plant's baseline NO_x level of about 67%. Thus, the project would demonstrate that the integration of LNBs and SNCR could provide a cost-effective level of NO_x control beyond that mandated by Title IV, allowing for the generation of NO_x credits.

A significant feature of the project was that it addressed two critical SNCR technical issues – unit size and coal-sulphur content. First, it represented the largest-scale demonstration of SNCR technology on a coal-fired boiler in the United States. The previous largest SNCR installation was on a 321 MW_e unit. Second, Unit 1 burns an eastern bituminous coal with a sulphur content of about 3.7%. An important SNCR operating issue is the potential formation of ammonium sulphate and bisulphate due to the reaction of sulphur oxides with ammonia that has 'slipped' through the SNCR system. Ammonium bisulphate can condense in the heat-transfer sections of regenerative air heaters. In addition, ammonia can adsorb on fly ash and ammonium salts can create a potential plume opacity problem. The amount of ammonium sulphate and bisulphate formed is a function of the sulphur in the combustion gases.

Fuel Tech provided and installed the SNCR equipment at the Cardinal plant. Urea could be injected at three levels (zones) in the furnace. Optimization of the SNCR unit was carried out between March 16 and April 27, 1999. The testing was performed at loads of 600, 450, and 350 MW_e. The tests comprised a wide variety of configurations covering the zones in service, injectors in service at each zone, chemical bias, the amount of urea injected, and other injection parameters.

NO_x reduction and NH_3 slip data are shown in Figure 7 for the test runs at 600 MW_e. The line in this and the next two figures defines the theoretical performance curve based on the measurements made during the optimization tests. These data show NH_3 slip below the 5 p.p.m. target can be achieved at NO_x reductions of 20 to 25% on a consistent basis. Several tests were also performed where NO_x reductions between 25 and 35% were achieved with an NH_3 slip at or below 5 p.p.m.

At intermediate load (450 MW_e), the corresponding NO_x reduction versus NH_3 slip data are shown in Figure 8. These data show that multiple-level injection provided the best combination of high NO_x reduction and low ammonia slip. These data also show that mid-load NO_x reductions between 30 and 35% can be achieved with NH_3 slip levels less than 5 p.p.m.

The NO_x reductions versus NH_3 slip data for tests performed at 350 MW_e are shown in Figure 9. These data indicate that NO_x reductions between 30 and 35% could be achieved at an NH_3 slip below 5 p.p.m. using multi-zone injection.

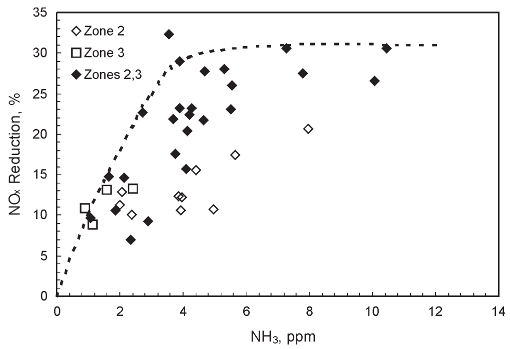

Figure 7 Relationship between NO_x reduction and NH_3 slip at 600 MW_e.

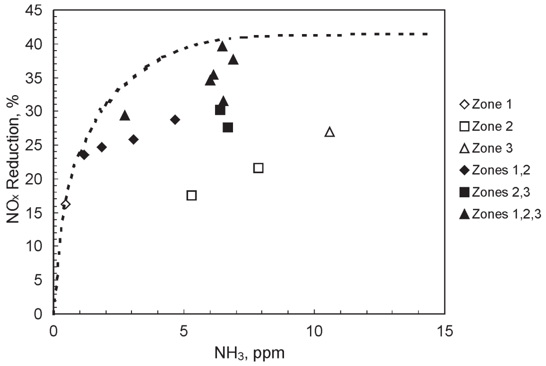

Figure 8 Relationship between NO_x reduction and NH_3 slip at 450$_e$ MW.

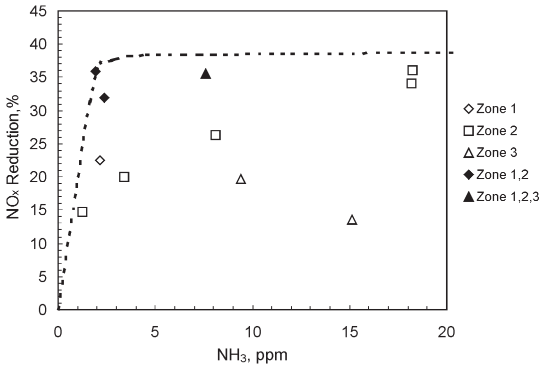

Figure 9 Relationship between NO_x reduction and NH_3 slip at 350$_e$ MW.

Based on the results of the optimization programme, long-term testing of the SNCR system at the Cardinal Plant was carried out between September 20 and November 19, 1999. During this time, the unit was held at various load points in order to verify that SNCR could successfully perform at full, intermediate, and minimum loads. The system provided approximately 30% reduction in NO_x across the load range while minimizing slip. SNCR performance, based on post-LNB NO_x emission levels, at the three primary test loads during the long-term runs is summarized below in Table 3.

Table 3 Results of SNCR long-term testing at AEP Cardinal Station.

Load, MW_e	NO_x Reduction, %	NH_3 Slip, p.p.m.
600	25	4
450	29	2
350	30	3

The most significant balance-of-plant concerns, air heater pluggage and fly ash contamination, were not a major problem during the long-term test programme, but a longer test period would be needed to fully evaluate the effect of SNCR on downstream plant operations.

3.4 Impact of SCR on mercury speciation

It is expected that SCR technology will play an expanded role in the electric-utility industry's strategy to comply with current and future NO_x regulations. Further, the need to control the emission of mercury from coal-fired power plants continues to remain a topic of debate. In response, the University of North Dakota Energy and Environmental Research Centre (UNDEERC) has initiated a nine-month effort to investigate the impact of SCR on mercury speciation – and on its subsequent capture. Specifically, UNDEERC is studying the effect of metal oxide catalysts, such as vanadium/titanium metal oxide (V_2O_5/WO_3-TiO_2), on promoting the conversion of elemental mercury to an oxidized and/or particulate form. Thus, an SCR process may improve the capture of mercury in a downstream control system such as a wet scrubber or baghouse.

The research is being carried out in UNDEERC's pilot-scale 550,000 Btu/hr pulverized coal-fired combustor. This unit is designed to generate fly ash and flue-gas chemistry representative of that produced in full-scale utility boilers. Three coals have been selected for testing, two eastern bituminous coals (low and high sulphur) and a Powder River Basin subbituminous coal. The project is being supported under a DOE Joint Venture Programme with UNDEERC, with co-funding provided by Ontario Power Generation, EPA, and EPRI.

4 Summary

A critical goal of DOE-NETL is to promote secure, competitive, and environmentally responsible energy systems that serve the needs of the public. An important component of the R&D programme to achieve this goal is the development of advanced NO_x control technology. This effort is driven by a host of environment issues surrounding the emission of NO_x including ground-level ozone, fine particulate matter, acidification, visibility impairment, nitrogen deposition, eutrophication, and climate change. DOE-NETL is maintaining a research portfolio focused on providing the electric-utility industry with a suite of cost-effective technological options to address current and future restrictions on NO_x emissions. In addition, the programme will provide an improved understanding of the impact of these advanced technologies on fundamental issues such as unburned carbon, waterwall wastage, and mercury speciation. This research will help meet the challenge of maintaining a balanced energy mix in the United States well into the twenty-first century.

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