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Project Summary

Selective Catalytic Reduction and NO_x Control in Japan

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A four-member study team traveled in Japan during March 1980 to assess NO_x flue gas treatment technology and related areas. The overall goal of the study was to obtain new information on current issues concerning application of this technology and update information previously published. A total of 28 equipment vendors, process operators, governmental agencies, and industry groups were contacted. There has been substantial progress recently with regard to commercial applications of selective catalytic reduction (SCR) technology to gas- and oil-fired boilers. In several applications, SCR systems are operated continuously and are successfully removing 80 percent of NO_x from the flue gas stream. Current development and demonstration efforts are aimed at applying SCR technology to coal-fired boilers since that fraction of Japan's total electric power generation is expected to increase to 12.5 percent in 1995 and since most of the new, coal-fired boilers will use flue gas treatment technology for NO_x control. Four SCR systems on coal-fired boilers are scheduled to start up in 1980 and 81. Thus, the Japanese activity in the NO_x control field should provide valuable information to interested parties in the U.S. in the next 4 years.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The Environmental Protection Agency is investigating methods of controlling nitrogen oxides (NO_x) emissions from fossil-fuel-fired utility and industrial boilers since these sources comprise a major source of NO_x emissions. Coal-fired boilers are of particular interest since they emit the highest concentrations of NO_x. Currently, NO_x emissions are reduced by the use of special "Low NO_x" burners and other combustion controls. Another technique, developed primarily in Japan, treats the flue gas to achieve 80-90 percent NO_x removal. This technique is usually termed selective catalytic reduction (SCR) or catalytic de-NO_x since it involves injecting ammonia (NH_3) into the flue gas prior to a catalytic reactor which reduces NO_x to N_2 according to the following reactions:

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O$$
$$2NO_2 + 4NH_3 + O_2 \rightarrow 3N_2 + 6H_2O$$

SCR has been widely applied commercially in Japan to several oil- and gas-fired boilers. Coal-fired applications will soon be on-line. However, because this technology has not been applied in the U.S., there are several unanswered questions regarding the application of SCR technology to coal-fired boilers. A study team consisting of J. David Mobley of EPA/IERL-RTP, Jumpei Ando of Chuo University, Gary D. Jones of Radian Corporation, and J. Douglas Maxwell of TVA, travelled in Japan during March 1980 to develop answers to these questions. Information was gathered from a wide variety of sources. Equipment vendors included those of SCR processes, SCR catalysts, other NO_x control processes, air preheaters, and instruments. The variety of installations *inspected included utility and industrial* boilers firing gas, oil, or coal. Governmental groups on both the national and local level were contacted as well as industry groups.

The results of the study can be broken down into four primary subject areas: SCR process design, application of SCR, SCR installations, and additional aspects of SCR.

SCR Process Design

Catalyst

The catalyst is the "heart" of an SCR system and, consequently, is the subject of many design considerations. Specially shaped catalysts that avoid being plugged by fly ash have been developed for use with oil- and coal-fired boilers. The shapes are typically classified as honeycomb, plate, or pipe. For gas-fired applications, the conventional pellet shape is preferred.

The same basic catalyst material is used by all SCR vendors and consists of TiO_2 and V_2O_5 as major components. Each vendor also includes some additional proprietary compounds to affect special properties, such as the SO_2 oxidation potential. The catalyst elements can be supplied in three forms: composed solely of catalyst material (homogeneous), or composed of either a ceramic or metallic substrate that is coated with catalytic material. There is no agreement on which type is best; each offers unique advantages. Of the systems currently under construction on coal-fired boilers, two are using a homogeneous catalyst and two are using a coated catalyst.

One aspect of using V_2O_5 as an SCR catalyst is that it will also catalyze the oxidation of SO_2 to SO_3; with a V_2O_5-rich catalyst, the conversion can be as high as 5 percent. The performance of one formulation can be shown as a function of temperature; however, note that other factors also affect the rate of SO_2 oxidation. High SO_3 concentrations can cause problems due to ammonium bisulfate (NH_4HSO_4) plugging of air preheaters downstream of the reactor. For applications where SO_2 is present, the catalyst formulation is modified and can include compounds that suppress the SO_2 oxidation reaction. This suppression is accomplished at the expense of NO_x removal; for example, reducing SO_2 oxidation from 2.5 to 0.5 percent reduces the NO_x removal efficiency by 20 percent.

The actual catalyst guarantee period depends on the characteristics of each application; however, the guarantees are usually 1 year for coal, 1 to 2 years for oil, and 2 to 3 years for gas although the experience on gas- and oil-fired boilers has been that actual catalyst life exceeds the guarantee. Several things can affect catalyst life. Although some catalyst poisons (such as alkaline oxides) are present in flue gas, their concentrations are low enough to prevent rapid changes in activity.

Temperature is important and there are both upper and lower limits. If the temperature is allowed to get too high, >450°C, the catalyst particles will undergo sintering and the catalyst activity will be reduced. Low temperatures, <300°C, can allow NH_4HSO_4 to form on the catalyst if sufficient SO_3 is present. Temperatures <100°C can allow water deposition which can permanently reduce the catalyst activity. The desired operating temperature range is 350 to 400°C: for some applications a temperature control system is recommended by the vendor to maintain a minimum temperature.

NO_x Removal

NO_x removals of 90 percent and higher are potentially obtainable with SCR systems, although most systems installed in Japan usually operate at 80 percent removal. The vendors indicate that 90 percent removal can be achieved, but will cost more since more catalyst and a higher NH_3:NO_x mole ratio will be necessary.

NH_3 Emissions

Increasing NO_x removal also has the effect of increasing NH_3 emissions. Keeping the NH_3 emissions low at 90 percent NO_x removal levels will require either additional catalyst or some other process for NH_3 removal. To go from 80 to 90 percent NO_x removal without increasing NH_3 emissions is estimated to require about 30 percent additional catalyst. The process vendors indicate that at either 80 or 90 percent NO_x removal, slip or unreacted NH_3 can be limited to ≤10 ppm.

Costs

The costs quoted for SCR systems by all of the vendors are similar. Investment costs for a typical 80 percent control system applied to an oil-fired boiler are about 3000 to 4000 yen/kW ($12 to $16/kW). A similar system applied to a *coal-fired boiler would be about* ¥6000 to 7000 yen/kW ($24 to $28/kW). These figures are for new units. The cost of SCR systems installed on existing boilers can be substantially higher. No general figures are available for retrofit systems due to the high variability between individual installations.

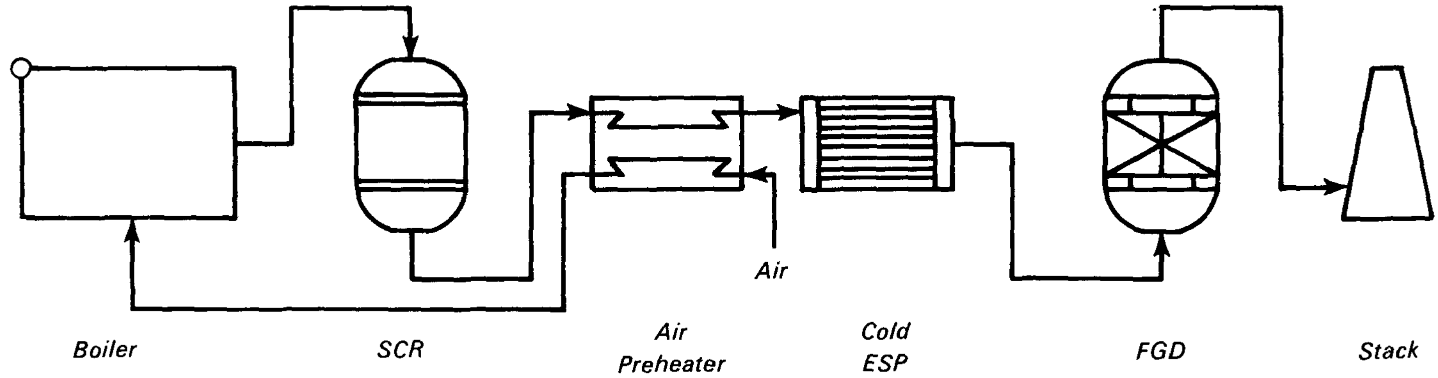
One vendor indicated that investment costs for a 90 percent NO_x removal system are 20 to 25 percent higher than those for an 80 percent system applied to an oil-fired boiler if increased NH_3 emissions are allowable. For 90 percent NO_x control with low NH_3 emissions, the costs will be 40 to 50 percent greater, rather than 20 to 25 percent.

Application of SCR

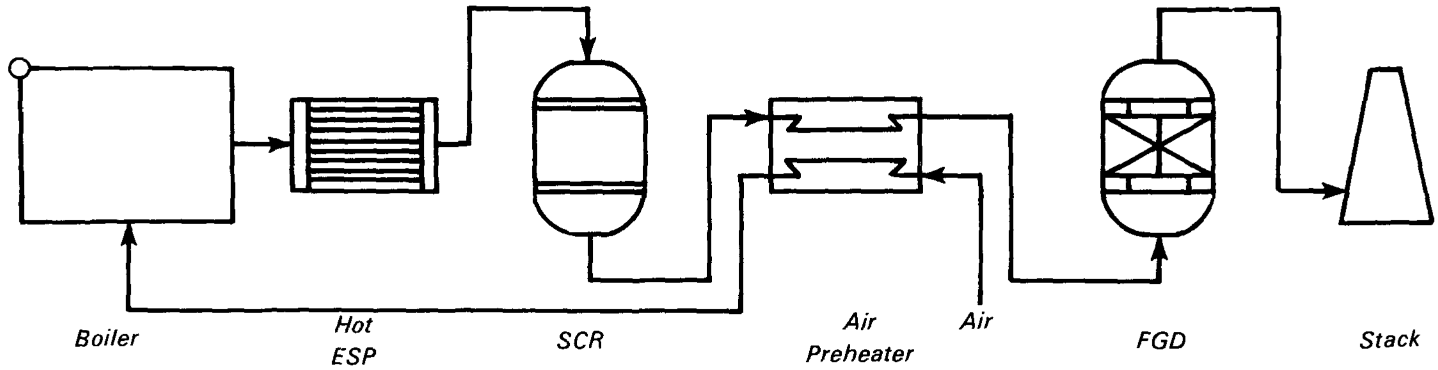
Flue Gas Processing Considerations

When installing a boiler which includes an SCR system (or retrofitting an SCR system to an existing unit), it is necessary to consider the flue gas processing system as a whole since the location of equipment relative to other pieces can have significant downstream impacts. The temperature requirements of the NO_x reduction reactions are 370 ±~50°C, and economics limit the location of the reactor to upstream of the air preheater. Temperature regulation may be necessary to limit temperature excursions outside this range. Where required, temperature control can be obtained by either load regulation or bypassing hot flue gas around the economizer and mixing it with the main gas stream.

In Japan, the particulate control devices are almost always electrostatic precipitators (ESP), installed either upstream of the SCR system or downstream of the air preheater. This option results in the two basic arrangements of flue gas processing equipment, shown in Figure 1.



Arrangement A. Cold-Side Particulate Removal — Example: Chugoku Electric Shimonoseki Power Station



Arrangement B. Hot-Side Particulate Removal — Example: Electric Power Development Company Takehara Power Station

Figure 1. *Basic equipment layout options for processing flue gas from a coal-fired boiler.*

In Japan, hot-side ESP's will be used with most of the SCR systems currently under construction on coal-fired boilers. There are several reasons for this: one is that, when these systems were designed several years ago, SCR catalysts had not been adequately proven with high ash loadings. Other equally important reasons are that cold-side ESP's are not effective with the low sulfur coal sometimes received in Japan, and the concern that NH_3 compounds in the collected fly ash will affect by-product utilization. However, it is anticipated that many SCR systems currently being designed will utilize cold-side ESP's.

Downstream Impacts and Countermeasures

The SCR system can potentially impact the air preheater, ESP, and FGD system. The most important of these is the air preheater. Several plants have reported problems with air preheater plugging and corrosion by NH_4HSO_4, product of a condensation reaction between NH_3, SO_3, and H_2O. In many cases, more rigorous soot blowing procedures have controlled the plugging to the extent that water washing the air preheater is only required once per year during a scheduled boiler outage. In other cases, however, water washing is necessary every few months, even with NH_3 and SO_3 concentrations of <4 ppm and a temperature of 140°C. The plugging occurs at the interface between the intermediate- and cold-temperature elements where the temperature is cool and soot blowing is least effective.

Where more severe problems are anticipated, such as after a hot-side ESP, a modified air preheater design will be used. This design consists of combining the intermediate and cold sections into a single element which permits more effective soot blowing which, for the new design, is done from both the hot and cold sides. Also, the large element is made of corrosion resistant, coated material similar to typical cold-end elements.

The NH_3 emitted by SCR systems is not expected to adversely impact the operation of a cold-side ESP; on oil-fired boilers in Japan, NH_3 is often injected upstream of these units for ash conditioning. However, the NH_3 can affect the fly ash utilization in by-products by depositing on the surface of the fly ash. The impact of SCR systems on baghouse efficiency is not known in Japan where it is being investigated at two pilot units.

Systems in which a limestone FGD unit exists downstream of an SCR unit will be operational in the near future in Japan. There are no adverse effects of SCR systems anticipated on the performance of downstream limestone/gypsum FGD systems. The FGD system will absorb NH_3; however, reliability and

SO_2 removal should not be affected.

Environmental Impacts

SCR systems on gas- and oil-fired boilers in Japan emit NH_3; however, *since the concentrations are 3-10 ppm,* it is not considered to be a serious air quality impact. Ammonia emissions from full-scale, coal-fired applications have not been quantified; however, downstream equipment will probably mitigate any air impacts.

Visible plumes of ammonium sulfite have been known to occur during certain atmospheric conditions with NH_3-based FGD systems. Based on Japanese experience with situations in which 10 ppm of NH_3 enters the FGD system, Japanese researchers do not feel that SCR systems will cause visible plume formation.

SCR systems can also have water impacts resulting from: NH_3 in the leachate from ash that has been used as landfill, NH_4HSO_4 in the air preheater wash water, NH_3 in FGD blowdown, and ammonia compounds in the water used to wash gypsum produced by an FGD system. Where the concentrations are high enough to cause water pollution problems, an activated sludge water treatment system may be necessary. Solid waste from SCR systems consists of spent catalyst material. A definite disposal method has not yet been established primarily because none of the commercial SCR units have required a catalyst change. This area requires further investigation for U.S. applications.

SCR Installations

Extent of Application

SCR systems have been installed on many gas- and oil-fired boilers and are operating successfully. One application to a coal-fired boiler and four others are scheduled to startup within a year.

With all of the coal-fired applications, the catalyst and reactor are designed in a vertical downflow arrangement to minimize the potential for plugging by fly ash. Soot blowers are installed on some units above the beds to control ash buildup. These are installed as a conservative design measure in case plugging develops; however, they are not expected to be necessary. There are up to four beds within the reactor consisting of one or two layers of catalyst modules, each of which contains many catalyst elements.

In some cases a dummy layer of catalyst modules is installed at the reactor inlet to prevent abrasion of the catalyst surface.

Operation and Maintenance

The labor requirements of the operating full-scale systems are small. No additional operating personnel are required and maintenance labor consists primarily of NH_3 and catalyst loading, and cleaning the air preheater during the annual outage. Since there have been no catalyst changes to date, the labor estimates for this work vary widely. Operators indicate that the SCR processes themselves are very reliable, essentially 100 percent. However, in some cases, a boiler shutdown has been necessary where air preheater plugging has occurred. In most cases, steps have been taken to reduce the plugging rate to the extent that cleaning is only required during normal boiler outages.

Additional Aspects of SCR

Government Agencies

Environmental regulation in Japan is similar to that in the US in that a federal agency establishes the minimum requirements for the country as a whole. Local governments have the option of adopting the federal standards or establishing their own, more stringent standards. The Japan Environment Agency recently modified standards for ambient air quality and established three concentration ranges based on a daily average sample. Above 0.06 ppm NO_2, countermeasures must be undertaken to reduce the level to 0.06. Within the range 0.04 to 0.06 ppm NO_2, no significant change is allowed; areas below 0.04 ppm NO_2 are allowed to deteriorate to 0.04 ppm. The Environment Agency also sets emission standards for point sources. Emission limits for new sources are shown in Table 1.

The local governments, city and prefectural, have required many of the SCR applications. This occurs primarily when the plant owners negotiate with the local governments over a plant siting or expansion. Citizen groups appear to have a significant voice in these deliberations. The cooperative spirit which exists between the local citizens, the plant personnel, and the government provides the driving force for the plant's compliance with the negotiated emission limits and the successful implementation of new control technology. Further, the local governments utilize sophisticated air quality monitoring systems to ensure that the public is protected and that regulations are complied with.

Industry Groups

The two industry groups contacted are looking to the future when coal will be a much more significant energy source. Coal-fired boiler capacity is expected to increase from 4410 MW in 1978 to about 34,500 MW by 1995 and NO_x control will probably be required on all of these. They feel that combustion modifications and SCR have been successfully demonstrated on gas- and oil-fired boilers and that these techniques will also be useful on coal-fired units.

Instrument Design

The application of SCR systems in Japan has necessitated the development of instruments capable of measuring NO_x and NH_3. Coal-fired applications present the most difficult situation for the instruments since the flue gas contains SO_2, SO_3, and fly ash. Chemiluminescent monitors, most commonly used for NO_x measurement, apparently operate satisfactorily with coal-fired flue gas.

Usually the NH_3 monitor is a chemiluminescent NO monitor with an up-

Table 1. *National Emission Limits for New Sources*

Fuel	*Unit Size 1000 Nm³/hr*	*O_2 Concentration %*	*No_x Emission Limit ppm*
Gas	*>500*	*5*	*60*
	40 - 500	*5*	*100*
	10 - 40	*5*	*130*
	<10	*5*	*150*
Oil	*>500*	*4*	*130*
	10 - 500	*4*	*150*
	<10	*4*	*180*
Coal	*All Sizes*	*6*	*400*

stream catalytic converter which converts NH_3 to NO. Sulfur oxides apparently interfere with the monitoring equipment and there have been additional problems with the converters giving incomplete NO conversions. An alternative NH_3 emissions monitor for coal-fired flue gas has not been perfected; however, several techniques are being developed.

Conclusions

SCR has been applied to many gas- and oil-fired boilers, both utility and industrial, and these are operating successfully. Full-scale tests on coal-fired utility boilers are just beginning; several units will be on-line in the next year or two. These full-scale units will be incorporating some new design features developed specifically to deal with coal-related problems such as air preheater plugging and catalyst erosion.

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The complete report, entitled "Selective Catalytic Reduction and NO_x Control in Japan," (Order No. PB 81-191 116; Cost: $20.00, subject to change) will be available only from:
National Technical Information Service
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