AF

No_x Emission Abatement Technologies

R. Goles

October 1991

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes **any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.** Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE MEMORIAL INSTITUTE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831;
prices available from (615) 576-8401. FTS 626-8401.

Available to the public from the National Technical Information Service,
U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161.

NO_x EMISSION ABATEMENT TECHNOLOGIES

R. Goles

October 1991

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY

The Hanford Waste Vitrification Plant (HWVP) will convert Hanford Site high-level liquid defense waste to a solid vitrified (glass) form suitable for final disposal in a geological repository. Future process flow sheet developments may establish a need for a NO_x scrubber in the melter off-gas system. Consequently, a technology review has been conducted to identify and compare applicable off-gas processing alternatives should NO_x emission abatement be required.

Denitrification processes can be separated into two distinct categories, wet or dry, depending upon whether or not NO_x is absorbed into an aqueous solution. The dry methods of removal are generally more efficient (>90%) than wet scrubbing approaches (>60%); however, most dry approaches are applicable only to NO_x . Of the dry removal methods, selective catalytic reduction (SCR) using NH_3 reductant and a hydrogen zeolite catalyst appears to be the most suitable technology for reducing HWVP NO_x emissions should emission abatement be required. SCR is a relatively simple, well established technology that produces no secondary waste stream and is applicable to a wide range of NO_x concentrations (500 to 30,000 ppm). This technology has been successfully applied to uranium dissolver exhaust streams and has, more recently, been tested and evaluated as the best available control technology for reducing NO_x emissions at the Idaho National Engineering Laboratory's waste calciner facility, and at DOE's West Valley Demonstration Project.

Unlike dry NO_x scrubbing methods, the wet techniques are not specific to NO_x , so they may support the process in more than one way. This is the only major advantage associated with wet technologies. Their disadvantages are that they are not highly efficient at low NO_x concentrations, they produce a secondary waste stream, and they may require complex chemical support to reduce equipment size. Wet scrubbing of HWVP process NO_x emissions is an option that is justifiable only if the technology is needed to eliminate other process emissions and scrubbing compatibility can be established.

CONTENTS

SUMMARY	iii
INTRODUCTION	1
NO _x CONTROL TECHNOLOGIES	3
WET TREATMENT METHODS	3
DRY TREATMENT METHODS	5
Selective Catalytic Reduction	6
Nonselective Catalytic Reduction	8
Selective Noncatalytic Reduction	8
Adsorption	9
Radiolytic Destruction	9
COMPARISON OF WET AND DRY EFFLUENT ABATEMENT TECHNOLOGIES	13
CONCLUSIONS	17
REFERENCES	19

INTRODUCTION

In the liquid-fed ceramic melter (LFCM) vitrification process, all feed component nitrogen is lost to the process exhaust (Goles and Nakaoka 1990). Since Hanford Waste Vitrification Plant (HWVP) feed component nitrogen will be almost entirely composed of NO_3^- and NO_2^- anions, process emissions of nitrogen will be primarily composed of the gaseous effluents NO_2 and NO , referred to in this paper as NO_x . The relative proportion to which each of the NO_x components is produced depends on the process oxidizing conditions. Without organic feed reductants, NO_2 emissions will predominate. As the carbon loading of LFCM feeds increases, the importance of NO emissions will also increase. For the HWVP reference feed composition, a low average NO_x oxidation state (high NO/NO_2) would be predicted. This prediction has been confirmed during cold simulation tests of the HWVP process where, nominally, 80% of the NO_x evolved was found to be NO .

Although NO is not a noxious environmental pollutant, it is quickly converted to NO_2 , which is. As a result, process emission limits for nitrogen oxides are based upon the magnitude of NO_x concentrations independent of composition. For HWVP, NO_x emissions may be high enough, depending upon the nitrogen feed loading, to require implementation of NO_x emission control technology. This report summarizes and compares applicable NO_x emission abatement technologies so that alternatives can be evaluated should NO_x emission control be required for the HWVP.

NO_x CONTROL TECHNOLOGIES

Although plant emission limits are independent of NO/NO₂ ratios, the average oxidation state of NO_x is quite important in evaluating technical alternatives for emission abatement. Nitrogen dioxide is much more reactive with an aqueous media than is NO; consequently, wet scrubbing strategies are strongly dependent upon the characteristics of the NO_x source term. Dry NO_x abatement technologies, on the other hand, are much less sensitive to NO/NO₂ ratios but are limited by a variety of other processing conditions. Since the wet- and dry-based technologies are so dissimilar, each will be discussed separately.

WET TREATMENT METHODS

Conventional aqueous NO_x scrubbing techniques are limited by low concentrations, high NO/NO₂ ratios, and by the fundamental chemistry governing NO₂ absorption (Counce 1984). All three limitations apply to HWVP. For an atmospheric-pressure nonrecycle water scrubber, maximum expected removal efficiency for NO₂ at low concentrations (≤ 1000 ppm) has been projected to be 60% (Counce 1984). This NO_x scrubbing efficiency (NO₂/NO_x=1) would be dramatically reduced if significant NO was present (as is the case for HWVP) or if the scrubbing liquor was recycled. The limitations to conventional wet scrubbing efficiencies can, however, be overcome to some extent through physical and chemical process design modification.

For a fixed scrubber design, NO_x scrubbing efficiency increases with rising effluent concentration. Therefore, process exhaust recycle to off-gas film coolers would act to enhance wet NO_x scrubbing efficiency if the column was located downstream of the recycle extraction point. Since recycle would only increase NO_x concentrations by a factor of ~2, scrubbing efficiency would not be significantly improved by this off-gas system modification alone.

Combining adsorption methods with wet scrubbing techniques is yet another way of overcoming concentration limitations to scrubbing. Since absorption is considered a dry removal process, it is discussed in more detail in the following section.

To overcome the NO limitation to wet NO_x scrubbing efficiencies, NO can be converted to NO₂. The oxidation-absorption technique (Fausett 1977) is a method by which NO is converted to NO₂ in the process exhaust prior to liquid contacting. Ozone (O₃) and chlorine dioxide (ClO₂) are common oxidants that have been successfully used for this purpose. The absorption-oxidation method (Fausett 1977) is similar to the previous technique, but liquid-phase reactions between an aqueous oxidant (H₂O₂, MnO₄⁻) and NO_(aq) are used. Because of low NO solubility, much longer gas-liquid contact times are required for this latter technique. Consequently, large absorbers with high liquid-to-gas ratios (L/G=15 l/m³) and low superficial velocities (1 to 3 m/s) are required to support it. NO_x removal efficiencies of 85 to 90% have been demonstrated by both of these chemically modified scrubbing techniques.

The basic chemistry governing aqueous NO_x absorption can also be modified by adding complexing agents to the scrubbing liquor that catalyze the NO_x-H₂O absorption process. Ferrous ethylene diamine tetraacetic acid (Fe²⁺EDTA) has been successfully used to increase scrubbing efficiencies for both NO and NO₂ without the use of expensive oxidants (Fausett 1977). Like the absorption-oxidation method, this technique requires an absorber with a large L/G ratio and low superficial gas velocities if NO is to be directly sorbed. However, prior gas-phase oxidation can significantly reduce the required size of the liquid-gas contactor. This oxidation-complexation technique for NO_x removal has been evaluated in fossil fuel power plant flue gas applications, with demonstrated scrubbing efficiencies of ~90% (Fausett 1977).

From the above discussion, it is apparent that wet scrubbing techniques can be effectively utilized in reducing NO_x emissions. These methods have been successfully applied to combustion gases that exhibit NO_x compositions (500 ppm) similar to those projected for the HWVP process. With proper design of the NO_x absorber and application of chemical methods to enhance absorption kinetics, wet scrubbing techniques could be used to reduce HWVP NO_x process effluent by 60-90% should emission abatement become necessary. A comparison of wet scrubbing approaches is presented in Table 1.

TABLE 1. Characteristics of NO_x Technologies

<u>Abatement Technology</u>	<u>Advantages</u>	<u>Disadvantages</u>
<u>Wet Treatment</u>	Multi-purpose, simple, developed	Waste generator, inefficient, cost
Water Scrubber	Simple	Extremely large equipment required
Oxidation-Absorption	Decreased equipment size requirements, more efficient	Gas-phase oxidants required, increased waste problems, more complex
Absorption-Oxidation	Increased scrub efficiency	Extremely large equipment required, increased waste, expensive oxidants
Oxidation-Complexation	High efficiency, reduced equipment size	Increased waste, costly oxidants, complex process
<u>Dry Reducing Methods</u>	High efficiency, simple, no waste, well developed	Requires reductant, aerosol sensitive, high temperatures, NO _x specific
SCR	Well established, efficient, safe	Requires NH ₃ , requires temperature control, NH ₃ and N ₂ O potential effluents
NCR	Efficient	High fuel use, costly, O ₂ depleted streams only
SNR	Extremely simple	Low efficiency, not well developed, very high temp, tight temp control required
<u>Other Dry Methods</u>		
Adsorption	Efficient	Large equipment required, high energy, complex, waste generator
Radiolytic Destruction	Simple, no service required, no aqueous wastes, remote, compatible with existing equipment	Extremely large radioactive source required, may require chemical additives

DRY TREATMENT METHODS

As the name implies, dry NO_x treatment methods do not require aqueous scrubbing support and do not generate a condensed-phase waste stream. Indeed, the main virtue associated with most dry technologies is that they eliminate rather than transform the pollution problem. For a variety of reasons, including simplicity, economy, and compatibility, dry denitrification methods have been extensively developed for the purpose of supporting effluent control at fossil fuel power plants. This is especially true in Japan. Essentially, all dry denitrification processes can be placed into the following five categories:

- selective catalytic reduction (SCR)
- nonselective catalytic reduction (NCR)
- selective noncatalytic reduction (SNR)

- adsorption
- radiolytic destruction.

The advantages of the NO_x abatement methods associated with each of these groups will now be discussed as they apply to the HWVP processing needs.

Selective Catalytic Reduction (SCR)

This technology utilizes NH_3 and an appropriate catalyst to selectively reduce NO_x to N_2 . In addition to this primary task, the catalyst must also be capable of minimizing competing reactions that can produce undesirable effluent such as N_2O . A variety of catalysts have been used to support SCR (Kiovisky et al. 1980); most of these catalysts are quite efficient (>90%) and essentially all are proprietary. Metal-based catalysts (noble and non-noble) have been used extensively in the denitrification of oxygen depleted flue gases. However, these catalysts generally have quite narrow operating temperature ranges that are limited at the low end by NH_4NO_3 formation and limited at the upper end by competing oxygen reactions that consume reductant and further increase reactor temperatures (Thomas and Munger 1978). Nevertheless, several of these metal based SCR systems have been examined in nuclear process applications involving high- O_2 (20%) process stream conditions. Bruggeman et al. (1981) evaluated $\text{Fe}_2\text{O}_3\text{-Cr}_2\text{O}_3\text{-C}$ and $\text{Pd-Al}_2\text{O}_3$ as SCR catalysts under simulated reprocessing conditions. Both catalysts lacked required efficiency. Titanium, which also has been evaluated under oxidizing conditions, has been found to be an effective but difficult-to-use SCR catalyst that can produce significant quantities of N_2O effluent (Hattori 1984).

Natural and synthetic zeolites have also been evaluated as potential SCR catalysts. Sodium zeolites have been satisfactorily used for NO_2 reduction but were found to be ineffective for NO . A platinum zeolite has been found to actually catalyze the conversion of NH_3 to NO_x , making it totally unsuitable as a NH_3 -based SCR catalyst. Hydrogen zeolite, on the other hand, has enjoyed great success as an SCR catalyst in oxygen-rich ($\text{O}_2 > 10\%$) process streams. It is efficient for reducing both NO_x components, has a very wide operating temperature range (300 to 600°C), is not poisoned by sulfur or iodine compounds, is not significantly affected by high off-gas water loadings, and produces

very little N_2O by-product. In comparative tests, hydrogen zeolites have been found to meet or exceed the performance of several metal-based SCR catalysts in oxygen-rich ($O_2 > 10\%$) process streams.

The apparent effectiveness of hydrogen zeolites as an SCR catalyst has led to several pilot-scale tests designed to further evaluate the applicability of the SCR technology to NO_x emission abatement in nuclear process streams. The Idaho National Engineering Laboratory (INEL) applied this technology to a simulated waste calciner exhaust stream (Thomas and Munger 1978). Greater than 99% abatement efficiency was obtained for both NO and NO_2 in the temperature range of 340 to 440°C using a NH_3/NO_x ratio of 1.3. Using the design criteria established at INEL, the National Lead Corporation (NLCO) constructed an SCR emission abatement plant to control NO_x tail gas emissions generated by uranium dissolution and purification processes (Leist 1975). NO_x abatement efficiencies >99% were demonstrated for wet (46°C dew point) process streams containing up to 0.5% NO_x (30% NO). Exxon Nuclear also applied the hydrogen zeolite SCR technology to its uranium dissolver off-gas stream (2.5% NO_x , 65% H_2O) and obtained NO_x destruction efficiencies that exceeded 99% (Mays and Schwab 1975).

Although the above examples illustrate the usefulness of (hydrogen zeolite) SCR methods for dealing with high NO_x concentrations, the technique has also been successfully applied to process streams with low (500 ppm) NO_x concentrations. Thomas examined the suitability of hydrogen zeolite as an SCR catalyst for removal of NO_x in simulated fossil fuel plant exhausts (500 ppm NO , 3000 ppm SO_2 , 12% CO_2 , and air with a 35°C dew point) (Thomas and Munger 1978). The study showed that NO could be selectively reduced to below 7 ppm before competing reactions with SO_2 limited the reduction process; however, the potential for using this technology for simultaneous SO_2 recovery as ammonium bisulfate and sulfate was also demonstrated. Norton has recently developed and patented a natural hydrogen zeolite catalyst that is highly specific for NO_x and exhibits negligible SO_2 activity--a highly desirable property for industrial power plant operations. This catalyst is currently being successfully used to reduce power plant boiler emissions of NO_x (500 ppm) at a fossil

fuel facility in Nuremburg, Germany (private communication with S. M. Turner, Norton Chemical Process Products Division, Norton Corporation).

Selective catalytic reduction techniques for NO_x destruction have been successfully demonstrated in small-, pilot-, and large-scale applications under a variety of processing conditions. The method is highly efficient (>90%), is applicable to a wide range of NO_x (500 to 25,000 ppm) and O_2 (2 to 20%) concentrations, and it eliminates a pollution problem without creating a waste stream. Minimal temperature control constraints ($T > 250^\circ\text{C}$) can easily mitigate all safety concerns regarding the potential production of ammonium nitrate salts. Since hydrogen zeolite catalysts have a very wide operating temperature range (300 to 600°C) and are normally controlled at -400°C , maintaining catalytic reactor temperatures above 250°C is a trivial process control task. Selective catalytic reduction methods for NO_x reduction have been shown to be effective and versatile as well as safe; furthermore, this technology is compatible and directly applicable to the HWVP process should NO_x emission abatement be required.

Nonselective Catalytic Reduction (NCR)

This process utilizes a fuel or reducing gas in combination with a catalyst to promote the reduction of all oxidizing agents in the treated process exhaust stream. Since O_2 is consumed preferentially by this nonselective method, all process oxygen has to be consumed before NO_x can be reduced (Fausett 1977). Due to the exothermic nature of the combustion process, catalytic burnup limits the application of the NCR technology to oxygen depleted ($\text{O}_2 < 3\%$) process streams (Thomas and Munger 1978). This method of NO_x destruction is clearly incompatible with reference HWVP process conditions and, consequently, will not be discussed further.

Selective Noncatalytic Reduction (SNC)

This thermal denitrification process utilizes NH_3 and high temperatures (1000°C) to selectively reduce NO to N_2 . The efficiency of this technology is expected to be <70% (Muzio et al. 1978). Although SNC is simple in comparison to other wet and dry denitrification processes, its practical application can be quite difficult. This difficulty is due to the fact that SNC reduction

efficiency is highly temperature dependent. If operating conditions vary from the narrow temperature range required (which is NH_3/NO dependent), additional NO effluent will be produced or unreacted NH_3 will be discharged. SNC has been exclusively applied to oxygen-depleted ($\text{O}_2 < 5\%$) process streams (flue gases) (Fausett 1977), and thus the effect of high (~20%) oxygen concentrations upon potential competing reactions (N_2O , NO production) and upon denitrification efficiency in general has not been established. The applicability of SNC to the HWVP process is doubtful.

Adsorption

The dry adsorption process is based upon a catalytically active sorption bed that not only physically removes NO_x from the process stream but also efficiently converts the NO component to the easily (wet) scrubbed NO_2 species. Typically, two beds are utilized to support a continuous denitrification process. The beds are cyclically employed so that one bed is always being loaded while the other is being regenerated. The highly concentrated NO_2 stream produced during regeneration is usually recovered (at high efficiency) with conventional scrubbing equipment. The PuraSiv-N molecular sieve process developed by Union Carbide has been successfully applied to the nitric acid plant tail gas emissions (~3000 ppm) with reported NO_x recovery efficiencies of >90% (private communication with L. L. Fornoff, Union Carbide Corporation, Material Systems Division). Application of this process would be quite useful in overcoming concentration and chemical (NO) limitations to wet scrubbing techniques previously discussed if HNO_3 recovery is considered useful and desirable. Otherwise, the adsorption system will add significant complexities and waste handling problems to the process. This technology is compatible with the HWVP process, but it may not be economically practical since it is not a stand alone system and must be supported by a NO_x removal method which could be used to mitigate the process emission problem directly.

Radiolytic Destruction

Extensive investigations have been conducted to evaluate the effectiveness of using ionizing radiation for the destruction of NO_x and SO_2 compounds (Fausett 1977). Again, most of this work has been directed at developing more efficient and cost effective methods for eliminating flue gas pollutants

generated by fossil fuel power plants. Due to the nature of the industry for which these methods are being developed, the source of ionizing radiation used in essentially all of these evaluations is a high-energy electron beam.

The radiolytic destruction method of pollution abatement is based upon the chemical transformation of the complex pollutants into simpler innocuous compounds that can be emitted into the environment or easily removed from the process exhaust. The mechanisms responsible for these chemical transformations are quite complex and composition dependent. In general, the processes of importance in radiation chemistry include ionization, the formation of excited electronic states, the transfer of excitation energy between molecules, molecular dissociation, electron capture, neutralization, and radical reactions. The relative importance of each of these processes is dependent upon the chemical reactants present. Consequently, chemical additives can and have been used to control the composition and yield of radiolytic products.

If typical flue gases are exposed to Mrad doses of ionizing radiation, >90% of the NO_x present will be radiolytically converted to N_2 and other condensed phase compounds (e.g., HNO_3) (Helfritch and Feldman 1984). Since the nature of the condensed phase radiolytic products of NO_x are in general easily removed by conventional flue gas treatment systems, application of radiolytic destruction can eliminate the need for specialized and costly NO_x (and SO_2) emission abatement equipment. A 10,000 m^3/h pilot plant using this technology has been built by the Ebara Manufacturing Corp. to treat NO_x and SO_2 emissions generated by an iron ore sintering plant operated by Nippon Steel Corporation (Kawamura and Shui 1984). The only active effluent removal component in the system was an electrostatic precipitator. Nevertheless, demonstrated removal efficiencies of 95 and 85% were achieved for NO_x and SO_2 , respectively. Ebara, supported by the U.S. Department of Energy, is currently applying this radiolytic NO_x and SO_2 abatement technology to a fossil fuel power plant operated by Indianapolis Power and Light (Fishlock 1987).

In gases, the observed radiolytic chemical reactions are found to be independent of the type and energy of the ionizing radiation used. In addition, the magnitude of the effects in a particular system is solely determined by the total energy absorbed -- the dose. Consequently, passive radioactive

sources (^{137}Cs) can effectively replace the complex, high-energy scanning electron source used almost exclusively in flue gas denitrification investigations. The required source strength of an isotopic irradiator would, admittedly, be exceedingly high, but high-intensity ^{137}Cs sources are available at Hanford that could be readily used to support this application. In addition, radionuclide based irradiators, unlike electron beam generators, are completely compatible with HWVP's remote design. Unfortunately, the use of high intensity radioactive sources for NO_x abatement is hardly a demonstrated technology. Consequently, although radiolytic destruction is interesting, it cannot be seriously considered as a viable NO_x emission abatement alternative for the HWVP. A comparison of dry scrubbing techniques is presented in Table 1.

COMPARISON OF WET AND DRY EFFLUENT ABATEMENT TECHNOLOGIES

NO_x abatement technologies have been examined for the purpose of identifying alternative off-gas treatment methods that are applicable to the HWVP process should plant emissions need to be controlled. A summary of the process characteristics of the various scrubbing technologies is presented in Table 1. Denitrification processes can be separated into two distinct categories, wet or dry, depending upon whether or not NO_x is absorbed into an aqueous solution. Dry methods for NO_x removal are generally superior to wet technologies. The advantages (Fausett 1977) of the dry NO_x technologies over wet scrubbing techniques for low to intermediate NO_x concentrations (500 to 30,000 ppm) can be generalized as follows:

- higher NO_x removal efficiencies (>90%)
- no secondary waste stream generated (in many cases)
- simpler systems with few process requirements
- lower capital investment and operating costs
- more-extensive large-scale proven applications.

The disadvantages of dry NO_x abatement systems include

- sensitivity to off-gas aerosol loadings
- control of consumption of NH_3 reductant may be required
- high ($\sim 400^\circ\text{C}$) reactor temperatures may be required
- potential for NH_4NO_3 formation.

The first and last areas of concern can easily be mitigated by proper positioning of the catalytic bed and by controlling its temperature, respectively. The remaining disadvantages are associated with the operating conditions (costs) of some of the dry technologies themselves, which have been previously established to be advantageous over wet techniques. Thus, there is no serious impediment to the application of compatible dry NO_x removal technologies to the HWVP process.

Of the applicable dry technologies, only SCR using a hydrogen zeolite catalyst can be considered an established technology. Furthermore, pilot scale tests conducted at both the INEL and WVDP sites have demonstrated SCR to be the best available control technology for abating NO_x emissions generated by high-level waste solidification operations. Both DOE sites will implement full scale SCR abatement of NO_x emission by FY 1996.

The only other dry technology that has had limited large-scale applications compatible with the HWVP process is the radiolytic destruction method. With an isotopic irradiator, NO_x destruction can be achieved, in principle, by a passive external add-on device whose operation is transparent to the melter off-gas processing system. Unfortunately, essentially all the developmental work on radiolytic destruction of NO_x has been conducted on flue gases that are not similar to the reference HWVP process stream. Since impact of composition differences upon radiolytic products is difficult to predict, and the use of radionuclide based irradiators for NO_x destruction has not been demonstrated, without additional development, this methodology cannot seriously be considered as an abatement technology for the HWVP.

Unlike dry NO_x scrubbing methods, the wet techniques are not specific to NO_x , so they may support the process in more than one way. The major advantages to wet scrubbing methods include

- multi-purpose scrubbing (NO_x , F^- , I, etc.)
- insensitivity to off-gas aerosol loadings
- back-up quenching capabilities.

On the other hand, wet system disadvantages include

- complexity resulting from NO_x insolubility
- waste formation
- high capital and operating expenses
- only moderate NO_x removal (~70%).

Wet scrubbing of low concentrations of NO_x is justifiable only if the technology is needed to eliminate other process effluents. Expensive chemical

additives would probably be necessary to provide efficient NO_x scrubbing for reasonably sized absorption equipment. On the other hand, some scrubbing approaches (absorption-oxidation) may not be compatible with other off-gas processing objectives (I_2 retention). Since specialized chemical recovery and/or regeneration equipment or processes could also be required to support the chemical optimization of NO_x removal, the best approach to wet scrubbing (absorption-oxidation, oxidation-absorption, absorption-reduction, etc.) is difficult to predict without conducting a thorough economic and technical analysis of the process involved. For HWVP, process compatibility and economics are major factors that will influence the potential applicability of the wet scrubbing technologies if NO_x abatement is needed.

In summary, both wet and dry effluent abatement technologies have been examined for the purpose of identifying alternatives for HWVP NO_x emission control should it be required. Dry techniques are efficient processes (>90%), that have been developed and applied to process streams similar to those associated with HWVP. The wet technologies become quite complex when applied to low concentrations of NO_x but are capable, in principle, of reducing NO_x concentrations by ~70%. Process compatibility, waste stream generation, and capital equipment and operating costs are major factors that will influence the applicability of wet methods for low level (500ppm) NO_x removal.

CONCLUSIONS

NO_x abatement technologies have been examined for the purpose of identifying alternative off-gas treatment methods that are applicable to the HWVP process should plant emissions need to be controlled. Of the applicable scrubbing methods, SCR using NH_3 reductant and a hydrogen zeolite catalyst appears to be the most suitable technology for reducing HWVP process emissions of NO_x . The use of radiolytic destruction techniques provides a potentially passive method for abating HWVP NO_x process emissions; however, this technology has not been demonstrated using ionizing radiation sources that are compatible with a canyon-remote environment, e.g., ^{137}Cs . Wet scrubbing of low concentrations of NO_x can be successfully accomplished using very large columns and/or chemical additives, but use of this technology is justifiable only if wet scrubbing is needed to remove other process effluents.

REFERENCES

Bruggeman, A., L. Meynendonckx, and W. R. A. Goossens. 1981. "Elimination of NO_x by Selective Reduction With NH_3 ." Proceedings of the 15th DOE Air Cleaning Conference, Department of Energy, Washington D.C.

Counce, R. M. 1984. "The Theory and Practice of Nitrogen Oxide Absorption." Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Department of Energy, Washington, D. C.

Fausett, H. L. 1977. Technical Assessment of NO_x Removal Processes for Utility Applications. EPRI AF-568, Electric Power Research Institute, Palo Alto, California.

Fishlock, D. 1987. "IAEA's Acid Rain Solution: Radiation." New Technology Week 1(14):8.

Goles, R. W., and R. K. Nakaoka. 1990. Hanford Waste Vitrification Program Pilot-Scale Ceramic Melter Test 23. PNL-7142, Pacific Northwest Laboratory, Richland, Washington.

Hattori, S., Y. Kobayashi, Y. Katoh, Y. Takimoto, and M. Kunikata. 1984. " NO_x Removal from Nuclear Fuel Reprocessing Plants Off Gas by Catalytic Reduction with NH_3 ." Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Department of Energy, Washington, D.C.

Helfritch, D. J., and P. L. Feldman. 1984. "Flue Gas SO_2/NO_x Control By Combination of Dry Scrubber and Electron Beam." Radiat. Phys. Chem. 24:129-143.

Kawamura, K., and V. H. Shui. 1984. "Pilot Plant Experiences in Electron-beam Treatment Of Iron-Ore Sintering Flue Gas and Its Application To Coal Boiler Flue Gas Cleanup." Radiat. Phys. Chem. 24:129-143.

Kiovisky, J. R., P. B. Koradia, and C. T. Lim. 1980. "Evaluation of a New Zeolytic Catalyst For NO_x Reduction with NH_3 ." I&EC Product Research & Development 19:218.

Leist, N. R. 1975. The Design and Operation of a Pilot Unit for NO_x Destruction with Ammonia and a Synthetic Mordinite Catalyst. NCL0-1139, National Lead Corporation, Ohio.

Mays, E. B., and M. R. Schwab. 1975. "Elimination of NO_x Fumes." Chemical Engineering, 82(4):112.

Muzio, L. J., J. K. Arand, and K. L. Maloney. 1978. Noncatalytic NO_x Removal with Ammonia. EPRI FP-735, Electric Power Research Institute, Palo Alto, California.

Thomas, T. R., and D. H. Munger. 1978. An Evaluation of NO_x Abatement by NH₃ Over Hydrogen Mordenite for Nuclear Fuel Reprocessing Plants. ICP-1133, Idaho National Engineering Laboratory, Idaho Falls, Idaho.

DISTRIBUTION

No. of
Copies

OFFSITE

- 12 DOE/Office of Scientific and
Technical Information

ONSITE

DOE Field Office, Richland

E. C. Norman

- 3 Westinghouse Hanford Company

HWVP PRMC H5-51

P. Felise

E. T. Weber

- 13 Pacific Northwest Laboratory

L.M.G. Ballou

J. M. Creer

R. W. Goles (5)

Publishing Coordination

Technical Report Files (5)