Selective Catalytic Reduction (SCR) Technology for the Control of Nitrogen Oxide Emissions from Coal-Fired Boilers

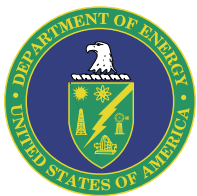## An Update of Topical Report Number 9

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CLEAN COAL TECHNOLOGY

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Executive Summary

The Clean Coal Technology Demonstration Program (CCTDP) is a government and industry co-funded effort to demonstrate a new generation of innovative coal utilization processes in a series of facilities built across the country. These projects are carried out on a commercial scale to prove technical feasibility and provide information for future applications. The goal of the CCTDP is to furnish the marketplace with a number of advanced, more efficient coal-based technologies that meet strict environmental standards. Use of these technologies is intended to minimize the economic and environmental barriers that limit the full utilization of coal.

To achieve this goal, beginning in 1985, a multi-phased effort consisting of five separate solicitations was administered by the U.S. Department of Energy (DOE) through its National Energy Technology Laboratory (NETL). Projects selected through these solicitations have demonstrated technology options with the potential to meet the needs of energy markets while satisfying relevant environmental requirements.

Part of this program is the demonstration of technologies designed to reduce emissions of oxides of nitrogen (NO_x) from existing coal-fired utility boilers. NO_x is an acid rain precursor and a contributor to the formation of ground-level atmospheric ozone, which is a health hazard and is also related to smog formation. NO_x emissions are regulated under the provisions of the Clean Air Act Amendments (CAAA) of 1990. In 1995, a CCTDP project was concluded that demonstrated selective catalytic reduction (SCR) technology for the control of NO_x emissions from high-sulfur, coal-fired boilers. The project was conducted by Southern Company Services, Inc., who served as a co-funder and as the host at Gulf Power Company's Plant Crist.

The SCR process consists of injecting ammonia (NH_3) into boiler flue gas and passing the flue gas through a catalyst bed where the NO_x and NH_3 react to form nitrogen and water vapor. The results of the CCTDP project confirmed the applicability of SCR for U.S. coal-fired power plants. In part as a result of the success of this project, a significant number of commercial SCR units have been installed and are operating successfully in the United States. By 2007, the total installed SCR capacity on U.S. coal-fired units will number about 200, representing about 100,000 MWe of electric generating capacity. This report summarizes the status of SCR technology.
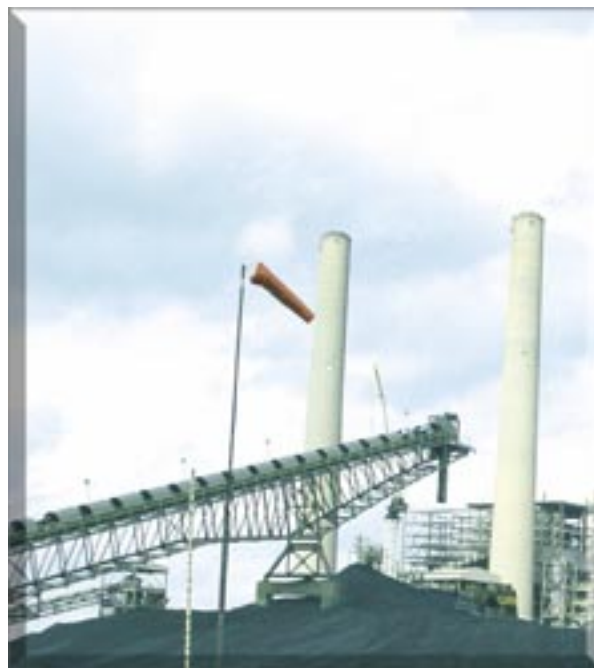
Winyah Generating Station.

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Background

The Clean Coal Technology Demonstration Program (CCTDP), sponsored by the U.S. Department of Energy (DOE) and administered by the National Energy Technology Laboratory (NETL), has been conducted since 1985 to develop innovative, environmentally friendly coal utilization processes for the world energy marketplace. The CCTDP, which is co-funded by industry and government, involves a series of demonstration projects that provide data for design, construction, operation, and technical/economic evaluation of full-scale applications. The goal of the CCTDP is to enhance the utilization of coal as a major energy source.

A major environmental issue in the generation of electric power is the emission of nitrogen oxides, collectively referred to as NO_x. NO_x consists primarily of nitric oxide (NO) and lesser amounts of nitrogen dioxide (NO_2). Among the chief technologies available for reduction of NO_x emissions is selective catalytic reduction (SCR). SCR technology was initially developed in the United States and then commercialized overseas, primarily in Japan and Europe. A major advantage of SCR is that the reaction products, nitrogen and water, are innocuous compounds already present in the air.

In 1995, a project was concluded under the CCTDP that evaluated the performance and economics of the SCR process for removing NO_x from the flue gas of boilers fired with U.S. high-sulfur coals. At that time there were uncertainties, primarily involving the formation of excessive amounts of ammonium-sulfur compounds with attendant plugging and corrosion of downstream equipment. There was also concern over the presence of trace metals in the coal, such as arsenic, since they can lead to catalyst deactivation.

The CCTDP project evaluated the performance and cost of SCR technology under typical U.S. boiler conditions. Over

SCR units at Cross 1 and 2.

a two-year test period at Southern Company's Plant Crist, eight commercially available catalysts were exposed to a slipstream of flue gas extracted from the inlet duct of the hot side electrostatic precipitator (ESP), thus containing full particulate loading. The fuel at Plant Crist was a typical Midwestern coal (Illinois No. 6) having 2.3% sulfur. The SCR test reactors were small-scale units that simulated full-scale operation in terms of the major variables of temperature and space velocity. Anhydrous ammonia (NH_3) was metered into a stream of heated dilution air and injected via nozzles into the flue gas upstream of each SCR reactor.

All of the catalysts performed well in both parametric and long term testing, achieving at least 80% NO_x removal. The success of this project provided a major impetus for widespread adoption of SCR on coal-fired boilers in the United States. Since that time, SCR has become the predominant post-combustion technology for meeting increasingly stringent NO_x emissions regulations.

NO_x Control Technologies

When the Clean Air Act Amendments (CAAA) of 1990 were first enacted, the primary technology available for NO_x reduction in utility boilers was combustion modification using low-NO_x burners (LNBs), frequently combined with overfire air (OFA). These technologies proved adequate to meet CAAA Title IV emissions requirements. In fact, field experience with combustion modification technologies demonstrated in the CCT Demonstration Program provided the data required to establish Title IV regulations. However, LNBs (with or without OFA) are not able to meet Title I target levels. As a result, utilities subject to the most stringent requirements are now using post-combustion technologies such as SCR, either alone or in combination with LNBs. To meet the further demands of the 2005 Clean Air Interstate Rule, more extensive application of SCR will be required, either alone or in combination with combustion modification technologies and advanced plant automation systems.

NO$_x$ Emissions Regulations

Technology development for NO$_x$ reduction is being forced by no less than three regulatory drivers based on the Clean Air Act Amendments (CAAA) of 1990, administered by the U.S. Environmental Protection Agency (EPA): Title IV, Title I, and the 2005 Clean Air Interstate Rule (CAIR).

- CAAA Title IV, addressing acid rain (SO$_2$ and NO$_x$), requires industrial and utility boilers to reduce NO$_x$ emissions in two phases, incorporating an allowance cap-and-trade approach. Phase I took effect in 1996, followed by a stricter Phase II in 2000. NO$_x$ emissions limits were established for each major boiler type. The more stringent limits in Phase II range from 0.40 lb/million Btu for tangentially fired boilers to 0.86 lb/million Btu for cyclone boilers larger than 155 MWe.

- CAAA Title I is more complex. It focuses on emissions of NO$_x$ as an ozone precursor. The CAAA specifies National Ambient Air Quality Standards (NAAQS) for ground-level concentrations of a number of atmospheric pollutants, including ozone. Title I addresses pollution control in areas that are designated as non-attainment with respect to ozone. Under Sec. 110 of Title I, EPA on November 7, 1997 proposed a State Implementation Plan (SIP) Call that specified NO$_x$ emission budgets for utility boilers in 22 eastern and midwestern states that corresponded to a NO$_x$ emission limit of about 0.15 lb/million Btu or about an 85% reduction from 1990 levels. The SIP Call also permitted trading of emission allowances between affected sources. These states were shown by the Ozone Transport Assessment Group (OTAG) to significantly contribute to ozone non-attainment in downwind states. The Title I emissions limits apply to the five-month ozone season (May 1–September 30). Implementation of Title I is designed to bring all applicable states into attainment.

Under the SIP Call, the states were required to have their emissions reduction measures, as specified in their respective SIPs, operable by May 31, 2003. Ultimately, the number of states was reduced to 19 and the compliance date was delayed to May 31, 2004. Georgia and Missouri, previously exempted, have a due date of May 1, 2007. The effect of the SIP Call was to significantly reduce NO$_x$ emissions from electric generating units irrespective of boiler type. All of the SIPs have been approved by the EPA.

- Under Section 110 of Title I, on March 10, 2005, EPA promulgated the CAIR. The CAIR is more stringent than the 1997 SIP Call because the NO$_x$ emissions cap is reduced to 0.125 lb/million Btu and is applied on an annual basis rather than only during the ozone season. The CAIR will take effect beginning in 2010 and will apply to 29 eastern states and the District of Columbia. Reductions in NO$_x$ emissions will be achieved through revised SIPs and a cap-and-trade program patterned after the one used successfully under Title IV. Alternative regulatory scenarios also have been proposed, but in any event it seems certain that control of NO$_x$ emissions will be increasingly more stringent.

The strictest NO$_x$ emission limit under Title IV is 0.40 lb/million Btu (for tangentially fired boilers). The more stringent regulatory requirement of 0.15 lb/million Btu in Title I is driven by health standards without regard for technology availability or cost, whereas Title IV was predicated on the availability and effectiveness of low-NO$_x$ burner technology. Some state and local permitting agencies have imposed even lower NO$_x$ emissions limits, as low as 0.03 lb/million Btu. Large-scale industrial boilers and turbines also are subject to NO$_x$ emissions regulations.

Plant Gorgas, Alabama Power Company Unit 10 has an SCR that has been operating for three ozone seasons.

The results to date of compliance with NO_x emissions regulations are impressive. According to EPA figures, total NO_x emissions from coal-fired electric generating units decreased from 6.7 million tons in 1990 to 4.2 million tons in 2003, a 37% reduction. During the same period, the amount of coal burned to produce electric power increased by about 27%. Reductions also have occurred for both average emissions and short-term peak NO_x emissions that are a concern on hot, high electricity demand days conducive to ozone formation. More than 99% of the affected power plants were in full compliance in 2003.

The CAIR, when implemented, will further reduce annual NO_x emissions from power plants to 1.6 million tons by 2010 and 1.3 million tons by 2015. For the 29 states and the District of Columbia affected by CAIR, power plant NO_x emissions will be reduced by 64% from 2002 levels.

NO_x emissions reductions already achieved have resulted in significant improvement in ambient ozone concentrations. By 2003, the mean measured ozone value was only slightly higher than the NAAQS of 0.08 ppm. Further reductions in NO_x emissions undoubtedly will bring more areas of the country into ozone attainment. Such progress, of course, requires NO_x emissions reductions not only from power plants but also from other major contributors to ground level ozone load, including vehicles (both highway and off-highway) and industrial boilers. Coal-fired electric generators accounted for only about 22% of all U.S. NO_x emissions in 2003, representing a significant reduction from earlier figures and thus demonstrating the success of the electric power industry in reducing NO_x emissions.

NO_x Emissions Regulations

History

The Clean Air Act originally was passed in 1967. It was amended in 1970, 1977, and most recently in 1990. The Clean Air Act Amendments (CAAA) of 1990 authorized the U.S. Environmental Protection Agency (EPA) to establish National Ambient Air Quality Standards (NAAQS) for six criteria atmospheric pollutants: carbon monoxide, lead, ozone, nitrogen dioxide, sulfur dioxide, and particulates. Updating the emissions standards every five years is mandated. For convenience, the nitrogen oxides regulations are written in terms of NO_x, which represents a mixture of nitric oxide (NO) and nitrogen dioxide (NO_2).

Ozone Formation

In the atmosphere, NO_x and volatile organic compounds (VOCs) react in the presence of sunlight to form ground-level ozone, which is the major ingredient of smog. Two major portions of the CAAA relevant to NO_x control for electric power generation are Title I and Title IV. Title I addresses ambient ozone, while Title IV regulates NO_x emissions for specific types of boilers, including stationary coal-fired power plants. Title IV is referred to as the Acid Rain Program. As opposed to Title IV, which specifies NO_x emissions directly, Title I focuses on health-based standards for ozone, which in turn are strongly influenced by NO_x emissions.

The current Title I NAAQS for ozone, established in 1997, is 80 ppb (0.08 ppm), maintained over an 8-hour period. To meet this standard, NO_x emissions from coal-fired power plants must be reduced to an average of 0.15 lb/million Btu of boiler heat input. Clearly, the NO_x emissions targets under Title I are much more stringent than those under Title IV.

The Clean Air Interstate Rule (CAIR), promulgated by EPA on March 10, 2005, addresses the problem of long-range emissions transport from 29 upwind states and the District of Columbia to downwind states that are not in attainment with respect to several criteria pollutants, especially ozone, SO_2, and particulates. CAIR calls for deeper emissions reductions than do existing regulations and incorporates cap-and-trade features to permit flexibility in reaching the designated targets.

NO$_x$ Control Technologies

The major NO$_x$ control technologies for boilers are:

- Low-NO$_x$ burners (LNB)

- Overfire air (OFA)

- Reburning

- Selective non-catalytic reduction (SNCR)

- Selective catalytic reduction (SCR)

The first three technologies involve modifying combustion in the boiler, whereas the latter two are post-combustion processes that are applied to the flue gas. Combustion modification technologies are relatively inexpensive, but alone they cannot achieve the degree of NO$_x$ reduction required by current emissions standards. For that purpose, post-combustion technologies must be applied. This Topical Report focuses primarily on SCR, the predominant post-combustion technology.

The SCR Process

History

Selective catalytic reduction of NO$_x$ using ammonia as the reducing agent was developed in the United States by Engelhard Corporation and patented in 1957. The original catalysts, consisting of platinum or platinum group metals, were unsatisfactory because of the need to operate in a temperature range in which explosive ammonium nitrate forms. Other base metal catalysts were found to have low activity. Research conducted in Japan in response to severe environmental regulations in that country led to the development of vanadium/titanium catalysts, which performed successfully. Ongoing catalyst development has been instrumental in promoting continued growth in SCR applications worldwide.

Process Description

In the SCR process as applied to coal-fired electric power plant operation, NO$_x$ in the flue gas exiting the boiler economizer is converted to nitrogen and water by reaction with ammonia in the presence of the catalyst. The process is termed "selective reduction" because it takes oxygen only from the nitrogen compounds and not from carbon, sulfur, or other oxygenated compounds. Of significant importance are side reactions that result in undesirable by-products, primarily ammonium sulfate, $(NH_4)_2SO_4$, and ammonium bisulfate, NH_4HSO_4. These by-products can cause fouling and corrosion of downstream equipment, most notably the combustion air preheater. Sulfur present in the coal is oxidized to SO_2 in the boiler. In turn, a small fraction of the SO_2 is oxidized to sulfur trioxide (SO_3) over the SCR catalyst, an undesirable side reaction. Major operating variables include inlet NO$_x$ concentration, NH_3/NO_x ratio, fuel sulfur and trace element (As, Ca, V) content, inlet dust loading, reactor temperature, and system pressure drop.

NO$_x$ Emissions Limits for Coal-Fired Boilers
Title IV of the Clean Air Act Amendments of 1990

	Emissions Limit lb/million Btu	Number of Affected Units
Phase I (1996–2000)		
Dry bottom wall-fired	0.50	119
Tangentially fired	0.45	132
Phase II (2000+)		
Dry bottom wall-fired	0.46	306
Tangentially fired	0.40	302
Wet bottom wall-fired (>65 MWe)	0.84	26
Cyclone-fired (>155 MWe)	0.86	56
Vertically fired	0.80	26
Cell burner	0.68	37
Total		1,004

Ammonia Slip

Unreacted ammonia in the flue gas downstream of the SCR reactor is referred to as ammonia slip. For power plants burning eastern U.S. coals, it is essential to hold ammonia slip to a very low level, typically around 2 to 3 ppm, to minimize formation of ammonium sulfate and bisulfate and to maintain acceptable ash quality for disposal or sale. For western subbituminous coals, such as those from the Powder River Basin (PRB), higher levels of slip can be tolerated due to the lower sulfur content of these fuels. An upper limit of 5 ppm may be acceptable. For European and Japanese installations, ammonia slip is typically between 1 and 4 ppm. These plants typically operate on coals having a maximum sulfur content of about 1.7%, and have an average maximum NO_x removal efficiency of 85%. Ammonia slip is a greater problem with high-sulfur coals because of higher SO_3 levels resulting from these coals. Oxidation of SO_2 in the SCR reactor to form SO_3 adds to the intrinsic SO_3 resulting from combustion. Additional reasons to limit ammonia slip are discussed later. The primary determinants of ammonia slip are catalyst volume and activity.

Operating Temperature

Since SCR catalysts are expensive, it is essential to operate at as high a temperature as possible to maximize space velocity and thus minimize catalyst volume. At the same time, it is necessary to minimize the rate of oxidation of SO_2 to SO_3, which is more temperature sensitive than the SCR reaction. The operating range for vanadium/titanium-based catalysts is 500 to 850 °F, and for zeolites 850 to 1050 °F. Zeolite catalysts are much more expensive than vanadium/titanium catalysts. Most installations use an economizer bypass to provide flue gas to the reactors at the desired temperature during periods of low flow rate, such as low-load operation.

NO$_x$ Formation

Nitrogen oxides (NO_x) are formed primarily by the reaction of atmospheric nitrogen and oxygen, through a fairly complex mechanism. Although molecular nitrogen and oxygen in the air are stable at ambient conditions, they react in the high-temperature environment associated with fuel combustion. The resulting NO_x is referred to as thermal NO_x. In addition, nitrogen in the fuel is oxidized under combustion conditions to form fuel NO_x.

The formation of nitric oxide via thermal fixation of atmospheric nitrogen is highly temperature sensitive. As originally defined by Zeldovich, the rate of thermal NO_x formation is exponentially dependent on temperature and is proportional to the square root of oxygen concentration. Reducing both the amount of oxygen available to the fuel and the combustion temperature are effective methods of controlling thermal NO_x. The mechanism by which fuel nitrogen is converted to NO_x is not fully understood but is known to depend on fuel/air ratio. Fuel NO_x can represent a significant fraction of the total NO_x emitted during the combustion of a high nitrogen content coal.

Nitrogen oxides are highly reactive compounds that play significant roles in air pollution. In addition to the formation of ozone, nitrates and sulfates formed by atmospheric oxidation of NO_x and SO_2 are major components of fine particulate matter that poses a human health problem when inhaled, and also an environmental problem for lakes and rivers. The fine particulate matter is deposited into lakes and rivers, contributing to excessive algae growth and oxygen depletion, conditions that can threaten aquatic life. These particulates also are largely responsible for atmospheric haze and visibility impairment.

About 55% of U.S. NO_x emissions are associated with combustion of transportation fuels in motor vehicles and non-road equipment. Another 40% results from fuel combustion in stationary sources, of which about one-half is emitted by electric generating plants. The remaining 5% is emitted as a by-product of industrial processes.

Regulations are in place that will substantially reduce NO_x emissions from the transportation sector in the near future. Emissions from the power generation sector, already heavily regulated, will be further reduced by additional controls, both existing and planned. Like most emissions sources, power plants have been the target of substantial regulatory activity since enactment of the Clean Air Act of 1970 and the Clean Air Act Amendments (CAAA) of 1990. Implementation of the CAAA Acid Rain Program (Title IV) has reduced annual NO_x emissions from coal-fired power plants from 6.7 million tons in 1990 to 4.2 million tons in 2003, while the amount of coal burned increased by about 27%. Now, as a result of the SIP Call of 1997, promulgated under Title I of the CAAA, and the Clean Air Interstate Rule (CAIR), also promulgated under Title I, this figure will decrease to 1.6 million tons by 2010 and 1.3 million tons by 2015.

U. S. Installations of SCR on Coal-Fired Utility Boilers
(2004 Data)

State	Units
Indiana	23
Ohio	20
Kentucky	14
North Carolina	14
Pennsylvania	14
Tennessee	14
West Virginia	14
Alabama	12
South Carolina	9
Michigan	8
Georgia	7
Illinois	7
Virginia	6
Texas	5
Massachusetts	4
New York	4
Wisconsin	4
Maryland	3
Missouri	3
New Jersey	3
New Hampshire	2
Florida	1
Total	**191**

Source: Argus SCR Report, May 2004

Reactor Placement

SCR systems can be installed either: (1) after the boiler economizer, *i.e.*, upstream of the air preheater (APH), ESP, and flue gas desulfurization (FGD) unit; or (2) downstream of the APH, ESP, and FGD. The former is referred to as hot side, high dust installation and the latter as cold side, low dust. In commercial practice, the hot side location is more commonly used because it eliminates the need to reheat the flue gas to reaction temperature, thereby minimizing loss of plant thermal efficiency. The flue gas velocity in the ductwork is generally limited to about 60 ft/sec to minimize erosion while maintaining the fly ash in suspension. The design mean gas velocity in the SCR reactor is 16–18 ft/sec.

A typical arrangement in coal-fired plants consists of a horizontal duct that conveys the flue gas to the vertical catalyst housing section, where the gas flows downward over the catalyst bed. Ammonia reagent is injected into the horizontal duct. Turning vanes provide the transition from horizontal to vertical flow. In some cases, the boiler configuration requires a vertical upward duct from the economizer exit (with ammonia injected in the duct) followed by two 90-degree turns to the vertical downflow SCR reactor. The ducting is designed to maintain uniform flow of flue gas and reagent while minimizing pressure drop.

The relatively high capital cost of an elevated, vertical SCR can be reduced by first removing a significant fraction of the NO_x with SNCR, thereby reducing the load on the SCR reactor. In such hybrid installations, excess reagent is used in the SNCR step, with the remaining NO_x removed in the SCR reactor. As a result, the SCR unit can be significantly smaller and can use a short, horizontal vessel with less ductwork, structural steel, and foundations.

Catalysts

SCR catalysts are made of a solid ceramic material that contains active catalytic components. The ceramic may be in the form of a honeycomb monolith or it may be coated onto ceramic, metal, or fiber substrate. Catalyst elements include titanium, vanadium, molybdenum, and tungsten, as well as zeolites. Catalyst formulations are tailored to specific performance needs. Vanadium is one of the catalyst elements responsible for the NO_x reduction reaction, but it also contributes to the oxidation of SO_2 to SO_3 in the reactor. This effect is discussed in greater detail later.

Three mechanical forms are used: plate, fiber, and honeycomb. Fiber catalysts, which are lighter than the others, have characteristics similar to both honeycomb and plate designs, but are fabricated on a fibrous base. As indicated previously, the most common arrangement involves downflow of dust-laden flue gas in vertical reactors. The catalyst typically is arranged in a series of two to four beds, or layers. The lowest layer often is not installed initially. This arrangement provides a safety factor to allow for catalyst aging and/or additional NO_x reduction requirements. As catalyst activity declines, additional catalyst layers are installed in the available spaces in the reactor. With continued aging, the catalyst is replaced on a rotating basis, one layer at a time, starting with the top. This strategy permits maximum catalyst utilization. Ammonia slip increases somewhat over time until it reaches the design limit, at which point fresh catalyst is added. Another option, rejuvenation and/or regeneration, is also in widespread use, as discussed later.

Mixing of Ammonia and Flue Gas

To ensure optimum performance of the SCR process, it is essential to achieve intimate and uniform mixing of ammonia and flue gas in the limited space at the inlet to the catalyst bed. At the same time, pressure drop must be minimized to avoid excessive parasitic power losses. Uneven ammonia distribution can cause poor SCR performance, because some portions of the flue gas stream are overtreated, resulting in higher than design ammonia slip, while other portions are undertreated, resulting in higher than design exit NO_x concentration. Maintaining the appropriate balance is particularly critical since overall NO_x removal targets are generally very high (90%+) and the ammonia slip limit is very low (2 to 5 ppm). Maximum permissible deviations specified in SCR plant designs are in the range of ±10 to ±20% for velocity, ±20 to ±50 °F for temperature and ±5% to as high as ±10% for NH_3/NO_x molar ratio. The more challenging the NO_x reduction and ammonia slip requirement, the lower the maximum deviation permitted. Thus, there is little margin for error for high removal systems, and this is compounded by the fact that NO_x distribution in the flue gas varies with boiler operating conditions.

To help ensure optimum reagent distribution, a number of proprietary ammonia injection grid designs and mixing systems have evolved that make extensive use of three-dimensional physical flow modeling and computational fluid dynamics (CFD) modeling. The latter takes into account fluid turbulence, heat transfer, mass transfer, and chemical kinetics. The resulting designs have proved very effective when scaled up for commercial application.

SCR unit at Trimble County 1.

Chemistry of the SCR Process

4NO nitric oxide	+	4NH$_3$ ammonia	+	O$_2$ oxygen	→	4N$_2$ nitrogen	+	6H$_2$O water
2NO$_2$ nitrogen dioxide	+	4NH$_3$ ammonia	+	O$_2$ oxygen	→	3N$_2$ nitrogen	+	6H$_2$O water
SO$_2$ sulfur dioxide		+	½ O$_2$ oxygen		→	SO$_3$ sulfur trioxide		
2NH$_3$ ammonia	+	SO$_3$ sulfur trioxide	+	H$_2$O water	→	(NH$_4$)$_2$SO$_4$ ammonium sulfate		
NH$_3$ ammonia	+	SO$_3$ sulfur trioxide	+	H$_2$O water	→	NH$_4$HSO$_4$ ammonium bisulfate		

Commercial Operating Experience with SCR

SCR Catalyst Developments

As mentioned previously, catalyst deactivation is a normal function of aging. The mechanisms for deactivation are complex, and include fouling (surface deposition), pore condensation and/or blocking, and poisoning. For a given situation, one or more of these mechanisms may be involved. All of them are functions of the nature and quantity of minerals in the coal, including compounds of sodium, potassium, calcium, arsenic, and others. The degree of deactivation is more pronounced with lower rank fuels such as lignite or subbituminous coals. So-called popcorn ash is one form of deposit that sometimes fouls catalyst surfaces. This has been encountered in a number of SCR installations. The mechanism for popcorn ash formation in the boiler is not clear. Popcorn ash can be removed from the catalyst bed by vacuuming during planned outages.

Catalyst physical characteristics include pitch and wall thickness, which determine the open area for the flue gas flow. Smaller pitch catalysts generally are used for relatively clean flue gas (natural gas firing), and larger pitch catalysts for coals, especially those with relatively high calcium oxide content (*e.g.*, PRB coals). Larger pitch is used for situations involving large ash size or loading. In the past, the standard pitch in coal applications was ~7.5 mm for honeycomb and ~6.0 mm for plate and fiber catalysts. With increased experience, pitch has been reduced, resulting in increased specific surface area and decreased overall catalyst volume requirements.

The choice of pitch is influenced by the location of the particulate collection device used in the power plant, and the ash loading of the flue gas. In hot side SCR installations, the catalyst is exposed to high particulate loading, requiring somewhat larger pitch and larger overall volume. On the other hand, large dust particles tend to have a cleaning effect on the catalyst and APH surfaces, potentially reducing plugging from deposits such as ammonium bisulfate. Cold side SCR installations have lower particulate size and loading, which can permit smaller pitch and smaller catalyst volume, thus reducing the size of the reactor, associated ducts, and structural supports. However, smaller particles may require more frequent sootblowing due to plugging of the catalyst pores.

Much of the work on catalyst development done in the past, as well as currently under way, involves elucidation of deactivation mechanisms and tailoring catalyst compositions to meet specific needs. SCR catalyst development is highly competitive, and thus is mostly proprietary. Since catalyst cost, both for initial inventory and for makeup, is the single most important factor in SCR economics, there is great incentive to develop catalysts with optimum performance characteristics.

It has become standard practice in the industry for suppliers to provide catalyst life guarantees, expressed in terms of the number of hours of operation that can be achieved before NO_x removal or ammonia slip reach unacceptable levels. Typical catalyst life guarantees are in the neighborhood of 16,000 hours, representing over two years of operation at 80% on-stream time.
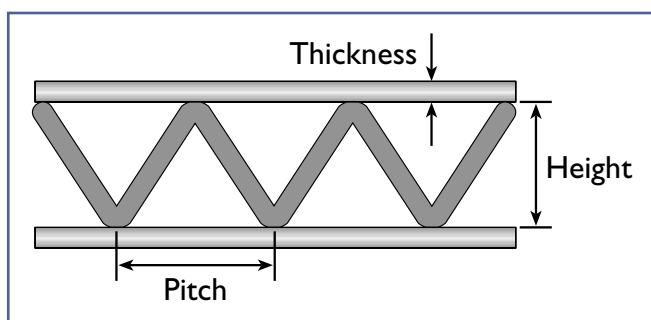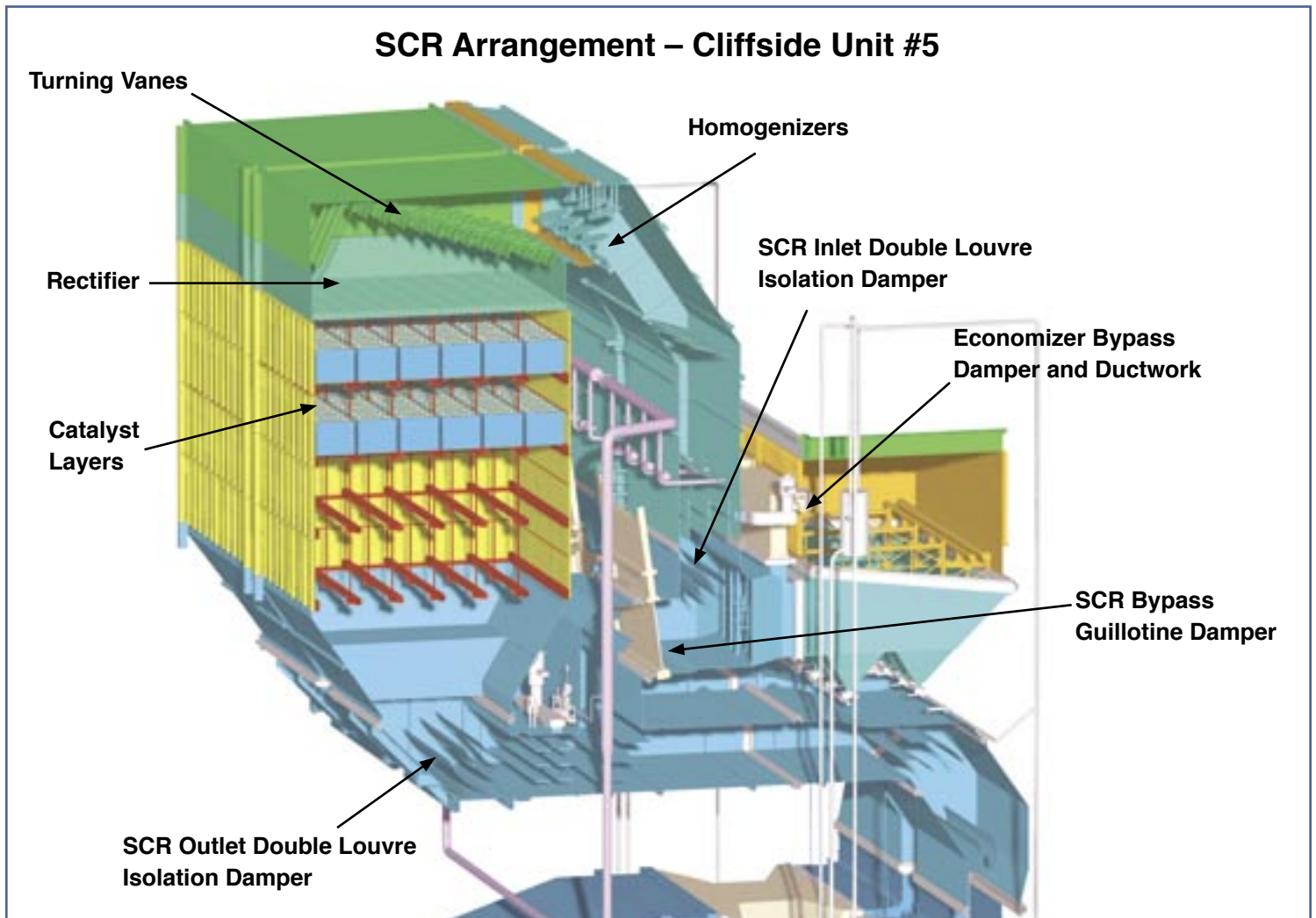

Catalyst Dimensions.

Some work has been done on catalysts that operate at much lower temperatures (<400 °F) than conventional SCR. These catalysts have higher surface area than conventional catalysts and also tend to have higher vanadium content, thus resulting in higher activity. Advantages include lower capital costs because the SCR unit can be placed downstream of the cold end ESP. This minimizes the ductwork required, reduces the need for sootblowing, and offers longer catalyst life. Low-temperature catalysts can be effective in hybrid installations where SNCR is employed upstream of the SCR unit. The main disadvantage of today's low-temperature catalysts is their susceptibility to ammonium bisulfate precipitation and particulate fouling.

Catalyst Rejuvenation and Regeneration

SCR catalysts must be cleaned or rejuvenated periodically to maintain optimum NO_x reduction performance. In most applications, cleaning involves sootblowing, using compressed air, steam, or sonic energy as the cleaning agent. Benefits claimed for sonic sootblowing include lower capital and operating costs than for steam sootblowing.

In addition to mechanical deposition, catalyst deactivation is caused by chemical depletion as a result of normal SCR operation. SCR catalyst deactivation is linked to the composition of the coal feedstock. Deactivation has been attributed in part to alkali and alkaline earth elements found in

SCR Arrangement – Cliffside Unit #5

Turning Vanes

Homogenizers

Rectifier

SCR Inlet Double Louvre Isolation Damper

Economizer Bypass Damper and Ductwork

Catalyst Layers

SCR Bypass Guillotine Damper

SCR Outlet Double Louvre Isolation Damper

subbituminous coals and in biomass co-fired with coal. Arsenic, lead, and zinc present in all ranks of coal are also major contributors to deactivation. In the case of arsenic, arsenic trioxide formed in the boiler reacts with vanadium in the catalyst to poison the catalyst. However, the presence of calcium oxide in the coal helps mitigate the deleterious effect of arsenic by forming calcium arsenide, a stable solid that becomes bottom ash in the boiler, thereby minimizing exposure of the catalyst to arsenic. Complicating matters further, calcium oxide can have adverse affects on catalyst— forming fine calcium sulfate particles that may plug catalyst micropores.

Eastern U.S. coals tend to have higher arsenic content and lower calcium oxide content than do PRB coals. Catalyst deactivation for eastern coals is caused primarily by arsenic, while for PRB coals the primary mechanism is calcium oxide poisoning. Because of the interaction of calcium oxide and arsenic, blends of eastern and PRB coals can reduce the deactivation rate. In many cases, fuels with low calcium content are supplemented with additional calcium by injecting limestone. This fuel treatment is recommended, provided the impacts of limestone addition on slag formation and ESP operation have

been properly evaluated. Alternatively, some plant operators have chosen to forego limestone injection in favor of accepting lower catalyst life guarantees. Studies are continuing to elucidate catalyst deactivation mechanisms and to devise methods for mitigating these effects.

Scheduled catalyst layer rotation was mentioned previously. Regenerating old catalyst in addition to layer rotation has been proven to reduce SCR operating costs by increasing the effective life of the individual catalyst layers. Proprietary regeneration techniques are applied either to the catalyst in place or to sections of catalyst removed and treated externally. Catalyst regeneration is often less expensive than replacement, but after two or three cycles of regeneration the effects of erosion or physical damage usually require catalyst replacement. Different methods of catalyst re-processing, including mechanical cleaning, heating with special gases, and washing with regenerating solutions, have been tested (including full-scale operations) with varying degrees of success. An additional benefit is that disposal of spent catalyst and release of toxic metals to the environment are minimized. A disadvantage of some regeneration approaches is that more frequent shutdowns may be

necessary for catalyst maintenance, and this may have a significant impact on the relative economics of regeneration versus replacing old catalyst with new catalyst.

Many plant operators make use of periodic catalyst testing by firms specializing in this service. A number of catalyst management strategies have been developed to optimize catalyst activity and life, using a variety of rejuvenation and/or regeneration methods, and software programs are available commercially that address these issues.

Reagent Preparation and Handling

Several methods are available to supply the ammonia reagent to an SCR facility. A simple technique is to use anhydrous ammonia, delivered from a supplier and stored on-site in pressure vessels. Although direct use of anhydrous ammonia has been applied successfully in some SCR installations and in other industries such as fertilizer manufacture, ammonia is designated as a hazardous chemical requiring special safety precautions for transportation and storage. Many utilities are reluctant to take the risk. Aqueous ammonia (diluted with 70–80% water) is a safer alternative, but its drawbacks include the energy required to vaporize the ammonia for injection into the flue gas, and the need for substantially larger storage and transportation capacities.

Another approach involves on-site conversion of granular or aqueous urea to ammonia by hydrolysis. Urea is a material that is widely used as a fertilizer and has fewer personnel safety concerns than either form of ammonia. At least two commercial processes are currently in use. In both processes, urea is dissolved in water and heated to initiate the hydrolysis reaction. The ammonia is stripped from the solution with steam and fed to the SCR system at a controlled rate. Claims made for the use of urea include increased safety and improved control of ammonia flow to the SCR reactor.

Plant Bowen, Georgia Power Company - Units 1 and 2 SCRs have been operating for 4 ozone seasons. Units 3 and 4 have been operating for 2 ozone seasons.

The Clean Coal Technology Demonstration Program

The Clean Coal Technology Demonstration Program (CCTDP) is a unique partnership between the federal government and industry that has as its primary goal the successful introduction of new clean coal 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 environmental concerns associated with the use of coal for electric power production. As the program has evolved, it has expanded to address the need for new, high-efficiency power generating technologies that allow coal to continue to be a fuel option well into the 21st century.

Begun in 1985 and expanded in 1987 consistent with the recommendations of the U.S. and Canadian Special Envoys on Acid Rain, the program has been implemented through a series of five nationwide solicitations, or rounds. Each round was associated with specific government funding and program objectives. After five rounds, the CCTDP consists of a total of 35 projects located in 17 states with a total investment value of over $3.7 billion. DOE's share of the total cost is about $1.4 billion, or approximately 38%. The projects' industrial participants (*i.e.*, non-DOE) have provided the remaining 62%, about $2.3 billion.

Processes demonstrated under the CCTDP have established a technology base that is enabling the nation to meet its energy goals under increasingly stringent environmental requirements. Also ready is a new generation of technologies that can produce electricity and other commodities, such as steam and synthesis gas, at high efficiencies consistent with concerns about global climate change.

Most of the CCTDP demonstrations have been 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 applications. Each application addresses one of the following four market sectors:

· Environmental control devices

· Advanced electric power generation

· Coal processing for clean fuels

· Industrial applications

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

Two follow-on programs have been developed that build on the successes of the CCTDP. The Power Plant Improvement Initiative (PPII) is a cost-shared program, patterned after the CCTDP and directed toward improved reliability and environmental performance of the nation's coal-burning power plants. Authorized by the U.S. Congress in 2001, the PPII involves six projects that focus on technologies enabling coal-fired power plants to meet increasingly stringent environmental regulations at the lowest possible cost.

The second program is the Clean Coal Power Initiative (CCPI), also patterned on the CCTDP, and authorized in 2002. The CCPI involves a 10-year program having a goal of accelerating commercial deployment of advanced technologies to ensure that the nation has clean, reliable, and affordable electricity. Total federal funding will be up to $2 billion, with a matching cost-share by industrial participants of at least 50%. As of April 2005, 10 CCPI projects have been awarded or are in negotiation.

Mercury Removal in SCR Systems

Flue gas from coal-burning power plants is considered a significant source of atmospheric mercury, which is widely recognized as a threat to public health. Typical mercury concentration in coal ranges from 0.08 to 0.20 micrograms per gram (μg/g). Volatilized during combustion, the mercury exits the boiler in the flue gas as either particulate mercury, oxidized mercury, or elemental mercury. The most common form of oxidized mercury is mercuric chloride ($HgCl_2$), which is highly water soluble and is readily removed from the flue gas in wet FGD systems. Experience indicates that additional oxidation of mercury compounds occurs in the SCR unit. The presence of chlorides (e.g., HCl) in the flue gas enhances this effect, while high concentrations of calcium oxide, which may remove some of the HCl, can have the reverse effect. Enhancement of mercury removal in downstream FGD systems thus appears to be an unanticipated benefit of SCR technology, with significant implications for multi-pollutant control strategies, and further investigation is under way. One study reported mercury removal efficiency of 89-95% for SCR in a power plant equipped with a lime spray dryer and 83% when using wet FGD with 15% flue gas bypass. Several studies have shown that SCR significantly improves mercury capture by wet FGD systems when bituminous coals are burned. For low-rank coals, which tend to have lower chlorine contents, the effect of SCR on improving mercury oxidation does not appear to be as great.

Oxidation of SO_2 to SO_3 over SCR Catalysts

Essentially all of the sulfur present in coal feedstocks is oxidized to SO_2 in the boiler, with a small percentage (about 1%) oxidized to SO_3. Likewise in the SCR reactor, a portion of the SO_2 is oxidized to SO_3 over the catalyst. SCR catalysts are designed to minimize this effect, but the flue gas exiting the SCR unit can contain sufficient SO_3 to cause problems downstream because it reacts with water vapor in the flue gas to form sulfuric acid (H_2SO_4). This acid, which is not removed in the FGD unit, is present as vapor adsorbed or condensed onto sub-micron particles in the flue gas. These particles can evade separation or capture and thus are present in the power plant stack. The resulting mist can settle as a plume over the surrounding area. The light-scattering properties of these fine particles can cause high opacity and impaired visibility. Another detrimental effect is corrosion of metallic surfaces, both inside and outside the power plant.

As mentioned previously, another effect of the presence of SO_3 in the SCR reactor is the reaction with ammonia slip to form ammonium sulfate and bisulfate. These compounds can cause plugging of the SCR catalyst as well as fouling and corrosion of the APH surfaces. Physical properties of the coal impact the selection of both catalyst pitch and chemical properties such as the allowable SO_2 oxidation rate. For high-sulfur coals (>1% S), SO_2 oxidation should be maintained at less than 1%. SCR units operating on low-sulfur coals (<1% S) can be designed for SO_2 oxidation rates somewhat greater than 1% without negative impact on plant operations. Because of the lower sulfur content of PRB coals, the allowable ammonia slip can be somewhat higher than for eastern coals.

Acid mist and ammonium sulfate/bisulfate formation have caused major operating problems at some SCR installations. Remedial measures include injecting alkaline compounds such as lime or magnesium oxide into the flue gas to neutralize the SO_3. However, despite numerous efforts along these lines, a significant problem remains at certain sites, and further work is in progress to elucidate reasons and develop solutions. Recently, successful mitigation of acid plume by injecting trona (a sodium-based mineral compound) has been reported.

Ammonia Deposition on Fly Ash

The presence of ammonia slip in the SCR reactor exit gas results in ammonia contamination of the fly ash, as a result of adsorption or other mechanisms. At a certain NH_3 level, the ash properties are adversely impacted. This effect can be detrimental to utilization and/or disposal of the ash. If the fly ash is used in the manufacture of construction building materials, there is an upper limit to the permissible ammonia content based on exposure limits for employees and product performance requirements. If the fly ash is disposed of as landfill, odor becomes a factor. Further, rainwater can leach ammonia as well as other components of the ash from landfills, resulting in violation of groundwater pollution regulations. These effects must be taken into account in the development of ammonia utilization strategies for SCR operations.

Considerable work has been done on removing ammonia from fly ash by a variety of methods including thermal treatment. One such process subjects the fly ash to fluidized-bed combustion to remove both the unburned carbon and the residual ammonia. The ammonia is oxidized to nitrogen and water. Another process treats the fly ash with an alkali such as calcium oxide to release the ammonia, which is recycled to the SCR unit. In addition, methods have been developed for removal of ammonia and other pollutants from the wastewater resulting from exposure of fly ash in landfills to rainwater.

Instrumentation and Control

NO_x emissions from power plants depend on a number of variables, including furnace and combustion system design, combustion temperature, residence time, coal/air distribution, nitrogen content of the fuel, and coal particle size. The goal of process optimization for SCR units operating on coal-fired boilers is to maximize NO_x reduction and catalyst life, while minimizing ammonia consumption and slip as well as overall power plant heat rate. Control strategies to accomplish
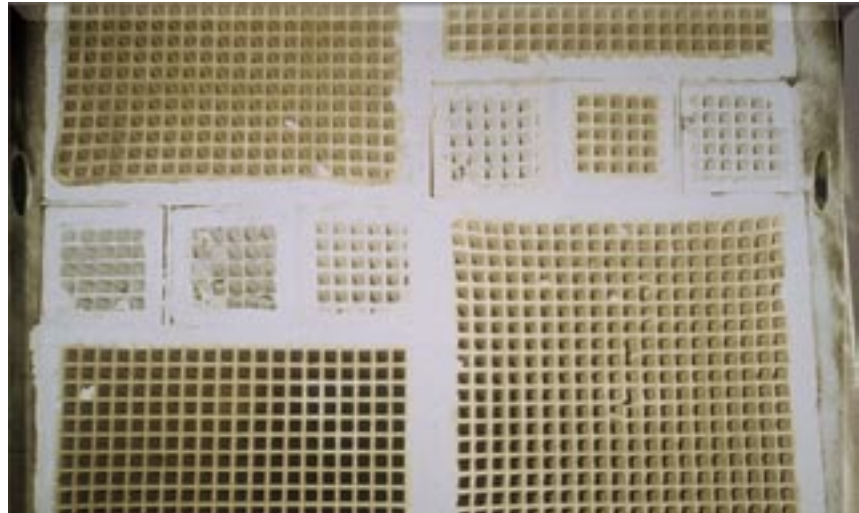
Clean catalyst in a SCR reactor.

Damage caused by erosion in the catalyst bed.

Plugging that may occur during normal operations of an SCR facility.

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this have become increasingly more sophisticated, involving advanced instrumentation, neural networks, optimization technologies, and modeling techniques. A number of control systems have been developed and are being applied commercially.

SCR control typically requires two basic loops, an ammonia injection control loop and an economizer bypass control loop. Essential to optimum operation and control of SCR systems is precise control of the NH_3/NO_x feed ratio at the reactor inlet, which requires the use of appropriate instrumentation. The primary variables are flue gas temperature and flow rate, NO_x concentrations in the SCR reactor inlet and exit gas streams, and ammonia slip in the reactor exit gas. The latter is critical from the standpoint of both environmental regulations and operating cost, and the NO_x concentration in the stack gas is, of course, the key variable in determining whether the plant meets mandated emissions limits.

SCR unit at Ghent 1.

NO_x concentrations are measured by dedicated NO_x analyzers operated as part of the SCR control system. For the feed-forward loop, the SCR has dedicated inlet and outlet NO_x analyzers that are independent of the stack continuous emissions monitor (CEMS). The stack CEMS can be used as an input for a feedback loop on single-stack units, but this approach provides response that is too slow for good control during load changes. So, dedicated NO_x analyzers are used for the SCR controls on most systems. The signal from the boiler combustion system O_2 analyzer is utilized for O_2 measurement at the economizer outlet (required for establishing total gas flow). These instruments must be calibrated regularly to insure accuracy. Because of the acid plume phenomenon mentioned previously, measurement of SO_3 in the flue gas is also critical. However, because continuous measurement systems for SO_3 are not available, measurement of SO_3 is very difficult and is normally only performed periodically.

Unfortunately, ammonia analyzers in coal-fired SCR applications have not proved to be reliable and can be used for reference only. Consequently, ammonia slip is most often determined by one of several means: periodic manual samples, calculation based on measured ammonia feed rate and inlet and outlet NO_x concentrations, and measuring the ammonia content of the fly ash.

The economizer bypass control is set to maintain a minimum temperature in the catalyst bed, particularly at low loads. For applications where the economizer bypass may affect the primary superheat and reheat sections, a more complicated temperature control system may be required, including a second control damper at the economizer outlet.

Market Analysis

The SCR process is applicable to all types of coal-fired boilers including stoker, cyclone, wall-fired, and tangentially fired units. EPA reports that, as of the end of 2003, there were 120 coal-fired units with a combined capacity of about 64,000 MWe that were retrofitted with SCR. According to the Argus SCR Report, total installed and projected U.S. SCR capacity on coal-fired units through 2007 will amount to about 200 units, representing about 100,000 MWe of generating capacity. These installations are primarily in the midwestern and eastern states designated by EPA in the 1997 SIP Call.

These capacity figures are based on EPA assumptions of SCR achieving 85–90% NO_x emissions reductions. Less effective performance would result in the need for additional SCR installations to reach the total emissions reduction targets. Furthermore, enactment of the Clean Air Interstate Rule could result in a doubling of total SCR capacity, to as much as 200,000 MWe by the year 2015. States outside the SIP Call region are beginning to impose NO_x emissions regulations comparable to those in the SIP Call region. If this trend continues, even more SCR capacity will be required.

A significant factor in application of SCR for NO_x control in the United States is the provision in Title I of the CAAA for emissions allowance trading. Emissions trading provides a mechanism for facility owners to comply with environmental regulations at the lowest overall cost. Trading is enhanced in highly liquid markets that are characterized by low transaction costs, good access to market information by all participants, and a large number of participants. Although the U.S. NO_x trading market does not meet all of these criteria, the mechanism has become very effective, with many facility owners participating to their benefit because it is usually most economical to install SCR to overcontrol NO_x on a large unit while avoiding expensive controls on smaller units. The price for 2005 vintage NO_x allowances is about $3,500/ton.

Future Projections

The CCTDP project conducted in 1995 and the subsequent widespread commercial application of SCR on U.S. coal-fired boilers have shown that this technology can achieve high levels of NO_x reduction at an acceptable cost. It is expected that design improvements will be made as further operating experience is gained. The degree to which SCR will be incorporated in new or future retrofit applications will depend on the severity of NO_x control standards yet to be promulgated. In fact, NO_x emissions criteria established to date are largely the result of the demonstrated technical and commercial success of SCR. Future application of NO_x reduction technologies such as SCR will involve consideration of multi-pollutant control strategies, including not only NO_x but also SO_2, particulates, and other components of flue gas such as mercury and volatile organic compounds.

Capital and Operating Costs of SCR

Incorporating SCR in a power plant is relatively expensive, in terms of both capital and operating costs. Most of the SCR installations in the United States to date have been retrofitted to existing plants. A recent survey in *Power* magazine provides available data on a total of 45 SCR installations representing a wide range of electric generating capacity. The results are summarized as follows:

Plant Generating Capacity, MWe	Avg. SCR Capital Cost, $/kW (2004)
100–399	123
400–599	103
600–900	81
>900	117

These cost figures were obtained from a variety of sources and were not necessarily developed on a comparable basis. They do, however, provide a range of figures that to some extent represent the current situation with respect to retrofit applications. Because of the limited amount of information, it is not possible to derive trends for cost vs. plant capacity. Installing SCR in new power plants is likely to be considerably less expensive because the major added costs associated with retrofits are eliminated.

The costs shown above are somewhat higher than those projected when SCR installation was just beginning in the United States. Several site-specific factors influence these figures, including the degree of difficulty involved in retrofitting, the availability of specialized construction labor, and the balance-of-plant modifications required for items such as upgraded or replacement fans, reinforcement of ductwork structural steel, extended ductwork, and the complexity of subsystems for loading and storage of reagent. There is some indication that capital costs will edge downward as more experience is gained. According to a limited amount of published data, the operating cost for SCR units is about $0.50–1.00/MWh (0.05–0.1 cents/kWh). This figure consists of operation and maintenance, including expenses for catalyst replacement, reagent, and labor.

Comparison to Other Technologies

The major commercially available technologies capable of achieving high levels of NO_x removal are the two post-combustion processes, SCR and SNCR. Since SNCR does not require a catalyst, it is considerably less expensive than SCR. However, the maximum demonstrated NO_x removal for SNCR is about 50–60%, whereas SCR can achieve 90% or greater NO_x removal. Recent work has shown the potential for meeting NO_x emissions targets by a layered approach involving a combination of the three major combustion modification technologies (LNB, OFA, and reburning) coupled with SNCR. This approach can be competitive with SCR in certain applications. Reburning, a well-established technology applied in a number of commercial installations both in the United States and in other countries, involves the staged introduction of fuel and air to reduce NO_x formation. Fuel is injected above the primary combustion zone to form a fuel-rich reburn zone, and OFA is injected to complete the combustion process and restore the normal level of excess air.

Another alternative is to achieve the required NO_x reduction without SCR by optimizing combustion modification technologies coupled with fuel/air balancing and advanced network controls. This approach, being investigated at Sunflower Electric's Holcomb Station under the DOE PPII, is being considered in applications where NO_x emissions are only somewhat higher than the target level of 0.15 lb/million Btu. All of these alternative options need to be evaluated on a case-by-case basis, taking into account NO_x removal requirements and relative costs.

Other approaches to NO_x reduction also are being developed. One of these is selective autocatalytic reduction, in which relatively small amounts of oxygen and a

hydrocarbon such as methane are injected into the flue gas along with ammonia. At elevated temperatures, the hydrocarbon autoignites, forming a plasma and creating radicals. These radicals cause the autocatalytic reduction of NO_x to nitrogen and water. Ultra-low NO_x burners also are being investigated as a means of reducing NO_x emissions to acceptable levels at the combustion stage, eliminating or limiting the need for post-combustion processes such as SCR. Another innovative approach to minimizing SCR load is to incorporate partial gasification of the fuel before injection into the reburning zone of the boiler.

Although the primary focus of this report is the application of SCR technology to coal-fired power plants, it should be pointed out that SCR also is used extensively to control NO_x emissions from gas turbines, and work is under way on treating diesel exhaust as well. In addition, NO_x emissions regulations recently have been promulgated for coal-fired industrial, commercial, and institutional boilers. SCR may well be applied in meeting these requirements. Furthermore, DOE-sponsored research is in progress on advanced alternative technologies that can meet even lower NO_x emissions limits than the 0.15 lb/million Btu target now in place.

Conclusions

Post-combustion NO_x control has become a mandatory component of coal-based electric power generation, and SCR has become the industry standard for achieving the reductions in NO_x emissions required by the CAAA. The SCR process is operating successfully in a wide variety of boiler installations. Additional work remains to be done in developing low temperature catalysts, *in-situ* catalyst regeneration processes, and hybrid SNCR/SCR systems, to name a few. An important source of ongoing information on SCR process developments is the annual Conference on Selective Catalytic Reduction and Non-Catalytic Reduction for NO_x Control, sponsored by NETL.

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SCR units at Plant Bowen.

List of Acronyms and Abbreviations