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Title: **Advanced Technology for Nitrogen Oxide (NO_X) Abatement in Flue Gas Using Microwave Irradiation of Coal Char**

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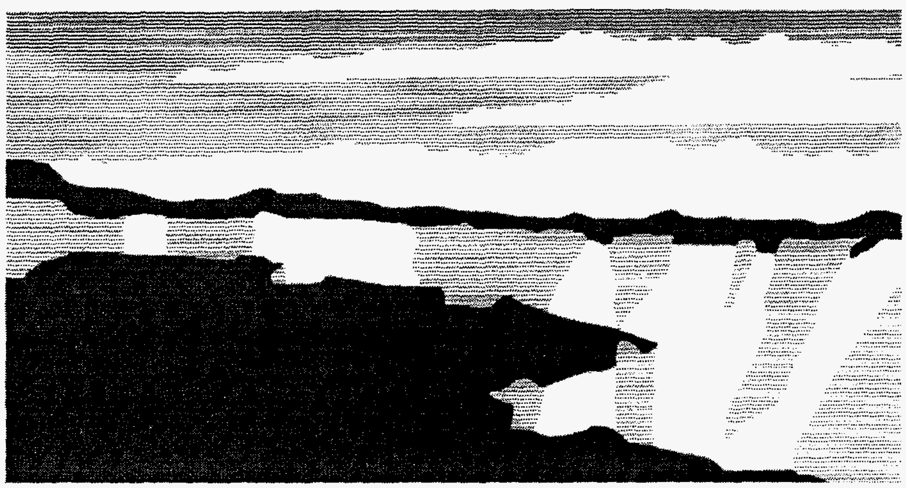

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Advanced Technology for Nitrogen Oxide (NO_X) Abatement in Flue Gas Using Microwave Irradiation of Coal Char

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Abstract

This is the final report of a one-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The project sought to demonstrate a new technology for the elimination of oxides of nitrogen (NO_X) emission from fossil-fuel-fired industrial processes and power plants. The current technologies are not capable of meeting the government mandates for NO_X emission scheduled for implementation in 1997. Our approach is completely new in that microwave energy is the driver for the NO_X removal process that operates at greater than 98% NO_X removal efficiency. Flue gas containing NO_X was passed through a bed of carbon-based reducing agent (coal char) that adsorbed the NO_X. The NO_X-laden char was irradiated with microwave energy that reduced the NO_X to nitrogen and carbon dioxide. The process has been empirically demonstrated in the laboratory on a small scale.

1. Background and Research Objectives

Power generating plants, industrial plants, and internal combustion engines emit more than 18 million tons of nitrogen oxide gases (NO_X) into the atmosphere in the United States each year. These oxides contribute to the formation of acid rain and street-level ozone and thereby pose significant health and environmental hazards. The response of the Environmental Protection Agency (EPA) to this problem has been to legislate anti-pollution laws and regulations, which will be enforced in increasingly strict stages beginning in 1997. The Clean Air Act Amendment of 1990, Title I, provides that the United States will meet a national air quality standard for ozone of 0.12 parts per million. Meteorological modeling indicates that this objective cannot be met without more stringent control of NO_X emissions from new and

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previously exempted sources. In addition, Title IV of the Clean Air Act places severe limits on NO_X emissions to limit the contribution of these sources to the acid rain problem.

Traditional methods for NO_X abatement from flue gases are inefficient and will not be able to achieve compliance with the Clean Air Act Amendment. This is an extremely large problem since almost all fossil-fuel-fired processes result in NO_X emissions. Until very recently, sufficient NO_X reduction was expected to be achieved by regulation of fossil-fuel-fired power plants alone. But more recently, urban-air-shed modeling studies indicate that NO_X abatement from industrial sources will also be required if street-level ozone concentrations are to be brought under control. To meet the requirements of the Clean Air Act, United States industry will be required to make large capital and research investments in new technologies.

A very promising new NO_X-abatement technology has been demonstrated on a laboratory scale by Dr. C.Y. Cha of the University of Wyoming. In this process NO_X is filtered from a carrier gas as it flows through a bed of carbon adsorption material (coal char), where the NO_X is removed as it is adsorbed onto the large surface area of the highly porous carbon. When the NO_X-laden carbon is irradiated with microwave energy, a reaction occurs between the surface-phase NO_X and the carbon surface, producing nitrogen and carbon dioxide. These gases subsequently desorb from the surface into the gas phase. The carbon in the bed is slowly consumed in this process and thus needs to be periodically replenished by adding additional material. Cha has demonstrated that this microwave-driven, NO_X-abatement process can economically remove and destroy more than 98% of the NO_X from an inert carrier gas. This result is dramatically superior to the simple thermally-driven char process in which only about 25% NO_X can be cleanly abated. Apparently, in the conventional thermal case, the heating proceeds so slowly that most of the NO_X desorbs from the surface before the reaction activation temperature can be reached.

The main technical objectives of this project were (i) to provide independent verification of Cha's reported results (that is, demonstrate at least 98% removal efficiency of several hundred parts per million NO from an inert carrier gas); (ii) to investigate NO_X removal from realistic exhaust gas mixtures that contain other reactive components such as oxygen, carbon dioxide, water, and hydrocarbon gases, which can have a profound affect on the chemistry of this process; (iii) to optimize the NO_X-abatement process over a wide range of controllable parameters such as gas flow rate, microwave power and pulse parameters, and the physical properties and impurities of the carbon bed material; and (iv) to consider scale-up issues and commercial feasibility (in close collaboration with Dow Chemical Company).

2. Importance to LANL's Science and Technology Base and National R&D Needs

This project supports Los Alamos core competencies in earth and environmental systems as well as complex experimentation and measurement. It builds upon LANL's ability to bring together multi-disciplinary research teams to solve large-scale, real-world technological problems that often do not lend themselves to convenient division among the traditional disciplines. The process investigated in this project is the most prominent of a number of emerging uses of microwave energy to enhance chemical processes in unique ways. The use of microwave energy to drive chemical processes creates a new range of physical and chemical conditions heretofore not explored in any detail. This opens up a great new range of possibilities for new chemistry and greater control over product purity and cost. Products of the chemical industry drive about one-third of the national economy and many of the large-scale production processes that make up this industry stand to benefit from the implementation microwave technology. In many of these processes, even incremental improvements can yield huge pay-offs. In addition to developing the NO_X technology, this project will help Los Alamos establish its expertise and reputation in the microwave chemical processing area that is currently experiencing very strong growth in the industrial sector.

3. Scientific Approach and Results to Date

The approach taken in this project has been to bring together complementary people and expertise, particularly expertise in microwave systems and experience in engineering gas phase flow systems, to address the outstanding issues of the NO_X abatement technology. Significant progress has been made in several different aspects of this project. Technical success was achieved in combining the gas-flow and diagnostic equipment with a microwave wave guide system into an integrated microwave enhanced chemistry apparatus. A diagram of this combined apparatus is shown in Figure 1. This apparatus gave us considerable freedom in varying the reactant gas mixture and flow rate and microwave irradiation parameters such as peak power and pulse profile.

All of the experimental objectives were met using this apparatus. First, Cha's results on the reduction efficiency of NO in an inert carrier gas were verified. The graph in Figure 2 shows three data sets where NO reduction efficiencies of 99% or greater were accomplished. The graph shows the NO concentration in the output stream in parts per million (ppm) versus time in hours. The input gas mixture consisted of 350 ppm NO in a helium carrier gas. Each of the three runs began with a clean carbon bed (i.e., no pre-adsorbed NO) and so the first part of each data set reflects the loading of the NO onto the char bed until saturation was reached.

The microwave irradiation was initiated only after this saturation was achieved. The time intervals of microwave exposure are indicated by the shaded regions on the graph. The incident microwave power was varied across the three data sets from 250 to 540 to 1020 watts as indicated in the figure.

Next, the more relevant case of NO_X abatement in realistic exhaust gas mixtures was considered. This case is significantly more complicated in that typical exhaust gas contains many components that can themselves react with the carbon bed, with the NO_X, and with each other. The carrier mixture used in this study contained nitrogen, oxygen, carbon dioxide, water, and hydrocarbon gases. Although it proved to be fairly easy to achieve high NO abatement efficiencies with this gas mixture, other important issues arose. Due to the relatively large amounts of oxygen present (approximately 6%), the carbon combustion rate tended to increase by several orders of magnitude and the associated heat of combustion caused "light-off" in the bed. That is, the combustion of the carbon increased uncontrollably, due to the heat generated, to the point that all of the oxygen was burned in the process. This resulted in unacceptably high carbon consumption rates and the production of large amounts of carbon monoxide, an undesirable byproduct. We found that it was possible to suppress this high-temperature carbon combustion as long as the bed temperature was kept below about 50 °C. This could be accomplished by keeping the average microwave power incident to the applicator at sufficiently low levels. However the efficiency of NO destruction is also a function of incident power, tending to decrease with decreasing power. By pulsing the microwave irradiation, however, we found that it was possible to improve the NO reduction efficiency while maintaining an adequately low bed temperature to suppress light-off. That is, we found reduction efficiency to be a sensitive function of the peak power of the radiation pulses whereas the average bed temperature was only sensitive to the average power of the irradiation. Optimization of the process was achieved by using the maximum peak power available (2000 watts), but varying the pulse length and repetition rate such that the bed temperature was maintained just below the light-off value. Clearly, microwave irradiation is essential here with its ability to be rapidly pulsed, something not possible with conventional heating. Through optimization over the microwave pulse parameters, 80% NO reduction was achieved with essentially no excess carbon combustion or carbon monoxide formation. At these reduction levels, this is still a very attractive technology for industrial pollution control. Further, by using higher peak power microwave sources further optimization to higher reduction efficiencies can be expected.

An important issue relating to the ultimate commercial implementation of this technology, which came up in the course of this project, is the uptake rate at which NO_X can be adsorbed onto the char. Because this rate at which a given volume of char can filter NO_X from a gas stream is limited, very large volume filter beds will be required in an industrial application.

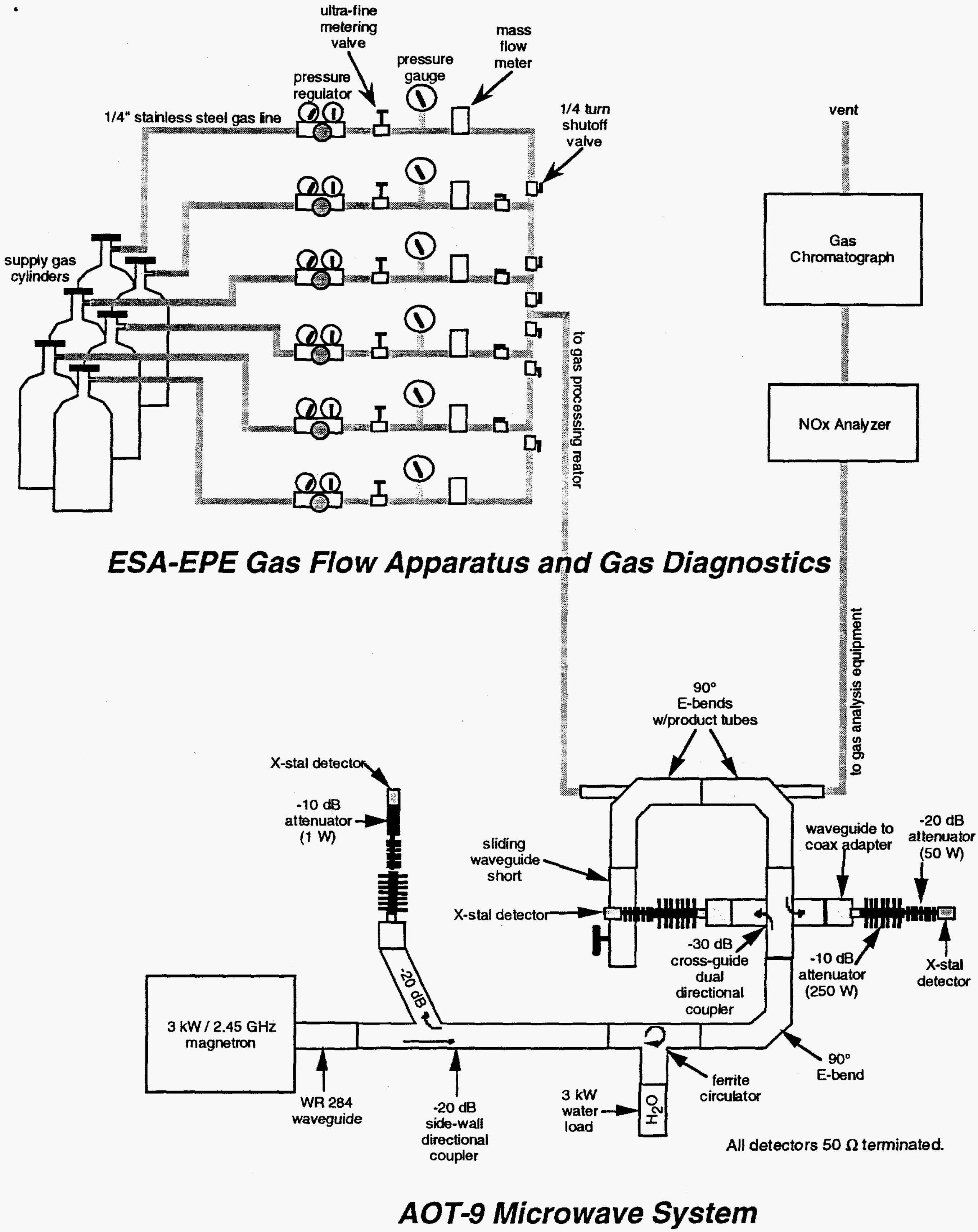

Figure 1. Experimental apparatus combining gas flow manifold, gas diagnostics, and microwave wave guide system.

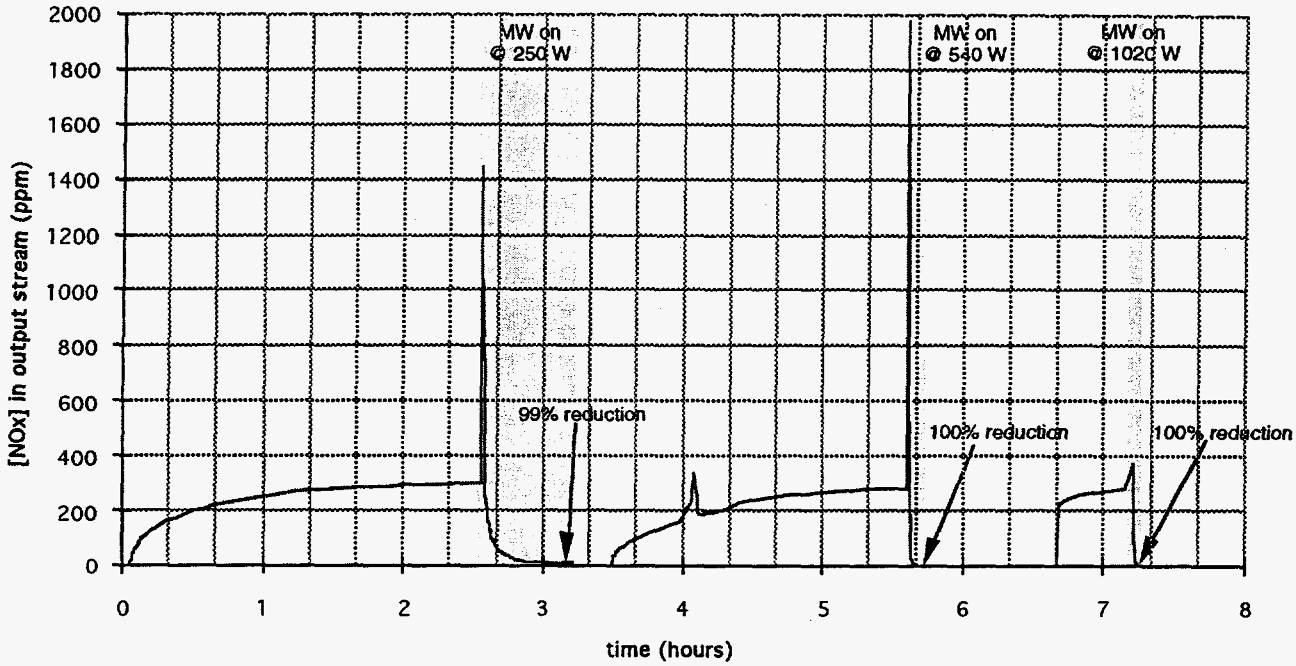

Figure 2. NO_X abatement data. Three data sets corresponding to incident microwave power levels of 250, 540, and 1020 watts.

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