

CLEAN COAL TECHNOLOGY



Technologies for the Combined Control of Sulfur Dioxide
and Nitrogen Oxides Emissions from Coal-Fired Boilers

Technologies for the Combined Control of Sulfur Dioxide and Nitrogen Oxides Emissions from Coal-Fired Boilers

A report on three projects conducted under separate cooperative agreements between:

The U.S. Department of Energy and

- ABB Environmental Systems
- The Babcock & Wilcox Company
- Public Service Company of Colorado



ABB



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COAL
TECHNOLOGY**

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Executive Summary

The Clean Coal Technology (CCT) Demonstration Program is a government and industry cofunded effort to demonstrate a new generation of innovative coal utilization processes in a series of “showcase” facilities built across the country. These projects are carried out on a scale sufficiently large to demonstrate commercial worthiness and to generate data for design, construction, operation, and technical/economic evaluation of full-scale commercial applications.

The goal of the CCT Program is to furnish the U.S. energy marketplace with a number of advanced, more efficient coal-based technologies meeting strict environmental standards. These technologies will mitigate the economic and environmental impediments that limit the full utilization of coal as a continuing viable energy resource.

To achieve this goal, beginning in 1985, a multiphased effort consisting of five separate solicitations was administered by the U.S. Department of Energy’s (DOE) Federal Energy Technology Center (FETC). Projects selected through these solicitations have demonstrated technology options with the potential to meet the needs of energy markets while satisfying relevant environmental requirements.

A significant part of this program is the demonstration of technologies designed to simultaneously reduce emissions of nitrogen oxides (NO_x) and sulfur dioxide (SO₂) from existing coal-fired utility boilers. NO_x and SO₂ are acid rain precursors, with NO_x also contributing to atmospheric ozone formation. Ozone is both a health hazard and a major component of smog. NO_x and SO₂ emissions are regulated under the provisions of the 1990 Clean Air Act Amendments (CAAA).

This report discusses three completed CCT projects involving combined SO₂ and NO_x removal processes.

- *SNOX™ Flue Gas Cleaning* was demonstrated at Ohio Edison’s Niles Station, Unit No. 2, where high-sulfur bituminous coal was the fuel. Haldor Topsoe supplied the technology, which consists of NO_x removal by selective catalytic reduction (SCR) and SO₂ removal by oxidation/hydrolysis to make sulfuric acid (H₂SO₄). The objectives of this demonstration project were to achieve greater than 95% SO₂ emissions reduction and greater than 90% NO_x emissions reduction. These objectives were achieved. Distinguishing features of the SNOX™ technology are high pollutant removal efficiencies and the production of sulfuric acid, which avoids the solid wastes associated with processes using sorbent injection.

- *SO_x-NO_x-Rox Box™ (SNRB™) Flue Gas Cleanup* was demonstrated at Ohio Edison’s R.E. Burger Plant, Unit No. 5. This technology, developed by The Babcock & Wilcox Company (B&W), consists of SCR for NO_x control and dry sorbent injection (DSI) for SO₂ control. The objectives of this project were to achieve 90% reduction of NO_x emissions, 70% reduction of SO₂ emissions with calcium-based sorbent and 90% reduction with sodium-based sorbent, and particulate emissions below 0.03 lb/million Btu. These objectives were achieved. A unique feature of the SNRB™ process is that all emissions reductions take place within a high-temperature baghouse.

- *Integrated Dry NO_x/SO₂ Emissions Control System* was demonstrated at Public Service Company of Colorado’s Arapahoe Station, Unit 4. This project demonstrated the integration of four technologies; the first three for NO_x removal and the fourth for SO₂ removal:

- (1) B&W’s DRB-XCL® low-NO_x burners (LNBs)
- (2) overfire air (OFA)
- (3) furnace injection of urea for selective noncatalytic reduction (SNCR)
- (4) DSI (plus humidification, when using calcium-based sorbents).

The project demonstrated that SNCR can be used in tandem with burner modifications to enhance NO_x emissions reduction. The objective of this project was to achieve at least 60% reduction in both NO_x and SO₂ emissions. This objective was achieved.

These three projects address somewhat different commercial applications. The SNOX™ process is well suited for areas where emissions constraints are very stringent and where emissions credits can be taken for reductions beyond those required by the CAAA. An advantage of the SNRB™ process is its simplicity, thus lending itself to smaller, new industrial plants and small utility boiler retrofit applications. The major market for the Integrated System is expected to be older units that fire a low-sulfur (<1%) coal and require both SO₂ and NO_x emissions reductions. All of these technologies have the added advantage that they are effective in removing hazardous air pollutants.

Technologies for the Combined Control of Sulfur Dioxide and Nitrogen Oxides Emissions from Coal-Fired Boilers

Background

The Clean Coal Technology (CCT) Demonstration Program, sponsored by the U.S. Department of Energy (DOE), is a government and industry cofunded technology development effort conducted since 1985 to demonstrate a new generation of innovative coal-utilization processes.

The CCT Program involves a series of “showcase” projects, conducted on a scale sufficiently large to demonstrate commercial worthiness and generate data for design, construction, operation, and economic/technical evaluation of full-scale commercial applications. The goal of the CCT Program is to furnish the U.S. energy marketplace with advanced, more efficient coal-based technologies meeting strict environmental standards. These technologies will mitigate some of the economic and environmental impediments that inhibit the full utilization of coal as an energy source.

Concurrent with the development of the CCT Program by DOE, the U.S. Environmental Protection Agency (EPA) has

promulgated regulations under the 1990 Clean Air Act Amendments (CAAA) to control emissions from a variety of stationary sources, including coal-burning boilers.

The CCT Program has opened a channel to policy-making bodies by providing data from cutting-edge technologies to aid in formulating regulatory decisions. For example, results from several CCT projects have been provided to EPA to help establish achievable nitrogen oxides (NO and NO₂, collectively referred to as NO_x) emissions targets for coal-fired boilers subject to CAAA compliance.

One of the major objectives of the CCT Program is to develop technologies that reduce emissions of sulfur dioxide (SO₂) and NO_x, which are major contributors to acid rain. Many U.S. coals have sulfur and nitrogen contents sufficiently high to generate SO₂ and NO_x emissions exceeding environmental standards. Consequently, the CCT Program includes projects to demonstrate technologies that reduce both SO₂ and NO_x emissions. This report reviews three CCT Program demonstration projects designed to accomplish that objective.

Emissions Standards

History

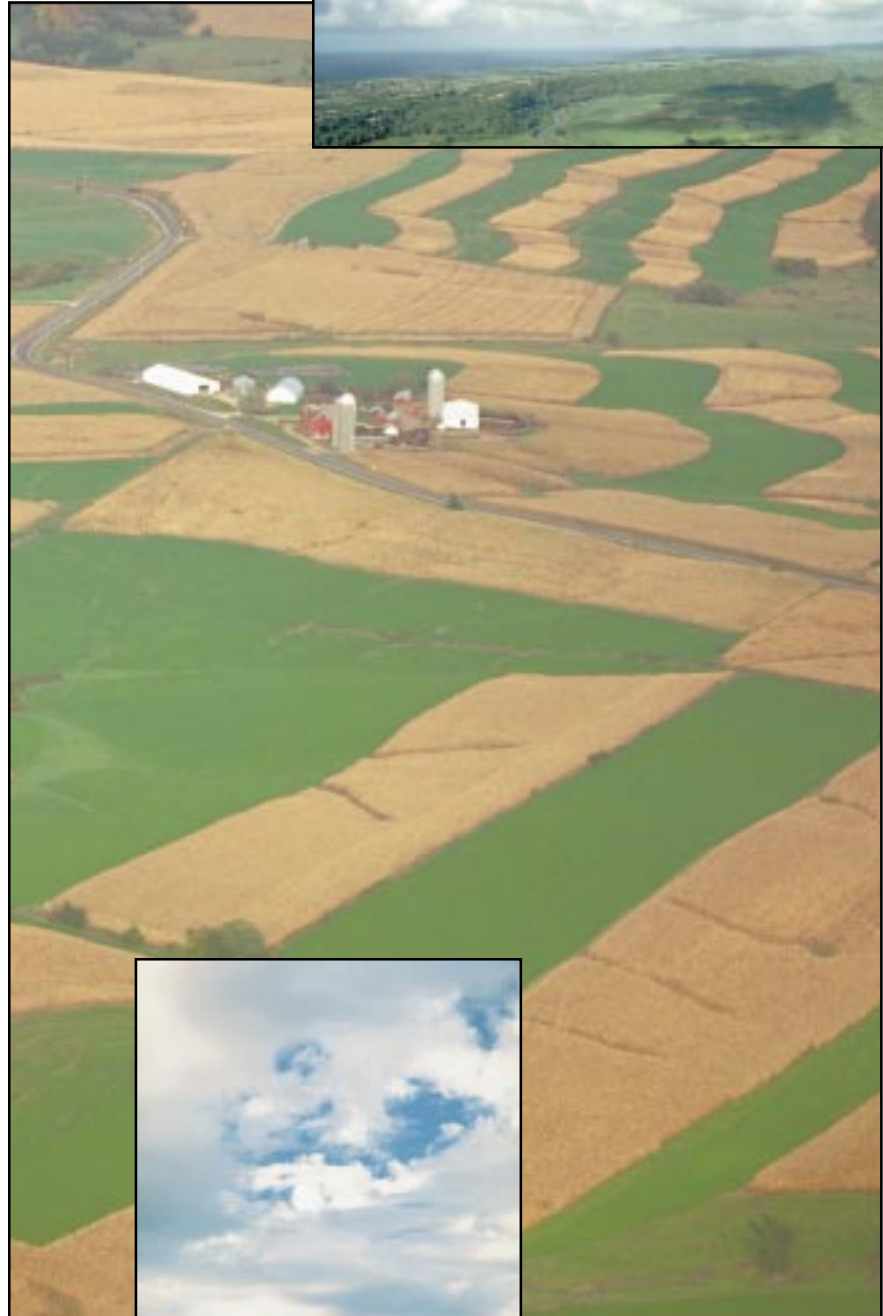
The Clean Air Act was originally passed in 1970, amended in 1977, and most recently in 1990. The 1990 CAAA authorize the EPA to establish standards for a number of atmospheric pollutants, including SO₂ and NO_x. The amendments establish performance standards for existing and new sources of these flue gas components. Updating emissions standards every five years is mandated.

NO_x Emissions Standards

NO_x emissions are generated primarily from transportation, utility, and other industrial sources. They are reported to contribute to a variety of environmental problems, including acid rain and acidification of aquatic systems, ground-level ozone (smog), and visibility degradation. For these reasons, NO_x emissions are regulated by various levels of government throughout the country.

SO₂ Emissions Standards

SO₂ is formed through the combustion of sulfur contained in fossil fuels. Burning typical medium- and high-sulfur coals produces SO₂ emissions that exceed the allowable limits under the CAAA. Two major portions of the CAAA relevant to SO₂ control are Title I and Title IV. Title I establishes National Ambient Air Quality Standards (NAAQS) for six criteria pollutants, while Title IV addresses controls for specific types of stationary boilers, including those found in coal-fired power plants. Title IV is often referred to as the Acid Rain Program.



CCT Program technologies are improving the quality of our environment.

NO_x and SO₂ Control Technologies

NO_x Control Technologies

Techniques for reducing NO_x emissions from fossil-fuel-fired boilers can be classified into two fundamentally different categories: combustion controls and post-combustion controls. Combustion controls reduce NO_x formation during the combustion process, while post-combustion controls reduce NO_x after it has been formed.

Combustion controls include low-NO_x burners (LNBs), reburning, overfire air (OFA), flue gas recirculation (FGR), and operational modifications. Post-combustion controls include selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR).

How NO_x Is Formed in a Boiler

Most of the NO_x formed during the combustion process is the result of two oxidation mechanisms: (1) reaction of nitrogen in the combustion air with excess oxygen at elevated temperatures, referred to as thermal NO_x; and (2) oxidation of nitrogen that is chemically bound in the coal, referred to as fuel NO_x. In addition, minor amounts of NO_x 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 NO_x, referred to as prompt NO_x.

For most coal-fired units, thermal NO_x typically represents about 25% and fuel NO_x about 75% of the total NO_x formed. However, for cyclones and other boilers that operate at very high temperatures, the ratio of thermal to fuel NO_x is different, and thermal NO_x can be considerably higher than fuel NO_x.

The quantity of thermal NO_x formed depends primarily on the “three t’s” of combustion: temperature, time, and turbulence. In other words, 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 NO_x level in the flue gas. Combustion modifications delay the mixing of fuel and air, thereby reducing temperature and initial turbulence, which minimizes NO_x formation.

SO₂ Control Technologies

Most SO₂ control technologies involve the addition of a calcium- or sodium-based sorbent to the system. Under the proper conditions, these materials react with SO₂ and sulfur trioxide (SO₃) to form sulfite and sulfate salts.

Sometimes the sorbent is injected directly into the furnace or flue gas duct, where the dry particles react with SO₂ and are subsequently removed by the boiler’s particulate control device. This is known as Dry Sorbent Injection (DSI). In other cases, the sorbent is dissolved in or slurried with water, and the flue gas contacts the solution or slurry in a scrubber. This approach is referred to as wet flue gas desulfurization (FGD). Because of their low cost, limestone and lime are the most frequently used sorbents.

Another approach, less frequently used, is to oxidize the SO₂ to SO₃ over a catalyst and absorb the SO₃ in water to form sulfuric acid.

Clean Coal Technology Combined SO₂/NO_x Control Demonstration Projects

This report discusses three CCT demonstration projects:

- SNOX™ Flue Gas Cleaning Demonstration Project
- SO_x-NO_x-Rox Box™ (SNRB™) Flue Gas Cleanup Demonstration Project
- Integrated Dry NO_x/SO₂ Emissions Control System

Each of these demonstration projects involves a unique combination of control technologies to achieve reduction of NO_x and SO₂ emissions.

NOx Reduction Technologies

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

Combustion Modifications

Low-NOx Burners — LNBS are designed to control the mixing of fuel and air so as to achieve staged combustion. Lower NOx results from a lower maximum flame temperature and a reduced oxygen concentration during some phases of combustion.

Overfire Air — Overfire air (OFA) is air that is injected into the furnace above the normal combustion zone. OFA is generally used in conjunction with operating the burners at a lower than normal air-to-fuel ratio, which reduces NOx formation. The OFA is then added to achieve complete combustion. OFA is frequently used in conjunction with LNBS.

Reburning — With reburning, part of the boiler heat input (typically 10–30%) is added in a separate reburn zone, where fuel-rich conditions lead to the reduction of NOx formed in the normal combustion zone. OFA is injected above the reburn zone to complete combustion. Thus, with reburn there are three zones in the furnace: (1) a combustion zone with a normal to slightly below normal air-to-fuel ratio; (2) a reburn zone, where added fuel results in a fuel-rich, reducing condition; and (3) a burnout zone, where OFA leads to completion of combustion. Coal, oil, and gas can all be used as the reburn fuel.

Flue Gas Recirculation — FGR, in which part of the flue gas is recirculated to the furnace, can be used to modify conditions in the combustion zone (lowering the temperature and reducing the oxygen concentration) to reduce NOx formation. Another use for FGR is as a carrier to inject fuel into the reburn zone to increase penetration and mixing.

Operational Modifications — These involve changing certain boiler operational parameters to create conditions in the furnace

that will lower NOx production. Examples are burners-out-of-service (BOOS), low excess air (LEA), and biased firing (BF). In BOOS, selected burners are removed from service by stopping fuel flow, but air flow is maintained to create staged combustion in the furnace. LEA involves operating at the lowest possible excess air level while maintaining good combustion, and BF involves injecting more fuel to some burners (typically the lower burners) while reducing fuel to other burners (typically the upper burners) to create staged combustion conditions in the furnace.

Post-Combustion Technologies

Selective Noncatalytic Reduction — In SNCR a reducing agent (typically ammonia or urea) is injected into the furnace above the combustion zone, where it reacts with NOx to form nitrogen gas and water vapor, thus reducing NOx emissions. The critical factors in applying SNCR are sufficient residence time in the appropriate temperature range and even distribution and mixing of the reducing agent across the full furnace cross section.

Selective Catalytic Reduction — In SCR a catalyst vessel is installed downstream of the furnace. Ammonia is injected into the flue gas before it passes over the fixed-bed catalyst. The catalyst promotes a reaction between NOx and ammonia to form nitrogen and water. NOx reductions as high as 90% are achievable, but careful design and operation are necessary to keep ammonia emissions (referred to as NH₃ slip) to a concentration of a few ppm.

Hybrid Process — SNCR and SCR can be used in conjunction with each other with some synergistic benefits. Also, both processes can be used in conjunction with LNBS.



Aerial view of Ohio Edison's Niles Station.

SNOX™ Flue Gas Cleaning Demonstration Project

Project Description

This project was selected during Round II of DOE's CCT Program. In December 1989, ABB Environmental Systems (ABBES) entered into an agreement with DOE to conduct this demonstration. Cofunders of the project were the Ohio Coal Development Office, the Ohio Edison Company, and Snamprogetti, U.S.A. Total project cost was \$31.4 million, with DOE supplying 50% of the funding.

The project was hosted by Ohio Edison at its Niles Station, Unit No. 2, a 108-MWe cyclone-fired boiler.

Haldor Topsøe developed and demonstrated abroad the key process steps of the SNOX™ technology, including SCR, SO₂ conversion, and the Wet-Gas Sulfuric Acid (WSA) tower. With each of these

process steps already demonstrated separately, this project demonstrated the effectiveness of integrating the several technologies to clean the flue gas from a utility burning U.S. coals.

In its usual operating configuration, flue gas from Unit No. 2 passes through a combustion air preheater and an existing electrostatic precipitator (ESP), exiting via a 393-foot stack. For the demonstration project, the SNOX™ unit was fed a flue gas slipstream equivalent to 35 MWe, taken after the boiler's air preheater and prior to the ESP.

The fuels burned during this project were bituminous coals from Ohio and Pennsylvania having a sulfur content of 2.5% to 3.1%. The objective of the demonstration was to achieve 90% or greater NO_x reduction and 95% SO₂ removal and to produce salable sulfuric acid as a by-product, while burning U.S. coals.

Although the demonstration was carried out at a relatively modest capacity, the sizes of the fabric filters used in the baghouse, the catalyst beds, and the equipment used in all of the process operations were representative of full-scale commercial equipment. Furthermore, since the chemical principles involved in NO_x reduction, SO₂ conversion, and acid condensation are independent of plant size, the results of this demonstration project should be applicable to any type or size of boiler.

The testing and operating activities on the SNOX™ demonstration unit spanned a period of 33 months. The unit was continuously operated for periods of up to two months, while a typical run time was several weeks. The unit was on stream for a total of 8000 hours, which included operation at both full and partial load.

Process Description

At operating temperature, the SO₂ conversion catalyst is very effective at particulate removal and retains about 90% of the dust that enters the reactor. Therefore, to avoid frequent catalyst cleaning, the flue gas first passes through a conventional pulse-jet baghouse, where particulates are significantly reduced. Most of the remaining dust is removed in the WSA condenser. After being heated in the flue gas heat

exchanger, the flue gas is sent to an SCR unit for NO_x removal, the first step of the SNOX™ process. In the SCR unit, ammonia (NH₃) is injected into the flue gas and reacts with NO_x over a catalyst to produce nitrogen and water vapor.

From the SCR unit, the flue gas goes to a second catalytic reactor where SO₂ is oxidized to SO₃. Following the SO₂ converter, the flue gas is reduced in temperature by the flue gas heat exchanger. The flue gas

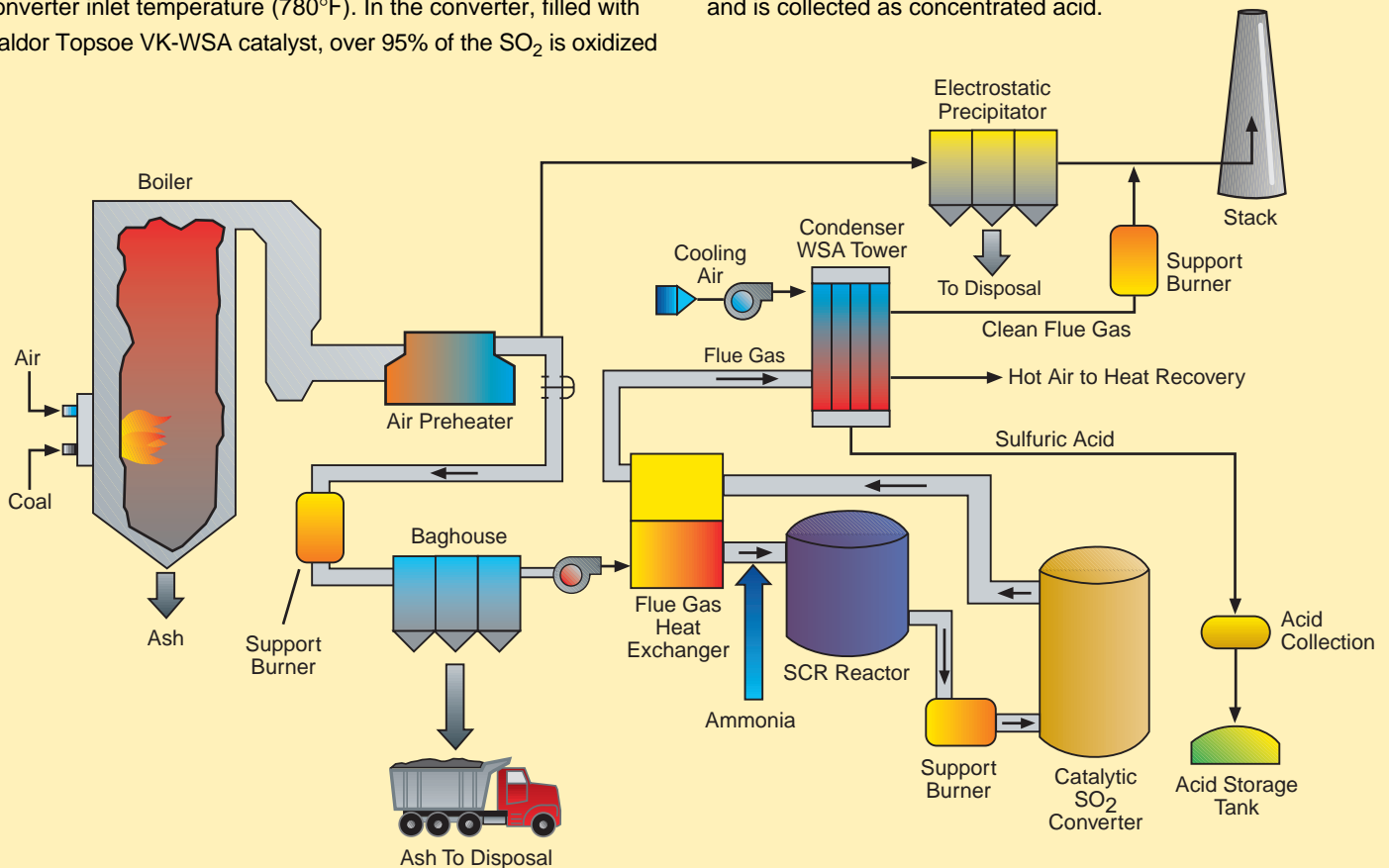
then passes through a novel glass-tube condenser (the WSA tower) where the SO₃ is hydrated to H₂SO₄, which is condensed to produce a high-quality, commercial-grade sulfuric acid.

Although not incorporated in this demonstration project, recovery of the exothermic heat of reaction would be included in future commercial designs, and the projected economics include credit for this recovered heat.

SNOX™ Process Description

Filtered flue gas is heated to the SCR reaction temperature (750°F) in the flue gas heat exchanger. Then in the SCR reactor, nitrogen oxides are selectively reduced with ammonia (NH₃) to elemental nitrogen over a Haldor Topsoe DNX catalyst, a titanium dioxide-based monolith with a high tolerance for both thermal shock and dust. The gas leaving the SCR reactor, containing residual NH₃ and a small amount of fine particulates, is heated with natural gas, oil, or steam to reach the optimum SO₂ converter inlet temperature (780°F). In the converter, filled with Haldor Topsoe VK-WSA catalyst, over 95% of the SO₂ is oxidized

to SO₃. In addition, unreacted NH₃ and unburned hydrocarbons in the flue gas are completely oxidized. This allows a high NO_x removal with a small SCR catalyst volume without having to be concerned about NH₃ emissions. Flue gas leaving the SO₂ converter goes to the hot side of the flue gas heat exchanger, where it is cooled. It is further cooled to about 210°F in the WSA condenser. As the flue gas is cooled, SO₃ and water react exothermically to form H₂SO₄, which condenses and is collected as concentrated acid.



SNOX™ process flow diagram.



Duct section being hoisted into place during SNOX™ technology installation.

SNOX™ SCR reactor.



Results

The SNOX™ unit consistently achieved the 90% NO_x removal target, with NO_x reductions generally being in the range of 93–94%. Although sulfur reduction varied with operating conditions, the 95% SO₂ removal target was also consistently met.

The by-product sulfuric acid had an average concentration of 94.7%, which exceeded the specification of 93.2%. Purity met the Federal Specification for Class I acid. A total of 5600 tons were produced during the demonstration. Some of the acid was used by Ohio Edison in their boiler water demineralizer systems. The rest was purchased by a regional marketer and sold for a variety of uses, such as metal pickling.

Costs

An economic estimate was prepared by ABBES for SNOX™ technology added to an existing 500-MWe power plant. This estimate takes into account improvements based on experience gained from the 35-MWe SNOX™ demonstration and from commercial installations in Europe.

For 95% reduction in SO₂ emissions and 90% reduction in NO_x emissions, the

estimated capital cost is \$305/kW. The levelized cost, which includes a credit (\$25/ton) for sale of the sulfuric acid produced and also a credit (\$2.00/million Btu) for heat recovery from the SNOX™ facilities, is 6.1 mills/kWh or \$198 per ton of NO_x plus SO₂ removed (constant dollars, 15-year project life).

ABBES compared SNOX™ economics with those for a combination of conventional technologies designed to achieve comparable emissions control. For this comparison study, the technologies selected by ABBES were wet-limestone, forced-oxidation FGD for control of SO₂ and SCR for control of NO_x. SNOX™ has a 13% lower capital requirement and over 50% lower operating and maintenance costs.

Conclusions

As a commercially proven technology, SNOX™ provides an innovative alternative system for the simultaneous control and maximum removal of NO_x, SO₂, and particulates. The capital cost is competitive for high removal, multipollutant control options. The operating cost is partially offset by the revenues generated from the by-product H₂SO₄ and by heat recovery from the exothermic reactions involved, making the SNOX™ technology attractive for new and retrofit applications.

Because the SNOX™ technology is applied to the flue gas downstream of the boiler, it is applicable to all electric power plants and industrial/institutional boilers, no matter what fuel is fired, provided both NO_x and SO₂ need to be removed. The only limitation is that sufficient space is required near the boiler flue duct so that the flue gas can be economically transported to the SNOX™ unit, processed, and returned to the stack.

SO_x-NO_x-Rox Box™ (SNRB™) Flue Gas Cleanup Demonstration

Project Description

This project was selected during Round II of DOE's CCT Program. In December 1989, The Babcock & Wilcox Company (B&W) entered into an agreement with DOE to conduct this demonstration. Cofunders of the project were the Ohio Coal Development Office, the Ohio Edison Company, EPRI, the Norton Company, the 3M Company, and Owens Corning Fiberglas Corporation. Total project cost was \$13.3 million, with DOE supplying 46% of the funding.

The project was hosted by Ohio Edison at its R.E. Burger Plant, which is located along the Ohio River in Dilles Bottom, Ohio. There are eight coal-fired boilers at the plant, supplying five generating units.

Flue gas feed to the SNRB™ demonstration unit was a 5-MWe equivalent slipstream from Boiler No. 8. This nominal 160-MWe wall-fired B&W boiler was built in 1955, before implementation of pollution controls on boilers. The flue gas tie-in was between the economizer and the combustion air heater, where the gas temperature was 600–650°F.

The goal of this project was to demonstrate SNRB™ technology for SO₂, NO_x, and particulate removal during extended operation with fully-integrated, commercial-sized components.

Over the period of the demonstration, the boiler was fired with Midwestern bituminous coal having an average sulfur content of about 3.5%.

Operations started in May 1992 and were completed in April 1993. The SNRB™ facility was operated for about 2300 hours, including more than 25 cold startup cycles.



**Aerial view of Ohio Edison's
R.E. Burger Plant.**

Process Description

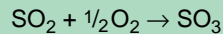
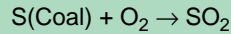
The SNRB™ process combines the removal of SO₂, NO_x, and particulates in one unit, a high-temperature baghouse located between the economizer and the combustion air heater. SO₂ is removed by dry injection of either a calcium- or sodium-based sorbent upstream of the baghouse. Particulates, including the sorbent, are removed by the fabric filter bags. NO_x is removed by SCR. NH₃ is injected upstream of the baghouse, and the cylindrical monolith SCR catalyst is contained within the bags in the baghouse.

An advantage of the SNRB™ approach is a reduction in equipment and space requirements for the emissions control system. Since particulate and SO₂ removals occur upstream of the air heater, fouling and corrosion potential are substantially reduced, allowing the air heater to operate at a lower flue gas outlet temperature. A further advantage is the potential for enhanced energy recovery and improved boiler efficiency.

SO₂ Formation

All coals contain sulfur. Some of this sulfur, known as organic sulfur, is intimately associated with the coal matrix. The rest of the sulfur, in the form of pyrites or sulfates, is associated with the mineral matter. High-sulfur bituminous coals contain up to about 4% sulfur, whereas low-sulfur Western coals may have a sulfur content below 1%.

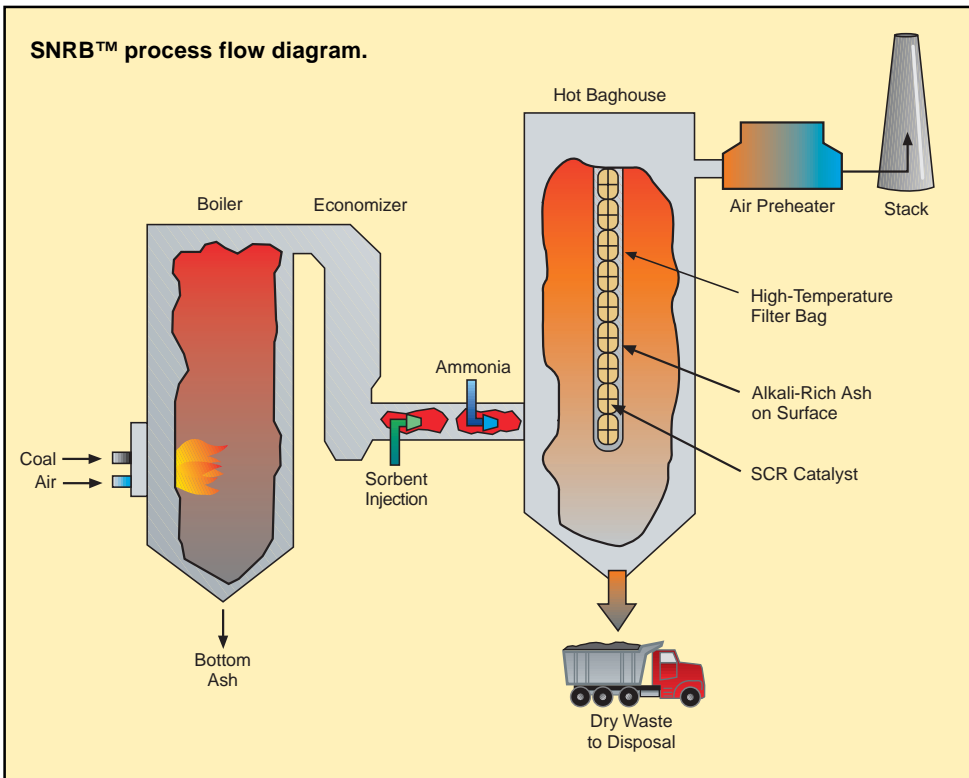
Upon combustion, most of the sulfur is converted to SO₂, with a small amount being further oxidized to sulfur trioxide (SO₃).



Because, in the absence of a catalyst, the formation of SO₃ is slow, over 98% of the combusted sulfur is usually in the form of SO₂.

Effective January 1, 2000, the SO₂ emissions limit for coal-fired power plants is 1.2 lb/million Btu. To comply with this regulation without FGD, the maximum sulfur content for a coal having a higher heating value of 12,000 Btu/lb is 0.72% by weight, assuming 100% conversion of sulfur to SO₂.

SNRB™ process flow diagram.



In some SNRB™ retrofit applications, modification of the heat transfer surfaces in the economizer and air heater may be necessary to optimize overall plant efficiency and emissions control performance. The economizer, which is usually the last water-cooled heat transfer surface in the boiler, heats the boiler feed water, and the air preheater recovers heat from the flue gas to heat the combustion air.

Results

At baghouse operating temperatures of 830°F and higher, use of a commercial hydrated lime sorbent at a Ca/S molar ratio of 1.8 or above resulted in over 80% SO₂ removal, with 40% to 45% calcium utilization. This is significantly improved performance compared with the 60% removal at 30% sorbent utilization typical of other dry calcium-based sorbent injection processes. This improved performance is due to enhanced contacting of flue gas with sorbent as the flue gas passes through the sorbent layer built up on the filter bags.

For sodium-based sorbents, two moles of sorbent are required per mole of SO₂ removed. To put sodium on the same basis

as calcium, the normalized stoichiometric ratio (NSR) is used. The NSR is defined as the ratio of the moles of Na injected to the moles of SO₂ in the flue gas divided by 2. Thus, an Na/S ratio of 2.0 is equal to an NSR of 1.0. In the test program, sodium-based sorbents achieved over 90% removal efficiency at an NSR of 1.0, with a sorbent utilization of 85%.

Ninety percent NO_x removal with less than 5 ppm NH₃ slip was achieved over the design temperature range (700–900°F) of the zeolite catalyst. NH₃ slip is defined as the amount of NH₃ remaining in the stack gas after NO_x reduction. Neither temperature nor flue gas flow rate had much effect on NO_x removal over the operating range evaluated. NO_x removal over the range from 50–95% was easily controlled by varying the rate of NH₃ injection.

A concern with SCR is the oxidation of SO₂ to SO₃, since SCR catalysts are capable of promoting this oxidation reaction. This can lead to downstream deposition of ammonium sulfates with attendant equipment fouling and corrosion. A goal of SCR catalyst development is to minimize this side reaction. SNRB™ provides the distinct advantage of significantly reducing the

amount of SO₂ in the flue gas before the gas contacts the SCR catalyst. Furthermore, in the SNRB™ demonstration, less than 0.5% of the SO₂ passing over the SCR catalyst was converted to SO₃. Consequently, the production of SO₃ in the SCR unit was very low.

The particle removal rate for the high-efficiency fabric filter baghouse exceeded 99%. Hazardous air pollutants (HAPs) testing showed high capture efficiencies in the baghouse for most trace elements.

In three periods of planned continuous operation for more than 200 hours each, system availability averaged 99%. No degradation of catalyst or filter bags was observed during the demonstration, and the SNRB™ unit had no effect on boiler performance, since it involved only post-combustion treatment of a flue gas slipstream.

A key initial market for the SNRB™ technology consists of retrofits to existing boilers with generating capacities of



SNRB™ baghouse with catalyst holder tube ready for lifting into baghouse.

Hazardous Air Pollutants

Hazardous air pollutants (HAPs), also referred to as toxic air pollutants or air toxics, are generally defined as atmospheric pollutants that are known or suspected to cause serious health problems. HAPs are emitted by motor vehicles and a variety of industrial sources and may exist as particulate matter or as gases. HAPs include metals and other particulates, gases adsorbed on particulates, and certain vapors, such as benzene, from fuels and other sources.

For coal-fired power plants, the HAPs of most concern are metals such as arsenic, cadmium, mercury, selenium, and vanadium, present in trace quantities in the mineral matter in coal. There is also concern over certain other elements such as fluorine.

DOE conducted on EPA's behalf a study to investigate the fate of HAPs at a number of coal-fired plants utilizing a variety of air pollution control technologies. The objective was to see how effective these technologies are for removing HAPs from flue gas. The CCT Program has made a significant contribution to this study through the participation of a number of its projects.

Emissions Standards

History

The Clean Air Act of 1970 established a major air regulatory role for the federal government. The Act was further extended by amendments in 1977 and most recently in 1990. The 1990 CAAA is one of the most complex and comprehensive pieces of environmental legislation ever written. It authorizes EPA to establish standards for a number of atmospheric pollutants, including sulfur dioxide (SO₂) and NO_x. Two major portions of the CAAA relevant to SO₂ and NO_x control are Title I and Title IV.

Title I

Title I establishes National Ambient Air Quality Standards (NAAQS) for six criteria pollutants, including SO₂, NO_x, and ozone (O₃). The NAAQS for ozone is 0.08 ppm (eight-hour average), and the NAAQS for SO₂ is 0.14 ppm (24-hour average).

NO_x and volatile organic compounds (VOCs) in the atmosphere react in the presence of sunlight to form ground-level O₃, which is a major ingredient of smog. Many urban areas do not meet the O₃ standard and are classified as nonattainment. A large number of power plants are situated within these nonattainment areas. This nonattainment status is attributable not only to NO_x emissions in a given locality but also to significant amounts of NO_x and VOCs transported by wind over a wide geographical region.

To address regional pollutant transport, EPA issued a rule governing NO_x emissions from electric power plants and other large stationary boilers in 22 Eastern states and the District of Columbia. EPA's rule sets statewide NO_x emissions budgets, which include budget components for the electric

power industry and certain industrial stationary sources. These sources are expected to make large NO_x emissions reductions to decrease transport of pollutants from one region of the country to another. The target NO_x emissions limit for utility boilers is 0.15 lb/million Btu. States must develop State Implementation Plans (SIPs) for NO_x to achieve the required statewide emissions budgets.

Title IV – The Acid Rain Program

The overall goal of the Acid Rain Program is to achieve environmental and public health benefits through reductions in emissions of SO₂ and NO_x. Both the NO_x and SO₂ control programs use a two-phase approach to achieve compliance.

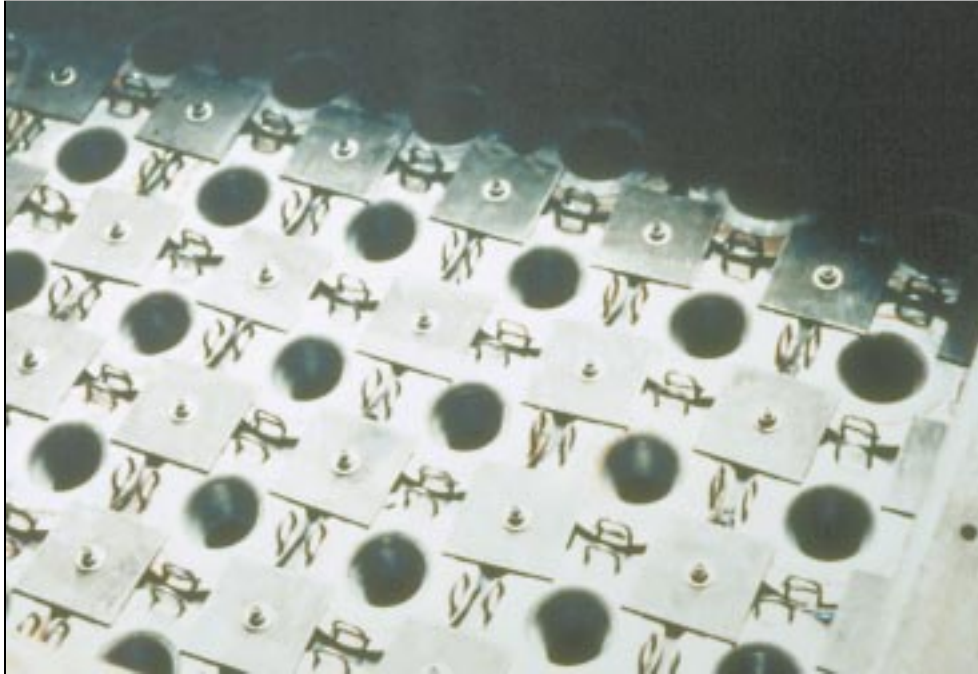
For NO_x control, Title IV focuses on a particular set of NO_x emitting sources—coal-fired electric utility plants. Phase I of the program, begun in 1996, has reduced NO_x emissions in the United States by over 400,000 tons/year. These reductions were achieved by the installation of low-NO_x burner (LNB) technology on dry-bottom, wall-fired boilers and tangentially fired (T-fired) boilers (Group 1). In Phase II, which begins in 2000, EPA has established lower emissions limits for Group 1 boilers and established limits for Group 2 boilers. Group 2 boilers include cell-burners, cyclones, wet-bottom boilers, and other types of coal-fired boilers. It is projected that the more stringent Phase II limits will result in an additional NO_x reduction of 820,000 tons/year.

The statute requires that NO_x emissions control costs for Group 2 boilers be comparable to the costs for Phase I, Group 1 boilers. The regulations allow for emissions averaging in which the emissions levels established by EPA are applied to an entire group of boilers

owned or operated by a single company.

A primary goal of the SO₂ control program is the reduction of annual SO₂ emissions by 10 million tons below 1980 levels. Phase I, which began in 1995, affects 263 units at 110 mostly coal-burning electric utility plants located in 21 Eastern and Midwestern states. An additional 182 units joined the program as substitution or compensating units, bringing the total of Phase I affected units to 445. Phase II, which begins in 2000, tightens the annual emissions limits and also sets restrictions on smaller plants fired by coal, oil, and gas. The Title IV, Phase I SO₂ emissions limit is 2.5 lb/million Btu. This decreases to 1.2 lb/million Btu in Phase II.

The Acid Rain Program introduces flexibility in achieving compliance through an allowance trading system that harnesses the incentives of the free market to reduce pollution. Affected utility units have been allocated allowances based on their historic fuel consumption. Each allowance permits emitting one ton of SO₂. For each ton of SO₂ discharged in a given year, one allowance is retired. Allowances may be bought, sold, or banked, and anyone may acquire allowances and participate in the trading system. However, regardless of the number of allowances held, a source may not emit pollutants at levels that would violate federal or state standards, including ambient air standards set under Title I to protect public health. In Phase II, the CAAA sets a permanent ceiling (or cap) of 8.95 million annual allowances allocated to utilities. This cap firmly restricts emissions and ensures that environmental benefits will be achieved and maintained.



SNRB™ baghouse catalyst bag insert holder.

100–200 MWe, representing a total U.S. market of about 15,000–20,000 MWe.

Costs

B&W prepared an economic estimate for a SNRB™ retrofit to a 150-MWe boiler, incorporating improvements based on experience gained from the 5-MWe demonstration. For 85% reduction in SO₂ emissions and 90% reduction in NO_x emissions, the estimated capital cost is \$253/kW. For a 15-year project life, the levelized cost on a constant dollar basis is 12.1 mills/kWh, equivalent to \$553/ton of SO₂ plus NO_x removed.

B&W compared SNRB™ economics with those for a combination of conventional technologies designed to achieve comparable emissions control. For a 100-MWe plant burning 1.5% sulfur coal, SNRB™ has significantly lower capital and levelized costs than a convention system consisting of separate dry lime scrubber, SCR, and fabric filter units.

Conclusions

The SNRB™ test program demonstrated the feasibility of controlling multiple emissions from a coal-fired boiler in a single processing unit. The emissions reductions for SO₂, NO_x, and particulates all exceeded the project goals. The SNRB™ system offers operating flexibility, control of multiple pollutants, and low space requirements.

Despite these advantages, the SNRB™ process probably would not be an economic choice for applications requiring SO₂ removals above about 85%. For lower levels of SO₂ removal, the economics for SNRB™ are more favorable than for installing separate units for the same levels of removal of SO₂, NO_x, and particulates.



View of Public Service Company of Colorado's Arapahoe Station.

Integrated Dry NO_x/SO₂ Emissions Control System

Project Description

This project was selected during Round III of DOE's CCT Program. In March 1991, Public Service Company of Colorado (PSCo) entered into an agreement with DOE to conduct this project. EPRI was a cofunder. Total project cost was \$27.4 million, with DOE supplying 50% of the funding.

PSCo hosted the project at its Arapahoe Station, Unit 4, in Denver, Colorado. The station includes four coal-fired steam generating units with a total capacity of 232 MWe. Unit 4 is a 100-MWe top-fired boiler with the burners mounted vertically on the roof. During the demonstration, Unit 4 burned two low-sulfur (0.4%)

Colorado bituminous coals (Cyprus Yampa Valley and Empire Energy).

The Dry NO_x/SO₂ Emissions Control System integrates four technologies to control NO_x and SO₂ emissions. For this project, the boiler was retrofitted with 12 B&W DRB-XCL[®] burners and six OFA ports 20 feet below the furnace roof. Two levels of SNCR injectors were installed, supplemented later with two lance injectors.

A dry sorbent injection (DSI) system was added, as well as

a new distributed control system and conversion of the fly ash collection system from wet to dry. This project was the first U. S. commercial-scale demonstration of low-NO_x burners (LNBS) on a top-fired boiler.

The test program began in August 1992 and was completed in November 1997. The project operated for more than 34,000 hours after the combustion modifications were installed.

Process Description

The four control technologies that make up the Integrated Dry NO_x/SO₂ Emissions Control System are LNBS, OFA, SNCR, and DSI plus flue gas humidification. NO_x reduction is accomplished through LNBS, OFA, and SNCR, while SO₂ control is achieved by DSI (using either calcium- or sodium-based sorbents) plus flue gas humidification with calcium-based sorbents (used to enhance SO₂ removal capabilities). NO_x reduction occurs in the furnace, while

SO₂ control is carried out in the ductwork downstream of the air preheater and upstream of the fabric filter dust collector.

Results

Before any modifications were installed, baseline tests of the original combustion system were made. Then, in order to adequately evaluate the contribution of each process to the Integrated System, the testing program addressed the performance of individual technologies, as well as various process combinations.

The SNCR process was tested with both urea and NH₃ injection, and DSI was tested with both calcium-based (calcium hydroxide) and sodium-based (sodium bicarbonate, sodium sesquicarbonate) sorbents. Flue gas humidification was used with the calcium-based sorbent.

Because the OFA ports could not be completely closed, it was not possible to test the LNBs without OFA, but they were tested with maximum and minimum OFA.

Additional tests were performed during urea, calcium, and sodium injection to determine the potential of these technologies for removing HAPs.

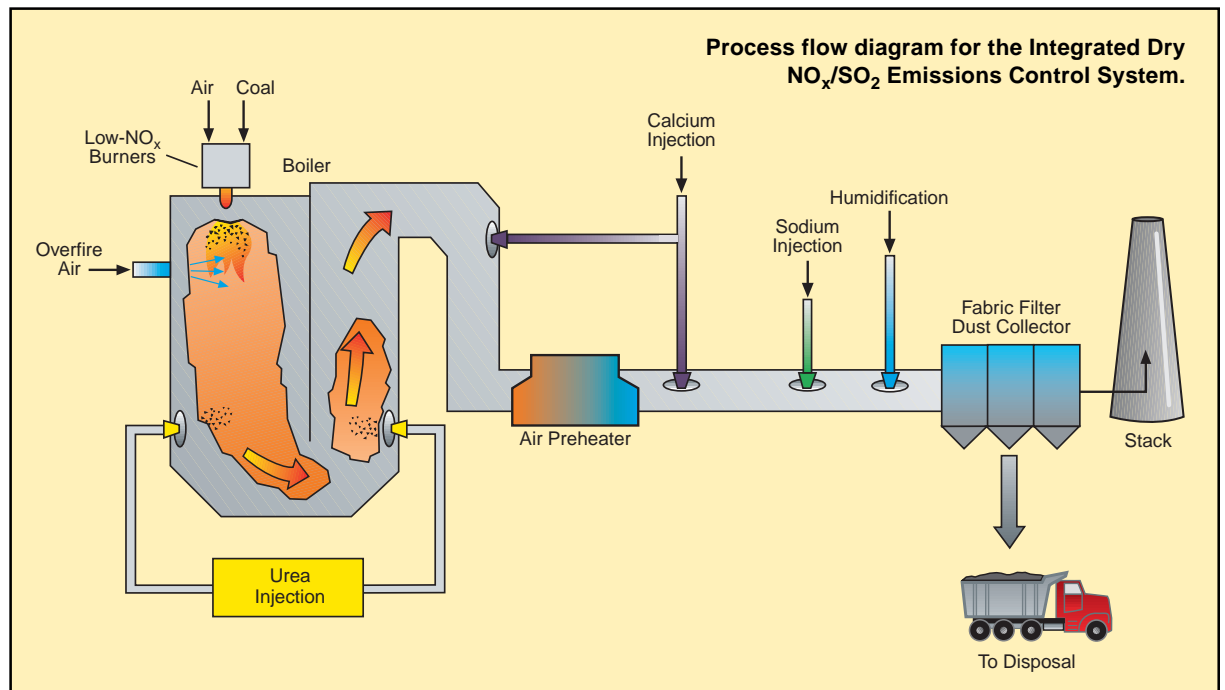
LNBs with OFA achieved a NO_x reduction of 62% to 69% across the 50 to 100-MWe load range. SNCR, using both stationary and retractable furnace injection lances, provided NO_x removals of 30% to 50% at a residual NH₃ content of 10 ppm. This increased the total NO_x reduction to greater than 80%. A residual NH₃ content of 10 ppm would be unsatisfactory from the standpoint of

atmospheric pollution and fly ash quality, but, as discussed subsequently, the incorporation of DSI in the process flow mitigated this problem.

Sodium bicarbonate injection achieved approximately 70% SO₂ removal at an NSR of 1.0. Sodium sesquicarbonate injection achieved the same SO₂ removal, but at an NSR of 2.0. Calcium hydroxide injection was less effective.

Sodium-based sorbents promote the oxidation of NO to NO₂. Although this does not increase the level of NO_x emissions, it can result in a visible plume. However, when sodium-based DSI was combined with SNCR, as in this demonstration, NO₂ emissions did not increase. Furthermore, the DSI system adsorbed most of the NH₃ in the flue gas, thus permitting optimum operation of the SNCR system without having to be concerned about residual NH₃.

During HAPs tests, the fabric filter successfully removed nearly all trace metal emissions, including 80% of the mercury.





Tip of low-NOx burner installed on Unit 4 at Arapahoe Station.



Sodium sorbent injection piping at Arapahoe Station.

Costs

For the Integrated Dry NO_x/SO₂ Emissions Control System, the capital cost is estimated to be \$190/kW for a 100-MWe unit burning low-sulfur coal. Levelized costs are not available.

Conclusions

This project demonstrated the first integration of sodium-based DSI and urea-based SNCR. The integration of these two technologies provides a synergistic effect. PSCO has patented the integration of these processes and intends to license third parties to market and install this technology. The Integrated Dry NO_x/SO₂ Emissions Control System successfully demonstrated the application of B&W DRB-XCL[®] low-NO_x burners to down-fired units with further decrease in NO_x emissions through use of SNCR.

The Integrated System is applicable to most utility coal-fired units and provides a lower capital cost alternative to conventional wet FGD processes for SO₂ control plus SCR for NO_x control. This system can be applied to any size unit, but is most applicable to older, small- to mid-size units.

Conclusions

Most coal-fired boilers are faced with the problem of controlling NO_x and SO₂ emissions. The three CCT projects reviewed in this report provide effective and innovative approaches to the combined control of these pollutants.

The SNOX[™] technology, through its use of SCR and a novel catalytic reactor/WSA tower to convert SO₂ to sulfuric acid, is able to achieve 90% or greater removals of both NO_x and SO₂. Its market potential will be in situations where stringent controls on emissions exist and there is a nearby market for the by-product sulfuric acid.



Urea injection system piping and compressor.

The SNRB™ technology is unique in that it combines NO_x, SO₂, and particulate removal into one unit with a relatively small space requirement. With a sodium-based sorbent and a high enough NH₃/NO_x ratio, 90% or greater removals of both NO_x and SO₂ are possible.

An advantage of the Integrated Dry NO_x/SO₂ Emissions Control System is that all emissions control takes place either in the furnace or the flue gas duct, so that little additional plot area is required. With this system, NO_x was reduced by 80% and SO₂ by 70%. The major market is expected to be older units that fire a low-sulfur (<1%) coal and require both SO₂ and NO_x reductions. While this is not a large market, the significant savings that are possible over competing technologies will provide a niche market for this technology.

These three technologies are potentially applicable to flue gas cleaning for all types of conventional coal-fired units, including stoker, cyclone, and pulverized coal-fired boilers. They are capable of high reduction

The Clean Coal Technology Program

The Clean Coal Technology (CCT) Program is a unique partnership between the federal government and industry that has as its primary goal the successful introduction of new clean coal utilization technologies into the energy marketplace. With its roots in the acid rain debate of the 1980s, the Program has met its objective of broadening the range of technological solutions available to eliminate acid rain concerns associated with coal use. Moreover, the program has evolved and has been expanded to address the need for new, high-efficiency power-generating technologies that will allow coal to continue to be a fuel option well into the 21st century.

Begun in 1985 and expanded in 1987 consistent with the recommendation of the U.S. and Canadian Special Envoys on Acid Rain, the program has been implemented through a series of five nationwide competitive solicitations. Each solicitation has been associated with specific government funding and program objectives. After five solicitations, the CCT Program comprises a total of 40 projects located in 18 states with a capital investment value of nearly \$6 billion. DOE's share of the total project costs is about \$2 billion, or approximately 34 percent of the total. The projects' industrial participants (i.e., the non-DOE participants) are providing the remainder—nearly \$4 billion.

Clean coal technologies being demonstrated under the CCT Program are establishing a technology base that will enable the nation to meet more stringent energy and environmental goals. Most of the demonstrations are being conducted at commercial scale, in actual user environments, and under circumstances typical of commercial operations. These features allow the potential of the technologies to be evaluated in their intended commercial applications. Each application addresses one of the following four market sectors:

- Advanced electric power generation
- Environmental control devices
- Coal processing for clean fuels
- Industrial applications

Given its programmatic success, the CCT Program serves as a model for other cooperative government/industry programs aimed at introducing new technologies into the commercial marketplace.

levels for the three major pollutants of concern: NO_x, SO₂, and particulates.

The commercial viability of these technologies has been demonstrated by these CCT projects, and they are ready to enter the marketplace as more stringent pollution regulations require their use.

SO₂ Emissions Control Technologies

Wet Scrubbing

Wet scrubbing, or wet flue gas desulfurization (FGD), is the most frequently used technology for post-combustion control of SO₂ emissions. Wet FGD is frequently added to existing boilers and has the advantage that no modifications to either the boiler or the particulate emissions control device are required. Typically, the flue gas is contacted with an aqueous slurry of limestone (CaCO₃) in a countercurrent absorber (scrubber), where the SO₂ reacts to form CaSO₃, which is then oxidized to CaSO₄ (gypsum).

Gas flow per unit cross sectional area, which determines scrubber diameter, must be low enough to minimize entrainment. Mass transfer characteristics of the system determine absorber height. These vessels and the accompanying equipment used for slurry recycle, gypsum dewatering, and product conveyance tend to be quite large.

Some variations of this technology produce high quality gypsum for sale. Less pure waste product may be sold for use in cement production. If neither of these options is practiced, the scrubber waste must be disposed of in a sludge pond or similar facility.

Dry and Semidry Sorbent Injection

A reactive calcium- or sodium-based sorbent is injected into the economizer or flue gas duct to react directly with the SO₂ in the flue gas.

The two most common calcium-based sorbents are limestone and slaked lime, Ca(OH)₂. Limestone, which generally requires a higher reaction temperature, is usually injected as a dry powder. Lime, on the other hand, is usually handled as a slurry that dries as soon as it is injected into the hot flue gas. This is referred to as semidry scrubbing, which dominates the sorbent injection market. All commercial semidry systems in the U.S. use lime and recycled fly ash as sorbent. These systems account for 8% to 10% of the installed FGD capacity in the U.S.

Upon injection, Ca(OH)₂ immediately begins to dehydrate. The escaping water vapor creates internal pores that provide access for SO₂ diffusion into the interior of the particles. The CaO produced by dehydration reacts with SO₂ to give CaSO₃, which can be oxidized to CaSO₄.

Typical sodium-based sorbents are sodium bicarbonate (NaHCO₃), sodium sesquicarbonate (NaHCO₃•Na₂CO₃•2H₂O), and sodium carbonate (Na₂CO₃). Below 300°F, NaHCO₃ reacts immediately with SO₂ to form Na₂SO₃. At higher temperatures, NaHCO₃ decomposes to Na₂CO₃ before reacting with SO₂.

As water and CO₂ are given off by the sorbent particles, additional surface area becomes available for reaction with SO₂. Although CO₂ is a product of CaCO₃, NaHCO₃, and Na₂CO₃ decomposition, the amount generated is minimal compared to that already present in the flue gas.

In some cases, flue gas humidification may be necessary for proper operation of the downstream particulate removal system.

Production of Sulfuric Acid

In this process option, the SO₂ in the flue gas is first converted to SO₃ by passing the flue gas over a catalyst bed. The SO₃ then reacts with water to form sulfuric acid, which is recovered for use or sale.

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List of Acronyms and Abbreviations

ABBES	ABB Environmental Systems	kW	kilowatt
BF	Biased firing	kWh	kilowatt hour
BOOS	Burners-out-of-service	LEA	Low excess air
Btu	British thermal unit	LNBS	Low-NOx burners
B&W	The Babcock & Wilcox Company	MWe	Megawatts of electric power
CAAA	Clean Air Act Amendments of 1990	NAAQS	National Ambient Air Quality Standards
CaCO ₃	Calcium carbonate (limestone)	Na ₂ CO ₃	Sodium carbonate
CaO	Calcium oxide (lime)	NaHCO ₃	Sodium bicarbonate
Ca(OH) ₂	Calcium hydroxide (slaked lime)	Na ₂ SO ₃	Sodium sulfite
CaSO ₃	Calcium sulfite	NH ₃	Ammonia
CaSO ₄	Calcium sulfate	NO _x	Nitrogen oxides
CCT	Clean Coal Technology	NSR	Normalized stoichiometric ratio
CO ₂	Carbon dioxide	OFA	Overfire air
DOE	U.S. Department of Energy	ppm	Parts per million
DSI	Dry sorbent injection	PSCo	Public Service Company of Colorado
EPA	U.S. Environmental Protection Agency	SCR	Selective catalytic reduction
EPRI	Electric Power Research Institute	SIP	State Implementation Plan
ESP	Electrostatic precipitator	SNCR	Selective noncatalytic reduction
FETC	Federal Energy Technology Center	SO ₂	Sulfur dioxide
FGD	Flue gas desulfurization	SO ₃	Sulfur trioxide
FGR	Flue gas recirculation	SNRB™	SO _x -NO _x -Rox Box™ Process
GR	Gas reburning	VOCs	Volatile organic compounds
HAPs	Hazardous air pollutants	WSA	Wet-Gas Sulfuric Acid
H ₂ SO ₄	Sulfuric Acid		