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# Proceedings of the Joint Symposium on Stationary Combustion NO<sub>X</sub> Control

Volume II Utility Boiler NO<sub>X</sub> Control by Flue Gas Treatment



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## Proceedings of the Joint Symposium on Stationary Combustion NO<sub>X</sub> Control

### Volume II Fun Utility Boiler NO<sub>X</sub> Control by Flue Gas Treatment

Symposium Cochairmen Robert E. Hall, EPA and J. Edward Cichanowicz, EPRI

Program Element No. N130

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### PREFACE

These proceedings document more than 50 presentations given at the Joint Symposium on Stationary Combustion NO<sub>x</sub> Control held October 6-9, 1980 at the Stouffer's Denver Inn in Denver, Colorado. The symposium was sponsored by the Combustion Research Branch of the Environmental Protection Agency's (EPA) Industrial Environmental Research Laboratory-Research Triangle Park and the Electric Power Research Institute (EPRI). The presentations emphasized recent developments in NO<sub>x</sub> control technology. Cochairmen of the symposium were Robert E. Hall, EPA, and J. Edward Cichanowicz, EPRI. Introductory remarks were made by Kurt E. Yeager, Director, Coal Combustion Systems Division, EPRI, and the welcoming address was given by Roger L. Williams, Regional Administrator, EPA Region VIII. Stephen J. Gage, Assistant Administrator for Research and Development, EPA, was the keynote speaker. The symposium had l1 sessions:

- I: NO<sub>X</sub> Emissions Issues
  Michael J. Miller, EPRI, Session Chairman
- II: Manufacturers Update of Commercially Available Combustion Technology Joshua S. Bowen, EPA, Session Chairman
- III: NO<sub>X</sub> Emissions Characterization of Full Scale Utility Powerplants
  David G. Lachapelle, EPA, Session Chairman
- IV: Low NO<sub>X</sub> Combustion Development Michael W. McElroy, EPRI, Session Chairman
- Va: Postcombustion NO<sub>X</sub> Control
  George P. Green, Public Service Company of Colorado,
  Session Chairman
- Vb: Fundamental Combustion Research
  Tom W. Lester, EPA, Session Chairman
- VI: Status of Flue Gas Treatment for Coal-Fired Boilers Dan V. Giovanni, EPRI, Session Chairman
- VII: Small Industrial, Commercial, and Residential Systems Robert E. Hall, EPA, Session Chairman
- VIII: Large Industrial Boilers
  J. David Mobley, EPA, Session Chairman
- IX: Environmental Assessment
  Robert P. Hangebrauck, EPA, Session Chairman
- X: Stationary Engines and Industrial Process Combustion Systems John H. Wasser, EPA, Session Chairman
- XI: Advanced Processes
  G. Blair Martin, EPA, Session Chairman

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### ASSESSMENT OF NO FLUE GAS TREATMENT TECHNOLOGY

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### **ABSTRACT**

The Environmental Protection Agency has maintained a program to further the advancement of  $NO_X$  control by flue gas treatment technology since the early 1970's. The program consists of technology assessment studies in conjunction with small scale experimental projects. These activities have shown that 80-90% reduction of  $NO_X$  emissions by selective catalytic reduction with ammonia has been commercially demonstrated on gas- and oil-fired sources in Japan, and that such processes are ready for test application on coal-fired sources. The Japanese experience, combined with experimental projects in the U.S., should establish the technology as a viable control technique for use in tackling  $NO_X$  environmental problems in the U.S. However, some significant technical concerns need to be addressed in demonstration projects before wide-spread application of the technology can be recommended.

### INTRODUCTION

The U.S. Environmental Protection Agency (EPA) has maintained a research and development program since the early 1970's to advance flue gas treatment technology for the control of nitrogen oxide  $(NO_{\hat{\mathbf{X}}})$ ' emissions from stationary combustion sources. The objective of this program has been to demonstrate the technical feasibility of a 90% reduction of  $NO_{\hat{\mathbf{X}}}$  emissions in a cost effective, energy efficient, and environmentally sound manner. Emphasis of the program has been on control of  $NO_{\hat{\mathbf{X}}}$  emissions from coal-fired boilers.

This program has been maintained since technology for highly efficient NO<sub>X</sub> reduction may be required as a control strategy for some of the environmental problems in the United States. Specifically, application of the technology may be required by regulations which address Prevention of Significant Deterioration, Visibility Protection, Acid Rain, or Nonattainment of National Ambient Air Quality Standards (NAAQS). The promulgation of a short term nitrogen dioxide (NO<sub>2</sub>) NAAQS may increase the number of nonattainment areas for NO<sub>2</sub> and may increase the need for application of flue gas treatment technology (1).

To ameliorate their environmental problems, the Japanese have developed and applied  $\mathrm{NO}_{\mathrm{X}}$  flue gas treatment technology to gas- and oil-fired sources and are planning applications on coal-fired sources. They have also conducted extensive research on dry and wet processes for simultaneous control of  $\mathrm{NO}_{\mathrm{X}}$  and sulfur oxide ( $\mathrm{SO}_{\mathrm{X}}$ ) emissions. Thus, a major portion of EPA's program has been to investigate Japanese technology for potential applications to coal-fired boilers in the U.S. Accordingly, this paper will include:

- \*Assessment of Technology in Japan
  - -Dry  $NO_{\mathbf{X}}$  Processes
  - -Dry Simultaneous NO<sub>x</sub>/SO<sub>x</sub> Processes
  - -Wet  $NO_X$  and  $NO_X/SO_X$  Processes
- \*Assessment of Technology for Application in the U.S.
  - -Cost Estimates
  - -Energy and Environmental Impacts
  - -Experimental Projects

Finally, conclusions that can be drawn from these assessment activities will be presented.

### ASSESSMENT OF TECHNOLOGY IN JAPAN

Japanese technology for control of  $NO_X$  and simultaneous control of  $NO_X$  and  $SO_X$  by flue gas treatment techniques is more advanced than any other country's. EPA has sponsored the publication of periodic reports and papers to facilitate the transfer of information from Japan to the United States. These documents have been mainly prepared by Jumpei Ando of Chuo University, Tokyo, Japan (2,3). Since most of the development and application of  $NO_X$  flue gas treatment technology has been in Japan, it is appropriate for any assessment to begin with the status of the technology in Japan.

### DRY NOx PROCESSES

Selective catalytic reduction (SCR) of  $NO_X$  with ammonia (NH<sub>3</sub>) is the only process that has achieved notable success in treating combustion flue gas for 90% removal of  $NO_X$  and has progressed to the point of commercial application. SCR processes are based on the preferential reaction of NH<sub>3</sub> with  $NO_X$  rather than with other flue gas constituents. Since oxygen  $(O_2)$  enhances the reduction, the reactions can best be expressed as:

$$4NH_3 + 4NO + O_2 \longrightarrow 4N_2 + 6H_2O$$
 (1)

$$4NH_3 + 2NO_2 + O_2 \longrightarrow 3N_2 + 6H_2O$$
 (2)

Equation 1 represents the predominate reaction since approximately 95% of the  $NO_X$  in combustion flue gas is in the form of nitric oxide (NO). Therefore, under ideal conditions, a stoichiometric amount of NH<sub>3</sub> can be used to reduce  $NO_X$  to harmless molecular nitrogen (N<sub>2</sub>) and water vapor (H<sub>2</sub>O).

In practice, an NH3:NO mole ratio of about 1:1 has typically reduced  $NO_X$  emissions by 90% with a residual NH3 concentration of less than 20 ppm.

The SCR processes require a reactor, a catalyst, and an ammonia storage and injection system. Due to increased pressure drop across the SCR reactor, some increase in boiler fan capacity, or possibly an additional fan, may be necessary.

The optimum temperature for the  $NO_X$  reduction reaction without a catalyst is about  $1000\,^{\circ}\text{C}$  ( $1830\,^{\circ}\text{F}$ ). However, the catalyst effectively reduces the optimum reaction temperature to the  $300\,^{\circ}$  to  $450\,^{\circ}\text{C}$  ( $570\,^{\circ}$  to  $845\,^{\circ}\text{F}$ ) range. To obtain flue gas temperatures in this range and to avoid the requirement for large amounts of reheat, the reactor is usually located between the boiler economizer and the air preheater. A typical flow diagram is shown in Figure 1. Obviously, the reactor and catalyst are the critical elements of the process and warrant further discussion.

Catalysts with vanadium compounds were found to promote the reduction of  $NO_X$  with  $NH_3$  and to be unaffected by the presence of  $SO_X$ . Titanium dioxide ( $TiO_2$ ) was found to be an acceptable carrier, since it is resistant to attack from  $SO_3$ . Therefore, many  $SO_X$  resistant catalysts are based on  $TiO_2$  and  $V_2O_5$ ; however, constituents and concentrations of . most catalysts are proprietary.

Reactor and catalyst configurations also vary with the application, primarily to accommodate the different particulate concentrations.

Natural-gas-fired boilers employ SCR catalysts (as spherical pellets, cylinders, or rings) and reactor vessels (as fixed packed beds).

However, designs for use with oil- and coal-fired boilers have to be capable of tolerating particulates (fly ash) in the flue gas stream. For these applications, a parallel flow catalyst is preferred. Parallel flow means that the gas flows straight through the open channels parallel to the catalyst surface. The particulates in the gas remain entrained while NO<sub>X</sub> reaches the catalyst surface by turbulent convection and diffusion. An alternative to the parallel flow catalysts is the parallel passage reactor. In this design, the catalyst material is arranged in channels and held in place by a metal screen. The operating principle is similar to that of the parallel flow catalyst.

The various parallel flow catalyst shapes are shown in Figure 2. The catalyst may be a homogeneous material or may be composed of an active material coated on the walls of a metallic or ceramic carrier. The parallel flow catalysts are normally manufactured in a unit cell configuration about 1 m<sup>3</sup> as shown in Figure 3. The cells are stacked in banks in the reactor as shown in Figure 4.

Even though much progress has been made in catalysts and reactor design, some problems still remain. The catalysts may not be resistant to all contaminants in flue gas or be able to tolerate high particulate loadings. In addition, fine particulates, smaller than about 1  $\mu\text{m}$ , may blind the catalyst surface. Long term operation without catalyst plugging or catalyst erosion needs to be demonstrated for coal-fired applications. Catalyst life also needs to be extended from the current guarantees of 1 to 2 years for applications with SO\_X and particulates in the gas stream.

One of the major concerns with SCR processes is the formation of solid ammonium sulfate  $[(NH_4)_2SO_4]$  and liquid ammonium bisulfate  $(NH_4HSO_4)$  downstream of the reactor. The formation conditions are

difficult to avoid since some unreacted NH3 from a SCR system and some SO3 from combustion of sulfur-containing fuels is expected. The biggest problem seems to be deposition of (NH4)2SO4 and NH4HSO4 on the air preheater. These compounds are corrosive and can form deposits which plug the air preheater. Air preheater problems were found to be most severe on high-sulfur oil-fired applications and coal-fired units which employ a hot electrostatic precipitator (ESP) ahead of the NO<sub>x</sub> reactor and the air preheater. Systems which accept full particulate charge of a coal-fired boiler through the air preheater should have less difficulty with pluggage of the air preheater. In these systems, the fly ash apparently scours the surface of the air preheater to remove any deposits, or the ammonium compounds deposit on the fly ash and are carried through the air preheater by the fly ash. However, increased soot blowing, from both the hot and cold sides, and water washing of the air preheaters appear necessary for most applications. The wash water from cleaning air preheaters as well as purge streams from a flue gas desulfurization (FGD) unit may require treatment to remove NH2 before being discharged. Modifications to the conventional air preheater design are also being developed to address the ammonium sulfate problem; however, these units have not been applied on full scale systems as yet.

Other concerns and potential problems include: emission of NH<sub>3</sub> and NH<sub>3</sub> compounds; causing or increasing the emission of undesirable compounds such as SO<sub>3</sub>; affecting the performance of downstream pollution control equipment such as FGD processes, ESPs, and baghouses; lack of proven NH<sub>3</sub> analytical control systems; sensitivity of the process to temperature changes due to boiler load swings; disposal or reclamation of spent catalysts in an environmentally acceptable manner; and reliability of the process and its effect on the boiler system's availability.

Despite these potential problem areas and uncertainties, the processes have been successfully installed and operated in Japan on gasand oil-fired boilers, and coal-fired units are being constructed. The companies listed in Table I were visited by a team of EPA, Tennessee Valley Authority, and Radian Corporation personnel in March 1980 to survey the status of the technology in Japan. The gas- and oil-fired operating units reported very high reliability and the capability to overcome operational difficulties. In general, no labor was added to operate a SCR system on retrofit applications, and new systems do not plan additional personnel for the NO<sub>x</sub> system. Generally, no maintenance was reported other than cleaning the air preheater during annual shutdowns. The control systems are usually fairly simple and employ a feedforward control system based on inlet  $NO_{\mathbf{x}}$  concentration with fine tuning supplied by feedback of the outlet NOx concentration. Most gas- and oil-fired applications also utilized an outlet NH3 emissions monitor; however, an NH3 emissions monitor has not been perfected for coal-fired applications. NH3 emissions were reported to be very low (0-5 ppm) in most cases. NH3 emission limitations are not prescribed by governmental agencies, but low NH3 emissions are deemed essential to minimizing problems with downstream equipment. No problems were noted with NH3 fuming or emissions of cyanide, nitrates, nitrosoamines, or other harmful species.

The  $\mathrm{NO_X}$  emission rates were very low when SCR units were employed. As shown in Table I, gas-fired units were controlled to about 10 ppm of  $\mathrm{NO_X}$  and oil-fired units to 15-35 ppm. Further, coal-fired units are being constructed which will demonstrate control to 40-80 ppm of  $\mathrm{NO_X}$ . It is important to note that the Japanese seem to prefer 80%  $\mathrm{NO_X}$  removal as the optimum control level since it minimizes capital and operating costs as well as energy and environmental impacts while maximizing operability, reliability, and system life.

The coal-fired applications of SCR technology in Japan are of particular interest to U.S. personnel since coal-fired sources represent a major NO<sub>X</sub> control problem in the U.S. Four SCR systems for coal-fired boilers were under construction in Japan and are shown in Table I. Further, construction of additional coal-fired boilers are being planned and SCR units are expected to be installed on most of these units. Therefore, the Japanese experience should provide valuable information for many years for U.S. personnel interested in NO<sub>X</sub> control.

The SCR units were installed to meet local governmental  $NO_X$  emission limitations and are not necessary to comply with the Japanese national standard. Compliance with local and national environmental agreements is achieved through a sincere cooperative spirit that exists between industry and government to solve the country's pollution problems. Local governments maintain a very comprehensive and extensive instrumentation system to monitor ambient pollutant concentrations and emission rates from major emitters. Although the local governments have no enforcement authority, violations are rarely, if ever, detected since the companies do not want to jeopardize the "good will" that exists between the industrial, governmental, and public sectors.

In summary, it should be emphasized that SCR technology is being successfully operated in Japan on large scale gas- and oil-fired sources and that large scale applications to coal-fired sources are under construction with equal success anticipated.

### DRY SIMULTANEOUS NOx/SOx PROCESSES

The Shell Flue Gas Treating process is a unique variation of SCR technology that can simultaneously remove both  $NO_{\mathbf{x}}$  and  $SO_{\mathbf{x}}$  from combustion

flue gas. The process uses copper oxide (CuO) supported on stabilized alumina placed in two or more parallel-passage reactors. The reactions, which characterize process operation, can be expressed as:

### Acceptance

$$CuO + 1/2 O_2 + SO_2 - CuSO_4$$
 (3)

$$4NO + 4NH_3 + O_2 \xrightarrow{CuSO_4} 4N_2 + 6H_2O$$
 (4)

### Regeneration

$$CuSO_4 + 2H_2 \longrightarrow Cu + SO_2 + 2H_2O$$
 (5)

$$Cu + 1/2 O_2 \longrightarrow CuO$$
 (6)

Flue gas is introduced at 385°C (725°F) into one of the reactors where the SO<sub>2</sub> reacts with CuO to form copper sulfate (CuSO<sub>4</sub>). The CuSO<sub>4</sub> and, to a lesser extent, the CuO act as catalysts in the reduction of NO<sub>x</sub> with NH<sub>3</sub>. When the reactor is saturated with CuSO<sub>4</sub>, flue gas is switched to a fresh reactor for acceptance of the flue gas, and the spent reactor is regenerated. In the regeneration cycle, hydrogen (H<sub>2</sub>) is used to reduce the CuSO<sub>4</sub> to copper (Cu), yielding a SO<sub>2</sub> stream of sufficient concentration for conversion to sulfur or sulfuric acid. The Cu in the reactor is oxidized, preparing the reactor for acceptance of the flue gas again. Between acceptance and regeneration, steam is injected into the reactor to purge the remaining flue gas or H<sub>2</sub> to eliminate any possibility of combustion. The process can also be operated in the NO<sub>x</sub>-only mode by eliminating the regeneration cycle, or in the SO<sub>x</sub>-only mode by eliminating the NH<sub>3</sub> injection.

The process has been installed in Japan on a heavy-oil-fired boiler treating 120,000  $\rm Nm^3/hr$  of flue gas. The unit has demonstrated 90%  $\rm SO_2$  removal and 70%  $\rm NO_X$  removal. UOP Process Division is the licensor of the process in the United States.

### WET NO<sub>x</sub> AND NO<sub>x</sub>/SO<sub>x</sub> PROCESSES

The wet  $\mathrm{NO_X}$  and simultaneous  $\mathrm{NO_X/SO_X}$  processes developed to date cannot compete economically with dry processes for removal of  $\mathrm{NO_X}$  from combustion flue gas. This is primarily due to the complexity, limited applicability, and water pollution problems associated with the wet processes.

Since  $\mathrm{NO}_{\mathbf{X}}$  SCR systems have been successfully installed and operated in Japan on gas- and oil-fired sources, it is expected that equal success could be obtained in the U.S. However, coal-fired boilers in the U.S. are the primary concern for control of  $\mathrm{NO}_{\mathbf{X}}$  from stationary combustion sources. Therefore, EPA has focused on technology assessment studies to determine cost estimates, energy requirements, and environmental impacts of applying the technology on coal-fired boilers in the U.S. In parallel with the technology assessment studies, small scale experimental projects have been undertaken to demonstrate the performance of the technology.

### COST ESTIMATES

Preliminary cost estimates for application of SCR processes to a coalfired utility boiler were made by the Tennessee Valley Authority through an interagency agreement with EPA. (This study was cosponsored by the Electric Power Research Institute.) The basis for this estimate was a new, 500 MW boiler firing Eastern bituminous coal with a heating value of 24.4 MJ/kg (10,500 Btu/1b), a sulfur content of 3.5%, and an ash content of 16%. The study assumed operation of the boiler for 7000 hr/yr and the SCR process for 90% reduction of  $NO_{x}$ . The estimates were based on a capital investment in mid-1979 and an annual revenue requirement in mid-1980. Results of that study, summarized in Table II, indicate that the capital costs of SCR processes will be about \$42/kW or \$9.8/acfm and that annual revenue requirements will be about 2.7 mills/kWh. (Since the volume of gas treated is the dominating parameter in determining the cost of installing an SCR system, the \$/acfm factor may be the most meaningful in determining representative costs of firing various coals in various sized plants.) The study also indicated that the dry simultaneous  $NO_X/SO_X$  process would be competitive with these costs if the cost of a FGD process for SO2 control were added to the cost of the NO<sub>x</sub> SCR process, but that the cost of wet simultaneous NO<sub>x</sub>/SO<sub>x</sub> processes would be significantly higher.

The cost of applying  $\mathrm{NO}_{\mathbf{X}}$  SCR technology to industrial boilers was estimated in a study for EPA by Radian Corporation (5). It was found that the fuel fired, boiler size, and design  $\mathrm{NO}_{\mathbf{X}}$  removal had significant effects on the system cost. The results of this study are summarized in Table III.

### ENERGY AND ENVIRONMENTAL IMPACTS

The SCR processes are projected to require about 0.2% of the boiler capacity. This assumes that flue gas reheat will not be required and does not include an energy requirement represented by raw materials (4).

The major concerns from an environmental impact viewpoint appear to be emissions and discharge of NH<sub>3</sub> and NH<sub>3</sub> compounds and disposal of spent catalysts. Operational techniques to limit NH<sub>3</sub> emissions and methods for regenerating or reclaiming the catalysts are under development to minimize problems in this regard (5).

### EXPERIMENTAL PROJECTS

To determine the actual performance and cost of applying SCR processes in the U.S., several demonstration projects have been planned as summarized in Table IV. Relative to the EPA pilot plant projects, the basic objective is to demonstrate the feasibility of the processes for 90% control of  $NO_X$  or  $NO_X/SO_X$  emissions from a coal-fired source.

The projects were divided into four phases:

I - Design

II - Procurement and erection

III - Startup, debugging, and optimization

IV - Long term operation and assessment.

Phase IV began in June 1980 for both projects. Therefore, it is not possible to provide results of the projects in this paper. However, it should be noted that numerous problems have been encountered in operating the pilot plants. One major problem area has been sampling and analytical techniques for NO, NO<sub>2</sub>, NO<sub>x</sub>, SO<sub>2</sub>, SO<sub>3</sub>, and NH<sub>3</sub>. Successes and failures of the pilot plants will be reported in full in the final project reports. Since papers on each project or process listed in Table IV will be presented at this Symposium, further details will not be presented in this paper.

If the coal-fired pilot plants and oil-fired demonstration scale plants are successful, then a coal-fired prototype or demonstration scale plant (10-100 MW) will be desirable. It is conceivable that such a plant could be built in conjunction with a new or expanding facility in an area with nonattainment, prevention of significant deterioration, visibility, or other environmental constraints. In this manner, the environmental impact of the facility could be minimized, and SCR technology for  $\mathrm{NO}_{\mathbf{x}}$  control could be commercially demonstrated.

### CONCLUSIONS

The following conclusions can be drawn from this assessment of  $\ensuremath{\text{NO}_{\times}}$  flue gas treatment technology.

- NO<sub>X</sub> flue gas treatment processes, based on selective catalytic reduction (SCR) of NO<sub>X</sub> with NH<sub>3</sub>, have been successfully demonstrated on commercial-scale gas- and oil-fired sources in Japan for >80% NO<sub>X</sub> removal.
- 2. SCR systems are being installed on commercial-scale coal-fired sources in Japan which will demonstrate the viability of the technology for coal-fired sources.
- 3. SCR systems are being evaluated on pilot-scale coal-fired sources and on a demonstration-scale oil-fired source in the U.S. for 90%  $NO_X$  control. One of the pilot plants is evaluating dry simultaneous  $NO_X/SO_X$  control. If these applications are successful, evaluation on a prototype-scale coal-fired facility will be desirable.
- 4. Although significant progress has been made in developing and applying the technology, several problem areas remain to be resolved (i.e., impact on downstream equipment, long term performance, and reliability).
- 5. Experimental projects, in conjunction with technology assessment studies, will enable a determination of the feasibility of applying  $NO_X$  SCR processes in the U.S.

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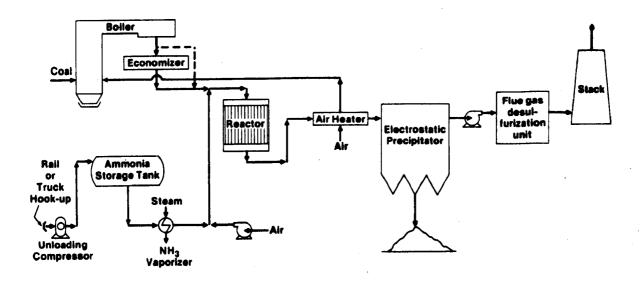


Figure 1. Typical flowsheet for NO<sub>X</sub> selective catalytic reduction processes (4).

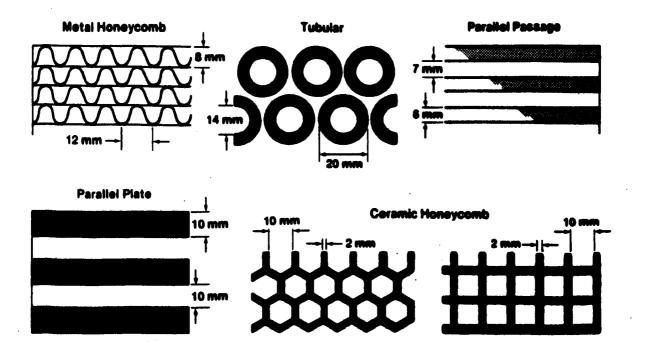


Figure 2. Types of parallel flow catalyst shapes for  $NO_X$  selective catalytic reduction processes (3).

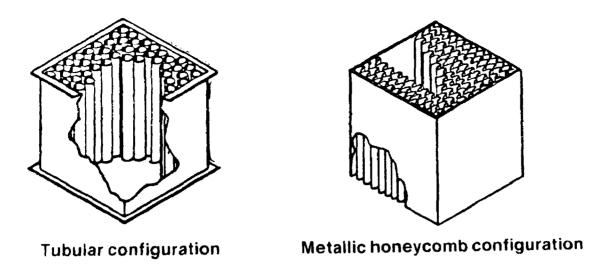


Figure 3. Unit cells of parallel flow catalysts for  $NO_X$  selective catalytic reduction processes (3).

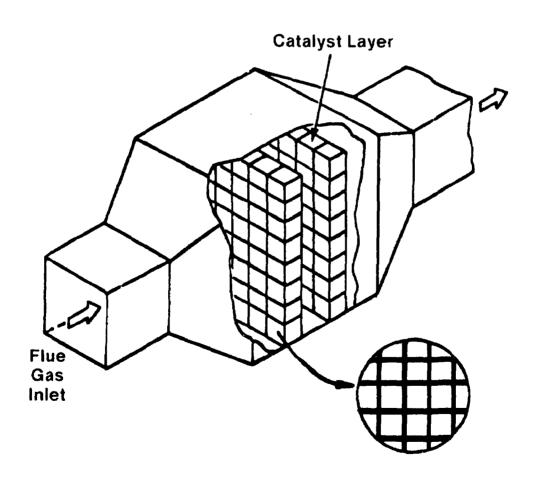


Figure 4. Parallel flow reactor for NO<sub>X</sub> selective catalytic reduction processes (3).

TABLE I. SELECTED INSTALLATIONS WITH NOx SELECTIVE CATALYTIC REDUCTION SYSTEMS IN JAPAN<sup>a</sup>

		Boiler			NO <sub>x</sub> SCR System					
Сощрапу	Station	No.	Fuel	Size (MW)	Size (MW)	NO <sub>x</sub> Removal (%)	Outlet NO <sub>x</sub> (ppm)	Vendor <sup>b</sup>	Startup	
Chubu Electric	Chita	5	Gas	700	700	80	8-10	в-н	Mar. 1978	
		6	Gas	700	700	80	8-10	в-н	Apr. 1978	
		4	011	700	700	80	20	MHI	Mar. 1980	
Chugoku Electric	Kudamatsu	2	Oil	375	375	80	32	IHI	Apr. 1979	
	•	3	011	700	700	80	26	IHI	Sep. 1979	
Fuji Oil	Sodegara	7	011	40	40	87	15	MHI	Jan. 1978	
			CO & 011	10	10	92	15	JGC	Jul. 1976	
Chugoku Electric	Shimonoseki	1	Coal	175	175	50	250	MHI	Apr. 1980	
Hokkaido Electric	Tomakomai	1	Coal	350	90	>80	<40	в-и	Oct. 1980	
Electric Power	Takehara	1	Coal	250	125	80	80	<b>B-</b> H	Jun. 1981	
Development Co.		1	Coal	250	125	80	80	KHI	Jun. 1981	

a) These installations were visited by a team of EPA, TVA, and Radian personnel in March 1980.

b) B-H Babcock-Hitachi

MHI Mitsubishi Heavy Industries

IHI Ishikawajima-Harima Heavy Industries

JGC JGC Corporation

KHI Kawasaki Heavy Industries

TABLE II. ESTIMATED COSTS FOR EMISSION CONTROL SYSTEMS FOR UTILITY BOILERS

	Capital Costs (\$/kW)				Annualized Costs (mills/kWh)			
Process Types	NO <sub>X</sub>	so <sub>2</sub>	PM	Total	NO <sub>X</sub>	so <sub>2</sub>	PM	Total
SCR, FGD, ESP	42	101	22	165	2.7	4.2	0.7	7.6
Dry Simultaneous NO <sub>x</sub> /SO <sub>x</sub>	<b>←</b> 1:	34>	29	163	<b>←</b> 6	.4>	0.9	7.3
Wet Simultaneous NO <sub>x</sub> /SO <sub>x</sub> /PM	<del></del>	200	<del>&gt;</del>	200	<del></del>	11.3	<del>&gt;</del>	11.3

NO<sub>X</sub> Control System
SO<sub>2</sub> Control System
Particulate Matter (PM) Control System

NO<sub>X</sub> Removal Efficiency
SO<sub>2</sub> Removal Efficiency
PM Removal Efficiency
Boiler Size
Fuel
Heating value
Sulfur content
Ash content

Basis for the Estimate:

Operation

Capital Investment

SCR Processes

Dry Simultaneous  $NO_x/SO_x$ Wet Simultaneous  $NO_x/SO_x/PM$ 

Annual Revenue Requirement

Selective Catalytic Reduction (SCR) with NH3 Limestone Flue Gas Desulfurization (FGD) Electrostatic Precipitator (ESP) for dry systems; wet scrubber for wet simultaneous systems 90% 90% 99.5% 500 MW, new Coal 24.4 MJ/kg (10,500 Btu/lb) 3.5% 16% 7000 hr/yr mid-1979 mid-1980 Average of UOP-Shell, Hitachi Zosen, Kurabo process costs UOP-Shell Average of Moretana Calcium and

Asahi ClO2 process costs

Source: Maxwell, Burnett, and Faucett (4).

22

TABLE III. ESTIMATED COST OF  $NO_{\mathbf{x}}$  SELECTIVE CATALYTIC REDUCTION SYSTEMS FOR INDUSTRIAL BOILERS

	Rating			Capital Costs	\$	Annual Costs \$			
Fue1	(MJ/s)	(MBtu/hr)	70% Control	80% Control	90% Control	70% Control	80% Control	90% Control	
Coal	8.8	30	181,900	213,100	249,600	104,200	117,500	133,900	
(Parallel Flow)	22.0	75	298,900	349,500	413,300	157,600	170,700	196,500	
	44.0	150	451,500	534,600	633,000	216,600	251,600	292,300	
***************************************	58.6	200	531,900	632,600	752,400	254,200	298,800	351,300	
Residual Oil	8.8	30	158,100	179,100	202,900	96,100	102,000	108,200	
(Parallel Flow)	44.0	150	378,200	436,200	503,100	181,180	200,900	222,860	
Residual Oil	8.8	<b>3</b> 0	144,600	154,700	166,700	120,400	125,100	129,900	
(Moving Bed)	44.0	150	259,700	300,200	347,000	167,700	184,800	203,600	
Distillate Oil	4.4	15	88,200	93,600	99,400	63,600	65,500	67,400	
(Fixed Packed Bed)	44.0	150	223,800	261,500	305,600	137,200	155,300	175,900	
Natural Gas	4.4	15	92,800	96,600	100,500	64,400	66,000	67,600	
(Fixed Packed Bed)	44.0	150	223,700	262,000	306,800	129,400	150,400	174,700	

Basis for Estimate:

Cost Basis Mid-1978

Load Factors
Coal 0.60
Residual 0il 0.55
Distillate 0il 0.45
Natural Gas 0.45

Coal Low Sulfur Western

Source: Jones and Johnson (5).

TABLE IV. PLANNED DEMONSTRATION PROJECTS OF  $NO_{\mathbf{x}}$  SELECTIVE CATALYTIC REDUCTION TECHNOLOGY IN THE U.S.

 Sponsor	Contractor	Location	Size (MWe)	Fuel	Planned NO <sub>X</sub> Reduction	Startup
 ronmental Protection ency	Hitachi Zosen	Georgia Power Company	0.5	Coal	90%	Fall 1979
 ronmental Protection ency	UOP Process Division	Tampa Electric Company	0.5	Coal	90%(a)	Fall 1979
tric Power Research stitute	Kawasaki Heavy Industries	Public Service Company of Colorado Arapahoe Station	2.5	Coal	90%	Winter 1980
nern California ison(b)	Kawasaki Heavy Industries	Southern California Edison Huntington Beach Station	100	011	90%	Fall 1981

a) 90% Reduction of  $SO_2$  also planned by the dry, simultaneous  $NO_X/SO_X$  process.

b) Required to meet regulations of the California Air Resources Board.

### STATUS OF SCR RETROFIT AT SOUTHERN CALIFORNIA EDISON HUNTINGTON BEACH GENERATING STATION UNIT 2

### By:

L. W. Johnson, C. L. W. Overduin, D. A. Fellows Souther California Edison Company P. O. Box 800 2244 Walnut Grove Avenue Rosemead, California 91770

### **ABSTRACT**

Utilities in the Southern California South Coast Air Basin are subject to a regulation (Rule 1135.1) requiring 90 percent NOx reduction. Rule 1135.1 is comprised of four basic compliance options of which the first two options require two stages with an intermediate milestone reduction and a demonstration unit of a 90 percent NOx reduction system.

This paper describes the Selective Catalytic Reduction (SCR) 107.5 MW Demonstration Facility SCE plans to install on one-half of Southern California Edison's Huntington Beach Unit 2, 215 MW boiler. The physical size, operation and maintenance, and controls for achieving 90 percent NOx reduction through normal load variations as well as the status of the project are discussed.

The system retrofit requirements are discussed with specific reference to the differences between the demonstration unit and other larger units and the site constraints for retrofit on the larger units. The operational and maintenance requirements for a systemwide retrofit and potential problem areas will also be reviewed.

The paper will present cost estimates for the Huntington Beach demonstration facility as well as SCE's projection of cost for adding SCR on the majority of its oil-fired units in the South Coast Air Basin. These costs will include capital as well as O&M. All costs will be presented in 1981 dollars.

### 1. INTRODUCTION

Utilities in the Southern California South Coast Air Basin are subject to a regulation (Rule 1135.1) requiring 90% NOx reduction. Over the past two years, many workshops and hearings have been held regarding the requirements of the rule. The latest version, adopted earlier this year, is comprised of four basic compliance options of which the first two options require two stages with an intermediate milestone reduction and a demonstration unit of a 90% NOx reduction system.

The demonstration unit requires the 90% NOx reduction on a unit of at least 100 megawatts in the Los Angeles basin by January 1, 1982. Depending on the options selected, a 90% NOx reduction basin wide must be achieved by 1988 or 1990. Considerable work is underway by the Southern California Edison Company to comply with the demonstration unit and much effort has gone into determining the impact of complying with the balance of the rule. This report describes the work performed to date.

### II. HUNTINGTON BEACH SCR DEMONSTRATION PROJECT

### FACILITY DESCRIPTION

The Huntington Beach Demonstration Facility employs a Selective Catalytic Reduction (SCR) process, in which nitrogen oxides in the boiler flue gas are reduced, using ammonia as a reducing agent. This facility will be capable of treating one half of the flue gas generated by Southern California Edison's Huntington Beach Unit 2, which is rated at 215 MW.

The SCR NOx removal process is depicted in Figure 1, and consists of the following five principle systems:

- o A reactor system consisting of a reactor to contain and support the catalyst modules, a ducting/damper system to divert flue gas through or past the reactor, and a reactor booster fan with bypass duct to draw the flue gas through the reactor.
- o An ammonia supply system consisting of a liquid ammonia storage tank, ammonia vaporizer and associated piping and controls to forward the ammonia vapor to the ammonia dilution skid.
- o An ammonia dilution and injection system consisting of a blower for diluting the ammonia with combustion air from the air preheater, and an ammonia flue gas mixer to inject the diluted ammonia vapor uniformly into the flue gas stream.

- o A control system consisting of ammonia injection controls, reactor booster fan control systems, a system to protect the boiler against negative pressure excursions and an emission monitoring system.
- o An electrical system utilizing the existing Unit 5 gas turbine auxiliary transformer which includes a 4160 V switchgear, 480 V switchgear, lighting facilities, communication system, grounding system, DC power system and all associated raceways and cable.

### **EQUIPMENT**

### Reactor

The reactor is designed for vertical flue gas flow and has a fixed catalyst bed. The catalyst bed is built from individual catalyst modules which can easily be removed from the reactor through a large access door utilizing a permanently installed hoist system.

### Reactor Booster Fan

The reactor booster fan is a centrifugal fan with air foil blading and is located downstream of the air preheater. The fan is directly coupled to a one speed electric motor and equipped with inlet vanes for flow control. A fan bypass duct and damper are provided to allow isolation of the fan from the existing boiler draft system.

### Ammonia Tank

The ammonia tank is a horizontal vessel designed for storage of liquid anhydrous ammonia. The tank is sized to satisfy maximum ammonia demand for approximately 14 days of continuous operation. The tank is equipped with an external electric vaporizer to insure adequate ammonia vaporization at high demand/low ambient temperature conditions.

### Equipment Data

Pertinent data for the above Apparatus as well as Huntington Beach Unit 2 are presented in Tables I and II.

### ARRANGEMENT

Refer to Figures 2 and 3 for plan and elevation views of the NOx removal system. The arrangement takes advantage of an existing duct elbow between the economizer outlet and inlet to the hot side of the vertical shaft air preheater. This duct will be replaced with new ducting including branches for reactor inlet and outlet, and a reactor bypass. The bypass will allow boiler operation during reactor inspection and service.

A portion of the stack breaching will be removed to accommodate the reactor booster fan and fan bypass duct. The fan can discharge either to the existing stack or, through damper manipulation, to its own new stub stack.

The latter discharge mode will allow visual monitoring of the treated flue gas opacity.

The reactor is located in a vertical position directly south of the unit and does allow for system expansion to treat all flue gas flow from Unit 2.

### CONTROLS AND INSTRUMENTATION

### Ammonia Injection

A control system will be installed at Huntington Beach to inject ammonia according to unit load and flue gas NOx content at a rate to maintain a preset 90% removal ratio and a NH<sub>3</sub> slip within 0 to 10 ppm. To set the NOx removal at 90%, NOx will be monitored before and after the reactor for calculation of the removal ratio. The differential signal between the calculated ratio and the setpoint will be fed back to the NH<sub>3</sub> injection controls for correction of the injection rate.

### Reactor Flue Gas Flow

A reactor booster fan is used to control the flue gas flow through the reactor and balance the flow between the east and west air preheaters. Air flow is controlled by inlet vanes which respond to boiler load and reactor differential pressure. The latter signal represents the actual flow through the reactor.

### Boiler Protection

The addition of the reactor booster fan has increased the occurrence of boiler negative pressure excursions with an associated increased risk of boiler implosion. Hence, a protective control system will be added to mitigate this increased risk. This control system will take the reactor booster fan out of the flue gas loop in response to boiler trips, low furnace pressure, FD fan trips and low reactor outlet pressure. The booster fan draft is eliminated by the opening of fast acting dampers in the booster fan by-pass duct and the fast closing of the fan isolation damper.

### Emission Instrumentation

Emission of NOx,  $NH_3$ ,  $SO_2$  as well as  $O_2$  levels will be monitored downstream of the air preheater. In addition, opacity of the flue gas subjected to the SCR process will be visually monitored at the new stub stack.

### TEST PROGRAM

The test program to be conducted at the Huntington Beach NOx removal facility will be designed primarily to determine the following process and operating parameters:

o NOx removal efficiency as a function of operating hours, ammonia injection rate, and  $NH_3$  slip. A removal rate of 90% for 7,000 hours with an  $NH_3$  slip of less than 10 ppm will be attempted.

- o Assessment of catalyst operating life. This is a most important parameter with a dominating impact on system operating cost.
- o Reactor and air preheater plugging as a result of deposits such as ammonia bisulfate.
- o Operating factors such as reactor draft loss and ammonia consumption.
- o Evaluation of ammonia injection controls, draft system controls, boiler protection controls and emission monitors and analyzers.

### PROJECT STATUS

As indicated by the basic project schedule shown in Figure 4, all engineering has been completed and the construction contract awarded. All major equipment has been procured and will be delivered to the site during the last quarter of 1980. One exception is the reactor itself, which is being fabricated in Japan at this time, and is scheduled for delivery in April, 1981. The system will be ready for testing on October 1, 1981. Testing for performance and compliance with the ARB Rule 1135.1 will be complete in January, 1982. Further testing will continue thereafter, to evaluate catalyst life and long-term operating effects.

#### III. SYSTEM RETROFIT

#### RETROFIT REQUIREMENTS

The retrofit requirements as previously illustrated in Figures 2 and 3 for the Huntington Beach Generating Station are not at all typical for SCE generating facilities in the Los Angeles basin. Most of the larger units have horizontal shaft air preheaters which are located closely behind the boiler economizer outlet. As illustrated in Figure 5, this means that for most units all equipment and ducting downstream of the economizer outlet, including the stack, will have to be relocated to make room for the reactor and the ammonia flue gas mixer. In many cases existing structures, equipment and other site restraints interfere with the required expansion of the backend of the boiler, and consequently dictate major site rearrangements.

#### OPERATING AND MAINTENANCE REQUIREMENTS

Table III tabulates the O&M requirements for all Los Angeles basin units involved, including major items such as electrical power consumption, system NH<sub>3</sub> consumption and the yearly new catalyst and used catalyst disposal requirements. Maintenance items such as the washing and cleaning of reactor, air preheater and reactor booster fan is not known and not included at this time.

#### PROBLEM AREAS

Since most Steam Generators require a major modification to the back-end of the unit in order to provide room for the installation of the reactor, retrofitting these units will require a long construction period and extended unit outages. In addition, the existing plants are already congested, which will add to this problem and create many construction restraints. Because of these problem areas and the existing unit overhaul schedule to work with, considerable risks will be encountered in meeting these schedules and supplying power to the consumers.

Some unknowns which may create further problems are catalyst availability and catalyst disposal after use. At present there are insufficient manufacturing facilities available for the new honeycomb DeNOx catalyst. The type of disposal required is not yet known nor the potential for reconditioning and reuse.

Perhaps the greatest risk to the system reliability waits at the completion of construction when startup and operation begin. Because of the booster fan required to overcome the pressure drop in the system, the larger units will be converted from forced draft units to a balanced draft operation. Controls to operate in this mode will be included and complete boiler implosion studies will be made to assess the need for boiler reinforcement. Mitigation of this risk is possible, elimination is not.

#### IV. COST

#### HUNTINGTON BEACH DEMONSTRATION UNIT

#### Capital Cost

Capital cost estimates have been prepared for the 107.5 MW Demonstration Project at Edison's Huntington Beach Generating Station. This estimate is provided for information, and caution should be exercised in using the figures for other applications. Since this is a demonstration project, some shortcuts have been assumed which would not be used in a commercial installation. In addition, due to the small scale of the demonstration, many of the modifications required to backfit an entire unit were not required.

The cost for this facility is estimated to be \$13,500,000 including all corporate overheads and AFUDC, but excluding testing cost. This is based on a late 1981 operating date and the costs are consistent with 1981 dollars.

#### O&M Cost

Estimates for operating labor and material for this project were obtained from Edison's Power Supply Department. The Engineering Department provided data for ammonia, energy and catalyst consumption which became the remaining components of O&M. Catalyst replacement was calculated based on the bids received for the project. These costs are shown in Table IV.

#### SYSTEM RETROFIT COST

#### Capital

Capital cost estimates for Catalytic DeNOx were prepared in May, 1978. The basis for these estimates follows:

Conceptual engineering was performed for providing catalytic DeNOx equipment at the following units:

Units	Unit Size (Each)
Alamitos 1 & 2	175 MW
Alamitos 3 & 4	320 MW
Alamitos 5 & 6	480 MW

The engineering consisted of the development of plot plans, flow diagrams, one-line diagrams and arrangement sketches. At the completion of this effort, a list of major equipment was prepared, along with engineering take-offs of bulk material quantities such as piping, cable, structural steel, and the equipment relocations and site modifications required. Using this information, cost estimates were prepared for the above units. This information was then used to estimate the cost for all other stations using \$/kW figures from the base estimates multiplied by a site factor to account for specific conditions at each station.

Consideration was given to the following criteria in arriving at the site factor for the units listed in Table II.

Space Availability
Unit Configuration
Size of Unit

The estimates were reviewed in December, 1979, to determine if there were any inconsistencies with information collected on the catalyst and the balance of plant for the Catalytic DeNOx Demonstrations Project. It was determined that no modifications to the total costs were required. The estimates were then escalated from 1978 to 1981. These capital costs are shown in Table V.

#### O&M Cost

The system O&M cost was based on MW/year from the Catalytic DeNOx Demonstration Project modified for the unit capacity factor.

For catalyst replacement, the cost per cubic foot of catalyst of the demonstration unit was used with the assumption that the catalysis reactor volume would be proportional to the flue gas flow to be treated.

The O&M as well as catalyst replacement costs are shown in Table V.

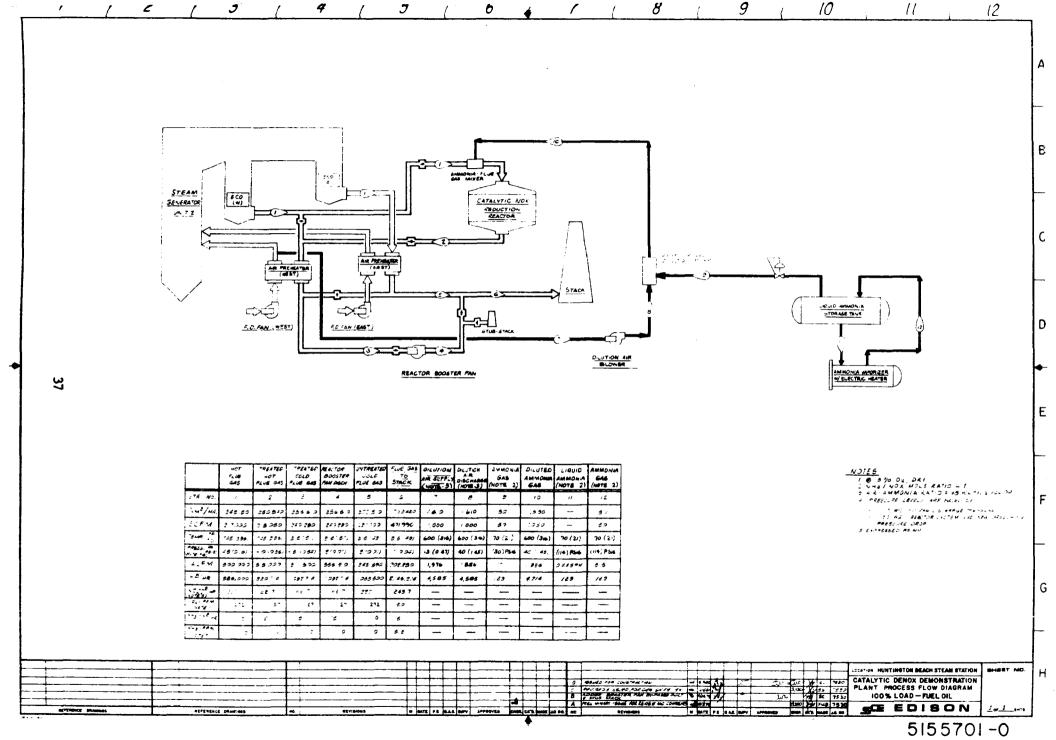
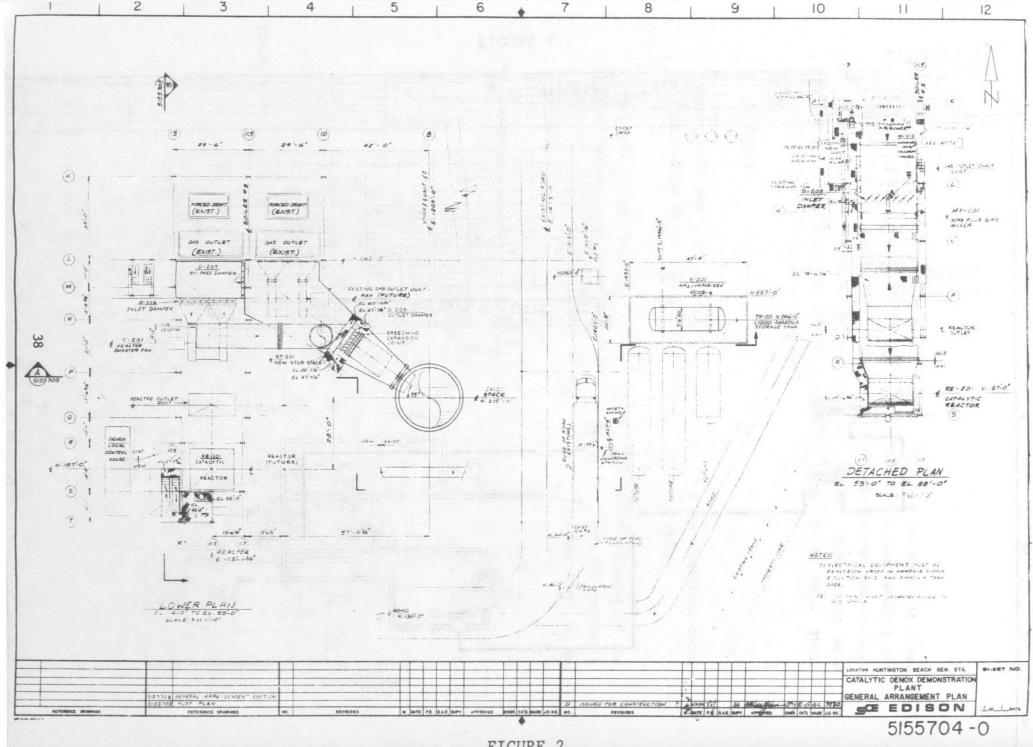


FIGURE 1



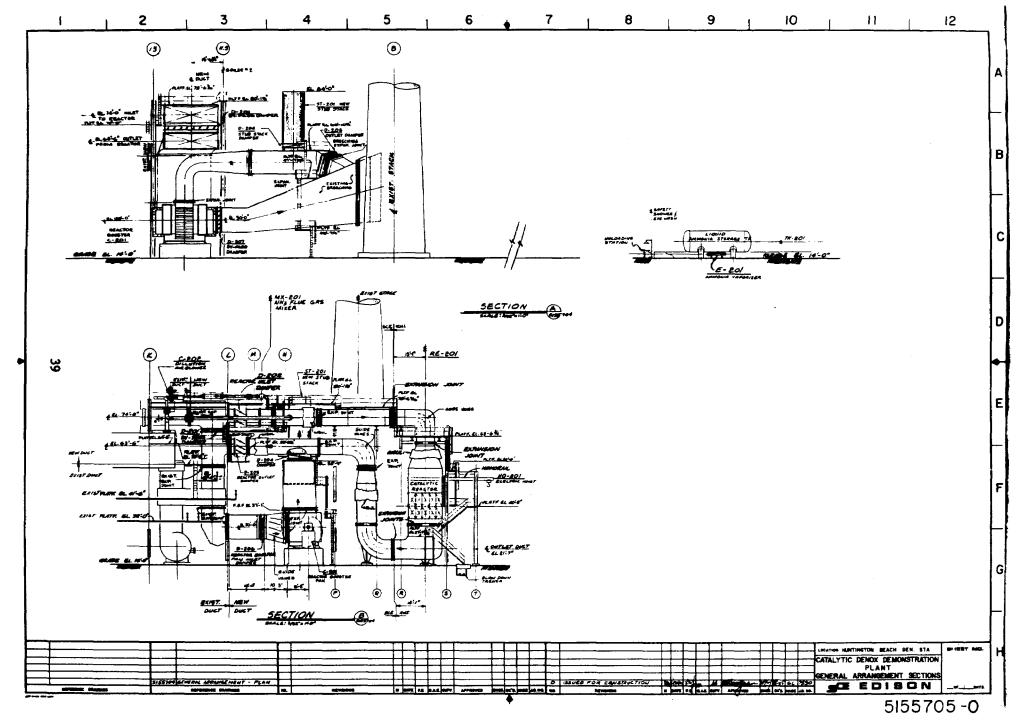
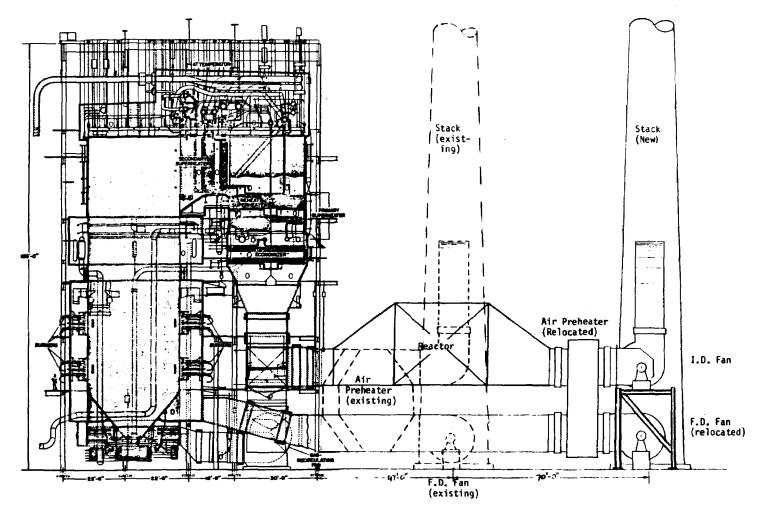


FIGURE 3

Huntington Beach 2 7-29-80

MAJOR ACTIVITIES FOR CATALYTIC DeNOx PROJECT

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SOUTHERN CALIFORNIA EDISON COMPANY ALAMITOS STEAM STATION - UNITS NO. 5 & 6 LOS ALAMITOS, CALIFORNIA

SHOWN RETROFITTED WITH SCR MOX REMOVAL REACTOR

## TABLE I

## HUNTINGTON BEACH UNIT 2 DATA

0	Electrical Output:	215 MW (107.5 MW for West half of unit)
0	Flue Gas Flow Rate:	100,000 to 500,000 ACFM at 750°F (half of total unit flow)
o	Flue Gas Temperature:	550° to 800°F (economizer outlet)
o	NOx Concentration:	180-280 vppm at 3% 0 <sub>2</sub> dry
o	SO <sub>2</sub> Concentration:	70-140 vppm at 11% and 4% $0_2$
o	Particulate Concentration:	.00502 gr/SCF
o	SO <sub>3</sub> Concentration:	1 - 5 vppm

#### TABLE II

#### MAJOR EQUIPMENT DATA

#### Catalytic Reactor:

- Size 16' (W) x 18' (L) x 23' (H)
- Space Velocity: 6655 hr<sup>-1</sup>
- Design Pressure: -17 to +15" WG
- Assembled Weight: 184,300 lbs
- Catalyst Shape: Square Honeycomb

#### Booster Fan:

- Type: centrifugal, air foil blading
- Operating characteristics: 371,800 CFM @ 17" WG SP.
- Flow control: inlet vanes
- Drive: 2000 HP Electric Motor

#### Ammonia Tank:

- Nominal Capacity: 12,000 gallon

#### Electrical Equipment:

- 4160 V Switchgear
- 480 V Circuit Breaker
- 480 V Motor Control Center

#### TABLE III

#### OPERATING AND MAINTENANCE DATA

Huntington Beach SCR Demonstration Unit (107.5 MW)

0	Power Requirement	1500 kW
	Power Consumption	7.5 x 10 <sup>6</sup> kWhr/yr
	NH <sub>2</sub> Consumption	320 tons/yr
	Catalyst Consumption*	20 tons/yr
	and Disposal	<u>-</u>

SCR NOx Removal Systems on all 175 MW and larger L.A. Basin Units (7720 MW)

o Power Requirements 83 MW
o Power Consumption 386 x 10<sup>6</sup> kWhr/yr
o NH<sub>3</sub> Consumption 21,500 tons/yr
o Catalyst Consumption\* 1,860 tons/yr
and Disposal

\*Based on two-year catalyst operating life

## TABLE IV

107.5 MW

## CATALYTIC DeNOx - DEMONSTRATION UNIT

## O&M COST (1981 \$)

Derate		\$	59,880
Energy			207,600
Chemicals			62,570
Labor and Supplies			134,250
Catalyst Replacement		<del></del>	986,740
Total O&M	,,·	\$1	,362,860
	SAY	\$1	,363,000

TABLE V

COST SUMMARY FOR CATALYTIC DeNOx REDUCTION TECHNIQUES

(\$  $\times 10^6$  all in 1981)

			Average (1)		Cat		
		Total Capacity (MW)	Capacity Factor(%)	Capital (\$)	0 & M (1-Year) (\$)	Catalyst (1-Year Life) (\$)	Replacement (3)
	Ormond 1 & 2	1500	55	220.0	7.2	10.1	5.1
	Redondo 7 & 8	960	59	190.0	4.9	6 • 4	3•3
	Alamitos 5 & 6	960	59	130.0	4.9	6 • 4	3.3
	Alamitos 3 & 4	640	53	80.0	3.0	4-1	2.1
	El Segundo 3 & 4	670	53	120.0	3.2	4.5	2.3
	Etiwanda 3 & 4	640	53	100.0	3.0	4.1	2.1
46	Huntington 3 & 4	440	47	60.0	2.0	2 • 8	1.4
	Huntington 1 & 2	430	47	60.0	1.8	2.7	1.4
	Mandalay 1 & 2	430	47	60.0	1.8	2.7	1.4
	Alamitos 1 & 2	350	40	50.0	1.4	2 • 4	1.2
	El Segundo 1 & 2	350	40	50.0	1.4	2 • 4	1.2
	Redondo 5 & 6	350	40	60.0	1.4	2.4	<u> 1.2</u>
	Total	7720		1180.0	36.0	51.0	26.0

NOTE: (1) Average capacity factor - Based on the latest information from System Planning - November 1979. These factors were used in the calculation of O&M.

<sup>(2)</sup> Catalytic DeNOx - The capital cost was based on the original estimate of May 1978. The O&M cost was based on the latest information available from the demonstration unit in Huntington Beach Station and also using the latest capacity factors available.

<sup>(3)</sup> Catalyst Replacement - Cost shown is an average cost for a 2-year life catalyst.

# COUNTERMEASURES FOR PROBLEMS IN NO $_{\mathbf{x}}$ REMOVAL PROCESS FOR COAL-FIRED BOILERS

By:

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#### ABSTRACT

The construction of many coal-fired power plants is being planned as a result of the recent petroleum shortage, and so the need for the De-NOx process for coal-fired boilers is increasing. However, there are many problems to be solved in the practical application of the De-NOx process for coal-fired boilers, because high concentrations of dust particles and SOx are contained in the flue gas. The major problems are: (1) catalyst bed pluggage and catalyst erosion by dust particles (2) the influence of unreacted NH3 and SO3 from the De-NOx reactor to the downstream equipment (3) deactivation of the catalyst by dust particles and SOx.

KHI has been working to solve these problems and to put the De-NOx process into practical use for coal-fired boilers with the co-operation of EPDC for many years. We have developed superior catalysts having several characteristics, which include long life, SOx resisting properties, dust resisting properties and low conversion of SO2 to SO3. Further, we have solved the above problems and have developed the most economical and stable De-NOx process.

This paper describes the problems involved in the practical application of the De-NOx process for coal-fired boilers and the countermeasures undertaken in KHI's De-NOx process.

#### INTRODUCTION

It was since 1973 that the regulation on NOx emission from fixed source was enforced in Japan. Many enterprises have been involeved in research and development of NOx removal technology. Kawasaki Heavy Industries Co., Ltd. (hereinafter referred to as "KHI") has been endeavoring to the NOx removal technology for many years, and has in particular engaged in the research and development with respect to the Selective Catalytic Reduction of NOx with NH3 since 1970. Finally, we have succeeded in the practical application of the DeNOx process for LNG-fired and heavy oil-fired boilers.

On the other hand, the practical application of the DeNOx process for coal-fired boilers is more difficult than for oil-fired boilers, because the flue gas of coal-fired boilers contains more dust particles and SOx as compared to that of oil-fired boilers. Since 1975, KHI has been cooperating with Electric Power Development Co., Ltd. (hereinafter referred to as EPDC) in the research of DeNOx process for coal-fired boilers. We have developed superior catalysts having several characteristics, which feature long life, SOx resisting properties, dust resisting properties and low conversion of SO2 to SO3. Furthermore, we have obtained the economical and stable DeNOx process. Consequently,KHI received an order for demonstration DeNOx plant for Takehara No.1 boiler of EPDC and are now doing our best to manufacture it. Furthermore, KHI received an order for the DeNOx pilot plant from Electric Power Research Institute in the U.S.A. and are now engaged in its operation.

In this paper, we describe the various problems involved in practical application of the DeNOx process for coal-fired boilers and countermeasures undertaken in KHI's DeNOx process.

#### VARIOUS PROBLEMS IN DeNOx PROCESS FOR COAL-FIRED BOILERS

Because of high concentration of dust, SOx and NOx in the flue gas out of the coal-fired boiler, the total system is required for flue gas treatment in order to resolve the environmental problems. The total flue gas treatment system is composed of dust collection, DeNOx and DeSOx. In order to improve the performance, reliability and economics of the total system, we must consider not only the performance and reliability of each component technic but also the most suitable combination of these components. Accordingly, the

performance and reliability of the DeNOx process have to be pursued considering its influence on the other process.

There are two kinds of DeNOx processes for coal-fired boilers, viz. low dust DeNOx process and high dust DeNOx process (cf. Figure 1). We have solved two problems in developing these DeNOx processes. The first problem is the influence of the unreacted NH3 and the generated  ${\rm SO}_3$  from the DeNOx reactor on the downstream equipment, and the second one is the influence of the high concentration of dust and  ${\rm SO}_3$  in the flue gas on the performance and stability of the DeNOx plant.

## Influence of the unreacted NH3 and the generated SO3 from the DeNOx reactor on downstream equipment

Influence of the unreacted NH3. In the low dust DeNOx process, the unreacted NH3 reacts with SO3 and forms ammonium bisulfate with the fall of the flue gas temperature at the air preheater. As the ammonium bisulfate is strongly corrosive and adhesive, it causes the corrosion or plugging of the air preheater. And as the unreacted NH3 is collected in the wet DeSOx process and accumulated in the absorbing liquid, there arises a need for removing NH3 in the waste water of the wet DeSOx process.

In the high dust DeSOx process, the unreacted NH<sub>3</sub> is fixed to the fly ash which is collected by the electrostatic precipitator. Figure 2 shows the relationship between the concentration of the unreacted NH<sub>3</sub> and the adsorbed ammonia on fly ash. The dotted line in the figure shows the calculated value assuming that all the unreacted NH<sub>3</sub> is adsorbed on the fly ash. According to the test results, most of the unreacted NH<sub>3</sub> less than 20ppm is proved to be adsorbed on the fly ash. Accordingly, the influence of the unreacted NH<sub>3</sub> on the air preheater and the wet DeSOx process in the high dust DeNOx process is less than in the low dust DeNOx process. But there are several problems such as leaching of NH<sub>3</sub> and offensive odor in utilizing the fly ash for cement admixing material or using it for reclamation.

As the unreacted NH3 will cause various sorts of troubles, it is necessary to decrease to the unreacted NH3 at least as low as 10ppm and preferrably below 5ppm (1).

Influence of SO3. The DeNOx catalyst generally works as shown in the formulas

(1) and (2). In addition to these NOx removal reactions, it works as  $SO_2$  oxidation catalyst as in the formula (3).

$$NO + NH_3 + 1/4 O_2 \longrightarrow N_2 + 3/2 H_2O$$
 (1)

$$NO_2 + 4/3 NH_3 - 7/6 N_2 + 2 H_2O$$
 (2)

$$SO_2 + 1/2 O_2 \longrightarrow SO_3$$
 (3)

In the low dust DeNOx process, SO $_3$  forms ammonium bisulfate through the reaction with the unreacted NH $_3$  and forms the sulfulic acid at low temperatures.

In the high dust DeNOx process, there is no influence of  $SO_3$  to the air preheater, because most of  $SO_3$  is adsorbed on the fly ash. But  $SO_3$  adsorbed on the fly ash accelerates the adsorption of  $NH_3$ .

As  $\mathrm{SO}_3$  from the DeNOx reactor will cause the above mentioned troubles, it is desirable to keep the oxidation of  $\mathrm{SO}_2$  as low as possible. It is necessary to decrease  $\mathrm{SO}_3$  below 10ppm in order to control influence of  $\mathrm{SO}_3$  to the minimum.

#### Influences on the performance and stability of DeNOx process

Influence of dust. Problems of the plugging of the catalyst bed and the erosion of the catalyst are perceived as the influence of the dust. The plugging of the catalyst bed will cause the increase of the pressure drop of the catalyst bed and the decrease of the NOx removal efficiency, whereas the erosion of the catalyst will cause the decrease of the NOx removal efficiency and the secondary environmental pollution by scattering the catalyst components. Accordingly, sufficient countermeasures are required against the plugging and the erosion caused by dust, especially in the high dust DeNOx process.

#### Influence caused by the particular properties of coal-fired flue gas.

Influences of SOx, halogen (HCl,HF), and alkali metals in dust, especially pottassium, on the catalytic performances may be anticipated. The decrease of the catalytic performance must be especially avoid, because it causes not only the lack of stability of the system but also the increase of running cost. Accordingly, it is necessary to develop catalysts which can resist against the particular properties of coal-fired flue gas.

Influence of the long-term low load operation and the repeat of star-up/shut-down. The long-term low load operation of boilers may result in the deactivation of the catalyst and the plugging of the catalyst bed due to the deposit of NH<sub>3</sub>, SOx and dust in accordance with the decrease of the gas temperature and the gas flow rate.

Repeat of start-up/shut-down of boilers may result in the catalytic deactivation due to the condensation of SOx and H<sub>2</sub>O on the catalyst surface and due to masking of the catalyst surface with dust.

It is necessary to perform the simulation tests of the long-term low load operation and the repeat of start-up/shut-down.

COUNTERMEASURES ADOPTED IN KHI's DeNOx PROCESS

### Control of the unreacted NH3 and the oxidation of SO2

Control of the unreacted NH3. The NH $_3$ /NOx mole ratio should be controlled less than 1 in order to control the unreacted NH $_3$  emission. For example, it is necessary to control the NH $_3$  injection whithin the region of NH $_3$ /NOx mole ratio between 0.83 and 0.93 in order to obtain the NOx removal efficiency more than 80% and to decrease unreacted NH $_3$  less than 5ppm (cf. Figure 3.).

For this purpose, it is required to control the NH<sub>3</sub> injection in accordance with the change of the volume of the treatment gas and that of the inlet NOx concentration. KHI developed an unique control system by combining the following two methods. One is a feed-forward control of the NH<sub>3</sub> injection, of which the amount is calculated by the boiler load and the inlet NOx concentration. The other is a feed-back control, in which the amount of NH<sub>3</sub> injection is corrected by the outlet NOx concentration or the NOx removal efficiency. The follow-up test results of our control system for boiler load variation was satisfactory. As shown in Figure 4, the unreacted NH<sub>3</sub> is controlled within 5ppm in spite of the change of the gas volume, the gas temperature as well as the inlet NOx concentration.

In large DeNOx plants, the mixing and dispersing of injected NH<sub>3</sub> are very important in order to decrease the unreacted NH<sub>3</sub>. We carried out cold-model tests as to the mixing and dispersing, and have found the most efficient

configulation. The length of the duct from the nozzle to the reactor is short and the pressure drop of the duct is quite small in this configulation.

Control of  $SO_2$  oxidation. Characteristic of a conventional catalyst are shown in Figure 5. The Oxidation rate of  $SO_2$  will increase as the NH<sub>3</sub>/NOx mole ratio and the space velocity decrease. Therefore, the development of catalysts with low oxidation rate of  $SO_2$  is indispensable for the simultaneous control of the unreacted NH<sub>3</sub> and the generated  $SO_3$  emission. KHI has endeavored to improve the catalysts in cooperation with the catalyst manufacturer.

We started with the  $V_2O_5-TiO_2$  basis catalyst, which has various features such as high NOx reduction activity and long life, etc.. Our points of the improvement are as follows :

- 1. Decrease of the amount of the active element (  $V_2O_5$  )
- 2. Addition of an active element as promoter
- 3. Improvement of the dispersion of the active elements

We succeeded in developing the catalysts with low oxidation rate of  $SO_2$  and high NOx reduction activity. Figure 6 shows the performance of the two improved catalysts. KHI has several catalyst with low oxidation rate of  $SO_2$  avilable, and select the most suitable catalyst for practical use in accordance with gas temperature and SOx concentration. Figure 7 shows the performance of the improved catalyst (K-llO) for coal-fired boiler load conditions. Thus it is possible to control  $SO_3$  emission from the DeNOx reactor below 10ppm by employing the improved catalysts with low oxidation rate of  $SO_2$ .

Treatment of the ash containing  $NH_3$ . As already mentioned,  $NH_3$  is contained in the fly ash collected by the electrostatic precipitator in the high dust DeNOx process. The ash containing  $NH_3$  being dumped for reclamation, there might arise problems of  $NH_3$  odor and leaching of  $NH_3$ . We carried out odor tests and leaching tests, and confirmed that it is necessary to make the amount of  $NH_3$  on the fly ash below  $20 \, \text{ppm}$  ( mg/kg ) for reclamation. We have been developing the  $NH_3$  removal technology out of the fly ash in cooperation with EPDC. Our  $NH_3$  removal process is a pyrolysis process by utilizing a part of flue gas of the boiler. We have carried out the bench scale tests and have confidence in the practical application of this process.

#### Means of stabilizing the catalystic performance

Countermeasures against dust. As already mentioned, there are two problems due to the dust, viz. the plugging of the catalyst bed and the erosion of the catalyst. Against the plugging of the catalyst bed, we have been employing fixed bed reactor with dust free type catalysts for many years. Through the pilot tests for the dust free type catalysts in high dust and low dust flue gases, we have found the most suitable catalyst shape, catalyst dimensions and gas flow rate for respective flue gas. As for the shape of the catalyst, pipe or honeycomb shape is most preferable, but for the high dust gas wider opening is required than for the low dust gas.

The erosion of the catalyst usually starts at the inlet part of the catalyst and gradually progresses (2). The countermeasure we adopted is hardening the inlet part of the catalyst as long as 1/100 to 1/50 of its length by the impregnant, of which effectiveness was confirmed by endurance tests. Our catalysts are made so as to contain in the active elements evenly into the inside, and so the performance of the catalysts will be maintained in spite of the progress of the erosion. Figure 8 shows the change of the catalyst erosion rate with time for a conventional catalyst and our improved catalyst. The inlet hardening could decrease the amount of erosion of about 6% per year to less than 1%.

Long-term stability of the catalytic performence. KHI's catalysts have various features such as high activity and long life. In our previous report, we had confirmed that the lifetime of the catalyst for the flue gas of the heavy oil-fired boilers was more than two years (2). For coal-fired boilers, we carried out the durability test at the pilot plants adopted to Takehara No.1 boiler of EPDC and could confirm that the lifetime of the catalyst was more than 8,000 hours. As shown in Figure 9, no change was noticed of NOx removal efficiency as well as the pressure drop due to the catalyst bed. For the improved catalyst with low oxidation rate of SO<sub>2</sub>, the durability test has been continued for more than 3,000 hours. According to Figure 10, no change is noticed of the NOx removal efficiency and of the oxidation rate of SO<sub>2</sub>. As a result, it was confirmed that our catalyst has a stable performance against SOx, halogen ( HCl, HF ) in the flue gas and potassium in

the dust.

In order to investigate the influence of the long-term low load operation and the repeat of start-up/shut-down of the boiler, we conducted simulation tests. As for the long-term low load operation, we conducted cyclic operations between a quarter and a half of the full load for about one month. It was found that the catalytic activity could be recovered by the full load operation at  $350^{\circ}$ C, although it was temporarily lowered by the low load operation at  $270^{\circ}$ C (cf. Figure 11). This temporary deactivation phenomenon is consider to be caused by the increase of  $50^{2-}_4$  and  $50^{0-1}_4$  is removed from the catalyst with the recovery of reaction temperature, and the catalytic activity is recovered. On the other hand, we repeated emergency stop and start-up five times. No deactivation of the catalyst was caused by such operations like as emergency stop and start-up (cf. Figure 11).

#### KHI's DeNOx PROCESS FOR COAL-FIRED BOILERS

The KHI's newly developed DeNOx process for coal-fired boilers has the following features :

- 1. It is possible to suppress the unreacted  $NH_3$  below 5ppm by means of our highly active catalysts, pertinent control of  $NH_3$  injection and uniform dispersion of  $NH_3$ .
- 2. It is possible to control the  $SO_3$  from the DeNOx reactor below 10ppm by employing the catalysts of low oxidation rate of  $SO_2$ .
- No plugging of the catalyst bed occurs by employing such dust free type catalyst as pipe-shaped or honeycomb-shaped.
- 4. The erosion of the catalyst will hardly occur even in the high dust DeNOx process by employing the hardening treatment of the inlet part.
- 5. Our catalysts are hardly subject to the influence of S0x, halogen in the flue gas and potassium in the dust .

In addition to this DeNOx process, KHI has the established technology of dust collection and wet DeSOx, and has been proceeding with the development of the comprehensive flue gas treatment system for coal-fired boilers. The flow

diagram of the toatl flue gas treatment systems and an example of material balance for a 500 MW coal-fired boiler are shown in Figures 12 and 13. As shown in these figures, it can be seen that the unreacted NH<sub>3</sub> is suppressed to 5ppm and the generated SO<sub>3</sub> is to 8ppm with the NOx removal efficiency as high as 90%. And it can be seen that unreacted NH<sub>3</sub> and generated SO<sub>3</sub> are all collected by the electrostatic precipitator in the high dust DeNOx process, and are partially collected by the wet DeSOx process in the low dust DeNOx process.

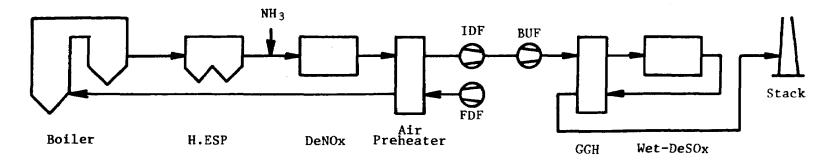
#### SUMMARY

Hereto described is the progress to date of the KHI's DeNOx process with many outstanding features for coal-fired boilers. KHI has almost accomplished the total system for flue gas treatment utiliziling this process. However, further study tasks are continuing for improvement of reliability and economics of the total systems.

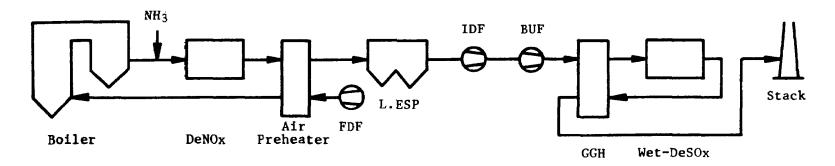
Finally, we wish to express our sincere appreciation to Mr. Nakabayashi, Mr. Shimizu and Mr. Mohri of Electric Power Development Co., Ltd..

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- Y. Nakabayashi, "Research and Development on NOx Removal in Japan and Results of EPDC's Research and Development of the DeNOx Process", EPRI FP-1109-SR Seminar Proceedings Special Report, pp18-1 - 18-25, July 1979
- 2. Y. Nakabayashi and S. Niwa, "Characteristics of Cylindrical De-NOx Catalysts for a Coal-fired Boiler", EPRI FP-1109-SR Seminar Proceedings Special Report, pp21-1 21-13, July 1979



## Low Dust DeNOx Process



## High Dust DeNOx Process

Figure 1. Total Flue Gas Treatment Process for Coal-Fired Boilers

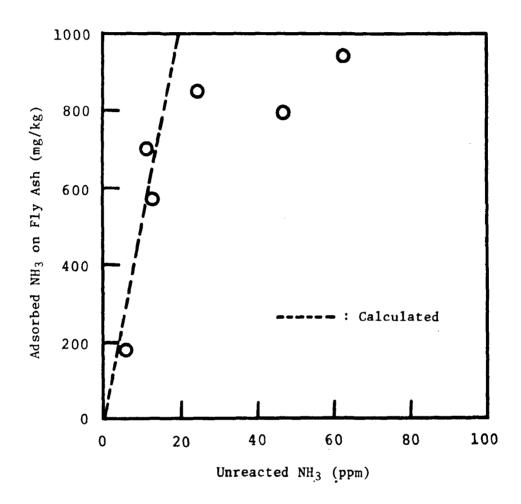


Figure 2. Adsorption of Unreacted  $\mathrm{NH}_3$  on Fly Ash

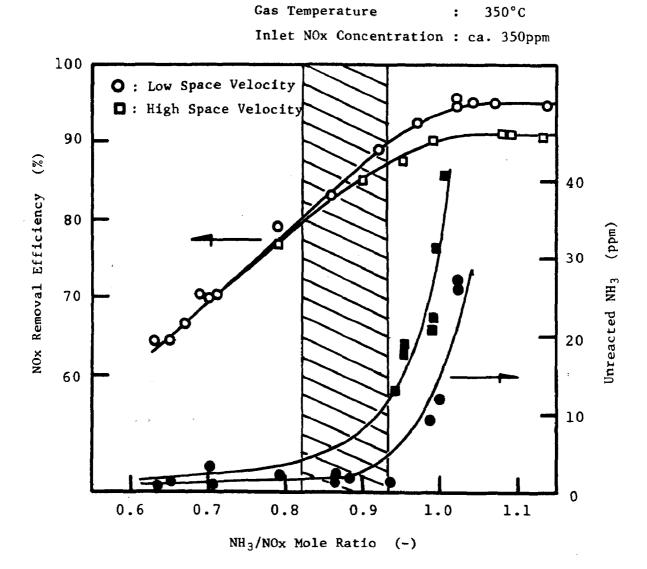


Figure 3. Control Region of NH<sub>3</sub>/NOx Mole Ratio

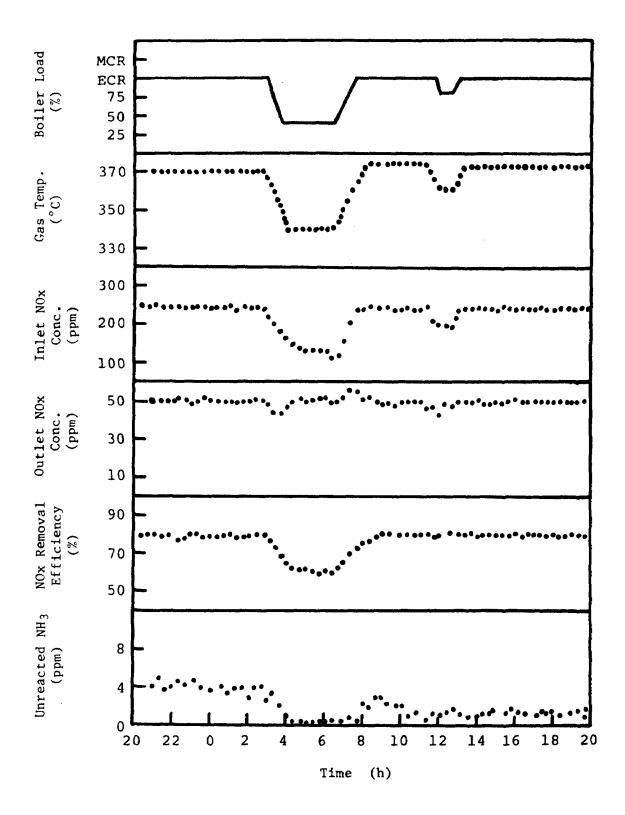


Figure 4. Follow-up Test for Boiler-Load Variation

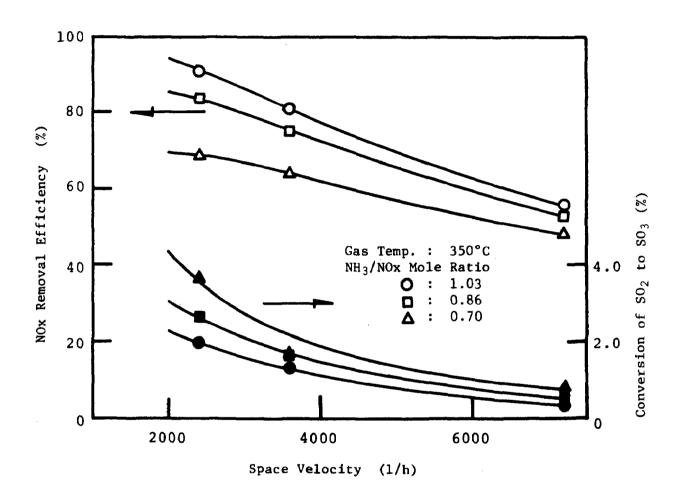


Figure 5. Characteristic of a Conventional Catalyst

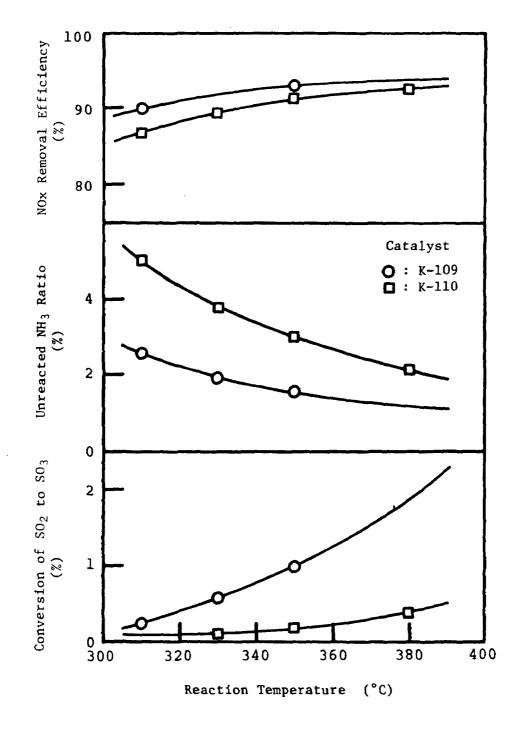


Figure 6. Performance of Improved Catalysts

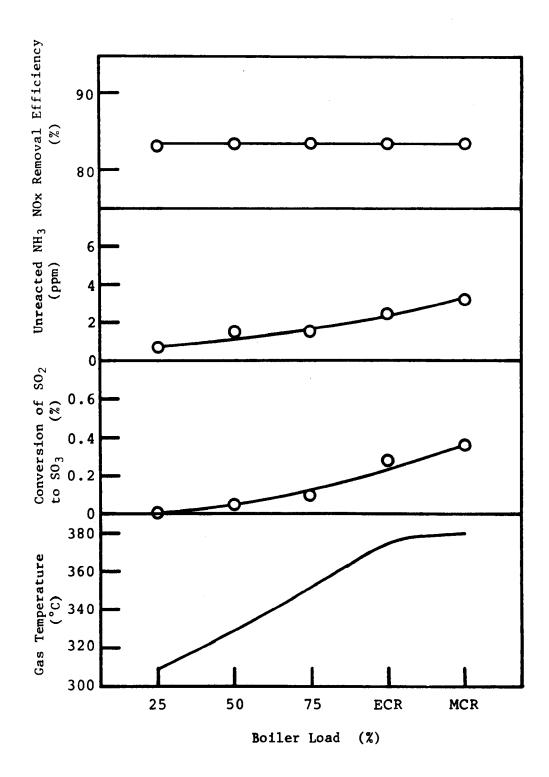


Figure 7. Performance of Improved Catalyst (K-110) for Boiler Load Conditions

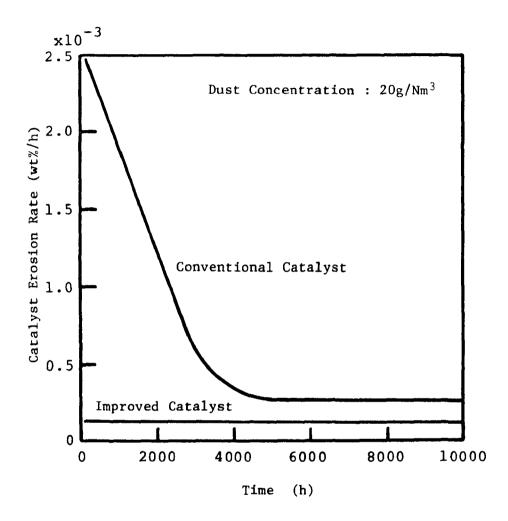


Figure 8. Change of Catalyst Erosion Rate with Time

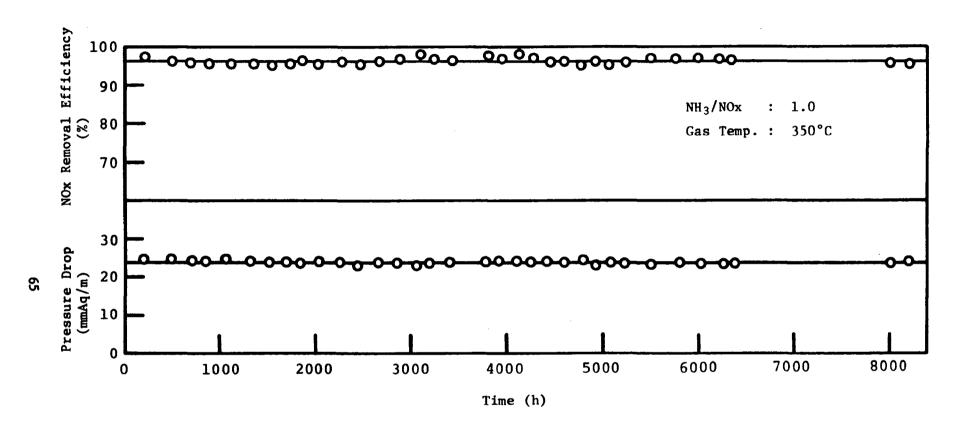


Figure 9. Durability Test of Pipe-Shaped Catalyst for the Economizer Outlet Gas of the Coal-Fired Boiler

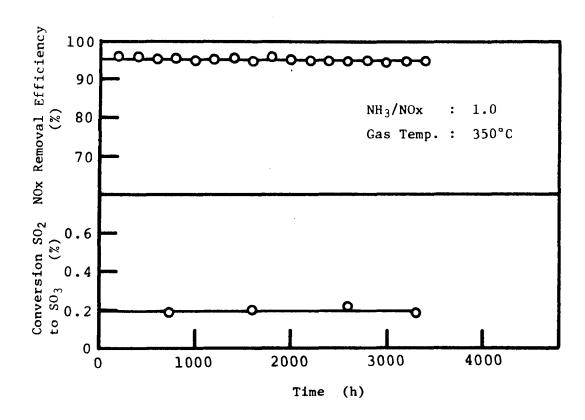
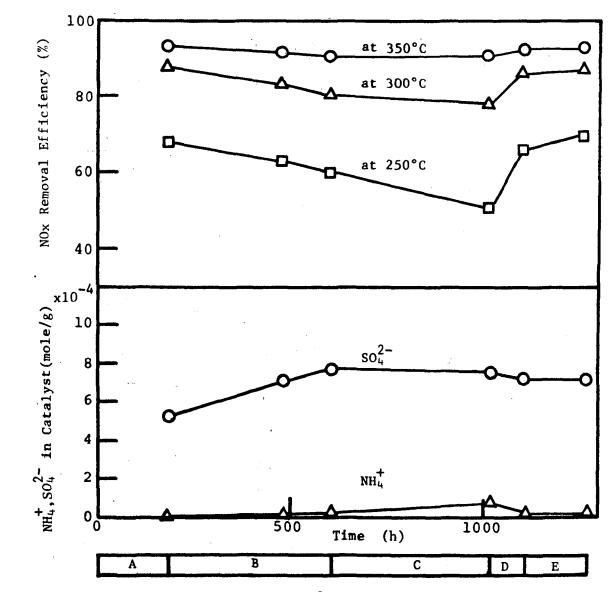


Figure 10. Durability Test of Improved Catalyst for the Economizer Outlet Gas of the Coal-Fired Boiler



A : Operation at 350°C

B : Cyclic Operation at 300°C and 270°C

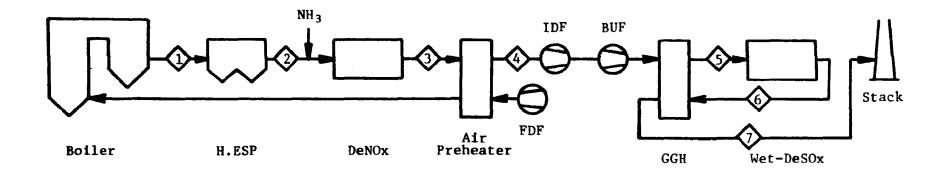
Without NH3 at 270°C

C : Cyclic Operation at 300°C and 270°C

D : Operation at 350°C

E : Cyclic Operation of Start-up and Shut-down

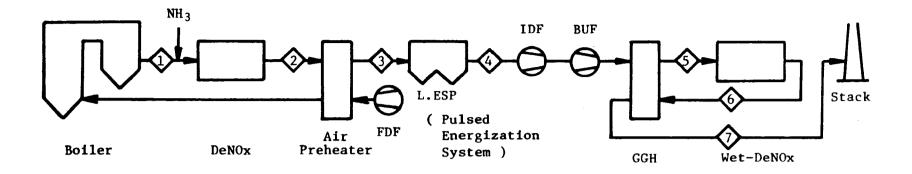
Figure 11. Change of Catalitic Activity and  $NH_4^+$ ,  $SO_4^{2-}$  in Catalyst with Low Load Operation and Start-up/Shut-down Operation



		<b>1</b>	2	<b>③</b>	<b>③</b>	<b>③</b>	<b>©</b>	♦ .
Gas Volume	Nm³/h (Wet Bas <b>is</b> )	1700000	1714000	1723600	1861500	1913600	1967400	1915300
Gas Temperature	°C	380	380	380	150	86	45	106
Conc. of Fly Ash	mg/Nm <sup>3</sup> (Dry Basis)	20000	30	30	30	30	20	20
Conc. of SOx	ppm (Dry Basis)	1000	990	985	910	889	50	50
Conc. of NOx	ppm (Dry Basis)	300	300	30	30	30	30	30
Conc. of NH <sub>3</sub>	ppm (Dry Basis)			5	5*	5*	4*	4*
Conc. of SO <sub>3</sub>	ppm (Dry Basis)	7	7	15	5 <b>*</b> 10	5 <b>*</b> 10	4 <b>*</b> 8	4* 8

\* : Reduced Value of NH4HSO4 to NH3 or SO3

Figure 12. Flow Diagram of Total Flue Gas Treatment Process and Example of Material Balance for a 500 MW Coal-Fired Boiler (Low Dust DeNOx Process)



		<b>�</b>	<b>②</b>	<b>③</b>	<b>\$</b>	<b>\$</b> >	<b>©</b>	$\Diamond$
Gas Volume	Nm <sup>3</sup> /h (Wet Basis)	1700000	1709600	1846400	1849700	1901500	1953600	1901800
Gas Temperature	°C	380	380	150	150	86	45	106
Conc. of Fly Ash	mg/Nm³ (Dry Basis)	20000	19900	18400	10	10	10	10
Conc. of SOx	ppm (Dry Basis)	1000	990	920	920	895	50	50
Conc. of NOx	ppm (Dry Basis)	300	300	30	30	30	30	30
Conc. of NH <sub>3</sub>	ppm (Dry Basis)		5	5 <sup>*</sup>	Trace	Trace	Trace	Trace
Conc. of SO <sub>3</sub>	ppm (Dry Basis)	7	15	5 <b>*</b> 10	Trace	Trace	Trace	Trace

\* : Reduced Value of  $NH_4HSO_4$ to  $NH_3$  or  $SO_3$ 

Figure 13. Flow Diagram of Total Flue Gas Treatment Process and Example of Material Balance for a 500 MW Coal-Fired Boiler (High Dust DeNOx Process)

### THE HITACHI ZOSEN NO $_{\mathbf{X}}$ REMOVAL PROCESS APPLIED TO COAL-FIRED BOILERS

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#### ABSTRACT

Hitachi Zosen is a leading supplier of flue gas treatment systems for the removal of nitrogen oxides. They have nine commerical plants in operation. Early in 1978 Chemico Air Pollution Control Corp. whollyowned subsidiary of Envirotech Corporation acquired the North American license for the Hitachi Zosen technology, a dry process with selective catalytic reduction using ammonia. Because many sources of flue gas which require nitrogen oxide removal also contain a high level of dust, Hitachi Zosen has expended a considerable effort in developing a catalyst bed which can operate without plugging even though the gas contains particulates. Pilot plant tests using a specially designed metallic catalyst have been successfully operated on very dirty gases from steel sinter operations, and coal-fired boilers. An extensive pilot plant program has been in operation for over a year at Georgia Power Company's Plant Mitchell in Albany, Georgia. This is a 0.5 MW equivalent demostration plant for coal-fired denitrification which is sponsored by U.S. EPA. Hitachi Zosen and Envirotech/Chemico believe that this testing has shown the effectiveness of the process over extended operating periods and the soundness of the control system and basic design.

Eventually a workable system will be evolved which can supply a continuous record of ammonia slippage.

#### STARTUPS AND SHUTDOWNS

Hitachi Zosen experience indicates that no special precautions are needed during starting up and shutting down of the Hitachi Zosen NO<sub>X</sub> removal system in coal-fired units except for the avoidance of fly ash accumulations. Ammonia injection should of course be stopped whenever the temperature drops below about 300°C to prevent ammonium sulfate/bisulfate formation on the catalyst. The reactor can follow the normal temperature changes along with the boiler with no difficulties. The catalyst has been tested through hundreds of temperature cycles without deterioration. If SO<sub>3</sub> condenses on the catalyst during shutdowns or startups, this produces no adverse effects and the SO<sub>3</sub> will evaporate when the temperature is raised to normal levels. The SO<sub>3</sub> condensate will not corrode the catalyst or reduce its activity.

Fly ash accumulations on the catalyst can be avoided by careful design of the ductwork to ensure that proper flow distribution is maintained at most flow rates. Soot blowing is an additional measure to avoid fly ash deposits.

#### INTRODUCTION

Hitachi Zosen began the development of selective catalysts for  $\mathrm{NO}_{\mathrm{X}}$  removal in late 1969 with basic research and laboratory testing. These were carrier-supported catalysts. By 1974, after extensive pilot plant studies, the catalysts were considered to be commercial. The Green Chemical Company, a wholly-owned subsidiary company, was then formed in June 1974 to manufacture these catalysts and started operation in 1975.

In early 1974 a contract was signed between the Idemitsu Kosan Company, LTD, one of the world's leading petroleum refineries in Japan, and Hitachi Zosen for the construction of an  $NO_X$  removal plant with a capacity of treating 350,000 NM $^3/H$  (218,000 SCFM) of flue gases. The plant was the first of its kind and size in the world. Construction work commenced at the Chiba Refinery in May, 1975 with test operation in November the same year.  $NO_X$  removal efficiencies of 95% were demonstrated.

Following the successful start-up of the Idemitsu-Kosan system, other plants soon went into operation including a 274,000 SCFM unit at a petrochemical plant, and two plants at steel manufacturing facilities. All of these have operated successfully. (See Table 1 for a listing of commercial plants built by Hitachi Zosen).

A particularly effective physical design of the catalyst structure has been developed by Hitachi Zosen. This structure is of a metallic corrugated shape. Figure 1 illustrates this design. The key features of the catalyst are:

- (1) All-alloy thin metal plate or wire mesh corrugated structure.
- (2) Due to substantially reduced pressure drop across the catalyst layer, operating power costs are much lower than with conventional catalysts.
- (3) A straight gas flow path prevents dust clogging.
- (4) It is applicable for gases with high  $SO_x$  concentrations and high dust loadings.

One of the primary applications for Hitachi-Zosen's corrugated NOXNON catalyst is the treatment of high temperature gases with high  $\mathrm{SO}_{\mathrm{X}}$  and high dust concentration such as coal fired boiler flue gases. Due to the catalyst's non-clogging feature, the  $\mathrm{NO}_{\mathrm{X}}$  removal system reactor can be installed directly behind the economizer. Thus, expensive flue gas pretreatment for dust removal is not required.

Development work by Hitachi Zosen has been conducted at various bench-scale and prototype pilot plants to ensure the successful application of NO<sub>X</sub> removal process technology for the treatment of dirty gases exhausted from power plants (coal-fired and high sulfur-containing heavy oil or residual oil fired), iron ore sintering plants, cement kilns, and other similar sources.

One of the most important of the pilot plant operations continues at the Electric Power Development Corporation (EPDC) power plant at the Isogo Station in Yokohama, Japan. This is a collaborative effort between EPDC and Hitachi Zosen under subsidy of the Japanese government. The excellent cooperation and contributions of the personnel of EPDC have resulted in the collection of a great deal of valuable data. Important developments, particularly in methods of gas distribution, have been achieved. There are three reactors installed, each treating about 200 NM<sup>3</sup>/Hr of gas, plus several smaller reactors for abrasion

testing. These reactors have operated essentially continually for several years.

NOXNON 600 catalyst was tested at this facility and some of the runs were over 6000 hours and were halted only because of the shutdown of the boiler for scheduled maintenance. As an example, one test was run for about 6300 hours at the end of which the targeted removal of 80% was still being attained. Ammonia slippage was very low as was the conversion of  $SO_2$  to  $SO_3$ . These tests have established the effectiveness of NOXNON 600 catalyst, its resistance to abrasion, its long-term reactivity, and the low tendency to convert  $SO_2$  to  $SO_3$ .

In addition to the work at EPDC, the pilot plant test facility for EPA, located in Albany, Georgia, has been operated for over a year. The results to date of this test program form the basis for this paper.

#### PROCESS DESCRIPTION

The Hitachi Zosen  $NO_X$  Removal Process is a selective catalytic reduction process in which nitrogen oxides in the boiler flue gas are reduced using ammonia as the reducing agent. The reduction reaction occurs in the presence of a proprietary catalyst.

Flue gas leaves the boiler economizer (Figure 2) at a temperature of  $665^{\circ}-745^{\circ}$  (normal temperature range), depending on the boiler load, and is diverted from the air preheater by means of dampers into the ductwork leading to the reactor.

Ammonia vapor, diluted with air or steam, is injected into the flue gas in this ductwork in approximately equimolar ratio to the desired  $\mathrm{NO}_{\mathrm{X}}$  removal requirements. A pictorial representation of the ammonia injection system is shown in Figure 3. Careful attention is given to the design of the ammonia injection system to produce uniform mixing throughout the bulk of the flue gas.

The flue gas, containing the ammonia, enters the top of the NO<sub>X</sub> reactor where it passes through a grid system which insures uniform gas distribution through the fixed catalyst bed, located below the grids. No dust removal is necessary prior to catalyst contact because of the non clogging design of the catalyst geometry. Nitrogen oxides are reduced by ammonia to ordinary nitrogen and water vapor over the catalyst surface. The treated gas then resumes the flow through the present boiler exit train, i.e., air preheater, and stack. Soot blowers are provided in the reactor for high dust laden gases to periodically clean the catalyst bed of any dust accumulation (Figure 4).

#### CATALYST CHARACTERISTICS

The gases to be treated flow in parallel to the plates of active catalyst material in Hitachi Zosen's corrugated catalysts, (see Figure 1). The catalyst bed consists of alternate rows of corrugated and flat plates. The distance between the flat plates is termed the pitch and this is typically between 6 and 16 millimeters. The pitch selection will depend upon the application and in particular the concentration of particulates in the gas to be treated. A larger pitch will have less tendency to retain accumulations of particulates but the larger pitch increases the required volume of catalyst.

#### CATALYST PREPARATION

The catalyst plates are arranged in a steel frame box supported by retainers. A standard module is 1.0 meter long, 1.0 meter wide, and 0.5 meter deep.

The activation of the catalyst follows after the corrugated catalyst assembly is made. In the NOXNON 500 catalyst, thin stainless steel plates are used. The surface is first converted to an aluminum alloy which is then treated with an aluminum dissolving solution rendering the surface layer porous. The steel surface may then be immersed in a solution containing active components which adhere to the porous surface layer. A permanent bond is obtained by further proprietary treatment.

A newer development is NOXNON 600 catalyst which, instead of plates, uses a metallic mesh material which is then coated with a porous carrier in which the active components are supported. The NOXNON 600 is considerably lighter in weight and contains more active material for a given volume of catalyst.

The active components of the catalyst consist of vanadium and titanium compounds. Other components are added to increase resistance to fly ash abrasion.

#### CATALYST ACTIVITY MEASURE

Catalyst activity is calculated by the following equation and is independent of  ${\rm NO}_{\rm x}$  concentration, and  ${\rm SO}_2$  or  ${\rm SO}_3$  concentrations.

$$- ln (1-X) = K (A/F)$$

where X = fraction removal of NO,

A = surface area of catalyst

F = flue gas volumetric flow rate

K = apparent reaction rate coefficient

#### MEASURE OF GAS VELOCITY

The concept of space velocity is normally applied to catalysts which are granular, cylindrical, ring, extrudate, and so on. However, the concept of space velocity is not useful in designing for the use of corrugated catalysts because of this unique structure. Instead of space velocity, Hitachi Zosen uses area velocity (A.V.), which is defined as flue gas volume flow rate per unit of apparent catalyst surface:

$$(M^3/M^2-Hr.)$$

The particular A.V. used in the Hitachi Zosen design depends upon the percent removal of  $\mathrm{NO}_{\mathbf{X}}$  required, the gas temperature, and other factors. However, the velocities are relatively high.

#### **ABRASION**

The catalyst can be abraded by the action of fly ash. Over a period of time the continuous abrasive blasting of the catalyst surface by the high velocity, dust laden flue gas will wear away the active catalyst material. The degree of abrasion or ash-cut has been studied by Hitachi Zosen and is estimated by the following equation:

$$A = a (c) (v^2) (t)$$

where A = decrease of catalytic activated layer by ash-cut

a = constant

c = concentration of fly ash

v = superficial linear velocity of gas across catalyst
 surface

t = elapsed operating time

A great deal of effort has gone into increasing the time that the catalyst can be used in contact with particulate laden flue gas. Special additives have been developed which are incorporated into the catalyst to increase its abrasive resistance. Precautions are taken to ensure good gas distribution so as to reduce areas of high gas velocity. These steps have been successful in allowing operations of a year or more at pilot operations in Japan where dust laden flue gases have been tested. Hitachi Zosen catalyst life of two years or more with typical coal-fired boiler conditions are obtainable.

#### CATALYST REGENERATION

The catalyst life is expected to be at least two years.

Regeneration of the catalyst is not needed during the planned catalyst life period. At the end of this period the old catalyst would be scrapped and new catalyst installed.

#### CHEMISTRY OF PROCESS

The following reactions are postulated:

The first two reactions are probably those that occur within the reactor. As we have experienced only very low levels of  $NO_2$ , the reactions (3) and (4) are not of significance. Reaction (5) will only begin to occur at very high temperatures of  $850^{\circ}F$  or higher.

The reaction of NH<sub>3</sub> and NO is probably a combination of the equations (1) and (2). It has been found by many investigators that one mole of ammonia will react with more than one mole of NO. At the Georgia Pilot Plant it appears that the ratio is around 8:9. That is, eight moles of ammonia will react with about nine moles of NO. This ratio holds up to about 80-85% removal of NO.

The efficiency will drop off as increased removals are sought and at a 90% removal the ratio is close to 1.0.

#### IMPORTANT VARIABLES

The most critical variables which affect the degree of  ${\rm NO}_{_{\rm X}}$  removal are the mole ratio of ammonia to  ${\rm NO}_{_{\rm X}}$ , the flue gas flow rate, and the reactor temperature.

#### MOLE RATIO

This variable is defined as the ratio of moles of ammonia fed into the reactor to the moles of  $\mathrm{NO}_{\mathrm{X}}$  in the flue gas to be treated. Typically a ratio of 0.9 to 1.0 is required for a  $\mathrm{NO}_{\mathrm{X}}$  removal of 90% or higher. Figure 5 shows the mole ratio curve prepared from data collected at the Georgia Power pilot plant with NOXNON 600. It illustrates that the ammonia will.remove somewhat more than one mole of  $\mathrm{NO}_{\mathrm{X}}$  per mole of ammonia feed up to a mole ratio of 0.7. Above this the removal becomes less efficient and the removal of more than 90%  $\mathrm{NO}_{\mathrm{X}}$  requires a lot more ammonia for the final quantities of  $\mathrm{NO}_{\mathrm{Y}}$  to be reacted.

This figure also shows the slippage of ammonia which becomes appreciable at high mole ratios. At a 90% removal of  $NO_X$  the slippage is still relatively low, below 10 PPM.

#### FLUE GAS FLOW RATE

Increased flow rates provide decreased contact time with the catalyst and consequent lower removal efficiencies. However, at the normal range of operations the changes in removal are not too obvious at varying flow rates. Figure 7 shows results of tests at the Georgia pilot plant over a range of velocities which indicate a general tendency toward lower removals at the higher flow rates but the data is mostly scattered in a narrow band. Figure 5 shows Mole Ratio vs. Removal data for two flow rates and the results are very close.

Tests at a wider range of flows performed in Japan (Figure 8) indicate a definite drop off of efficiency at very high flow rates. Very high flow rates are not recommended because of increased pressure drops. Figure 6 shows the pressure drops found at the Georgia pilot plant over the test conditions. These flows would probably be typical of commercial operations.

#### TEMPERATURE

The flue gas temperature in the reactor is an important variable. The effects are shown in Figures 8, 9 and 10. Normally the reactor temperature is held above 330°C (620°F) to avoid the possibility of ammonium sulfate/bisulfate formation and the reactivity of the catalyst is very high at this level. At area velocities of about 10 (Figure 8) temperatures above 300°C have little effect on the percent NO conversion. In effect, the normal expected operating temperature ranges (above 300°C) have essentially no influence over the removal efficiency.

#### EFFECTS OF SULFUR OXIDES

The NOXNON 500 and 600 catalysts have been developed to be able to withstand high levels of sulfur oxides. They have been run for several thousand continuous hours of testing in contact with flue gases containing 400 to 500 PPM SO<sub>2</sub> with no measureable decrease in activity due to the sulfur oxides.

Data from Japan provides indication of the fact that there is no effect of  $SO_2$  concentrations on the removal of  $NO_X$ . These tests were run with NOXNON 600 catalyst at 10 M<sup>3</sup>/M<sup>2</sup>-Hr., with a gas composition of 500 PPM  $NO_X$  and a mole ratio of 1.0.

so,	0,	% NO Conversion						
(PPM)	(%)	200°C 250°C 300°C 350°C						
0	10	55 73 86 94						
250	10	54 73 85 93						

For  $\mathrm{SO}_3$ , promoted catalyst life tests with concentrated  $\mathrm{SO}_3$  have been used to obtain data. Even with contact with  $\mathrm{SO}_3$  concentrations of 30,000 PPM over several hours the reaction rate of NOXNON 500 and 600 showed no decrease after several such exposures.

#### EPA FLUE GAS DENITRIFICATION DEMONSTRATION PLANT

In May of 1978 Hitachi Zosen received an order from the United States Environmental Protection Agency (E.P.A.) for a 1,700 Nm<sup>3</sup>/h (0.5 MW equivalent) capacity demonstration plant for its coal-fired power plant flue gas denitrification process.

As part of its energy strategy, the United States Government plans to establish regulations which will require newly installed power plants to use coal as their energy source instead of heavy fuel oil or natural gas. But most U.S. coals have high  $\mathrm{SO}_{\mathrm{X}}$  (sulfur oxides) and ash concentrations, and no commercial technology has been introduced to remove  $\mathrm{NO}_{\mathrm{X}}$  (nitrogen oxides) from flue gases from power stations using these coals.

Therefore, the E.P.A. decided to construct a flue gas denitrification demonstration plant to carry out technological and economical assessments on high  ${\rm SO}_{\rm X}$  and dust-containing coal-fired power station flue gas.

The objective of the pilot plant program is to demonstrate Hitachi Zosen's catalytic reduction process for the treatment of flue gas with ammonia for the removal of 90% of NO<sub>X</sub>. The program will evaluate the performance of the NOXNON 500 and 600 catalysts, which have been developed by Hitachi Zosen for flue gas of high sulfur oxide and high dust content which is exhausted from coal-fired boilers.

The original planned duration of the test program included a three-month start-up and test operation period plus three months of continuous operation. Varying levels of nitrogen oxide and sulfur oxide concentrations were tested along with varying

temperatures and gas flow rates. This program has been extended and the test operation will be run for over a year.

Through the generosity and support of Georgia Power Company the pilot plant host site is the Plant Mitchell of GPC at Albany, Georgia. The flue gas is obtained from Unit #3. This unit was built in 1964 and consists of a coal-fired C-E Boiler and a Westinghouse Turbine-Generator with a capacity of 165 MW. Steam temperature is  $1000^{\circ}$ F and the pressure 1800 PSI.

Flue gas for the pilot plant is obtained by connecting a 14" duct into the power plant duct after the economizer. The slipstream of flue gas is first passed through an electric heater to control the gas temperature, ammonia is injected into the gas, and the gas then passes through the reactor vessel where the nitrogen oxide is converted to nitrogen. The cleaned gas is repressurized in a blower and returned to the power plant duct. Additional quantities of nitrogen oxide and sulfur trioxide are generated, as required, to vary the concentration in the flue gas.

Construction and operation of the pilot plant was carried out by Chemico Air Pollution Control Corporation (CAPCC), Hitachi Zosen's North American licensee. Fabrication and procurement was done within the United States as far as possible.

In implementing the contract with E.P.A., Hitachi Zosen provided the basic design, proprietary catalysts, and fabrication of the  $\mathrm{NO}_{\mathbf{X}}$ ,  $\mathrm{SO}_3$  generating unit needed for the system. CAPCC, as major subcontractor, provided the detailed engineering and design, procurement, erection and operation of the pilot plant.

The initial NOXNON 500 catalyst charge provided excellent results for about 2000 hours of operation. Based on experience in Japan the sootblower was not operated because the pressure drop remained constant. After this period of time however, there occurred a drop in removal efficiency and an increase in

pressure drop. Removals decreased from the normal 90% level to the 80% level. Intensive efforts were made to check and recheck the analytical and control equipment to make certain that it was the catalyst at fault. It became obvious that the catalyst activity had definitely decreased below the acceptable level.

A spare batch of NOXNON 500, which was at the site, was installed to replace the initial batch. The replacement material was exactly the same shape and composition as the original.

Samples of the original batch were flown back to Japan for intensive investigations. The results of these indicated that there was no loss of catalyst activity. The material was as chemically sound as originally provided. It appeared that the loss of activity was due to a masking of a portion of the catalyst surface due to a buildup of fly ash within the catalyst bed. This phenomenon was not exhibited in Japan during long-run tests at a coal-fired station. Analyses were made of the fly ash from Georgia as compared to that in Japan. One possible significant difference was that the Georgia material contained a relatively high level of unburned coal or carbon. Physical tests were performed on the fly ash and it showed that the Georgia material had a greater tendency to agglomerate at high temperatures.

One of the contributing factors to the fly ash build-up on the catalyst was the design of the ductwork. When the ducts were disassembled for an inspection of the system it was found that the design allowed a large accumulation of fly ash in a horizontal run above the reactor. Some of this fly ash undoubtedly loosened and fell onto the catalyst bed, thus blocking a portion of the flow through the reactor. This probably contributed to the lower than expected performance of the pro-

cess. The ductwork was changed soon after the inspection was made and such problems were not encountered later.

The replacement NOXNON 500 also gave excellent results for some months of operation. However, again after some 2000 hours of operation it began to exhibit a loss of activity. This happened despite daily operation of the sootblower. This catalyst was taken out of the reactor, cleaned with compressed air, and was replaced. The pressure drop, which had increased at the same time the removal decreased, returned to a normal level but the activity did not recover.

The loss of activity of the second batch of catalyst was found to be due to a physical blinding of the surface of the catalyst by fly ash. The degree of fly ash accumulation was surprising. Investigations into the causes continue. One clue is the tendency to an increased pressure drop when the system is shutdown and restarted. One theory is that deposits on the elements of the flue gas heater are released when the system is They are then carried with the flue gas during cooled down. restarts, into the reactor. The extreme temperatures on the heater elements over long periods of time probably cause physical or chemical changes in the deposited fly ash which might make it more adhesive. This altered fly ash could stick to the catalyst surface. This situation was not encountered during many months of operation in Japan probably because of the difference in the heater design and the difference in the fly ash composition. If this did actually occur in the pilot plant, it would not be expected to happen in commercial systems where flue gas heaters are not required.

The third batch of catalyst was then installed. This catalyst was of Hitachi Zosen's latest design: NOXNON 600. This is made from a metallic mesh instead of from sheets of steel and is consequently much lighter in weight. Of more significance is the fact that the pitch of this catalyst was larger:

14 mm vs. 8 mm. The openings are much wider and the tendency to clog is greatly reduced.

Because of the wider pitch, the reactor had to be lengthened to accept the new catalyst which required more volume. This NOXNON 600 catalyst has been in service since April 1980 and, as of this writing at the end of July, is providing excellent results. It is planned to test this catalyst at least until October.

#### PROCESS DESIGN CONSIDERATIONS

#### AMMONIUM SULFATE/BISULFATE FORMATION

The formation of ammonium sulfate (or ammonium bisulfate) produced by the reaction between ammonia and sulfur trioxide can cause difficulties in the process. Deposits can result in a decrease of catalyst activity due to adhesion on the catalyst-activated surface and/or blockage of the catalyst layer.

There is an increased tendency towards ammonium sulfate formation at higher levels of ammonia and sulfur trioxide and at lower temperatures. The best means of prevention is to maintain a temperature so high that this compound cannot form. This is normally around 330°C (620°F). If the reactor temperature, for some reason, begins to drop and approaches this temperature it would be best to stop the ammonia flow until the required minimum temperature is reattained. If deposits are formed on the catalyst by a process upset the sootblower can be effective in removing these.

Deposits on downstream equipment such as air preheaters are best avoided by maintaining a low level of ammonia slippage.

AMMONIA CONTROL SYSTEM

The pilot plant at Georgia was designed to simulate as closely as possible the control system for a full-sized commercial plant. The ammonia feed system is completely automatic and requires only that the operator select the desired mole ratio. From that point on the ammonia feed will be delivered as required to satisfy the selected ratio of ammonia to incoming nitrogen oxide.

In this control system, the flue gas flow rate and the incoming nitrogen oxide concentration are measured and the signals are multiplied to provide the mass flow of  $NO_X$  entering the reactor. This quantity is then multiplied by the set mole ratio to determine the amount of ammonia required. This signal, in turn, is relayed to the ammonia flow meter to set the ammonia flow.

This system has worked exceptionally well. This ammonia control system can probably be applied to a full-scale unit along with some refinements, such as using the outlet  $\mathrm{NO}_{\mathrm{X}}$  or outlet ammonia concentration to provide a signal to fine tune the ammonia feed rate.

One potential drawback to this control system is the measurement of the flue gas flow rate. This is difficult to do on a large boiler system because usually there are no straight runs of duct in which a flow element could be installed. Most of the flue gas passes through headers without any significant ducting. If there were a straight run, either before or after the reactor, a flow element could be installed to provide a pressure drop signal as a measure of gas flow. Such a system is used at the Georgia pilot plant and might be practical even for larger boilers.

In the absence of a flue gas flow meter other signals are available. One of these is the steam flow rate to the turbine. This is a measure of the fuel burned and indirectly a measure of the combustion gases produced. Combining this signal with the inlet  $\mathrm{NO}_{\mathrm{x}}$  level will provide a fairly accurate measure of the  $\mathrm{NO}_{\mathrm{x}}$  mass loading. This control can then be fine tuned using the outlet  $\mathrm{NO}_{\mathrm{x}}$  level or the percent removal of  $\mathrm{NO}_{\mathrm{x}}$ .

#### REACTOR INSTRUMENTATION

Both temperature and pressure drop are required measurements for the reactor. The temperature of the flue gas has an effect on the removal efficiency. Normally, the temperature would not be controlled and the reactor would accept whatever the gas temperature would be at the economizer exit. At the Georgia pilot plant an electric heater is supplied to control the temperature because of the high heat loss from the ductwork, typical of pilot plants.

A low temperature at the reactor inlet is of concern because of the risk of ammonium bisulfate formation. At low temperatures an interlock is provided to shut down the ammonia supply by over-riding the ammonia flow controller.

The pressure drop across the reactor is an important measurement. This will indicate whether there is any tendency to plugging by fly ash or by other deposits within the catalyst bed. At an increased pressure drop the sootblower is actuated to clean the catalyst.

#### SOOTBLOWER CONTROL SYSTEM

Soot blowers are installed to periodically clean the catalyst bed. They can use steam or air. The system can be completely automated and initiated manually by a push button. At the Georgia pilot plant the system is set up the same as would be provided for a commercial system. Upon initiation steam or air is admitted into the electric heater which raises the temperature of the gas to 650°F. The gas is released to atmosphere until the temperature is sufficiently high. When the temperature is high enough a valve opens permitting the gas to flow into the sootblower and at the same time the sootblower moves forward across the top of the reactor blasting the hot gas down through the catalyst. After one cycle, forward and back, the system automatically stops, the remaining gas is released to the atmos-

phere, and the system shuts off. There are interlocks provided to ensure proper gas pressure and to ensure that the sootblower moves properly. The air or steam pressure and temperature are monitored. At the Georgia pilot plant the sootblower has been run once per shift since the NOXNON 600 catalyst was installed. Both air and steam are being tested.

#### AMMONIA SLIPPAGE

A certain amount of ammonia can be lost from the process without reacting. This can happen particularly at high  $\mathrm{NO}_{\mathrm{X}}$  removals when there would have to be an excess of ammonia available to approach complete reactions with the  $\mathrm{NO}_{\mathrm{X}}$ . Experience has shown that slippage of ammonia is negligible below about 80%  $\mathrm{NO}_{\mathrm{X}}$  removal. At 90% removal or above the slippage can amount to 10-20 PPM or higher.

The control of ammonia slippage is important mainly because of the risk of accumulations of ammonium sulfate/bisulfate on downstream equipment. In Japan some clients have requested targets as low as 10 PPM. The need for a continuous measurement of ammonia slippage is very obvious.

At the Georgia pilot plant ammonia slippage has been essentially nil below a mole ratio of 0.9. As the ammonia feed is increased, however, the slippage rapidly increases as can be seen in Figure 5.

The pilot plant in Albany, Georgia was originally designed to provide a continuous read out of ammonia levels in the flue gas leaving the reactor. The design used a chemiluminescent analyzer with two converters. The sample of flue gas drawn through the sample line was split. Half was sent through a high temperature converter to convert all of the ammonia and NO<sub>2</sub> to NO. The other half of the sample stream was passed through a low temperature converter which converted only the NO<sub>2</sub> to NO and did not affect the ammonia. The analyzer was designed to deter-

mine the NO level in each of the two streams in the two chambers and by difference provide the ammonia concentration. In principle this system should work but the problem was to ensure that the ammonia reached the converter.

Ammonia can be easily lost from the flue gas sample before it reaches the analyzer. It can react with SO<sub>2</sub> or SO<sub>3</sub> or it can be absorbed by condensed moisture or by deposits of solid material. We have even experienced the absorption of ammonia by certain gas filters.

Despite heroic efforts at the pilot plant it was not possible to make the ammonia analytical system operable. A good sample to the analyzer could not be ensured. Eventually they went to wet sampling methods using impingers to absorb ammonia from gas samples and a Hach colorometric method to determine the concentration of ammonia in the absorbent. This system has provided the necessary data but would not be a practical system for a commercial plant.

Further efforts continue in the design and testing of a continuous ammonia slippage monitoring system. A gas conditioning system has been designed by GCA and Charlton Associates to treat a sample of flue gas at the sample point in the duct before the sample can cool down. A sample of flue gas is filtered and then passed through an absorbent to remove sulfur trioxide. gas is either passed through a stainless steel converter to convert the ammonia to NO or is bypassed around the converter. The gas sample is then dried and sent by the sample line to the analyzer at the control rooms. By comparing the NO level of the sample passed through the converter and the sample bypassing the converter, the ammonia analysis can be determined. This system avoids the problem of the reaction of ammonia with sulfur trioxide by absorbing the sulfur trioxide and avoids the problem of the reaction with sulfur dioxide by maintaining a minimum temperature above which the reaction cannot take place.

## FIGURE 1 NOXNON 600 CATALYST CONFIGURATION

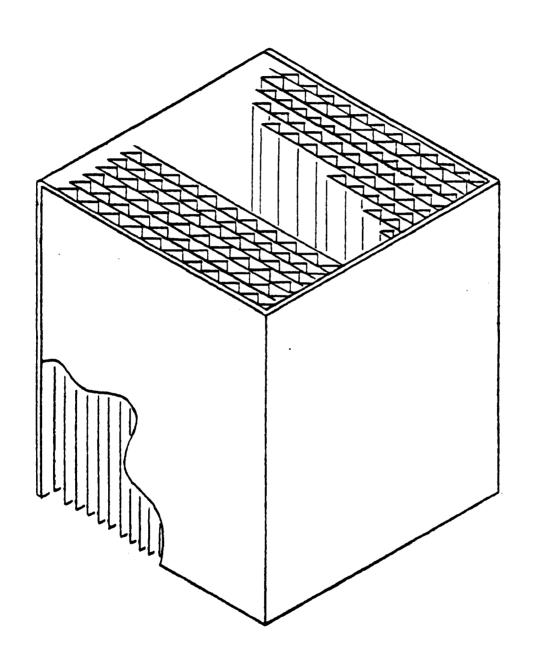
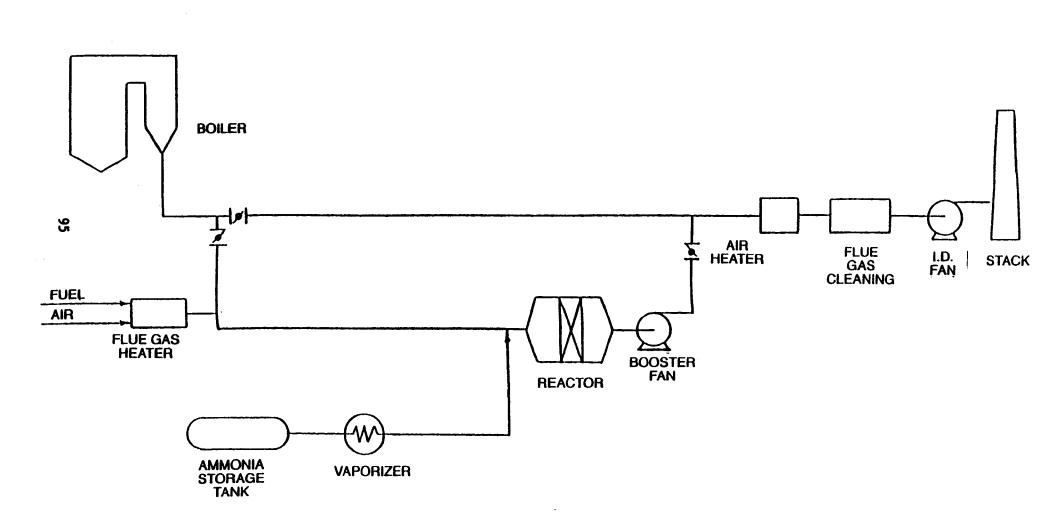
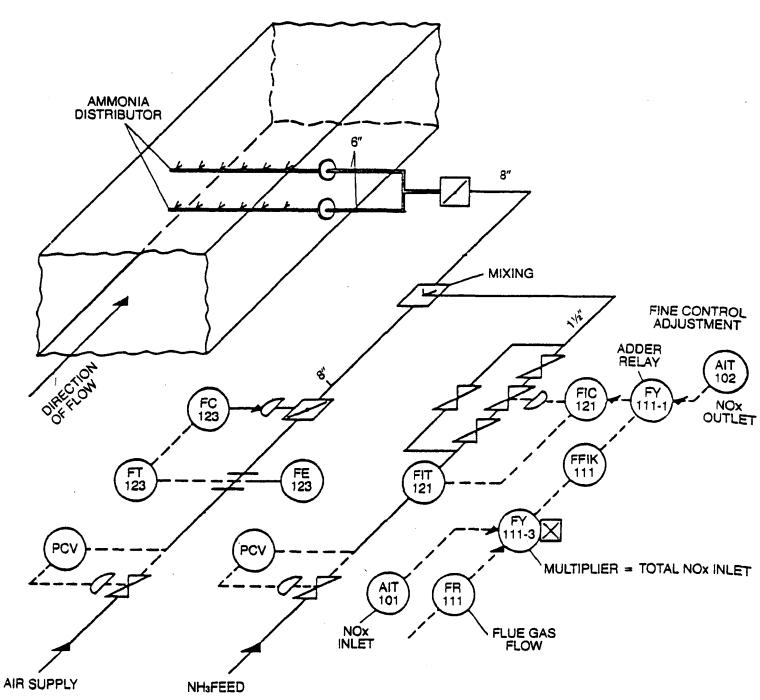


FIGURE 2

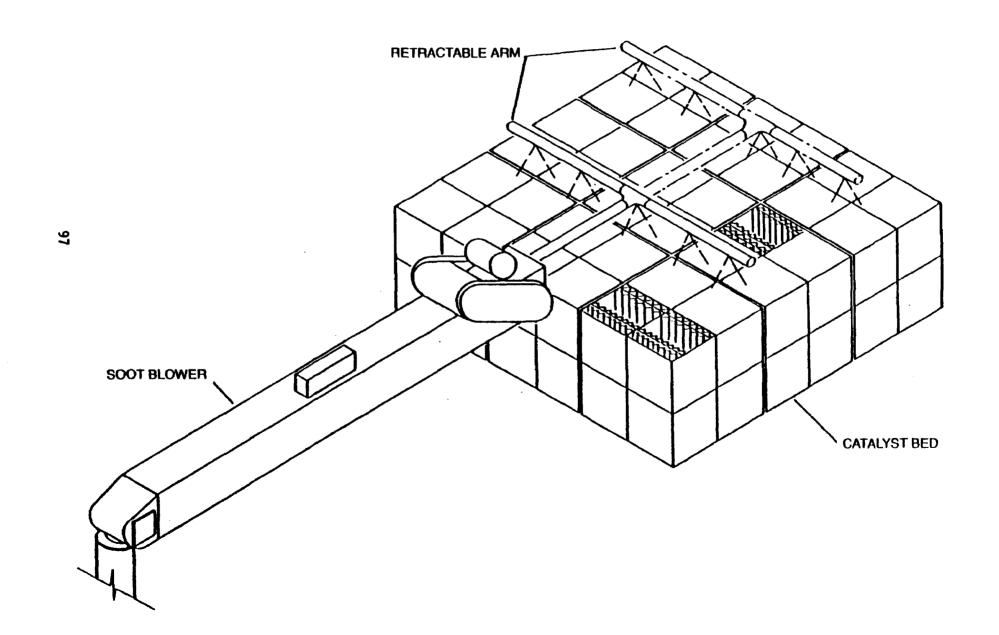
TYPICAL FLOW SHEET COMMERCIAL SYSTEM



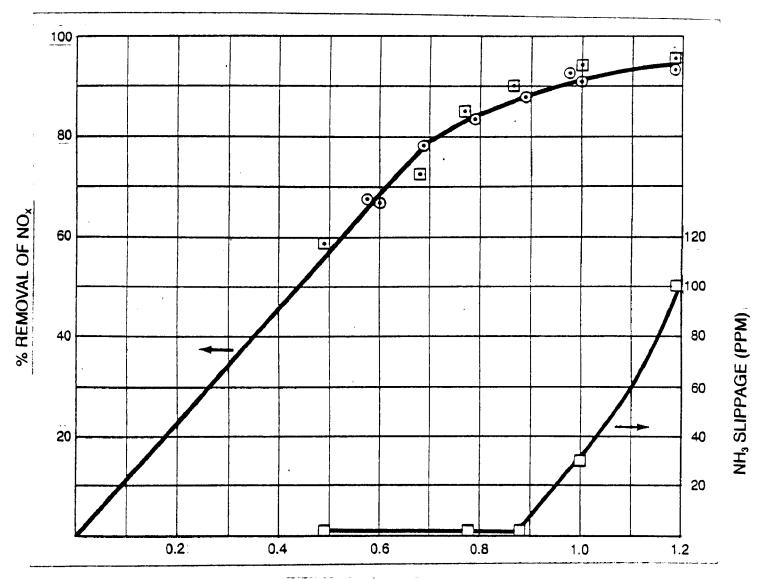
### FIGURE 3 AMMONIA INJECTION SYSTEM



# FIGURE 4 SOOT BLOWER



### NO<sub>x</sub> REMOVAL vs. MOLE RATIO



MOLE RATIO NH√NO<sub>x</sub>

Catalyst: NOXNON 600

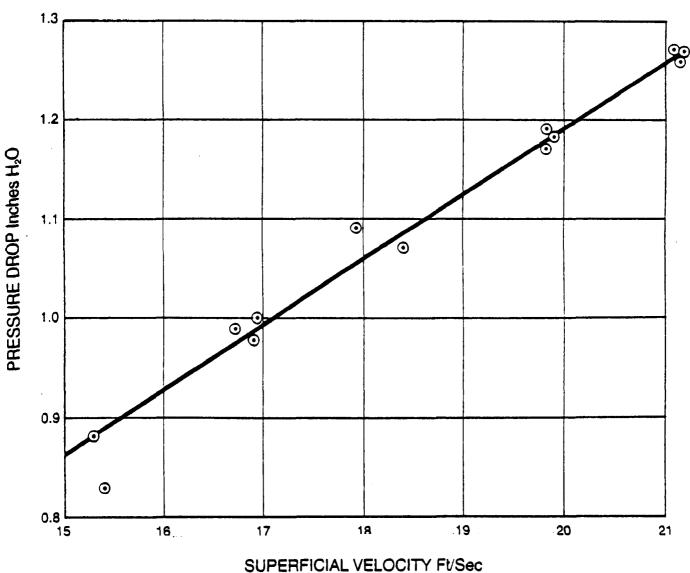
Temp.: 700-710°F

Flow Rate: ○ 1500 SCFM

□ 1300 SCFM

Inlet NO<sub>x</sub>: 400-500 PPM

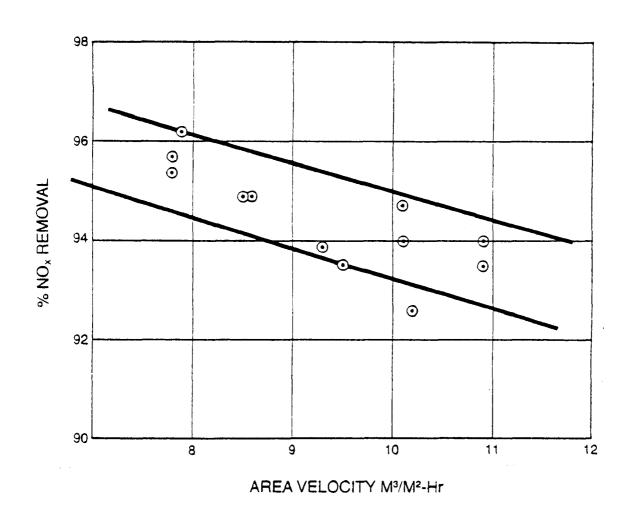
### FIGURE 6 PRESSURE DROP vs. VELOCITY



**NOXNON 600** 

Bed Depth: 2.0 Meters

Temperature: 715°F

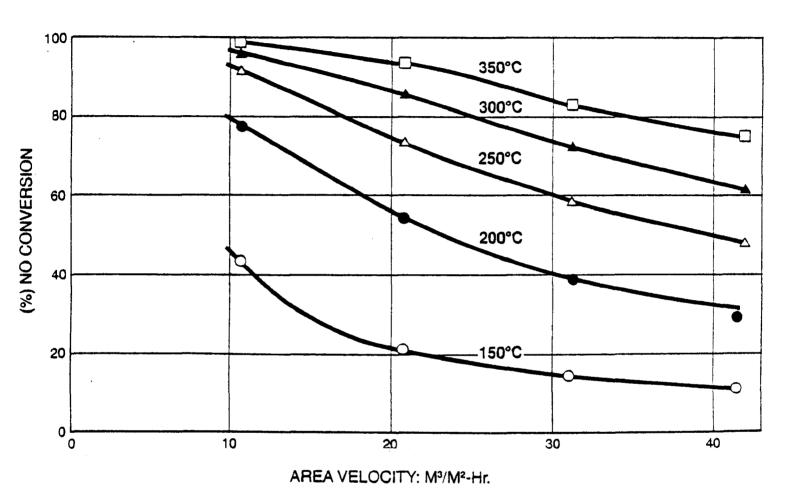


Mole Ratio: 1.0

Reactor Temp.: 720°F

Inlet NO<sub>x</sub> Conc.: 400-450 PPM

### FIGURE 8 AREA VELOCITY vs. NO CONVERSION



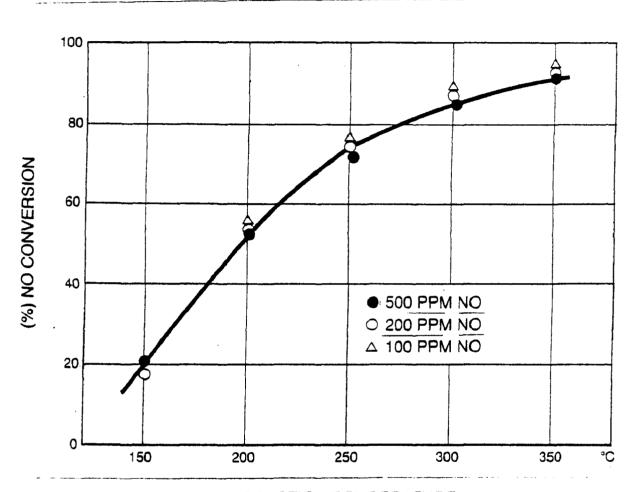
Catalyst: NOXNON 600

Gas Composition: 500 PPM NO

500 PPM NH<sub>3</sub>

250 PPM SO<sub>2</sub>

6% O<sub>2</sub>, 10% H<sub>2</sub>O, 10% CO<sub>2</sub>



**REACTION TEMPERATURE** 

Catalyst: NOXNON 600

Area Velocity: 20.4 M³/M²-Hr.

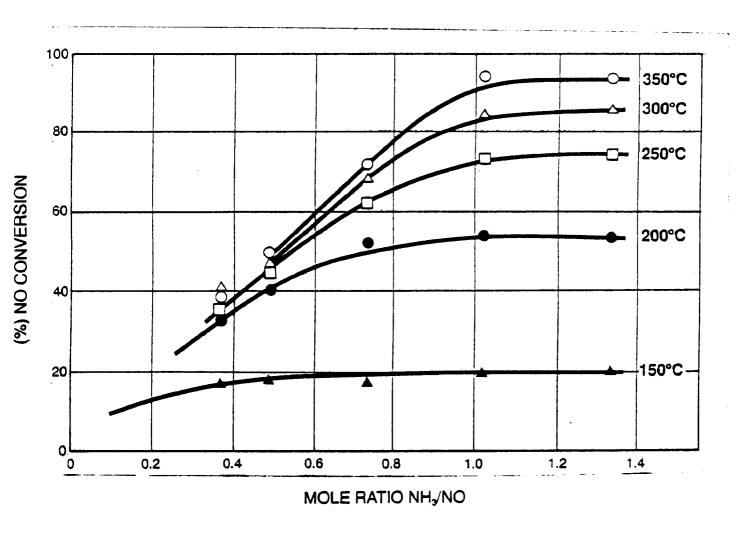
Gas Composition: 250 PPM SO<sub>2</sub>

6% O₂

10% H<sub>2</sub>O

10% CO<sub>2</sub>

## FIGURE 10 MOLE RATIO vs. NO CONVERSION



Catalyst: NOXNON 600

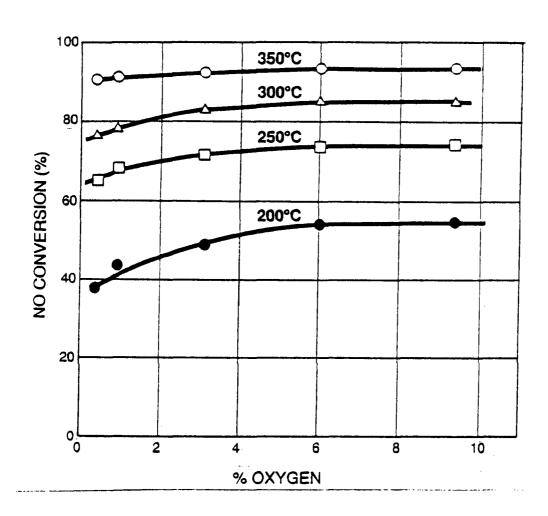
Area Velocity: 20.4 M³/M²-Hr.

Gas Composition: 500 PPM NO

250 PPM SO<sub>2</sub>

6% O<sub>2</sub>, 10% H<sub>2</sub>O, 10% CO<sub>2</sub>

# FIGURE 11 OXYGEN LEVEL vs. NO CONVERSION



Catalyst: NOXNON 600

Area Velocity: 20.4 M³/M²-Hr.

Gas Composition: 500 PPM NO

500 PPM NH<sub>3</sub>

250 PPM SO<sub>2</sub>

10% H<sub>2</sub>O, 10% CO<sub>2</sub>

## TABLE 1

## HITACHI ZOSEN LIST OF COMMERCIAL PLANTS

Customer	Treating Capacity, Nm <sup>3</sup> /hr.	Flue Gas Source	Process	Completion
Osaka Gas Co., Sakai	53,000	LNG or naphtha-fired furnace	Ammonia Reduction	1975
Daiki Engineering, Chiba	5,000	LPG-fired furnace	Ammonia Reduction	1975
Idemitsu Kosan, Chiba	350,000	CO boiler and gas- fired heater	Ammonia Reduction	1975
Shin-Daikyowa Petro- chemical, Yokkaichi	440,000	Fuel oil-fired boiler with wet-type desulfurization	Ammonia Reduction	1975
Hitachi Zosen, Osaka	6,000	Gas-fired annealing furnace	Ammonia Reduction	1975
Toshin Steel Mill, Himeji	70,900	Kerosene-fired steel heating furnace	Ammonia Reduction	1976
Kawasaki Steel, Chiba	762,000	lron ore sintering plant with wet-type desulfurization	Ammonia Reduction	1976
Nippon Satetsu, Himeji	10,000	Fuel oil-fired steel heating furnace	Ammonia Reduction	1977
Kansai Oil Co. Sakai	150,000	Fuel oil fired boiler	Ammonia Reduction	1979
	Osaka Gas Co., Sakai Daiki Engineering, Chiba Idemitsu Kosan, Chiba Shin-Daikyowa Petro- chemical, Yokkaichi Hitachi Zosen, Osaka Toshin Steel Mill, Himeji Kawasaki Steel, Chiba Nippon Satetsu, Himeji Kansai Oil Co.	Customer Capacity, Nm³/hr.  Osaka Gas Co., Sakai Daiki Engineering, 5,000 Idemitsu Kosan, 350,000 Idemitsu Kosan, Chiba Shin-Daikyowa Petrochemical, Yokkaichi Hitachi Zosen, 6,000 Toshin Steel Mill, 70,900 Kawasaki Steel, 762,000 Nippon Satetsu, 10,000 Himeji Kansai Oil Co. 150,000	Customer  Capacity, Nm³/hr.  Capacity, Nm³/hr.  Capacity, Nm³/hr.  Flue Gas Source  LNG or naphtha-fired furnace  LNG or naphtha-fired furnace  LPG-fired furnace  LPG-fired furnace  LPG-fired furnace  CO boiler and gas-fired heater  Shin-Daikyowa Petrochemical, Yokkaichi  Hitachi Zosen, Osaka  Toshin Steel Mill, Himeji  Kawasaki Steel, Chiba  Nippon Satetsu, Himeji  Kansai Oil Co.  CO boiler and gas-fired heater  Fuel oil-fired boiler with wet-type desulfurization  Gas-fired annealing furnace  Kerosene-fired steel heating furnace  Iron ore sintering plant with wet-type desulfurization  Fuel oil-fired steel heating furnace  Fuel oil fired	Customer Capacity, Nm³/hr. Flue Gas Source Process  Osaka Gas Co., Sakai Daiki Engineering, Chiba Idemitsu Kosan, Chiba Shin-Daikyowa Petrochemical, Yokkaichi Hitachi Zosen, Osaka Toshin Steel Mill, Himeji Kawasaki Steel, Chiba Nippon Satetsu, Himeji Kansai Oil Co.  Capacity, Nm³/hr. Flue Gas Source Process  Flue Gas Source Process  Ammonia Reduction Ammonia Reduction Process  Ammonia Reduction Process  Ammonia Reduction Reduction Reduction Process  Ammonia Reduction Reduction Reduction Process  Ammonia Reduction Reduction Process  Ammonia Reduction Reduction Process  Ammonia Reduction

# BABCOCK-HITACHI NO $_{\mathbf{x}}$ REMOVAL PROCESS FOR FLUE GASES FROM COAL-FIRED BOILERS

### By:

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#### **ABSTRACT**

In the previous symposium in 1978 we presented a paper titled "SOME EXPERIENCES OF NOx REMOVAL IN PILOT PLANTS AND UTILITY BOILERS". In that paper we stated the history of developments of our process, catalyst characteristics and several operating experiences. This time we intend to introduce some of the improvements and developments which we have achieved since then.

As far as coal-fired applications are concerned, there are two systems required. The first one is DeNOx with low dust loading where the DeNOx reactor is located downstream of the hot electrostatic precipitator (EP), and the second is DeNOx with high dust loading where the DeNOx reactor is located upstream of the cold EP. Although the selection of EP system should be determined mainly from the standpoint of performance of collecting fly ash through the boiler, our DeNOx process is applicable in either case.

As for the two commercial DeNOx plants with low dust loading from coalfired boilers, we have already completed the design and manufacture, and they will go into commercial operation in November of 1980 and in July of 1981, respectively. Concerning the DeNOx with high dust loading, we will introduce in this paper the results of abrasion and performance tests under dust concentration of 15 to 20 g/Nm<sup>3</sup>, through which we have confirmed the reliability of the catalyst.

Furthermore, another important thing in the Selective Catalytic Reduction (SCR) process is to reduce the conversion of SO<sub>2</sub> to SO<sub>3</sub> in order to minimize the influence on the downstream equipment. We have developed a catalyst with the lowest conversion rate less than 0.5% without decreasing the NOx conversion activity at the rated load.

#### INTRODUCTION

Recently the use of coal as a substitute fuel for fossil oil has become of world wide importance. Since the characteristics of coal vary according to the location of mining, those of the coal-fired flue gas also vary. Generally, it contains not only more dust  $(15\sim25~{\rm g/Nm^3})$  than heavy oil-fired flue gas  $(0.05\sim0.2~{\rm g/Nm^3})$ , but also higher NOx (Nitrogen Oxides) and higher SOx (Sulfur Oxides) concentration. This means that the DeNOx system, which is one of the environmental protection systems for coal-fired flue gas, demands more sophisticated technology than for heavy oil-, or gas-fired flue gas.

It was introduced at the previous symposium that Babcock-Hitachi had produced a high activity catalyst through extensive R&D, and has succeeded in developing it into a thin plate catalyst (a parallel flow type) for dirty gas. The plate type catalyst has been adopted for 20 plants out of 23 plants which have been ordered or are intended to be ordered soon. 10 of these plants are already operating satisfactorily or are under preoperation stage. Recently, demand for a DeNOx system for coal-fired flue gas has been increasing rapidly. Our first new DeNOx plant for low dust coal-fired flue gas (Hokkaido Electric Power Co., Tomato Atsuma P.S. Unit 1), the first in Japan to be installed in a new coal-fired P.S., will be in commercial operation this coming November, while the second plant (Electric Power Development Co. (EPDC) Takehara P.S. Unit 1) will also be in commercial operation next July. In addition to these, several plants including those for high dust flue gas are now being designed.

This paper describes the features of the DeNOx system for coal-fired flue gas in comparison with that for the oil-fired flue gas and includes an example of a DeNOx system for a coal-fired 500 MW boiler.

Furthermore, a pilot plant test using the plate type catalyst for a coal-fired flue gas has been conducted at Takehara P.S. on a co-study basis with EPDC.

#### ITEMS CONSIDERED IN THE DESIGN OF THE DeNOx SYSTEM FOR COAL-FIRED FLUE GAS

As the characteristics of coal-fired flue gas differ much from those of the heavy oil-fired flue gas, the peculiarities of the coal-fired flue gas have to be taken into consideration when designing the DeNOx system. Fig. 1 is a summary of the items considered and the measures taken in each item.

Our company has been producing the catalyst, which is a key point in the DeNOx technology, in accordance with our policy of manufacturing in our own works, from mixing of material to final product, under strict quality control since the start of development. Our success in developing a catalyst with high performance in practical use, is the result of a long program of catalyst improvement based on severe tests conducted in our research laboratory, factory, and in the field.

The catalyst, is composed of many thin plate elements, and its shape resembles the N.F elements of an air heater, so it is free from dust accumulation and has a low pressure drop. Additionally it has high erosion resistance and rigidity because it contains inner layer metal plate. It was confirmed that the performance of the catalyst had not decreased against the influence of SOx, halogen compounds (HCl, HF), and alkaline metals (K, Na) in the dust, even after 10,000 hours test with an actual coal-fired flue gas. Especially, the conversion ratio of SO<sub>2</sub> to SO<sub>3</sub> which is apt to cause problem in the downstream equipment is small enough to say that our DeNOx catalyst for coal-fired flue gas has a great advantage. Moreover, based on the results of model testing, improvements have been made to the reactor structure to prevent dust accumulation and non-uniform gas flow, and to simplify the loading and removal of catalyst.

#### SYSTEM FOR DeNOX

The flue gas treatment system for coal-fired boiler consists of three major systems, namely DeNOx, DeSOx and Dust Collection. Each system, in addition to fulfilling its own particular function, should, as part of the

general boiler system, contribute to the total reliability and stability of the system over a long period of time, and be economical. Taking the above into consideration, we put two treatment systems for coal-fired flue gas into practical use. Those are shown in Fig. 2.

One is called a low dust DeNOx system, in which the boiler flue gas is first treated through a hot EP, and then the low dust flue gas is treated in the DeNOx reactor. The other is called a high dust DeNOx system, in which the boiler flue gas is treated in the DeNOx reactor without any pre-treatment after the economizer and followed by a cold EP.

The selection of the DeNOx system depends on the EP type which should be determined mainly from the standpoint of performance of collecting fly ash of which properties vary widely according to the location where fuel coal is mined. As far as DeNOx system is concerned, tests using actual flue gas have been conducted sufficiently for both systems, and it has been confirmed that both are practical as DeNOx system. The construction or the design of actual plants for each system is being carried out at present.

#### DeNOx SYSTEM WITH LOW DUST LOADING

We introduced some results of a low dust loading test for practical use at the previous symposium. We had performed a pilot plant test, in which we carried out long duration operation of more than 10,000 hours and confirmed various characteristics of the low dust loading. For instance, no change is found in DeNOx efficiency and reactor pressure drop, see Fig. 3, which shows that the DeNOx system can be operated quite stably. As shown in Fig. 3, DeNOx inlet dust concentration of 20, 100, 200 mg/Nm<sup>3</sup> were used and in addition 12 g/Nm<sup>3</sup> was experienced at hot EP trip, but there was no change in DeNOx efficiency nor reactor pressure drop. During this test the annual maintainance inspection, lasting 74 days, for the boiler unit was carried out and during this shutdown the DeNOx plant was exposed to the atmosphere, however this had no influence on the catalyst.

Summarizing the above, we can come to the conclusion that our DeNOx system has high reliability for practical use at low dust loading. The result of actual operation of the DeNOx system at Hokkaido Electric Power Co., and at plant of Electric Power Development Co. are expected to be excellent.

#### DeNOx WITH HIGH DUST LOADING

A high dust DeNOx system treats the flue gas from a boiler without any pretreatment. Therefore, the main aim in developing this system is to obtain an erosion resistant catalyst, which can withstand large amounts of, coarse, and hard dust composed of mainly silica and aluminum. For the purpose of overcoming this problem, an accelerated erosion test for the plate type catalyst was conducted with actual fly ash in Hitachi research laboratory. Fig. 4 shows the test equipment and Fig. 5 shows the evaluation of the test results.

According to our investigation of abrasion phenomenum, the erosion caused by abrasion with dust on the plate catalyst starts almost at the front edge of the plate, and then gradually proceeds downward. The catalyst material is integrally coated to stainless steel support plate which works as a protector against the abrasion by particulates. It was confirmed that our catalyst has extraordinary high erosion resistance. Summarizing the test results, the weight loss of the catalyst caused by abrasion can be estimated using the equation in Fig. 5.

Moreover, in order to conduct an abrasion test on the catalyst in an actual gas, the abrasion test catalyst was installed upstream of a hot E.P. in the same pilot plant as used for the low dust test mentioned in preceeding section. The test with high dust loading was conducted at a velocities of 4, 6, 8 m/s for about 3,000 hours. The result showed little or no erosion nor deactivity of the catalyst employed in the test. The result of this accelerated test with a gas velocity of 6 to 8 m/s was converted to the actual operating basis where the gas flows at a velocity of 4 to 5 m/s, and the

result of this evaluation was that the operating duration would be equivalent to as much as 17,000 to 23,000 hours. Accordingly it is apparent that the catalyst can be used for commercial plant.

Fig. 6 shows the result of a high dust performance test, using the catalyst that had been used for the abrasion test mentioned above.

Up to now approximately 4,000 hours of performance testing, and as far as abrasion is concerned, a total of 7,000 hours of testing has been completed satisfactorily. It can be seen from Fig. 6 that little or no deactivity of the catalyst nor increase in pressure drop across the reactor has occurred over the test duration, as was expected.

Several inquiries have been received regarding commercial DeNOx systems for high dust loading and some of them are at present under design.

## LOW CONVERSION OF SO2 TO SO3

As illustrated in Fig. 1 when designing a DeNOx system for coal-fired flue gas, such points as "minimum effect on the downstream equipment" should be taken into consideration. To achieve this, the following conditions are essential.

- 1) A low conversion rate of SO<sub>2</sub> to SO<sub>3</sub>
- 2) Minimized slip NH3 from the DeNOx system

 $SO_3$ , mainly converted from  $SO_2$ , and  $NH_3$  in the flue gas react to form  $NH_4HSO_4$ , as in the following reaction formula (1).

$$NH_3(g) + SO_3(g) + H_2O(g) = NH_4HSO_4(liq.)$$
 (1)

Fig. 7 shows the relation between the precipitation temperature of NH4HSO4 and SO3 concentration with the parameter of NH3 concentration. From Fig. 7, under the ordinary SO3 and slip NH5 concentration at reactor outlet, it is

understandable that  $NH_{4}HSO_{4}$  is formed at around 230~250 °C, which usually coincides with the temperature range in the intermediate section of the air heater downstream.

NH<sub>4</sub>HSO<sub>4</sub> becomes such a sticky liquid over 150 °C that it is likely to adhere to the elements of the air heater and cause some dust to accumulate on the elements, ultimately resulting in problems, such as increase in pressure drop and plugging. In order to prevent this, it is preferable to slightly modify the air heater with regard to element shape, section deviding, and soot blowing procedure.

Most important is to minimize the formation of NH<sub>4</sub>HSO<sub>4</sub>, which causes trouble downstream. In order to prevent SO<sub>3</sub> and NH<sub>3</sub> from forming NH<sub>4</sub>HSO<sub>4</sub>, the SO<sub>3</sub> and NH<sub>3</sub> concentrations in the flue gas have to be kept as low as possible. To minimize slip NH<sub>3</sub>, the reactor should be designed under an appropriate Space Velocity (SV), which is the gas quantity (Nm<sup>3</sup>/h) divided by the catalyst volume (m<sup>3</sup>), and operated with low (NH<sub>3</sub>)/(NO<sub>3</sub>) mole ratio, then the reaction between NH<sub>3</sub> and NO<sub>3</sub> should be maximum. The slip NH<sub>3</sub> can be kept at less than 5 ppm.

Regarding SO<sub>3</sub> which forms  $NH_4HSO_4$ , in addition to the SO<sub>3</sub> originated in the combustion flue gas, SO<sub>3</sub> is converted from SO<sub>2</sub> under the presence of the DeNOx catalyst. In the case of coal-fired gas, SO<sub>2</sub> is contained at the inlet of DeNOx reactor in such quantity, about  $1000 \sim 2000$  ppm, that large quantitis of SO<sub>3</sub> will be formed if we use an ordinary catalyst. It means  $20 \sim 40$  ppm of SO<sub>3</sub> will be formed newly with a catalyst which has a conversion ratio of SO<sub>2</sub> to SO<sub>3</sub> of 2 %, for instance. Needless to say the lowest conversion of SO<sub>2</sub> to SO<sub>3</sub> should be required for DeNOx catalyst for coal-fired flue gas.

Generally speaking, the catalyst which has high activity for NOx decomposition presented by formula (2) has also high conversion activity of SO<sub>2</sub> to SO<sub>3</sub> expressed in formula (3).

$$4NO + 4NH_3 + O_2 \Rightarrow 4N_2 + 6H_2O$$
 (2)

$$80_2 + 1/2 0_2 = 80_3$$
 (3)

In other words if SO<sub>2</sub> oxidation activity is restrained, the NOx decomposing activity will also be greatly reduced. However, we succeeded in achieving low SO<sub>2</sub> oxidation activity without decreasing the NOx decomposing activity at boiler rated load by the selection of the proper catalyst component.

Fig. 8 shows the pilot plant test results using actual flue gas. During about 6,000 hrs operation conversion of \$02 to \$03 has been kept at less then 0.5 % without any tendency to increase.

#### DESIGN EXAMPLE FOR A 500 MW COAL-FIRED UTILITY BOILER

For reference, we would like to present an example of a design of a reactor for a 500 MW coal-fired utility boiler.

Design conditions are as follows.

Gas flow rate	1,500,000 Nm <sup>3</sup> /h at MCR
No. of reactors	2/boiler
Gas temperature	370 °C
NOx at inlet	500 ppm (5% 0 <sub>2</sub> , dry basis)
SOx at inlet	1,000 ppm
Dust concentration	20 g/Nm <sup>3</sup>
DeNOx efficiency	Case-1 80 %
	Case-2 90 %
Slip NH3	10 ppm
	(5% 0 <sub>2</sub> , dry basis)

The outline of the reactor designed, the shape of the catalyst and the catalyst loading procedure are illustrated in Fig. 9 and 10.

The basic dimensions of the catalyst element are about 1 mm thickness and 10 mm pitch. Gas from the economizer comes down, parallel to each catalyst plate, to avoid dust accumulation and two stages of catalyst bed are contained in the reactor. Generally soot blowers to clean the catalyst

would be installed, however in the case of our plate type catalyst, which consists of thin catalyst plates arranged in parallel, very little dust accumulation on the catalyst was experienced, since there are very few corners in the flow path and the flow velocity is evenly distributed. In our design usually soot blowers for the catalyst are not necessary.

Fig. 11 shows the expected performance of the DeNOx for the boiler load at a DeNOx efficiency of 90 % (case-2). Generally, the DeNOx efficiency has a tendency to increase a little with lower load, which is caused by the relation of both the decreased gas flow rate and the decreased gas temperature. The pressure drop of a reactor can be designed to be as low as about 50 mmH<sub>2</sub>O and the SO<sub>2</sub> oxidation ratio, to be as low as 0.3~0.5 %. Our study is based on a NOx concentration of 500 ppm at the DeNOx reactor inlet (boiler outlet), however, in Japan boiler combustion technology has improved so much that the NOx emitted by a new boiler can be less than 200 ppm.

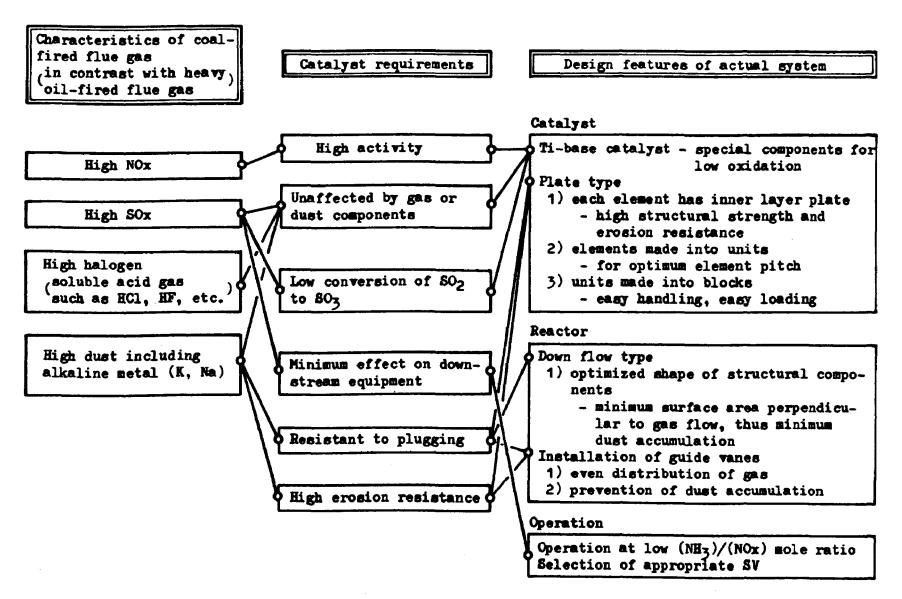
SV is influenced by the reactor inlet NOx concentration, so that under the condition of constant DeNOx efficiency and slip NH<sub>3</sub>, SV could be increased in proportion to lower NOx. The relation between SV and NOx concentration is shown in Fig. 12. Incidentally, if the NOx concentration at the DeNOx reactor inlet decreased from 500 ppm to 200 ppm, SV would increase by about 25 % and the quantity of catalyst required would decrease by about 20 %.

DeNOx efficiency is related to the quantity of catalyst, as shown in Fig. 13. It can be seen from Fig. 13 that the quantity of catalyst would decrease by about 30 %, in case the DeNOx efficiency lowered from 90 % to 80 %.

#### SUMMARY

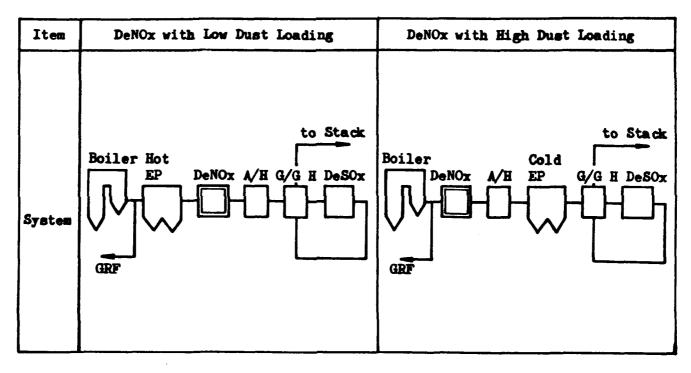
In this paper we have related some of our recent studies with the Babcock-Hitachi dry catalytic NOx removal process. Regarding the DeNOx efficiency, we can obtain as high a value as we wish, by increasing the catalyst quantity, but the most important points for the DeNOx system in

coal-fired plant are the catalyst characteristics of erosion resistance, anti-plugging and low conversion of  $SO_2$  to  $SO_3$ . We believe that our plate type catalyst, which we manufacture in our own works under strict control, should satisfy all of those requirements. Hitachi Ltd., including Babcock-Hitachi, as not only a utility boiler manufacturer but also as an integrated machine manufacturer, highly experienced in the field, will continue to strive to improve this technology with all its technological resources.



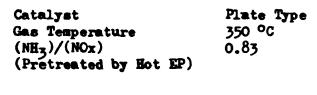
Remarks: NOx (Nitrogen Oxides), SOx (Sulfur Oxides), SV (Space Velocity)

Fig. 1 Consideration on DeNOx System for Coal-Fired Flue Gas



Remarks: GRF (Gas Recirculation Fan) G/G H (Gas/Gas Heater)

Fig. 2 Flue Gas Treatment System for a Coal-Fired Boiler



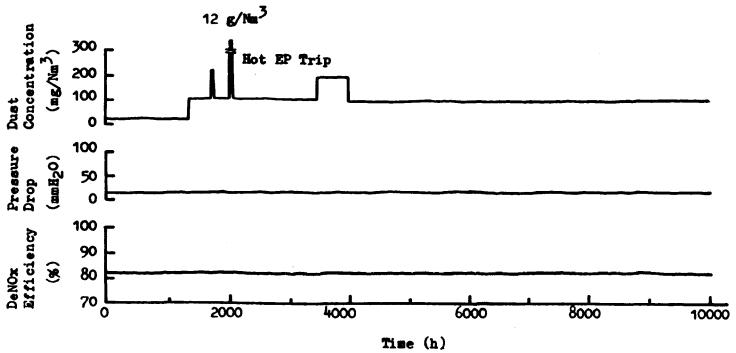


Fig. 3 Pilot Plant Test of Parallel Flow Reactor Treating Flue Gas from Coal-Fired Utility Boiler

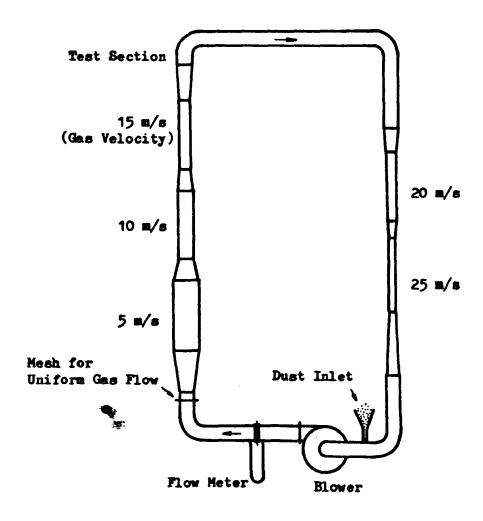


Fig. 4 Equipment for Catalyst Abrasion Test

Experimental Equation  $W = K \cdot V^{2.9} \cdot C^{1.7} \cdot T^{0.64}$ 

W: Weight Loss of Catalyst (wt%)

K : Constant

V : Gas Velocity (m/s)

C: Dust Concentration (g/Nm<sup>3</sup>)

T : Operating Time (h)

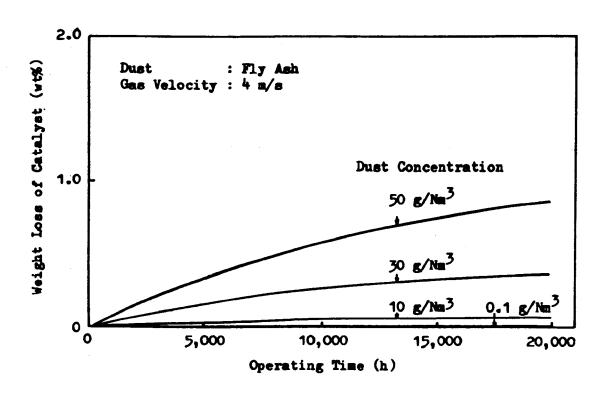


Fig. 5 Anticipated Weight Loss of Plate Type Catalyst vs Operating Time

Test Purpose	Abrasion Test	Performance Test
Gas Temperature	346 ~ 360 °C	348 ~ 353 °C
NOx	244 ~ 345 ppm	285~ 472 ppm
SOx	1100 ~ 1700 ppm	(1100 ~ 1700) ppm
Dust Concentration	16.1~19.0 g/Nm <sup>3</sup>	17.3~20.2 g/Nm <sup>3</sup>
LV	-	4 m/s
(NH3)/(NOx)	0	0.83

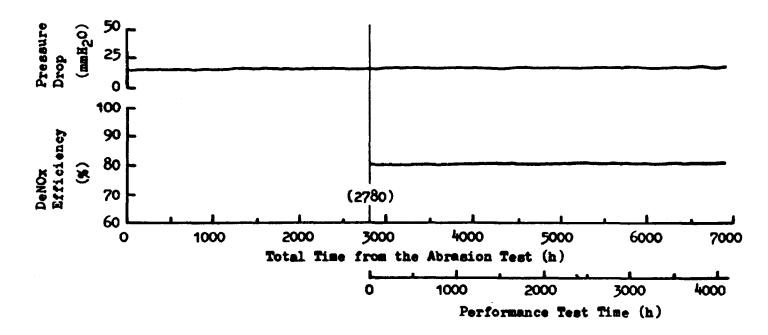
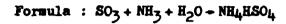


Fig. 6 Operation Chart of the Parallel Flow Type DeNOx Pilot Plant for Coal-Fired Boiler Flue Gas



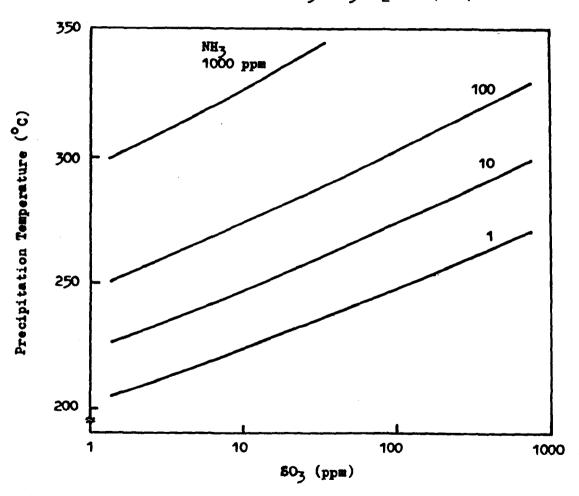


Fig. 7 Precipitation Temperature of Ammonium Bisulfate (NH4HSO4)

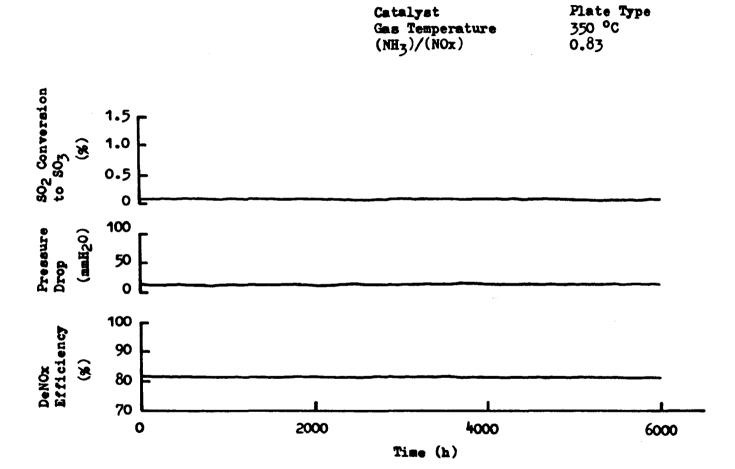


Fig. 8 Pilot Plant Test of a Parallel Flow Reactor Treating Flue Gas from a Coal-Fired Utility Boiler

CASE-1   CASE-2			(m)	
L 13 13 H 18 19  Reactor  Catalyst  Rlock		CASE-1	CASE-2	Gas Flo
R 18 19  Catalyst Rlock	W	11	11	
H 18 19  Catalyst  Rlock	L	13	13	Reactor
Block	H	_18	19	
				Block

Fig. 9 Outline of Reactor for 500 MW Coal-Fired Boiler

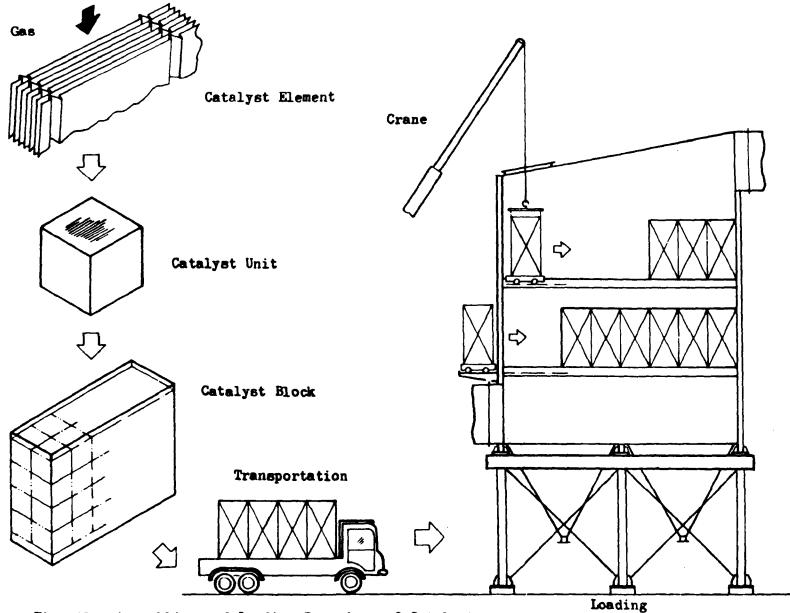


Fig. 10 Assembling and Loading Procedure of Catalyst

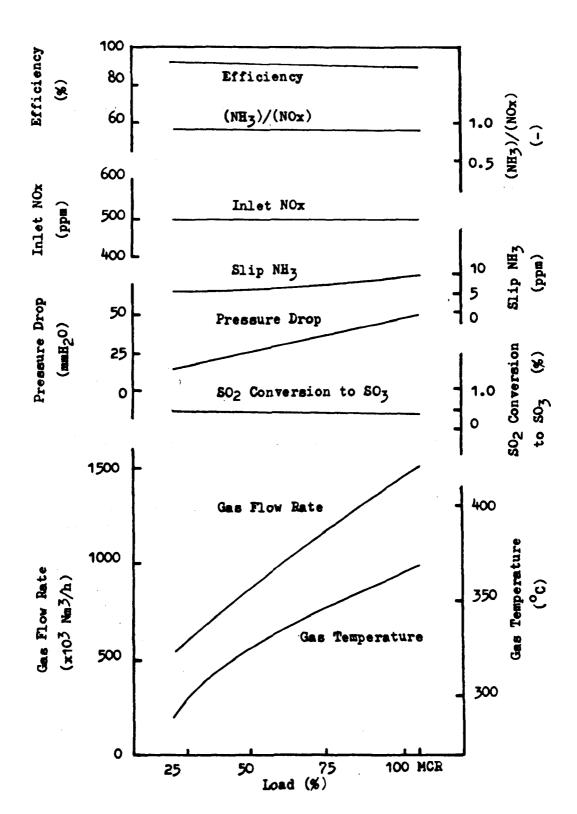


Fig. 11 Expected Performance with Load Conditions

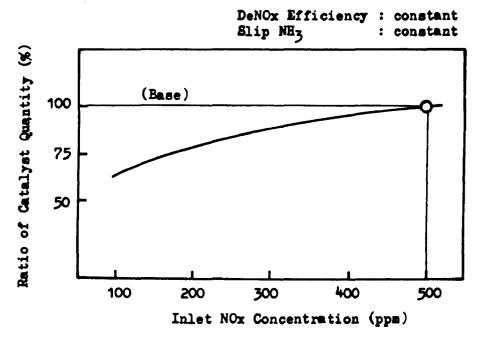


Fig. 12 NOx Concentration vs Catalyst Quantity

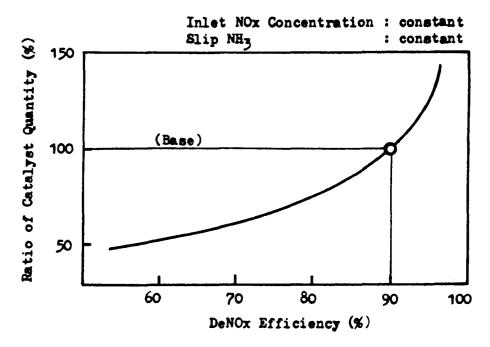


Fig. 13 DeNOx Efficiency vs Catalyst Quantity

#### TEST SUMMARY OF AN INTEGRATED FLUE GAS TREATMENT SYSTEM

Utilizing the Selective Catalytic Reduction Process for a Coal-Fired Boiler

By:

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#### ABSTRACT

This research program was initiated to investigate whether denitrification of flue gases from a coal-fired boiler can be effectively accomplished by using the selective catalytic reduction process in combination with desulfurization and dust control as an integrated system.

An experimental system was designed to handle between 1000 and 2000 NM<sup>3</sup>/ hr of flue gas and measure such parameters as catalyst life, catalyst plugging, catalyst abrasion rate due to ash, air heater plugging, hot ESP, wet ESP, and bag filter efficiencies, along with the efficiency of a limestone desulfurization system.

The results of this testing have shown that this integrated approach to flue gas cleanup is feasible and may be incorporated into a full-scale, coal-fired boiler flue gas design. This testing will be continued to develop a more reliable integrated flue gas treatment system prior to commercialization.

#### INTRODUCTION

Recent attempts to substitute coal for oil as an energy source for power plants have created a decrease in the overall air quality. To correct this trend, the need for an integrated flue gas treatment system has become apparent. This new integrated system must reduce the concentrations of dust,  $SO_X$ , and  $NO_X$  in the flue gases below those created by the former use of heavy oil.

To meet these needs, IHI started a joint research and development project for an integrated coal flue gas treatment system with Electric Power Development Co., Ltd. To prepare for this project, IHI started denitrification testing in January 1978 on flue gas from a coal-fired boiler at Isogo Power'Station of Electric Power Development Co., Ltd. In December 1978 a desulfurizing system was installed on the downstream side of the denitrification system. Various tests were conducted concerning denitrification, desulfurization, and dust collecting functions of this integrated flue gas treatment system. These tests have provided the opportunity to analyze the problems and possible solutions concerning the eventual commercialization of this integrated flue gas treatment system.

Foster Wheeler is presently marketing the IHI Selective Catalytic Reduction Process in the United States.

#### TEST OBJECTIVE

The individual techniques to treat air pollution emissions from coal-fired boilers, such as  $\mathrm{NO}_{\mathrm{X}}$ ,  $\mathrm{SO}_{\mathrm{X}}$ , and dust, have been well established. However, these tests were aimed at integrating these techniques into a total system, including waste water treatment. The following discussion is concerned with various problems and their solutions regarding such an integrated flue gas treatment system.

The major test objectives were as follows:

- 1. Determining DeNO, catalyst life
- 2. Preventing the catalyst layer from plugging with ash
- 3. Preventing the abrasive wear on the catalyst due to ash
- 4. Detecting ways to prevent plugging of the gas air heater (GAH)
- 5. Developing of a low SO3 conversion ratio catalyst
- 6. Determining dust collecting efficiencies of a high temperature electrostatic precipitator, bag filter, and a wet type electrostatic precipitator
- 7. Measuring characteristics of the desulfurization system, such as efficiency of removal, effect of dust, quality of gypsum, method of waste water treatment, and ways to prevent plugging of the reheat system.

#### TEST SYSTEM

Two  $\mathrm{DeNO}_{\mathrm{X}}$  systems were selected as the best available for this testing. The first was chosen to handle the high dust condition directly from the boiler and located ahead of the air heater and cold ESP. The other  $\mathrm{DeNO}_{\mathrm{X}}$  system was installed between the hot ESP and the air heater to handle the low dust condition. (Reference Figure 1 on the following page.) In this test, the flue gas reheater was a standard design, and the FGD system used a common wet limestone process.

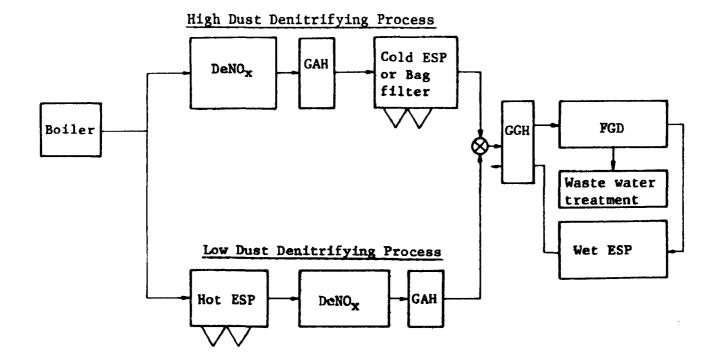


FIGURE 1

# SECTION 4 SPECIFICATIONS OF TEST EQUIPMENT

The specifications of the major test equipment used for this program are shown in Table I below, and Figure 2 on following page shows the Schematic Flow Diagram.

TABLE I

Test Equipment	Specifications			
Denitrifi- cation device	Denitrifying method	Selective catalytic reduction process using NH <sub>3</sub>		
device	Type of reactor	Vertical downward flow type fixed bed		
	Shape of catalyst	Square honeycomb type catalyst		
	Capacity	1000 NM $^3$ /h x 2 for high dust 1000 NM $^3$ /h x 2 for low dust		
		High Dust	Low Dust	
Air heater (GAH)	Type	(Regenerative type)	(Generative type)	
		High temperature element: DU	NF-Single layer	
		Medium and low temperature element: NF		
	Capacity	2000 NM <sup>3</sup> /h x 1	2,000 NM <sup>3</sup> /h x 1	

TABLE I (cont'd.)

Test Equipment		Specifications			
		High temper- ature electro- static precipi- tator	Bag filter	Wet electro- static precipi- tator	
Dust collector	Туре	Dry horizontal gas flow type	Reverse washing bottom inlet type	Wet type flat plate	
	Capacity	2000 NM <sup>3</sup> /h	2000 NM <sup>3</sup> /h	1000 NM <sup>3</sup> /h	
Reheater Type (Generative type)			)		
(GGH)	Capacity	1000 NM <sup>3</sup> /h x 1			
Desulfuri- zation device	Туре	Limestone wet process FGD system (Both separation and mixing of ash are possible)			
device	Capacity	$1000 \text{ NM}^3/\text{h} \times 1$			
		Dust removing tower, absorption tower, mist eliminator, gypsum recovery process, waste water treatment equipment			

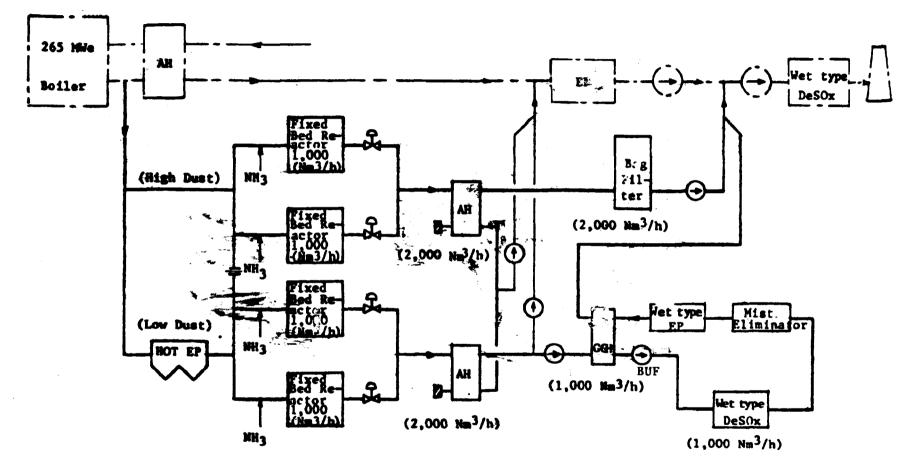


Fig. 2 Schematic Test flow diagram of integrated flue gas treatment systems

#### TEST SCHEDULE

This test and research project was initiated as a result of early  $\mathrm{DeNO}_{\mathbf{x}}$  testing that started January 1968. It soon became apparent that there was a need to combine  $\mathrm{NO}_{\mathbf{x}}$ ,  $\mathrm{SO}_{\mathbf{x}}$ , and dust removal in one integrated system and that this combined system looked viable based on the early individual testing. Table II shows the schedule of the testing from 1977 through 1980.

TABLE II

				·
	1977	1978	1979	1980
	Denitrifica- tion device High Temperature ESP, GAH	Desulf zation device	filter	
DeNO <sub>x</sub> test		DeNO <sub>X</sub> test (High/Low dust)  Test to check the construction of the reactor  Performance test  Test to check pitch of honeycomb mesh  Development of low SO <sub>3</sub> conversion ratio catalyst  Measures to prevent plugging of GAH  Measures to prevent plugging and wear of catalyst		
Integrated flue gas treatment test			(Low dust dust syst  Desulfuri  Measures to ging of GAH  Long-term plugging	of catalyst system, High em)  zing test prevent plug-

#### SECTION 6

## DISCUSSION OF TEST RESULTS

The results of this integrated flue gas treatment test can be summarized as follows:

## DENITRIFICATION EQUIPMENT

### Life Test of Catalysts

Tests to determine the life of the catalyst are now in progress. Some catalysts have passed 9,000 hours during low dust conditions and 7,000 hours for high dust conditions. No significant deterioration was observed under either of these conditions, and the catalysts have shown that a stable  $\text{DeNO}_{\chi}$  efficiency of greater than 80% is possible. These tests will be continued in order to test their performance over a longer period.

Figures 3 and 4 on the following pages show the life test results of catalyst at low and high dust conditions.

# Plugging of Catalyst Layer

At the high dust conditions, no deposits or ash accumulation were observed on the catalyst. While at the low dust conditions, the ash has a smaller grain size and is more prone to sticking to the catalyst without the benefit of having the larger particles perform a self-cleaning action.

As a result of this non-self-cleaning action, partial deposits or accumulations of ash were observed in this case. It was shown, however, that the plugging of the catalyst layer can be prevented by considering such factors as the flue gas flowrate, mesh size of honeycomb catalysts, catalyst support construction, and adoption of a catalyst layer using seamless catalyst in the flow direction.

Pig. 3 Results of test for the concentration of high dust

Place of test Catalyst		Isogo Power Plant  Square mesh honeycomb type catalyst (A-III type)		
8	Concentration of S	Ox		300 ∿ 900
피	Reaction temperatu	re	*c	350
Tours	NH3/NOx	i	Mol ratio	0.9
ပို	SV value		h-1	3,300

(800 ~ 900ppm after Feb. 14, 1980)

Concentration of high SOx
(SO2, SO3 injected)

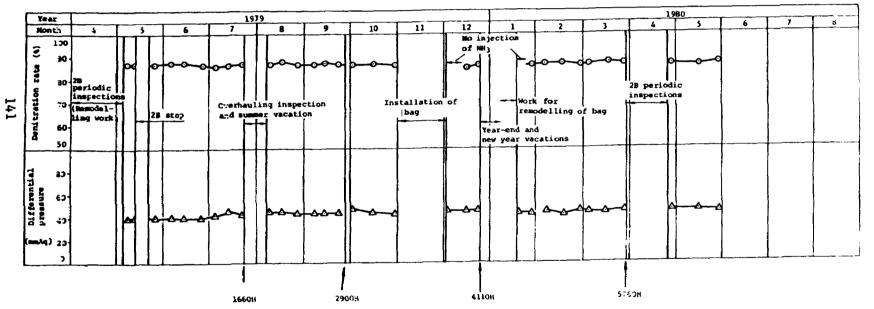
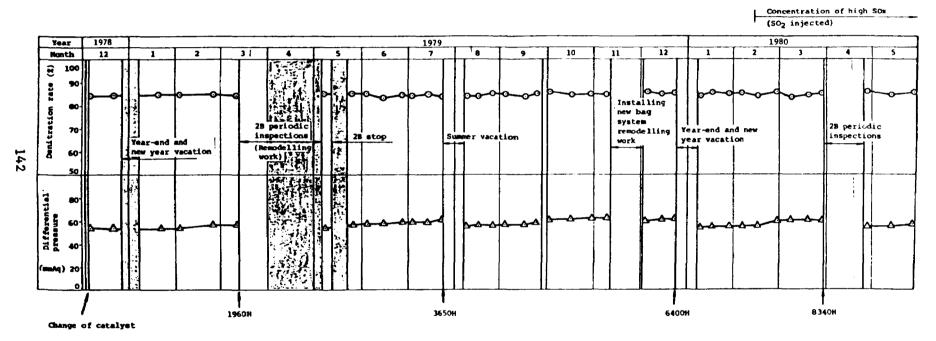


Fig. 4 Results of test for the concentration of low dust

Catalyst S		Isogo Power Plant		
			Square mesh honeycomb catalyst (A-II type)	
	Concentration of	dust	g/Nm³	0.1~0.2
6	Concentration of	SOx		300 ∼ 900
dit	Reaction tempera	ture	°c	350 °C
Š	HH <sub>3</sub> /NOx		Mol ratio	0.9
•	SV value		h-1	2,800

(800  $\sim$  900ppm after Peb. 14, 1980)



### Erosion of Catalyst

The most significant factor which affects the life of the catalyst in coal-fired applications is the abrasive wear caused by the ash in the combustion gas. However, these tests have shown that this wear can be effectively controlled by designing for the following conditions: selecting an appropriate flowrate for gas, choosing a catalyst having a high resistance to wear, and eliminating the seams in the catalyst in the direction of gas flow. The results of the test for the 7,000 hr catalyst life test showed no significant wear during inspection when the above design considerations were taken.

During this series of testing, an accelerated wear test was conducted over a relatively short period of time, where the flowrate of gas and the concentration of dust were extremely high when compared with those at normal conditions.

The result of this test showed the abrasive wear of the catalyst to be calculated as 0.1mm/year, indicating that the catalyst can be used effectively for several years at normal conditions. At low dust conditions, the concentration is 1/100 of that under high dust conditions and the grain size is smaller, thereby eliminating the concern about abrasive wear of the catalyst.

## Development of Low SO3 Conversion Ratio Catalyst

The rate of conversion at which parts of SO<sub>2</sub> contained in the gas are converted into SO<sub>3</sub> in the catalyst layer affects the equipment on the downstream side through corrosion or plugging. However, it has been shown through research that a catalyst having an extremely low conversion rate of SO<sub>2</sub> to SO<sub>3</sub> of less than 1% can be manufactured.

### PLUGGING OF GAS AIR HEATER (GAH) ELEMENTS

This plugging, caused by an  $NH_3/SO_3$  based product consisting of unreacted  $NH_3$  from the  $DeNO_x$  process and  $SO_3$  contained in the flue gas, is one of the problems that emanate from the operation of the  $DeNO_x$  process. However, this problem can be solved by considering the design of the heater element, soot blowing design, and frequency of soot blowing.

Where there is a high dust concentration in the flue gas, a large cleaning effect from the dust can be expected so that the GAH can be normally operated by using the soot blower several times a day.

In the case of a low dust concentration flue gas, a cleaning effect from the dust cannot be expected, thus creating a greater possibility for the heater elements plugging. However, even for this low dust condition, plugging of the elements can be prevented by designing the element shape to produce an improved overall soot blowing effect. For example, according to the results of a test conducted for over 3,000 hours, close inspection showed no significant plugging of heater elements. This test is still in progress in order to check whether it is possible to operate continuously over a long period of time using these design improvements.

Additional test results showed no significant corrosion or decrease in the wall thickness of the heater element for either the low or high dust conditions.

Figure 5 shows the results of pressure loss testing for the GAH for both high and low dust conditions.

#### DUST COLLECTOR

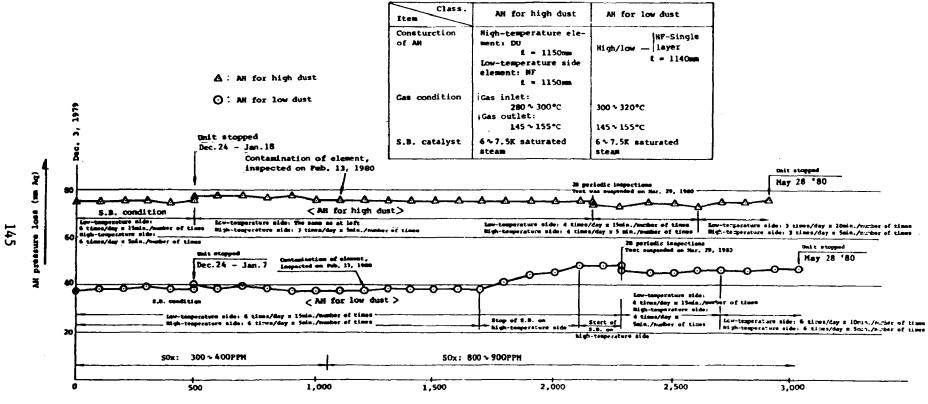
## High Temperature Electrostatic Precipitator (Hot ESP)

Currently there are various opinions as to the efficiency of the hot ESP. According to the results of these tests, the collection efficiency of a hot ESP proved highly effective on dust in the flue gas from coal containing 0.6% sulfur.

## Wet Type Electrostatic Precipitator (Wet ESP)

At present, the concentration of dust at the outlet of a desulfurization system has to be about 20 mg/NM<sup>3</sup> in Japan. However, in order to meet more strict requirements in the future, a wet ESP was installed on the outlet side of the desulfurization device used for this testing. The next test series showed the dust collection efficiency of wet ESP to be about 93%. These results show that the concentration of dust at the outlet of the FGD unit can be maintained under 10 mg/NM<sup>3</sup> when this wet ESP is used, even if the scale effect is taken into consideration.

Pig. 5 | Pressure loss in GAH



Operating hours

### Bag Filter

A Sumitomo bag filter is used with the high dust  $DeNO_X$  process as the low temperature dust collector. This same bag filter was used successfully in a joint development project by Electric Power Development Co., Ltd., Sumitomo Heavy Industry Co., Ltd., and IHI. The results of this test confirmed the expected efficiency of the bag filter, as it succeeded in maintaining the concentration of dust below 10 mg/NM $^3$  with a pressure drop of 150 mm W.G. for the case where the flue gas had passed through the  $DeNO_X$  process. A long-term test to determine the life span of the bag filter material is now in progress.

#### FGD PROCESS

## Desulfurization Efficiency

When a boiler is fired with several kinds of coal, the  $SO_2$  concentration of the flue gas may vary over a wide range.

In this test it was shown that a satisfactory desulfurization efficiciency of over 96% can be obtained for  $SO_2$  concentrations ranging from 300 to 1,000 ppm at the inlet.

# Dust Removing Efficiency

When an FGD unit is installed as shown in Figure 1, the dust at the inlet is affected by the unreacted ammonium from the  $\mathrm{DeNO}_{\mathrm{X}}$  process, dust contained in the flue gas, and other various gases such as fluorine, chlorine, etc.

According to the results of this series of tests, the effects of such dust and gases are relatively small. For instance, the concentration of dust at the outlet of the FGD system is less than 20/NM<sup>3</sup>, thus showing that this FGD system has sufficient dust removing capability.

## Quality of Gypsum from the FGD Process

In the case of coal-fired boilers, traces of various impurities contained in the coal accumulate in the circulating scrubbing liquid of the FGD system after a period of time. This test showed that some of these impurities affect the gypsum crystallization and also control of pH in the scrubbing liquid. However, it was also shown that such problems can be solved

and that gypsum of the desired quality can be obtained by taking appropriate measures such as injection of an additive. Furthermore, it was confirmed that an NH3/SO3 based product coming from the upstream side of the desulfurizing system does not significantly affect the quality of gypsum and that to obtain gypsum of high quality, the ash-separation method is more effective than the ash-mixed method.

## Plugging of GGH Element

The GGH element is subject to plugging by mist deposits from the FGD process; but by operating the soot blower more frequently, this problem is not severe. During these tests, no significant corrosion or decrease in the thickness of the element wall was noticed.

### FGD Booster Fan

SUS material is used for the blades, and a rubber lined casing is used for corrosion protection. The booster fan has provided adequate and stable performance during these tests.

### Waste Water Treatment System

Traces of various substances such as heavy metals, fluorine, chlorine, etc., contained in the coal and unreacted ammonium from the  $DeNO_X$  process will accumulate in the circulating scrubbing liquid of the FGD system. Normally this liquid is blown down to protect the materials of construction from corrosion. This waste water stream is normally sent to an integrated waste water treatment system. A research program is now being conducted to develop a more economical waste water treatment system for FGD applications.

SECTION 7

EVALUATION OF TEST RESULTS AND CONSIDERATIONS FOR COMMERCIAL PLANTS

	Evaluation	Evaluation of	Test Results	Considerations for
	Item	High Dust Denitrifying Process	Low Dust Denitrifying Process	Commercial Plants
tability	Process	Boiler DeNO <sub>X</sub> GAH Bag filter DeSO <sub>X</sub>	Boiler Hot ESP DeNO <sub>X</sub> GAH DeSO <sub>X</sub>	In setting up the planning for actual plants, it is an important point to determine the process arrangement which depends on whether a low temperature or a high temperature dust collecting method is selected. The selection of the method largely affects the economy and general arrangement of the system and can be determined by considering the kind of coal used.
Adaptab	Denitrification Function	<ul> <li>This process will not cause any serious plugging of the catalyst layer or the GAH and is useful from the viewpoint of the denitrification function.</li> <li>As for wear, partial erosion was observed where there was a drift in the gas flow, but such phenomenon can be prevented by correcting the gas flow on the upstream side of the catalyst layer.</li> </ul>	• In this process, the catalyst layer, especially the element of the GAH, is subject to more plugging compared with the high dust process, but this process has proved its effect in the DeNO <sub>X</sub> function if proper consideration is given to the construction of the catalyst layer and the form of the GAH element and if soot blowis used appropriately.	• When applied to the actual system, unlike the case of the test plant, considerable changes will be expected in the gas flow, causing partial erosion of the catalyst at the upstream side. To minimize the erosion effect, the method of regulatthe gas flow must be considered carefully.
	-			To solve the plugging problem that occurred in the low dust AH, it is necessary not only to select an appropriate value for the space velocity but also to follow the instructions given in the column at the left. Also, it is desirable to apply a close control on the NHJ/NO <sub>x</sub> mol ratio to restrict unreacted NH3 for partial load and to adopt a low conversion ratio for SO3 for the catalyst.

SECTION 7 (cont'd.)

EVALUATION OF TEST RESULTS AND CONSIDERATIONS FOR COMMERCIAL PLANTS (cont'd.)

	Evaluation	Evaluation	of Test Results	Considerations for
	Item	High Dust Denitrifying Process	Low Dust Denitrifying Process	Commercial Plants
Adaptability	Dust Collecting Function	• Unlike the low dust denitrification method, the NH <sub>3</sub> -SO <sub>3</sub> -based products from the GAH and ash containing NH <sub>3</sub> are collected almost completely by the dust collector installed on the downstream side. Therefore, the method of treating NH <sub>3</sub> ash becomes one of the problems to be considered. Thus, IHI is now conducting research on this problem in parallel with research on the effective use of NH <sub>3</sub> ash.	and will not adversely affect the down- stream side of the system. Since it does	<ul> <li>When using an ESP as the low temperature dust collector, its efficiency should be considered with the integrated system's dust collecting efficiency, including the dust removing efficiency of the wet type desulfurization equipment.</li> </ul>
	Desulfurization Function	• Tests on the desulfurization function in conjunction with the high dust denitrification process have not been conducted yet; however, the NH3-S03-based product and mist produced in GAH are collected almost completely by the dust collector on the downstream side of this process. Therefore, this process is more advantageous than the low dust denitrification process as far as the treatment of waste water and the removal of dust are concerned. Future testing will check treatment of waste water, integrated dust collecting efficiency, the requirement for a wet ESP, and the effect on the bag filter caused by the NH3-S03-based product.	treatment system with a conventional desulfurizing system without a DeNO <sub>X</sub> section, the problem of an NH3-SO3-based product arises from unreacted NH3 entering the desulfurizing system and accumulating in the circulating liquid. This problem is solved by removing the ammonius through the waste water teatment system. Testing has shown that this process has an excellent desulfurizing efficiency and dust removing capability.	The desulfurization process is conventional, but it is necessary to develop more economical techniques to treat chemical oxygen demand and to eliminate formation nitrogen from unreacted NH3.

## SECTION 8

### CONCLUSION

This paper is a summary of the results of testing an integrated flue gas treatment system for a coal-fired boiler. Through these tests and associated research, it has been shown that the denitrification, dust collecting, and desulfurizing processes which have been tested are effective and that they can be incorporated into a full-scale integrated flue gas treatment system. These tests have provided the confidence that both the high and low dust  $\mathrm{DeNO}_{\mathrm{X}}$  systems tested will be put into commercial use in the near future.

This test and research program will be continued in order to develop a more reliable integrated flue gas treatment system.

# SECTION 9

## APPENDIX

The Appendix Figures A-1, A-2, A-3, and A-4 show photographs of the denitrification, bag filter, and desulfurization test equipment, respectively.

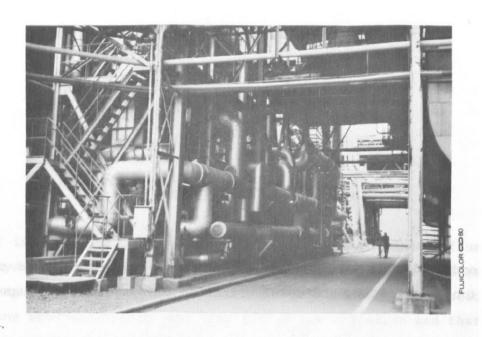
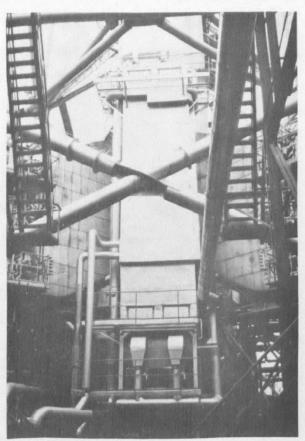


Figure A-1 Denitrification Test Equipment



FUJICOLOR COD 80

Figure A-2 Bag Filter

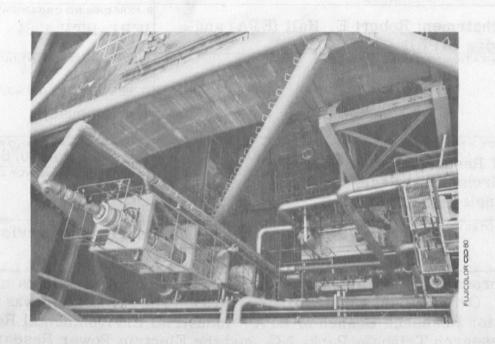


Figure A-3 Desulfurization Test Equipment

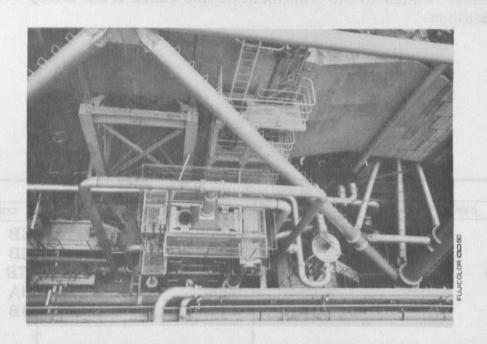


Figure A-4 Desulfurization Test Equipment

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 $^{15. \text{ SUPPLEMENTARY NOTES}}$  EPA-600/7-79-050a through -050e describe the previous symposium.

The proceedings document the approximately 50 presentations made during the symposium, October 6-9, 1980, in Denver, CO. The symposium was sponsored by the Combustion Research Branch of EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, and the Electric Power Research Institute (EPRI), Palo Alto, CA. Main topics included utility boiler field tests; NOx flue gas treatment; advanced combustion processes; environmental assessments; industrial, commercial, and residential combustion sources; and fundamental combustion research. This volume relates to the treatment of flue gases from utility boilers to control NOx emissions.

17.	KEY WORDS	S AND DOCUMENT ANALYSIS		
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