An Overview of NO_x Control Technologies Demonstrated under the Department of Energy's Clean Coal Technology Program

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ABSTRACT

The Clean Coal Technology (CCT) Demonstration Program, sponsored by the U. S. Department of Energy (DOE), is a government and industry co-funded technology development effort to demonstrate a new generation of innovative coal utilization processes. The CCT Program began in 1987 and will continue through the year 2000. The purpose of the Program is to remove the economic and environmental impediments associated with the development and demonstration of new coal-burning technologies. It also has opened a channel to policy-making bodies, such as the Environmental Protection Agency (EPA) and state regulators, to provide data to aid in formulating regulatory decisions. Data provided by several of the CCT projects were used to confirm that the emissions limits set by the EPA under the Clean Air Act Amendments (CAAA) of 1990 for nitrogen oxides (NO_x) from coal-fired power plants are attainable at reasonable cost. The CCT Program comprises 40 demonstration projects, of which 14 involve some form of NO_x control, either alone or in combination with control of sulfur dioxide (SO_2), a major contributor to acid rain. This paper provides an overview of the NO_x control projects, including both pre- and post-combustion technologies that have been successfully demonstrated.

HOW NO_x IS FORMED IN A BOILER

Nitrogen oxides (NO_x) are formed as a result of combustion processes, primarily in the transportation, utility, and industrial sectors. NO_x emissions contribute to a variety of environmental problems, including acid rain and acidification of aquatic systems, ground-level ozone (smog), and visibility degradation. In power plant boilers, NO_x is formed primarily in two ways: (1) high temperature thermal fixation of nitrogen in the combustion air with excess oxygen, producing thermal NO_x; and (2) conversion of nitrogen that is chemically bound in the coal, producing fuel NO_x. The amount of NO_x formed depends on flame temperature, nitrogen content of the coal, quantity of excess air used for combustion, the degree of turbulence, and the residence time at high temperature. An increase in any of these factors results in increased NO_x formation.

NO_x REGULATIONS UNDER THE CLEAN AIR ACT AMENDMENTS OF 1990

History

The Clean Air Act was originally passed in 1967. It was amended in 1970, 1977, and most recently in 1990. The Clean Air Act Amendments (CAAA) of 1990 authorize the U.S. Environmental Protection Agency (EPA) to establish standards for a number of atmospheric pollutants, including NO_x. Two major portions of the CAAA relevant to NO_x control are Title I and Title IV. Title I establishes National Ambient Air Quality Standards (NAAQS), while Title IV addresses controls for specific types of boilers, including stationary coal-fired power plants. Under Title IV, Congress authorized EPA to establish the Acid Rain Program.

Ozone Formation

Title I addresses emissions of six criteria pollutants, including ozone. Ground-level ozone is a major ingredient of smog. Since NO_x is an ozone precursor, in some situations the control of NO_x is required to achieve compliance with ozone limits. The current NAAQS for ozone is 0.12 ppm (1 hour average). At this level, many large- and medium-sized urban areas are classified as nonattainment, and many power plants are within these nonattainment areas. Updating Title I standards every five years is mandated. A number of studies have indicated that the current NAAQS for ozone is inadequate to protect either human health or the environment. Therefore on July 16, 1997, EPA proposed a more stringent ozone limit, 0.08 ppm (8 hour average). An area would be considered nonattainment when the fourth highest daily maximum 8-hour concentration, averaged over three years, exceeds 0.08 ppm.

States have the responsibility to develop State Implementation Plans (SIPs) that spell out their strategies for achieving EPA's ozone regulations. Recommendations for attaining the ozone standards have been developed by the Ozone Transport Assessment Group (OTAG) that was created under the auspices of EPA to develop a strategy for reducing ozone and ozone-precursor emissions on a regional scale. OTAG comprises the 37 contiguous eastern states plus the District of Columbia (DC); it excludes the 11 westernmost states. EPA's final rules based on OTAG recommendations will be issued by September 1998.

EPA's proposed regional rule of October 16,1997, covers boilers having a capacity of 250 million Btu/hr or more, located in 22 states and DC within the OTAG region. For these boilers, the power plant NO_x emission limit under Title I is reduced to 0.15 lb/10^6 Btu, which represents a severe reduction for a significant number of units.

Acid Rain

Title IV uses a two-part strategy for NO_x control. The first stage (Phase I) went into effect January 1, 1996 and will reduce NO_x emissions in the United States by over 400,000 tons/yr between 1996 and 1999. This will be achieved through decreasing emissions from dry bottom wall-fired boilers and tangentially fired (T-fired) boilers (Group 1). In Phase II, which begins in the year 2000, more stringent standards are set for the remaining Group 1 boilers that were not regulated in Phase I, and initial regulations are established for other boilers known as Group 2 boilers (cell-burners, cyclone boilers, wet bottom wall-fired boilers, and other types of coal-fired boilers). In Phase II, annual NO_x

reduction is estimated to be 2,060,000 tons. The statute requires that emission control costs for Group 2 boilers be comparable to those for LNBs applied to Group 1 boilers.

Implementation

The following table gives the current and proposed NO_x emissions limits for Title IV:

	NO_x Emissions Limit, lb/10^6 Btu	
	Phase I	**Phase II**
Implementation Period	**1996-2000**	**2000+**
Status of Regulations	**Promulgated**	**Proposed**
Group 1 Boilers		
Dry Bottom Wall-Fired	**0.50**	**0.46**
Tangentially Fired	**0.45**	**0.40**
Group 2 Boilers		
Wet Bottom Wall-Fired (>65 MWe)	**NA**	**0.84**
Cyclones (>155 MWe)	**NA**	**0.86**
Cell Burners	**NA**	**0.68**
Vertically Fired	**NA**	**0.80**
Fluidized Bed	**NA**	**Exempt**

NA = Not applicable

TECHNOLOGY DEVELOPMENT

NO_x control technologies can be categorized as (a) combustion modifications or (b) post-combustion processes. Combustion modifications include such technologies as low-NO_x burners (LNBs), overfire air (OFA), coal reburning (CR), and gas reburning (GR). Some combustion modification processes incorporate flue gas recirculation (FGR). Post-combustion technologies include selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR). Some technologies are combination processes which involve control of both NO_x and SO_2. The Clean Coal Technology (CCT) Program administered by the U.S. Department of Energy (DOE) includes projects that incorporate all of these technologies. They are discussed in the following section.

Combustion Modification for NO_x Control
Combustion modification is widely used to provide low-cost NO_x emissions reductions from existing plants. Three CCT projects demonstrate modified combustion processes, including the use of specially designed LNBs alone or in conjunction with OFA ports. Data from two of these projects, *Southern Company Services 180-Megawatt Demonstration of Advanced Tangentially Fired Combustion Techniques for the Reduction of NO_x Emissions from Coal-Fired Boilers* and *Demonstration of Advanced Combustion Techniques for a Wall-Fired Boiler,* were used by the EPA to set the regulatory limits for NO_x emissions under the CAAA for Title IV, Group 2 boilers.

Commercial applications of LNB technology include a wide range of wall-fired utility, T-fired utility, and industrial boilers in the United States and abroad. In the U.S. alone, the technology can be utilized for 422 existing wall-fired boilers and nearly 600 T-fired utility units. For T-fired boilers, the technology is known as the Low NO_x Concentric Firing System (LNCFS™). Also being developed in conjunction with the wall-fired boiler project is an artificial-intelligence-based software package for optimizing NO_x reduction and boiler efficiency. This is discussed in a subsequent section.

Also under the CCT program, *The Babcock & Wilcox (B&W) Company's Full-Scale Demonstration of Low-NO_x Cell Burner Retrofit* demonstration project won the 1994 Research and Development 100 Award from *R&D Magazine*. The goal of the demonstration was to achieve at least 50% NO_x emissions reduction without degrading boiler performance. The 24 two-nozzle cell burners on the Unit No. 4 boiler at Dayton Power and Light's J. M. Stuart Plant, located near Aberdeen, Ohio, were replaced with Low-NO_x Cell Burners (LNCB®). NO_x emissions and boiler performance data before and after the conversion were analyzed to determine NO_x emissions reduction and the impact on overall boiler performance. The results showed that NO_x emissions were reduced by more than 50%, while carbon in the fly ash and carbon monoxide (CO) emissions were at acceptable levels. Other boiler performance parameters (e.g., flue gas temperatures, thermal efficiency, slagging tendency, and electrostatic precipitator performance) were normal.

Thus, the technology was successfully demonstrated and is expected to have direct commercial application to more than 23,000 MWe of existing generating capacity produced by power plant boilers configured with standard cell burners. B&W has already installed its LNCB® technology on more than 4,600 MWe of capacity in the U.S. -- in each case achieving more than 50% NO_x reduction.

Reburn technologies for NO_x control are being demonstrated in four projects under the CCT Program. These technologies include gas and coal reburning and are capable of reducing NO_x emissions by 60 to 75%. *Energy and Environmental Research Corporation* demonstrated gas reburning in two CCT projects utilizing three type of units, wall-fired (172 MWe), T-fired (80 MWe), and cyclone (40 MWe). To accomplish reburning, natural gas is mixed with recirculated flue gas and injected into the furnace slightly above the main burners. OFA is injected just above the gas injection point. Mixing of the gas, flue gas, and OFA creates a reburn zone where the nitrogen oxides are converted to nitrogen and oxygen. Reburning alone is capable of reducing NO_x by up to 65% and over 70% when coupled with LNBs. Gas reburn technology was recently selected to receive the 1997 J. Deane Sensebaugh Award from the Air & Waste Management Association for the successful commercialization of a new environmentally friendly technology.

The CCT Program includes two coal reburning projects. The first is with *B&W* utilizing standard pulverized coal on a cyclone unit (100 MWe). The second is with *New York State Electric and Gas (NYSEG)* (T-fired, 150 MWe) and *Eastman Kodak Company* (cyclone, 50 MWe), demonstrating the next step in reburn technologies, using micronized coal (80% passing 325 mesh). Coal reburning is accomplished in a manner similar to natural gas. The NO_x removal rate for B&W's unit exceeded 50%. The Micronized Coal Reburn NO_x removal rate is expected to be the same as with natural gas, 65%. Either reburn technology can be utilized in conjunction with LNBs.

As discussed above, NO_x regulations will very likely become more stringent in many areas of the U.S. where ozone nonattainment prevails. Technologies that produce commercially salable by-products instead of solid waste will be in demand. Many NO_x- and SO_2-control technologies can be combined to meet future requirements. FETC is managing a number of combined SO_2/NO_x control technology projects under the CCT Program that have achieved SO_2 removal efficiencies of over 90% and NO_x reduction efficiency above 70%. These are discussed subsequently.

Post-Combustion NO_x Removal Processes

SCR is a post-combustion process in which NO_x in the flue gas is selectively reduced by reaction with ammonia (NH_3) over a catalyst to give innocuous nitrogen and water vapor. The optimum temperature range is 650 to 750^{o}F, which corresponds to the temperature at the economizer outlet. SCR systems can be located between the economizer and air heater, between the hot ESP and air heater, or downstream of a flue gas desulfurization (FGD) system. The preferred location is between the economizer and air heater, which avoids the need to reheat the flue gas. NO_x reductions of 80-90% have been achieved.

SCR has been demonstrated in a CCT project conducted by *Southern Company Services* at its Plant Crist station near Pensacola, FL, using high-sulfur coal. The process is now in commercial use at six U.S. power plants ranging in capacity from 220 to 425 MWe. A number of issues affect SCR. Catalyst activity significantly affects SCR economics because catalyst cost constitutes 15-20% of the capital cost. Unreacted NH_3 (NH_3 slip) exiting the stack is undesirable, especially when firing high-sulfur coals, because SCR catalysts tend to oxidize SO_2 to SO_3 that can then react with NH_3 to form ammonium bisulfate. The latter is a sticky solid that can plug downstream equipment such as air heaters. If the SCR unit were installed downstream of the FGD system, gas reheat would be required which would drive costs up. The oxidation of SO_2 tends to occur at the upper end of the SCR operating temperature range, which is also the temperature range in which the best rate of NO_x reduction occurs. Thus operation of SCR requires balancing removal efficiency against bisulfate formation.

For those utilities subject to the most stringent NO_x emissions limits, SCR may be the only NO_x removal technology capable of meeting the regulations.

SNCR also involves reacting NO_x with a reducing agent, which can be ammonia or urea. As the name implies, no catalyst is used. Typical operating temperatures are in the 1600-2000^{o}F range.

These temperatures require that the reducing agent be injected into the top or back pass of the furnace. SNCR typically removes 25 to 45% of the NO_x in the flue gas. Several CCT projects, including the NYSEG Project, involve demonstration of SNCR on coal-fired units.

Public Service Company of Colorado demonstrated SNCR as one of several NO_x/SO_2 technologies comprising its CCT project *Integrated Dry NO_x/SO_2 Emission Control System*. The technologies were tested on a 100 MWe vertically fired B&W unit. NO_x control was accomplished using LNBs, SNCR, and OFA. Urea was injected into the furnace at the nose, using stationary and retractable lances. The retractable lances were adjusted according to the system load. SO_2 control was accomplished by injecting dry calcium- or sodium-based sorbents into the flue gas stream. The combination of the three NO_x control technologies reduced NO_x by more than 80%. Concurrent operation of the SO_2 reduction technology had no effect on NO_x reduction.

With SNCR, as in the case of SCR, it is necessary to minimize deposition of ammonium bisulfate in the air heater to avoid plugging problems. Because of the need to achieve thorough mixing of the flue gas and reagent, there is a practical limit to the size of boiler that can effectively accommodate SNCR. To date, this limit has been about 200 MWe. Another significant issue is the generation of N_2O, a greenhouse gas that depletes ozone in the upper levels of the atmosphere.

SNCR is considerably less expensive than SCR. However, SNCR's NO_x removal capability is considerably less than that of SCR. If very high NO_x removal rates are needed, SNCR alone cannot do the job.

Combined NO_x/SO_2 Removal Processes

ABB Environmental Systems demonstrated that its *SNOX^{TM} Flue Gas Cleaning Demonstration Project* can remove over 95% of SO_2, more than 90% of NO_x, and 99.9% of particulates from flue gas while producing commercial-grade sulfuric acid as a by-product. Attractive process features include the elimination of solid waste and a very low parasitic energy consumption (i.e., none to 0.5%). Due to the exothermic nature of sulfuric acid condensation, the SNOX^{TM} process can even be optimized to produce rather than consume energy. The demonstration was conducted on a 35 MWe flue gas slipstream at Ohio Edison's Niles Station in Niles, Ohio. Commercial SNOX^{TM} plants have been started up in Denmark and Sicily. In Denmark, a 305-MWe plant has been designed and constructed; it has operated since August 1991. The plant in Sicily, operating since March 1991, has a capacity of about 30 MWe and fires petroleum coke.

B&W also demonstrated a technology known as the *SOx-NOx-Rox Box^{TM} Flue Gas Cleanup System* or SNRB^{TM} . The process combines the removal of SO_2, NO_x, and particulates in one unit, a high-temperature baghouse. NO_x removal is accomplished by injecting ammonia to selectively reduce NO_x in the presence of a catalyst. Although the technology was demonstrated only on a 5 MWe scale, it was large enough to simulate commercial-scale operation since the fabric filters used in the baghouse were full size. One reason the scale was kept small was so that it could provide the necessary temperature control of the flue gas to test a wide range of sorbents to remove SO_2. The technology performed extremely well. NO_x removal averaged over 90% and SO_2 removal averaged over 80%. Particulate removal was over 99%.

Artificial Intelligence

Experience with several CCT projects involving combustion modification has shown a tendency toward increased loss-on-ignition (LOI) values in the fly ash. LOI is a measure of the unburned carbon (UBC) that escapes from the furnace to the particulate control system. Increased carbon content changes the electrostatic properties of the fly ash and decreases the resistivity of the particles; this adversely affects ESP performance and fly ash marketability. Fly ash can be sold as road construction material, among other things, provided that the UBC content meets specifications. Although UBC can be reduced by increasing coal fineness, this problem remains significant and is receiving considerable attention.

Experience also has indicated that combustion modification leads to difficulties in boiler optimization. This is a result of several factors, including (1) heightened awareness of the impact of combustion conditions on NO_x emissions and plant efficiency, including UBC, and (2) increased sensitivity of combustion conditions to process adjustments.

Artificial intelligence can aid in optimizing boiler control with a view to minimizing UBC and maximizing overall efficiency. The Plant Hammond wall-fired NO_x reduction project has been expanded to demonstrate the effectiveness of advanced digital control and optimization methodologies as applied to the NO_x abatement technologies being demonstrated at that site. This extension includes (1) design and installation of a distributed digital control system, (2) instrumentation upgrades, and (3) advanced controls/optimization design and implementation. The optimization methodology being developed in this project has been termed the Generic NO_x Control Intelligent System (GNOCIS). Several on-line carbon-in-ash monitors are being evaluated for this application.

The GNOCIS test work at Southern Company's Plant Hammond has demonstrated an efficiency gain of 0.5%, a reduction in LOI of 1-3%, and a reduction in NO_x emissions of 10-15%. Based on the success of this project, Southern Company has installed GNOCIS software in six other power plants, and additional commercial installations are anticipated.

ECONOMICS

Based on the results of the CCT demonstration projects, economics have been estimated for full-scale commercial implementation of the various technologies. The results are summarized in the following table:

Technology	**Power Plant Capacity, MWe**	**NO_x Removal, %**	**Capital Cost, $/kW**
LNB (wall-fired)	500	48	6

LNB/OFA (wall-fired)	500	68	13
LNCFS™ (T-fired)	200	37	7
LNCB® (cell burners)	600	50	9
GR	300	67	17
CR (cyclones)	600	50	43
SNOX™	500	90	305*
SNRB™	150	90	260*
SCR	500	80	50
SNCR	200	40	15

* Includes removal of SO_2 and particulates

These capital costs provide a measure of the relative overall economics. Levelized costs, e.g. $/ton of NO_x removed, can also be a useful criterion, but since the economics developed by the CCT project participants were not done with consistent economic assumptions, it is not feasible to make valid comparisons of the several processes based on levelized costs. Therefore the above table does not include this figure.

CONCLUSIONS

A number of processes for control of NO_x from coal-fired boilers have been developed under the auspices of the DOE's CCT Program. No single technology is appropriate for all applications. Choice of technology involves consideration of boiler type, regulatory requirements, and site specific conditions.

Unit Conversions

1 lb/10^6 Btu = 0.43 kg/GJ