



## Project Summary

# Hitachi Zosen NO<sub>x</sub> Flue Gas Treatment Process: Volume 1. Pilot Plant Evaluation

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The EPA/Hitachi Zosen pilot plant project — evaluating selective catalytic reduction (SCR) of NO<sub>x</sub> on a coal-fired source — operated for a year and a half. The objective of the contract, to operate at 90% NO<sub>x</sub> removal over a 90-day period, was exceeded. The 0.5 MW pilot plant was installed on a slipstream from Georgia Power Co.'s Plant Mitchell near Albany, GA.

A newly developed catalyst, NOXNON 600, was successfully applied and demonstrated 90% NO<sub>x</sub> removal efficiency for over 5600 hours. Tests to operate the catalyst outside of design specifications showed good tolerance to adverse situations.

The pilot plant project was the first demonstration and evaluation of NO<sub>x</sub> SCR technology on a coal-fired source in the U.S. Project results indicate that the process may be usable as a NO<sub>x</sub> control option; however, some technical concerns remain before the technology can be considered commercially available and demonstrated for coal-fired sources in the U.S.

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

### Introduction

As part of the effort to assess technology for control of nitrogen oxide (NO<sub>x</sub>) emissions, EPA sponsored the design, construction, and testing of a pilot-scale unit (0.5 MW equivalent) which demonstrated the operation of Hitachi Zosen's process on flue gas from a coal-fired boiler. This report gives details of the Hitachi Zosen process, designed to limit NO<sub>x</sub> emissions from coal-fired steam generators, and results of the demonstration program.

This flue gas treatment (FGT) process uses selective catalytic reduction (SCR) of NO<sub>x</sub> with ammonia which can achieve over 90% reduction in NO<sub>x</sub> emissions.

Strict air pollution laws in Japan led to the construction of several full-scale systems for the removal of NO<sub>x</sub> from flue gas. Hitachi Zosen took the lead in this area with the construction of the first large commercial unit in 1974. Hitachi Zosen now has nine commercial plants in operation.

To further the goal of controlling stationary source NO<sub>x</sub> emissions, EPA sought to enhance the reliability and effectiveness of technology to reduce these emissions. One aspect of EPA's involvement includes sponsoring programs designed to demonstrate this technology. Because combustion modifications can achieve only limited

reduction in NO<sub>x</sub> emissions, some emphasis has been placed on developing FGT processes. In particular, SCR technology appears to be a very promising method of reducing stationary source NO<sub>x</sub> emissions by over 90%. Consequently, the EPA has acted to demonstrate some of the more advanced SCR systems.

EPA initiated programs to demonstrate two SCR processes on a 0.5 MW scale. The processes are:

The Shell/UOP Simultaneous SO<sub>2</sub>/NO<sub>x</sub> Removal System.

The Hitachi Zosen NO<sub>x</sub> Removal System.

The EPA-sponsored programs demonstrated these processes on flue gas from coal-fired boilers. The demonstration programs were expected to answer many of the questions which remain concerning the application of SCR technology. In addition, these programs should provide an improved basis for estimating the costs of applying SCR technology.

### Process Description

One method of removing NO<sub>x</sub> from flue gas is using SCR. Ammonia is the most practical reductant because it reacts selectively and quantitatively with NO<sub>x</sub> to produce innocuous nitrogen and water. Ammonia is available, relatively inexpensive, safe to handle, and easy to store.

Ammonia will react with NO<sub>x</sub> without a catalyst in a narrow temperature range at around 1000°C. By using a suitable catalyst, the required temperature can be lowered to a more practical range of 300-450°C.

Carrier-based catalysts have been developed and used by Hitachi Zosen in several plants in Japan. These are pellets of alumina, silica, titania, or other materials into which the catalyst is impregnated.

With dust-containing gases, carrier-based catalysts are not practical because the bed of catalyst will be plugged by the particulate matter in a short time. However, parallel-flow honeycomb catalysts have been developed and applied to the removal of NO<sub>x</sub> from dusty gases. These catalysts permit lower operating costs because of their low pressure drops.

The honeycomb catalysts developed by Hitachi Zosen have been extensively tested in several pilot plants and were used in this test program. They are termed NOXNON 500 and NOXNON 600.

### Overall Process Scheme

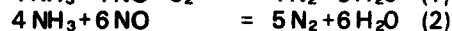
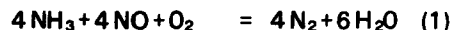
The process consists of the injection of a small amount of ammonia into the flue gas and passage of the flue gas over a catalyst. The ammonia reacts almost entirely with the NO<sub>x</sub> in the flue gas to form small quantities of nitrogen gas and water vapor, both of which are normal constituents of the atmosphere and are environmentally acceptable.

Ammonia is injected into the flue gas from a boiler through the economizer and the air preheater (see Figure 1). At this point the gas temperature is about 400°C which is suitable for the catalytic reduction of NO<sub>x</sub>. This gas enters the reactor, passes over the catalyst, and then reactions proceed. The flue gas then passes through the usual air heater, particulate control, SO<sub>2</sub> control, fan, and stack. The ammonia for the reaction is vaporized with steam and is diluted with air (or steam) before injection into the duct. If the flue gas temperature is too low for optimum

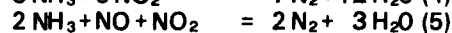
removal efficiencies, additional heat can be added by auxiliary burners; or economizer bypass.

### Chemistry

The exact relationships, between ammonia and NO<sub>x</sub> are not completely understood. However, certain reactions are probably involved which may or may not include oxygen. Virtually all of the NO<sub>x</sub> in combustion gas is present as NO, so the following equations are only for NO reactions:



If NO<sub>2</sub> is also present, the following reactions may also occur:



The reactions of ammonia with NO<sub>x</sub> over the catalyst occur below 300°C. Without a catalyst the reaction will only occur in a narrow temperature range of

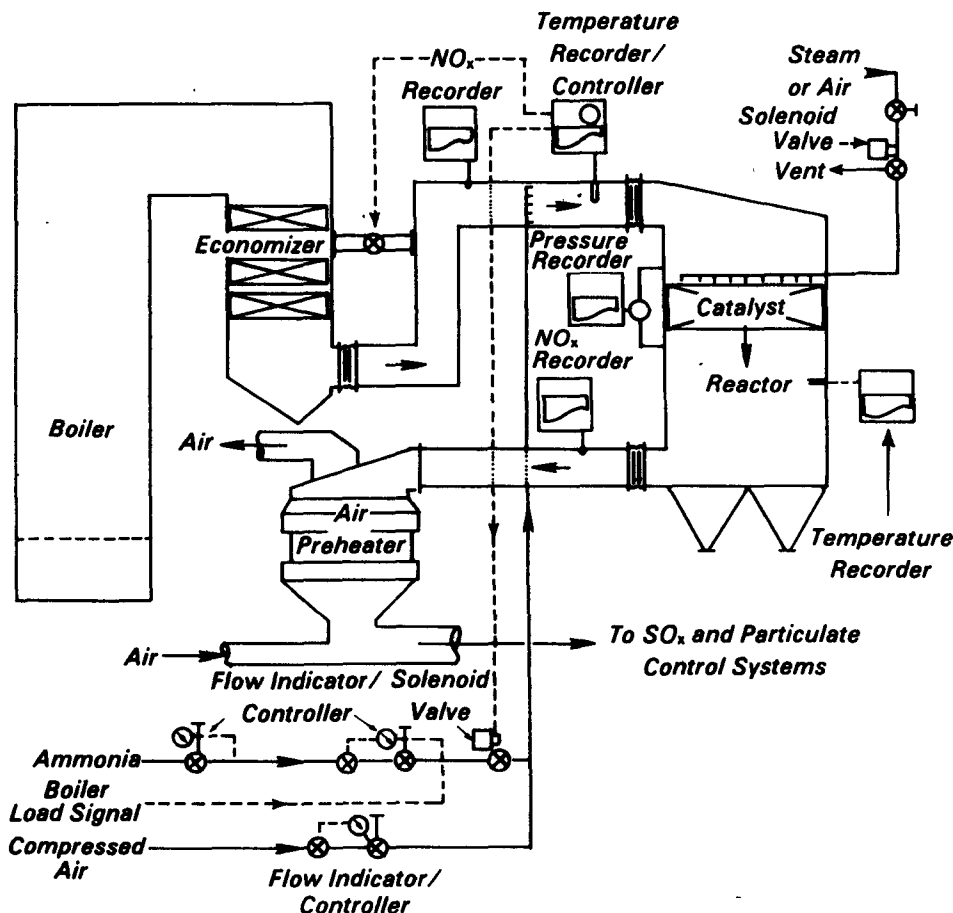


Figure 1. Typical commercial system flow diagram.

950-1000°C. Below this temperature the reaction rate is very low.

### Factors Affecting the Catalytic Reduction of NO<sub>x</sub>

In any chemical reaction, there are factors which can influence the rate and extent of the reaction. These factors include the reaction temperature, concentrations of reactant species, and other parameters specific to the reaction system. The following discussion briefly examines the major influences on the catalytic reduction of NO<sub>x</sub> by ammonia.

The most critical variables which affect the degree of NO<sub>x</sub> removal are the mole ratio of ammonia to NO<sub>x</sub>, the flue gas flow rate, and the reactor temperature.

### Ammonia Emissions

A small amount of ammonia will invariably pass through the reactor and exit with the flue gas due to incomplete reactions between the NO<sub>x</sub> and NH<sub>3</sub>. Ammonia slippage (unreacted ammonia) is of concern, and efforts are normally made to control slippage to levels of 10-20 ppm. A high slippage of ammonia can be considered a pollutant and, in some cases in Japan, agreements with local governments have set allowable levels at around 10 ppm. Another reason for the concern with ammonia slippage is that flue gas desulfurization following the denitrification system could absorb any ammonia in the flue gas and ammonium salts can build up. Bleeding of the ammonium salts from the system can cause water pollution control problems in certain cases. The degree of slippage is a function of the NH<sub>3</sub>/NO<sub>x</sub> mole ratio, the area velocity, and the temperature at which the reaction occurs. For commercial application to coal-fired utility boilers, ammonia slippage would be expected to be less than 5 ppm at an NO<sub>x</sub> removal efficiency of 80%, and less than 10 ppm at an NO<sub>x</sub> removal efficiency of 90%.

### Ammonium Sulfate/Bisulfate

The presence of sulfur trioxide (SO<sub>3</sub>) in the flue gas can lead to a reaction with ammonia to form ammonium sulfate and ammonium bisulfate. When burning heavy fuel oil, approximately 2-4% of the SO<sub>x</sub> in the flue gas are present as SO<sub>3</sub>. For coal combustion, SO<sub>3</sub> accounts for approximately 1% of the

total SO<sub>x</sub>. The reaction of ammonia with this SO<sub>3</sub> will not occur above about 300°C (572°F). At very low concentrations, a temperature of around 200°C (392°F) might be sufficient to avoid this formation.

To avoid such deposits it is necessary that the temperature in the reactor be maintained at a minimum of 320°C (608°F) as long as ammonia is injected into the reactor.

The formation of bisulfate can be minimized to a large extent by maintaining a low ammonia level in the flue gas exiting the reactor. The presence of fly ash also reduces the problem both by scouring the deposits from surfaces and by providing surfaces on which the deposits will form instead of on metallic surfaces.

### Application of Process to Boilers

A schematic flow diagram of the process as applied to a boiler is shown in Figure 1. Flue gas leaving the economizer at a temperature of about 390°C (734°F) is first mixed with ammonia in quantities needed to meet NO<sub>x</sub> removal requirements. The gas is then passed through the fixed-bed catalyst reactor. No dust removal is required prior to the catalyst because of the non-clogging design of the catalyst geometry. NO<sub>x</sub> is reduced by ammonia to innocuous nitrogen and water. The denitrified gas then resumes its passage through the normal boiler train: air preheater, dust collection, etc.

Ammonia required for the process is first vaporized with steam and then diluted with either air or steam to aid in the distribution of ammonia into the flue gas.

The reactor is supplied with retractable soot blowers in which blasts of steam or hot air are occasionally applied to the catalyst bed to remove deposits of dust which may adhere to the surface of the catalyst.

### Catalyst Description

A particularly effective physical design of the catalyst structure has been developed by Hitachi Zosen. This structure is honeycomb shaped as shown in Figure 2. Overall, the catalyst has four characteristics:

- The structure is a thin plate honeycomb.
- Due to substantially reduced pressure drop across the catalyst layer, operating power costs are

much lower than with conventional catalysts.

- A straight gas flow path prevents dust clogging.
- It is applicable for gases with high SO<sub>2</sub> concentrations.

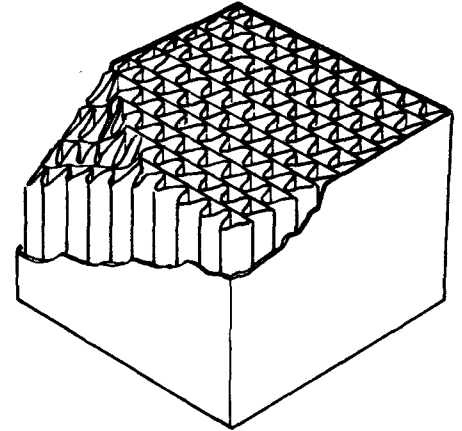


Figure 2. Configuration of NOXNON 500 or 600 series catalyst.

As a result, in treating high-temperature gases with high SO<sub>x</sub> and dust concentrations (such as coal-fired boiler flue gas), the NO<sub>x</sub> removal reactor can be installed immediately downstream of the economizer. In addition, dust elimination or other pretreatment is unnecessary and, with the low pressure drop, operating costs are low.

The honeycomb catalysts are referred to by Hitachi Zosen as NOXNON 500 or NOXNON 600. These catalysts are manufactured as plates and are fabricated so as to form parallel flow gas passages which provide excellent contact with the flue gas with minimum impingement of fly ash on the catalyst surface.

The catalyst plates are arranged in a steel frame box supported by retainers. A standard module is 1 meter long, 1 meter wide, and 0.5 meters deep. The catalyst is activated after the corrugated catalyst assembly is made. In the NOXNON 500 catalyst, thin stainless steel plates are used. The newer NOXNON 600 catalyst, instead of plates, uses a stainless steel wire mesh as a base metal to give mechanical strength upon which catalytic components are cemented. 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.

The catalyst has an expected life of about 2 years. Regeneration of the catalyst is not needed during the planned catalyst life. At the end of the useful life of the catalyst, it would be removed from the reactor vessel and scrapped. The catalyst does not contain any hazardous materials and can be disposed of by recovering the metal in the catalyst or by disposing of the spent catalyst as industrial waste. No special precautions are required in the disposal process.

### Reactor Design

Reactor vessels housing the catalyst are carbon steel and are divided into chambers each having its own fly ash hopper (see Figure 3). Flue gas flows downward in the reactor. Flue gas usually enters at the top of the reactor at one side, passes through the gas distribution grid and catalyst bed, and exits the opposite side of the reactor at the bottom. Fly ash that drops out of the flue gas stream is collected in the bottom fly ash hoppers and is disposed of periodically.

### Demonstration Program

A potential market for the denitrification of power plant flue gases may be at coal-fired stations. Limited test work has been done by Hitachi Zosen on flue gas from coal combustion. The EPA-sponsored demonstration at Georgia Power Co.'s Plant Mitchell was an excellent site for testing the Hitachi Zosen process. The power plant burns typically medium sulfur coal with relatively high ash levels. Adverse effects, if any, on the catalyst by this flue gas could be readily evaluated.

The demonstration plant was highly instrumented and provided much useful data to supplement data collection from previous work in Japan. Several parameters were evaluated, and their effects on removal efficiencies and ammonia utilization were checked. The data were collected and evaluated to establish valid characterizations of the process. Long-term tests provided information on the aging tendencies of the catalyst when applied to flue gas from American coal burning boilers. Additionally, the data collected during these tests were useful in establishing operating and

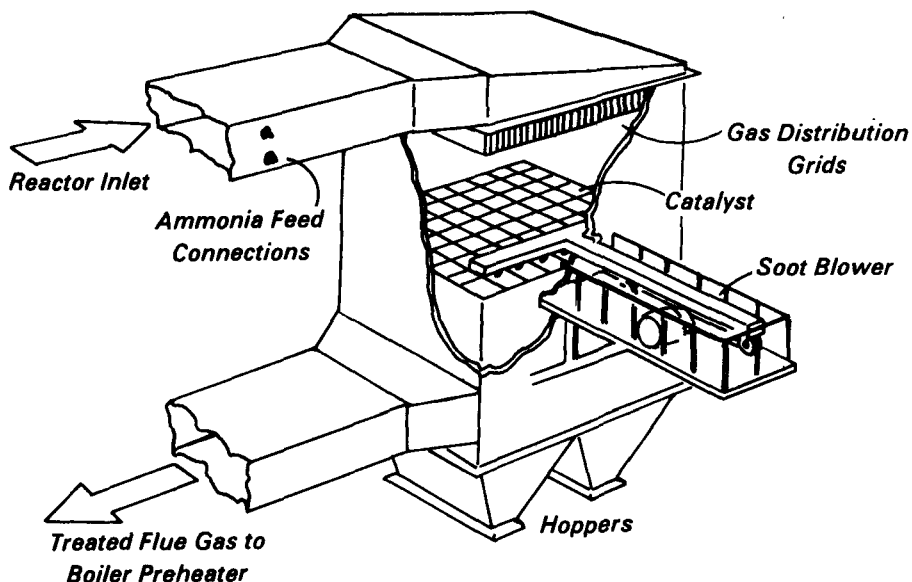


Figure 3. Typical reactor arrangement.

capital cost requirements for commercial installations.

Hitachi Zosen, with Chemico Air Pollution Control Corp. (CAPCC), a Division of Envirotech Corp., as their major subcontractor, provided the test plant and operated the unit. This included detailed engineering, procurement, fabrication, transportation, erection, test operation, and continuous demonstration operation of the pilot plant. The work was in four phases:

Phase I (engineering) started with the basic design package which was designed by Hitachi Zosen in Japan. Based on these designs, CAPCC prepared a detailed design.

Phase II included start-up, debugging, and parametric tests.

Following the successful completion of the system optimization test, Phase III, the pilot plant was to be operated continuously (24 hours/day, 7 days/week) for at least 3 months as Phase IV. The period of continuous operation was to include no less than 75 days of cumulative operation in compliance with contract objectives and guarantees.

Additional tests were conducted as an addition to the original scope of work. Phase V was run to examine the response of the SCR system's performance to transient operating conditions.

### Schedule

The period of performance for completion of the work related to the pilot plant was originally 18 months. The award of the contract was May 1978. The design and specification period, Phase I, took approximately 3 months. Phase II required approximately 9 months for procurement, fabrication, transportation, and erection. These schedules were as originally expected. However, since start-up and debugging required about 2 months, the first tests were not started until August 1, 1979. This first charge of catalyst showed less than optimum results after some 4 months of operation, and it was decided to replace it in December 1979. After some 4 additional months of operation, the NOXNON 500 catalyst again began to provide less than expected results, and a decision was made to install a third charge of catalyst.

The third charge was the new NOXNON 600. This was installed on April 18, 1980, and was used for some 9 months until the plant was shut down on February 2, 1981.

### Description of the Test Plant

The following description of the pilot unit includes summaries of major items of equipment (see Figure 4):

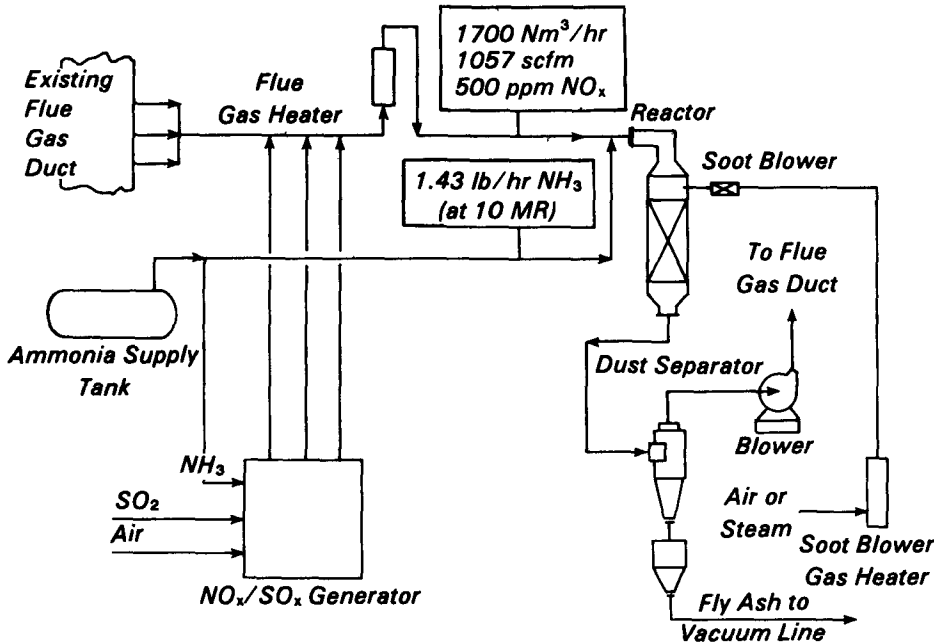


Figure 4. Process flow diagram of pilot plant.

Flue gas to be used as input to the pilot plant was drawn from the boiler duct downstream of the economizer and introduced to the reactor through a 14-in. diameter\* pipeline.

An electric heater was provided in the piping between the flues and the reactor. This flue gas heater was used to control flue gas temperature to the reactor.

Gaseous ammonia was injected to the gas stream after the heater and before the reactor. The gas then flowed down through the reactor in contact with the catalyst. The  $\text{NO}_x$  in the gas reacted with the ammonia to form small quantities of gaseous nitrogen and water, which remained in the gas stream.

Dust settling in the reactor was kept within acceptable bounds, using a soot blowing apparatus with either steam or air.

A blower, downstream of the reactor, overcame the flue gas pressure drop through the pilot plant. A cyclone dust separator, ahead of the boiler, prevented erosion of the blower by dust.

Since it was planned to test the performance of  $\text{NO}_x$ ,  $\text{SO}_2$ , and  $\text{SO}_3$ , and since only fixed concentrations of these

substances were available from the Albany, GA, plant boiler operation,  $\text{SO}_2$  was purchased for addition to the flue gas, and a  $\text{NO}_x$ ,  $\text{SO}_2$ , and  $\text{SO}_3$  generating unit was provided to manufacture these materials for addition to the flue gas. This permitted testing at  $\text{NO}_x$ ,  $\text{SO}_2$ , and  $\text{SO}_3$  levels higher than those available from the boiler.

The operating conditions controlled in the pilot unit are:

- Flue gas flow rate.
- Flue gas temperature.
- The amount of charged ammonia.
- Soot blowing gas pressure and temperature.

### Host Site

The pilot plant was at Unit 3 of Georgia Power Co.'s Plant Mitchell, Albany, GA. This unit has a pulverized-coal-fired Combustion Engineering boiler, initially operated on April 18, 1964, with a 125 MW nameplate rating. The boiler has a rated steam capacity of 1,075,000 lb/hr of steam to the turbine at 1800 psig. The unit is tangentially fired with CE standard tilting burners for steam temperature control. The pilot plant was located outside Unit 3.

### Data Collection

The pilot plant operation was closely monitored by an array of instruments

and analyzers. Continuous analyzers of the latest design measured the  $\text{NO}_x$  levels both entering and leaving the reactor.

Much of the data at the pilot plant were collected in a data logger which frequently scanned the instruments and stored the data. Visual and paper tape displays were provided.

### Relating the Results to a Full-Scale Operation

The test unit included a section of catalyst which had the same depth as that to be used in a full-scale plant. The results could therefore be directly extrapolated to a large plant assuming that the velocity, mole ratio, and temperature remained the same and the gas distribution through the catalyst bed was properly designed.

Ammonia consumption could also be determined from the test results because it is calculated as a mole ratio of ammonia to  $\text{NO}_x$  in the flue gas.

## NOXNON 600 Tests

### Introduction

Primarily because of clogging of the catalyst channels by fly ash due to narrow clearances, the initial and second charges of catalyst did not perform as expected. The installation and testing of NOXNON 600 catalyst having wider channels was proposed by Hitachi Zosen and accepted by EPA.

NOXNON 600 is produced from thin stainless steel wire mesh (as a base metal to give mechanical strength) to which catalytic components are cemented. This technique has been proven in applications on oil- and coal-fired combustion flue gas in Japan.

### Catalyst Performance

Starting from April 22, 1980, the demonstration operation with NOXNON 600 ran for more than 9 months until February 2, 1981. The operation with combustion flue gas from the Unit 3 Boiler of Georgia Power Co.'s Plant Mitchell was for 5620 hours. The program was terminated due to scheduled maintenance of the power plant requiring moving of the pilot plant.

The pilot plant program required achieving  $\text{NO}_x$  removal efficiency of more than 90% continuously for more than 3 months. Afterwards, the project scope was extended and transient tests were included in the scope of the

(\* Readers more accustomed to metric units are asked to use the conversion factors at the end of this summary.

contract along with an extension of the operating period.

Catalyst life tests were run to confirm the expected catalyst life. From April 22, 1980, the pilot plant was operated maintaining NO<sub>x</sub> removal efficiency of more than 90% until the end of October. After October, a nominal 80% NO<sub>x</sub> removal was accepted to decrease ammonia slip as far as possible.

Following the catalyst life test, further testing was carried out to determine the effects of transient conditions on the catalyst and to provide an extended operating time so that at least 5000 hours of operation could be obtained to evaluate the long-term effectiveness of the NOXNON 600.

At various times, tests were run to determine catalyst efficiency.

During the operating time with flue gas, tests were run including catalyst life tests, catalyst performance tests, and transient tests. Controllability and reliability of the entire system were also evaluated at the same time.

Operating conditions were varied for the performance and transient tests. Therefore, when the activity of the catalyst was to be evaluated, operating conditions were set at certain consistent levels each time. These conditions were a flue gas flow rate of 1000 - 1200 scfm, a reactor temperature of 700 - 720°F, and a NH<sub>3</sub>/NO<sub>x</sub> mole ratio of 1.0. Under these conditions, the NO<sub>x</sub> removal efficiency was measured over a period of several hours to determine the condition of the catalyst:

|  | %Removal    |
|--|-------------|
| At the beginning of the operation          | : 90 - 94   |
| At the end of 8/80(2500 hours)             | : 90 - 94   |
| 11/80 (4000 hours)                         | : 90 - 92   |
| Before transient tests 12/80 (4420 hours)  | : 90 - 91.5 |
| During transient tests 1/5/81 (5000 hours) | : 90 - 91   |
| After regeneration 1/27/81 (5500 hours)    | : 91 - 94.5 |

### Operating Variables

#### Mole Ratio

During the test operation, the pilot plant was operated at a selected mole ratio. The control system was designed to automatically provide this mole ratio by using the flue gas flow rate signal and the inlet NO<sub>x</sub> concentration analysis to determine the quantity of NO<sub>x</sub> in the inlet stream. From the inlet NO<sub>x</sub> quantity and the selected mole ratio,

the required ammonia was automatically calculated. This signal was then relayed to the ammonia control system which set the ammonia control valve setting to provide the required ammonia flow.

Figure 5 is a mole ratio curve which shows that 80% removal requires a mole ratio of about 0.85, while 90% removal requires a mole ratio of 1.0.

#### Flue Gas Flow Rate

The NOXNON 600 catalyst for the pilot plant was designed to operate at 1057 scfm, equivalent to an area velocity (A.V.) of 9.6 Nm<sup>3</sup>/m<sup>2</sup>-hr. However, operating at a much higher flow rate of 1650 scfm (an A.V. of 15) provided the desired 90% NO<sub>x</sub> removal efficiency. Therefore, it was anticipated that between the designed flow rate of 1057 scfm and the normal operating flow rate of 1500 scfm, the NO<sub>x</sub> would be unchanged. As seen in Figure 6, the flow rate had little or no effect on NO<sub>x</sub> removal.

#### Influence of SO<sub>x</sub> Concentration on NO<sub>x</sub> Removal Efficiency

Concentrations of SO<sub>x</sub> measured in the pilot plant deviated widely between approximately 500 ppm and 1,500 ppm depending on the variations of sulfur in the coal.

Through the whole operating period, NO<sub>x</sub> removal efficiency was not affected by SO<sub>x</sub> concentrations in the flue gas in this relatively wide range.

#### Operating Temperature

In the pilot plant, the operating temperature was varied between 640°F (338°C) and 780°F (415°C) without affecting NO<sub>x</sub> removal efficiency. Optimum operating temperatures from fundamental experiments in the laboratory are between 572°F (300°C) and 750°F (400°C).

#### NO<sub>x</sub> Concentration

On August 24, 1980, the NO<sub>x</sub> generator was operated to increase the

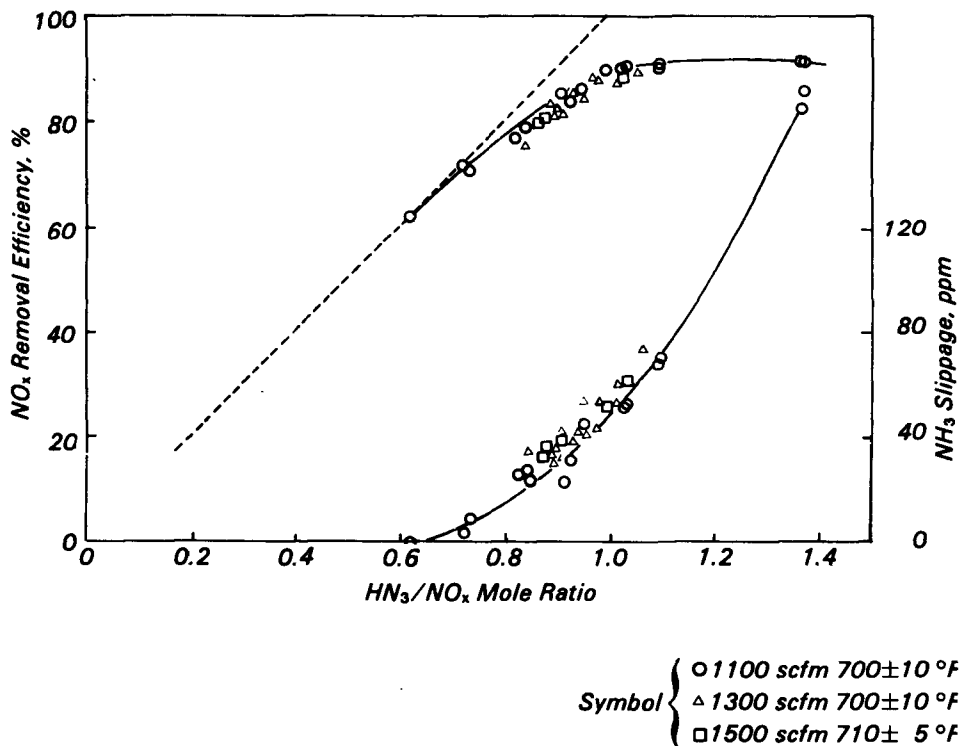


Figure 5. Effect of NH<sub>3</sub>/NO<sub>x</sub> ratio on NO<sub>x</sub> removal efficiency and NH<sub>3</sub> slippage.

concentration of  $\text{NO}_x$  at the inlet of the reactor, and the influence of  $\text{NO}_x$  concentration was investigated.

Between 400 ppm and 900 ppm of inlet  $\text{NO}_x$  concentrations, the concentration of  $\text{NO}_x$  did not influence  $\text{NO}_x$  removal efficiency. This data is consistent with fundamental data.

### Ammonia Slippage

Since a continuous ammonia analyzer was not available during the operation of the pilot plant, ammonia had to be measured by wet analysis.

Figure 5 shows ammonia slippage when mole ratio was varied at operating times of 3200 hours to 4500 hours. General conclusions from these tests include:

- Flue gas flow rate had little effect on ammonia slippage.
- Ammonia slippage seemed to increase slightly with increased operating time.
- Ammonia slippage was apparent even at low mole ratios; e.g., at 0.6.

The reason for this higher than expected slippage may have been clogging by fly ash and fibers of asbestos yarn used to seal a clearance between the catalyst box and reactor. These fibers, along with fly ash between

the first and second catalyst layers, probably reduced the effective catalyst surface and adversely affected the apparent catalyst activity resulting in relatively high ammonia slippage. Even a slight reactivity loss, while producing little loss in  $\text{NO}_x$  removal efficiency, can significantly increase ammonia emissions.

### Oxidation of $\text{SO}_2$ to $\text{SO}_3$

On July 23-25, 1980, operating conditions were maintained constant so that  $\text{SO}_3$  measurements could be obtained at the inlet and outlet of the reactor.

The results indicated an average oxidation rate of 1.8%.

Prior experimental results indicated that, at these conditions, the ratio should have been somewhat lower: 1.0 - 1.5%.

### Transient Tests

After the  $\text{NO}_x$  removal efficiency of more than 90% was demonstrated in the continuous run of 3 months as required in the contract, a decision was made to extend the scope of the contract. This was done to supplement the originally planned operation of the pilot plant to further establish the suitability and reliability of Hitachi

Zosen's catalyst for commercial operations on coal-fired combustion flue gas. Five transient tests were performed.

### Emergency Shut-off of Ammonia Feed

The EPA pilot plant was provided with a trip system for ammonia supply. The purpose of the trip system was to shut off the ammonia feed when temperature in the reactor decreased to 600°F and allow it to introduce ammonia into the system when temperature in the reactor increased and returned to 600°F. The purpose of this test was to confirm the reliability of the trip system.

The results of this test proved that:

- Automatic shut-off and supply of ammonia operated smoothly.
- $\text{NO}_x$  removal efficiency and pressure drop at a temperature of 700°F was constant and unchanged through the three repeated tests.

### Cold Start-up

A commercial boiler normally starts operating after a long shutdown with the reactor and ductwork filled with ambient air. When operation commences, flue gas is introduced into the system as the temperature rises and the flue gas can be relatively cool for a time when in contact with the catalyst. There is a risk of formation and deposition of sulfuric acid mist, ammonium sulfate, and ammonium bisulfate. Previous experience proved that sulfuric acid mist does not deteriorate the catalyst, and ammonium sulfate and ammonium bisulfate can be removed when the temperature rises. The purpose of this test was to prove that cold start-up with flue gas does not affect the  $\text{NO}_x$  removal reaction.

On December 27, 1980, the blower was started and flue gas was introduced into the system directly. The system was heated up with flue gas and put into operation.

Twelve hours after commencing the start-up, operating conditions became stable at the same conditions before cold start-up. The results proved that cold start-up does not influence catalytic performance.

### Boiler Shutdown and Start-up

From time to time a power plant boiler shuts down and starts up, and an  $\text{NO}_x$  removal system must follow such transitions.

The purpose of this test was to confirm the controllability of the  $\text{NO}_x$  removal system during the shutdown

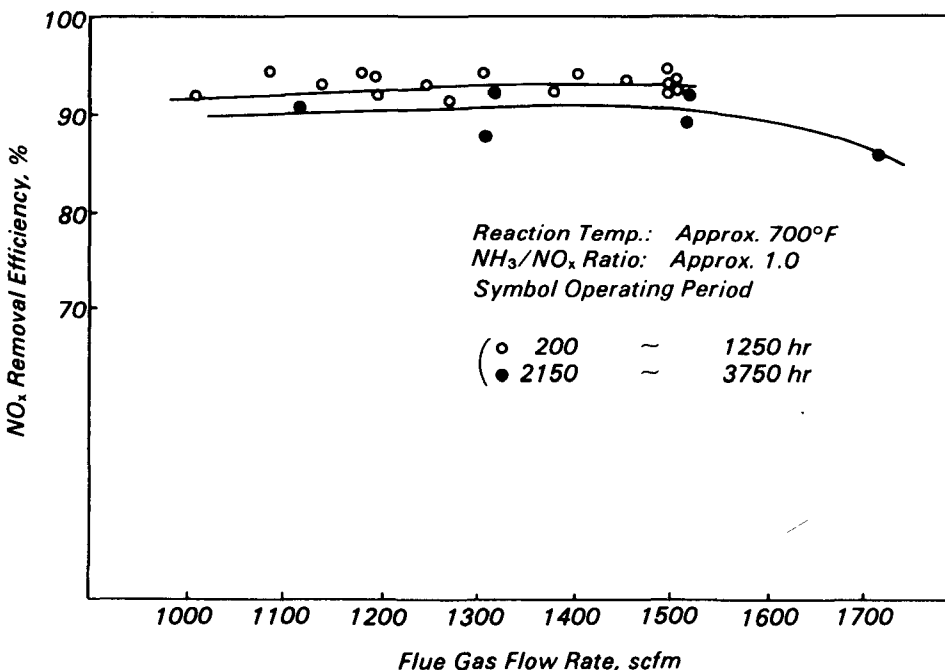


Figure 6. Effect of flue gas flow rate on  $\text{NO}_x$  removal efficiency.

and start-up of the boiler. When the host boiler was shut down for maintenance, this test was executed with the NO<sub>x</sub> removal system shutting down and starting up along with the boiler with no purging of the reactor. The results proved that the system could withstand the transient shutdown and start-up of the boiler.

### Sudden Load Change

A power plant boiler may occasionally change load suddenly, complying with variations of power consumption. The NO<sub>x</sub> removal system should follow these sudden boiler load changes. Two levels of operating conditions were adopted as representative load levels:

- High load: 1,300 scfm, 700°F, 0.8 mole ratio.
- Low load: 900 scfm, 610°F, 0.8 mole ratio.

The above conditions were altered every 2 hours and continued for 24 hours. No serious problem seemed to be caused by these sudden load changes.

### Sootblower Requirements

From the beginning of pilot plant operation with the third charged catalyst, NOXNON 600, the sootblower was operated three times a day, one cycle at a time, to prevent clogging by fly ash. This frequency seemed to be the minimum to prevent clogging; however, there was no experience in operating the pilot plant without the sootblower. Thus, a trial was made to operate without the sootblower.

At a flow rate of 1,100 scfm, 700°F, and 0.8 mole ratio, sootblower operation was halted and the progress of pressure drop increase was observed. Pressure drop appeared unstable (increasing gradually from 1.20 to 1.25 in. H<sub>2</sub>O to 1.35 to 1.40 in. H<sub>2</sub>O in approximately 28 hours) and seemed to continue to increase. The sootblower was then restarted. This test indicated that operation of the sootblower was necessary.

### Changes in Pressure Drop in Relation to NO<sub>x</sub> Removal Efficiency

In the treatment of coal-fired combustion flue gas, it is realized that adhesion and clogging caused by fly ash, along with the formation and deposition of ammonium sulfate and bisulfate, cause problems. In NO<sub>x</sub> removal systems, these problems are first noticed as an increase in pressure drop. Therefore, changes in pressure drop were carefully

studied during the operation of the EPA pilot plant.

Since operating conditions varied from time to time in accordance with the needs of the pilot plant operation and the scope of the contract, changes of pressure drop were evaluated by a ratio of measured value to a calculated value.

Several experiments were carried out. The tendency for pressure drop changes is summarized below.

- When the pilot plant was shut down, for some reason the pressure drop generally increased after the next startup. The increased pressure drop usually continued at this high level although the sootblower was operated three times a day. However, the previous pressure drop was restored after a few days of continuous operation.
- When the flue gas flow rate was changed, the pressure drop increased at times. For example, in November 1980 after some 20 days of steady operation the flow rate was changed from 1300 to 1500 scfm and after 2 days at this higher flow rate the pressure drop suddenly increased and did not return to its original level for several days.
- Whenever Georgia Power's Unit 3 boiler operated the economizer sootblower repeatedly, the pressure drop across the catalyst increased.
- On December 18, 1980, the water tubes of the boiler were washed with pressurized water to remove slag on the tube surface while the boiler was in operation. This abnormal maintenance work caused a serious increase in pressure drop.
- Operation of the sootblower seems to be necessary for coal-fired combustion flue gas. The pilot plant was operated for 28 hours without the sootblower, and the pressure drop increased sharply. After the sootblower was returned to operation, it took 3 days to restore the pressure drop.

### Regeneration of Catalyst

When the catalyst was washed with warm water, NO<sub>x</sub> removal efficiency was restored to its initial efficiency. The pressure drop also seems to have been partially restored. However, the catalyst was partially clogged with asbestos fibers, which is not typical. The effectiveness of water washing for pressure drop restoration, therefore, is uncertain. Also, the waste wash solution contains dissolved metals from the fly ash which

would require water treatment before disposal.

### Abrasion of Catalyst

Among the four blocks of catalyst, only some of the flat plates in the top block showed a loss of catalyst by abrasion. This was probably due to the vibration of the loose plates caused by the impact of the flue gas flow. There was no abrasion in the other blocks. A commercial system would incorporate a tighter catalyst structure to avoid such vibration.

### Clogging

Anticipated clogging of the catalyst by fly ash was prevented by operating the sootblower three times a day. Reducing this frequency of operation may be possible but it was not tested. Apart from the clogging anticipated by fly ash, asbestos fibers unfortunately led to plugging of about 30-35% of the catalyst passages between the top catalyst blocks and the second catalyst blocks. This was caused by asbestos rope used to seal passages between the catalyst box and the reactor shell. This asbestos would not be used in a commercial plant.

### Conclusions

The contract objectives were exceeded. NO<sub>x</sub> removal efficiency of more than 90% was demonstrated during an operating period of about 5000 hours at the designed capacity of 0.5 MW equivalent. Following this period, transient tests were run. These demonstrated that the performance was not adversely affected by such conditions as sudden boiler load changes, cold start-ups, low boiler loads, or by boiler shutdowns and start-ups. The pilot plant operation was terminated after 5620 hours of cumulative operating time only because the host boiler had to undergo major modifications necessitating that the pilot plant be moved.

During operation of the pilot plant, catalysts had to be replaced twice. The first and second charges of catalyst had relatively narrow clearances between the catalyst plates and were clogged by fly ash from the flue gas resulting in increased pressure drops through the catalyst beds. There was also a slight decrease in apparent catalyst activity, although true catalyst activity did not decrease. These catalysts, which were NOXNON 500, had been tested in pilot plants in Japan for application to coal-fired combustion flue gases from



boilers prior to application to the EPA pilot plant. However, clogging caused by fly ash was not experienced even though no sootblower was installed or operated in those pilot plants.

There were obviously significant differences in the character and composition of the fly ash in the U.S. as compared to that in Japan.

Nevertheless, the tests with NOXNON 600 were highly successful. This catalyst has somewhat wider clearances between the catalyst plates, helping to avoid fly ash clogging problems.

Fly ash contained in flue gas varies in its characteristics and behavior relative to its clogging tendency in catalyst beds. This depends on the source and composition of the coal. At present, qualitative measurements to estimate the tendencies to agglomeration and cohesion of fly ash are available through chemical analysis and thermal processing tests. However, more adequate and accurate methods to estimate the behavior of fly ash in catalyst beds will be required. It is expected that this method when further amplified will be useful in selecting adequate linear velocity to prevent clogging and abrasion caused by fly ash, and to determine the necessity and operating conditions of the sootblower. Improvement of catalyst configuration to prevent clogging by fly ash is also expected.

Testing for catalyst regeneration by washing was examined just before the pilot plant was dismantled. The results were very encouraging. The regenerated catalyst exhibited properties of a virgin catalyst. However, due to the limited time available, potential problems related to the catalyst regeneration were not clarified. For example, the method of drying the catalyst and the reactor after regeneration without encountering corrosion or fly ash clinging, and the treatment of waste washing solution should be investigated before commercial application. This area should be studied further since the cost of this technology could be substantially reduced if the catalyst life could be extended by *in situ* regeneration techniques.

Based on tests with NOXNON 600 both at the EPA pilot plant and in Japanese pilot plants, 90% NO<sub>x</sub> removal could be expected at a NH<sub>3</sub>/NO<sub>x</sub> mole ratio of 0.92 - 1.0. Although ammonia slippage from the EPA pilot plant measured about 40 ppm under these conditions, ammonia slippage would be expected to be less than 10 ppm based

on Japanese tests. The differential was probably due to the asbestos clogging in the EPA pilot plant. For 80% NO<sub>x</sub> removal the required mole ratio would be 0.82 - 0.85 with ammonia slippage of 5 ppm or less, again based on tests in Japan.

The pressure drop with NOXNON 600 catalyst in the pilot plant and expected commercially is only 1.0 - 1.4 in. H<sub>2</sub>O. Such low pressure drops required very little power consumption resulting in low operating costs.

The controllability of the pilot plant was satisfactory. However, the control system for commercial plants would differ slightly from the pilot plant control system due mainly to the problems in measuring flue gas flow. The reliability of the system was very good as verified by the high onstream factor achieved. A desirable addition to the control system would be a continuous analyzer to monitor slippage of ammonia.

The NO<sub>x</sub> removal efficiency of this SCR system in commercial applications would be expected to be the same as that experienced in the EPA pilot plant if

the size and configuration of the catalyst, superficial linear velocity of the flue gas across the catalyst, and the temperature of flue gas were the same. Therefore, the data is directly applicable for scaling of commercial systems.

The pilot plant project was the first demonstration and evaluation of NO<sub>x</sub> SCR technology on a coal-fired source in the U.S. The project results indicate that the process may be usable as a NO<sub>x</sub> control option; however, some technical concerns remain before the technology can be considered commercially available and demonstrated for coal-fired sources in the U.S.

### Metric Conversion

Readers more familiar with metric units are asked to use the following factors to convert certain non-metric units used in this summary.

| Non-metric       | Multiplied by | Yields metric   |
|------------------|---------------|-----------------|
| °F               | 5/9(°F - 32)  | °C              |
| ft <sup>3</sup>  | 28.3          | l               |
| in.              | 2.54          | cm              |
| in. <sup>2</sup> | 6.45          | cm <sup>2</sup> |
| lb               | 0.45          | kg              |

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*The complete report, entitled "Hitachi Zosen NO<sub>x</sub> Flue Gas Treatment Process: Volume 1. Pilot Plant Evaluation," (Order No. PB 83-113 829; Cost: \$20.50, subject to change) will be available only from:*

*National Technical Information Service  
5285 Port Royal Road  
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