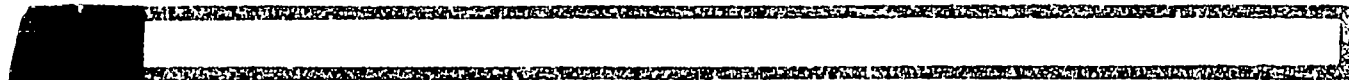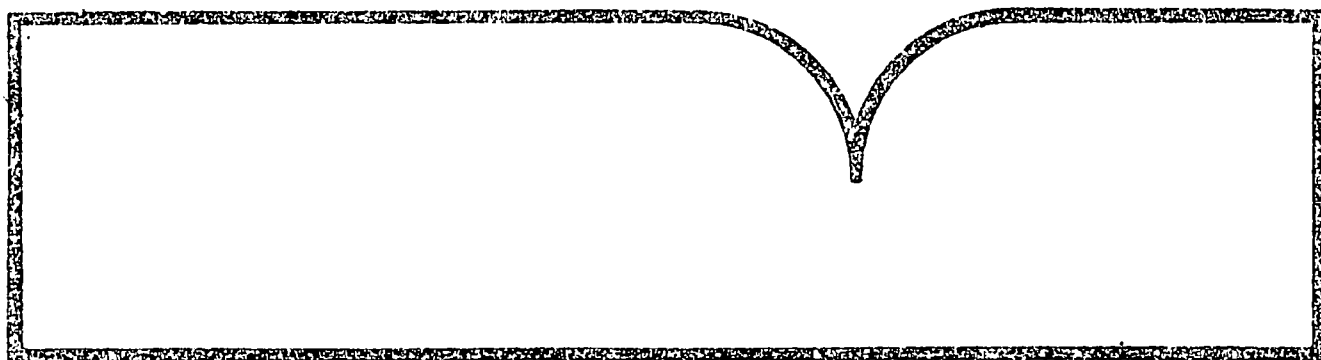Hitachi Zosen NOx Flue Gas Treatment Process
Volume 1. Pilot Plant Evaluation

Hitachi Shipbuilding and Engineering Co. Ltd.
Tokyo (Japan)

Prepared for

Industrial Environmental Research Lab.
Research Triangle Park, NC

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EPA-600/7-82-057a

September 1982

PB83-113829

Hitachi Zosen NO$_x$ Flue Gas Treatment
Process; Vol. 1. Pilot Plant Evaluation

by

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Prepared for

U.S. Environmental Protection Agency
Office of Research and Development
Washington, D.C. 20460

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16. ABSTRACT The report gives results of a pilot plant evaluation of the Hitachi Zosen NOx flue gas treatment process. The project--evaluating selective catalytic reduction (SCR) of NOx on a coal-fired source--operated for 1-1/2 years. A newly developed catalyst, NOXNON 600, was successfully applied at the 0.5 MW pilot plant on a slipstream from George Power Co.'s Plant Mitchell near Albany, GA: it demonstrated 90% NOx removal efficiency for over 5600 hours. Tests to operate the catalyst outside of design specifications showed good tolerance to adverse situations. The project was the first demonstration and evaluation of NOx SCR technology on a coal-fired source in the U.S. Project results indicate that the process is a viable NOx control technology option; however, some technical concerns remain before the technology can be considered commercially available and demonstrated for coal-fired sources in the U.S.

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ABSTRACT

The EPA-Hitachi Zosen pilot plant project - evaluating selective catalytic reduction of NO_x on a coal-fired source - operated for a year and a half. The objective of the contract, which was to operate at 90% NO_x removal over a 90-day period, was met and exceeded. The 0.5 MW pilot plant was installed on a slip stream from Georgia Power Company's Plant Mitchell near Albany, Georgia.

A newly developed catalyst, NOXNON 600 was successfully applied and demonstrated 90% NO_x removal efficiency for over 5,600 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_x selective catalytic reduction technology on a coal-fired source in the U.S. The project results indicate that the process may be useable as a NO_x 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.

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FIGURES

Figures (Continued)

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ACKNOWLEDGEMENTS

This project could not have been accomplished without the enthusiastic support of Georgia Power Company. The site that was provided at Plant Mitchell proved to be ideal. The cooperation of the Georgia Power Company personnel both at Atlanta and Albany is gratefully acknowledged. Mr. W. E. Ehrensperger, Senior Vice President, and his staff were particularly supportive. In addition, Mr. Harry Maheras, Mr. Cam Daniel, and Mr. Jim Lightfoot went out of their way to be helpful. The constructive advice and comments from Dr. W. B. Harrison, Mr. Randall Rush, and Mr. David Burford of Southern Company Services was also appreciated.

EPA also wishes to acknowledge the financial support to this project of the U. S. Department of Energy. Without their resources, this project could not have been completed as planned. The DOE project monitors were Mr. Bill Fedarko, Mr. Ed Trexler, and Mr. John Williams. They are with DOE's Fossil Energy Office.

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_____EXECUTIVE SUMMARY _ . ___ _____ _ _ __ ___ ____

INTRODUCTION

As part of the effort to assess technology for control of NO_x emissions, EPA has 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 presents details of the Hitachi Zosen process, designed to limit NO_x 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_x with ammonia which can achieve over 90 percent reduction in NO_x emissions.

Strict air pollution laws in Japan led to the construction of several full-scale systems for the removal of NO_x from flue gas. Hitachi Zosen took the lead in this area with the construction of the first large commercial unit in 1974.

To further the goal of controlling stationary source NO_x emissions, EPA has 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 are capable of achieving only limited reduction in NO_x emissions, some emphasis has been placed on developing flue gas treatment processes. In particular, SCR technology appears to be a very promising method of reducing stationary

source NO_x emissions by over 90 percent. Consequently, the
EPA has acted to demonstrate some of the more advanced SCR
systems.

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

1)   The Shell-UOP Simultaneous SO_x/NO_x Removal System

2)   The Hitachi Zosen NO_x 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

A method of removing nitrogen oxides (NO_x) from
flue gas is through the use of selective catalytic reduction.
Ammonia is the most practical reductant for this purpose be-
cause it reacts selectively and quantitatively with NO_x to
produce innocuous nitrogen and water. Ammonia is available,
relatively inexpensive, safe to handle, and easy to store.

Ammonia will react with NO_x 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 level of between 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

x

removal of NO_x 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 nitrogen oxides 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 between 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_x. 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_2 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 relations between ammonia and nitrogen oxides are not completely understood. However, certain reactions are probably involved which may or may not include oxygen. Virtually all of the NO_x in combustion gas is present as NO so

xi
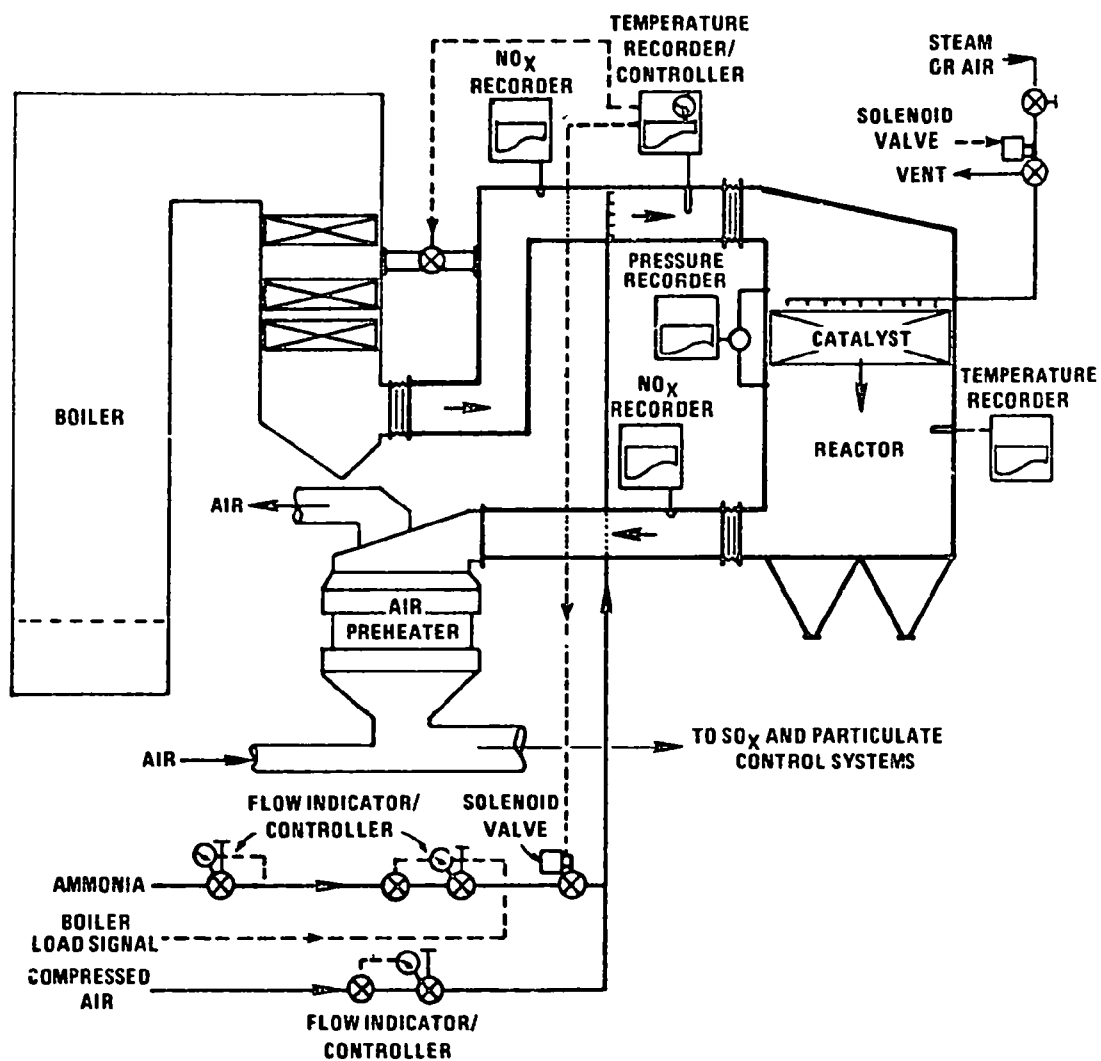
Figure 1. Typical commercial system flow diagram.

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the following equations are only for NO reactions:
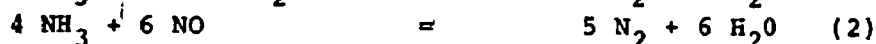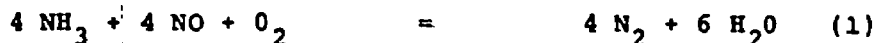
$$4\ NH_3 + 4\ NO + O_2 \quad = \quad 4\ N_2 + 6\ H_2O \quad (1)$$

$$4\ NH_3 + 6\ NO \quad = \quad 5\ N_2 + 6\ H_2O \quad (2)$$

If NO_2 is also present, the following equations represent reactions which may also occur:

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

$$8\ NH_3 + 6\ NO_2 \quad = \quad 7\ N_2 + 12\ H_2O \quad (4)$$

$$2\ NH_3 + NO + NO_2 \quad = \quad 2\ N_2 + 3\ H_2O \quad (5)$$
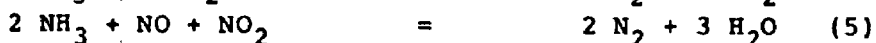
The reactions of ammonia with NO_x over the catalyst occur below 300°C. Without a catalyst the reaction will only occur in a narrow temperature range of 950-1000°C. Below this temperature the reaction rate is very low.

Factors Affecting the Catalytic Reduction of NO_x --

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_x by ammonia.

The most critical variables which affect the degree of NO_x removal are the mole ratio of ammonia to NO_x, the flue gas flow rate, and the reactor temperature.

Ammonia Emmissions --

A small amount of ammonia will invariably pass through the reactor and exit with the flue gas due to incomplete reactions between the NO_x and NH_3. Ammonia slippage (unreacted ammonia) is of concern and efforts are normally made to control slippage to levels of 10 to 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

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around 10 ppm. Another reason for the concern with ammonia slippage is that flue gas desulfurization following the de-nitrification system could absorb any ammonia in the flue gas and a build-up of ammonium salts can occur. 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_3/NO_x 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_x removal efficiency of 80%, and less than 10 ppm at an NO_x removal efficiency of 90%.

Ammonium Sulfate/Bisulfate --

The presence of sulfur trioxide (SO_3) 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 percent of the sulfur oxides in the flue gas are present as the trioxide. For coal combustion, SO_3 accounts for approximately one percent of the total SO_x. The reaction of ammonia with this sulfur trioxide will not occur above approximately 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 off of surfaces and by providing surfaces on which the deposits will form instead of forming on metallic surfaces.

A schematic flow diagram of the HZ 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_x 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. Nitrogen oxides are 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 is 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 the following characteristics:

    (1)   The structure is a thin plate honeycomb.

    (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_2 concentrations.
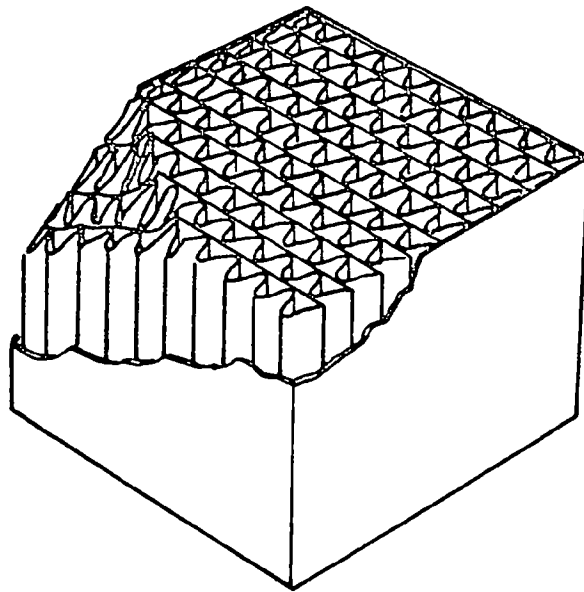
Figure 2. Configuration of NOXNON 500 or 600 series catalyst.

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As a result, in treating high-temperature gases with high
SO$_x$ and dust concentrations such as coal-fired boiler flue
gas, the NO$_x$ removal reactor can be installed immediately
downstream of the economizer. In addition, dust elimination
or other pretreatment is unecessary 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 Series. These catalysts are manu-
factured in the form of plates and are fabricated so as to
form parallel flow gas passages which provide excellent con-
tact 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 1.0 meters long,
1.0 meters wide, and 0.5 meters deep.

The activation of the catalyst follows after the corru-
gated catalyst assembly is made. In the NOXNON 500 catalyst,
thin stainless steel plates are used.

A newer development is NOXNON 600 catalyst which, 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 cata-
lyst.

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

The catalyst is designed with an expected life of approxi-
mately two 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 haz-
ardous materials and can be disposed of by recovering the
metal in the catalyst or by disposing of the spent catalyst as

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industrial waste. No special precautions are required in the disposal process.

Reactor Design

Reactor vessels housing the catalyst are of carbon steel construction and are divided into chambers each having its own fly ash hopper (see Figure 3). The reactor is oriented so that flue gas flows downward. Flue gas usually enters at the top of the reactor at one side, passes through the gas distribution grid and catalyst bc , 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
9 1 ⁖

A potential market for the denitrification of power plant flue gases may be at coal-fired stations. There has been limited test work done by Hitachi Zosen on flue gas from coal-combustion. The EPA-sponsored demonstration program at Plant Mitchell of the Georgia Power Company provided an excellent site for testing of 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 were conducted to provide information on the aging

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GAS DISTRIBUTION
GRIDS

REACTOR INLET

AMMONIA FEED
CONNECTIONS

CATALYST

SOOT BLOWER

TREATED FLUE GAS TO
BOILER PREHEATER

HOPPERS
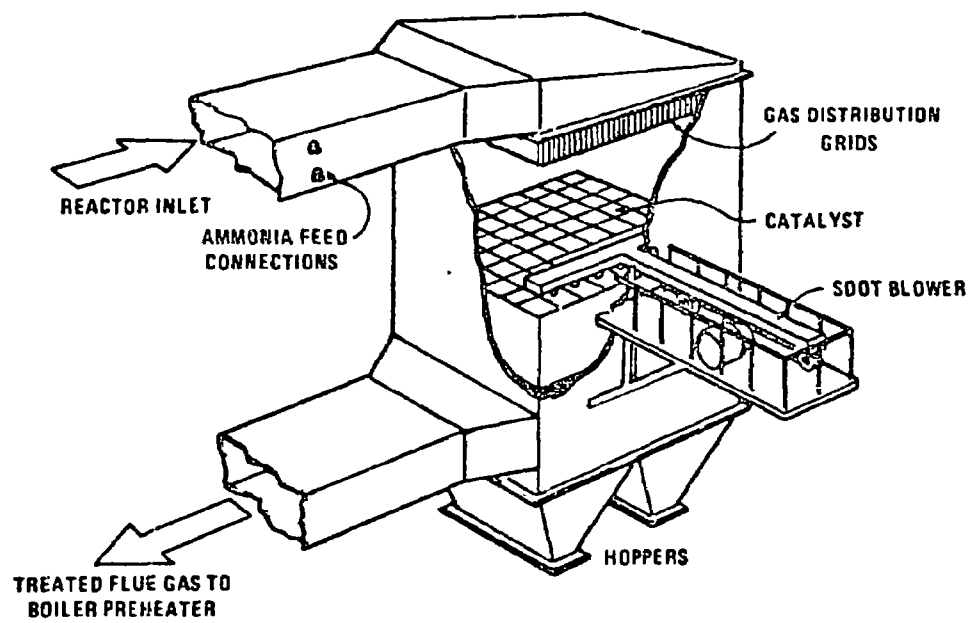
Figure 3.   Typical Reactor Arrangement.

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 capital cost requirements for commercial installations.

Hitachi Zosen, with Chemico Air Pollution Control Corporation, Division of Envirotech Corporation (CAPCC) 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 performed 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 startup, debugging, and parametric tests.

Following the successful completion of the system optimization tests, Phase III, the pilot plant was to be continuously operated (24 hours/day, 7 days/week) for at least three 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.

An additional series of 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 eighteen (18) months. The award of the contract was May 1978. The design

and specification period, Phase I, took approximately three
months. Phase II required approximately nine months for
procurement, fabrication, transportation and erection.
These schedules were as originally expected. However,
startup and debugging required about two months and so the
first tests were not started until August 1, 1979. This
first charge of catalyst showed less than optimum results
after some four months of operation and it was decided to
replace it in December of 1979. After some four additional
months of operation, once again, the NOXNON 500 catalyst
began to provide less than expected results and a decision
was made to install a third charge of catalyst.

   The third charge was a new type: NOXNON 600. This was
installed April 18, 1980 and was utilized for some nine
months until the plant was shut down on February 2, 1981.

Description of the Test Plant

   The following is a description of the pilot unit in-
cluding summaries of major items of equipment (see Figure
4):
   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 fourteeninch 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 at the temperature
required.
   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 NO_x
in the gas reacted with the ammonia to form small quantities
of gaseous nitrogen and water, which remained in the gas
stream.
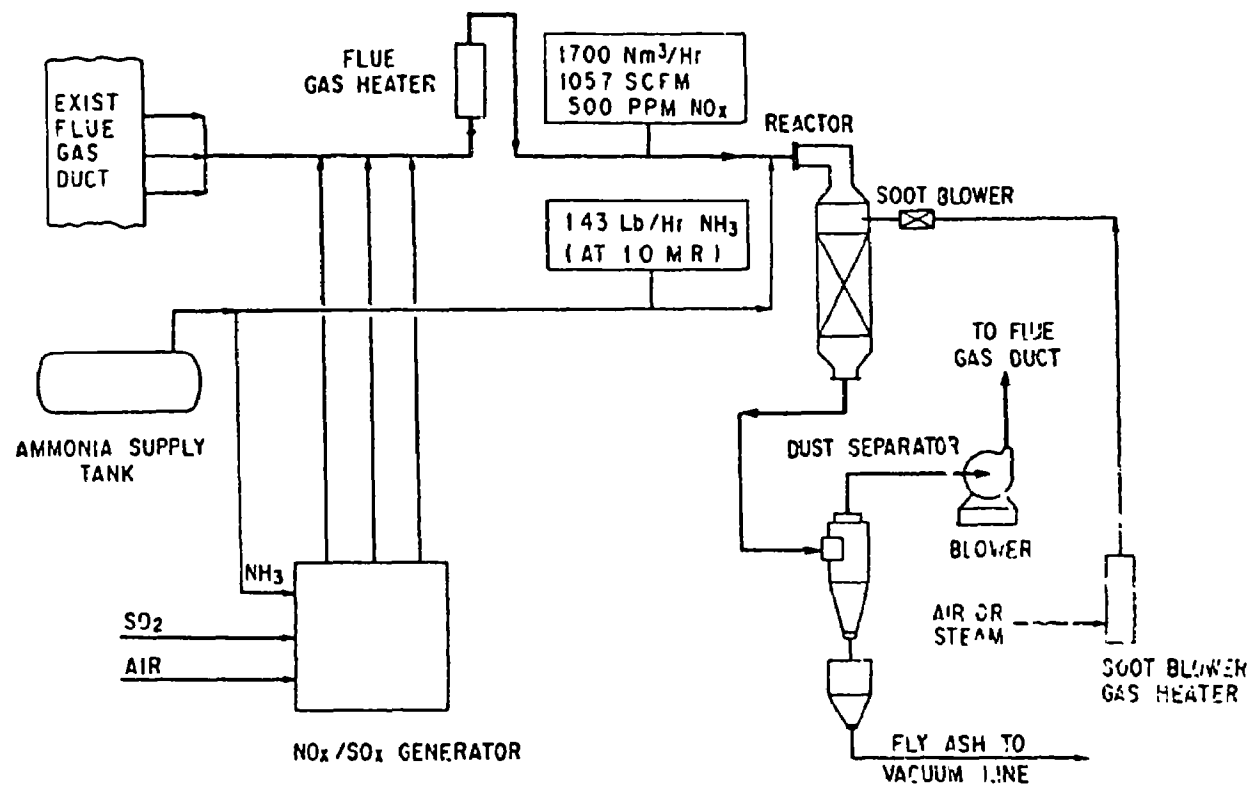
Figure 4. Process Flow Diagram of Pilot Plant.

Dust settling in the reactor was kept within acceptable bounds through the use of a soot blowing apparatus which used either steam or air.

A blower was located downstream of the reactor to over-come the flue gas pressure drop through the pilot plant. A cyclone dust separator was supplied ahead of the boiler to prevent erosion of the blower by dust.

Since it was planned to test the performance of NO_x, SO_2, SO_3, and since only fixed concentrations of these substances were available from the Albany, Ga. plant boiler operation, SO_2 was purchased for addition to the flue gas, and a NO_x, SO_2 and SO_3 generating unit was provided to manufacture these materials for addition to the flue gas. This permitted testing at NO_x, SO_2 and SO_3 levels higher than those available from the boiler.

The operating conditions which were controlled in the pilot unit are as follows:

1. Flue gas flow rate.
2. Temperature of flue gas.
3. The amount of charged ammonia.
4. Soot blowing gas pressure and temperature.

Host Site

. The pilot plant was located at the Unit #3 of Plant Mitchell, Georgia Power Co., Albany, Georgia. This unit has a pulverized coalfired Combustion Engineering boiler which was initially operated 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 outdoors of the Unit #3.

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Data Collection

The pilot plant operation was closely monitored through the use of an array of instruments and analyzers. Continuous analyzers of the latest design provided measurements of the NO_x levels both entering and leaving the reactor.

Much of the data at the pilot plant was collected in a data logger which frequently scanned the instruments and stored the data. Visual display and paper tape display 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 fullscale plant. The results could therefore be directly extrapolated to a large plant assuming that the velocity, mole ratio, and temperature remain 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 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 charge and the second charge of catalyst did not achieve their expected performances. 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

catalytic components are cemented. This technique has been
proven in applications on oilfired and coalfired combustion
flue gas in Japan.

Catalyst Performance

Starting from April 22, 1980, the demonstration operation
with NOXNON 600 ran for more than nine months until February 2,
1981. The operation with combustion flue gas from the Unit #3
Boiler of Georgia Power Company, Plant Mitchell was for 5,620
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 NO_x removal
efficiency of more than 90 percent continuously for a period
of more than three months. Afterwards, the project scope was
extended and transient tests were included in the scope of the
contract along with an extension of the operating period.

Catalyst life tests were run to confirm the expected cat-
alyst life. From April 22, 1980, the pilot plant was operated
maintaining NO_x removal efficiency of more than 90 percent until
the end of October. After October, a nominal 80 percent NO_x
removal was accepted in order 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 5,000 hours of operation could be obtained to
evaluate the long term effectiveness of the NOXNON 600.

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

During the operating time with flue gas tests were run
including catalyst life test, catalyst performance test, and
transient tests. Controllability and reliability of the entire

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system was also evaluated at the same time.

Operating conditions were varied for the performance tests and transient tests. Therefore, when the activity of the catalyst was to be evaluated the 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_3/NO_x mole ratio of 1.0. Under these conditions the NO_x removal efficiency was measured over a period of several hours to determine the condition of the catalyst.

At the beginning of the operation		: 90 - 94% removal
At the end of August '80	(2500 hours)	: 90 - 94%
November 1980	(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. This was accomplished by using the flue gas flow rate signal and the inlet NO_x concentration analysis to determine the quantity of NO_x in the inlet stream. From the inlet NO_x 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 a removal of 80% requires a mole ratio of about 0.85 while a 90% removal requires a mole ratio of 1.0.

NO$_x$ REMOVAL EFFICIENCY (%)

NH$_3$ SLIPPAGE (PPM)

HN$_3$/NO$_x$ MOLE RATIO (—)

SYMBOL ⎧ ○ 1100 SCFM 700±10 °F
⎨ △ 1300 SCFM 700±10 °F
⎩ □ 1500 SCFM 710±5 °F
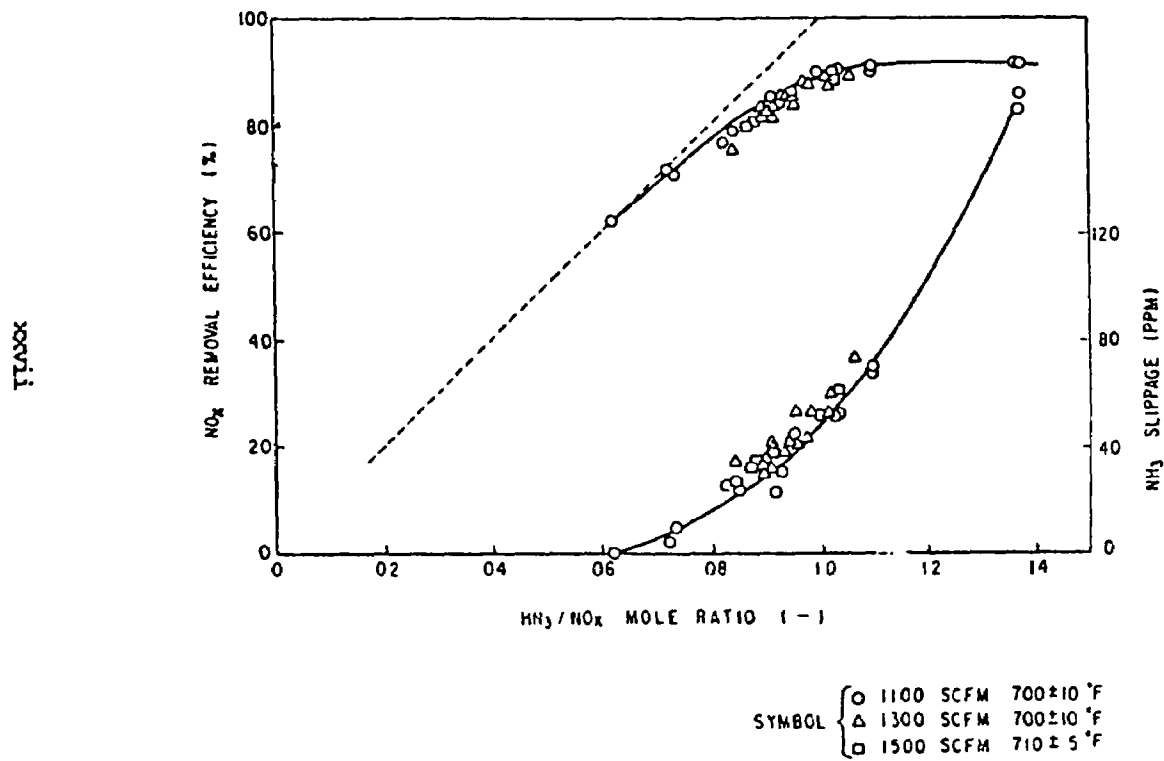
Figure 5. Effect of NH$_3$/NO$_x$ Ratio on NO$_x$ Removal Efficiency and NH$_3$ Slippage.

The NOXNON 600 catalyst for the pilot plant was designed to operate at 1057 SCFM which is equivalent to an Area Velocity (A.V.) of 9.6 Nm^3/m^2-hr. However, operations at a much higher flow rate of 1650 SCFM (an A.V. of 15) provided the desired 90 percent NO_x 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_x would be unchanged. As seen in Figure 6 the flow rate had little or no effect on the removal through the program.

Influence of SO_x Concentration on NO_x Removal Efficiency --

Concentrations of SO_x 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_x removal efficiency was not affected by SO_x 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), and NO_x removal efficiency was not affected in this temperature range. Optimum operating temperatures obtained from fundamental experiments in the laboratory are between 572°F (300°C) and 750°F (400°C).

NO_x Concentration --

On August 24, 1980, the NO_x Generator was operated to increase the concentration of NO_x at the inlet of the reactor, and the influence of NO_x concentration was investigated.

EPA Form 2220-6 (4-80)

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REACTION TEMP. · APPROX 700 °F
NH₃/NOₓ RATIO · APPROX 10
SYMBOL OPERATING PERIOD

o   700 ~ 1250ᴴ
•   2150 ~ 3750ᴴ

NOₓ REMOVAL EFFICIENCY (%)

FLUE GAS FLOW RATE (SCFM)

xxix
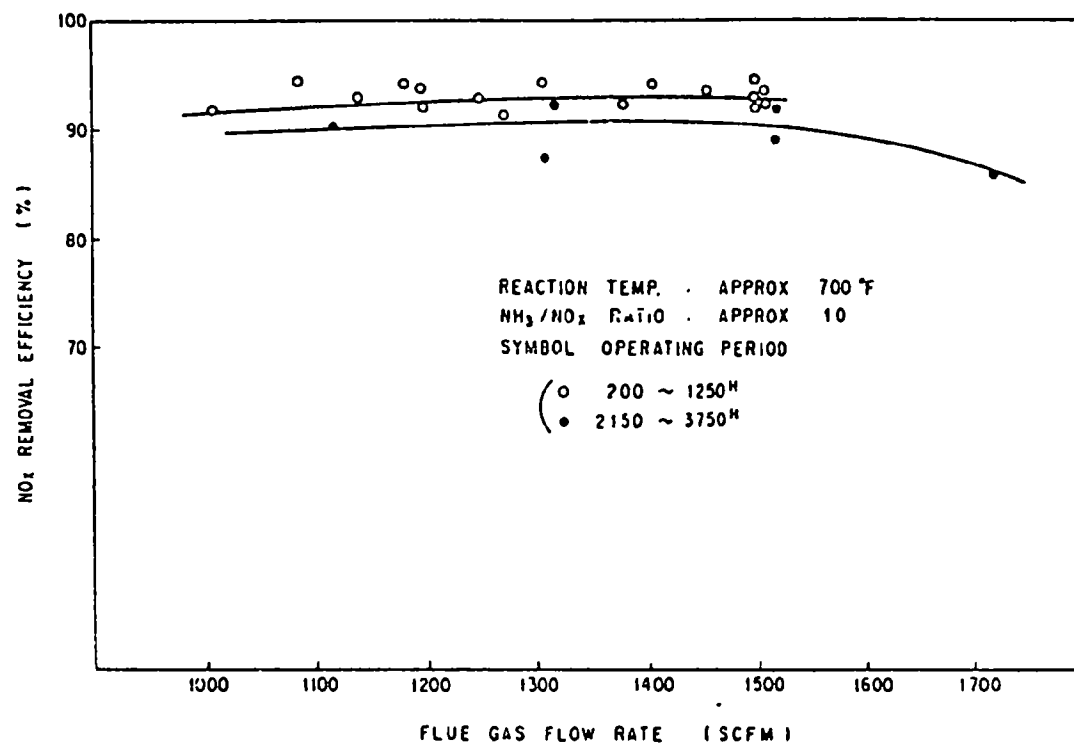
Figure 6. Effect of Flue Gas Flow Rate on NO_x Removal Efficiency.

Between 400 ppm and 900 ppm of inlet NO_x concentrations, the concentration of NO_x did not influence 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 slip when mole ratio was varied at operating times of 3200 hours to 4500 hours. General conclusions from these tests:

- 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, for example at a mole ratio of 0.6.

The reason for this higher than expected slippage may have been due to 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 slip. Even a slight reactivity loss while producing little loss in NO^x removal efficiency, can cause a significant increase in ammonia emissions.

Oxidation of SO_2 to SO_3 --

On July 23, 24 and 25, 1980 operating conditions were maintained at a constant level so that SO_3 measurements could be obtained at the inlet and outlet of the reactor.

The results indicated an average oxidation rate of 1.8%.

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From prior experimental results, it was expected that at these conditions the ratio would be somewhat lower: about 1.0 to 1.5%.

Transient Tests

After the NO_x removal efficiency of more than 90 percent was demonstrated in the continuous run of three 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. The following 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 returns 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.
- 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 into operation after a long shut down with the reactor and ductwork filled with ambient

air. When operation commences flue gas would be introduced into the system as the temperature rises and the flue gas could 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 cause any trouble to the 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 performances.

Boiler Shut-down and Start-up --

From time to time a power plant boiler shuts down and starts up and an NO_x removal system must follow such transition periods. The purpose of this test was to confirm the controllability of the NO_x removal system during the shut-down and start-up of the boiler. When the host boiler was shut-down for maintenance, this test was executed with the NO_x removal system shutting down and starting up along with the boiler with no purging of the reactor. From the results, the system proved that it could withstand the transient period of shut-down and start-up of the boiler.

Sudden Load Change --

The boiler for a power plant may occasionally change load

suddenly complying with variations of power consumption. The NO$_x$ removal system should follow these sudden boiler load changes. The following 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 once every two hours and continued for 24 hours. There seemed to have been no serious problem 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, in order 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, 0.8 mole ratio, sootblower operation was halted and the progress of pressure drop increase was observed. Pressure drop appeared unstable and increased gradually from 1.20 to 1.25 inches H$_2$O to 1.35 to 1.40 inches H$_2$O in approximately 28 hours, and seemed to continue to increase. The sootblower was then restarted at this time. From this test it was concluded that operation of the sootblower was necessary.

Changes in Pressure Drop in Relation to NO$_x$ 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$_x$ 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, and the tendency for pressure drop changes are summarized below.

- When the pilot plant was shut down for some reason the pressure drop generally increased after the next start-up. 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 twenty days of steady operation the flow rate was changed from 1300 SCFM to 1500 SCFM and after two 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 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 twenty-eight hours without the sootblower and the pressure drop increased sharply. After the

sootblower was returned to operation it took three days to restore the pressure drop.

Regeneration of Catalyst

When the catalyst was washed with warm water the NO_x 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 a typical condition. 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 operation of 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 to 35 percent 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.

The contract objectives were met and exceeded. NO_x removal efficiency of more than 90% was demonstrated during an operating period of approximately 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 shut-downs 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 the operation of the pilot plant, catalysts had to be replaced twice. The first and second charge 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 the apparent catalyst activity, although the 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

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 ade-
quate linear velocity to prevent clogging and abrasion caused by
fly ash, and to determine the necessity and operating conditions
of the soot blower. 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 avail-
able, 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 wash-
ing solution should be investigated before commercial appli-
cation. 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_x removal could be expected
at a NH_3/NO_x 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_x 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 between 1.0 and 1.4

inches H_2O. 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 was verified by the high onstream factor achieved. A desirable addition to the control system would be a continuous analyzer to monitor slippage of ammonia. A desirable addition to the control systems would be a continuous analyzer to monitor slippage of ammonia.

The NO_x removal efficiency of this SCR system in commercial applications would be expected to be the same as that experienced in the EPA pilot plant provided that the size and configuration of the catalyst, superficial linear velocity of the flue gas across the catalyst, and the temperature of flue gas are 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_x selective catalytic reduction technology on a coal-fired source in the U.S. The project results indicate that the process may be useable as a 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.

SECTION 1

INTRODUCTION

Nitrogen oxides in the atmosphere have been determined to have adverse effects on human health and welfare. The Clean Air Act of 1970 and subsequent amendments to this act requires the reduction in the emission rates of nitrogen oxides along with sulfur oxides, particulates, carbon monoxide, and hydrocarbons. A projection indicates that, while emission rates of the other four pollutants will stabilize in the near future, the nitrogen oxide (NO_x) emissions will continue to increase and by 1990 will be 50 percent higher than in 1975. Because of this the EPA has placed special emphasis on the development and demonstration of technology to reduce NO_x emissions.

As part of the effort to advance the technology for control of NO_x emissions, EPA has 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 presents details of the Hitachi Zosen process, designed to limit NO_x emissions from coal-fired steam generators, and results of the demonstration program.

THE PROBLEM OF NO_x CONTROL

Over 95 percent of man-made NO_x emissions result from combustion process. Nitrogen and oxygen can combine to form several nitrogen oxide compounds. However, for most combustion processes, the only nitrogen oxide compounds produced in significant concentrations are nitrogen oxide (NO) and nitrogen dioxide (NO_2).

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Typically, NO represents 90 to 95 percent of the NO_x emissions from combustion. But this NO gradually oxidizes to NO_2 once it is exposed to oxygen in the atmosphere.

Sources of man-made NO_x can be classified as either mobile or stationary. Each of these sources contributes approximately 50 percent to the annual NO_x emissions in the U.S. Mobile sources primarily consist of automobiles and trucks while the majority of stationary source emissions results from utility and industrial steam generators.

METHODS TO CONTROL NO_x EMISSIONS FROM STATIONARY SOURCES

NO_x is formed in the combustion process by one of two routes. It can result from combination of fuel-bound nitrogen with oxygen, or it can result from the thermal fixation of atmospheric nitrogen and oxygen in the combustion zone. In both cases the end result is primarily NO. This is because the residence time in most combustion units is too short for a significant amount of NO_2 to form. The formation of NO_x from fuel-bound nitrogen is relatively insensitive to combustion temperature while NO_x formed by thermal fixation depends primarily on the reaction temperature. Another significant factor which influences the formation of NO_x is the amount of trace oxygen available for reaction.

The contribution of the two mechanisms to total NO_x emissions varies widely depending on the type of fuel and the combustor in use. For example, NO_x emissions from a natural gas fired boiler result almost exclusively from thermal fixation. But, up to 80 percent of the NO_x emissions from a coal fired boiler can result from oxidation of fuel-bound nitrogen.

Because of the way NO_x is formed, there are three methods of controlling NO_x emissions. These are pre-combustion, combustion, and post-combustion control. Pre-combustion control is designed to limit the oxidation of fuel-bound nitrogen by

removing the nitrogen from the fuel prior to combustion. Currently, this control technique is primarily limited to switching from a high to a low-nitrogen fuel. Techniques are under development for removing nitrogen from fuel, but success has been limited.

Combustion control, or combustion modification, is a second technique which limits the formation of NO_x by adjusting temperature and the availability of free oxygen in the combustion zone. Combustion modification techniques have been demonstrated in the past and to a large extent they are considered available technology. In fact, it was these developed techniques which permitted utility boilers to meet New Source Performance Standards (NSPS) for NO_x emissions.

Employing conventional combustion modifications, NO_x emissions can be reduced up to 50 percent with some loss of boiler efficiency. And recently, more sophisticated techniques employing burner designs which carefully control temperature and the amount of oxygen in the flame have demonstrated somewhat higher reduction in NO_x emissions with little or no loss in boiler efficiency. Combustion modifications are relatively simple and inexpensive. Their primary limitation is the relatively low degree of NO_x removal which can be achieved. Although this level of removal has been able to comply with current NSPS, combustion modifications alone may not be adequate control technique for complying with future NO_x emission standards.

A third method for limiting stationary source NO_x emissions is post-combustion control, known as flue gas treatment (FGT). FGT is a general classification for a variety of processes which remove NO_x from the products of combustion. This technology was first developed in Japan and it has reached a relatively advanced state for use on gas and oil fired boilers. However, it remains to be demonstrated in the United States. Also, very little work has been done with respect to applying FGT to flue gas from coal fired boilers.

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There are many different flue gas treatment processes, but
in general, they are characterized by high NO_x control efficien-
cies. However, this additional removal is attainable only with
a significant increase in costs. Typically, greater than 80
percent reduction in NO_x emissions is possible with an FGT pro-
cess as compared to a maximum of about 50 percent with combus-
tion modifications.

One of the most advanced, promising, and cost effective FGT
processes is selective catalytic reduction (SCR) which can
achieve over 90 percent reduction in NO_x emissions.

In summary, there are three methods available to control
NO_x emissions. Pre-combustion control is primarily limited to
switching the type of fuel burned in order to reduce the quantity
of fuel bound nitrogen and thus, NO_x emissions. Combustion
modification involves careful control of the temperature and the
oxygen available in the combustion zone. This method of control
is relatively well developed technology, and it is relatively
inexpensive. However, the NO_x removal efficiency associated with
combustion modifications is limited to about 50 percent. Flue
gas treatment is the third method of controlling NO_x emissions.
NO_x removal efficiencies greater than 80 percent are possible
and SCR processes are capable of over 90 percent reduction in
NO_x emissions. But, FGT processes have not been developed in
this country to the extent combustion modifications have. In
addition, the costs associated with FGT appear significantly
higher than those of combustion modifications.

DEVELOPMENTAL HISTORY

Strict air pollution laws in Japan led to the construction
of several full-scale systems for the removal of NO_x from flue
gas. Hitachi Zosen took the lead in this area with the construc-
tion of the first large commercial unit in 1974. Hitachi Zosen
now has nine commercial plants in operation.

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Early in 1978 Chemico Air Pollution Control Corporation
(CAPCC) (a division of Envirotech) acquired the North American
license for the Hitachi Zosen technology for the process to
remove NO_x from flue gas by selective catalytic reduction using
ammonia. CAPCC is a major supplier of wet scrubbers and other
flue gas desulfurization systems designed to control air pollu-
tion emissions from electric utility boilers.

Hitachi Zosen began the development of catalysts in late
1959 with basic research and laboratory testing. The original
work was done with carrier-supported catalysts. The process was
tested in 1973 in a 10,000 Nm^3/hr (6000 scfm) pilot plant that
was used for the denitrification of flue gases from a boiler
plant. The data collected from an extensive series of tests
provided the basis for the first NO_x removal plant which was for
Idemitsu Kosan.

In early 1974 a contract was signed between the Idemitsu
Kosan Company, one of the world's leading petroleum refineries,
and Hitachi Zosen for the construction of a NO_x removal plant
with the capacity of treating 350,000 Nm^3/hr (218,000 scfm) of
flue gas. The plant was the largest of its kind in the world.
Construction work commenced at the Chiba Refinery in May 1975
with test operations in November of the same year. The plant
operated successfully with removals as high as 95 percent. Fol-
lowing the start-up of the Idemitsu Kosan system other plants
were constructed.

SELECTIVE CATALYTIC REDUCTION DEMONSTRATION PROGRAM

To further the goal of controlling stationary source NO_x
emissions, EPA has sought to enhance the reliability and effect-
iveness of technology to reduce these emissions. One aspect of
EPA's involvement includes sponsoring programs designed to dem-
onstrate this technology. Because combustion modifications are
capable of achieving only limited reductions in NO_x emissions,

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some emphasis has been placed on developing flue gas treatment processes. In particular, SCR technology appears to be a very promising method of reducing stationary source NO_x emissions by over 90 percent. Consequently, the EPA has acted to demonstrate some of the more advanced SCR systems.

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

1) The Shell-UOP Simultaneous SO_x/NO_x Removal System

2) The Hitachi Zosen NO_x 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.

SECTION 2

PROCESS DESCRIPTION

The best method of removing nitrogen oxides (NO_x) from flue gas is through the use of selective catalytic reduction. Many reducing agents can be used for this purpose including hydrogen, carbon monoxide, hydrogen sulfide, ammonia, methane, and cther hydrocarbons. However, ammonia is the most practical reductant for this purpose because it reacts selectively and quantitatively with NO_x to produce innocuous nitrogen and water. Ammonia is available, relatively inexpensive, safe to handle, and easy to store.

Ammonia will react with NO_x without a catalyst in a narrow temperature range at around $1000^\circ C$. By using a suitable catalyst the required temperature can be lowered to a more practical level of between $300-450^\circ C$. Several different catalysts have been tested for this purpose. Platimum compounds are the most efficient catalysts to promote the selective reduction of nitrogen oxides. However, they cannot be applied to most flue gases due to the poisoning of the catalyst by sulfur oxides and other elements or compounds.

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. Alumina is a very suitable material but it has been found to react with sulfur oxides, thus decreasing the available surface area and catalyst activity. Other carrier materials, however, have been found to be less affected by SO_x and these have been successfully applied in commercial systems.

With dust-containing gases, carrier-based catalysts are not

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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_x from dusty gases. The 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 nitrogen oxides in the flue gas to form small quantities of nitrogen gas and water vapor, both of which are usual constituents of the atmosphere and are environmentally acceptable.

Figure 2-1 illustrates a typical flow sheet of a commercial system. Ammonia is injected into the flue gas from a boiler between the economizer and the air preheater. At this point the gas temperature is about 400°C which is suitable for the catalytic reduction of NO_x. This gas enters the reactor, passes over the catalyst, and then reactions proceed. The flue gas then passes through the usual air heater, precipitator, SO_2 control system, 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.

Figure 2-1. Typical commercial system flow diagram

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Chemistry

The exact relations between ammonia and nitrogen oxides are not completely understood. However, certain reactions are probably involved which may or may not include oxygen. Virtually all of the NO_x in combustion gas is present as NO so the following equations are only for NO reactions:

$$4\ NH_3 + 4\ NO + O_2 \quad = \quad 4\ N_2 + 6\ H_2O \qquad (1)$$

$$4\ NH_3 + 6\ NO \quad = \quad 5\ N_2 + 6\ H_2O \qquad (2)$$

If NO_2 is also present, the following equations represent reactions which may also occur:

$$4\ NH_3 + 2\ NO_2 + O_2 \quad = \quad 3\ N_2 + 6\ H_2O \qquad (3)$$

$$8\ NH_3 + 6\ NO_2 \quad = \quad 7\ N_2 + 12\ H_2O \qquad (4)$$

$$2\ NH_3 + NO + NO_2 \quad = \quad 2\ N_2 + 3\ H_2O \qquad (5)$$

At high temperatures (above $1000^\circ C$), additional reactions can take place as illustrated by equations 6, 7 and 8:

$$4\ NH_3 + 3\ O_2 \quad = \quad 2\ N_2 + 6\ H_2O \qquad (6)$$

$$4\ NH_3 + 5\ O_2 \quad = \quad 4\ NO + 6\ H_2O \qquad (7)$$

$$4\ NH_3 + 4\ O_2 \quad = \quad 2\ N_2O + 6\ H_2O \qquad (8)$$

The reactions of ammonia with NO_x over the catalyst occur below $300^\circ C$. Without a catalyst the reaction will only occur in a narrow temperature range of $950-1000^\circ C$. Below this temperature the reaction rate is very low while at higher temperatures reactions (6), (7) and (8) readily occur producing more NO_x than there was originally.

With the presence of sulfur oxides in the flue gas, certain undesirable reactions can also take place. These are the reactions between ammonia and sulfur trioxide to form ammonium sulfate or ammonium bisulfate.

$$2\ NH_3 + SO_3 + H_2O \quad = \quad (NH_4)_2SO_4$$

$$NH_3 + SO_3 + H_2O \quad = \quad NH_4HSO_4$$

A discussion of the reactions which form ammonium sulfate/bisulfate is presented later.

Kinetics of the Reaction

It appears from the results of laboratory and pilot plant tests that the reduction of nitrogen oxide with ammonia is similar to a first order reaction. With sufficient ammonia present the rate of disappearance of nitrogen oxide is proportional to the concentration of nitrogen oxide. In practical terms, at a fixed gas velocity and a fixed volume of catalyst the percent removal of NO_x (at any one ratio of ammonia to NO_x) would be the same at any NO_x concentration. This can be shown as follows:

A first order reaction can be expressed mathematically

as: $$\frac{dC}{dt} = kC$$ where t = residence time
C = concentration
k = a constant

Integrating this expression under the conditions t = 0 and the initial concentration as C_o, the result is:

$$\frac{C}{C_o} = e^{-kt}$$

If t is a constant at a fixed velocity of the gas and a fixed volume of catalyst, then C/C_o (or the fraction removal) would also be a constant. This has been substantiated by experimentation.

Regeneration of the Catalyst

The catalyst is designed with an expected life of approximately two 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.

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_x by ammonia.

The most critical variables which affect the degree of NO_x removal are the mole ratio of ammonia to NO_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 NO_x in the flue gas to be treated. Typically a ratio of 0.9 to 1.0 is required for a NO_x removal of 90 percent or higher. Ammonia will remove one mole of NO_x per mole of ammonia up to a mole ratio of 0.7. Above this the removal becomes less efficient and the removal of more than 90 percent NO_x requires a lot more ammonia for the final quantities of NO_x to be reacted.

Flue Gas Flow Rate --

The gas velocity through the reactor is an important parameter. At lower velocities the contact time with the catalyst is increased and better removals are obtained. Operating at higher velocities will, of course, allow the system to use less catalyst and reduce the costs. Higher velocities will, however, increase the pressure drop and will also lead to increased erosion of the catalyst surface particularly when significant quantities of fly ash are present in the gas. (The degree of erosion is very slight and the catalyst is non-toxic and, therefore, there would

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be no air pollution problems.) The increased erosiion will
decrease the life of the catalyst. Hitachi Zosen has performed
a great deal of research work to develop catalysts which are
resistant to erosion and they have established optimum gas
velocities for obtaining high removal effiencies with low
pressure drops and long catalyst life. Tests at a wider range
of flows performed in Japan indicate a definite drop off of
efficiency at very high flow rates. Very high flow rates are
not recommended because of increased pressure drops.

The concept of space velocity is normally applied to cat-
alysts 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:

Nm^3/m^2-hr

The particular A.V. used in the Hitachi Zosen design depends
upon the percent removal of NO_x required, the gas temperature,
and other factors.

Temperature --

The flue gas temperature in the reactor is an important
variable. Normally the reactor temperature is held above 320°C
(608°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 Nm^3/m^2-hr tempera-
tures above 300°C have little effect on the percent NO_x removal
efficiency. In effect, the normal expected operating temperature
ranges (above 300°C) have essentially no influence over the
removal efficiency.

Sulfur Oxide Concentration --

The presence of SO_x has essentially no effect on NO_x removal since the catalyst has been formulated to tolerate the presence of these compounds.

Oxygen Concentration --

A certain amount of oxygen in the flue gas is necessary to promote NO_x reduction. With flue gas containing 2 percent or more oxygen, there is little effect of oxygen on the NO_x removal. However, at oxygen levels below one percent the removal is adversely affected.

Moisture Content of Flue Gas --

Moisture in the flue gas tends to decrease some of the catalytic activity. However, this decrease is small and levels off above 5 percent moisture in the flue gas.

NO_x Concentration --

The performance of the catalyst is not affected by the usual inlet NO_x concentrations expected from a boiler provided ammonia is injected in stoichiometric quantities.

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_x and NH_3. Ammonia slippage (unreacted ammonia) is of concern and efforts are normally made to control slippage to levels of 10 to 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.

gas desulfurization following the denitrification system could
absorb any ammonia in the flue gas and a build-up of ammonium
salts can occur. 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_3/NO_x mole ratio,
the area velocity, and the temperature at which the reaction
occurs. For commercial application to coal-fired utility boil-
ers, ammonia slippage would be expected to be less than 5 ppm
at an NO_x removal efficiency of 80%, and less than 10 ppm at
an NO_x removal efficiency of 90%.

Ammonium Sulfate/Bisulfate

The presence of sulfur trioxide (SO_3) 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 percent of the sulfur oxides in the flue gas are present as
the trioxide. For coal combustion, SO_3 accounts for approxi-
mately one percent of the total SO_x. The reaction of ammonia
with SO_3 will not occur above approximately 300°C (572°F). At
very low concentrations a temperature of around 200°C (392°F)
might be sufficient to avoid this formation. Figures 2-2 and
2-3 show the relationship between the ammonia concentration,
sulfur trioxide concentration and temperature of the flue gas
below which the reaction to form ammonium sulfate or ammonium
bisulfate will occur.

The condition in which ammonium sulfate and bisulfate are
formed depends on the equilibrium in the reactions described
below:

$$2\ NH_3 + SO_3 + H_2O = (NH_4)_2SO_4 \quad (1)$$
$$NH_3 + SO_3 + H_2O = NH_4HSO_4 \quad (2)$$
$$(NH_4)_2SO_4 + SO_3 + H_2O = 2\ NH_4HSO_4 \quad (3)$$
$$NH_4HSO_4 + NH_3 = (NH_4)_2SO_4 \quad (4)$$

2-9
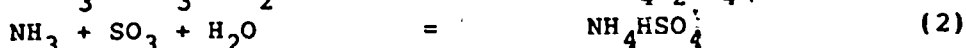
tion equilibrium when the vapor pressure is atmospheric and the moisture content is ten percent. Solid lines shown as A and B, and dotted lines C and D indicate the equilibrium between equations (1), (2), (3) and (4), respectively.

The area surrounded by the solid line A and B, the vertical axis, and the horizontal axis represents the condition at which $(NH_4)_2SO_4$ and NH_4HSO_4 cannot be formed.

In the area surrounded by the solid line A, the dotted line C, and the vertical axis where the concentration of ammonia is higher than the concentration of line A, only $(NH_4)_2SO_4$ is produced according to equation (1). In the area to the right of solid line B and below dotted line D only NH_4HSO_4 is formed according to equation (2).

In the area to the right side of dotted line C and the upper side of dotted line D, both $(NH_4)_2SO_4$ and NH_4HSO_4, produced by equations (3) and (4), coexist.

For instance, with a concentration of 10 ppm NH_3 and 50 ppm SO_3, ammonium bisulfate (NH_4HSO_4) will form if the temperature drops to around 230°C (446°F). (See Figure 2-3).

Double salts of ammonium sulfate and ammonium bisulfate can also form upon the cooling of flue gas. The flue gas temperature leaving the air preheater is in the range of 150-200°C (302° - 392°F) and deposits can, therefore, form. The deposits are often double salts of ferrous sulfate and ammonium bisulfate. The ferrous ions are the results of the corrosion of steel by ammonium bisulfate which is very corrosive.

It is also known that certain deposits can form on catalyst surfaces at 300°C (572°F) or higher even though the gas concentration is relatively low in SO_3 and NH_3. This condition could be the result of localized high concentrations on the surface of the catalyst due to the oxidation of sulfur dioxide and the absorption of ammonia.

To avoid such deposits Hitachi Zosen recommends that the temperature in the reactor be maintained at a minimum of 320°C

2-10
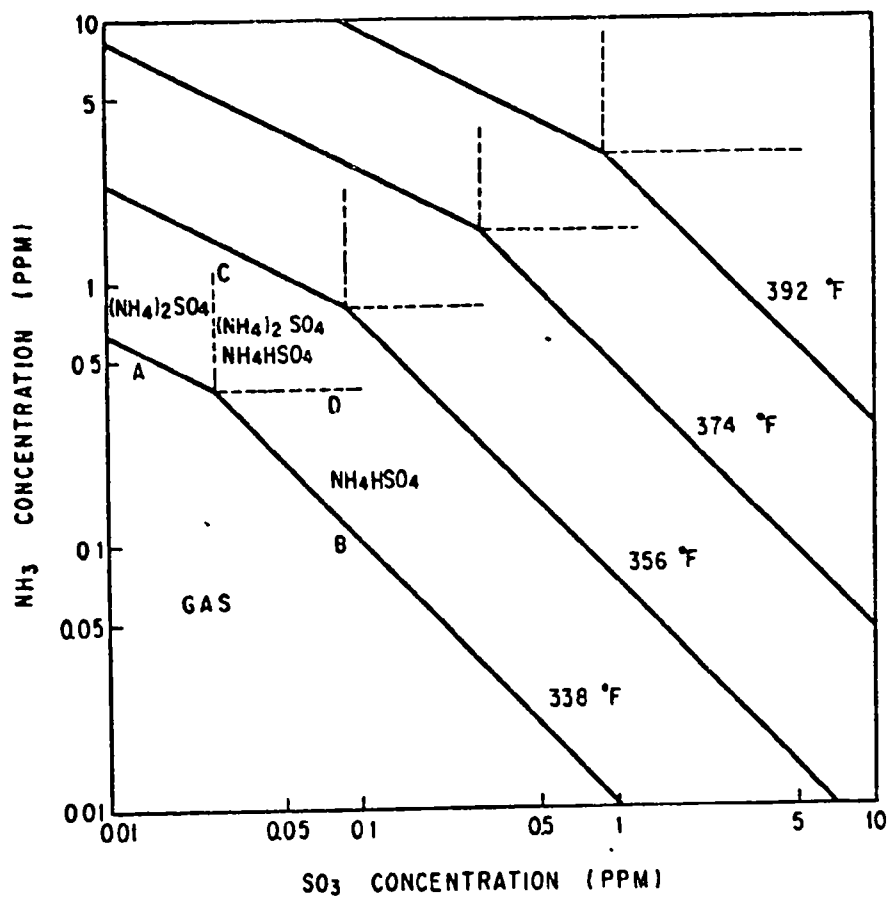
Figure 2-2.   Formation of Ammonium Sulfate
(Low Temperature Range)
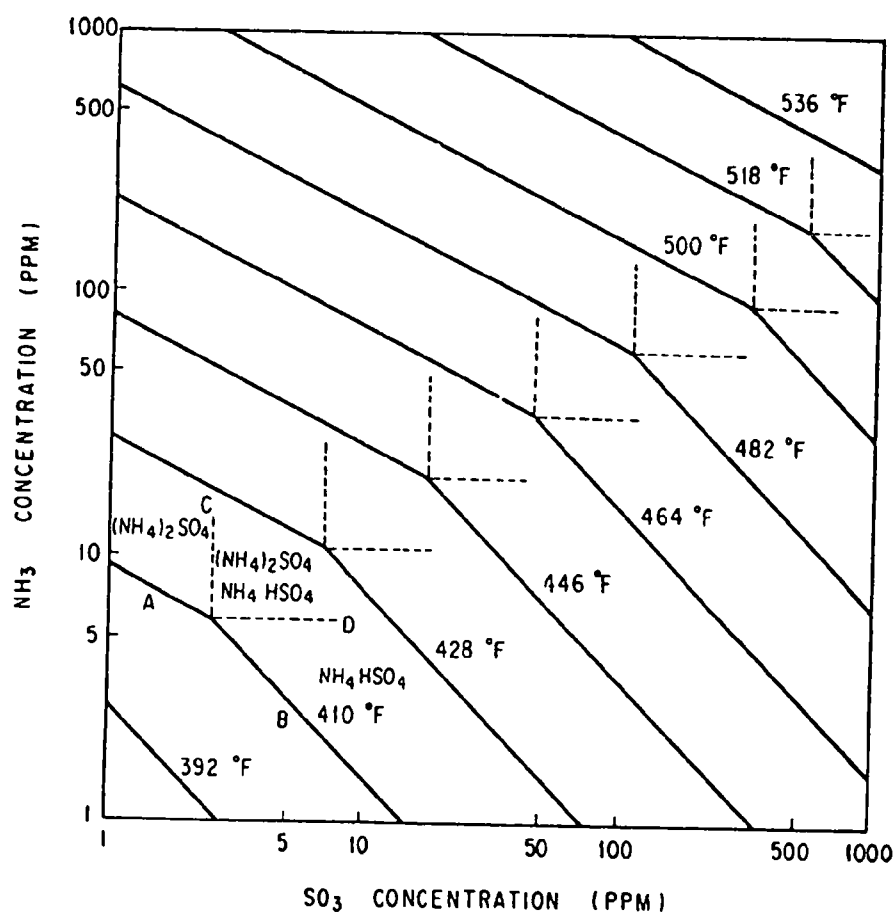
Figure 2-3. Formation of Ammonium Sulfate

(High Temperature Range)

2-12

(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 off of surfaces and by providing surfaces on which the deposits will form instead of forming on metallic surfaces. If preheater deposits do form, sootblowing is effective for removal, as is occasional water washing.

APPLICATION CF PROCESS TO BOILERS

A schematic flow diagram of the HZ process as applied to a boiler is shown in Figure 2-4. 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_x 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. Nitrogen oxides are 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 is then diluted with either air or steam to aid in the distribution of ammonia into the flue gas. The diluted ammonia is fed into the duct through a network of nozzles or perforated pipes at some distance before the reactor to allow complete diffusion 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.

The NO_x removal efficiency depends mainly upon the gas temperature, the ratio of ammonia to NO_x, and the gas velocity. Removals of 80 percent or higher can be readily attained under

2-13

GAS DISTRIBUTION GRIDS

REACTOR INLET

AMMONIA FEED CONNECTIONS

CATALYST

SOOT BLOWER

TREATED FLUE GAS TO BOILER PREHEATER

HOPPERS
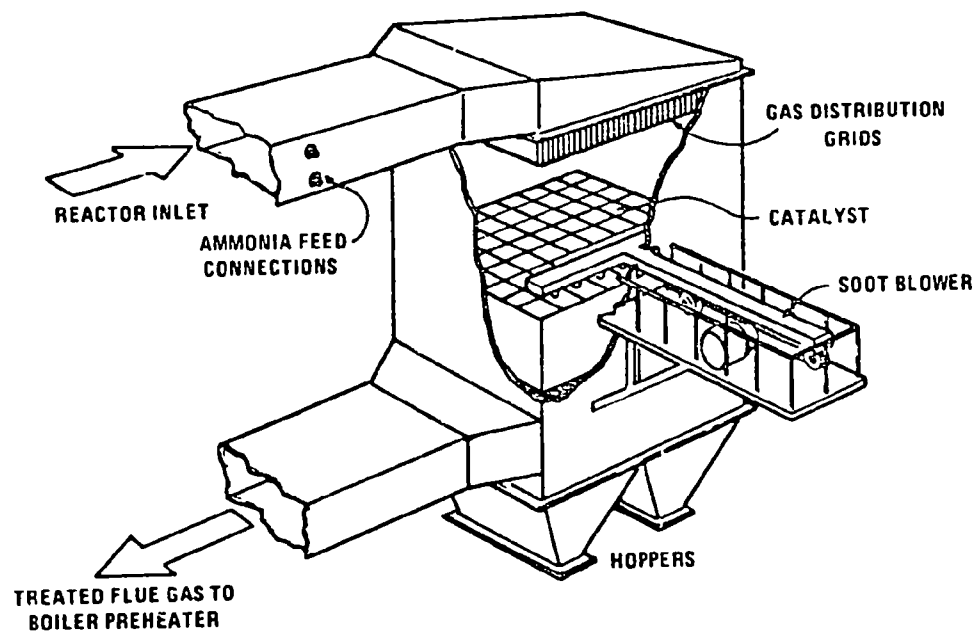
Figure 2-4. Typical Reactor Arrangement

2-14

proper conditions of operation.

Minimum design temperature for the system is around 320°C (608°F). At temperatures lower than this ammonium sulfate or bisulfate produced by the reaction between ammonia and SO_3 may crystallize on the catalyst surface resulting in a decrease of catalyst activity due to adhesion on the catalyst activated surface and/or blockage of the catalyst layer.

The gas temperature exiting the economizer is normally higher than 320°C. In other cases additional heat may be required. This additional heat can be attained by an auxiliary burner which provides high temperature flue gas into the flue gas leaving the economizer. An economizer bypass is also possible. Most of this additional supplied heat would be recovered in the air preheater.

On the other hand, at temperatures higher than approximately 450°C (842°F), while the efficiency of the catalyst for NO_x removal is not affected, the activity of the catalyst for NO_x formation by oxidation of ammonia may become appreciable. This activity, objectionable from the standpoint of NO_x removal, appears at 450-470°C (842-878°F). Although slight at such temperatures, it gradually increases with further increase in temperature. Because of the undesirable effects discussed above, the optimum reaction temperature range is 330-400°C (626-752°F).

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 2-5. Overall, the catalyst has the following characteristics:
    (1) The structure is a thin plate honeycomb.
    (2) Due to substantially reduced pressure drop across the catalyst layer, operating power costs are much lower than with conventional catalysts.
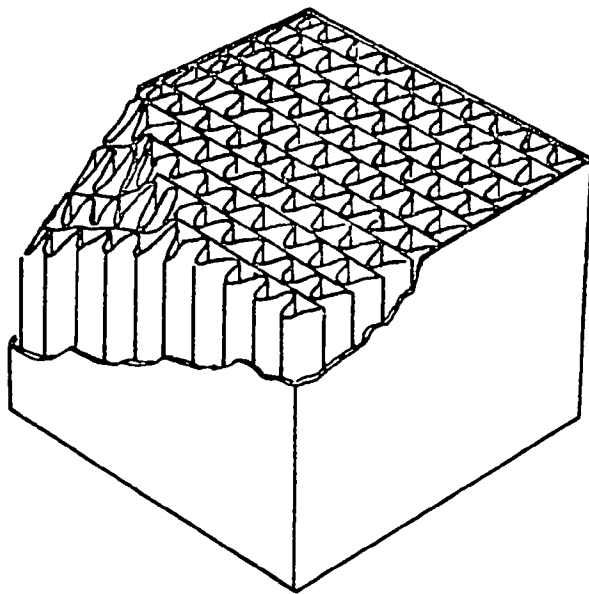
Figure 2-5 Configuration of NOXNON 500 or 600 series catalyst.

(3) A straight gas flow path prevents dust clogging.

(4) It is applicable for gases with high SO_2 concentrations.

As a result, in treating high-temperature gases with high SO_x and dust concentrations such as coal-fired boiler flue gas, the NO_x 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 Series. These catalysts are manufactured in the form of 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 gas passages are created by thin plates folded in a plicated pattern and inserted between flat plates that act to separate one folded plate from adjacent folded plates. The resulting passages are then oriented in the gas stream to be parallel to the direction of gas flow. The problem of erosion by dust is minimized by the selection of the proper linear velocity of flue gas over the catalyst layer. This has been demonstrated at a coal-fired power plant in Japan where tests have been run for several thousand hours without significant deterioration of the 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 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.

REACTOR DESIGN

Reactor vessels housing the catalyst (see Figure 2-6) are of carbon steel construction and are divided into chambers each having its own fly ash hopper. The reactor is oriented so that flue gas flows downward. This minimizes the buildup of fly ash on the reactor internals. 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.

It is important to provide good gas distribution that smooths and evenly distributes the flue gas flow as it enters the reactor. The catalyst cells thereby get equal exposure to the flue gas and gas channeling is decreased. Channeling could lower NO_x removals and decrease ammonia utilization. Fly ash erosion could also result at higher gas velocities through a channeled area.

Proper gas distribution is provided by ensuring, for one thing, that the gas velocity entering the inlet duct is evenly distributed. Even gas distribution at the inlet duct will depend upon the design of the ductwork upstream and turning

GAS IN →

GRIDS

CATALYST

GAS OUT →

FLY ASH HOPPERS
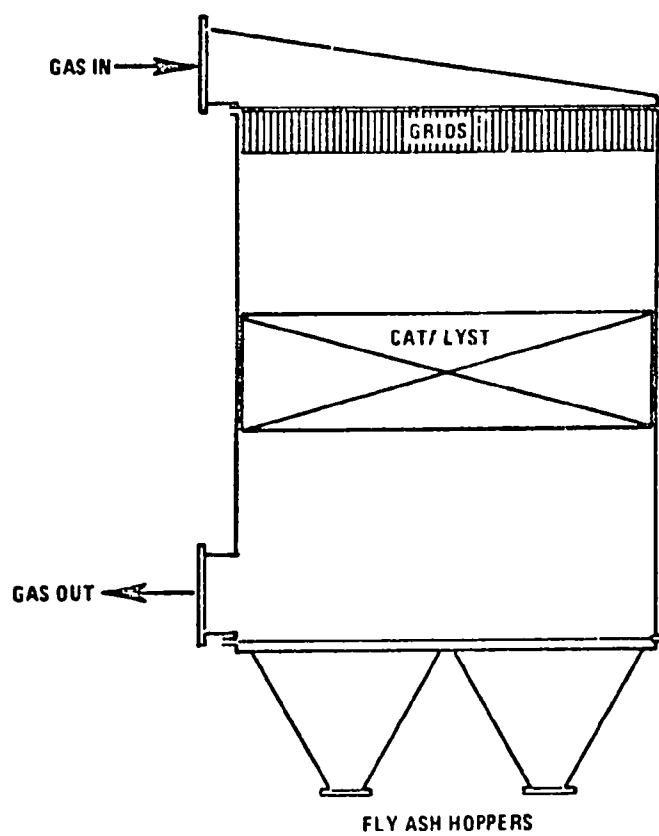
Figure 2-6. Elevation view of typical reactor design

2-19

vanes may be required at bends in the ductwork. Additional design factors for even distribution are incorporated in the reactor design. The slope of the oblique top plate is one of the critical parameters. Specially designed grids above the catalyst bed are also very important. Other factors are the spacing between the bottom of the catalyst layer and the top of the outlet duct, and the width to length ratio of the reactor. These parameters have been established through extensive experimental efforts.

Space above the catalyst bed is also required for the installation of soot blowers and for monorails required for the removal of the catalyst blocks.

SECTION 3

DEVELOPMENT HISTORY

Hitachi Zosen began the development of selective catalysts for NO_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 percent were demonstrated.

Following the successful start-up of the Idemitsu-Kosan system, other plants soon went into operation including a 440,000 NM^3/H (274,000 SCFM) unit at a petrochemical plant, and two plants at steel manufacturing facilities. All of these have operated successfully. (See Table 3-1 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.

One of the primary applications for Hitachi-Zosen's corrugated NOXNON catalyst is the treatment of high temperature gases with high SO_x and high dust concentration such as coal-fired

3-1

TABLE 3-1. LIST OF COMMERCIAL PLANTS BUILT BY HITACHI ZOSEN

	Customer	Treating Capacity, Nm^3/hr	Flue Gas Source	Process	Completion
1.	Osaka Gas Co., Sakai	53,000	LNG or naptha-fired Furnace	Ammonia Reduction	1975
2.	Dakai Engineering, Chiba	5,000	LPG-fired furnace	Ammonia Reduction	1975
3.	Idemitsu Kosan, Chiba	350,000	CO boiler and gas-fired heater	Ammonia Reduction	1975
4.	Shin-Daikyowa Petrochemical, Yokkaichi	440,000	Fuel oil-fired boiler with wet-type desulfurization	Ammonia Reduction	1975
5.	Hitachi Zosen, Osaka	6,000	Gas-fired annealing furnace	Ammonia Reduction	1975
6.	Toshin Steel Mill, Himeji	70,900	Kerosene-fired steel heating furnace	Ammonia Reduction	1976
7.	Kawasaki Steel, Chiba	762,000	Iron ore sintering plant with wet-type desulfurization	Ammonia Reduction	1976
8.	Nippon Satetsu, Himeji	10,000	Fuel oil-fired steel heating furnace	Ammonia Reduction	1977
9.	Maruzen Oil, Sakai	150,000	Fuel oil-fired boiler	Ammonia	1979

boiler flue gases. Due to the catalyst's non-clogging feature, the NO_x removal system reactor can be installed directly behind the economizer. Thus, expensive flue gas pre-treatment 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_x 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 has been at the Electric Power Development Company, Ltd. (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. This has resulted in the collection of a great deal of valuable data. Important developments, particularly in methods of gas distribution, have been achieved. There were three reactors installed, each treating about 200 Nm^3/hr of gas, plus several smaller reactors for abrasion testing. These reactors 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 shut-down 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 percent was still being attained. Ammonia slippage was very low as was the conversion of SO_2 to SO_3. These tests 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.

3-3

SECTION 4

DEMONSTRATION PROGRAM

The success of Hitachi Zosen in the design and construction
of several full-scale denitrification plants in Japan has prompt-
ed the EPA to fund further research on flue gas from coal-fired
boilers. This work was initiated to accurately define the cost,
process performance, complexity, reliability, and the process
impact on the operations of power plant equipment.

The principal potential market for the denitrification of
power plant flue gases will probably be at coal-fired stations.
There has been limited test work done by Hitachi Zosen on flue
gas from coal combustion. The EPA-sponsored demonstration pro-
gram at Plant Mitchell of the Georgia Power Company provided an
excellent site for testing of 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 was collected and evaluated to establish valid characteriz-
ations of the process.

Long term tests were conducted to provide 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
capital cost requirements for commercial installations.

SCOPE OF WORK

Hitachi Zosen, with CAPCC 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 performed in four phases:

Phase I (engineering) started with the basic design package which was prepared by Hitachi Zosen in Japan. Based on these designs, CAPCC prepared a detailed design. This was sufficient for the issuing of requisitions for all equipment, vessels, instruments, electricals, piping, insulation, and a construction subcontract. During Phase I, equipment with unusually long lead times were identified so that procurement of these items could be initiated during Phase I. Also included in Phase I were a detailed capital and operating cost estimate including off-sites and interfaces with the host boiler and spare parts. At the end of Phase I, detailed reports were submitted. These consisted of a Process Design Manual, containing all the drawings, specifications, and engineering documents generated, and a cost estimate for the project.

Phase II included the procurement and construction parts of the pilot unit. All components were procured, fabrication of equipment and vessels was completed, the pilot plant was erected, mechanical acceptability was demonstrated, spare parts and other supplies were obtained, startup and operating personnel were selected and trained, and all arrangements for the purchase of materials and utilities was completed. In addition, an Operating Manual was prepared.

Phase III included startup, debugging, and parametric tests. Parameters and conditions were to be varied so as to optimize the plant performance. The ability of the system to respond to variations in inlet conditions was tested during this phase.

Following the successful completion of the system optimiza-

...tion tests, the pilot plant was to be continuously operated (24
hours/day, 7 days/week) for at least three 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.

An additional series of 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. The object was to quantify any changes in catalyst
activity due to transient conditions. The transient conditions
tested would be similar to those which would be experienced by
an SCR system operated in conjunction with a utility boiler,
including: start-up and shutdown, temperature variations, and
flow rate variations. In addition, tests were to be conducted
to examine the effects of changing the reactor sootblowing
frequency on reactor performance. Further, catalyst regeneration
procedures were to be evaluated.

SCHEDULE

The period of performance for completion of the work relat-
ed to the pilot plant was originally eighteen (18) months. The
award of the contract was May 1978. The design and specification
period, Phase I, took approximately three months. Phase II re-
quired approximately nine months for procurement, fabrication,
transportation and erection. These schedules were as original-
ly expected. However, start-up and debugging required about two
months and so the first tests were not started until August 1,
1979. This first charge of catalyst, however, showed less than
optimum results after some four months of operation and it was
decided to replace it in December of 1979. After some four ad-
ditional months of operation, the second batch of NOXNON 500
catalyst began to provide less than expected results and a
decision was made to install a third charge of catalyst.

4-3

The third charge was a new type: NOXNON 600. This was installed April 18, 1980 and was utilized for some nine months until the plant was shut down on February 2, 1981.

Milestone Dates

Contract Awarded	05/26/78
Phase I Work Plan Submitted	09/15/78
Last Purchase Order Placed	12/18/78
Delivery of First Piece of Equipment	01/24/79
Mechanical Completion	06/15/79
First Catalyst Installed	08/01/79
Second Catalyst Installed	12/10/79
Third Catalyst Installed	04/18/80
Plant Shut Down Permanently	02/02/81

DESCRIPTION OF THE TEST PLANT

The following is a description of the pilot unit including summaries of major items of equipment (see Figure 4-1):

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 fourteen-inch diameter pipeline. The boiler flue gas was withdrawn through three points from the duct to insure that the pilot plant gas is representative of that in the flues, particularly with respect to fly ash (dust) content and particulate size distribution.

The pipeline was insulated and was provided with sampling nozzles for continuous analysis of nitrogen oxides, sulfur dioxide and oxygen.

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 at the temperatures required. The flue gas heater was also used to warm up the catalyst, equipment, and piping to prevent corrosion by the sulfur trioxide
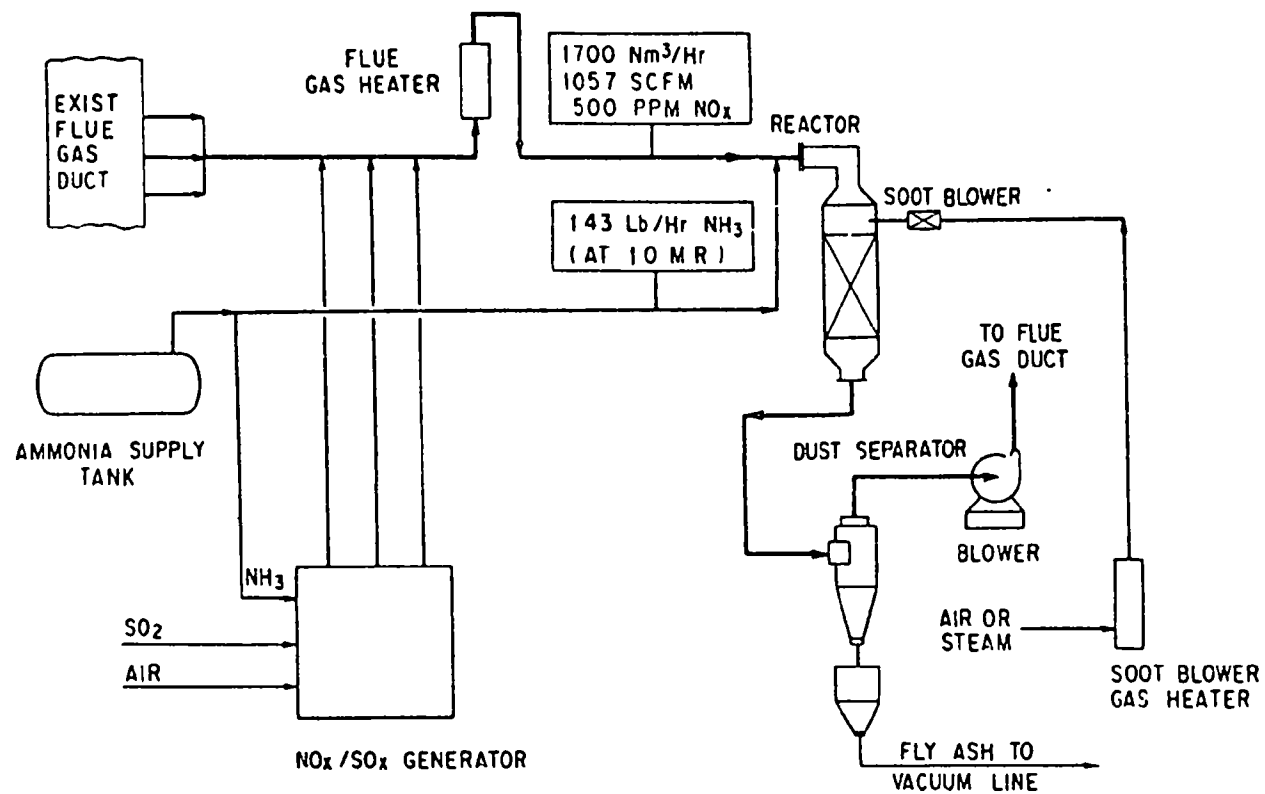
Figure 4-1.   Process Flow Diagram of Pilot Plant

present in the flue gas when starting up from a cold state. At
temperatures below the dew point of sulfur trioxide this corros-
ive compound would condense on the surface of catalyst, equipment
and piping if there were no provisions for warm-up with heated
air. Finally, the flue gas heater was used for purging flue gas
from the catalyst layer, equipment, and piping with hot air
prior to the long-term shutdown of the pilot plant. This was
required to prevent the condensation of sulfur trioxide upon
cooling of the equipment.

Gaseous ammonia was injected into the gas stream after the
heater and before the reactor. The gas then flowed down through
the reactor in contact with the catalyst. The NO_x in the gas
reacted with the ammonia to form small quantities of gaseous
nitrogen and water, which remained in the gas stream.

Fly ash from the flue gas settled in the reactor and would
have tended to block flow through the reactor and blind off and
partly inactivate the catalyst. These effects of dust settling
in the reactor were kept within acceptable bounds through the use
of a soot blowing apparatus which used either steam or air.

A blower was located downstream of the reactor to overcome
the flue gas pressure drop through the pilot plant. A cyclone
dust separator was supplied ahead of the blower to prevent eros-
ion of the blower by dust. The dust was collected in a hopper
below the separator. Dust (ash) collected in the hoppers was
periodically discharged into the boiler plant vacuum system.

The soot blowing gas heater heated compressed air or steam
used for the soot blowing apparatus. Since it was necessary to
perform soot blowing during continuous operation without shutting
down the system or reducing the flow rate of gas to be denitri-
fied, air or steam was heated up to the denitrification reaction
temperature.

The ammonia supply consisted of an ammonia tank with connec-
tion hoses, valves, safety relief valves, excess flow valves,
pressure gauge, and percentage liquid full gauge. The ammonia
supply tank was filled from a delivery trailer when the ammonia

4-6

supply ran low. The ammonia was stored as a liquid at approximately ambient temperature. The ammonia was supplied to the process in a gaseous state and a small electrically heated vaporizer was supplied with the tank.

Since it was planned to test the performance of NO_x, SO_2, SO_3, and since only fixed concentrations of these substances were available from the Albany, Ga. plant boiler operation, SO_2 was purchased for addition to the flue gas, and a NO_x, SO_2 and SO_3 generating unit was provided to manufacture these materials for addition to the flue gas. This permitted testing at NO_x, SO_2 and SO_3 levels higher than those available from the boiler.

The NO_x/SO_x unit consisted of an SO_2 storage facility, an air blower, two small converters, piping and instrumentation and related control systems. One converter, for the oxidation of ammonia, contained platinum gauze catalyst. The other converter, containing vanadium catalyst, was for production of sulfur trioxide from sulfur dioxide.

Compressed air was required for oxidation of NH_3 and SO_2. The air was mixed with ammonia or sulfur dioxide upstream of the converters. The air pressure provided the pressure necessary to inject the produced NO_x and SO_3 into the pilot plant flue gas stream. The unit was arranged in one compact skid-mounted package including a control panel. This unit was fabricated in Japan and transported to the United States.

The operating conditions which were controlled in the pilot unit are as follows:

1. Flue gas flow rate.
2. Temperature of flue gas.
3. The amount of charged ammonia and its pressure.
4. Soot blowing gas pressure and temperature.

CATALYST SPECIFICATIONS

The first two charges of catalyst (as described later in Sections 5 and 6) were replaced. These were both NOXNON 500
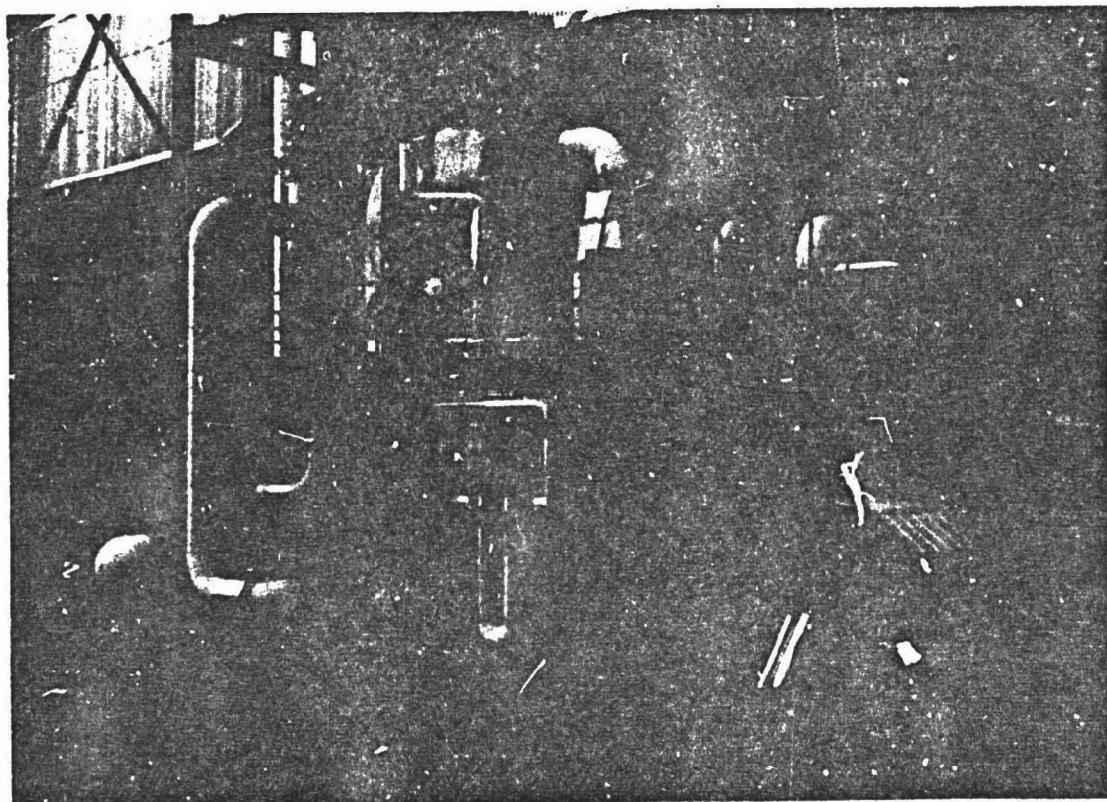
Figure 4-2.   Photograph of the pilot plant.

series. The third charge of catalyst, which was used until the end of the test program, was NOXNON 600. Specifications of these catalyst changes are shown below:

	First Charge Catalyst	Second Charge Catalyst	Third Charge Catalyst
Type	NOXNON 500	NOXNON 500	NOXNON 600
Size			
Length	270 mm	250 mm	525 mm
Width	540 mm	500 mm	525 mm
Height	1500 mm	1500 mm	2130 mm
Actual Catalyst Volume	0.437 M^3	0.375 M^3	0.587 M^3
Surface Area	245 M^2	218 M^2	217 M^2
Pitch	8 mm	8 mm	12.6 mm
Void	0.713	0.746	0.838
Linear Velocity*	1.62 Nm/sec	1.89 Nm/sec	1.71 Nm/sec
Area Velocity*	6.94	7.80	7.83
Space Velocity*	3,890	4,580	2,900

* Flue gas flow rate at 1,057 scfm (1,700 Nm^3/h)

CONTROL SYSTEM

In the control system of the EPA pilot plant, the system was operated under a set NH_3/NO_x mole ratio. The flue gas flow rate and incoming NO_x concentration were measured and the signal was multiplied to provide the mass flow of NO_x entering into the reactor. This quantity was then converted to the set mole ratio to determine the amount of ammonia required.

This system showed reliable controllability in the pilot plant. However, the control system for commercial plants would differ slightly from the pilot plant control system.

Due to the very large flow rate, the direct measurement of flue gas may not be practical, and some other measurements which are proporional to the boiler load, for example, the amount of fuel burned in the boiler, the steam generated, or the power

generated are available as representing the plant load instead of the direct measurement of flue gas flow rate.

The product of the above signal and the concentration of NO_x at the inlet of reactor would be used as a control signal to regulate the selected NH_3/NO_x mole ratio or the selected NO_x removal efficiency. This feed forward system provides a fairly accurate control of ammonia flow rate. Along with this the other feedback systems which measure the outlet concentration of NO_x, or possibly the NH_3 emission, can provide signals to fine tune the ammonia feed rate.

HOST SITE

The pilot plant was located at the Unit #3 of Plant Mitchell, Georgia Power Co., Albany, Georgia. This unit has a pulverized coal-fired Combustion Engineering boiler which was initially operated 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 outdoors of the Unit #3.

Georgia Power Co. is headquartered in Atlanta and serves about a million customers in Georgia. Plant Mitchell has three units, two of which have nameplate ratings of 22.5 MW while the Number 3 is rated at 125 MW.

The boiler normally burns coal from Kentucky or other mid-south coals with sulfur contents below 2 percent. The coal composition can vary widely. Typical flue gas compositions to the pilot plant are shown in Table 4-1 below.

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TABLE 4-1 TYPICAL FLUE GAS COMPOSITION

Component	Concentration
N_2	74.8%
O_2	3-7%
CO_2	13.8%
H_2O	8.6%
NO_x	500 ppm
SO_2	400 ppm
SO_3	5 ppm

Gas temperatures were 550-650°F and the dust content of the gas was typically 6.6 grains/SCFD. The pressure at the inlet duct to the pilot plant was about 6 inches H_2O.

TABLE 4-2 TYPICAL COAL ANALYSES

AS BURNED COAL ANALYSES

DATE	MOISTURE (%)	ASH (%)	SULFUR (%)	BTU/lb
05/08/80	5.50	11.92	1.26	12,368
05/19/80	4.28	12.98	1.40	12,414
07/11/80	4.92	10.58	1.04	12,564
08/05/80	4.11	11.87	1.32	12,549
08/11/80	4.60	13.37	1.26	12,250
10/09/80	4.32	10.33	1.22	12,840
10/21/80	3.72	9.43	1.19	13,059
11/10/80	4.05	10.62	1.13	12,835
11/17/80	3.48	10/92	1.18	12,874
12/09/80	3.99	12.38	1.11	12,544
01/07/81	4.12	11.63	1.24	12,709

DATA COLLECTION

The pilot plant operation was closely monitored through the use of an array of instruments and analyzers. Continuous analyzers of the latest design provided measurements of the NO_x

4-11

level both entering and leaving the reactor.

In addition, similar analyzers were to be used for measuring the ammonia slippage past the reactor. These analyzers were manufactured by Thermal Electron Corporation and use the principle of chemiluminescence. The ammonia was to be measured by splitting a sample of flue gas into two portions, converting the ammonia in one of the portions to NO_x, analyzing NO_x levels in each of the portions, and the difference in the analyses would be a measure of the ammonia level.

The measurements that were recorded by CAPCC personnel during the plant operations are shown in Table 4-3.

TABLE 4-3 DATA COLLECTED

Parameter	Method of Measurement
Temperature	
Flue Gas Heater Inlet	Duplex thermocouple
Reactor Inlet	Duplex thermocouple
Reactor Outlet	Duplex thermocouple
Soot Blower Inlet	Duplex thermocouple
Pressure	
Pressure drop across reactor	Pressure difference
Gas Flow Rate	
Flue Gas Flow Rate	Venturi flow meter
NH_3 Flow Rate	Rotameter with magnetic float
Analytical Measurements	
Inlet and Outlet NO_x	Chemiluminescence
Inlet or Outlet SO_2	Pulsed Fluorescent
Outlet NH_3	Chemiluminescence
Inlet O_2	Paramagnetic
Other Recorded Data	
Percent NO_x Removal	Calculated automatically
Mole ratio NH_3/NO_x	Calculated automatically

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In addition to the data collected by CAPCC personnel, out-side stack testing consultants were employed to take air pollution measurements of SO_2, NO_x, particulates, and HCl. This was done to check the analytical instruments and to obtain data for which no analyzers are installed.

Much of the data at the pilot plant was collected in a data logger which frequently scanned the instruments and stored the data. Visual display and paper tape display were provided. It was originally intended that the data logger would store the data on magnetic tapes in cassettes. The cassettes were to be sent to CAPCC's New York office to be transmitted through a terminal to a computer. This data would then be processed by electronic data processing methods to evaluate various factors and then stored for future use. However, the magnetic tape recorded was found to be incompatible with the equipment in New York and the tapes could not be used.

PROGRAM MANAGEMENT

For the overall management of the project, Mr. Shingo Tanaka was appointed project manager by Hitachi Zosen. Mr. Tanaka directed the progress of the work and coordinated with EPA and with CAPCC. The Project Director, Mr. H. Inaba, was located in Tokyo and supplied overall direction, superintending the efforts of those department managers concerned with research, development, design, procurement, erection, and commercial aspects of Hitachi Zosen's NO_x removal process.

Mr. Richard Wiener represented CAPCC on this project and acted as Project Coordinator. He was responsible for the contract between Hitachi Zosen and CAPCC and for liaison with EPA, Georgia Power, and Hitachi Zosen.

The Project Engineer for CAPCC was Mr. Rafat Morcos who was responsible for all in-house activities (technical and commercial) during the engineering, procurement, and erection stages of the project. The Field Engineer was Mr. Atef Demian.

The Project Officer for EPA was Mr. J. David Mobley who was responsible for the entire project. Working with EPA was the Radian Corp. of Austin, Texas providing technical support and reviewing all aspects of the project.

EXPECTED RESULTS

The purpose of the program was to demonstrate the application of the catalytic process to coal-fired boilers for 90 percent reduction of NO_x emissions. It was expected that the program would demonstrate the applicability and efficiency of the process for the removal of high percentages of nitrogen oxide from flue gas typical of that produced from boilers burning U.S. coal. The program tested several parameters so that when the tests are completed it would be possible to predict the removal efficiencies under various conditions of operation.

The energy requirements could be evaluated by determining the pressure drops across the catalyst reactor over a period of time. Unexpected increases in pressure drop would of course add to the energy needs of the process. Sootblowing capabilities could establish the effectiveness of this approach to the maintenance of low pressure drops.

Materials of construction, although not a problem in this process, were evaluated to determine if unusual corrosion effects were found. Most of the equipment was of carbon steel construction which was suitable for this process. High temperature operation could cause problems with some materials and this was to be studied.

The analytical system for NO_x, SO_2, and NH_3 was of considerable interest and the effectiveness, accuracy, and reliability of these instruments could be applied to commercial applications. Chemiluminescence analysis, if successfully applied here, would be used in full-scale plants. The SO_2 analyzer (Pulsed fluorescent) would also be followed closely for future use.

The control system was essentially the same as that which

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would be used for commercial plants. The normal control method would use a feed-forward control in which the flue gas flow and NO_x concentration in the feed will be used to set the ammonia feed. The success or failure of the control methods could be applied to commercial designs.

Additional items which were tested during the demonstration program are the instruments and controls and the handling by data logger. Certain variables such as "percent NO_x removal" and "mole ratio" were calculated and recorded. All of these systems were evaluated for commercial applications.

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 remain the same and the gas distribution through the catalyst bed was properly designed. From these pilot plant results the gas velocity required to produce a certain removal efficiency could be determined and the amount of catalyst required could, therefore, be calculated.

Ammonia consumption could also be determined from the test results because it is calculated as a mole ratio of ammonia to NO_x in the flue gas. Ammonia slippage would also be expected to be the same in a commercial unit.

Energy consumption is a function of the pressure drop through the catalyst bed and through the reactor inlet, reactor outlet, dampers, ducts, etc. This test work could only be used to establish the expected pressure drop through the catalyst bed. A commercial unit would produce different pressure drops depending upon the plant arrangement. However, these pressure drops can normally be calculated and the major pressure drop of interest is the one through the bed of catalyst.

The results from the pilot plant should be close to that

·expected in a commercial unit except ror variations in the type
of ash and the design of the soot blowers. The expected life
of the catalyst is about two years. The testing, however, could
only extend about six months. Extrapolation of any measurable
decrease in reactivity would give an indication of the expected
catalyst life.
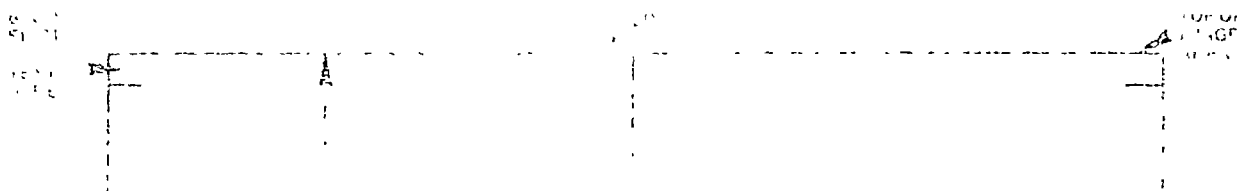
SECTION 5

NOXNON 500 TESTS

PRELIMINARY OPERATIONS

On June 4, 1979 construction of the pilot plant was essentially completed. Checking of the piping and instruments was then begun by CAPCC and HZ personnel. Orientation lectures for the training of newly hired operators was started a week later and trial practice runs were provided for the operators starting on June 18th.

Debugging of the plant required several weeks and the plant was not ready for operation until the end of July. Many of the problems were related to the electrical work. This included errors in the wiring installations, faulty connections in the motor control center, and mistakes in the connections to the control panel. In addition to the electrical system, several adjustments were required for the analyzer system to make it operable. Another problem was with the blower motor which burned out and required replacing.

Before the NOXNON 500 catalyst was loaded into the reactor the whole flue gas system was operated with circulating air. This was for training of the operators, for calibration and adjustment of the controllers, for testing of the Blower and Flue Gas Heater, for checking of the sample lines to the analyzer, and for general testing of the whole system.

On July 16, 1979 continuous operation with flue gas was started without the catalyst. During this time various systems were tested including the analyzers, controllers, soot blower,

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ammonia supply system, NO_x/SO_x generator, and ash removal equip-
ment.

Although calibration and adjustment of the analyzers and
controllers were not completed, it was decided to install the
catalyst into the reactor on August 2nd, which was approximately
one month later than originally scheduled.

FIRST CHARGE

On August 2, 1979 the first charge of NOXNON 500 was loaded
into the reactor and the testing commenced. Testing continued
until the beginning of December providing 2168 hours of operating
time on flue gas and 154 hours on hot air.

Overall data with the first charge of NOXNON 500 is shown in
Figure 5-1. When operations were initiated with flue gas at the
beginning of August, 1979, the removal efficiency was better than
90 percent at design conditions at 1.0 mole ratio. The removal
seemed to stabilize at approximately 92-93 percent with a 1.5
inches H_2O pressure drop across the reactor. This efficiency
was attained until the middle of September.

On September 14th, after about 900 hours of operating time
the pressure drop suddenly increased and the removal of NO_x
dropped sharply. The next day the plant was shut down and the
top surface of the catalyst layer was inspected from the man-
hole of the reactor. About half of its cross-section was found
to be covered with a thick layer of fly ash. It appeared that
the fly ash had built up on the side of the reactor in front of
the manhole and had then slid down for some reason covering a
large part of the catalyst. It was decided to close up the
reactor again and operate the soot blower for several cycles
with superheated steam. This reduced the pressure drop from
1.85 inches to the normal 1.4. Removal efficiency also increased
significantly.

It should be explained that up to this time the sootblower
had not been operated. This was based on experience at the

5-2
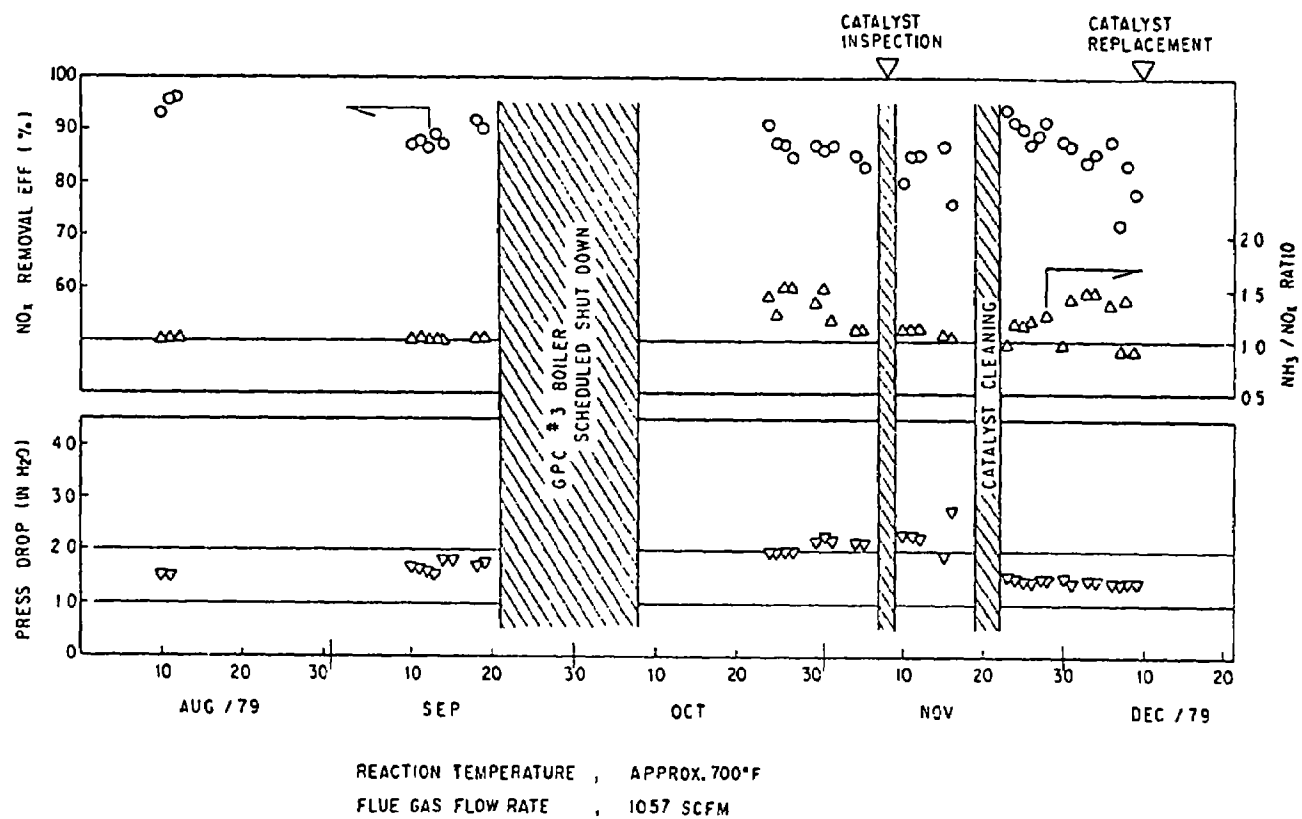
Figure 5-1. Operating Data with the First Charge of NOXNON 500.

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FIRST
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Isogo Power Station pilot plant in Japan. Similar reactors
there operated on fly ash loaded flue gas for long periods of
time without the need for sootblowing. Clogging by fly ash
did not occur. The sootblower was installed at the EPA pilot
plant only for the purpose of emergency use. It was not plan-
ned to be run regularly. However, differences in the fly ash
characteristics were obvious and the decision was made to
operate the sootblower at least once a day from this date.

By the middle of October (operating time 1170 hours) the
pressure drop appeared to be gradually increasing. The pressure
drop continued to increase and on October 30th there was a rapid
increase to 3.2 inches H_2O. The sootblower was operated repeat-
edly so as to continue the operation. This measure brought a
stable pressure drop of approximately 2.4 inches H_2O, still some-
what high. However, the NO_x removal efficiency was not restored.

On November 9th the reactor and ductwork was dismantled to
find the reasons for the low removal efficiency. It was sur-
prising to discover that the ductwork leading to the reactor was
heavily built up with deposits of fly ash. About three-quarters
of the elbow before the reactor was plugged. The reason for the
fly ash deposit was the expansion of the gas in the transition
piece which was located in the horizontal. The lowered velocity
permitted the particulates to drop out at that point. Deposits
were also found at the sootblower opening and at the manhole
shelf.

The catalyst assembly could only be cleaned by lifting it
out of the reactor and setting it on the ground. The upper sur-
face of the catalyst looked to be full of fly ash. Most of the
openings were full. An air hose was used to clean out most of
the fly ash. Further quantities of fly ash were removed by
poking rods into the openings.

The ductwork was modified so that the transition piece was
located in a vertical position above the reactor. Baffle plates
were installed at the manhole and at the sootblower opening to
prevent fly ash deposits at those locations. The catalyst was

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reinstalled and the system was bolted together and put back into operation on November 23rd.

The initial results indicated better NO_x removals. However, after about a half-day the removal dropped below 90 percent although the pressure drop was substantially lower than before cleaning of the catalyst.

On November 30th, the system was shut down for an inspection of the catalyst through the manhole. The surface looked clean and the changes in ductwork, along with the baffles at the manhole and at the sootblower, had been effective in avoiding piles of fly ash. However, inspection of catalyst by insertion of rods indicated that about 30 to 35 percent of the openings were plugged again.

The catalyst in place was rodded out as far as possible and compressed air was also used to try to clean it. After this effort, the system was once more operated to evalute the NO_x removal efficiency by varying the mole ratio. NO_x removals were found to be mostly in the 80-85 percent range even with sootblowing three times per day.

A decision was made to replace the catalyst as it appeared that the activity had decreased to the point where the expected 90 percent removal could not be attained. The catalyst had been in service some 2500 hours. The reason for the activity decline was not certain. However, it appears that there were two major reasons. One was the buildup of the fly ash within the layers of the catalyst caused in part by the inappropriate duct design thus blocking off a portion of the surface. The second major cause was a definite detrition of active material from the metallic substrate. Possibly 10 to 20 percent of the catalyst may have been lost.

On December 10, 1979 the original catalyst load was replaced by the spare catalyst which had been stored at the site. The removed catalyst was inspected and tests were run in an effort to determine the cause of the loss of activity. This is discussed in a later section, "Catalyst Evaluation."

The second charge of catalyst was loaded into the reactor on December 10, 1979. This charge was the same as the original except that it was in six blocks instead of four. The total volume, composition, and pitch were the same.

The system was started up again on the evening of December 10th and removals of over 90 percent were readily attained. Figure 5-2 shows overall performance with the second charge of NOXNON 500 catalyst. Based on the experience of the first charge, the sootblower was operated three times a day from the beginning. This frequency seemed the minimum required to prevent clogging by fly ash. The system was operated until March 3, 1980 with a stable 90-92 percent NO_x removal and a pressure drop of less than two inches H_2O.

Typically, over a two day period of February 25th and 26th the NO_x removal averaged 93.1 percent. The pressure drop across the catalyst bed was 1.72 inches H_2O. (These results are all at a flue gas rate of 1057 SCFM, a reactor inlet temperature of 710°F, and a mole ratio of 1.0).

On March 4th the pilot plant was shut down so that the flue gas heaters could be replaced. Several of the elements had burned out. The shut down and start up followed the normal procedure. When the system went back into operation, it was found that the operating results were significantly different. The following data are for the indicated days showing the average of percent NO_x removal and pressure drop:

3/5	91.8%	2.43 inches H_2O
3/6	92.8	2.25
3/7	91.4	2.45
3/9	89.8	2.59

There was a small but significant drop in removal but a relatively large increase in pressure drop. All of the instruments and controls were thoroughly checked and found to be sat-

CATALYST INSPECTION

CATALYST INSPECTION AND CLEANING

NOₓ REMOVAL EFF (%)

100
90
80
70
60

REACTION TEMP : APPROX 700 °F
FLUE GAS FLOW RATE : APPROX 1,057 SCFM

REPLACEMENT OF ELEMENTS IN FLUE GAS HEATER

CHRISTMAS HOLIDAYS

GPC #3 BOILER SHUT DOWN

GPC #3 BOILER SHUT DOWN

GPC #3 BOILER SHUT DOWN

NH₃/NOₓ RATIO

20
15
10
05

PRESS DROP (IN H₂O)

40
30
20
10
0

10 20 30 10 20 30 10 20 10 20 30 10 20

DEC /79   JAN /80   FEB   MAR   APR /80

Figure 5-2. Operating Data with the Second Charge of NOXNON 500.

isfactory. The cause of the pressure drop increase was of concern.

It was decided to inspect the surface of the catalyst as there was a possiblility that the shut down may have caused an influx of fly ash onto the catalyst. The system was shut down again by proper procedures, the manhole was opened for inspection, the inspection was made, the manhole was closed and the system restarted. No unusual accumulations of fly ash were seen on the top of the catalyst bed.

Surprisingly, when the system was restarted the removals decreased substantially and the pressure drop went up even higher.

3/10	84.0%	3.25 inches H_2O
3/11	85.6	3.22

No cause for this drop in performance could be found and once more all the instrument systems were checked and found to be working well.

On March 14th the system was closed down again, the catalyst was removed, each of 'the six blocks were cleaned with compressed air, and then replaced. The pressure drop went down but the removals did not improve. It appeared that there was a permanent loss of catalyst activity.

3/15	86.2%	2.12 inches H_2O
3/16	86.6	2.09
3/17	87.0	2.00
3/18	86.2	2.04

On March 21st the boiler was shut down for two weeks of maintenance. It was decided to circulate hot air through the reactor so as to avoid cooling it down. For two weeks the temperature was maintained at 400°F and then increased to 550°F two days prior to the reintroduction of flue gas. On April 9th the pilot plant was restarted and was run until the next day when the boiler was forced to go down due to a tube leak.

The results were even worst this time. A six hour period on April 11th indicated an average removal of 78.2 percent and

·a pressure drop of 2.17 inches. (At a mole ratio of 1.01, 1056 SCFM, 724°F). For some reason the removal had decreased from the previous 86 percent even though no flue gas had been run through it and a cooling and reheating cycle had not been experienced.

There was no apparent cause found for this degeneration. However, it seemed obvious that the catalyst was no longer acceptable. The catalyst was then removed and a new charge of catalyst was installed on the week of April 14th. The catalyst had been in operation with flue gas for 2184 hours.

When the Type 500 catalyst which had been removed from the reactor was inspected it was found to be in relatively good physical shape. For an inspection, several plates were stripped off of several of the blocks. (There were six blocks piled in three layers). There was certainly a quantity of fly ash deposited within the internal layers, but when this was brushed off or the plate was lightly tapped, the surface appeared mechanically sound. Except for the top two or three inches of the upper layer, there was no indication of erosion. Also, there was no sign of delamination, cracking, or peeling of the active material. The upper few inches was worn down to the stainless base due to the action of the fly ash or soot blower. But the remainder of the five feet length of catalyst was in excellent shape.

The cause of the loss of activity could have been due to the accumulation of fly ash or to a coating of the surface by some foreign material or to some other reason. Samples were prepared and sent to Japan for examination along with samples of fly ash. See "Catalyst Evaluation" for the results of these tests.

A thorough investigation of the operating data during the shut downs and start ups was done and it indicated that all proper procedures were followed. There was probably no chance of sulfur trioxide condensing during any of these times because when flue gas was present the temperatures were always above 400°F, which is considerably above the dewpoint.

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CATALYST EVALUATION

Inspection of the first charge of catalyst

The first charge of catalyst was disassembled on the ground.
The frame box was cut, each block of catalyst was opened, and
each corrugated plate and flat plate was pulled out of the block
for inspection. There were four blocks of catalyst, two on the
upper level and two on the lower level.

Abrasion --

Partial abrasion was observed at the upper part of the
catalyst. The depth of the abraded surface of catalyst was
about 25 cm (10 inches) at maximum and 5 cm (2 inches) at minimum.
Therefore, the amount of abraded surface was estimated at less
than 15 percent of the total catalyst surface. (The term abraded
surface refers to the surface where all or a majority of catal-
ytic components had come off and base metal of stainless steel
was exposed.)

Plugging by fly ash--

The top surface and bottom surface looked relatively clean.
Less than 15 percent of the openings were plugged. Periodic
operation of the soot blower appeared to have been effective.
However, when all the flat plates and corrugated plates were
pulled out of the box, it was found that almost all of the sur-
face except for the abraded portion was covered with accumula-
tions of fly ash. Passages for flue gas were originally tri-
angular but due to the accumulation of fly ash they appeared
round. The lower blocks, especially, featured the above-mention-
ed accumulations. This accumulation was rather hard, it could
be scraped off by rods, but did not seem to be removable by soot
blower.

Test pieces for catalytic activity examination were taken from the top catalyst blocks. Figure 5-3 describes the location of test pieces cut out of the top catalyst block. Figure 5-4 shows a schematic explanation of the testing apparatus.

Experimental Conditions--The following conditions were maintained in the testing apparatus:

Area Velocity : 20 $Nm^3/m^2.h$

Size of Test Pieces : 20 mm x 50 mm

Composition of mixed gas (volume on wet basis)

NO	390 ppm
NH_3	390 ppm
SO_2	225 ppm
O_2	3.6%
CO_2	10.8%
H_2O	10.0%
N_2	balance

Experimental Data--Catalytic activity of the test pieces were examined along with three virgin test pieces which were produced at the same time as the first charged catalyst and kept in storage by Hitachi Zosen.

Catalytic activity of the unwashed used catalyst was tested at first with the test pieces. Afterwards they were tested after washing with water at 40°C (104°F) in order to remove adhered rly ash from the activated surface, and then compared with the test pieces of virgin catalyst.

Results--Figure 5-5 shows the results of catalytic activity testings.

- Catalytic performance of the upper section of used catalyst which was abraded by fly ash or by soot blowing with steam had decreased considerably.
- Catalytic activity of the lower section which had not
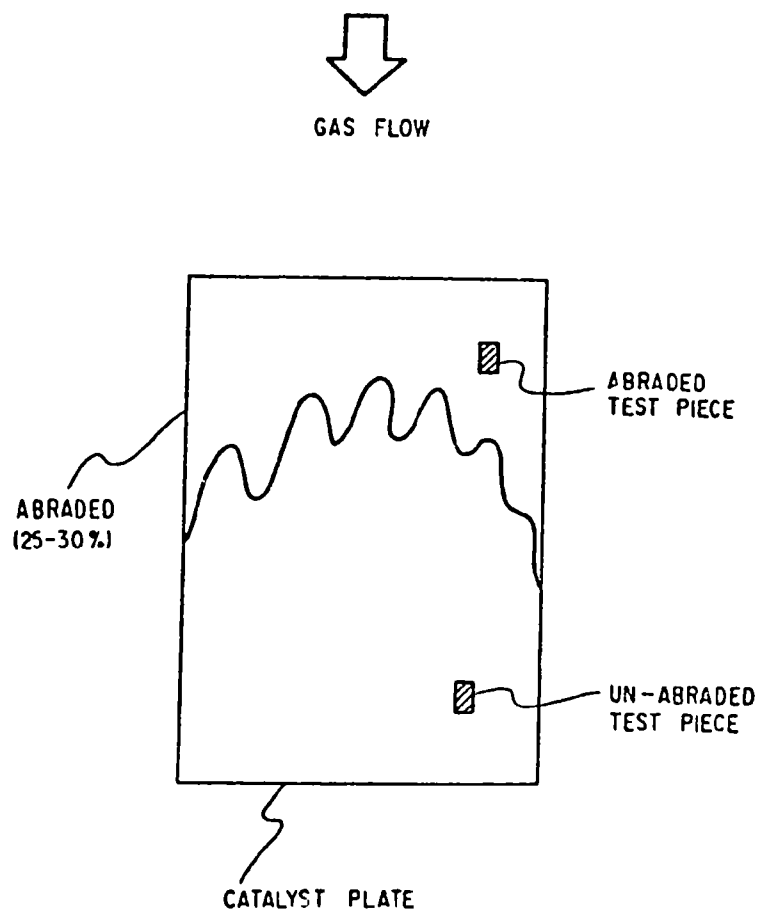
5-11

Figure 5-3. Location of Test Pieces.

Figure 5-4. Test Equipment for Determining Catalyst Activity.

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SYMBOL	CATALYST / DESCRIPTION
△	VIRGIN CATALYST
○	UN-ABRADED (AFTER WASHING)
●	UN-ABRADED (BEFORE WASHING)
◆	ABRADED

TEST CONDITIONS

TEST PIECE SIZE $20^{mm} \times 50^{mm}$ (PLATE)

A V $20 \ Nm^3/m^2 \ H$

GAS COMPOSITION (WET BASIS)

NO	390	PPM
NH_3	390	PPM
SO_2	225	PPM
O_2	3 6	%
CO_2	10 8	%
H_2O	10 0	%
N_2	BALANCE	

NH_3/NO MOLE RATIO = 10
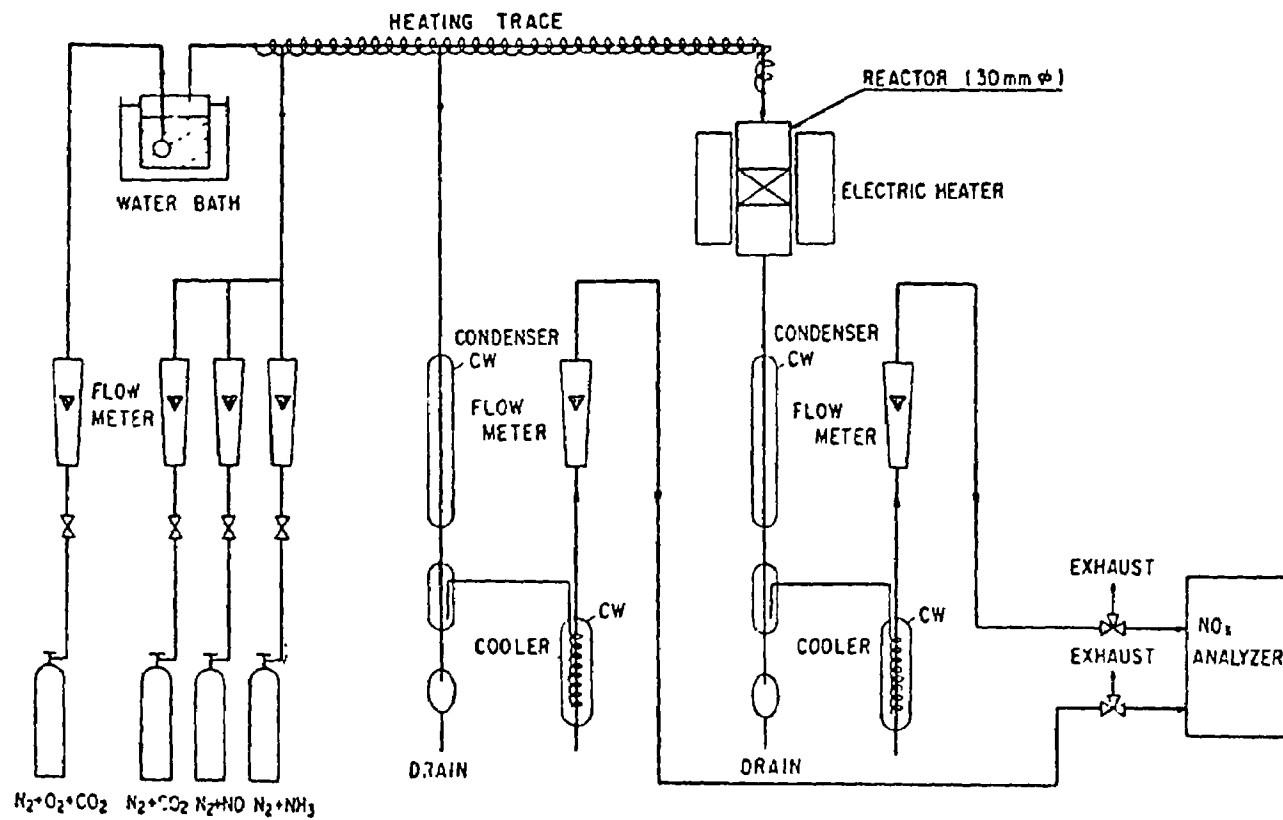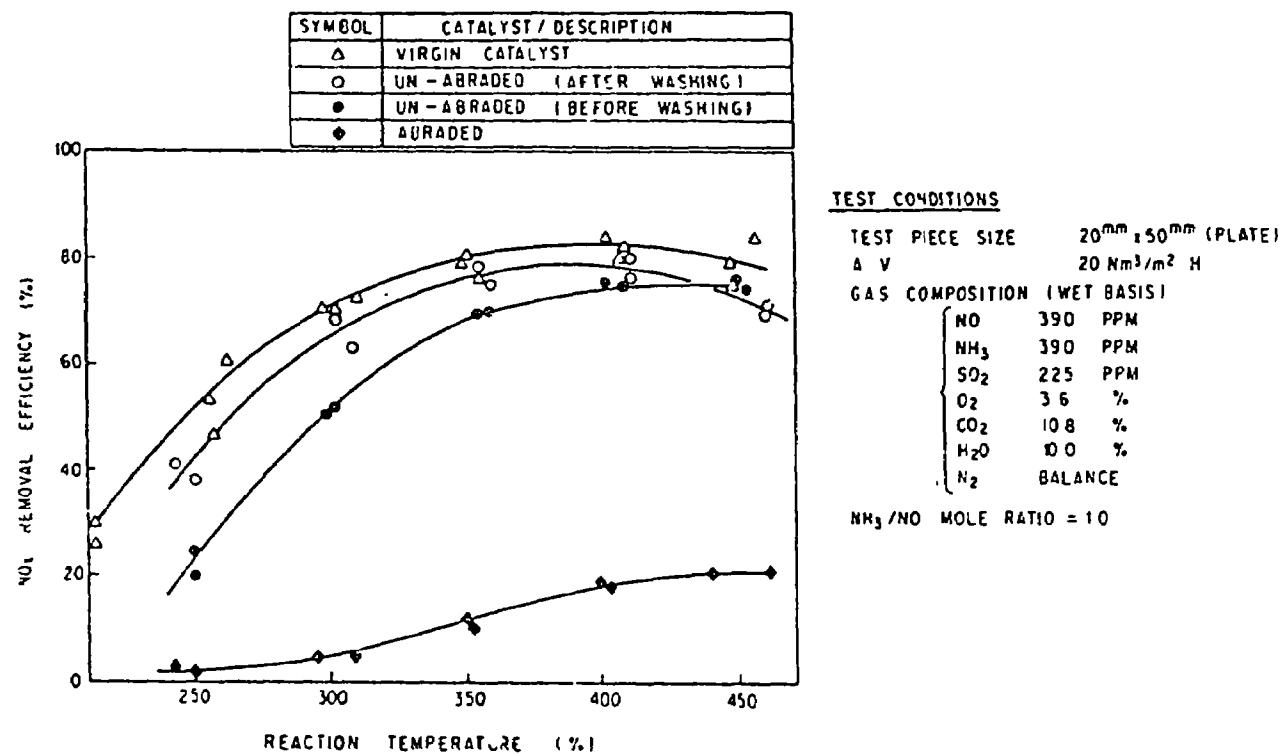
Figure 5-5. Evaluation of Used NOXNON 500 (First Charge).

abraded but was masked by adhered fly ash also decreased.
- Testing of the lower section after water washing showed
  a slight decrease of catalytic performance in comparison
  with the virgin catalyst. However, it was concluded that
  the catalytic performance of the lower and unabraded
  section still retained its original activity after the
  adhered fly ash was removed from the surface.

Inspection of the Second Charge of Catalyst

The second charge of catalyst were taken out of the reactor
and disassembled on the ground on April 11, 1980. There were six
blocks of catalyst. Two at the top, two in the middle, and two
at the bottom.

Abrasion--
   Abrasion was observed mainly at the upper part of the top
catalyst blocks and slightly lower side of the top block. There
appeared to be no abrasion in the middle and bottom blocks. The
ratio of abraded surface area to the total surface area was
estimated to be about 10 percent. Abrasion seemed more severe
under the sootblower discharge nozzles.

Plugging--
   Inspections were first made of the six blocks of catalyst
prior to removal of the catalyst from their containers. The
upper surfaces varied in the degree of apparent plugging. The
top catalyst appeared relatively clean with only about 5 to 7
percent of the channels plugged. The middle blocks, however,
were significantly plugged, estimated at 40 to 50 percent of the
total number of channels. Also, about 25 to 30 percent of the
channels in the bottom blocks appeared plugged.
   Three bundles of catalyst were taken out of the containers,
one each from the top, middle, and bottom. These were dis-
·assembled for inspection. A substantial amount of adhered and

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accumulated fly ash was found in a majority of the channels between the catalyst plates, especially in the middle bundle, although narrow paths for flue gas still remained.

Catalytic Activity--

The test pieces for catalytic activity examination were cut out of the top and bottom catalyst bundles respectively. The size of test pieces, testing apparatus, and the experimental conditions were the same as those used for the first charge of catalyst.

Experimental Data--Catalytic activity of the test pieces taken from the top and bottom catalyst bundles were tested before and after washing with water and shown in Figure 5-6 and was compared with the fresh catalyst.

Results--Catalyst activity before water washing was considerably lower than the original. However, with water washing the activity was restored almost to the original activity.

Investigation of the Deterioration of Catalyst

The application of NOXNON 500 to coal-fired flue gas was examined at the pilot plants of EPDC's Isogo Power Station and at the Georgia Power Company's Plant Mitchell. Radical differences between the two plants in respect to clogging by fly ash were observed.

At the EPDC pilot plant, the flue gas source had been divided into two streams. One stream was pre-treated through an electrostatic precipitator or cyclone dust separator, producing a low-fly-ash-containing flue gas. The other stream was not pretreated, so that high-fly-ash-containing flue gas was sent directly into the catalytic reactors. There were not soot blowers operating in either case.

At the EPDC pilot plant, clogging by fly ash did not occur.

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SYMBOL	CATALYST / DESCRIPTION
△	VIRGIN CATALYST
○	TOP CATALYST (AFTER WASHING)
●	TOP CATALYST (BEFORE WASHING)
□	BOTTOM CATALYST (AFTER WASHING)
◙	BOTTOM CATALYST (BEFORE WASHING)

TEST CONDITIONS

TEST PIECE SIZE . $20^{mm} \times 50^{mm}$ (PLATE)
A V · $20 \, Nm^3/m^2 \, H$
GAS COMPOSITION (WET BASIS)
- NO 390 PPM
- NH_3 390 PPM
- SO_2 225 PPM
- O_2 3 6 %
- CO_2 10 8 %
- H_2O 10 0 %
- N_2 BALANCE

NH_3/NO MOLE RATIO = 10
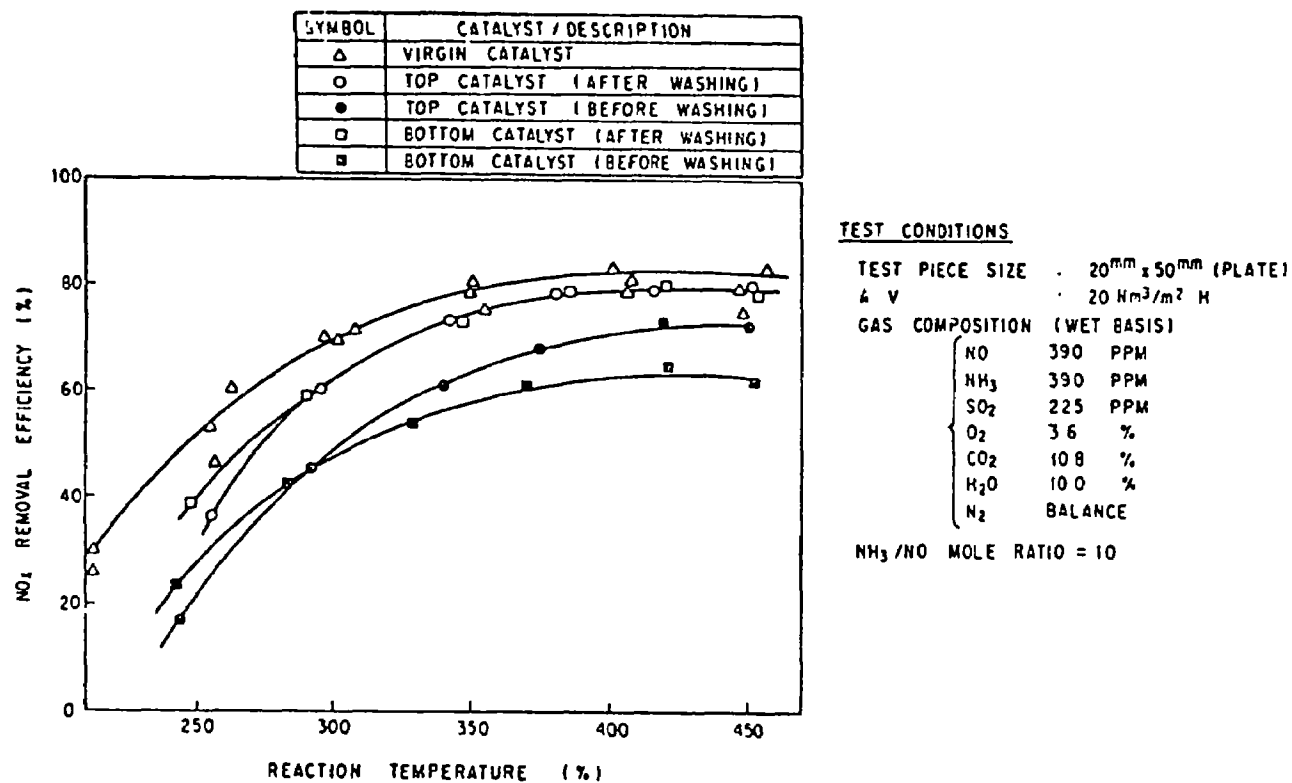
Figure 5-6. Evaluation of Used NOXNON 500 (Second Charge).

The low-fly-ash-loaded flue gas produced an increase in pressure drop, but no clogging. The high-fly-ash-containing flue gas also did not cause clogging and there was no increase in pressure drop. Pressure drop was stable and constant during more than 8,000 hours of operation.

However, at the EPA pilot plant NO_x removal efficiency decreased and pressure drop increased with both the first charge and the second charge of catalyst after about 2200 hours of operation. A major factor for these undesirable results was the character and the behavior of the fly ash which was obviously different from that of the fly ash at the EPDC pilot plant.

In order to investigate the cause of the decrease in NO_x removal efficiency and increase in pressure drop, the following studies were conducted.

Measurement of Particle Size Distribution of Fly Ash--

Samples--The following samples were collected:

Sample	Date	Location
A	09/15/79	upper surface of catalyst bed
B	11/08/79	top catalyst block
C	11/08/79	bottom catalyst block
D	11/08/79	upper surface of catalyst bed
E	04/17/80	top catalyst block
F	04/17/80	middle catalyst block
G	04/17/80	bottom catalyst block

Method--Particulate size distribution was measured by electron microscope.

<u>Results--</u>

Sample	Median Diameter (Micron)	Rosin-Rammler's Index No.*	Log Standard Geometric Deviation
A	13	3.2	1.6
B	21	3.0	1.7
C	15	3.3	1.8
D	16	3.3	2.0
E	18	3.1	1.8
F	19	3.4	1.6
G	18	3.2	1.6

* The above Rosin-Rammler's distribution is based on ANSI/ASTM E 20-68 and DIN 66145.

The results showed no significant difference in particle size at the various sampling points.

Pore Size Distribution of Catalyst--

Pore size distribution of NOXNON 500 catalyst was measured by a Mercury Penetration Method and is shown in Figure 5-7.

The results indicate that the pore size of the catalyst is very small, measured in angstrom units, compared to the particle size of the fly ash, measured in microns. There could be no possibility of the pores of the catalyst being plugged by fly ash.

Chemical Analysis of Fly Ash--

<u>Methods</u>--100 grams of fly ash were pulverized to less than 100 mesh, dried at 120°C for two hours, and one gram of this was weighed accurately.

This one gram of fly ash was mixed with concentrated HCl and HNO_3 and evaporated to dryness, heated with HCl solution, dissolved and filtered.

Al, Fe, V, Ti, Mg, Ca, Na, K and Li were then determined by an atomic absorption spectrophotometer from the filtrate. 10%
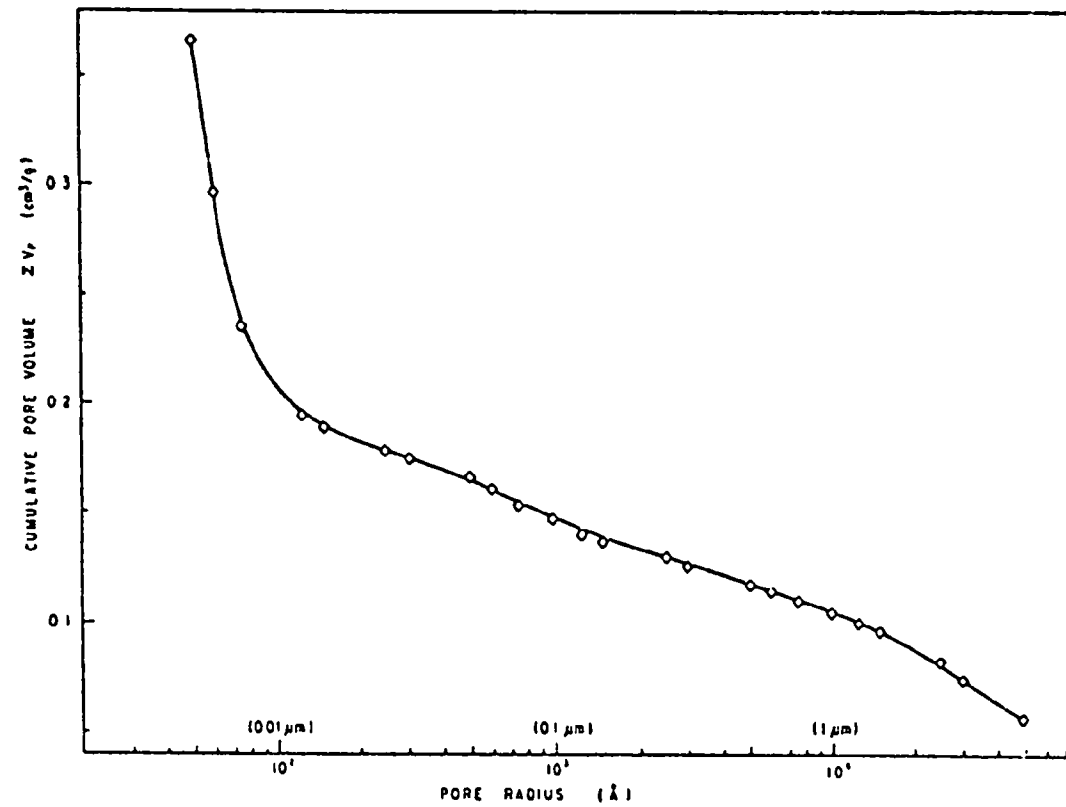
Figure 5-7. Pore Size Distribution of NOXNON 500.

of $BaCl_2$ solution was added to a definite amount of filtrate, and SO_4 group was determined by measuring the weight of precipitated $BaSO_4$.

The residue was ignited at 800°C and SiO_2 in the residue was vaporized with HF, and a decrease in weight was determined as the amount of SiO_2.

Non-volatile residue was melted with $K_2S_2O_7$, dissolved into water, and the amount of Al, Fe, V, Ti, Mg, Ca and Li was determined by an atomic absorption spectrophotometer and added to the amount included in the filtrate.

Ammonia was extracted with hot water and determined by an Indophenol process.

Results--In order to investigate the characteristics of Georgia Power fly ash, a comparison was made of the chemical analysis of the Georgia Power fly ash and a typical fly ash produced from a Japanese utility company.

Table 5-1 describes the chemical analysis of the fly ash collected from Georgia Power and a Japanese utility. R_f shown at the bottom of the table was cited from "K.H. Haller: Design of Large Coal Fired Steam Generators, Babcock & Wilcox Co., Technical Paper BR-1082, 1 - 8 (1977)," and shows the fouling tendency of fly ash in relationship to the composition of the fly ash as described in Table 5-2.

The analytical results in Table 5-1 show no particular reason for the causes of the shorter catalyst life in the EPA pilot plant. From the fouling factor evaluation (R_f) it would seem that Japanese fly ash would have more of a tendency to foul than the Georgia Power fly ash. The only significant analytical difference appears to be the potassium level which is about ten times higher in the Georgia Power Co. fly ash. However, it is not known whether this would be a factor that would increase fouling tendencies. As neither the chemical analysis nor the fouling factor seemed to offer an explanation for the shorter catalyst life, further investigations were required.

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TABLE 5-1   CHEMICAL ANALYSIS OF FLY ASH (wt. %)

	GEORGIA POWER COMPANY PLANT MITCHELL								TYPICAL JAPANESE FLY ASH					
DATE SAMPLED	Oct/78	9/15/79	11/8/79	11/8/79	11/8/79	4/17/80	4/17/80	4/17/80						
LOCATION SAMPLED	ESP	Top of Catalyst	Upper Catalyst	Bottom Catalyst	Top of Catalyst Layer	Top Catalyst	Middle Catalyst	Bottom Catalyst	Low Fly Ash	Low Fly Ash	High Fly Ash	High Fly Ash	Low Fly Ash	Low Fly Ash
FLUE GAS				HIGH FLY ASH LOADING FLUE GAS										
SiO_2	50.5	53.1	52.0	51.4	51.7	46.0	44.0	41.0	44.3	45.1	46.0	47.8	41.7	44.5
Al_2O_3	29.1	26.2	26.3	26.3	25.7	28.2	27.0	26.8	21.4	21.5	22.3	22.3	20.8	22.5
Fe_2O_3	11.0	9.0	10.2	9.4	9.7	10.0	10.7	9.87	5.0	5.2	6.3	6.4	12.0	5.7
V_2O_5	0.04	0.04	0.04	0.11	0.04	0.036	0.071	0.161	0.071	0.079	0.024	0.024	0.034	0.055
TiO_2	2.3	1.8	1.8	2.0	1.8	2.84	2.84	3.34	1.37	1.44	1.22	1.22	1.34	1.25
Na_2O	0.067	0.11	0.14	0.16	0.14	0.162	0.162	0.189	0.96	0.90	0.36	0.28	0.35	0.59
K_2O	0.241	0.96	0.96	1.75	0.55	1.18	1.45	1.45	0.085	0.082	0.162	0.165	0.191	0.246
LiO_2	0.034	0.079	0.068	0.073	0.056	0.022	0.024	0.026	0.027	0.025	0.010	0.011	0.011	0.017
MgO	0.76	0.78	0.91	0.76	0.81	0.75	0.90	0.85	1.69	1.76	2.73	1.96	2.03	1.88
CaO	0.161	0.01	0.60	0.07	0.29	0.38	0.15	0.15	0.80	1.05	6.34	6.86	4.55	4.72
SO_4	5.1	1.2	2.4	5.2	2.0	3.4	6.0	6.5	10.8	9.1	3.2	1.9	4.3	5.3
NH_4	nil	0.005	0.012	0.006	0.019	0.006	0.011	0.014	0.297	0.044	0.010	0.08	0.020	0.013
TOTAL	99.30	94.18	95.43	97.23	93.21	93.0	93.3	90.3	86.8	86.3	86.8	89.0	87.3	86.8
R_ℓ	0.26	0.06	0.13	0.27	0.10	0.18	0.36	0.38	0.46	0.40	0.23	0.14	0.43	0.34

TABLE 5-2   FOULING FACTOR OF FLY ASH

Bituminous Ash

Defined as $(CaO + MgO) < Fe_2O_3$

$$\text{Fouling Factor } (R_f) = \frac{CaO + MgO + Fe_2O_3 + Na_2O + K_2O}{SiO_2 + Al_2O_3 + TiO_2}$$

R_f	Fouling Tendency
below 0.2	low
0.2 - 0.5	medium
0.5 - 1.0	high
above 1.0	severe

Lignitic Ash

Defined as $(CaO + MgO) > Fe_2O_3$

Na_2O (%)	Fouling Tendency
below 3	medium
3 - 6	high
above 6	severe

Influence on Catalyst Activity Caused by Fly Ash--

In order to evaluate the influence on catalyst activity by fly ash a test piece of catalyst (50 mm x 20 mm) was put into fly ash and kept at a temperature of 380°C (716°F) for approximately 500 hours. The fly ash had been collected at Georgia Power Company, Plant Mitchell on September 15, 1979 from the upper surface of catalyst layer, and chemical analysis data is shown in Table 5-1.

200 milliliters of this fly ash was put into a steel cylinder and the test piece was buried in the fly ash and kept at 380°C for 500 hours in an electric furnace. This method, though simple, demonstrates that when any component which is poisonous to catalyst activity is present in fly ash, the catalyst is deteriorated by this test.

The catalytic activity of the test piece after this test is shown in Figure 5-8 along with its initial activity. The results proved that there was no component in the fly ash which deteriorated catalytic performance of NOXNON 500.

Observation of Fly Ash by Spectroelectronic Microscope--

By observing fly ash with a spectroelectronic microscope, Georgia Power fly ash was found to contain some porous substance which may have been an unburned hydrocarbon or unburned pulverized coal. Japanese fly ash does not contain these porous substances.

Shown on Figure 5-9 are microphotographs of Georgia Power fly ash at magnifications of 2700 and 900. They suggest that some high boiling point compound condensed and acted as an adhesive leading to agglomeration or conglutination of the particles of fly ash.

Thermal Processing of Fly Ash--

The above microscopic examination of the fly ash indicated the possibility of a compound found in Georgia Power fly ash which was a high boiling point compound. However, the mechanism

SYMBOL	TEST PIECES PROCESSED
O	VIRGIN CATALYST
△	500 HRS ON STREAM
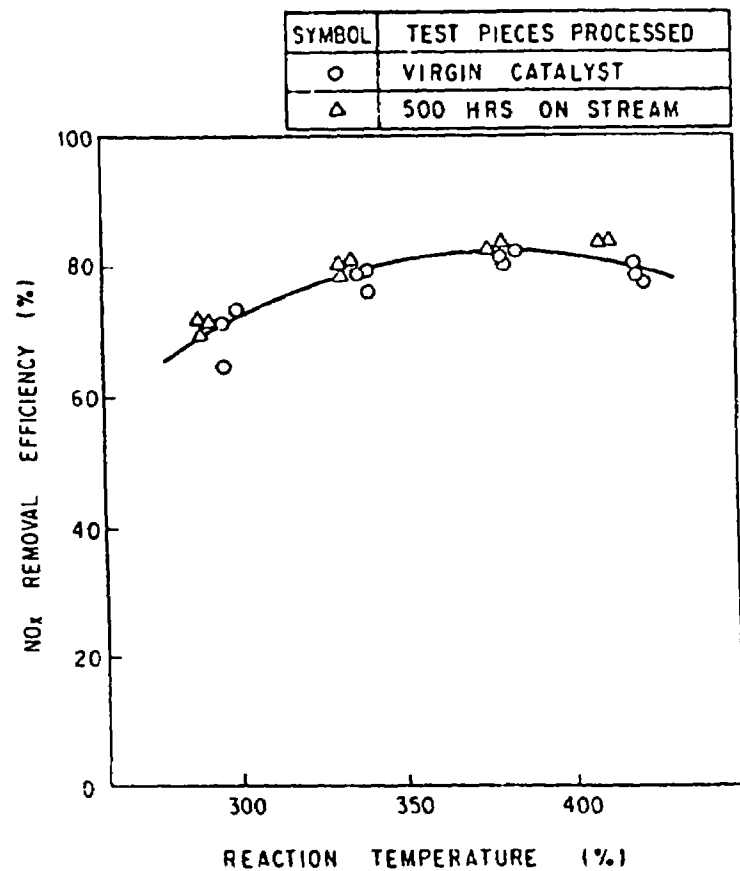
TEST CONDITIONS

A V , 24 Nm^3/m^2 H

GAS COMPOSITION (WET BASIS)

NO	220	PPM
NH$_3$	220	PPM
O$_2$	5	%
CO$_2$	10	%
H$_2$O	10	%
N$_2$	BALANCE	
NH$_3$/NO M R = 10		

PROCESSING TEMP : 380 °C
FLY ASH : FROM GPC

Figure 5-8. Effect of Thermal Processing in Fly Ash.
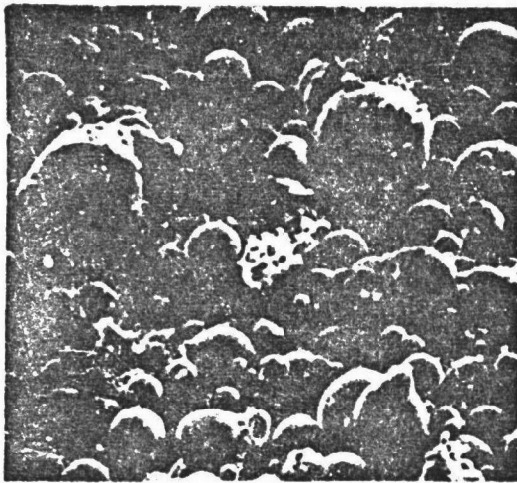
FLY ASH FROM ESP   (x900)
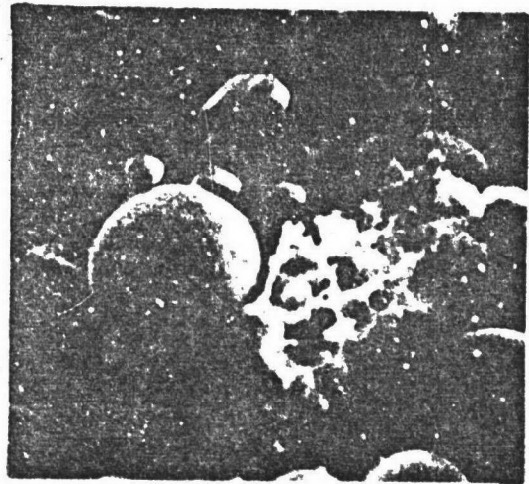
FLY ASH FROM ESP   (x2700)
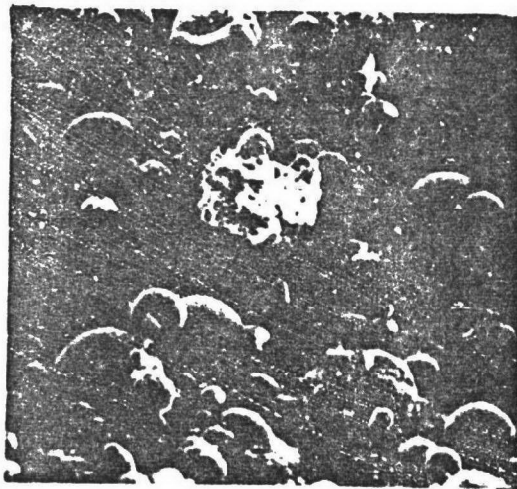
FLY ASH FROM TOP CATALYST
                    (x900)

FLY ASH FROM TOP CATALYST
                    (x2700)

Figure 5-9. Microphotographs of Fly Ash

of agglomeration was not clear. Therefore, the behavior of fly ash mixed with coal at the operating temperature was investigated.

Since the temperature and time are functions which influence adhesion and agglomeration of fly ash, the following qualitative tests were run using Georgia Power fly ash as well as EPDC fly ash to investigate cohesive characteristics of Georgia Power fly ash.

Fly ash from Georgia Power and EPDC was screened on a 70 mesh screen, introduced into lidded steel cylinders of 30 millimeters diameter, and processed at the conditions listed below in an electric furnace.

	PRETREATMENT	TEMPERATURE	TIME
Step 1	screened through 70 mesh sieve.	380°C	100 hours
Step 2	after Step 1 is finished, again screened through 70 mesh.	380°C	150 hours
Step 3	after Step 2 is finished, screened through 70 mesh sieve and mixed with 5% active carbon.	380°C	150 hours
Step 4	after Step 3 is finished, screened through 70 mesh sieve and mixed with 5% pulverized coal from EPDC.	380°C	200 hours

Figure 5-10 show photographs of fly ash after these thermal processing steps were completed.

The photographs from Step 1 at the top show that Georgia Power fly ash produced large size agglomerates which have the same diameter as the internal diameter of the steel cylinders used for the thermal processing after Step 1 processing, whereas EPDC fly ash did not agglomerate.

The photographs from Step 2 show that Georgia Power fly ash again produced agglomerates although the size was smaller. EPDC fly ash did not agglomerate.

From the Step 3 photographs it was observed that 5% active

Step 1 Processing
        Fly Ash of GPC

Screened by 70 mesh sieve.
380°C x 100 Hours

Step 1 Processing
        Fly Ash of EPDC

Screened by 70 mesh sieve.
380°C x 100 Hours

Step 2 Processing (GPC)
After Step 1, screened
again by 70 mesh.
380°C x 150 Hours

Step 2 Processing (EPDC)
After Step 1, screened
again by 70 mesh.
380°C x 150 Hours
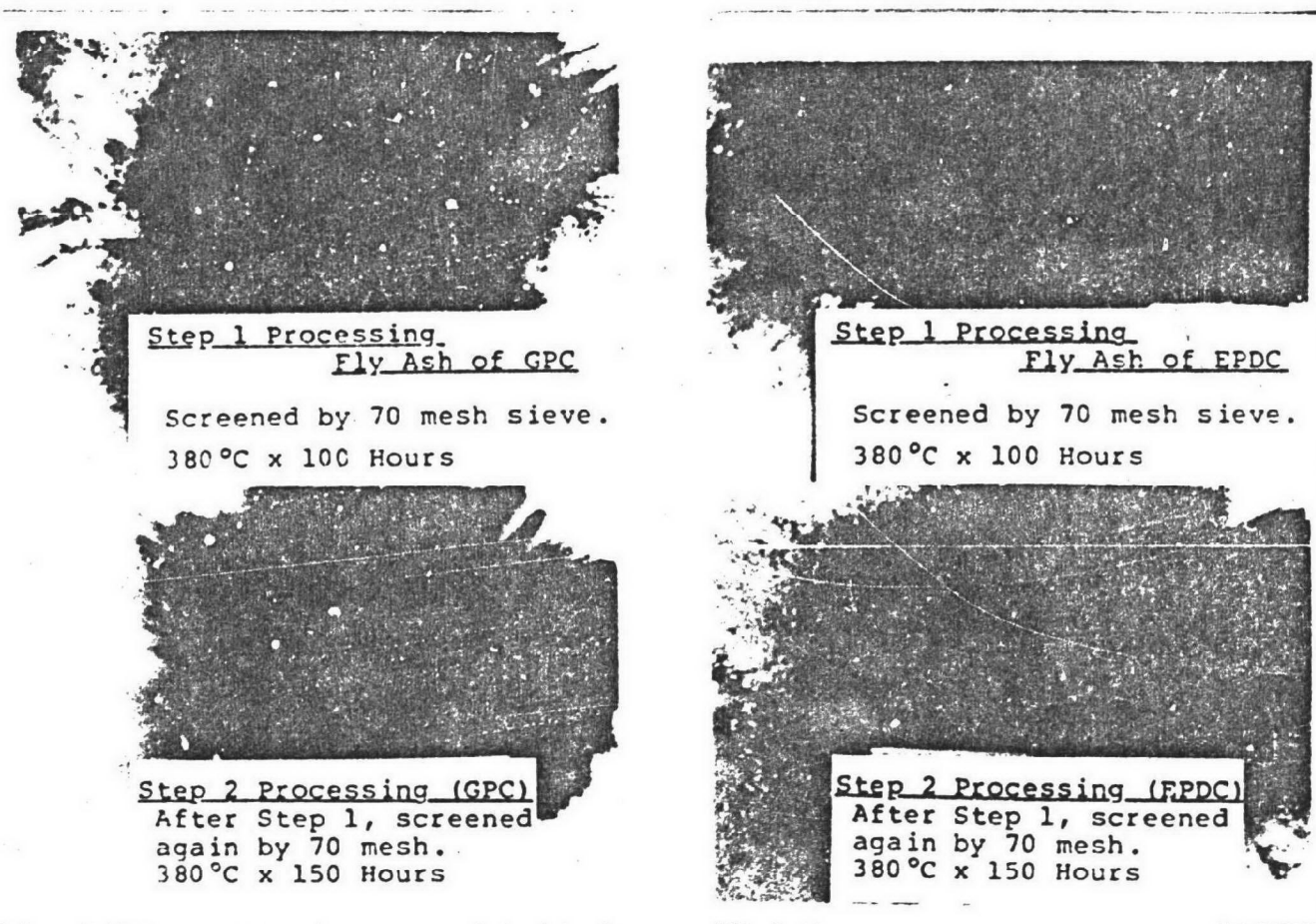
Figure 5-10.   Photographs of Fly Ash after Thermal Processing.

Step 3 Processing (GPC)
After Step 2, screened
again and mixed with
active carbon.
380°C x 150 Hours

Step 3 Processing (EPDC)
After Step 2, screened
again and mixed with
active carbon.
380°C x 150 Hours

Step 4 Processing (GPC)
After Step 3, screened
again and mixed with
pulverized coal.
380°C x 200 Hours

Step 4 Processing (EPDC)
After Step 3, screened
again and mixed with
pulverized coal.
380°C x 200 Hours
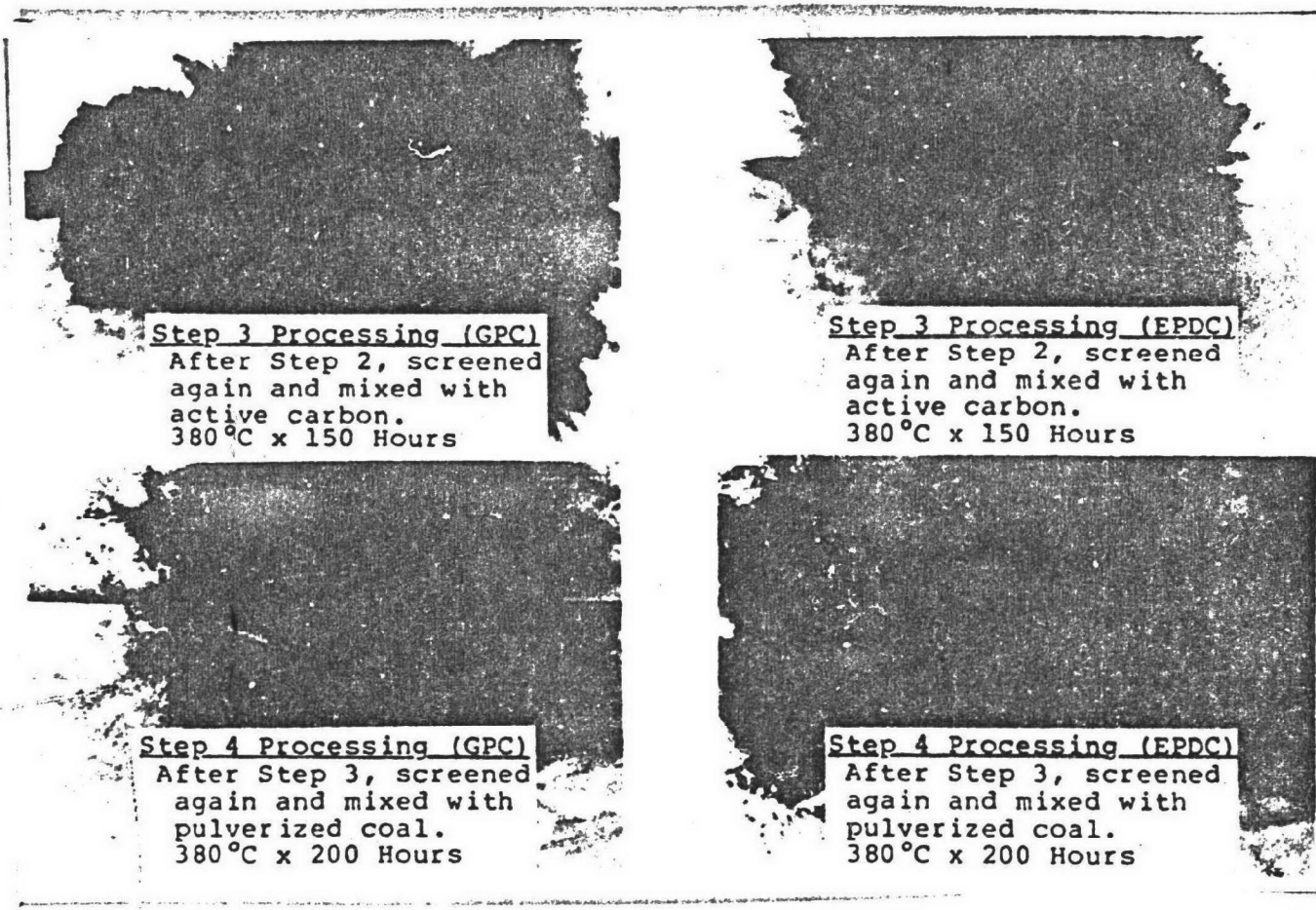
Figure 5-10 (continued). Photographs of Fly Ash after Thermal Processing

carbon mixed with fly ash seemed not to promote the cohesion of fly ash. However, a small amount of granular agglomerates were found in Georgia Power fly ash.

The bottom two photographs showed proof that Step 4 processing produced large size agglomerates for Georgia Power fly ash, whereas there were very small size agglomerates in EPDC fly ash.

From the above quantitiative testings,

- Georgia Power fly ash appears more cohesive than EPDC fly ash.

- Unburned pulverized coal or hydrocarbons derived from coal appear to seriously increase the cohesion of fly ash at the operating temperature of SCR.

Discussion

NOXNON 500 catalyst was replaced twice due to an increase in pressure drop and a decrease in NO_x removal efficiency after operations of approximately 2,100 hours and 2,300 hours. Phenomena observed were:

Catalytic activity did not deteriorate--

After the fly ash which adhered to the surface and was plugging the channels of the catalyst was removed, the catalyst itself retained its original catalytic performance.

Fly ash produced in the Unit No. 3 boiler does not include components poisonous to the catalyst.

Abrasion was observed at the upper part of the catalyst--

It is unknown whether the abrasion was caused by ash-cut or condensate in the steam used for the soot blower. However, the ratio of abraded surface to the total catalyst surface was not high, probably less than 10 to 15 percent. The abrasion, of course, contributed to the decrease in NO_x removal efficiency, but this was not a major factor compared with fly ash plugging.

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·Accumulation of fly ash on the catalyst surface--

    Since this accumulation was caused mainly by electrostatic
forces (Van der Waals Force), it could be removed easily by the
soot blower operation. During catalyst cleaning a tremendous
amount of fly ash was removed by blowing with compressed air.

Chemical analysis data of fly ash--

    These did not lead to any definite explanation for the plug-
ging of the catalyst channels by fly ash. However, fly ash pro-
duced in the Plant Mitchell Boiler No. 3 does agglomerate at the
reactor operating temperatures. Inspection of the catalyst,
which was disassembled at the site, indicated that the agglomer-
ated fly ash caused plugging of the channels between the catalyst
layers. The mechanism of agglomeration is not clear. However,
since the agglomeration leads to adhesion of fly ash in the cat-
alyst channels, fly ash adhering to the catalyst surface should
be removed as soon as possible by soot blowing.

Soot Blower Operations--

    It was concluded that the soot blower should be operated for
the treatment of Appalachian-produced coal-fired combustion flue
gas. The minimum operation frequency was determined from exper-
ience.

    However, a problem still remains. There seems to be a
possibility that abrasion of catalyst was caused by condensate
which existed in steam pipes between the nozzles of soot blower
and the block valve when the soot blower was not operated. Also,
this condensate may cause agglomeration of fly ash leading to
plugging of the catalyst. Therefore, steam for the soot blower
was repaced with air to prevent possible abrasion caused by con-
densate.

SECTION 6

NOXNON 600 TESTS

INTRODUCTION

     Primarily because of clogging of the catalyst channels by
fly ash due to narrow clearances and partially due to abrasion
by fly ash and by soot blowing, the initial charge and the second
charge of catalyst did not achieve their expected performances.
The installation and testing of NOXNON 600 catalyst which has
wider channels was proposed by Hitachi Zosen. This plan was
accepted by EPA.

     The development of NOXNON 600 catalyst is based on the con-
cept that a highly active catalyst which consists of thin plates
should be the most effective. Therefore, NOXNON 600 is produced
from thin stainless steel wire mesh as a base metal to give mech-
anical strength to which catalytic components are cemented. The
thickness of the catalyst is approximately 0.8 millimeters and
the principle catalytic components are vanadium and titanium
oxide.

     NOXNON 600 has been proven in applications on oil-fired and
coal-fired combustion flue gas in Japan.

     Before NOXNON 600 was installed in the reactor, the reactor
had to be rebuilt because the length of the NOXNON 600 catalyst
bed was longer than the one of NOXNON 500. The soot-blower was
changed to use hot air instead of superheated steam.

     Starting from April 22, 1980, the demonstration operation
with NOXNON 600 continued for more than nine months until Feb-
ruary 2, 1981. The operation with combustion flue gas from the

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Unit #3 Boiler of Georgia Power Company, Plant Mitchell exceeded 5,600 hours.

The pilot plant program required achieving NO_x removal efficiency of more than 90 percent continuously for a period of more than three months. Afterwards, the project scope was extended and transient tests were included in the scope of the Contract along with an extension of the operating period.

Tests with NOXNON 600 were, in general, as follows:

Catalyst life tests were run to confirm the expected catalyst life. From April 22, 1980, the pilot plant was operated maintaining NO_x removal efficiency of more than 90 percent until the end of October. After October, a nominal 80 percent NO_x removal was accepted in order to decrease ammonia slip as far as possible. However, from time to time, operating conditions such as flue gas flow rate, temperature, and mole ratio of ammonia to NO_x were varied to evaluate catalyst performance.

Following the catalyst life test, further testing of the Hitachi Zosen catalyst was needed to establish its suitability for commercial applications on coal-fired boilers. The purpose of this testing was, first, to determine the effects of transient conditions on the catalyst; second, to provide an extended operating time so that at least 5,000 hours of operation could be obtained to evaluate the long term effectiveness of the NOXNON 600. During this testing period, catalytic performance was evaluated by varying the flow rate, temperature and mole ratio. Transient tests consisted of power plant load excursions, emergency shut-off of ammonia feed, cold start-up, boiler shut down and start-up, sudden load changes, operation without soot blower, soot blower operation with ambient air and regeneration of catalyst.

These tests established the applicability of Hitachi Zosen catalyst NOXNON 600 to coal-fired boilers in the United States.

CATALYST LIFE TEST

Operating Period

    Operating hours with NOXNON 600 were,

operation with flue gas	:	5,620 hours
operation with air	:	920 hours
shut down	:	320 hours
TOTAL :		6,860 hours

Operation with air was done with air heated by the flue gas heater to maintain the temperature in the reactor so that the time for restartup could be reduced and to avoid thermal expansion and contraction of the catalyst. Ammonia was not injected.

    Interruptions in flue gas testing were caused approximately 80 percent by maintenance of Unit #3 Boiler of Georgia Power Company, and 20 percent by troubles with the soot blower and analyzers inside the pilot plant.

    Approximately 25 percent of complete shut downs were caused by problems with the Blower, the Flue Gas Heater and the rotary valve of the Dust Separator; 40 percent was due to catalyst inspection and holidays; and 20 percent was for troubles at the Unit #3 Boiler.

Timetable

    Major milestones of the pilot plant are described below.

4/14/80 -- 4/16/80	Reconstruction of reactor and ducting.
4/17/80	Catalyst charge.
4/22/80	Flue gas introduction into the system. Due to trouble of Unit #3 Boiler, flue gas was replaced with air on the same day.

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4/24/80 -- 4/26/80	Operation with flue gas. Due to boiler trouble, again replaced with air.
5/8/80 -- 5/14/80	Operation with flue gas at 1,200 scfm.
5/14/80	Catalyst inspection. Packing added to clearance between reactor wall and catalyst.
5/16/80 -- 5/26/80	Phase IV Test with 1,500 scfm.
5/24/80 -- 5/25/80	Flow rate test.
5/27/80 -- 6/27/80	Operation at 1,500 scfm.
(6/5/80 -- 6/8/80)	Stack test by Radian Corporation.
6/27/80	Heater bundle of Flue Gas Heater replaced.
	The second catalyst inspection.
7/3/80 -- 7/4/80	Flow rate test.
7/7/80 -- 7/9/80	NH_3 mole ratio test.
7/8/80	Flue gas flow rate changed from 1,500 scfm to 1,300 scfm.
7/20/80 -- 8/8/80	Stack test by Radian Corporation.
8/4/80 -- 8/5/80	SO_2 concentration test.
8/6/80 -- 8/9/80	Mole ratio test.
8/24/80	NO_x level test.
8/27/80	Maintenance of soo· blower.
9/12/80 -- 9/13/80	Mole ratio test.
9/21/80 -- 9/26/80	Shut down due to troubles with Blower and rotary valve.
10/28/80--10/30/80	Flow rate test.
11/7/80	Temperature test.
11/8/80	Flow rate was changed from 1,300 to 1,100 scfm.
11/9/80--11/17/80	Mole ratio test.
11/18/80	Mole ratio test.
11/20/80--11/25/80	80 percent removal test.
11/24/80	Flow rate was changed to 1,300 scfm.
11/26/80--11/28/80	Thanksgiving day holidays.

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12/5/80 -- 12/6/80	Mole ratio test.
12/8/80 -- 12/9/80	80 percent removal test.
12/10/80	Emergency shut-off of ammonia feed.
12/11/80 -- 12/17/80	Power plant load excursions.
12/18/80	Mole ratio test
12/19/80 -- 12/22/80	Operation with air due to increase in pressure drop.
12/24/80 -- 12/25/80	Christmas holiday.
12/26/80	Catalyst inspection.
12/27/80	Cold start up.
1/1/81 and 1/4/81	Boiler shut down and start-up.
1/5/81 -- 1/6/81	Sudden load change.
1/7/81	Flow rate was changed to 1,100 scfm.
1/9/81	Operation without soot blower.
1/15/81	Mole ratio test.
1/26/81	Regeneration of catalyst.
1/30/81 -- 1/31/81	Mole ratio test.
1/31/81 -- 2/1/81	Flow rate test.
2/2/81	Operations ended.

Operations

The catalytic performance of NOXNON 600 was investigated for the factors of NO_x removal efficiency, mole ratio of ammonia to NO_x, operating temperature, and pressure drop across the catalyst layer as summarized in Figure 6-1.

Furthermore, the relationship between actual pressure drop and calculated pressure drop across the catalyst layer is expressed in Figure 6-2.

Operating data used in Figure 6-1 are, in most cases, the average of four measurements between 12:00 to 13:00. When some special experiments were examined during this period and the average seemed unrepresentative, the data were deleted, or otherwise, collected from another period when the operating conditions

6-5
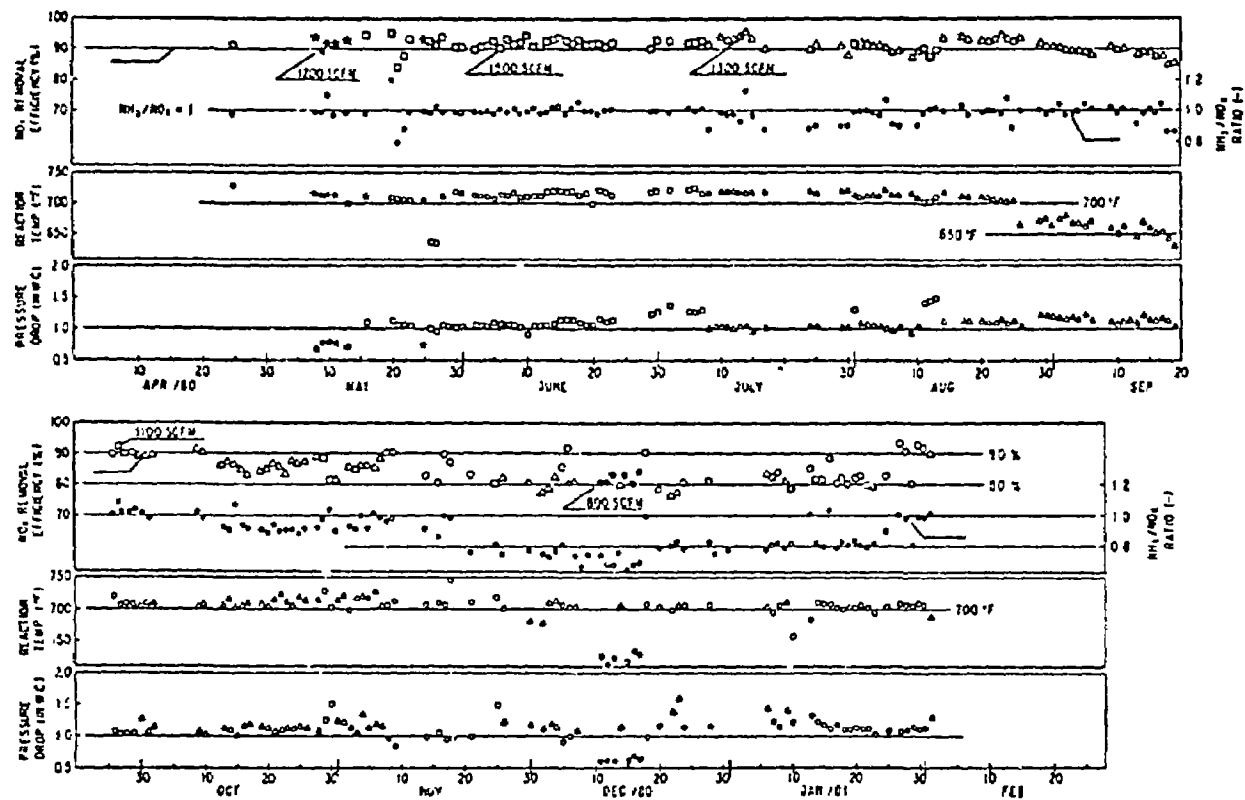
Figure 6-1. Operating Data with NOXNON 600 (5,620 hours).
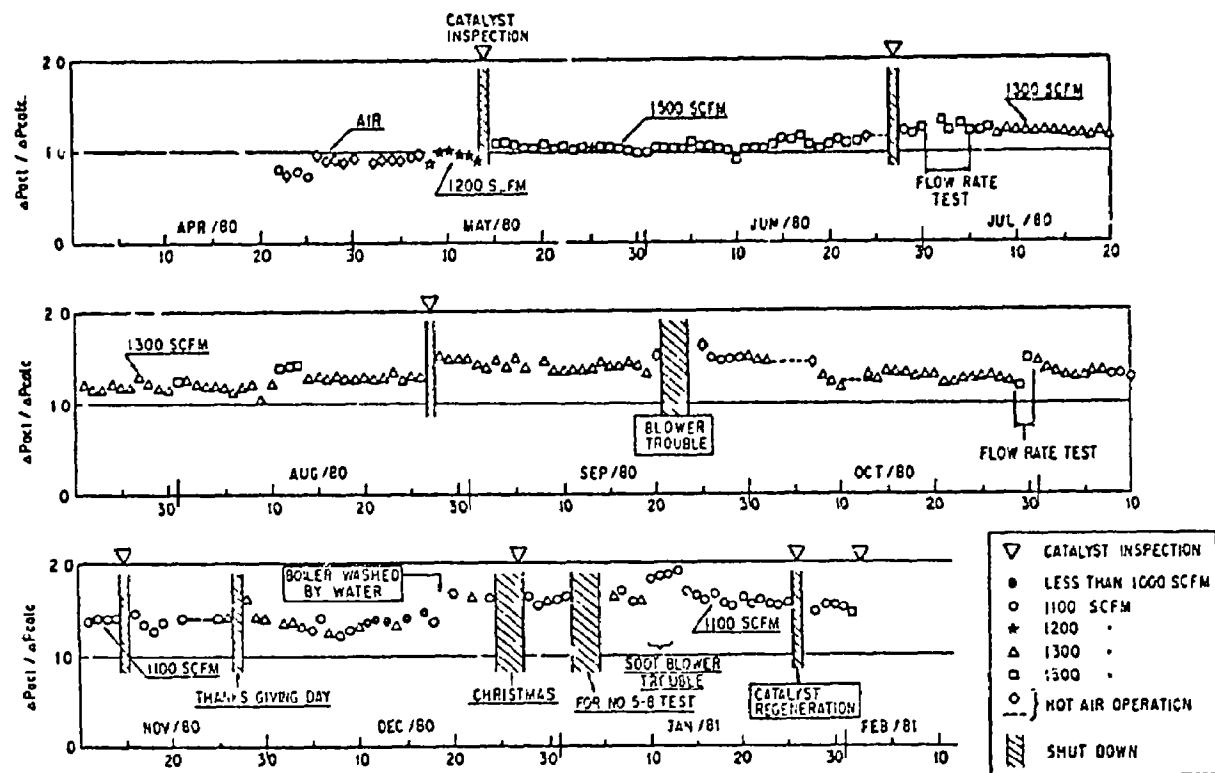
Figure 6-2. Pressure Drop across Catalyst Layer (Actual/Calculated).

were more stable.

A. Period from 0 hours to 200 hours (4/22/80 to 5/14/80).

The operation of NOXNON 600 with combustion flue gas began
on April 22, 1980. However, due to problems at the Unit #3
Boiler, the pilot plant was forced to watch and wait for flue
gas operation while maintaining the operating temperature in
the reactor with heated air. Therefore, actual start-up with
flue gas began on May 8, 1980.

During this period, standard operating conditions were,

Flow rate	:	1,200 scfm
Temperature	:	700°F
NH_3/NO_x mole ratio	:	1.0

During the first week of operation NO_x removal efficiency
was 88 percent to 93 percent which was lower than the expected
efficiency. At the same time, pressure drop was almost the same
as calculated pressure drop (0.77 inches H_2O). However, due to
the very low pressure drop, non-uniformity of flue gas distribu-
tion was suspected, and the catalyst was inspected from the man-
hole of the reactor on May 14, 1980. The results of the observa-
tion will be described later in "Catalyst Inspections On Site,"
and summarized as follows:

(1) The seal of the clearance between the reactor wall and
the catalyst appeared insufficient, and additional
asbestos yarn was supplemented.

(2) From the appearance of the adhered fly ash over the
reactor wall, some non-uniformity of flue gas distrib-
ution was assumed.

Subsequently, after this observation of the inside of the
reactor, the flue gas flow rate was increased to 1,500 scfm
although this flow rate corresponds to 0.7 MW which is 40 percent
larger than the designed capacity of 0.5 MW equivalent. This

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increased flue gas flow rate would provide better gas distribution.

B. Period from 200 hours to 1,320 hours (5/14/80 to 7/7/80).

Until 7/7/80, the pilot plant was operated at the flue gas flow rate of 1,500 scfm. NO_x removal efficiency was between 90 percent to 94 percent and seemed stable. During this period it was found that the NO_x removal efficiency appeared to increase slightly when the flow rate was increased. This tendency was noticed especially in the performance test run in the middle of May.

This tendency was possibly caused by:

-   Increase in the flue gas flow rate decreasing the deposits of fly ash on the catalyst surface.
-   Higher superficial linear velocity contributing to the uniformity of flue gas flow distribution due to higher pressure drop.

Meanwhile, the pressure drop started at 1.0 to 1.1 inches H_2O at the beginning of the operation, and increased very gradually up to 1.3 inches H_2O. From this experience, the pressure drop seemed to be more stable at the flow rate of 1,300 scfm, and catalyst inspection on June 27, 1980 showed that abrasion of catalyst seemed to be in progress although very slow. Flue gas flow rate was changed to 1,300 scfm, equivalent to 0.6 MW.

C. Period from 1,320 hours to 4,000 hours (7/8/80 to 11/9/80).

During this period, catalytic performance seemed stable. At the end of August when the operating time was 2,500 hours, performances were:

    flow rate   : 1,300 scfm
    operating temperature : 700° to 720°F
    NH_3/NO_x mole ratio   : 1.0

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NO_x removal efficiency	:	90 to 94 percent
Pressure drop	:	1.10 to 1.30 inches H_2O

At the end of September, for the purpose of confirming catalytic performance at more than 90 percent NO_x removal at the design condition, a performance test was run at the flow rate of 1,100 scfm. More than 90 percent NO_x removal was proven through the performance test. However, from the end of August, from time to time, pressure drop across the catalyst layer fluctuated as did the NO_x removal efficiency. This tendency was especially observed when the Unit #3 Boiler operated the soot blower at its economizer repeatedly and frequently. When the pressure drop increased and the NO_x removal decreased due to the frequent soot blower operation at the economizer, the unstable pressure drop and NO_x removal efficiency were restored after a few days by operating at a lower gas flow rate: 1,100 scfm.

Between October 13, 1980 and November 8, 1980 (3,340 hours to 3,960 hours), mole ratio of ammonia to NO_x was decreased to approximately 0.9 to obtain less ammonia slip.

Flue gas flow rate	:	1,300 scfm
Operating temperature	:	700° to 720°F
NH_3/NO_x mole ratio	:	0.88 to 0.94
NO_x removal efficiency	:	83 to 88 percent

On November 8 and 9, 1980, catalytic performance of more than 90 percent was again confirmed at the designed capacity, 1,100 scfm. At this time, operating time had been 4,000 hours. NO_x removal efficiency at a flow rate of 1,300 scfm and mole ratio of 1.0 was approximately 89 percent.

D. Period from 4,000 hours to 4,230 hours (11/10/80 to 11/21/80).

At the design condition, a mole ratio test and a temperature test were run to check the catalyst activity prior to entering into transient tests. Afterwards, an 80 percent test was run.

The purpose of the 80 percent test was to reduce ammonia slip as far as possible. The target of NO_x removal was decreased to 80 percent from 90 percent, and NH_3/NO_x mole ratio was set so as to maintain the NO_x removal at slightly higher than 80 percent. The operating conditions were:

flue gas flow rate	:	1,100 scfm
temperature	:	$700^{\circ}F$
NH_3/NO_x mole ratio	:	approximately 0.85

E. Period from 4,230 hours to 4,420 hours (11/24/80 to 12/4/80).

During this period the operation was continued at the following conditions:

flue gas flow rate	:	1,300 scfm
temperature	:	$700^{\circ}F$
NH_3/NO_x mole ratio	:	approximately 0.8
NO_x removal	:	slightly greater than 80 percent

The purpose of this test was to investigate operations with less ammonia slip and 80 percent NO_x removal at the flow rate of 1,300 scfm.

After Thanksgiving Day, temperature and pressure drop fluctuated widely resulting in changes in NO_x removal. These fluctuations were not caused by any problems in Unit #3 Boiler.

F. Period from 4,420 hours to 5,030 hours (12/5/80 to 1/7/81).

To evaluate the catalyst performance while transient tests were being executed, a performance test was run on 12/5/80 and 12/6/80. The result was:

flue gas flow rate	:	1,100 scfm
temperature	:	$700^{\circ}F$
NH_3/NO_x mole ratio	:	0.99 to 1.02
NO_x removal	:	90 to 91.5 percent

On 12/7/80, transient tests commenced. The results of the

6-11

transient tests are described later in "Transient Tests." While the transient tests were being run, operating conditions were sharply varied, and NO_x removal efficiency was difficult to estimate, however, at the conditions of:

flue gas flow rate : 1,100 scfm

temperature : approximately 700°F

NH_3/NO_x mole ratio : approximately 1.0

NO_x removal efficiencies were:

12/5/80 and 12/6/80 : 90 to 91.5 percent

12/18/80 : 89 to 91 percent

1/5/81 : 91 percent

From 12/11/80, while "Power Plant Load Excursions" test was run, the pressure drop gradually increased, and on 12/18/80, when Georgia Power Company had trouble with leakage in water tubes of Unit #3 Boiler, the pressure drop increased rapidly and seriously. In order to reduce the pressure drop, the pilot plant was operated at 1,100 scfm and 1,300 scfm, and air operation was also tried. However, higher and unstable pressure drop continued until 1/7/81.

G.   Period from 5,030 hours to 5,470 hours (1/7/81 to 1/26/81).

Transient tests continued during this period, and the pilot plant was shut down on 1/26/81 for regeneration of catalyst.

H.   Period from 5,470 hours to 5,620 hours (1/27/81 to 2/2/81).

After the regeneration of catalyst, the catalyst performance was examined. NO_x removal efficiency after the regeneration of catalyst was the same as the initial efficiency, and the pressure drop decreased and seemed stable.

6-12

CATALYST PERFORMANCE TESTS

While catalyst life test and transient tests were being run, catalyst performance was evaluated by varying operating conditions.

The purpose of the catalyst performance tests were (1) to evaluate the catalyst performance from time to time over the elapsed operating time, and (2) to investigate influences of operating variables on the catalyst performance. The performance tests were examined approximately once every 1,000 hours of operation.

Flue Gas Flow Rate

In the middle of May, the first catalytic performance test was performed. As shown in Figure 6-3, when the flue gas flow rate increased, the NO_x removal efficiency increased slightly. This rather contrary tendency seems peculiar to high fly-ash-loaded combustion flue gas from a coal-fired boiler. When the flow rate is somewhat low, fly ash clinging to the catalyst surface results in partial masking of the catalyst surface and, to some extent, to non-uniformity of flue gas flow through the catalyst channels. When the flow rate is increased, deposited fly ash is blown off the catalyst increasing the apparent NO_x removal efficiency. (At the same time, the fact that should be noted is pressure drop across the catalyst layer of NOXNON 600 was lower than that of NOXNON 500 due to wider channels between catalyst layers). From these results, the flue gas flow rate in the catalyst life test and the transient tests were increased to 1,300 scfm and 1,500 scfm although designed flow rate was 1,057 scfm.

After approximately 1,000 hours of operating time elapsed, the tendency to increased NO_x removal with an increase in the flue gas flow rate disappeared, and NO_x removal decreased when

6-13
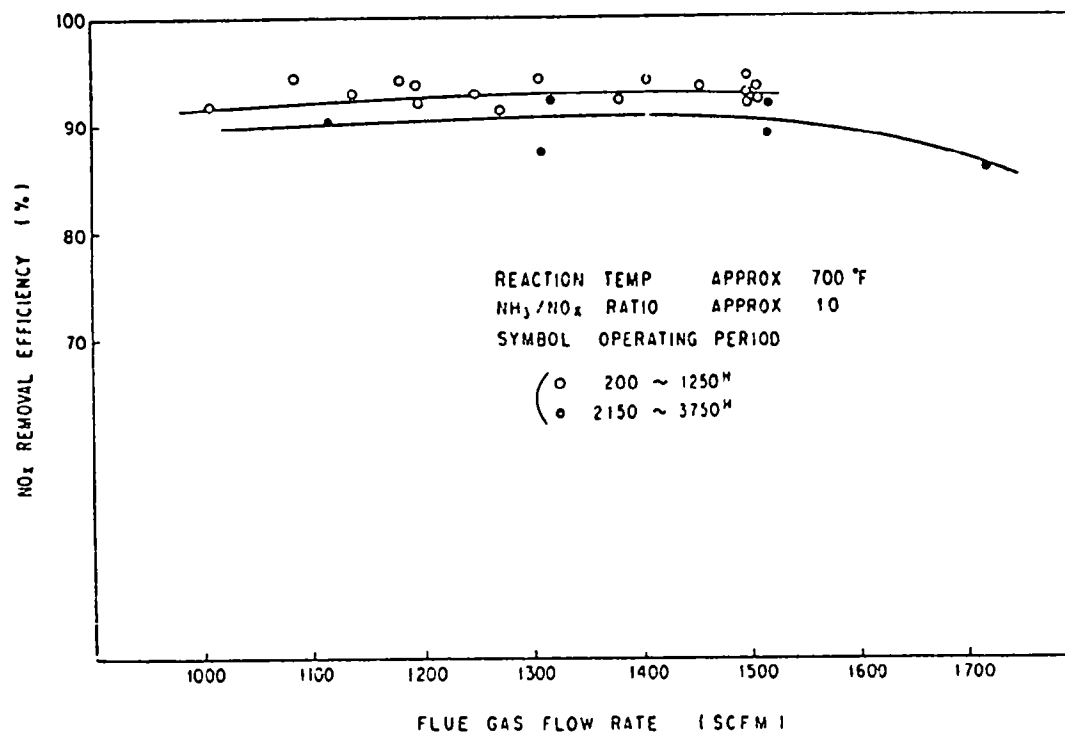
Figure 6-3. Effect of Flue Gas Flow Rate on NO_x Removal
Efficiency.

the flow rate of flue gas increased. This is as normally seen
in pilot plants applied to oil-fired combustion flue gas. It
was assumed that with the elapsed operating time fly ash deposit-
ed on the catalyst surface, and this thin fly ash layer stabil-
ized in equilibrium with the flue gas flow, especially at the
corners of the triangular channels, and resulted in a decrease
of NO_x removal efficiency.

Mole Ratio

During the test operation the pilot plant was operated at a
selected mole ratio. The control system was designed to auto-
matically provide this mole ratio. This was accomplished by
using the flue gas flow rate signal and the inlet NO_x concentra-
tion analysis to determine the quantity of NO_x in the inlet
stream. From the inlet NO_x 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.

The actual mole ratio tended to vary somewhat because of
fluctuations in NO_x levels, flue gas flow rates, flue gas
pressures, etc. Efforts were constantly made to be certain of
the accuracy of these measurements (see Appendix) but it is
believed that there may have been a consistent error.

The fraction NO_x removal is often higher than the mole ratio
especially at lower levels. Fundamental experiments, however,
have proven that this is not possible. In the presence of oxygen
the NO_x removal cannot be greater than the equivalent amount of
ammonia available. The reaction between NO and NH_3 is an equi-
molar reaction.

Although all of the instruments were checked and rechecked
and calibrated several times, there is a suspicion that the
ammonia flow meter may have been in error, particularly at low
flows. It may have been reading too low. The ammonia flow rate

6-15

·was very low (about 0.5 scfm) and the very narrow clearance
between the rotor and the tapered tube could readily catch small
particles such as dust, rust, and even moisture causing inaccu-
rate readings. This may have happened despite the installation
of very fine micron filters before the flowmeter.

Because of the questions regarding the ammonia flowmeter
the data in the later tests were recalculated to obtain mole
ratio curves (Figure 6-4, 6-5 and 6-6). The ammonia feed was
calculated by adding together the ammonia slippage and the
ammonia theoretically required to react with the NO_x removed.
The mole ratio was then determined by dividing the calculated
moles of ammonia by the moles of inlet NO_x. (The NO_x was assumed
to be all NO).

Figure 6-4 shows the results of the test at an 80% removal.
Figures 6-5 and 6-6 are mole ratio curves taken over different
operating periods. The mole ratio was calculated by the methods
described above. From these results it can be seen that flue
gas flow rates had no measurable effect on NO_x removal. A NO_x
removal of 80% required a mole ratio of about 0.85 while a 90%
removal requires a mole ratio of 1.0.

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. The method was refined to the point where by
August 15, 1980 the data was considered reliable. The accuracy
of the method was checked by Radian Corp. and found to be sat-
isfactory. Data relative to ammonia slippage was collected after
that time.

Figures 6-4, 6-5 and 6-6 show ammonia slip under various
conditions. Figure 6-4 shows ammonia slip measured during the
80 percent removal test. Figures 6-5 and 6-6 show ammonia slip
when mole ratio was varied at operating times of 3200 hours to
4500 hours and 4600 hours to 5600 hours respectively. General

CONDITION :
REACTION TEMP . 705 ~ 718 °F
INLET NOx : 367 ~ 431 PPM
NO REMOVAL EFF : 80.0 ~ 80.7 %
OPERATING PERIOD . 4200 ~ 4

SYMBOL { △ 80 % NOx REMOVAL
         □ NH₃ SLIPPAGE

NH₃ / NOx MOLE RATIO (-)

NH₃ SLIPPAGE (PPM)

FLUE GAS FLOW RATE (SCFM)
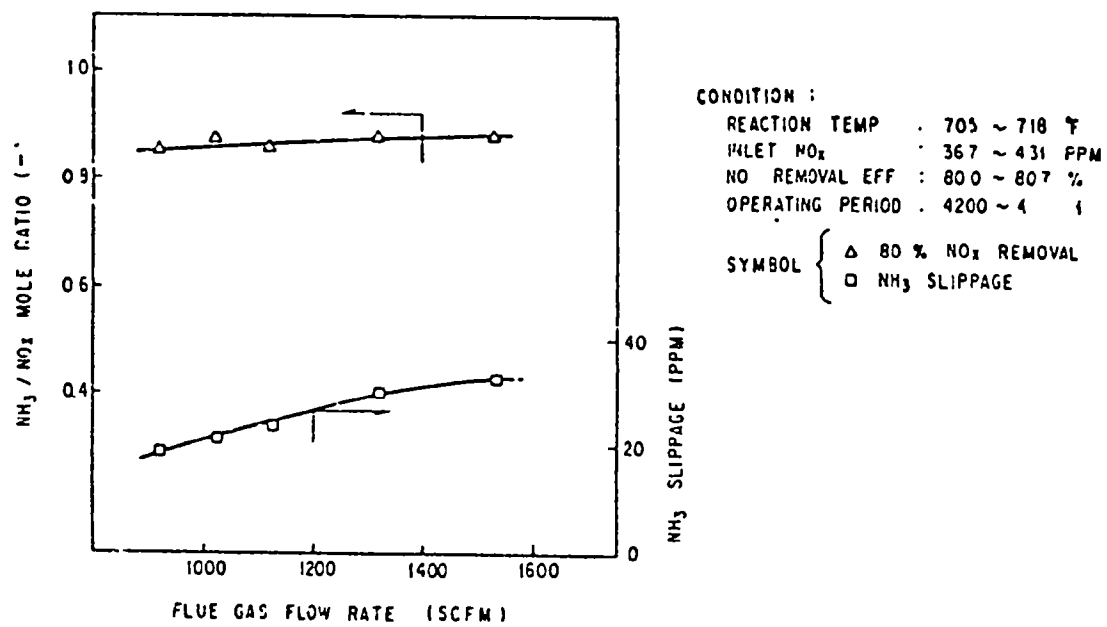
Figure 6-4 80 percent Removal Test.
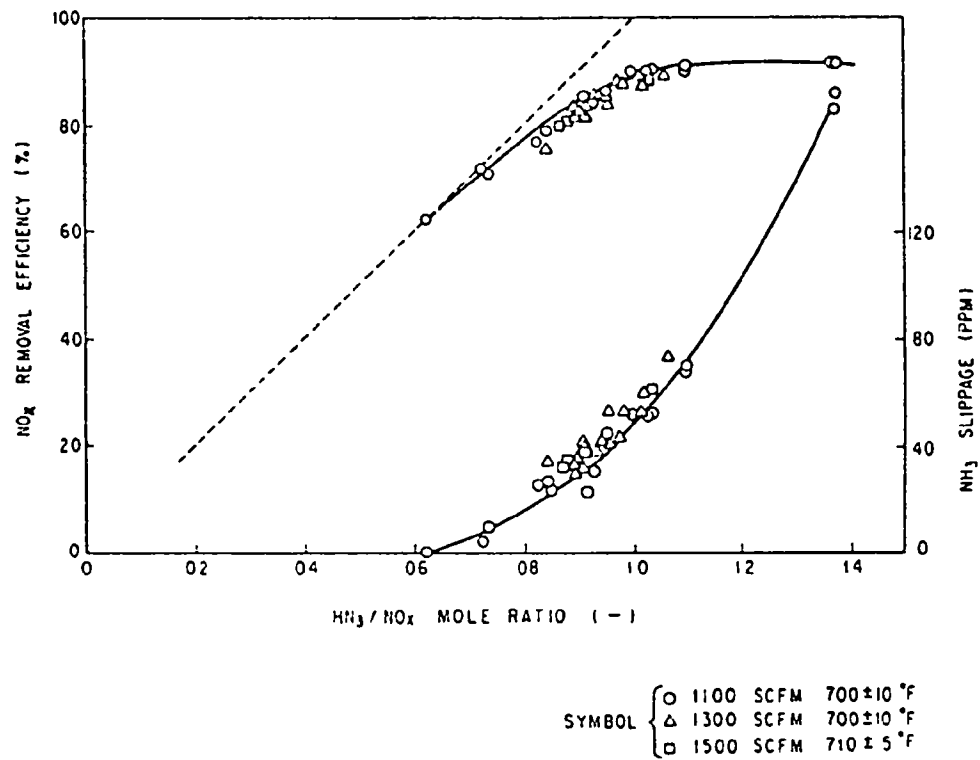
Figure 6-5. Effect of NH_3/NO_x Ratio on NO_x Removal
Efficiency and NH_3 Slippage. (3200-4500 hrs)
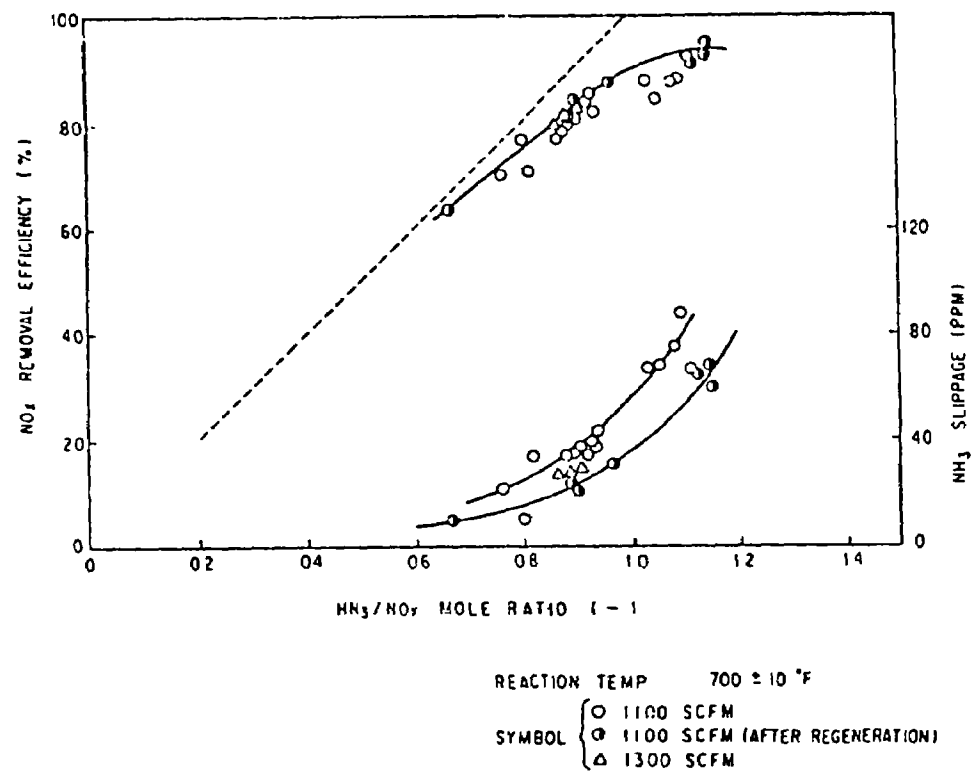
Figure 6-6. Effect of NH_3/NO_x Ratio on NO_x Removal
Efficiency and NH_3 Slippage. (4600-5600 hrs)

conclusions from these tests:

- 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, for example at a mole ratio of 0.6.

The reason for this higher than expected slippage may have been due to clogging by fibers of asbestos yarn used to seal a clearance between the catalyst box and reactor (see "Catalyst Inspections"). These fibers along with fly ash between the first and second catalyst layers may have reduced the effective catalyst surface and adversely affected the apparent catalyst activity resulting in relatively high ammonia slip.

Operating Temperature

The influence of operating temperature on NO_x removal efficiency is shown in Figure 6-7, "Reaction Temperature vs. NO_x Removal Efficiency." In the pilot plant, the operating temperature was varied between 640°F (338°C) and 780°F (415°C), and NO_x removal efficiency was not affected in this temperature range. Optimum operating temperatures obtained from fundamental experiments in the laboratory are between 572°F (300°C) and 750°F (400°C).

The results obtained in the EPA pilot plant are fairly consistent with the fundamental experimental data. At the beginning of operations, NO_x removal efficiency did not vary between 640°F and 780°F, and after approximately 4,100 hours of operation, NO_x removal at a temperature higher than 730°F seemed slightly lower. However, the difference was very small. It would seem, therefore, that between 640°F and 780°F, the effect of temperature on removal efficiency is almost negligible.
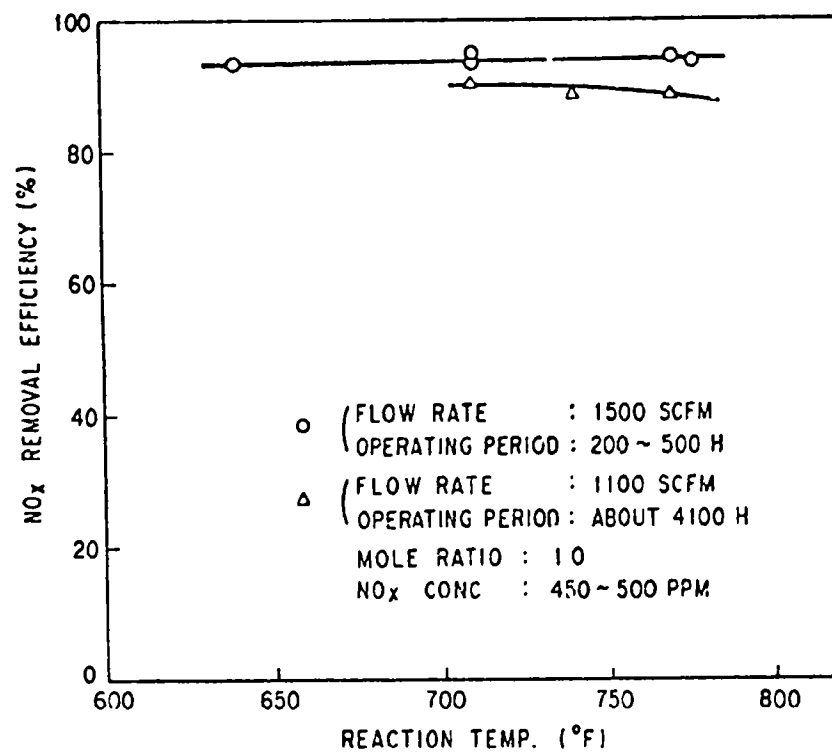
Figure 6-7   Effect of Reaction Temperature on NO$_x$ Removal Efficiency.

NO$_x$ Concentration

On August 24, 1980, the NO$_x$ Generator was operated to in-
crease the concentration of NO$_x$ at the inlet of the reactor, and
the influence of NO$_x$ concentration was investigated. Operating
conditions were:

flue gas flow rate	:	1,320 scfm (average)
operating temperature	:	700°F
NH$_3$/NO$_x$ mole ratio	:	1.0
NO$_x$ concentration	:	773 - 906 ppm

NO$_x$ removal efficiency during this test was compared with
that obtained on August 22, 1980 after approximately 2,400 hours
of operation. Results are shown in Figure 6-8.

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

Influence of SO$_x$ Concentration on NO$_x$ Removal Efficiency

It has been well established through fundamental experiments
and other pilot plant's data in Japan that catalyst activities are
not affected by SO$_x$ concentration. In operation at the EPA pilot
plant, even though the load of Unit #3 Boiler had been relatively
stable at approximately 135 MW, concentrations of SO$_2$ measured in
the pilot plant deviated widely between approximately 500 ppm and
1,500 ppm depending on the variations of sulfur in the coal.
Therefore, various data under the same operating conditions ex-
cept for SO$_2$ concentrations were selected and summarized in Fig-
ure 6-9. This showed that through the whole operating period,
NO$_x$ removal efficiency was not affected by SO$_2$ concentrations in
the flue gas between the relatively wide range of 500 ppm and
1,500 ppm.
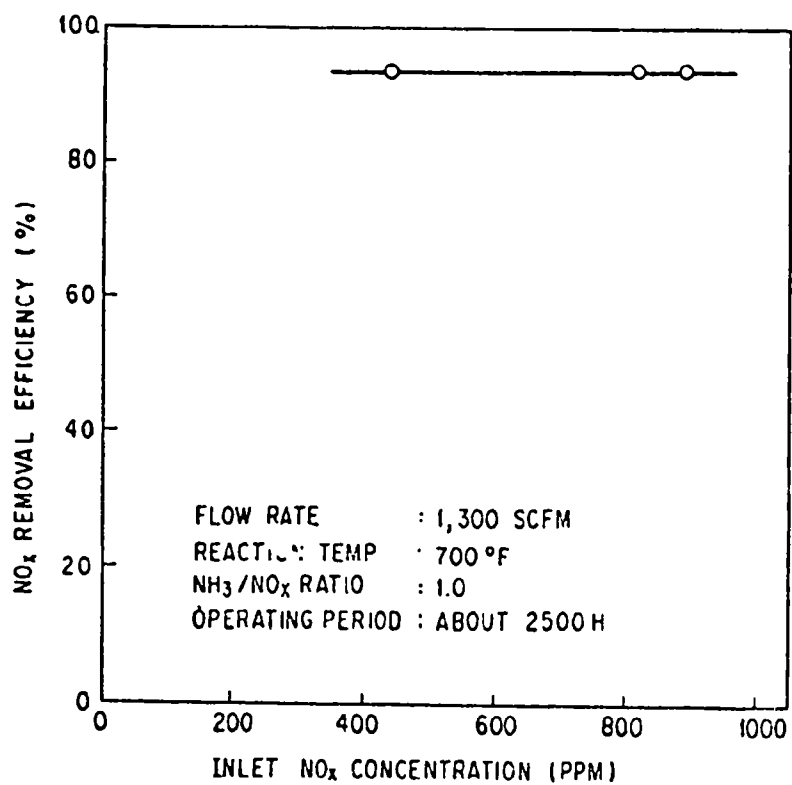
Figure 6-8   Effect of NO_x Concentration on
NO_x Removal Efficiency.

6-23
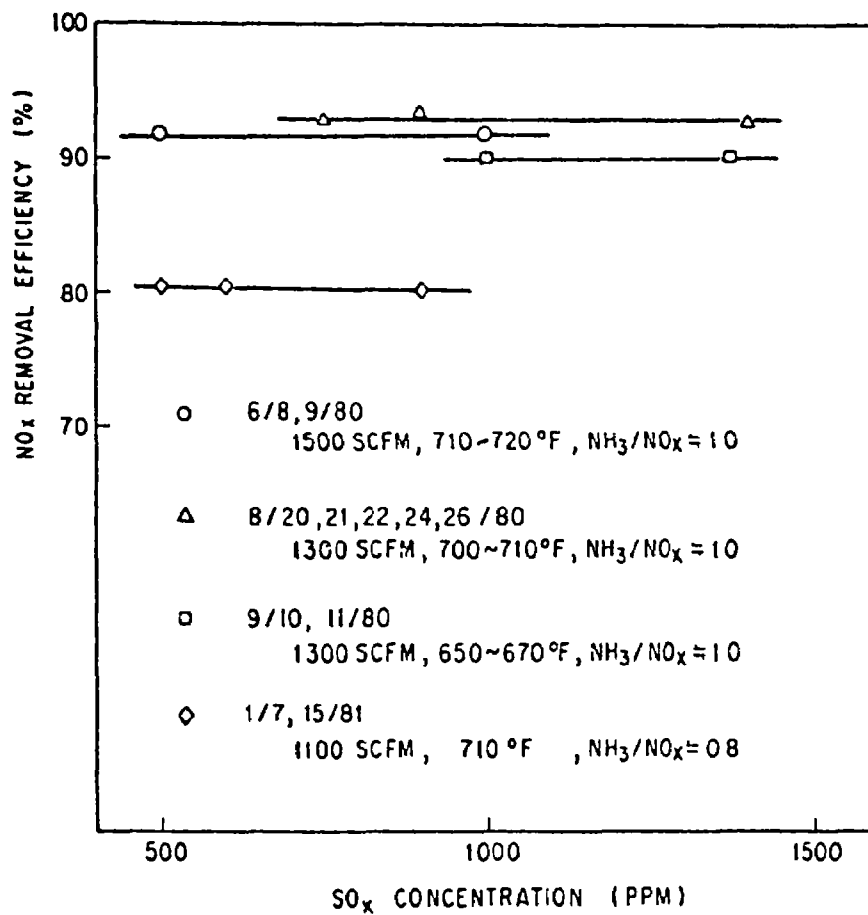
Figure 6-9   Effect of SO$_x$ Concentration on
NO$_x$ Removal Efficiency.

Oxidation of SO$_2$ to SO$_3$

On July 23, 24 and 25, 1980 operating conditions were main-
tained at a constant level so that SO$_3$ measurements could be
obtained at the inlet and outlet of the reactor. The following
were the conditions of operation. The SO$_2$ and SO$_3$ measurements
were made by Radian.

Date	7/23/80	7/24/80	7/25/80
Time	1630	1650	1625
Flue Gas Flow Rate (scfm)	1320	1320	1320
Temperature °F (°C)	715 (380)	715 (380)	716 (380)
Inlet SO$_2$ - ppm	820	579	679
Inlet NO - ppm	473	514	468
Oxygen - %	5.7	5.8	5.5
NH$_3$/NO$_x$ mole ratio	0.915	0.931	0.91
NO$_x$ removed	91.2	92.4	91.9
Inlet SO$_3$ - ppm	11.0	7.5	6.6
Outlet SO$_3$ - ppm	25.3	14.8	21.9
SO$_2$ to SO$_3$ conversion - %	1.7	1.3	2.3

These results indicate an average oxidation rate of 1.8%.
It was expected that at these conditions the ratio would be
lower: about 1.0 to 1.5%. Those expectations were based on
experimental laboratory data and on pilot plant data from Japan.

Experimental data results are shown in Figure 6-10. At the
catalytic composition as used for the EPA pilot plant, the per-
cent oxidation would be expected to be 0.5%. However, this ex-
perimental data was performed at a somewhat lower temperature
(350°C vs. 380°C) and a higher Area Velocity (12.1 vs. 9.6).

Data from a coal-fired pilot plant in Japan indicated an
oxidation rate of between 0.89 and 1.11% at the same temperature
and area velocity as used for the EPA pilot plant tests.

Of course, measurements of SO$_3$ are quite difficult and
require a high degree of technical skill and experience. Even

6-25
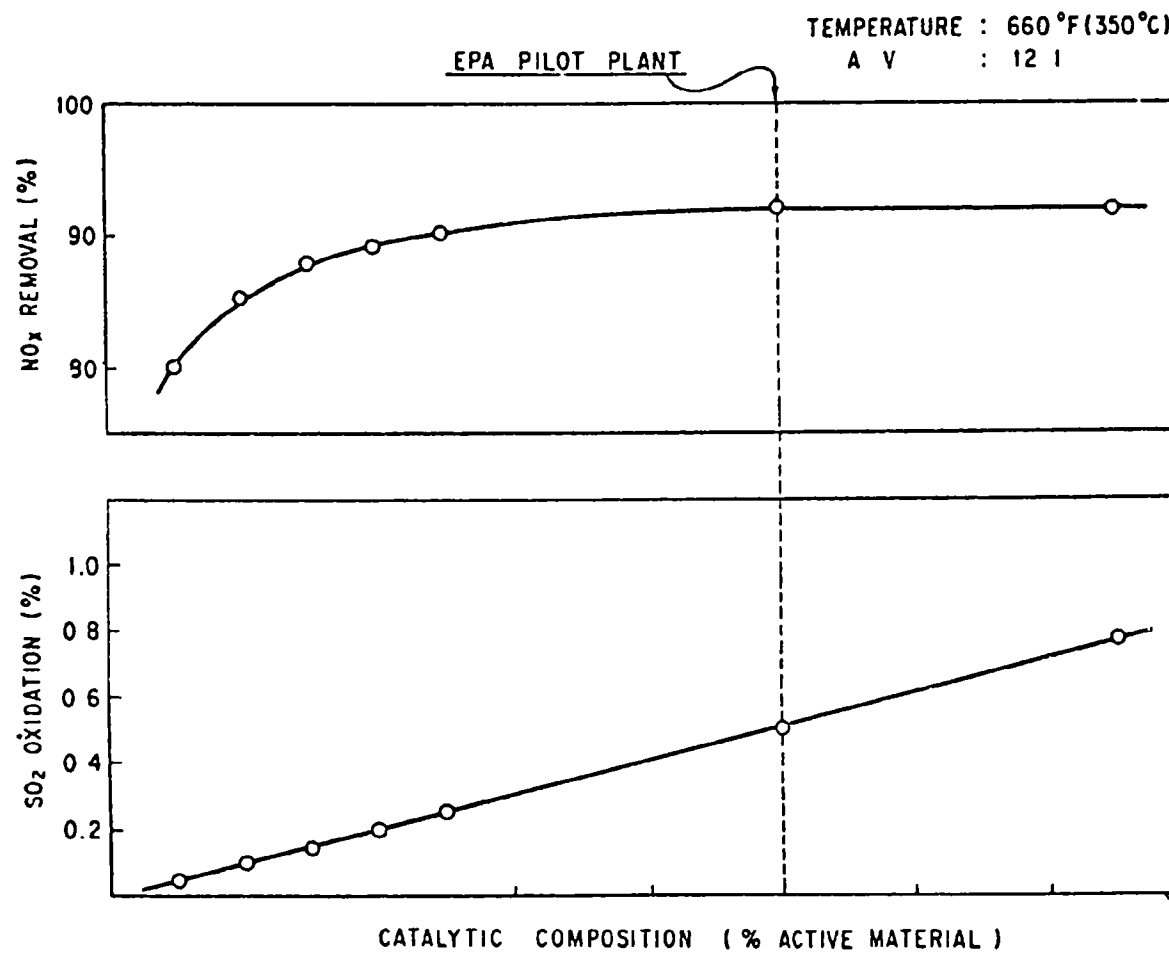
Figure 6-10 Typical SO$_2$ to SO$_3$ Conversion.

with that, the presence of fly ash always influences the analytical results. Although there were no questions about the techniques followed in obtaining this data, the expected scattering of data would require many points to provide a reasonable statistical confidence in the results. Unfortunately, only three points were obtained due to the difficulties and the limitations of funds. Nevertheless, the data was relatively consistent and the oxidation rate was close to what would have been expected.

The results of these tests showed an oxidation rate which was relatively low. However, these rates can be reduced even further by varying the catalyst composition. The need for very low oxidation would depend on the particular application of the process.

TRANSIENT TESTS

After the NO_x removal efficiency of more than 90 percent was demonstrated in the continuous run of three 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. For one thing, the continuous long term run would be extended to reach at least 5,000 hours of operation, if possible. Also, a series of tests were planned to determine the effects on the catalyst and the system of transient conditions which could be anticipated in commercial operations.

As Georgia Power Company planned modifications of its Unit #3 Boiler starting from February, the testing could not be extended beyond that period since the pilot plant would have to be removed to enable the modifications to be made.

The following transient tests were performed:

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Power Plant Load Excursions

A commercial power plant varies boiler load as required by power consumption. The change in boiler load is relatively rapid which can cause sharp variations in flue gas flow rate and temperature. The purpose of this test is to confirm the controllability of the system and the reliability of the catalyst for the transient conditions of load excursions. Two ranges of conditions were tested. One was at "full load" which is at high flue gas flow rate and high temperature, and the other was at "low load" with low flow rate and low temperature.

Operating conditions during daytime (9:00 am to 5:00 pm):

      flue gas flow rate: 800 scfm (approx. 1,300 Nm^3/h)

      temperature   : 610°F (320°C)

      NO_x removal   : 80 percent

Operating conditions during nighttime (5:00 pm to 9:00 am):

      flue gas flow rate: 1,300 scfm (approx. 2,000 Nm^3/h)

      temperature   : 700°F (370°C)

      NO_x removal   : 80 percent

The above conditions were repeated 6 times from December 11, 1980 to December 18, 1980. Transition from one condition to another was done rapidly.

Before and after the tests, catalytic performances were confirmed at the following conditions:

      flue gas flow rate: 1,100 scfm (1,700 Nm^3/h)

      temperature   : 700°F (370°C)

      NH_3/NO_x mole ratio: 0.8 and 1.0

Figure 6-11 shows the results of this test. During these repeated tests, NO_x removal efficiency at a given mole ratio showed no change. Pressure drop at the low load did not change, however, pressure drop at the high load increased gradually from 1.03 inches H_2O to 1.16 inches H_2O.

Usually pressure drop increased due to soot blowing applied
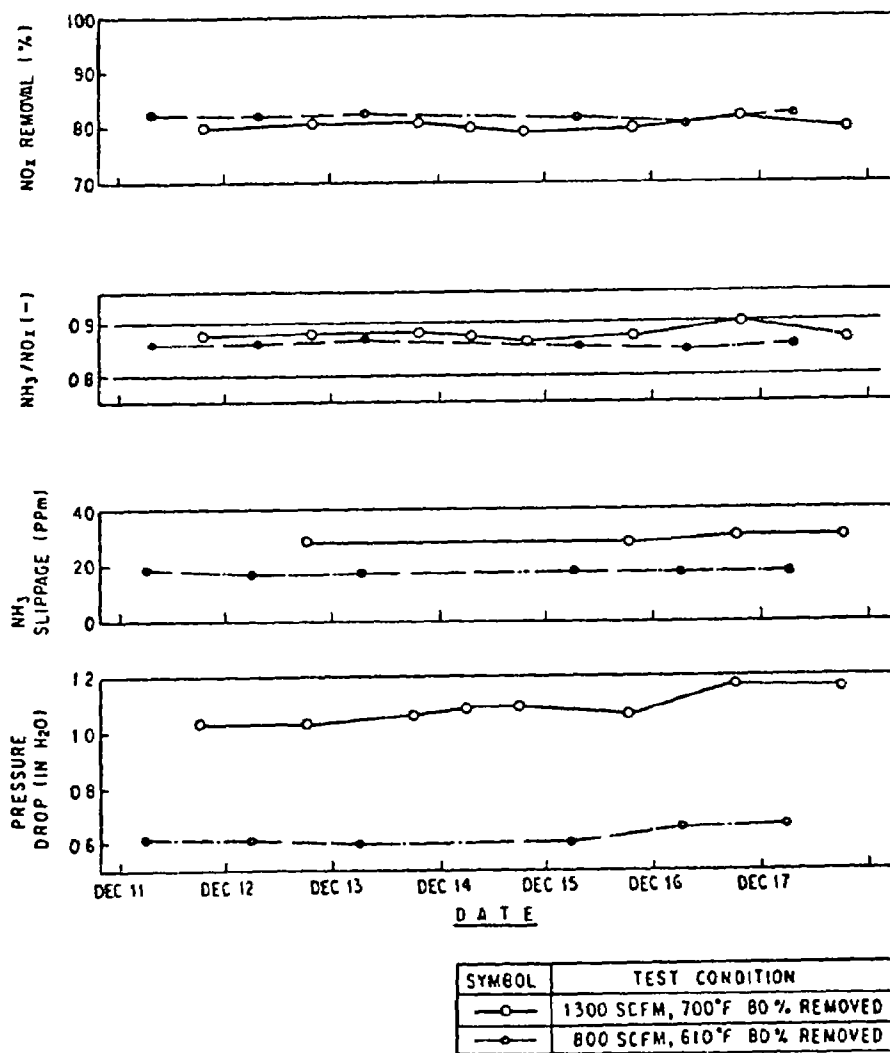
Figure 6-11. Test of System Performance under Plant Load Excursions.

to the economizer in Unit #3 Boiler, therefore, this increased pressure drop did not seem to be related to the variation in operating conditions. Catalytic performances before and after this test were the same: 83.6 percent at mole ratio 0.8 and 90.3 percent at mole ratio 1.0. These results proved that the system and catalyst can withstand sudden load changes in the boiler.

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 (315°C), and allow it to introduce ammonia into the system when temperature in the reactor increased and returns to 600°F. The purpose of this test was to confirm the reliability of the trip system.

On December 10, 1980, the following tests were performed. While the pilot plant was being operated at the conditions of 1,300 scfm, 700°F, and 0.8 mole ratio, the temperature was allowed to decrease from 700°F to 550°F. During this change, the automatic shut-off of ammonia feed at a temperature of 600°F was confirmed.

Afterwards, the temperature in the reactor was allowed to increase from 550°F to 700°F and it was confirmed that ammonia feed was started and introduced automatically into the system when the temperature of 600°F was reached. This experimental work was repeated three times in order to prove the repeatability of this system. The experimental data is shown in Figure 6-12.

The results of this proved that:

- Automatic shut-off and supply of ammonia operated smoothly.

- NO_x removal efficiency and pressure drop at a temperature of 700°F was constant and unchanged through the three repeated tests.
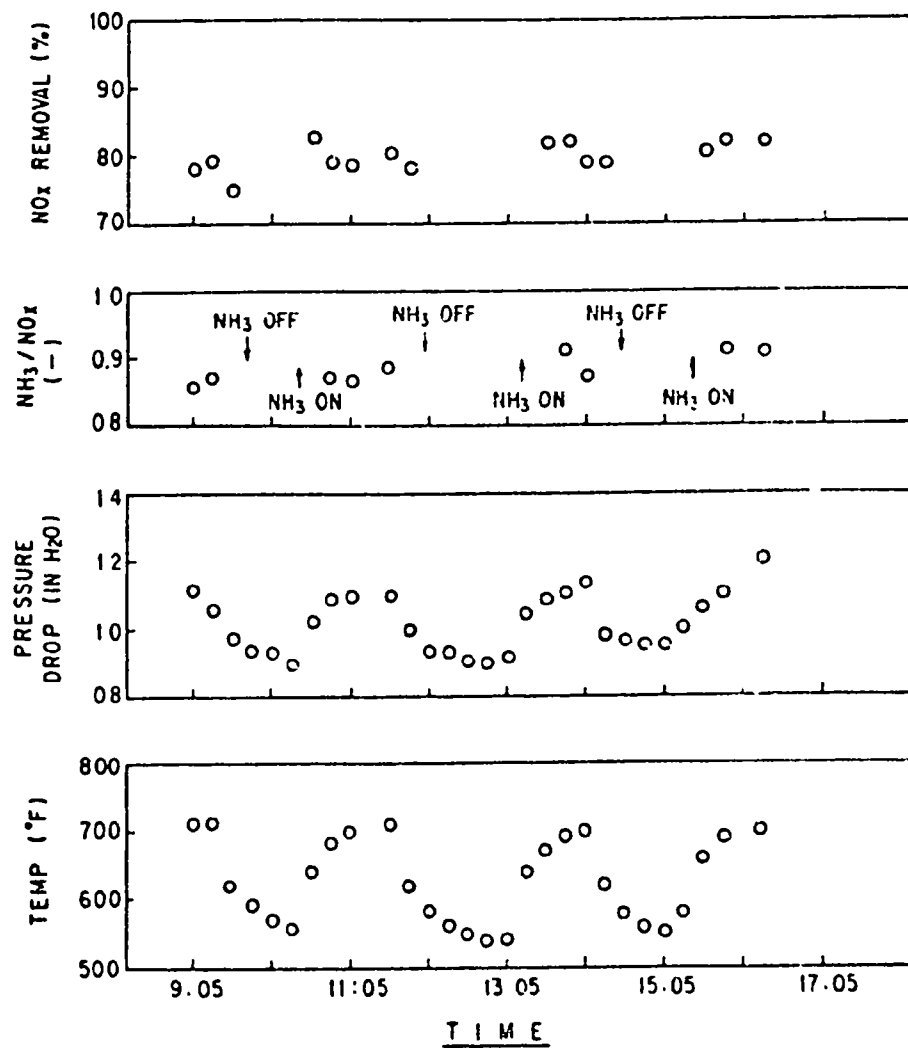
Figure 6-12.   Test of System Performance with
Emergency Shut-off of Ammonia Feed.

6-31

Cold Startup

The EPA pilot plant was provided with an air intake valve and air circulation piping line for startup and shut down in order to prevent the formation and deposit of sulfuric acid mist, ammonium sulfate, and ammonium bisulfate. During the pilot plant operation, the catalyst was routinely heated up with hot air prior to the introduction of flue gas. However, a commercial boiler normally starts into operation after a long shut down with the reactor and ductwork filled with ambient air. When operation commences flue gas would be introduced into the system as the temperature rises and the flue gas could 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 experiences 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 cause any trouble to the NO_x removal reaction.

Before the pilot plant was shut down for Christmas holidays on December 24, 1980, operating conditions were 1,100 scfm, 700°F, 1.1 to 1.2 inches H_2O, and 80 percent NO_x removal with 0.8 mole ratio. When the plant was shut down, the whole system was purged with air. 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.

Operating conditions before and after the cold start-up are shown in Figure 6-13. After the start-up, relatively higher pressure drop and lower NO_x removal efficiency in comparison with previous operating conditions continued for approximately a half day, and these were restored as time elapsed. These phenomena are normally observed when the pilot plant is shut down and started up. After a half-day after commencing the start-up, operating conditions became stable at the same conditions before
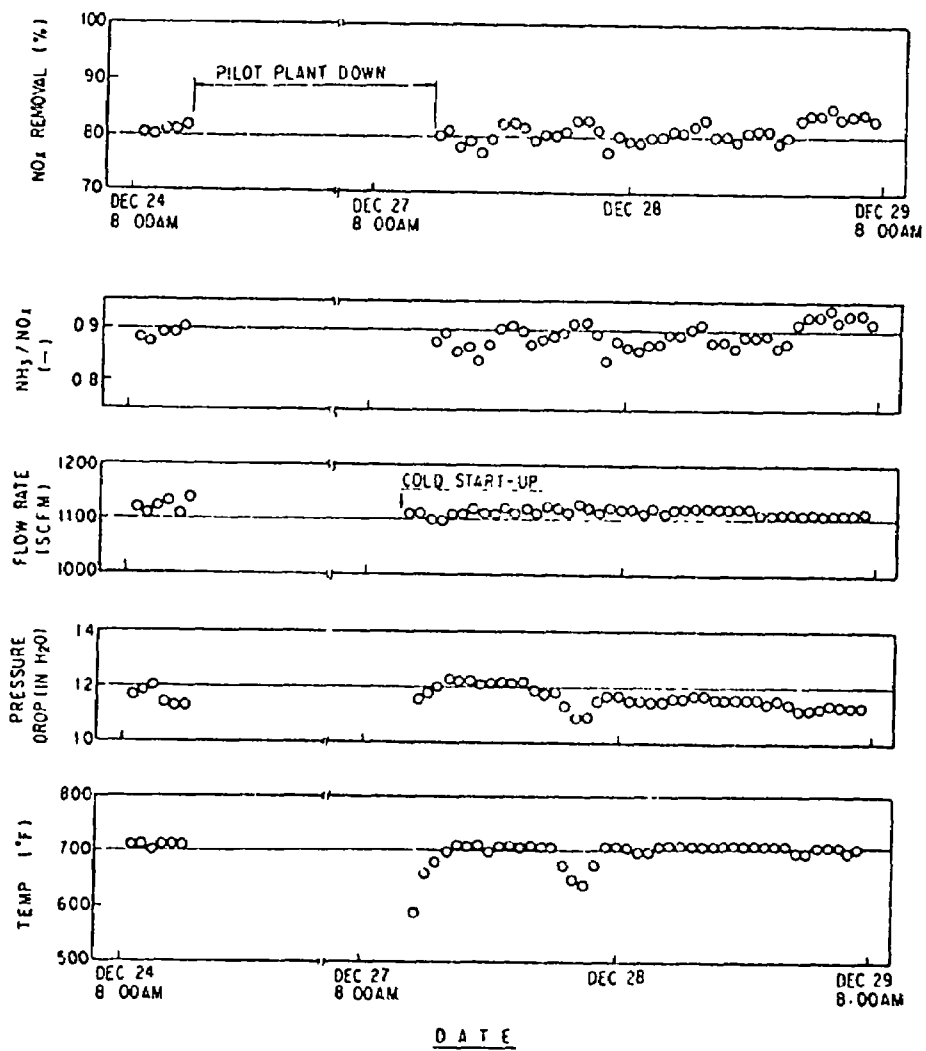
Figure 6-13.   Test of System Performance with
Cold Start-up.

cold start-up. The results proved that cold start-up does not influence catalytic performances.

Boiler Shut Down and Start-up

From time to time a power plant boiler shuts down and starts up and an NO_x removal system must follow such transition periods. The purpose of this test was to confirm the controllability of the NO_x removal system during the shut-down and start-up of the boiler, and the ability of the catalyst to withstand these transient operating conditions.

When the host boiler was shut-down for maintenance from January 1 to January 4, 1981, this test was executed. As shown in Figure 6-14, when the fan of the host boiler was switched off, the blower of the NO_x removal system was switched off at the same time on January 1, 1981. When the fan was switched on to purge the furnace and ductwork, the blower was again switched on, and then switched off following the operation of the fan. Before the start-up on January 4, 1981, the whole system was left as it is.

On 1/4/81 the fan of the host boiler was started up and the blower of the NO_x removal system was also started at the same time. The Flue Gas Heater in the pilot plant was not switched on, and cool flue gas was introduced into the system as temperature of flue gas in the boiler furnace rose. Thus, the pilot plant was filled with the same flue gas as the boiler during the transient period of boiler shut-down and start-up. During this test, ammonia was automatically cut off and fed at 600°F.

After the operating conditions became stable, catalytic performance was examined through a mole ratio test.

The result was:

flue gas flow rate	:	1,100 scfm
temperature	:	700°F
NO_x removal efficiency	:	at 0.8 of mole ratio = 83%
		at 1.0 of mole ratio = 91%
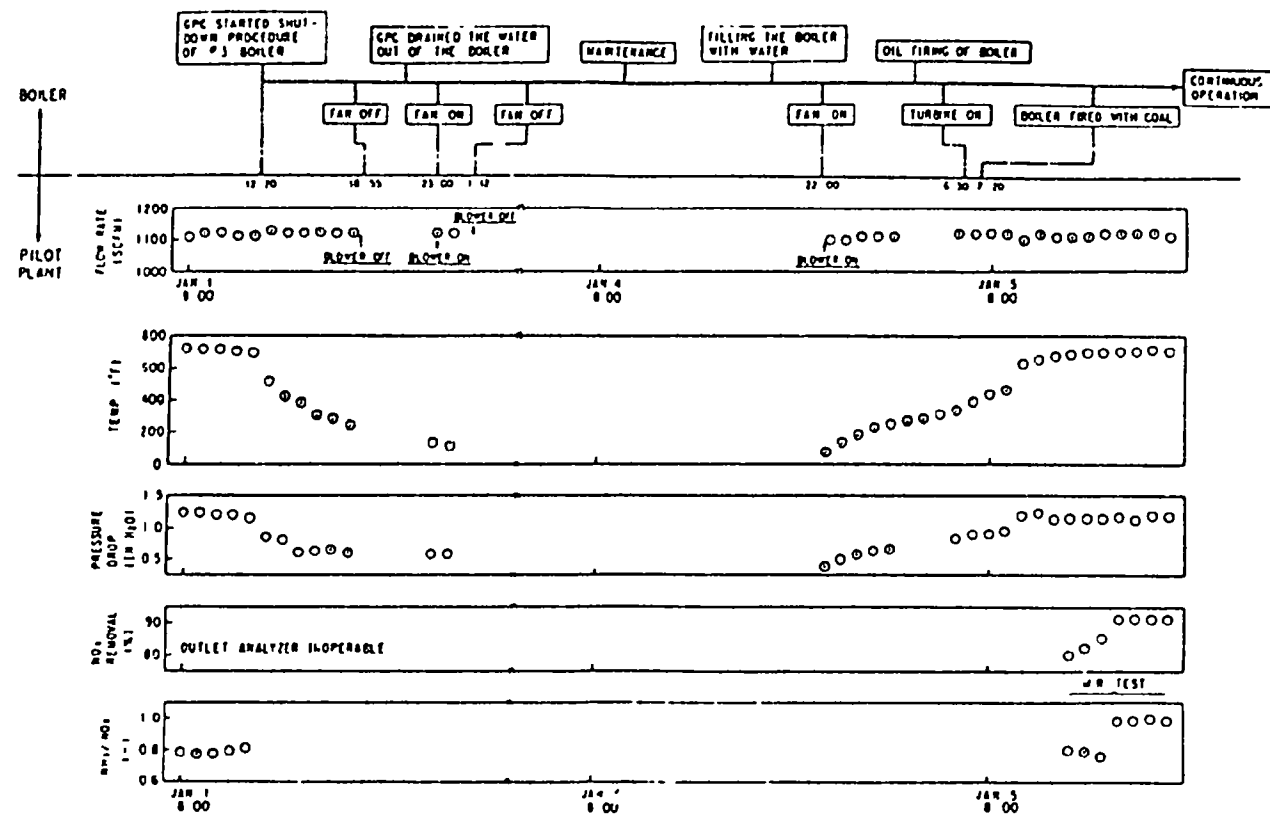
Figure 6-14. Test of System Performance with Boiler Shutdown and Start-up.

Pressure drop across the catalyst layer was 1.15 inches H_2O, and there was no change through this test.

From these results, the system proved that it could withstand the transient period of shut-down and start-up of the boiler.

Sudden Load Change

The boiler for a power plant may occasionally change load suddenly complying with variations of power consumption. The NO_x removal system should follow these sudden boiler load changes.

The following 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 once every two hours and continued for 24 hours from 11:00 AM on January 6, 1981. After this test was completed, catalytic performance was examined by a mole ratio test. The development of this test is shown in Figure 6-15.

The results showed a gradual increase in pressure drop and a slight decrease in NO_x removal efficiency. Catalytic performance checked by a mole ratio test also showed a slight decrease in NO_x removal efficiency:

81 percent at 0.8 mole ratio
87 percent at 1.0 mole ratio

Possibly, fly ash accumulated in ducting at the upstream of the reactor during low load operations and was blown off by the increased flue gas flow when the high load was reinstated. This fly ash settled on the catalyst resulting in slighly lower NO_x removal and slightly higher pressure drop. Experience in this pilot plant has shown that normal operating results are usually restored after a few days when this occurs. Therefore, there seems to have been no serious problem caused by these sudden load changes.

6-36
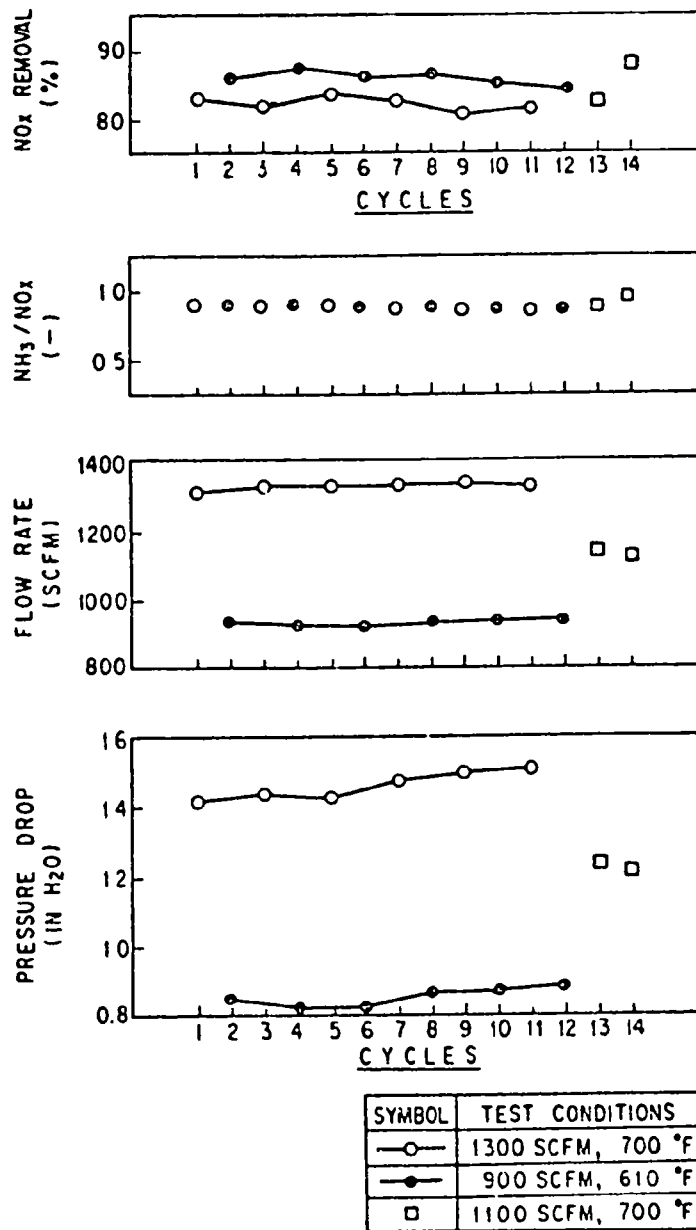
Figure 6-15. Test of System Performance with Sudden Load Change.

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, in order 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 on January 11, 1981. At a flow rate of 1,100 scfm, 700°F, 0.8 mole ratio, operation without sootblower was tried and pressure drop increase was observed. (See Figure 6-16). The pressure drop appeared unstable and increased gradually from 1.20 to 1.25 inches H_2O to 1.35 to 1.40 inches H_2O in approximately 28 hours, and seemed to continue to increase. The sootblower had to be operated at this time. From this observation it was concluded that operation of the sootblower was necessary.

From the beginning of the pilot plant operations with the third charged catalyst, the sootblower had been operated with air heated by the sootblower heater in order to prevent sudden shrinking and damage of catalyst due to cold air. The temperature of the air was 650°F. On January 12, 1981, after the 28 hours pilot plant operation without the sootblower, unstable and increasing pressure drop was being experienced, and it was decided to operate the sootblower. However, due to problems in the sootblower heater, heated air was not available. Thus, the sootblower was operated with ambient air as a necessity. One travel of the sootblower requires two minutes, and the temperature at the outlet of reactor decreased by only 5° to 10°F. There was no change in NO_x removal efficiency and pressure drop after this sootblower operation. Therefore, from this time to the end of the pilot plant operation, ambient air was used for the sootblower and it is possible that it can also be used in commercial systems.

6-38
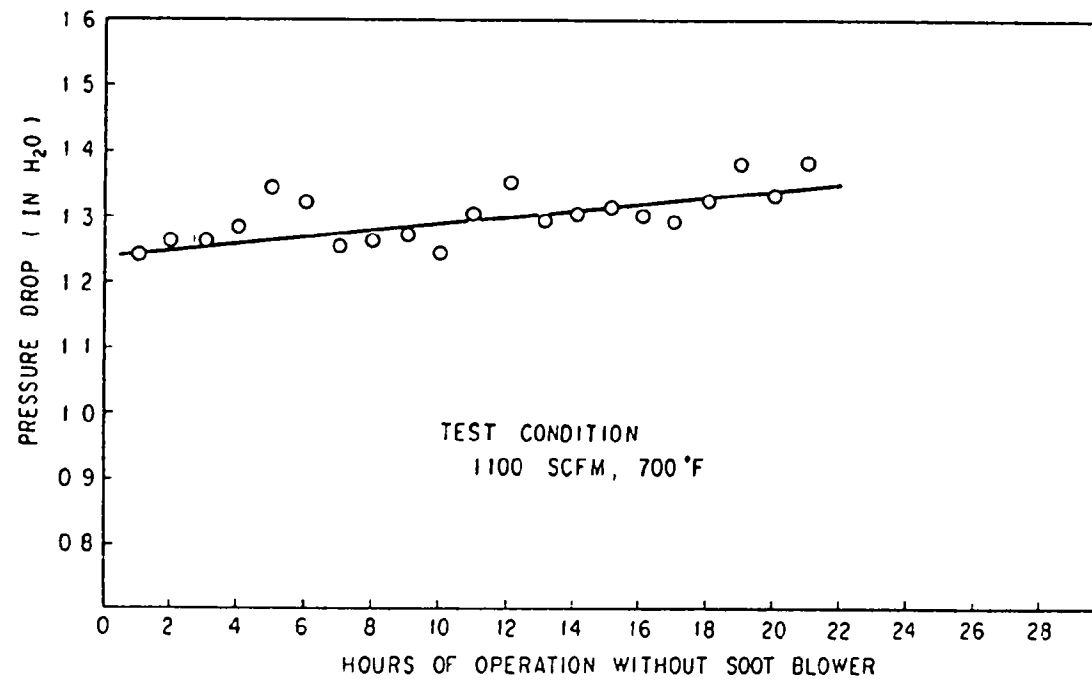
Figure 6-16.   Test of System Performance without Soot Blower.

REGENERATION OF CATALYST

Experience with NOXNON 500 catalyst indicated that washing with warm water could remove ammonium sulfate and ammonium bisulfate deposited on the surface along with fly ash which had adhered to the catalyst surface. This resulted in some degree of regeneration of deteriorated catalyst. Since the catalytic composition of NOXNON 600 is the same as that of NOXNON 500 the same effect could be expected in applying this procedure to NOXNON 600. Furthermore, preliminary tests were run in Hitachi Zosen's laboratory to confirm that the regeneration of NOXNON 600 by washing with warm water is applicable. Therefore, this test was run on site.

Preliminary Tests

Preliminary tests were tried at Hitachi Zosen's laboratory prior to the test on site. These tests evaluated the following:
- The influence of water washing on catalyst activity
- The chemical analysis of waste wash water.
- An abrasion test of the catalyst after water washing.

The following samples of catalyst were tested:
- Virgin catalyst which was produced for the EPA pilot plant and was stored at Hitachi Zosen's laboratory.
- Used catalyst from the EPDC pilot plant.
- Virgin catalyst from EPDC pilot plant (for chemical analysis of waste wash water only).

Due to the presence of sulfate and bisulfate, the pH of the water would be expected to become somewhat acidic (pH 2 to 3) when the used catalyst was washed by water. Therefore, instead of using ordinary water, a dilute (.001 N) sulfuric acid solution having a pH of about 3 was used.
- Amount of wash solution: 10 liters/sq. meter of catalyst.
- Washing times: 0, 5, 10, 30, 60, 120 minutes.

- Washing procedure: Samples of catalyst were soaked in the solution.
- Drying: at 110°C (230°F) overnight
- Analysis of solution: filtered and analyzed by atomic absorption spectrophotometry.
- Abrasion tests: Catalyst samples were tested by placing on vibrating screen with an alumina ball.

The results of these tests showed:
- Water soaking of the virgin catalyst for less than 60 minutes caused only a slight decrease in removal efficiency. However, soaking for 120 minutes caused a significant decrease in NO_x removal. (See Figure 6-17).
- Water soaking of used catalyst also showed the tendency to a decrease in NO_x removal efficiency depending on the soaking time. (See Figure 6-18).
- Chemical analysis of waste wash solution (see Table 6-1) indicates a definite increase in leached vanadium with increased soaking time.
- The abrasion tests (see Figure 6-19) indicated that the strength of the catalyst did not change after water soaking.

In conclusion, water washing with warm water appears to be applicable provided that the washing times are kept relatively short. Since water washing is obviously effective in removing sulfates and bisulfates along with adhered fly ash on the catalyst surface, it appeared possible that the catalyst could be regenerated to some extent by this procedure.

Site Tests

In the preliminary tests samples of catalyst were soaked in washing solutions. However, soaking in water is impractical for commercial systems. Therefore, instead of soaking in wash water

6-41
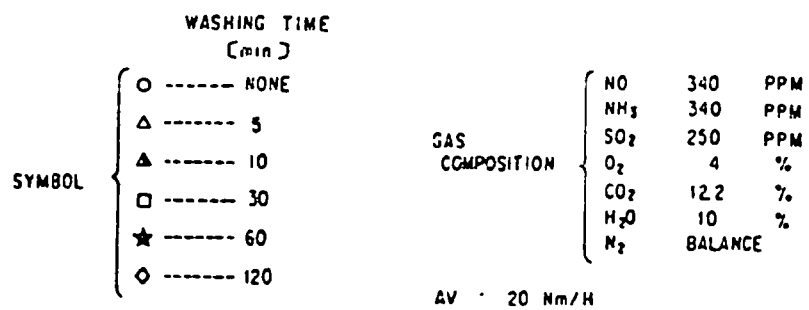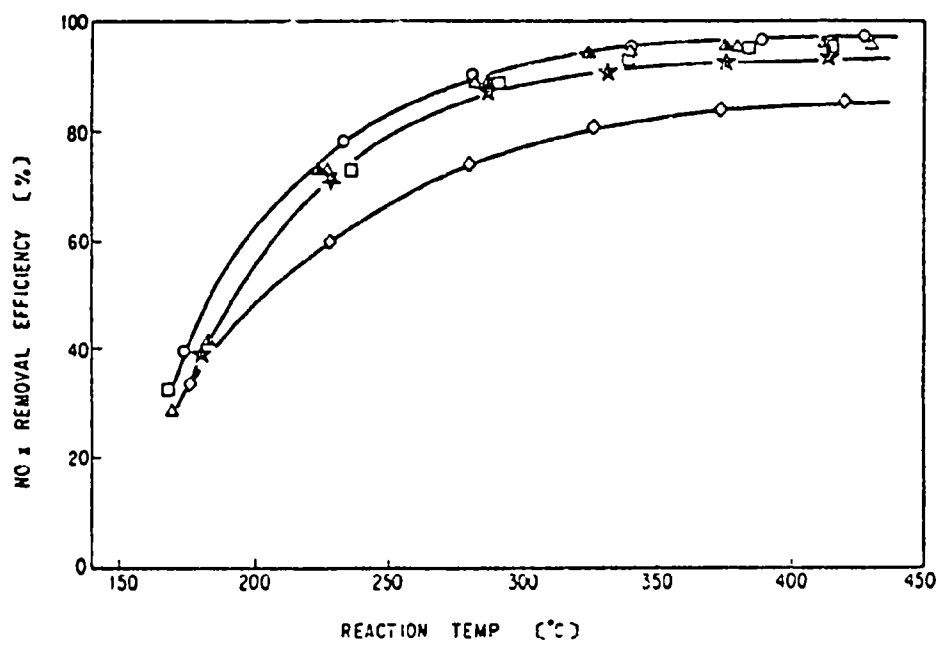
Figure 6-17   Influence of Water Washing on
                Catalytic Activity (Virgin Catalyst).
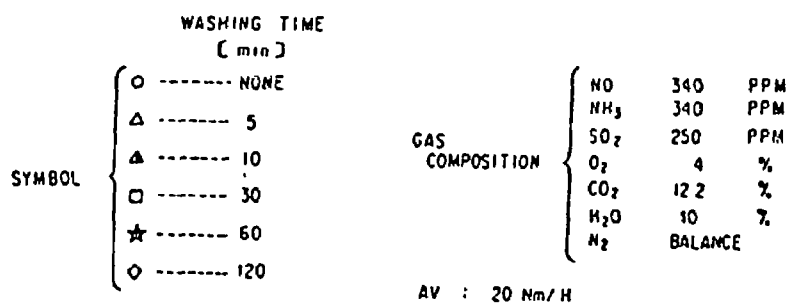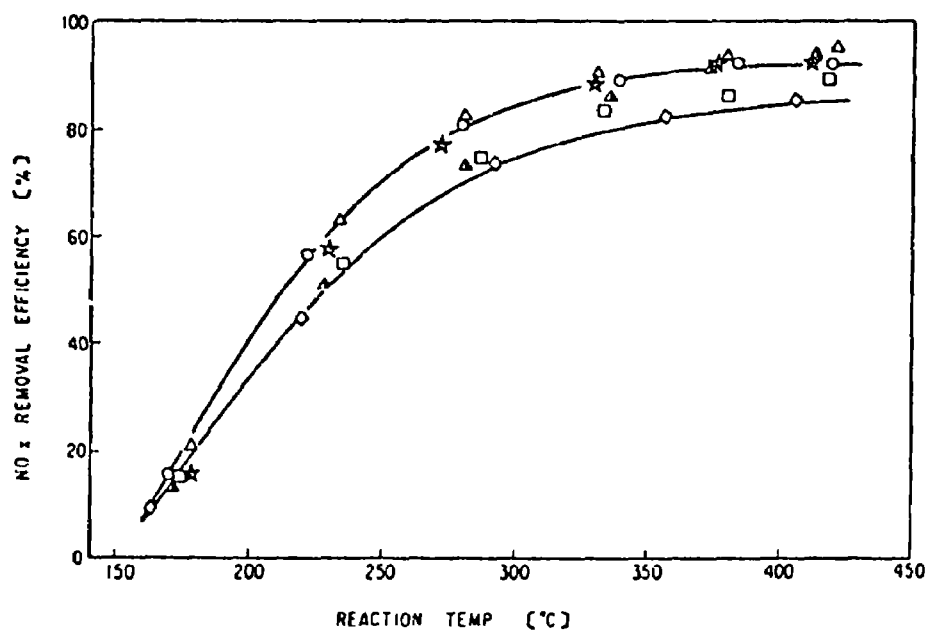
Figure 6-18   Influence of Water Washing on
                Catalytic Activity (Used Catalyst).
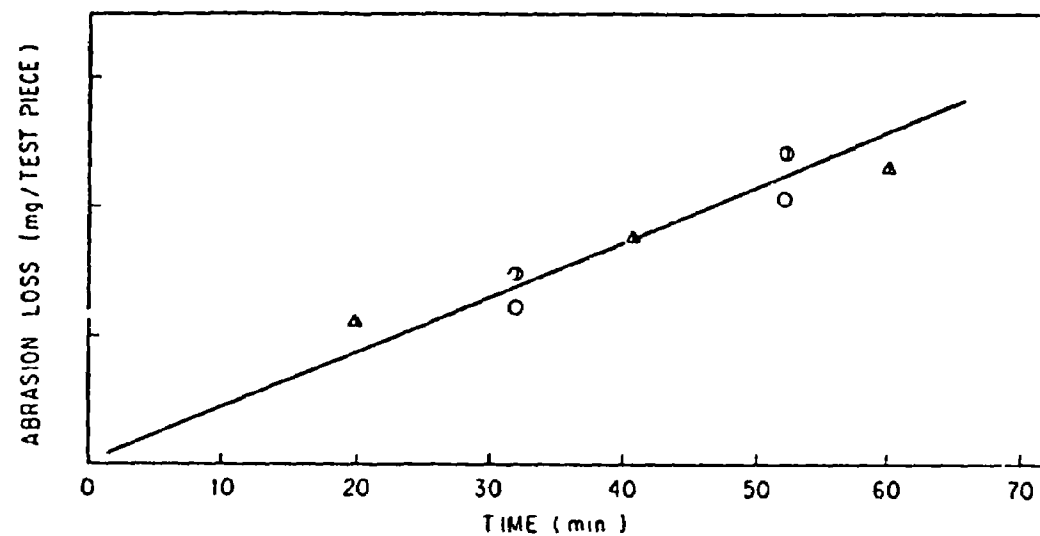
Figure 6-13 Abrasion Test.

TABLE 6-1.   ANALYSIS OF SAMPLES OF WASH WATER

Catalyst	Washing Time (minutes)	Na	K	Fe	V	Al	Ti
Virgin Catalyst	5	na	na	na	3.6	3.8	none
	10	na	na	na	3.5	2.7	none
	30	na	na	na	6.2	2.9	none
	60	na	na	na	7.0	3.1	none
	120	na	na	na	9.0	2.0	none
Used Catalyst	5	9.6	5.0	1.2	3.9	35.3	none
	10	10.0	5.0	1.2	4.4	41.1	none
	30	10.6	5.6	1.6	5.6	44.8	none
	60	11.5	5.6	1.0	5.6	59.5	none
	120	10.0	5.6	0.3	6.3	41.8	none

NOTE: na = not analyzed

the catalyst was washed with warm water delivered from a water hose. The manhole at the upper side of the reactor and the 14" elbow at the bottom of the reactor were removed. Water at 104°F was directed from the 1" hose back and forth across the catalyst surface as evenly as possible. The water flow was 2.7 gpm and this continued for two hours.

Samples of the waste wash water were taken every 15 minutes and the pH of the samples were measured right after sampling. Chemical analyses were performed later.

After the water washing the manhole and elbow were reinstalled, the blower was restarted, and the catalyst was dried with air at 600 scfm and 230°F overnight.

The analysis of the samples of wash water are shown in Table 6-2.

The regeneration procedure was evaluated by running a mole ratio test at 1,100 scfm and 700°F. The results were compared to an earlier mole ratio test performed before the catalyst was washed. The improved results can be seen from curves on Figure 6-23.

Before the washing test a sample of catalyst was taken for analysis. After the plant was dismantled other samples were taken for analysis. These analyses were performed by Hitachi Zosen in Japan.

After the regeneration the activity of the catalyst appears to have been restored to its original condition as indicated by mole ratio tests. (See Figure 6-20). This would indicate that it may be practical to restore catalyst activity by water washing.

Table 6-3 (Analysis of Used Catalyst) shows the results from Hitachi Zosen's laboratory. Except for vanadium, the elements and groups shown in this table are not included in the catalyst and are, therefore, derived from the fly ash and appear to be removed from the catalyst to some extent by water washing.

The vanadium content of the catalyst decreased with water washing. The actual amount of lost vanadium cannot be estimated precisely because the catalyst is a solid and uniform sampling

TABLE 6-2. SAMPLES OF WASH SOLUTION

Sample No.	Time of Sample	pH of Sample	Color of Sample	Chemical Analysis (mg/l)					
				Na	K	Fe	V	NH_4	SO_4
1	13:35	2.0	Dark Green	46	360	6	470	190	2860
2	13:40	1.8	Dark Green	185	1140	38	780	74	7580
3	13:50	1.8	Dark Green	175	1160	45	650	74	7280
4	14:05	2.1	Green	83	383	9.2	490	29	1060
5	14:20	2.4	Green	29	188	2.4	220	5.0	2850
6	14:35	2.4	Green/Gray	47	306	24	140	6.2	1910
7	14:50	2.7	Gray/Blue	19	125	1.0	80	8.4	650
8	15:05	2.8	Gray/Yellow	13	88	0.6	65	11.0	280
9	15:20	3.1	Gray/Blue	11	63	0.4	45	5.0	190
10	15:30	3.9	Yellow/Green	6.5	35	0.4	27	3.6	10
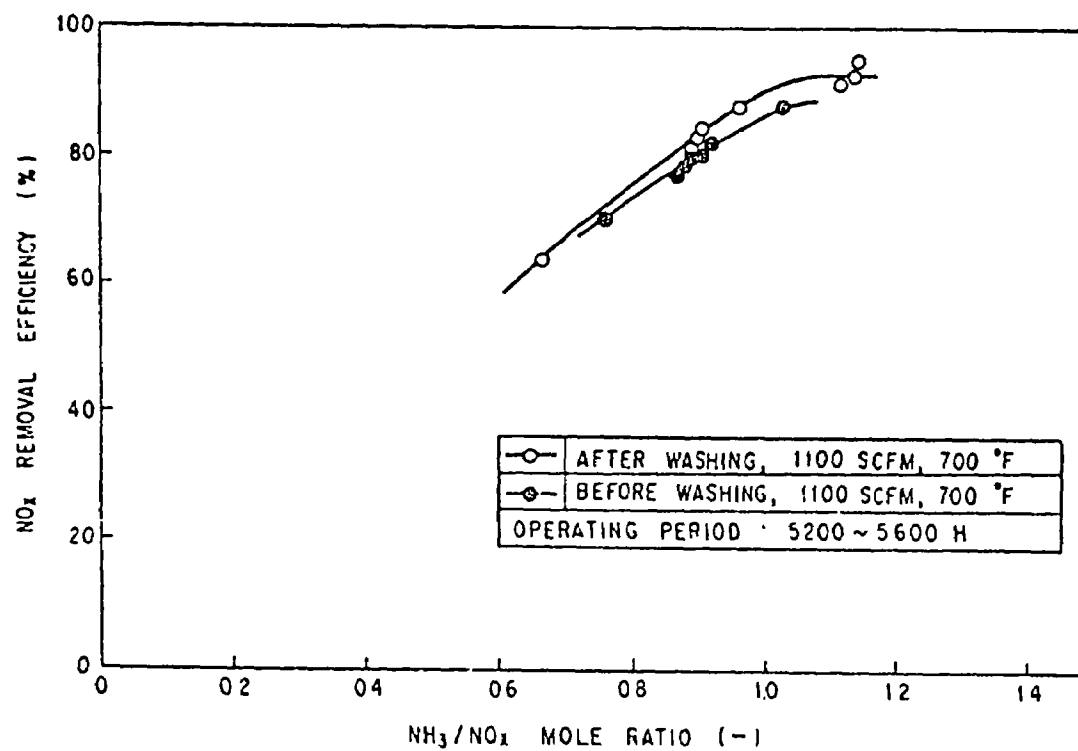
Figure 6-20 NO_x Removal Efficiency after Water Washing.

TABLE 6-3 - ANALYSIS OF USED CATALYST

(wt %)

Sample	Na	K	S	NH_4	Decreased V *
Before Washing	.09	.32	.18	.008	0
After Washing					
Top Block**	.07	.18	.07	.002	4.1
Top Block	.06	.13	.17	.002	2.8
Second Block	.06	.12	.13	.002	10.6
Third Block	.09	.22	.20	.002	2.3
Bottom Block	.07	.12	.18	.002	7.8

* Percent of decreased vanadium as compared to amount of vanadium in sample before washing.

** This test piece was from the used catalyst immediately after water washing. Other test pieces were taken from the used catalyst after three days of operation following the regeneration of catalyst.

·is difficult. Also, fly ash interferes with this analysis.
Nevertheless, though the loss was small, repeated water washing
is not recommended.

. The waste wash solution includes various elements which
leached from the fly ash as shown in Table 6-2 (Samples of Wash
Solution), and some of these metals are toxic. Water treatment
before disposal would be required.

CHANGES IN PRESSURE DROP IN RELATION TO NO_x REMOVAL EFFICIENCY

In the treatment of coal-fired combustion flue gas, it is
well-known that the adhesion and clogging caused by fly ash,
along with the formation and deposition of ammonium sulfate and
bisulfate, cause problems. In NO_x removal systems, these prob-
lems are first noticed as an increase in pressure drop. There-
fore, 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.

Calculation of Pressure Drop

The following equation has been established by Hitachi Zosen
for calculation of pressure drop on NOXNON 500 catalyst and
NOXNON 600 catalyst:

$$dPcal = \frac{n}{e^2}(f.\frac{L}{D_e} + C).\frac{r_g \cdot V_i^2}{2g}$$

whereas,

dPcal	= pressure drop (mmAq)
n	= number of catalyst layers (-)
e	= void fraction (-)
f	= friction coefficient (-)
C	= constant (-)

L   = length of catalyst layer (m)
D_e = hydraulic diameter (m)
r_g = density of gas (kg/m^3)
v_i = superficial gas velocity (m/s)
g   = acceleration of gravity (m/s^2)

The above equation has proven to be accurate in the measurement of pressure drops from various pilot plants.

Data from the EPA pilot plant obtained in flow rate tests performed in May 1980 were evaluated according to this equation. These data were plotted in Figure 6-21 and clearly show the pressure drop variation through changes in flue gas flow rate, temperature, and other operating conditions. Actual measured pressure drop was slightly higher than calculated. However, in the range of 1000 to 1500 SCFM the flue gas flow rate did not influence the pressure drop variation.

Pressure Drop Observations

On May 14, 1980, at the first catalyst inspection, additional asbestos yarn was added to ensure the seal between the reactor shell and catalyst box. As a result, dPact/dPcal increased from 0.9 to 1.0 - 1.1 and stable operation continued at this level until the second catalyst inspection on June 27, 1980.

From this date, several experiments were carried out, and the tendency for pressure drop changes are 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, from time to time, 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

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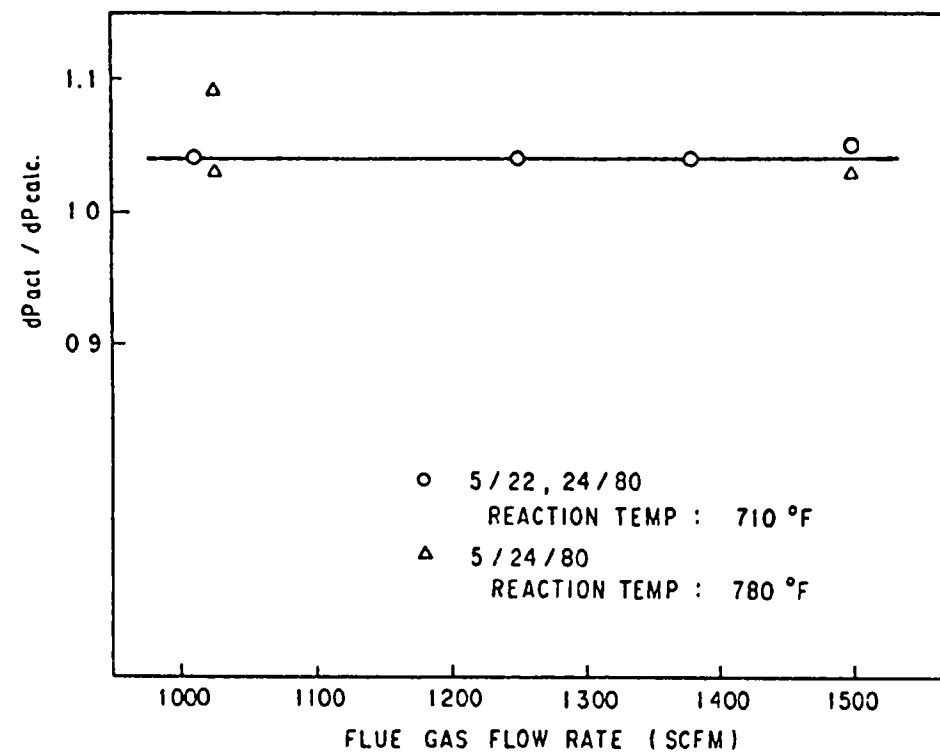
Figure 6-21   Effect of Flow Rate on dPact/dPcalc.

after some twenty days of steady operation the flow rate was changed from 1300 SCFM to 1500 SCFM and after two 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 Unit #3 Boiler operated the economizer soot blower 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 surfaces while the boiler was in operation. This abnormal maintenance work caused a serious increase in pressure drop.

- Operation of the soot blower seems to be necessary for coal-fired combustion flue gas. On January 10-11, 1981 the pilot plant was operated for approximately 28 hours without the sootblower. The pressure drop increased and after the sootblower was returned to operation it took three days to restore the pressure drop.

As described above, the pressure drop across the catalyst bed often drifted upward for a while and then gradually decreased. Overall, however, the operation was relatively stable except when the boiler tubes were water washed in an abnormal manner on December 12, 1980 causing a permanent increase in pressure drop. The washing obviously released a large amount of accumulated fly ash from the tubes and some of this material obviously entered the reactor. This unusual slug of material could have led to this pressure drop increase.

Relationship between Pressure Drop and NO_x Removal Efficiency

Relationship between pressure drop and NO_x removal efficiency observed before and after the first and second catalyst inspections are as follows:

6-53

date	flow rate (scfm)	pressure drop (inches H_2O)	NO_x removal (%)
05/15/80	1,500	1.1 - 1.14	91.5 - 95.0
06/24/80	1,500	1.08 - 1.11	89.5 - 92.0
06/26/80	the first catalyst inspection (1,200 hours)		
06/29/80	1,500	1.19 - 1.26	89.5 - 93.0
07/04/80	1,500	1.23 - 1.32	91.5 - 94.0
08/22/80	1,300	1.08 - 1.15	90.5 - 94.5
08/24/80	1,300	1.05 - 1.09	90.5 - 94.5
08/27/80	the second catalyst inspection (2,500 hours)		
08/27/80	1,300	1.27 - 1.32	88.5 - 91.5
08/28/80	1,300	1.15 - 1.29	89.5 - 92.5

After approximately 1,200 hours of operation, even though the pressure drop increased, NO_x removal was not influenced. However, after 2,500 hours operation, NO_x removal decreased with an increase in pressure drop.

When the pilot plant was shut down and started up, the pressure drop generally increased, and these increases were stepwise. The increase in pressure drop seems to have resulted from an increase in flue gas flow rate which was caused by partial clogging of flue gas passages by adhesion of fly ash.

The relationship between pressure drop and NO_x removal efficiency is shown in Figure 6-22. According to this data, NO_x removal efficiency decreased along with an increase in pressure drop.

Operating data after regeneration of catalyst shows the same NO_x removal efficiency as the initial efficiency. There was a definite decrease in pressure drop after regeneration, however, it was still about twenty percent above the initial level. Before regeneration it was about forty percent above the initial level.

Although the surface of the catalyst was cleaned by water washing, clogging by asbestos yarn found between catalyst blocks increased the pressure drop.

6-54
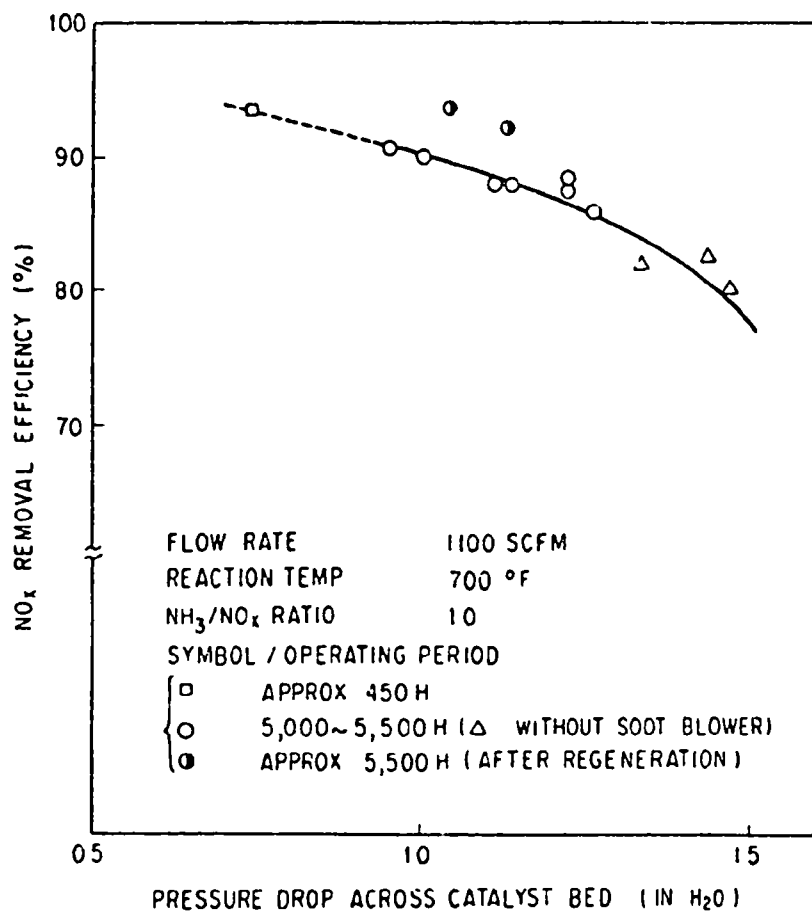
Figure 6-22.   Effect of Pressure Drop on NO_x

Removal Efficiency.

6-55

During the final stages of the pilot plant operation runs were made on two occasions without the use of the sootblower. These trials produced a rapid increase in pressure drop. It was concluded that the sootblower was required to maintain stable operation of the system.

CATALYST INSPECTIONS

Inspections

During the operation of pilot plant, the catalyst was inspected from time to time while the pilot plant was shut down.

05/14/80   The manhole was opened and the top surface of catalyst was observed through the manhole. The reason for this inspection was a relatively low NO_x removal efficiency and low pressure drop across catalyst.
Additional packing for sealing clearance between the catalyst and reactor wall was added.

06/27/80   Element of Flue Gas Heater was broken and replaced. During this shut down the catalyst was inspected.

08/27/80   The manhole of the reactor was opened to repair the Soot Blower. The catalyst was observed at this time.

11/15/80   The pressure drop fluctuated and increased, and the top surface of the catalyst was inspected from the manhole prior to starting mole ratio test. Nothing unusual was found at the top of catalyst.

12/26/80   The pilot plant was shut down for the Christmas holiday, and the catalyst was observed prior to the start-up.

01/26/81   Before regeneration of catalyst, the catalyst was observed and a few plates were sampled from the top layer.

02/02/81   The operation of pilot plant was terminated due to modification of Unit #3 Boiler of Georgia Power Company. The catalyst box was placed on the ground and all the catalyst plates were dismantled, inspected and sampled. Samples of catalyst were air mailed to Hitachi Zosen's Technical Research Institute, Osaka, Japan.

Observations

The first inspection on May 14, 1980--

On May 14, 1980 the pilot plant operation was shut down and the catalyst was observed from the manhole.

The catalyst had been installed in the reactor on April 22, 1980, however, due to repeated leakage from water tubes of Unit #3 Boiler, introduction of flue gas was stopped three times, and air was introduced into the system during these shut downs. Therefore, operating time with flue gas was only 185 hours. During the operation of 185 hours, NO_x removal seemed lower than designed efficiency.

Operating conditions before the shut down were:

flue gas flow rate	: approx. 1,200 scfm (1,930 Nm^3/h)
temperature	: 700°F (370°C)
NO_x concentration	: approximately 480 - 510 ppm
SO_2 concentration	: 600 - 1,000 ppm
pressure drop	: 0.76 - 0.79 inches H_2O (19 - 20 mm Aq)
NO_x removal efficiency	: approximately 91%

Pressure drop was almost the same as the calculated value, however, NO_x removal of 91% was lower than expected because designed NO_x removal at the beginning of the operation was 95%, and it seemed that there was no allowance to continue the operation for more than six months or one year. For this reason, it

was decided to shut down and observe the catalyst from the man-hole.

When the catalyst was observed it was seen that some of the corrugated plates had moved upward a few inches. None of the flat plates had moved. However, the upward displacement of these plates did not affect the efficiency of the system.

Some catalyst plates at the soot blower side were loose, and a few plates could be moved by hand. Bypassing of flue gas through these loose plates could be expected. These loose plates were tightened with added asbestos yarn.

The movement of the corrugated plates was caused by vibration of catalyst plates. It is required that in commercial applications a method of tightening and fixing in place the catalyst plates needs to be devised.

From the traces of fly ash adhering to the inside of the reactor shell, it was assumed that there might be partial flue gas flow through the clearance between the catalyst box and the reactor shell. Additional asbestos yarn was added to make sure of the seal.

No abrasion caused by fly ash or soot blowing was found.

Other Catalyst Observations from the Manhole--

From time to time the catalyst was observed when the operation was shut down for some reasons. Appearance of the catalyst was almost the same each time.

- Clogging and masking by fly ash was not observed. The operation and frequency of soot blower seemed suitable.
- Wearing away of catalyst components from the stainless wiremesh proceeded gradually through each inspection. However, the extent of the wear could not be observed until the catalyst was disassembled on 2/2/81. In general, abrasion seemed not serious.
- From the traces of fly ash on the inner surface of the reactor shell, the flue gas flow distribution seemed

acceptable.

- The upward displacement of some of the catalyst plates
  did not continue after the first inspection. It quickly
  reached a stable configuration.

Catalyst inspection after the operation terminated--

On February 2, 1981 the pilot plant operation was completed.
The catalyst was removed from the reactor, placed on the ground,
disassembled, and all of the catalyst plates were inspected. The
catalyst layers consisted of four blocks. The clearances in
between were carefully observed.

- Abrasion

  Abrasion was observed at only the flat plates in the
  top block. These appeared relatively abraded and a por-
  tion of the catalyst material had been completely worn
  away. Corrugated plates were not abraded. All of the
  plates in the other blocks were unabraded and looked
  like new catalyst.

- Clogging and Adhesion by Fly Ash

  The catalyst surface and the passages between the
  catalyst plates appeared to be clean. There was no
  fly ash adhering to and clogging the activated catalyst
  surface in the passages.

  However, in the clearances between the top block and
  the second block, fibers of asbestos yarn along with
  fly ash were found. The upper surface of the second
  block seemed to be blocked as much as 15 to 20 percent.

- In the clearance between the second and third block fine
  fly ash was found on the catalyst supports, piled in the
  clearance area, and blocking about 15% of the passages
  of the third block.

Discussion

In summary, approximately 30 to 35 percent blockage caused by fly ash was found in the clearance areas between the blocks of catalyst. Passageways between the plates of catalyst, however, were clean. The blockage could affect NO_x removal efficiency and pressure drop during operations. However, this did not prevent a successful operation of the project.

EVALUATION OF NOXNON 600 CATALYST

NO_x Removal Efficiency

Operation of the pilot plant with NOXNON 600 catalyst began on April 22, 1980 a..d terminated on February 2, 1981 due to scheduled maintenance of the boiler. The operating time with flue gas was 5,620 hours and tests examined during this period included catalyst life test, catalyst performance test, and transient tests. Controllability and reliability of the entire system was also evaluated at the same time.

Operating conditions were varied for the performance tests and transient tests. Therefore, when the activity of the catalyst was to be evaluated the 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_3/NO_x mole ratio of 1.0. Under these conditions the NO_x removal efficiency was measured over a period of several hours to determine the condition of the catalyst.

At the beginning of the operation   : 90 - 94%
At the end of August '80   (2500 hours): 90 - 94%
November 1980   (4000 hours): 90 - 92%
Before transient tests 12/80   (4420 hours): 90 - 91.5%
During transient tests 1/5/81   (5000 hours): 90 - 91%

During the transient tests, the "Sudden Load Change" test,

and the "Operation Without Soot Blower" caused an increase in the pressure drop and a decrease in the NO_x removal efficiency. Experience has shown that these deviations would be corrected if the pilot plant was operated under stable conditions with periodic operation of the sootblower. However, due to the limited period of time available, the testing program had to proceed and there was no opportunity for the catalyst restoration by the usual method. Before the regeneration of the catalyst the NO_x removal efficiency had slipped to 88 percent.

After the regeneration of the catalyst the efficiency was restored to 91.0 - 94.5 percent which was equivalent to the activity when initially tested.

Although approximately 30 to 35 percent of the cross-section area of the catalyst was found to be clogged by fibers of asbestos yarn along with fly ash (see "Catalyst Inspections on Site") satisfactory NO_x removal efficiency was achieved throughout the pilot plant operational period of over 5,000 hours.

It must be emphasized that the problems with the asbestos yarn were peculiar to this pilot plant and would not be expected in commercial applications.

Flue Gas Flow Rate

The NOXNON 600 catalyst for the pilot plant was designed to operate at an Area Velocity (A.V.) of 9.6 (1057 SCFM). However, operations at a much higher flow rate, at an A.V. of 15 (1650 SCFM) provided the desired 90 percent NO_x 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_x removal would be unchanged. As seen in Figure 6-3, the flow rate had little or no effect on the removal through the program.

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Mole Ratio

Mole ratio of ammonia to NO_x is an important variable affecting NO_x removal efficiency. One mole of ammonia reacts with at least one mole of NO_x and for a 90 percent removal a typical required mole ratio would be 0.9 to 1.0.

Ammonia Slippage

Since a continuous ammonia analyzer was not available during the operation of the pilot plant, ammonia had to be measured periodically by wet analysis. The method was not considered reliable until after August 15, 1980 and all data relative to ammonia leakage was collected after that time. By then the catalyst was probably partially clogged by asbestos fibers and fly ash and this caused a higher than expected ammonia slippage. At the flue gas flow rate of 1100 SCFM and an operating temperature of 700°F, the following results were collected in the operating period between 3200 hours and 4500 hours.

Mole Ratio	NO_x Removal (%)	Ammonia Slippage (PPM)
0.6	60	0
0.7	70	4 - 10
0.8	78	20 - 30
0.9	86	35 - 40
1.0	90	50 - 60

Operating Temperature

Between 640°F and 780°F the reactor operating temperature had no effect on NO_x removal efficiency.

NO_x Concentration in the Flue Gas

NO_x concentration does not affect NO_x removal efficiency within the typical boiler range of between 300 and 1,000 ppm.

SO_x Concentration in the Flue Gas

So_x concentration does not affect NO_x removal efficiency.

Power Plant Load Changes

The system can follow boiler load excursions in commercial systems without problems. This was proven by cycling the system three times between 0.4 MW equivalent and 0.95 MW equivalent without any consequent effect on catalyst activity.

Emergency Shut-off of Ammonia Feed

Automatic shut-off of ammonia is required to avoid build-up of deposits at low temperatures. This system was incorporated in the pilot plant and proved to be reliable through several cycles of temperature excursions.

Cold Start-up

The system exhibited no problems when it was started up with cold flue gas without prior heating of the system with hot air. This proved that in a commercial system no auxiliary heating is required for startups.

Boiler Shutdown and Startup

The system was allowed to shut down and start up with the boiler with no air purging of the system and without auxiliary heating. The system withstood this transient period without difficulty.

Sudden Load Change

Six cycles of sudden load changes were tried. This increas-

ed the pressure drop somewhat and the NO_x removal efficiency decreased slightly. However, experience has shown that normal conditions are usually restored after awhile during normal plant operations. Therefore, it is probable that sudden load changes will not cause serious problems.

Operation Without Sootblower

This brought an obvious decrease in NO_x removal efficiency and an increase in pressure drop. Therefore, periodic operation of the sootblower is imperative. The pilot plant normally operated the sootblower three times each day and this frequency seems adequate.

Regeneration of Catalyst

When the catalyst was washed with warm water the NO_x removal efficiency was restored to its initial efficiency. The pressure drop also seems to have been partially restored, however, this system was partially clogged with asbestos fibers which is not a typical condition. 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 flue gas flow. There was no abrasion in the other blocks. A commercial system should incorporate a tighter catalyst structure to avoid such vibration.

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Clogging

Anticipated clogging of the catalyst by fly ash was prevent-
ed by operation of 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 to 35 percent 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.

SECTION 7

CONCLUSIONS

NO_x REMOVAL EFFICIENCY

NO_x removal efficiency of more than 90% was demonstrated during the operating period of approximately 5,000 hours (from April 1980 to the beginning of January 1981) at the designed capacity of 0.5 MW equivalent.

Following the above period, transient tests were run and the operation of the pilot plant was terminated after 5,620 hours of cumulative operating time. Performance was not adversely affected by sudden boiler load changes, cold startups, sudden boiler shutdowns and start-ups, and low boiler load operations.

Therefore, it was expected that NO_x removal of more than 90% would have continued longer, for example, more than one or two years as was demonstrated in other pilot plant operations in Japan.

AMMONIA SLIPPAGE

Ammonia slippage i this pilot plant was higher than expected as shown below:

Mole Ratio	NO_x Removal (%)	Ammonia Slippage (ppm)
0.6	60	0
0.7	70	4 - 10
0.8	78	20 - 30
0.9	86	35 - 40
1.0	90	50 - 60

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These results probably resulted from the decrease in
apparent surface area of catalyst caused by clogging of the
clearances of catalyst plates with fibers of asbestos yarn
along with fly ash as described in "Catalyst Inspections."

Recent representative ammonia emissions with NOXNON 600
catalyst for coal-fired combustion flue gas demonstrated at a
pilot plant in Japan are, as referred to in Figure E-2 and E-3
in "Appendix E," less than 5 ppm at the NO_x removal efficiency
of more than 80% and less than 10 ppm at the NO_x removal effi-
ciency of more than 90%.

A commercial system would be expected to experience low
ammonia slippage similar to that found in tests in Japan. The
catalyst in the EPA pilot plant was obviously clogged by asbestos
and fly ash to an extent that a significant decrease in avail-
able surface area was found. Such a problem could not occur in
a commercial plant as asbestos would not be used for this purpose.
NOXNON 600 catalyst operating under typical conditions in the
presence of only fly ash would remain unclogged as shown by the
experience in Japanese pilot plant operations. Therefore, the
ammonia slippage data from the EPA pilot plant is not considered
valid and the Japanese experience seems more reliable.

PRESSURE DROP ACROSS CATALYST BED

The pressure drop in commercial applications would be ex-
pected to be the same as that experienced in the EPA pilot plant
provided that the size and configuration of catalyst, superficial
linear velocity of the flue gas across the catalyst, and the
temperature of flue gas are the same.

The pressure drop with NOXNON 600 catalyst in the EPA pilot
plant was between 1.0 and 1.4 inches H_2O. Such low pressure
drops reduce power consumption resulting in lower operating
cost.

CONTROL SYSTEM

The analyzers required frequent backflushing and cleaning of sampling probes and sampling tubes, the regular replacement of filters in the sample gas conditioner, and also frequent instrument air purging of the impulse lines of the controllers and indicators. However, in general, controllability and reliability of the pilot plant operation was satisfactory.

A desirable addition to the control system would be a continuous ammonia analyzer to measure slippage from the system.

FLY ASH 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 abrasion caused by fly ash, and to determine the necessity and operating conditions of the soot blower. Improvement of catalyst configuration to prevent clogging by fly ash is also expected.

REGENERATION OF CATALYST

Testing for catalyst regeneration was examined immediately before the pilot plant was dismantled, and the results were very encouraging. The regenerated catalyst exhibited properties of a virgin catalyst.

However, due to the limited time available, problems related to the catalyst regeneration had not been clarified. For ex-

ample, 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 investi-
gated 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 regen-
eration techniques.

APPENDIX A

EQUIPMENT DESCRIPTION

FLUE GAS INTAKE SYSTEM

Flue gas to be used as input to the pilot plant was drawn from the boiler flues downstream of the economizer and introduced to the Flue Gas Heater. Multiple withdrawal points from the flues were used to ensure that the pilot plant gas was representative of that in the flues, particularly with respect to fly ash (dust) content and particle size distribution.

REACTOR

The gas flows down through the reactor in contact with the catalyst and the NO_x content of the gas stream was reduced by reaction with the ammonia to form small quantities of gaseous nitrogen and water which remain in the gas stream.

The operating conditions in the reactor were specified so as to simulate the operating conditions of boilers in power plants.

Fly ash from the flue gas can settle out in the reactor and tend to block flow through the reactor and to blind it off and so partly inactivate the catalyst. These effects of dust settling in the reactor were kept within acceptable bounds through the use of soot blowing apparatus. This was demonstrated by the soot blower (which will be described in a later section).

The reactor size was 10' high, 23½" square, with a 16" transition at the bottom. The sootblower, described below, was attached to the reactor. The catalyst is described in Section 4 of this report.

FLUE GAS HEATER

The flue gas heater was designed to heat 1600 SCFM of flue gas from 550°F to 770°F.

The heater consisted of one unit containing 108 tubular elements connected into six subcircuits. The six subcircuits were divided into two groups, each of three circuits. One of the groups was on on-off control and the other was on automatic temperature control.

The entire heater was rated at 162 KW at 460 V, 3 phase.

The elements were contained in a weatherproof box of welded steel construction with a gasketed cover. A thermocouple which senses gas temperature was mounted on the back plate.

SOOT BLOWER

This was a retractable soot blower designed for side wall entry complete with all controls, supports, drive mechanism and control panel. The main lance was equipped with a removable cross T-head tube 3 inches in diameter 21½ inches long and having 11 x 3/16" diameter holes for gas injection in a vertical downward direction.

Lance traverse was 30-inches at an approximate speed of 24 inches per minute. Controls were provided for continuous extension and reaction of lance throughout the pre-set operating cycle.

The lance seal was gas-tight to prevent air leakage into the duct.

Blowing medium could be steam or air at 130 psig and a temperature of 660°F. A maximum design temperature and pressure were 840°F and 150 psig. The soot blower and auxiliaries were suitable for outdoor operation.

The blower was furnished with blowing medium inlet valve with an outside adjustable blowing pressure control.

The operation was semi-automatic. Operation was initiated

A-2

by a pushbutton. All subsequent operations were automatic. Initial operating cycle was set for 2.5 minutes. Upon completion of the cycle the lance would retract, the unit would shut down and be ready for re-start. All components such as motor, limit switches, etc. were wired to terminal strips in a junction box for connection. The unit was driven by an electric motor.

HEATER FOR SOOT BLOWER

Service Conditions

A stream of 400 SCFM of air entering at 130 psig and ambient temperature was to be heated to 660°F ± 20°F. The same heater was also required to heat an alternate flow of 1760 lb/hr of steam entering at 130 psig and 360-500°F to 600°F ± 20°F.

Description

One circulating heater rated at 96 KW (48 KW/bundle) at 460 V, containing two bundles each of 15 General Electric Calrod stainless sheathed tubular hairpin elements, was installed.

The pressure drop was below 0.1 psig at stated flow and temperature. The heater was mounted and wired together with the control enclosure on a structural steel base.

The control assembly was in a weather-proof enameled steel vented enclosure with all components mounted and wired.

DUST SEPARATOR SYSTEM

This consisted of a vertical standing cyclone having an inlet and outlet flanged connection, motor driven rotary valve to discharge collected particulates, and solids receiving hopper, all mounted on a fabricated steel supporting structure.

The system was designed to remove particulates larger than 30 microns at a design flow with a minimum efficiency of 95%.

Collected particulates were discharged into a receiving hopper
by means of a motor-driven rotary valve. The hopper was designed
to discharge solids into a vacuum removal system.

BLOWER

Performance and Characteristics

ACFM	3,700
S.P.	21" hot, 50" cold
BHP	17.4 hot, 41.4 cold
RPM	3550
Weight	2230 lb.
Operating Temp.	770°F hot, 70°F cold
Tip Speed	26062 Ft/Min.
Rotation Discharge	CCW-UB

Coolant Water

Max. Pressure	80 psig
Normal Pressure	15 psig
Flow Rate	0.7 GPM

Motor Specification
25 HP
460 Volt
3600 RPM

AMMONIA SUPPLY UNIT

Ammonia Storage Tank

1000 gallon water capacity having a design working
pressure of 250 psig.

Instrumentation and Controls

Relief Valves --

One d al relief valve assembly consisting of one three-way
dual control valve and two Safety Valves having a relieving cap-
acity of 1500 CFM air per minute at 250 psig. The dual control
valve permitted the shutting off of one relief valve at a time
leaving one protecting the tank at all times. Each relief valve
had full discharge capability for the size of the tank.

Valves and Fittings --

Tank and openings were fitted with excess flow check valves
between the openings and the shut-off valve with the exception
of the relief valve assembly.

Vaporizer Assembly

Immersion Heater with one external temperature cut-out
switch.

APPENDIX B

INSTRUMENTS AND CONTROLS

CONTINUOUS ANALYZERS

Description of Equipment

Flue gas samples were drawn continually from the ducts before
and after the reactor. Comparisons between the inlet and outlet
samples were made to determine the removal of NO_x. The inlet port
was located downstream of the flue gas heater and upstream of the
ammonia feed point. The outlet was located in the duct downstream
of the cyclone and before the fan.

The inlet port and outlet port of the reactor had sample
probes (sintered 20 micron stainless steel) that removed large
particles. The probes were connected via heated lines to the
sample gas conditioners where the sample stream was again filter-
ed (5 microns) and diluted with dry air to reduce the dew point
of the sample stream.

The sample gas from the pick off point was conducted to the
Sample Gas Conditioner via electrically heated sample tubing.
The heated tube was teflon and was controlled at a temperature
of $170^{\circ}F$. Heating maintained the sample temperature above the
dewpoint and prevented alteration of the gas constituents.

Sample Gas Conditioner --

The Sample Gas Conditioner was a module which prepared a flue
gas sample for analysis; the gas was filtered and the dewpoint was

lowered by dilution. The module contained a network of control
flow orifices which accurately diluted the sample for NO/NO$_x$ and
SO$_2$ analysis. This network was contained within a heated chamber
which was controlled at a temperature above the sample gas dew-
point.

Control Module --

Periodically the stack monitoring sequence was interrupted
to check the operation of the analyzers. First the analyzer
was purged with zero gas, which was then followed by a calibra-
tion gas (supplied in compressed gas cylinders) purge. The
sample lines were periodically purged with a short surge of com-
pressed air in order to clear the lines and the stack probe
filter of particulates.

Model 10A Chemiluminescent NO/NO$_x$ Analyzer (Inlet Sample) --

For measurement of NO concentrations, the gas sample to be
analyzed is blended with O$_3$ (ozone) in a reaction chamber. The
resultant chemiluminescence is monitored through an optical fil-
ter by a high sensitivity photomultiplier positioned at one end
of the chamber. The filter/photomultiplier combination responds
to light in a narrow wavelength band unique to this NO/O$_3$ reac-
tion. The flow parameters can be adjusted in such a way that the
output from the photomultiplier tube is linearly proportional to
the NO concentration.

To measure NO$_x$ concentrations (i.e., NO plus NO$_2$), the
sample gas flow is first diverted through a NO$_2$-to-NO converter.

Model 14D NH$_3$ Analyzer (Outlet Sample) --

This analyzer is the same as the NO$_x$ analyzer described
above except for the following changes:

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The TECO Model 14D has much higher sensitivity. This higher sensitivity is required because it measured the outlet stream which contained low levels of NH_3 and NO_x. The Model 14D also utilized a dual chamber design which was to continuously monitor the NH_3 concentration.

The chemiluminescent NO_x analyzer was converted to a NH_3 analyzer by adding a molybdenum converter to the inlet of one of the reaction chambers (resulting in the measurement of NO and NO_2) and using a stainless steel converter with the other reaction chamber (resulting in the analysis of NO, NO_2 and NH_3). The analyzer read the output signal from both chambers and automatically subtracts the two signals, and the resultant difference is the NH_3 concentration.

Pulsed Fluorescent SO_2 Analyzer --

The SO_2 Analyzer within the system used Thermo Electron's pulsed Fluorescent technique. Within the Analyzer a high-intensity ultraviolet lamp is pulsed to excite SO_2 molecules, which fluoresce in the blue region of the light spectrum, between 1900 and 2300 angstroms. A characteristic fluorescent light is emitted as the excited molecules returned to the ground state.

This specific illumination is passed through a narrow band filter and impinges upon the sensitive surface of a photomultiplier tube. The emitted light is proportional to the concentration of SO_2 molecules in the sample. The amplified and conditioned signal provides a meter reading and an electronic analog signal for recorder output.

Operating Problems

Dry Ammonia Analysis --

The analytical system was originally intended to provide

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continuous ammonia readings in the flue gas after the reactor.
However, it was not a workable system. The outlet TECO Model 14D
analyzer was provided with two converters. Converters are re-
quired to convert any NO_2 or NH_3 which may be present to NO
because the analyzer can only detect NO. The design provided
a molybdenum converter which would convert only NO_2 but not NH_3.
The second converter, the stainless steel unit, converts both
the NO_2 and the NH_3. The gas sample would be split between two
reactor chambers: one equipped with a molybdenum converter, the
other with a stainless steel converter. The analyzer could read
the output from both chambers and could automatically subtract
the two signals, the resultant difference being the ammonia con-
centration.

The problem which finally defeated the system was the loss
of ammonia in the sample line and probably in the probe. If
ammonia was introduced directly into the analyzer the ammonia
could be detected. However, samples from the duct lost its
ammonia content upon passage through the teflon tubing (heated
to 170°F). The ammonia probably reacted with sulfuric acid mist
deposited in the tubing. Possibly there was also a reaction with
SO_2. Whatever, the system could not be used to detect ammonia.

Problems with Flue Gas Sample Probes --

The analytical system for the determination of NO_x had sev-
eral unexplained problems for some time. Major efforts to deter-
mine their causes were undertaken. It appears that the causes
were catalytic material which had become imbedded in the sample
probe.

The first indication of something amiss occurred when it was
found that the outlet NO_x reading (after the reactor) was affect-
ed by the rate of sampling flow through the sample line. This
effect was noticed when the oxygen analyzer pump was turned on or
off. The oxygen analyzer draws the sample from the same sample

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probe and line as the NO_x analyzer. There is a tee in the line before the NO_x gas conditioning system and the gas is drawn throu_h one leg of the tee to the oxygen analyzer. When the oxygen pump was turned on the amount of sample drawn from the outlet duct through the probe and sample line was doubled. When this happened the apparent NO_x reading dropped substantially.

Concerted efforts to determine the cause of this phenomenon were unsuccessful. The analyzer gave accurate results when the standard gas was fed through the sample line with or without the oxygen pump in operation. There were no leaks found in the system and the same event occurred after washing of the sample line.

The next unexplained event occurred when the solenoid valves on the backflush (blowback) lines were replaced. Every 15 minutes the sample lines are blown back for a few seconds with high pressure instrument air to remove dust from the probes. It was found that the solenoid valves which supply the air had orifices much smaller than specified. The amount of air supplied for blowback was lower than desired. Larger solenoid valves were obtained and installed. These provided considerably more air.

When these new solenoid valves were tested two differences in the operation were discovered. One, the oxygen pump did not appear to affect the outlet NO_x readings. The second and probably more significant change was that the NO_x concentration in the outlet dropped substantially to an unusually low level. Again, a series of tests were run to determine whether the analyzer was operating correctly and all indicated that it was. No explanation could be found. Eventually, after a few weeks the oxygen pump began to affect the readings but the overall outlet NO_x levels remained low.

Some weeks later, the next event took place. The outlet sample probe was replaced by a new one and the apparent NO_x outlet level rose sharply from 40 PPM to 62 PPM under the existing operating conditions. Replacing the original probe restored

the original condition. The new probe was temporarily replaced by another new probe which was found to provide identical results. Apparently, the original probe was reducing the NO_x level for some reason.

The original outlet probe had been installed on the inlet line to replace the original inlet probe which had developed a crack.

One interr_ing experiment was then tried. Ammonia was introduced before the inlet probe to see if there was an effect on the NO_x reading at the inlet probe. Theoretically it should not because no catalyst is present. However, when ammonia was sent into the line upstream of the inlet probe the apparent NO_x in the inlet dropped from 580 PPM to 140 PPM. This test was run for about twenty minutes. It was apparent that a reaction between ammonia and NO_x took place and could only have occurred within the probe itself. The old probe obviously had catalyst imbedded within it!

The catalytic material within the probe must have been picked up over a period of time because of the erosion of some of this material from the reactor. Some of the fine particles of active material were probably imbedded in the sintered stainless steel of the probe.

After this the probes were often replaced temporarily to make certain that the permanent probes were functioning properly. This condition of probes may exist in other installations and should be considered when experimental or full-scale operating plants are evaluated. Such probes, if contaminated with catalyst, will produce artificially high removal indications.

VENTURI FLOW TUBE

Flue gas flow was measured by means of a Vickery-Simms Venturi tube equipped with pressure taps. From the reduced pressure at the venturi throat the flow rate was measured. The

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Venturi was about 60" long with a throat diameter of 7 3/8".
The unit was installed in a 14" pipe duct with flanges.

DATA LOGGER

Data Acquisition System (Logger) for scanning and logging analog inputs.

The system included internal visual display and recording of all data and a cassette tape recorder for peripheral output recording. The data logger had the following specifications:
    (1) Enclosure: Portable for desk top with dust cover
    (2) Input Terminals: Screw Type
    (3) Inputs: 12 linear, 4-20 MA (includes 2 spare points), 6 non-linear direct thermocouple ISA Type K (includes 2 spare points)
    (4) Fixed Data Entry: For data identification associated with a test
    (5) Day Calendar
    (6) Serial Output
    (7) Power Supply: 120 V, 60 Hz
Cassette Recorder Specification:
    (1) Enclosure: Desk top mounting
    (2) Recording Media: Magnetic tape cassette
    (3) Storage Capacity: It is compatible with T.I. 733 ASR
    (4) Power Supply: 120 V, 60 Hz

APPENDIX C

AMMONIA FEED MEASUREMENT

The feed of ammonia into the process was a major problem and was still subject to question after the program was completed. The major problem was to accurately measure a continuous, very low flow of gaseous ammonia. The flow was only about 0.5 SCFM. The flowmeter originally selected for this installation finally proved to be the best one to be found. This was a unit provided by Ramapo Instrument Co. It was a rotometer in which a differential transformer was attached to the float. The position of the float was precisely measured by the differential transformer which develops an output voltage dependent upon its position which is proportional to the flow of fluid.

This unit caused a considerable degree of trouble for some time. Most problems were due to plugging of the opening with bits of dirt. A very fine filter was installed just before the meter which helped to avoid most of the plugging. The installation of damping fluid for some reason helped to avoid further problems.

The unit was calibrated frequently using a dry test meter. The ammonia was bypassed after the control valve through the meter for an accurately determined period of time and the volume of gas was determined. This dry test meter was then often checked by a standardized unit with air. At one time a cylinder of ammonia was used as the source of ammonia for most of a day. The cylinder was weighed before and after the test and the results indicated the meter to be very accurate.

Other meters were tried at times because of the early difficulties with the Ramapo. One was a mass flow meter which worked

extremely well for a day or so but which eventually became in-
operable. After several such trials it was abandoned. It seemed
that ammonia was leaking into the electronics causing corrosion.
Another unit tried was a turbine meter which after installation
and failure the vendor admitted to its inapplicability.

APPENDIX D

WET AMMONIA ANALYSIS

Because of the failure of the dry ammonia analytical system,
ammonia slippage had to be determined by wet methods. There was
much trial and error experimentation before a satisfactory pro-
cedure could be attained. The problem was to obtain an accurate
volume of flue gas (which was at $700^{\circ}F$, and which contained quan-
tities of fly ash and sulfur oxides) to absorb all of the ammonia
present in this gas in a suitable absorbent, and to analyze the
absorbent for its ammonia content.

The sampling probe proved to be a big problem because of the
possibility of acid mist deposition at temperatures below $300^{\circ}F$
with consequent reaction with ammonia in the gas. After many
suggestions and trials a very simple procedure was adopted. The
probe consisted of a 3/8" SS tube inserted through a 3" nozzle
into the duct. The tube passed through a drilled hole in a 3"
cap which sealed the nozzle. The tube was welded to the cap to
prevent leakage. A short length of teflon tubing was then used
to connect the stainless steel tube to the sample impinger train.
After each analysis, the stainless probe and the teflon tube were
rinsed with distilled water and this water was added to the liquid
in the impinger train. In this way no special precautions were
required to avoid condensation in the sample lines.

The ammonia was absorbed in a standard impinger train set
in a tray of ice. Sulfuric acid was used as an absorbent. The
gas was drawn through the system by a vacuum pump and the volume
was measured with a dry test meter.

The absorbent was analyzed by a Nessler method using Hach

equipment and chemicals. The sample had to be neutralized before analysis.

This procedure was checked by Radian and by Hitachi Zosen and was considered to be reliable.

APPENDIX E

CORRECTION OF NH_3/NO_x MOLE RATIO

The operation of the EPA pilot plant which was to demon-
strate the performance of Hitachi Zosen's NO_x removal process
for the application to coal-fired utility boilers was successful-
ly completed on February 2, 1981. However, there seemed to be a
few questionab$_{i}$ operating data which were inconsistent with the
results of fundamental experiments obtained in the laboratory of
Hitachi Zosen s Technical Research Institute and experimental
results collected at various pilot plants in Japan. One of them
is the high NO_x removal efficiency when the mole ratio of NH_3 to
NO_x is lower than approximately 0.8 to 0.9, and the other is
the high ammonia slip even though the mole ratio is fairly low.

In the presence of oxygen, the amount of ammonia for reduc-
ing NO_x is equimolecular to the quantity of NO_x in the flue gas.
This is proven from fundamental data and from results collected
at pilot plants. Figure E-1 shows the results of fundamental ex-
periments obtained in the laboratory. Figure E-2 and Figure E-3
are experimental results collected at a pilot plant applied to
flue gas exhausted from a coal-fired utility boiler in Japan. In
this pilot plant, the flue gas source from the boiler was divided
into two streams. One of them was directly introduced into the
NO_x removal reactor as a high fly-ash-loaded flue gas. The other
was introduced into a reactor after the fly ash was removed by a
high temperature electrostatic precipitator as a low fly-ash-loaded
flue gas. Figure E-4 and Figure E-5 show the results obtained in a
pilot plant which is applied to oil-fired boilers in Japan.

All of these data indicate that the catalytic reduction of

NO_x with ammonia is an equimolecular reaction between NH_3 and NO_x. When the EPA pilot plant was operated, all the analyzers and flow meters which measure the mole ratio and ammonia slip were frequently and carefully calibrated and adjusted.

It is well understood that all flow meters and analyzers involve some errors. However, because of frequent and careful calibrations and adjustments, direct readings of flow meters and analyzers including the results of wet ammonia analysis were considered to be fairly accurate.

On the other hand, the concept of the equimolecular reaction between ammonia and nitrogen oxide which had been proven through fundamental experiments through various pilot plant operations is also an indisputable theoretical principle.

From the above circumstances, an attempt was made to correct the NH_3/NO_x mole ratio based on the assumption that the selective catalytic reduction of NO with NH_3 is an equimolecular reaction.

After August 1980, ammonia emission could be measured by wet analysis, and this wet analysis also made the correction of NH_3/NO_x mole ratio possible based on an ammonia balance. The correction was made as described below:

The amount of ammonia injected into the reactor was determined as the total of an equimolecular amount of ammonia to remove NO_x and the amount of ammonia emitted from the reactor. The amount of NO_x introduced into the reactor was not changed. Thus, the mole ratio of ammonia to NO_x was corrected.

The basis of this correction in which the NH_3 flow meter was deemed inaccurate was brought about from the very precise and susceptible configuration of the rotometer, especially, very narrow clearance between the rotor and the tapered tube which could result in erroneous flow rate measuring, particularly, at the very low flow rate. The phenomena is not anticipated in commercial systems because of the large flow rate of ammonia. However, when the small flow rate is applied to pilot plant operations, the likelihood of an erroneous measurement seems to require taking this into consideration. This correction is also

supported by the fact that higher NO_x removal efficiencies were observed when the mole ratios were relatively low.

As a result of this correction, for example, the effect of NH_3/NO_x mole ratio on NO_x removal efficiency and ammonia slippage after 3.200 hours operation, which is described in Section 6, "Evaluation of NOXNON 600 Catalyst," was revised as follows:

Mole Ratio		NO_x Removal	Ammonia Slippage
Direct Reading	Revised	(%)	(ppm)
0.5	0.6	60	0
0.6	0.7	70	4 - 10
0.8	0.8	78	20 - 30
0.9	0.9	86	35 - 40
1.0	1.0	90	50 - 60

In this correction, it should be emphasized that the NO_x removal efficiencies and ammonia emissions were not changed, and only traversed along with the corresponding mole ratios.
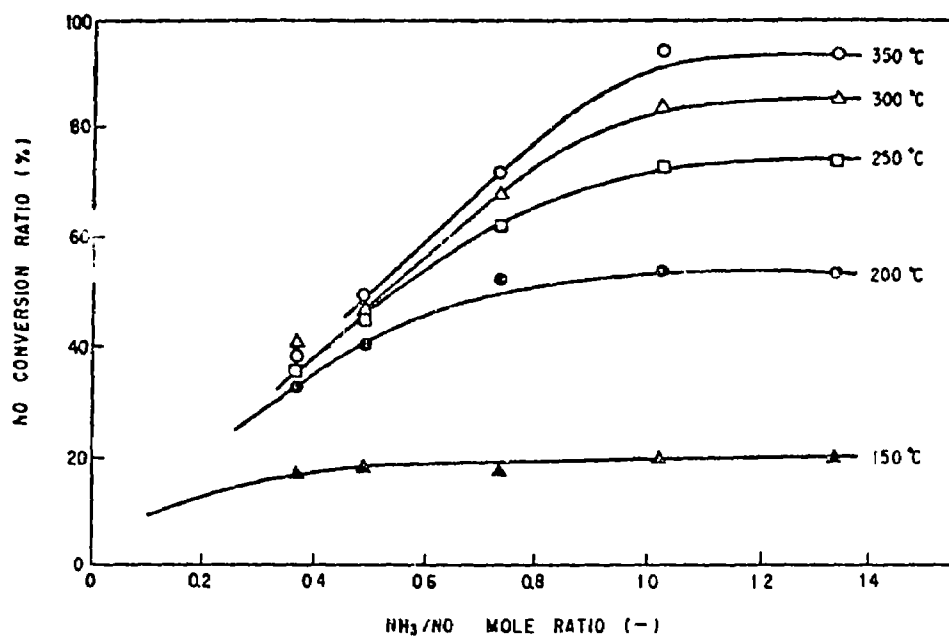
SOURCE   SYNTHETIC GAS (FROM CYLINDERS)
TEST CONDITION
    A V   :   20. 4 Nm³/m² H
    GAS COMPOSITION   ·
        ⎧ NO   500 PPM
        ⎪ SO₂   250 PPM
        ⎨ O₂   6 %
        ⎪ CO₂   10 %
        ⎪ H₂O   10 %
        ⎩ N₂   BALANCE

Figure E-1.   Effect of NH_3/NO Ratio on NO Conversion
              Rate.
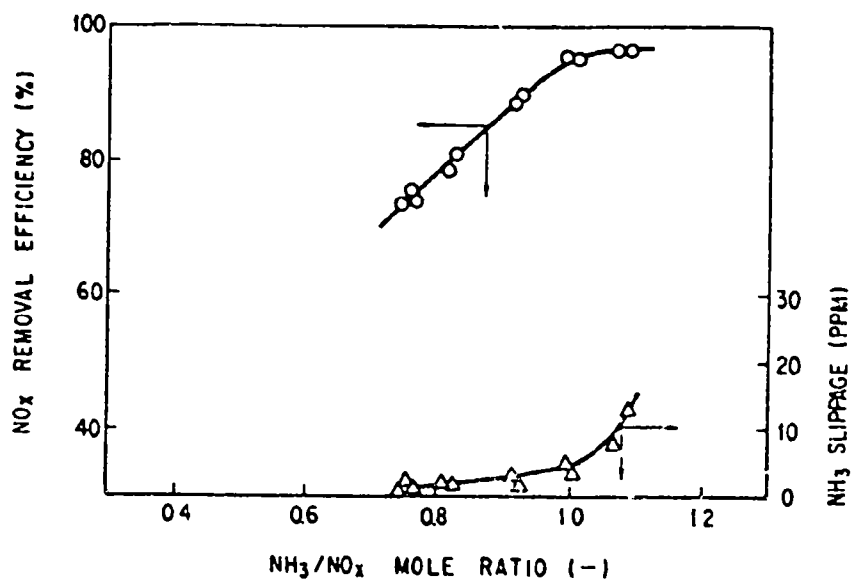
SOURCE : COAL - FIRED UTILITY BOILER

(HIGH FLY ASH LOADING GAS)

TEST CONDITION

FLY ASH CONCENTRATION : APPROX 14 9/Nm³
(58 grain/SCFD)
A V : 89 Nm³/m²·H
REACTION TEMPERATURE : 350 °C (662 °F)

Figure E-2. Effect of NH_3/NO_x Ratio on NO_x Removal Efficiency.

(Reported at "The U.S.-Japan Exchange of NO_x Control Technical Information Conference" by Hitachi Zosen on May 27, 1981)
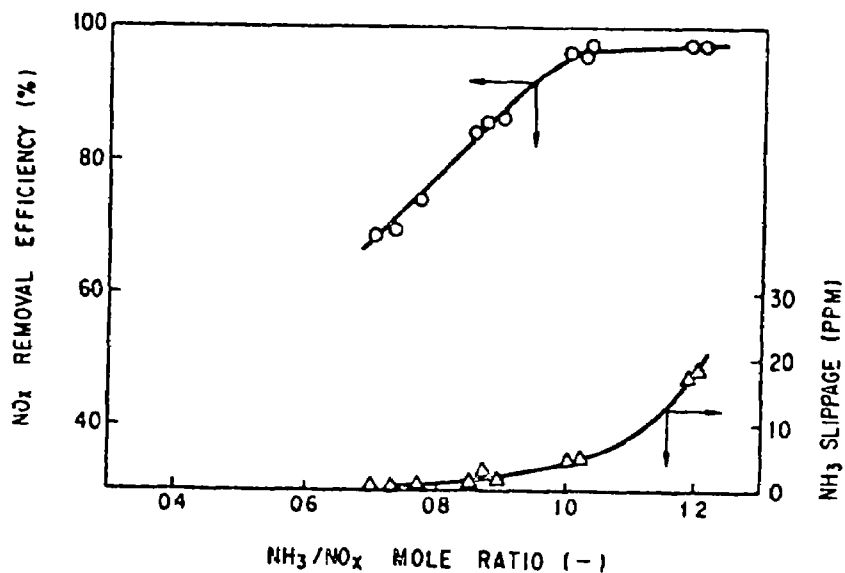
SOURCE · COAL - FIRED UTILITY BOILER

(LOW FLY ASH LOADING GAS)

TEST CONDITION

FLY ASH CONCENTRATION : 0 1 ~ 0 2 g/Nm^3

(0.04 ~ 0.08 grain/SCFD)

A V : 10 2 $Nm^3/m^2 H$

REACTION TEMPERATURE : 350 °C (662 °F)

Figure E-3. Effect of NH_3/NO_x Ratio on NO_x Removal Efficiency.

(Reported at "The U.S.-Japan Exchange of NO_x Control Technical Information Conference" by Hitachi Zosen on May 27, 1981)

SYMBOL	TEMPERATURE
O	375 °C (707 °F)
△	356 °C (673 °F)
□	339 °C (642 °F)
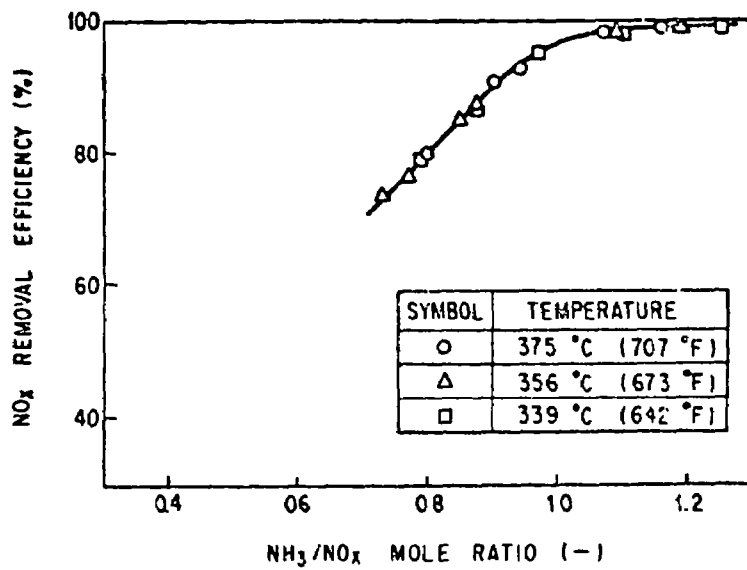
SOURCE : OIL - FIRED STEAM GENERATOR

TEST CONDITION

A V   : 850~864 Nm³/m²·H

GAS COMPOSITION :

NO_x 97~140 PPM
SO_x 32 PPM   (SO_3 : 7 PPM)
O_2 5 6 %
CO_2 10 5 %
H_2O 11 5 %
N_2 BALANCE
DUST   25 ~45   mg/Nm³   (Dry Basis)

Figure E-4.   Effect of NH_3/NO_x Ratio on NO_x
Removal Efficiency.
(Normal Flow Rate)

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NOₓ REMOVAL EFFICIENCY (%) vs NH₃/NOₓ MOLE RATIO (—)

SYMBOL	TEMPERATURE
○	375 °C (707 °F)
△	356 °C (673 °F)
□	339 °C (642 °F)
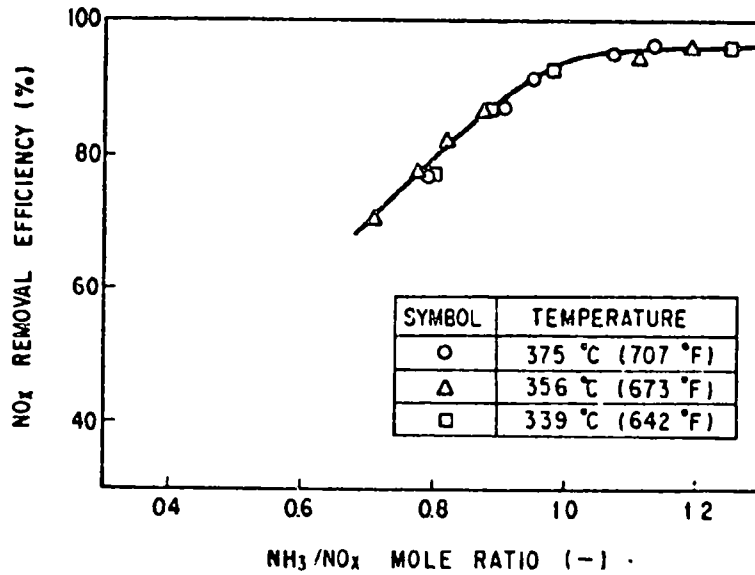
SOURCE : OIL – FIRED STEAM GENERATOR

TEST CONDITION

A. V : 119 ~ 121 $Nm^3/m^2 \cdot H$

GAS COMPOSITION :

NO_x 97 ~ 140 PPM
SO_x 32 PPM (SO_3 . 7 PPM)
O_2 56 %
CO_2 105 %
H_2O 114 %
N_2 BALANCE
DUST   25 ~ 45 mg/Nm^3 (Dry Basis)

Figure E-5. Effect of NH_3/NO_x Ratio on NO_x
Removal Efficiency.

(High Flow Rate)

E-8

APPENDIX F

ABBREVIATIONS

abs	absolute	hr	hour
acf	actual cubic feet	in.	inch
Btu	British thermal unit	kg	kilogram
°C	degrees Celsius	kW	kilowatt
cm	centimeter	l	liter
dia	diameter	lb	pound
ESP	electrostatic precipitator	m	meter
°F	degrees Farenheit	mg	milligram
FGD	flue gas desulfurization	min	minute
FGT	flue gas treatment	mm	millimeter
ft	feet	MW	megawatt
ft/sec	feet per second	Nm^3	normal cubic meter
g	gram	ppm	parts per million
gal	gallon	psi	pounds per square inch
gpm	gallons per minute	SCR	selective catalytic reduction
gr	grain		
hp	horsepower	sec	second
		scf	standard cubic feet

APPENDIX G

CONVERSION FACTORS

To convert from	To	Multiply by
British thermal unit	gram-calories	252
degrees Fahrenheit-32	degrees Centigrade	0.5555
feet	centimeters	30.48
cubic feet	cubic meters	0.02832
feet per minute	centimeters per second	0.508
cubic feet per minute	cubic meters per second	0.000472
gallons	liters	3.785
gallons per minute	liters per second	0.06308
grains per standard cubic foot	grams per normal cubic meter	2.288
pounds	kilograms	0.4536
standard cubic feet per minute (60°F)	normal cubic meters per hour (0°C)	1.607
tons (short)	metric tons	0.90718

H-1

EPA PROJECT

N° 600 LOG CHART ③
(5-2-'80 ~ 5-11-'80)

SOOT BLOWER CYCLE

DATE		TIME	PSIG	°F
5-8	1ST	—	—	—
	2ND	14:57	89	780
	3RD	22:23	27	780
5-9	1ST	08:01	86	760
	2ND	14:02	42	740
	3RD	23:09		
5-10	1ST	08:02	40	700
	2ND	17:32		
	3RD	—	—	—
5-11	1ST	00:16	13	623
	2ND	14:16		
	3RD	08:02	62	501

EPA PROJECT

N°600 LOG CHART (5)
(3-12-'80 ~ 3-21-'80)

SOOT BLOWER CYCLE

DATE		TIME	°F	PSIA
3-12	1 ST	9:11	820	19
	2 ND	15:19		
	3 RD	17:45	730	49
3-13	1 ST	8:00	810	33
	2 ND	16:16	749	42
	3 RD	23:37	770	42
3-14	1 ST	8:19	760	44
	2 ND	—	—	—
	3 RD	9:00	770	42
3-15	1 ST	9:11	760	42
	2 ND	17:33	760	44
	3 RD	23:55	773	64
3-16	1 ST	7:57	777	42
	2 ND	16:09	770	96
	3 RD	23:37	770	99
3-17	1 ST	8:19	749	44
	2 ND	17:12	770	42
	3 RD	0:10	775	43
3-13	1 ST	9:14	770	44
	2 ND	19:12	770	43
	3 RD	0:15	775	44
3-19	1 ST	8:13	770	39
	2 ND	17:11	772	42
	3 RD	0:15	770	44
3-20	1 ST	8:26	770	44
	2 ND	15:94	775	42
	3 RD	0:00	752	44
3-21	1 ST	8:09	770	44
	2 ND	16:50	770	99
	3 RD	23:30	770	99

NH3 SLIPPAGE

TEST №	ppm	
3-3-1	1.0	
3-3-2	0	
3-3-3	0.7	
3-3-4	0.8	
3-3-5	0	

INLET NOx (ppm) ──△──
OUTLET NOx (ppm) ──●──
600 400 200 0

C-3

NOx REMOVAL (%) ──●──
100 90 80 70 60
[NH3/NO2] (—)
1.4 1.2 1.0 0.8 0.6 0.4

Δ TEMP. °F ──■──
BOILER LOAD (%) ──○──
800 600 400

FLUE GAS FLOW RATE SCFM ──□──
FILTER PRESSURE DROP (in H.C.) ──▲──
1600 1400 1200 1000
1.2 1.0 0.8 0.6

SO2 CONC. (ppm) ──●──
O2 CONC. (%) ──○──
200 100 0

NORMAL SHUT DOWN FOR CATALYST INSPECTION

REPLACED OUTLET FILTER WITH KLEENZOR FILTER

TEST 3-3-1
TEST 3-3-2
TEST 3-3-3

TEST 3-3-4
TEST 3-3-5

PROBLEM WITH DATA LOGGER AND OUTLET ANALYZER

DATE (HOURS)	3/12 8:00	3/13	3/14	3/15	3/16	3/17	3/18	3/19	3/20	3/21	3/22 8:00
	(137.25)	(161.25)	(183.25)	(200.92)	(224.92)	(248.92)	(272.92)	(296.92)	(320.92)	(344.92)	(368.92)

TYPING GUIDE SHEET

INLET NO$_x$ (ppm)

OUTLET NO$_x$ (ppm)

INLET LEAK TEST

OUTLET ANALYSIS SYSTEM HAS PROBLEMS

NO$_x$ REMOVAL (%)

NH$_3$/NO$_x$

NH$_4$/NO$_x$

H-4

TEST

R. TEMP (°F)

BOILER LOAD (q°C)

FLUE GAS FLOW RATE SCFM

FLOW RATE TEST

PRESSURE DROP in w.c.

SO$_2$ CONC. (PPM)

O$_2$ CONC. (qPC) (VOL %)

DATE (HOURS)

5/22 8:00 (348.92) 5/23 (372.92) 5/24 (396.92) 5/25 (440.92) 5/26 (464.92) 5/27 (488.92) 5/28 (512.92) 5/29 (536.92) 5/30 (560.92) 5/31 (584.92) (608.92)

DATE		NH$_3$ SLIPPAGE		SOOT BLOWER CYCLE		
		TIME	PPM	TIME	°F	PSIG
5-22	1ST		8	9	15	44
	2ND				40	44
	3RD				70	45
5-23	1ST				50	44
	2ND				30	44
	3RD				55	44
5-24	1ST		10		50	42
	2ND				50	46
	3RD				52	64
5-25	1ST				55	40
	2ND				40	44
	3RD				40	44
5-26	1ST				55	45
	2ND				50	62
	3RD				50	42
5-27	1ST				50	40
	2ND				50	40
	3RD				40	42
5-28	1ST				50	42
	2ND				50	40
	3RD				50	42
5-29	1ST				50	40
	2ND				60	45
	3RD				60	42
5-30	1ST				50	40
	2ND				70	42
	3RD				62	42
5-31	1ST				55	40
	2ND				55	45
	3RD				90	45

TYPING GUIDE SHEET

G-5

OUTLET ANALYSING SYSTEM HAS PROBLEMS

OUTLET NH3 (PPM)
OUTLET NOx (PPM)
NOx REMOVAL (%)
NH3/NOx (—)
R. TEMP (°C)
BOILER LOAD (%)
FLUE GAS FLOW RATE (SCFM)
PRESSURE DROP (m.m.C.)
SO2 CONC. (PPM)
O2 CONC. (VOL%)

DATE		NH3 SLIPPAGE		SOOT BLOWER CYCLE		
		TIME	PPM	TIME	°F	PSIG
6-1	1ST			1:18	750	49
	2ND			19.52	750	42
	3RD			22.50	80	42
6-2	1ST			8.01	750	44
	2ND			18.52	770	42
	3RD			22.52	740	44
6-3	1ST			8.44	775	44
	2ND			13.52		
	3RD			6.17	760	44
6-4	1ST			8.18	770	44
	2ND			12.12	770	44
	3RD			9.31	740	44
6-5	1ST			7.39	765	49
	2ND			5.18	760	44
	3RD			22.34	760	44
6-6	1ST			9.09	730	42
	2ND			15.31	772	42
	3RD			19.44	772	42
6-7	1ST			17.21	733	46
	2ND			19.41	880	45
6-8	1ST			8.52		44
	2ND			12.44	801	42
	3RD			18.52	740	42
6-9	1ST			11.44	760	42
	2ND			5.31	750	42
	3RD			13.53	741	42
6-10	1ST			7.31	750	42
	2ND			13.53	50°C	45
	3RD			19.4	84°	45

DATE (HOURS)
6/1 8:00 (608.92) 6/2 (632.92) 6/3 (656.92) 6/4 (680.92) 6/5 (704.92) 6/6 (728.92) 6/7 (752.92) 6/8 (776.92) 6/9 (800.92) 6/10 (824.92) 6/11 8:00 (848.92)

EPA PROJECT
N° 600 LOG CHART Ⓖ
(6-11-'80 ～ 6-20-'80)

EPA PROJECT

N°600 LOG CHART ⑪
(6-21-'80 ~ 6-30-'80)

PARTIAL SHUT DOWN DUE TO
GAS SHUT DOWN #2 BOILER.

INLET NOₓ (ppm) — NO_x
600
400
200
0

OUTLET NOₓ (ppm)
300
100
0

NOₓ REMOVAL (%)
100
90
80
70
60

NH_3/NO_x
1.1
1.0
0.9
0.8

FLOW RATE TEST

R. TEMP (°C)
800
600
400

BOILER LOAD (MW)
200
80
0

FLUE GAS FLOW RATE SCFM
1600
1400
1200
1000

PRESSURE DROP (mmAq)
in. H₂O

SO_3 CONC. (ppm)
2000
1000
0

O_2 CONC. (VOL%)
9.0
7.0
5.0
3.0

DATE (HOURS)
7/1 8:00   7/2   7/3   7/4   7/5   7/6   7/7   7/8   7/9   7/10   7/11 7:00

(1219.09) (1243.09) (1267.09) (1291.09) (1315.09) (1339.09) (1363.09) (1387.09) (1411.09)

N° 600 LOG CHART
(7-1-'80 ~ 7-10-

DATE	TIME	PPM	TIME
7/1	1ST		9:10
	2ND		14:55
	3RD		—
7/2	1ST		4:00
	2ND		3:00
	3RD		20:00
7/3	1ST		7:53
	2ND		18:03
	3RD		1:55
7/4	1ST		9:50
	2ND		17:00
	3RD		0:55
7/5	1ST		8:17
	2ND		23:06
	3RD		23:05
7/6	1ST		7:00
	2ND		14:00
	3RD		22:30
7/7	1ST		9:00
	2ND		22:50
	3RD		20:30
7/8	1ST		9:00
	2ND		19:41
	3RD		0:37
7/9	1ST		0:00
	2ND		22:10
	3RD		23:00
7/10	1ST		8:55
	2ND		15:50
	3RD		23:40

NH₃ TEST #1   NH₃ TEST #2
NH₃ TEST #3   NH₃ TEST #4

N° 609 LOG CHART
(9-11- 60 ~ 9-20- '8

DATE		NH3 SLIPPAGE		SOOT BL CYCLE	
		TIME	PPM	TIME	DP
9/11	1ST			8:00	76
	2ED			15:32	78
	3RD			0:57	85
9/12	1ST			8:09	79
	2ED			15:55	72
	3RD			0:52	79
9/13	1ST			7.59	76
	2ED			15:07	76
	3RD			0:11	51
9/14	1ST			1:59	92
	2ED			15:59	79
	3RD			7:12	84
9/15	1ST			7:03	81
	2ED		0.8	14:01	89
	3RD			21:59	84
9/16	1ST		2.0	9:17	79
	2ED			16:59	79
	3RD			-	-
9/17	1ST		0.1	9:12	91
	2ED		6.1	16:59	81
	3RD			22:59	79
9/18	1ST		<1	9:12	79
	2ED			15:42	79
	3RD			23:59	71
9/19	1ST			9:04	71
	2ED			16:29	91
	3RD			22:59	79
9/20	1ST			8:11	79
	2ED			15:29	71
	3RD			22:59	70

INLET NOx (ppm)   OUTLET NOx (ppm)
600
400
200
0

PROBLEM WITH OUTLET ANALYZER

NOx REMOVAL (%)
100
90
80
70
60

NH3/NOx

PROBLEM WITH GCA SYSTEM. OUTLET NOx ANALYZER NOT REFEED

6-H

GAS TEMP °F
800
600
400

BOILER LOAD (MW)
200
100

FLUE GAS FLOW RATE SCFM
1600
1400
1200
1000

PRESSURE DROP

SO2 CONC. (PPB)
2000
1000
0

O2 CONC. (VOL %)

DATE
9/11 8:00   9/12   9/13   9/14   9/15   9/16   9/17   9/18   9/19   9/20   9/21 8:00
(HOURS)
(1411.09) (1935.09) (1052.09) (1463.09) (1509.09) (1231.09) (1559.09) (1577.09) (1603.09) (1629.09) (1851.09)

TYPICAL GUIDE SHEET

TYPICAL CONE E SHEET

EPA PROJECT

N°600 LOG CHART

(9-21-'80 ~ 9-30-)

PROBLEM WITH OUTLET ANALYZER

PROBLEM WITH SAMPLE LINE HEATERS.

INLET NOx (ppm)

OUTLET NOx (ppm)

NOx REMOVAL (%)

NH3/NOx

NH3/NOx

R. TEMP. (°F)

BOILER LOAD (MW)

FLUE GAS FLOW RATE (SCFM)

FILTER PRESSURE DROP (in. W.C.)

SO2 (ppm)

O2 CONC. (vol.%)

DATE (HOURS)

DATE	TIME	SLIPPAGE PPM	SOOT CYCLE	
9/21	1ST		1.0	8
	2ND	6.1	1.01	8
	3RD		27.75	8
9/22	1ST	6.1	1.19	8
	2ND		12.94	8
	3RD		12.22	8
9/23	1ST	6.1	1.19	8
	2ND		12.52	8
	3RD		9.13	8
9/24	1ST	1.0	1.10	10
	2ND		11.22	11
	3RD		11.50	11
9/25	1ST	6.0	1.10	11
	2ND		11.22	11
	3RD		9.53	11
9/26	1ST	6.8	1.01	12
	2ND		5.59	11
	3RD		9.01	11
9/27	1ST		1.57	11
	2ND		11.52	11
	3RD		9.09	11
9/28	1ST	6.8	1.70	12
	2ND		11.97	12
	3RD		11.40	11
9/29	1ST	6.1	1.00	11
	2ND		7.59	11
	3RD		9.19	11
9/30	1ST	1.2	9.01	11
	2ND		11.33	11
	3RD		0.09	11

EPA PROJECT

N°600 LDS CHART (1)
(7 31-'80 ~ 8-9-'80)

II-H

TYPING GAUGE SHEET

INLET NOx (ppm) | NO₃ OUTLET NOx (ppm)

PROBLEM WITH OUTLET ANALYZER

REPLACED OUTLET PROBE WITH MONS-3

NOx REMOVAL (%) | NH₃/NOx

BOILER LOAD (MW)

FLUE GAS FLOW RATE (SCFM) | PRESSURE DROP

SO₂ TEST BY RADIAN

SO₂ CONC. (PPM) | O₂ CONC. (VOL %)

DATE (HOURS):
7/31 8:00 (1890.34) | 8/1 (1914.34) | 8/2 (1938.34) | 8/3 (1962.34) | 8/4 (1986.34) | 8/5 (2010.34) | 8/6 (2034.34) | 8/7 (2058.34) | 8/8 (2082.34) | 8/9 (2106.34) | 8/10 8:00 (2130.34)

		NO₃ SLIPPAGE		SOOT BLOW CYCLE	
DATE		TIME	PPM	TIME	°F
7/31	1ST				
	2ND				
	3RD				
8/1	1ST				
	2ND				
	3RD				
8/2	1ST				
	2ND				
	3RD				
8/3	1ST				
	2ND				
	3RD				
8/4	1ST				
	2ND				
	3RD				
8/5	1ST				
	2ND				
	3RD				
8/6	1ST				
	2ND				
	3RD				
8/7	1ST				
	2ND				
	3RD				
8/8	1ST				
	2ND				
	3RD				
8/9	1ST				
	2ND				
	3RD				

EPA PROJECT

N° 600 LOG CHART
(8-10-'80 ~ 8-19-'80)

H-12

TYPING GUIDE SHEET

DATE (HOURS)

EPA PROJECT

N°630 LOG CHART ⑤
(8-20-90 ~ 8-29-90)

H-13

TYPING GUIDE SHEET

INLET NOx (ppm)   OUTLET NOx (ppm)

NOx REMOVAL (%)   NH3/NOx

R. TEMP. (°C)   BOILER LOAD (MW)

FLUE GAS FLOW RATE SKFM   PRESSURE DROP

SO3 CONC. (PPM)   O2 CONC. (VOL%)

DATE (HOURS)

H-14

		NH3 SLIPPAGE		SPOT BLOWS CYCLE		
DATE		TIME	PPM	TIME	°F	R
8/30	1ST					
	2ND					
	3RD					
8/31	1ST					
	2ND					
	3RD					
9/1	1ST					
	2ND					
	3RD					
9/2	1ST					
	2ND					
	3RD					
9/3	1ST					
	2ND					
	3RD					
9/4	1ST					
	2ND					
	3RD					
9/5	1ST					
	2ND					
	3RD					
9/6	1ST					
	2ND					
	3RD					
9/7	1ST					
	2ND					
	3RD					
9/8	1ST					
	2ND					
	3RD					

EPA PROJECT

N°600 IDG CHART ⑱
(9-9-'80 ~ 9-18-'80)

H-15

TYPING GUIDE SHEET

PROBLEM WITH LOW TEMPERATURE

TURNED OFF FEED

H.R. TEST

DATE	NH₃ SLIPPAGE		SOOT BLOWER CYCLE		
	TIME	PPM	TIME	°F	PSIG
9/9	1ST			7:51	650	48
	2ND		50	8:32	640	48
	3RD		50		650	50
9/10	1ST			1:51	670	50
	2ND		40	9:14	640	20
	3RD		63	10.51	660	50
9/11	1ST			1:53	660	48
	2ND		92	9:29	660	20
	3RD		80	11:50	672	46
9/12	1ST			8:03	695	48
	2ND			9:59	662	50
	3RD			9.15	690	48
9/13	1ST			9.51	685	46
	2ND			10.49	665	46
	3RD			11.56	670	48
9/14	1ST			7:54	600	52
	2ND			11.11	648	46
	3RD			8:10	670	48
9/15	1ST			1.51	680	20
	2ND			5:01	680	50
	3RD			8.18		44
9/16	1ST	8.00	83	9.51	670	46
	2ND	11.13	68	15.56	650	44
	3RD			0.50	649	44
9/17	1ST	10.51	39	1.51	600	44
	2ND	5.07	93	6.10	640	48
	3RD			9.01	440	44
9/18	1ST	11.75	93	6.10	662	44
	2ND	8.00		1.03	910	66
	3RD			9.01	448	44

DATE (HOURS)

SO₂ CONC. (PPM)   O₂ CONC. (%)

EPA PROJECT

NOₓ LOG CHART ⑭
(9-19-'80 ~ 9-29-'80)

INLET NOₓ (ppm)

NOₓ REMOVAL (%)

H-16

R. TEMP. (°F)

FLUE GAS FLOW RATE (SCFM)

SO₂ CONC. (ppm)

OUTLET NOₓ (ppm)

NH₃/NOₓ

FEED LOAD (%PC)

PRESSURE DROP (in. H₂O)

O₂ CONC. (%PC)

PROBLEM WITH LOW TEMPERATURE

COMPLETE SHUT DOWN. THE FAN AND ROTARY VALVE WAS REPAIRED AND ALSO THE HEATER BUNDLE WAS REPLACED.

PROBLEM WITH FLOW RATE

EMERGENCY SHUT DOWN. MAINTENANCE WAS PERFORMED ON THE FAN.

		NH₃ SLIPPAGE		SOOT BLOWER CYCLE		
DATE		TIME	PPM	TIME	°F	FREQ
9/19	1ST	12.01	46	7.52	653	47
	2ND			8.03	665	50
	3RD			0.00	660	44
9/20	1ST			1.52	632	47
	2ND			–	–	–
	3RD			–	–	–
9/21	1ST			–	–	–
	2ND			–	–	–
	3RD			–	–	–
9/22	1ST			–	–	–
	2ND			–	–	–
	3RD			–	–	–
9/23	1ST			–	–	–
	2ND			–	–	–
	3RD			–	–	–
9/24	1ST			–	–	–
	2ND			–	–	–
	3RD			–	–	–
9/25	1ST			5.12	670	48
	2ND			8.01	630	50
	3RD			0.03	650	47
9/26	1ST			8.24	670	50
	2ND			3.97	664	92
	3RD			0.03	648	44
9/27	1ST			6.05	670	40
	2ND			11.53	672	50
	3RD			10.57	650	44
9/28	1ST			8.03	665	40
	2ND			12.21	673	50
	3RD			0.50	673	39

DATE (HOURS)   9/19 8:00   9/20   9/21   9/22   9/23   9/24   9/25   9/26   9/27   9/28   9/29 8:00
(3061.94) (3085.94)   (3120.50)   (3144.50)   (3168.50)

EPA PROJECT

No 600 LOG CHART

(9-29-'80 ~ 10-8-'80)

EPA PROJECT

N°602 LOG CHART ⑬
(10-9-'80 ～ 10-18-'80)

TYPING GUIDE SHEET

H-18

NOₓ INLET (ppm) 600 400 200 0

OUTLET NOₓ (ppm)

NOₓ REMOVAL (%) 100 90 80 70 60

NH₃/NOₓ

INITIAL SHUT DOWN BECAUSE OF
LEAKAGE HAS CAUSED IN THE #9 BOILER.
ITS SYSTEM HAS OPERATED WITH HOT AIR,
DURING ITS REPAIRING.

R. TEMP. (°F) 800 600 400

BOILER LOAD (MW) 300 200 100 0

FLUE GAS FLOW RATE (SCFM) 1600 1400 1200 1000

PRESSURE DROP

PROBLEM WITH
PRESSURE DROP.

SO₃ CONC. (ppm) 2000 1000 0

O₂ CONC. (%)

DATE (HOURS) 10/9 9:00 (3289.00) 10/10 (3313.00) 10/11 10/12 10/13 (3357.75) 10/14 (3365.75) 10/15 (3389.75) 10/16 (3411.75) 10/17 (3433.75) 10/18 (3455.75) 10/18 9:00 (3455.75)

こ TYPING GUIDE SHEET

EPA PROJECT

N° 600 LOG CHART (14)

(10-19-'90 ~ 10-29-'90)

6T-H

INLET NOx (ppm)

NOx REMOVAL (%)

R. TEMP.

FLUE GAS FLOW RATE (SCFM)

SO₂ CONC. (ppm)

DATE (HOURS)

PROBLEM WITH HEAT FOR ANALYSIS

BLOW BACK TEST NOTE
1900 SCFM

		RATIO	SOOT BLOWER CYCLE		
DATE	TIME	PPM	TIME	°F	
7/9	1ST	0:19	91	7:28		
	2ND			8:02	637	77
	3RD	4:15	77	13:13	660	52
10/20	1ST	13:27	94	2:15	670	90
	2ND			15:09	642	70
	3RD	1:13	94	17:59	690	44
10/21	1ST			8:10	599	90
	2ND			14:51	690	47
	3RD			10:02	650	80
10/22	1ST			8:00	610	90
	2ND			13:10	650	80
	3RD			23:51	660	44
10/23	1ST			1:53	600	90
	2ND			15:51	690	70
	3RD			23:17	670	43
10/24	1ST	0:52	59	8:00	660	90
	2ND		95	17:21	690	91
	3RD	1:10	94	23:27	670	92
10/25	1ST			8:45	660	90
	2ND			15:10	640	70
	3RD			23:30	660	42
10/26	1ST			9:45	660	90
	2ND			15:15	660	90
	3RD			0:02	650	62
10/27	1ST			8:00	665	92
	2ND			15:21	660	91
	3RD			23:16	670	92
10/28	1ST			8:05	690	96
	2ND	9:27	88	8:10	600	98
	3RD	1:20	50	9:10	670	68

EPA PROJECT

N°600 LOG CHART ㉒
(10-29-'80 ~ 11-7-'80)

PROBLEM WITH
OUTLET ANALYZER

FLOW RATE TEST

		NH3 SLIPPAGE		SOOT BLOWER CYCLE		
DATE		TIME	PPm	TIME	°F	PSIG
10/29	1ST			7:57	650	46
	2ND			—	—	—
	3RD			23:30	650	46
10/30	1ST			8:03	657	46
	2ND			15:40	632	50
	3RD			23:00	690	48
10/31	1ST			8:03	658	46
	2ND			7:30	660	48
	3RD			23:50	655	50
11/1	1ST			8:22	662	49
	2ND			15:30	660	70
	3RD			23:40	655	48
11/2	1ST			8:11	650	48
	2ND			15:44	650	50
	3RD			23:45	640	50
11/3	1ST			8:23	650	48
	2ND			15:44	650	50
	3RD			23:17	690	50
11/4	1ST	11:70	53	15:23	640	45
	2ND	15:34	59	15:40	640	48
	3RD			23:30	670	47
11/5	1ST			—	—	—
	2ND	06:36	61	16:14		
	3RD			23:38	650	46
11/6	1ST			7:53	650	46
	2ND			15:53	650	40
	3RD			23:56	650	47
11/7	1ST	16:30	93	7:59	698	48
	2ND	16:52	60	16:03	672	48
	3RD			23:35	688	58

DATE
(HOURS)

EPA PROJECT

N°600 LOG CHART ㉑

(11-8-'80 ~ 11-17-'80)

INLET NOx (ppm)

OUTLET NOx

NOx REMOVAL (%)

NH3/NOx (—)

NH3/NOx

MOLE RATIO TESTS

PARTIAL SHUT DOWN BECAUSE OF PROBLEM WITH GAS BURNER.

TEMP TESTS

TEMP (°F)

BOILER LOAD (% PC)

FLUE GAS FLOW RATE (KFM)

PRESSURE DROP (IN. H2O)

HOT AIR CONDITION

LESS THAN 50% PC

SO2 CONC. (ppm)

O2 CONC. (% PC)

	NH3 SLIPPAGE		SOOT BLOWER CYCLE		
DATE	TIME	ppm	TIME	°F	PSIG
11/8	1ST		7:35		
	2ND	11:17	11:11	490	42
	3RD			490	42
11/9	1ST		5:13	440	40
	2ND			490	42
	3RD			42	40
11/10	1ST		5:30	100	42
	2ND		8:34	450	44
	3RD		21.35	452	40
11/11	1ST		15:20		
	2ND				
	3RD		22.31	450	44
11/12	1ST		—	—	—
	2ND	17	—	—	—
	3RD		—	—	—
11/13	1ST	85	—	—	—
	2ND	17	—	—	—
	3RD		—	—	—
11/14	1ST		—	—	—
	2ND		—	—	—
	3RD		—	—	—
11/15	1ST		—	—	—
	2ND		—	—	—
	3RD		3.30	495	44
11/16	1ST		9.35	490	50
	2ND		15.12	490	56
	3RD		2.35	452	42
11/17	1ST		3.05	440	44
	2ND		15.35		
	3RD			450	

DATE (HOURS)

INLET NOx (ppm) — NO. NOx

OUTLET NOx (ppm)

600
400
200
0

MAINTENANCE
(ANALYZER & WATER LEVEL)

NOx REMOVAL (%)

100
90
80
70
60

PARTIAL SHUT DOWN
BECAUSE ... OF BOILER GAS
TROUBLES

COMPLETE SHUT DOWN
FOR TH. TROUBLE ACTION
PRIMARY

NH3/NOx (—)

NH3 LINE REPAIRED
WITH HEATING PIPE.

SET FLOW WITH
EACH LINE.

NH3/NOx

TEMP. TEST

NOx REMOVAL TEST

TEMP. (°F)

800
600
400

BOILER LOAD (MW)

H-22

FLUE GAS FLOW RATE

1600
1400
1200
1000

NORMAL OPERATION

PRESSURE DROP

LOAD THROUGH BY-PASS.

SO2 CONC. (ppm)

2000
1000
0

O2 CONC. (vol %)

DATE (HOURS)

11/18 9:00 (4158.17)   11/19 (4182.17)   11/20 (420 17)   11/21 (4230 17)   11/22   11/23   11/24   11/25 (4262.67)   11/26 (4286.69)   11/27   11/28 8:00

DATE		NH3 SLIPPAGE		SOOT CYCLE	
		TIME	PPm	TIME	
11/18	1ST	10:45	70	9:00	
	2ND	01:30	72	13:90	
	3RD	20:13	26	9:23	
11/19	1ST	11:23	26	7:31	
	2ND			15:03	
	3RD			17:14	
11/20	1ST			1:51	
	2ND	11:53	23	1:15	
	3RD	21:30	23	17:34	
11/21	1ST	11:30	21	1:31	
	2ND	16:13	27	15:18	
	3RD	21:13	22	21:33	
11/22	1ST			1:54	
	2ND			—	
	3RD			17:03	
11/23	1ST			1:31	
	2ND			14:31	
	3RD			17:71	
11/24	1ST			7:19	
	2ND	2:50	23	9:50	
	3RD			9:21	
11/25	1ST	10:53	22	1:03	
	2ND	16:10	23	9:15	
	3RD			17:73	
11/26	1ST			7:76	
	1ST			15:10	
	3RD				
11/27	1ST			7:05X9	
	2ND			9:01	
	3RD				

EPA PROJECT

N° 600 10% CHART
(11-28-'80 ~ 12-9-'80)

FOR OUTLET SAMPLE PROBE
AND LINE CLOGGED.

PROBLEM OF
OUTLET SAMPLING SYSTEM

PROBLEM OF NH₃ INJ. RATE

SO₃ REMOVAL
TEST

EMERGENCY SHUT OFF OF
NH₃ FEED (TEST "5-B")

POWER PLANT LOAD EXCURSIONS (TEST "5-2")

H-24

TYPING GUIDE SHEET

DATE
(HOURS)

	600 SLIP RATE		OUTLET GAS CYCLE	
DATE	TIME	PPM	TIME	°F
12/8	1ST			
	2ND			
	3RD			
12/9	1ST			
	2ND			
	3RD			
12/10	1ST			
	2ND			
	3RD			
12/11	1ST			
	2ND			
	3RD			
12/12	1ST			
	2ND			
	3RD			
12/13	1ST			
	2ND			
	3RD			
12/14	1ST			
	2ND			
	3RD			
12/15	1ST			
	2ND			
	3RD			
12/16	1ST			
	2ND			
	3RD			
12/19	1ST			
	2ND			
	3RD			

PARTIAL SHUT DOWN

H-25

		FEED SLIPPAGE		SOOT BLOW CYCLE	
DATE		TIME	PPM	TIME	°F
12/19	1ST		39	7:15	441
	2ND	13.98	61	3:52	444
	3RD				
12/19	1ST				450
	2ND				
	3RD				
12/20	1ST				
	2ND				
	3RD				
12/21	1ST				
	2ND				
	3RD				
12/22	1ST				
	2ND				
	3RD				
12/23	1ST				
	2ND				
	3RD				
12/24	1ST				
	2ND				
	3RD				
12/25	1ST				
	2ND				
	3RD				
12/26	1ST				
	2ND				
	3RD				
12/29	1ST				
	2ND				
	3RD				

DATE
(HOURS)

12/18 8:00   12/19   12/20   12/21   12/22   12/23   12/26   12/27   12/28 8:00
(4751.84) (4153.84) (4157.42)   (4017.05) (4039.05)   (4063.23)

TYPING GUIDE SHEET

EPA PROJECT

№600 IDS CHART
(12-28-'80 ~ 1-6-'81)

INLET NOₓ (ppm)

NOₓ REMOVAL (%)

H-26

FLUE GAS FLOW RATE SCFM

SO₂ CONC. (ppm)

NO₃ OUTLET NOₓ (ppm)

NH₃/NOₓ

BOILER LOAD (%PC)

FEED PRESSURE DROP (in. H₂O)

O₂ CONC. (%PC) (VOL %)

		N₂ PURGE		SOOT BLOWER CYCLE		
DATE		TMR	PPₒₐ	TMR	°F	PH
12/28	1ST					
	2ND					
	3RD					
12/29	1ST					
	2ND					
	3RD					
12/30	1ST					
	2ND					
	3RD					
12/31	1ST					
	2ND					
	3RD					
1/1	1ST					
	2ND					
	3RD					
1/2	1ST					
	2ND					
	3RD					
1/3	1ST					
	2ND					
	3RD					
1/4	1ST					
	2ND					
	3RD					
1/5	1ST					
	2ND					
	3RD					
1/6	1ST					
	2ND					
	3RD					

DATE (HOURS)

12/28 Sun   12/29   12/30   12/31   1/1   1/2   1/3   1/4   1/5   1/6   1/7 Sun

EPA PROJECT

N°608 LOG CHART (2)

(1-7-'81 ~ 1-16-'81)

EPA   PROJECT

№ 600 LOG CHART ㉔
(1-17-'81 ~ 1-26-'81)

H-28

INLET NOx (PPM)

OUTLET NOx (PPM)

REMOVAL % (NO_x)

NH3/NOx (—)

BOILER LOAD (GPC)

FLUE GAS FLOW RATE (SCFM)

PRESSURE DROP

SO_2 CONC. (PPM)

O_2 CONC. (GPC)

SHUT DOWN FOR CATALYST WASHING

CATALYST DRYING WITH HOT AIR OPERATION

PROBLEM WITH PRESSURE GAGE

ΔP TRANSMITTER

MR TODA

DATE (HOURS)	1/17 8:00 (5257.92)	1/18 (5281.92)	1/19 (5305.92)	1/20 (5329.92)	1/21 (5353.92)	1/22 (5377.92)	1/23 (5401.92)	1/24 (5425.92)	1/25 (5449.92)	1/26 (5473.92)	1/27 8:00

DATE		NH3 SLIPPAGE		SOOT BLOW CYCLE		
		TIME	PPM	TIME	°F	PSH
1/17	1ST			9.00	44	28
	2ND			15:17	31	30
	3RD			23:55	26	32
1/18	1ST			8:00		34
	2ND			15:50	41	32
	3RD			23:36	42	34
1/19	1ST			8:12	34	35
	2ND			15:48		36
	3RD			23:15	42	36
1/20	1ST			8:15	49	38
	2ND	11:00	26	15:58		58
	3RD			23:38	40	58
1/21	1ST	14:30	36	8:10	36	42
	2ND	16:00	29	15:42		40
	3RD			23:58	46	32
1/22	1ST	16:40	38	8:10	43	42
	2ND	22:00	48	16:17	44	42
	3RD			0:15	44	28
1/23	1ST	8:30	41	8:00	39	40
	2ND	21:13	44	15:37	42	42
	3RD			23:57	42	36
1/24	1ST			8:20	28	30
	2ND			15:44	31	40
	3RD			23:35	42	42
1/25	1ST			8:13	32	42
	2ND			15:57	32	32
	3RD			23:38	44	40
1/26	1ST			8:00		
	2ND			—	—	—
	3RD			—	—	—

EPA PROJECT

N°600 LOG CHART
(1-27-'81 ~ 2-1-'81)

INLET NOx RB?(] (ppm) 600 400 (ppm) 200 0

RB?[] OUTLET NOx (ppm)

NOx REMOVAL (%) 100 90 80 70 60

NH₃/NOx (—)

NH₃/NOx

M.R. TEST

TEMP. 600 400

BOILER LOAD (°F C) (MW)

FLUE GAS FLOW RATE 2000 1500 1000 500 0 SCFM

FLOW RATE TEST

REACTOR PRESSURE DROP (mmAq in. H₂)

SO₃ CONC. 1000 (ppm)

O₂ CONC. (VOL %)

H-29

		IN-G SURFACE		SOOT BLOWER CYCLE		
DATE		TIME	FLO₂	TIME	°F	PSIG
1/27	1ST					
	2ND				04	47
	9TH				60	36
1/28	1ST	8:00	64	8:10	50	48
	2ND	11:08	65	13:53	85	40
	3RD	16:50	23	17:18	77	42
1/29	1ST	9:30	23	8:09		41
	2ND			16:21	89	42
	3RD	11:02	31	21:31	58	42
1/30	1ST	16:10	0	11:50		52
	2ND			13:43		48
	3ST			21:40	58	48
1/31	1ST	11:10	19	8:20	57	43
	2ND	16:55	21	15:55	62	43
	3RD	21:12	63	20:56	62	48
2/1	1ST	9:93	70	8:20	44	42
	2ND	9:55	74	14:15	60	22
	3RD	23:30		3:15	74	33

DATE 1/27 8:00 1/28 1/29 1/30 1/31 2/1 2/2 2/3
(HOURS) (5497.24) (5521.24) (5545.24) (5569.24) (5593.24) (5617.24)

END

DATE
FILMED
11-30-82

NTIS