REQUIRED DEVELOPMENT AND INSTALLATION OF NO_x ABATEMENT PROCESS AT THE IDAHO CHEMICAL PROCESSING PLANT

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ABSTRACT

Waste management operations associated with the reprocessing of nuclear fuel at the Idaho Chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory (INEL) result in the release of NO_x to the environment. Although these releases are within existing guidelines, the NO_x causes the off-gas to be highly visible. An increase in NO_x emissions will occur around 1995 when the fuel reprocessing rate at the ICPP increases. The State of Idaho, in a Permit to Construct for the new Fuel Processing Restoration Facility, has required that an NO_x abatement system be installed.

Because the NO_x offgas stream at the ICPP differs greatly from other industrial NO_x streams, testing will be required before an abatement process is constructed. The selective catalytic reduction (SCR) process, where ammonia is injected into the stream to reduce the NO_x in the presence of a catalyst, has been selected for pilot plant development. The pilot plant will be used to verify the proposed process, obtain design data, and determine process removal efficiency. Based on the pilot plant results, a plant-scale facility will be constructed to treat the off-gas that is released.

INTRODUCTION

The Idaho Chemical Processing Plant (ICPP) is a government-owned, contractor-operated facility for reprocessing nuclear fuel and for managing the resulting radioactive waste, located at the Idaho National Engineering Laboratory (INEL) in south-eastern Idaho. During the waste management operations, a highly visible plume of colored gas containing oxides of nitrogen (NO_x) is released to the environment. The NO_x concentrations have been shown by actual measurements, as well as by diffusional modeling, to be well below existing guidelines and limits.

A new facility at the ICPP is being constructed which will increase the fuel reprocessing rate and in turn increase the quantity of waste processed at the ICPP. Processing this waste is projected to increase NO_x releases by more than 40 tons/year. Consequently, in compliance with State regulations, a Permit to Construct was obtained from the State of Idaho (1,2,3). The State permit specifies NO_x release limits on an hourly and annual basis for the ICPP and also requires installation of NO_x abatement equipment to treat the main ICPP stack off gas-stream. It allows for construction and operation of a pilot plant to test the chosen abatement system and to obtain the information required for design of a plant-scale NO_x treatment facility.

Laboratory tests have been completed, a 1/40-scale pilot plant is now in operation, and conceptual design of the full-scale facility is underway. Conclusions based on laboratory tests and the experimental goals for the pilot plant are included in this paper.

DESCRIPTION OF THE ICPP

Facility Location

The ICPP operated by Westinghouse Idaho Nuclear Company, Inc. (WINCO) is located in southeastern Idaho, about 48 miles west of Idaho Falls. The nearest permanent residents are located about 11 miles from the ICPP at Atomic City. US Highway 26, a public roadway, traverses the INEL at a distance of about 4 miles from the ICPP, and Craters of the Moon National Monument, a Class I air attainment area, is located about 26 miles to the south west.

Process Description

Nuclear fuel from INEL test reactors and from the Naval propulsion program is received at the ICPP and stored underwater in the Fluorinel and Storage (FAST) fuel storage basins. During processing, the entire fuel element is dissolved in acid, and the unused uranium is separated from the fission products and cladding material through a liquid/liquid extraction process. The liquid waste material, including dissolved fission products and cladding material, is stored underground in (300,000 gal) stainless steel tanks. In the New Waste Calcining Facility (NWCF), the liquid waste is sprayed into a hot (500oC) fluidized bed calciner, and the liquid evaporates, leaving the solids adhering to the bed particles. The solid calcine is then pneumatically transported to large bins inside concrete vaults for interim storage.

SOURCES AND IMPACTS OF NOx AT THE ICPP

Sources

The calcination in the NWCF is the major NO_x source at the ICPP. The waste contains nitric acid from the fuel dissolution process, and calcium nitrate, added to minimize the fluoride volatility in the calciner. When the waste is sprayed into the hot calciner, the nitrates decompose, and NO_x is formed. After extensive treatment in the NWCF offgas cleanup system, NO_x, water vapor, and other gases are released to the ICPP stack. At full design throughput, the NWCF can release as much as 388 lb/hr of NO_x. This corresponds to 2000 scfm of a 3% NO_x stream with a dew point of 70°C.

Impacts of NO_x Releases

The NO_x released from the ICPP consists primarily of NO_2, a reddish-brown gas which creates a colored plume during NWCF operations. Under certain conditions, this plume is visible from the public highway traversing the INEL. Measurements have shown that the concentration of

NO_x at the highway is well below EPA limits, typically 2 to 6 $\mu g/m^3$ compared to EPA ambient air standards of 100 $\mu g/m^3$. In addition, the ICPP is located about 26 miles from the Craters of the Moon National Monument. This monument is a Class I area as designated by the Clean Air Act, and as such no degradation of the air quality is permitted. Even though modeling studies(3) have shown no decrease in visibility at the National Monument, there is some concern that the NO_x emissions could impact the quality of the air at that location.

Future Impacts

A new ICPP facility, the Fuel Processing Restoration (FPR) Facility, will replace several of the processes presently being conducted in the existing CPP-601 building, including uranium separation, uranium conversion to a solid, and low-level waste cleanup. The ICPP throughput will also be increased when the FPR becomes operational in FY-1995, increasing the amount of high-level waste sent to the NWCF. This, in turn, will increase the quantity of NO_x released to the atmosphere by more than 40 tons/yr. Consequently, a State of Idaho Permit to Construct was required before significant construction of the FPR could begin.

Based on past NWCF operations, the Permit to Construct set limits for both hourly and annual NO_x releases of 388 lbs/hr and 1700 tons/year. In addition, the State required construction of an NO_x abatement facility. Either a thermal decomposition or selective catalytic reduction process was identified as acceptable by the State, using previous scoping studies conducted by and for WINCO. Although no post-abatement limits were set by the State, design goals of 90% for the thermal decomposition and 99% for the SCR process were identified in the permit. Authority was also granted to construct and operate a pilot plant to obtain NO_x abatement data prior to construction of the full-scale facility. The actual efficiency required for the abatement process will be determined through pilot plant testing and emission testing during operation of the full-scale facility.

NO_x LABORATORY AND PILOT PLANT ACTIVITIES

The offgas stream from the NWCF varies greatly from typical NO_x sources such as those from power-generating plants or nitric acid plants. About 90% of the NWCF NO_x is NO_2, only 10% NO, a complete reversal of the normal distribution. The oxygen and moisture contents are also quite high, about 14% and 25% respectively. Also, the NO_x concentration in the NWCF offgas may range as high as 30,000 parts per million (ppm), much higher than the 500-1000 ppm normally encountered in power plants. Though the offgas is thoroughly filtered before release, gaseous impurities, such as iodine, mercury, fluorides, and chlorides, are present in the offgas and may impact the abatement process. Consequently, any abatement system used at the ICPP will be an extension of existing technology. The uncertainties involved in the process, and the large cost associated with constructing the main facility, make pilot plant verification essential before a plant-scale facility is constructed.

Potential NO_x Abatement Processes

Various methods of NO_x abatement have been used in industry, and these methods have been reviewed and studied both by WINCO and other outside contractors at WINCO's request. The methods fall into the following general groups:

1. destruction of nitrates before calcination
2. wet scrubbing
3. sorption on solid media
4. thermal destruction without a catalyst
5. thermal destruction with a catalyst
6. newly proposed methods such as plasma arc destruction, electron beam reaction, etc.
7. selective catalytic reduction

The advantages and disadvantages of each of these methods, as they apply to the ICPP NWCF offgas stream, have been documented previously (4,5,6). The unique offgas chemistry described above, and the prerequisite that no additional waste streams be generated, lead to the conclusion that the selective catalytic reduction (SCR) process would best serve the needs of the ICPP.

Description of the SCR Process

The SCR process has been successfully tested on more conventional gas streams throughout the world. It is the primary process now in use for cleaning the exhaust from stationary power plants in Japan and Europe. The process destroys the NO_x by reducing it with ammonia in the presence of a catalyst selective to the reduction of NO_x over other oxidants (even in an oxygen environment). Various catalysts can be used, based on titanium oxide, vanadium oxide, copper oxide, and silver. A Norton Company proprietary zeolite (NC-300) will be used as the catalyst for the ICPP pilot plant tests. An earlier version of this catalyst (Zeolon 900) was used previously in laboratory tests at the ICPP and worked well at concentrations up to 15,000 ppm (7).

Laboratory Studies

A two-step development program is in progress, consisting of laboratory testing which began in FY-1988, followed by a pilot plant testing program which began early in FY-1989.

The laboratory tests have measured:

1. pressure drop through a catalytic bed as a function of flow rate, bed depth, catalyst geometry, and catalyst size.
2. bulk gas and catalyst surface temperatures at various conditions.
3. reaction efficiencies as a function of space- and face velocity.

The laboratory tests were conducted using pure NO_2 in air rather than actual NWCF process offgases. Test

parameters were chosen to investigate effects on catalyst surface temperature, bulk gas temperature, and reactor temperature profile as the gas face velocity, residence time, and bed depth were varied. The initial tests have been completed (8,9), indicating the following operating envelope for the pilot plant:

Face Velocity:	5-7 fps as measured at reactor conditions
Inlet Temperature:	240-350°C
NH_3/NO_2 Ratio:	0.75:1 to 1.3:1
Bed Depth:	10 to 18"
Catalyst:	Norton Company's NC-300 Raschig Rings

Pilot Plant Design and Description

The pilot plant design (10,11) is based on laboratory tests and modifications identified since the completion of the initial laboratory tests. Major requirements of the design were:

1. to limit the temperature of the catalyst during the exothermic NH_3/NO_x reaction to below 500°C to prevent catalyst sintering and activity loss;
2. to provide precise control of gas flow rates over a temperature range from ambient to 500°C and,
3. and to prevent deposition of ammonium nitrate within the system by assuring that surfaces are maintained above the compound's decomposition temperature, 210°C.

To meet these requirements, three fixed-bed catalytic reactors are used, with piping and valving to allow operation in any series/parallel mode (Fig. 1). Air, ammonia and NO_x bearing NWCF off gas, may be combined with the exhaust from any stage as feed to any other stage. This arrangement gives the operator complete control of the extent of reaction in each bed, thereby distributing the heat generation and controlling the catalyst temperature. Gas flow measurements are made with turbine meters on individual streams, and a pitot-tube measurement on the total flow to each reactor. Preheating the feed to Stage I, mixing of the hot effluent with cooler gases downstream, and heat tapes throughout prevents the deposition of ammonium nitrate on cold surfaces, while lowering the overall temperature by mixing the concentrated NO_x with less reactive, cooler streams. The pilot plant tests will determine which operating mode is required and the number of reactor beds needed for NO_x abatement.

A slipstream of up to 50 scfm drawn downstream of the NWCF off-gas system is HEPA filtered as it enters the pilot plant. After preheating, the NO_x bearing gas (up to 3%) can be mixed with ammonia and preheated air (if desired) before the first reactor vessel. More NWCF off-gas, air, and ammonia can be added between subsequent reactors. All flows are drawn through the system by the vaccum from a steam jet, and the entire pilot plant is contained within a ventilated enclosure to assure that any NO_x releases from the pilot plant do not reach the surrounding occupied area.

Flows, pressures, and temperatures are recorded, and samples of gas can be directed to a flow-through diode-array spectrophotometer for on-line analysis of NO, NO_2, and NH_3. Calculated values such as NH_3/NO_x mole ratios, abatement efficiencies, and face velocity in each reactor, are carefully monitored and logged by a management control system. Process modifications, and alarms are continuously recorded.

Pilot Plant Investigations

The pilot plant will provide design-basis information for the full-scale NO_x abatement facility. Specific objectives of the test program are to:

1. Determine the effects that the high water content (20-30%) of the NWCF off-gas has on the SCR process in terms of NO_x reduction efficiency, required NO_x reaction temperature, reactor temperature rise and catalyst surface temperature.
2. Verify the catalyst life in the presence of minor impurities (F^-, Cl^-, heavy metals, radionuclides) existing in the NWCF off-gas.
3. Verify lab-scale test results in regard to: 1) the magnitude and location of the maximum catalyst surface temperature, 2) the recommended face velocity, 3) the recommended bed depth, and 4) the recommended initial gas inlet temperature.
4. Determine the number of reactors required to maximize NO_x reduction under the design constraints (i.e, a temperature range of 230 to 500°C).
5. Determine the flow scheme necessary to obtain optimum operation.
6. Determine the need for: 1) dilution air, 2) recycle and 3) parallel and/or series operation.
7. Determine the NH_3:NO_x ratio required to achieve maximum NO_x abatement (with minimum NH_3 discharge) for various operating modes.
8. Demonstrate control of the process and data acquisition through the use of a microprocessor based computer system.
9. Verify the reliability, accuracy and precision of the on-line, "real time", sampling techniques and analytical instrumentation.
10. Determine the optimal layering of various catalyst shapes/sizes.

FULL-SCALE NO_x ABATEMENT FACILITY

Conceptual Design

Conceptual design was done by Fluor Technology, Inc. during FY-1988. The cost estimate and design information from the Conceptual Design Report (12), was used to

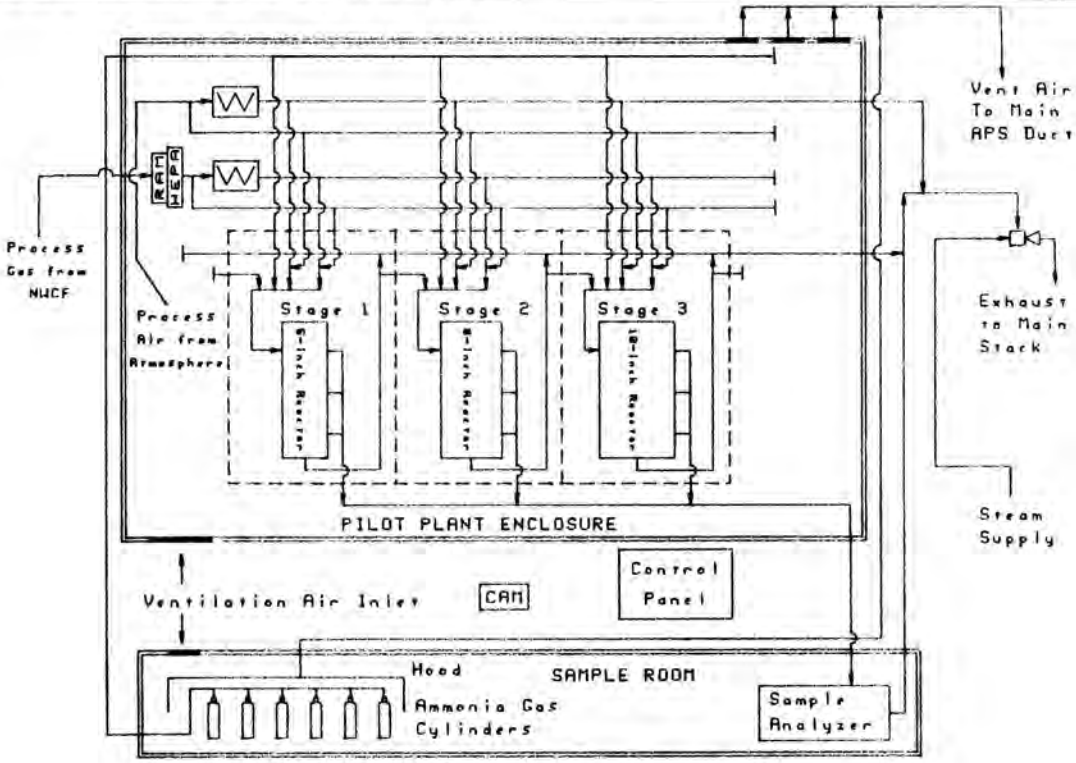

Fig. 1. NO_x Abatement Pilot Plant.

prepare the Congressional Project data sheet for DOE earlier this year.

Facility Description

The conceptual design places the new treatment facility (111' x 46') next to the NWCF, directly over the existing off-gas line to the main stack. The new facility will be self contained, equipped with heating and ventilation, fire protection, waste systems, and necessary utilities. Process equipment will include prefilters, preheaters, the reactor vessel or vessels containing the catalyst, off-gas blowers, and catalyst handling and decontamination systems. The ammonia storage tanks, two tanks containing 18,000 gallons each, will be sited away from any normally occupied areas.

No additional effluents which have the potential of exceeding existing regulations will be created in the facility. The process system will be capable of continuous operation with minimum downtime. It will be capable of reliable startup and operation on an intermittent basis or after a prolonged shutdown.

SCHEDULE AND STATUS

The present schedule is based on having the NO_x abatement facility operable when the FPR facility is completed in 1995. The schedule is complicated by the fact that the pilot plant can effectively operate only during periods of NWCF operation. Furthermore, the government funding cycle requires that certain funds be requested at specific times.

The pilot plant design work began early in FY-1988. The design, including specifications for the instrumentation and samplers, required about six months. Installation of the pilot plant was completed in November. After SO Testing, the pilot plant began integrated operation with the NWCF on November 28. On the project side, the Schedule 44 Project Data Sheet was submitted earlier this year. Conceptual design began in FY-1988, and will continue, incorporating the pilot plant data, late in FY-1989. Advanced conceptual design will be completed in FY-1990. Title design will begin in FY-1991 and be completed in 1992, with construction beginning in FY-1992 and continuing through FY-1994.

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