

QUARTERLY REPORT

FOR

DOE/PC/94223--T8

GRANT NO. DE-FG22-94PC94223

ENTITLED

"ADVANCED COMBUSTOR DESIGN CONCEPTS
TO CONTROL NOX AND AIR TOXICS"

SUBMITTED BY

COLLEGE OF ENGINEERING

UNIVERSITY OF UTAH

DR. DAVID W. PERSHING

DR. JOANN LIGHTY

DR. PHILIP SMITH

JENNIFER SPINTI

JOHN VERANTH

STEFAN DOMINO

SEPTEMBER 30, 1996

US/DOE PATENT CLEARANCE IS NOT REQUIRED PRIOR
TO THE PUBLICATION OF THIS DOCUMENT

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

lh

RECEIVED
USDOE/PETO
36 NOV 14 AM 10:45
ACQUISITION & ASSISTANCE DIV.

MASTER

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

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 any of their employees, make any warranty, express 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. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

1.0 Program Overview

Direct coal combustion must be a primary energy source for the electric utility industry and for heavy manufacturing during the next several decades because of the availability and economic advantage of coal relative to other fuels and because of the time required to produce major market penetration in the energy field. However, the major obstacle to coal utilization is a set of ever-tightening environmental regulations at both the federal and local levels. It is, therefore, critical that fundamental research be conducted to support the development of low-emission, high efficiency pulverized coal power systems.

The University of Utah, Massachusetts Institute of Technology (MIT), Reaction Engineering International (REI) and ABB/Combustion Engineering have joined together in this research proposal to develop fundamental understanding regarding the impact of fuel and combustion changes on ignition stability and flame characteristics because these critically affect: NO_x emissions, carbon burnout, and emissions of air toxics. Existing laboratory and bench scale facilities are being used to generate critical missing data which will be used to improve the NO_x and carbon burnout submodels in comprehensive combustion simulation tools currently being used by industrial boiler manufacturers. To ensure effective and timely transfer of this technology, a major manufacturer (ABB) and a combustion model supplier (REI) have been included as part of the team from the early conception of the proposal.

ABB/Combustion Engineering is providing needed fundamental data on the extent of volatile evolution from commercial coals as well as background information on current design needs in industrial practice. Since they will ultimately be a recipient of the enhanced design methodology, they are also providing ongoing review of the practical applicability of the tools being developed. MIT is responsible for the development of an improved char nitrogen oxidation model which will ultimately be incorporated into an enhanced NO_x submodel. Reaction Engineering International is providing the lead engineering staff for the experimental studies and an overall industrial focus for the work based on their use of the combustion simulation tools for a wide variety of industries. The University of Utah is conducting bench scale experimentation to (1) investigate alternative methods for enhancing flame stability to reduce NO_x emissions and (2) characterize air toxic emissions under ultra-low NO_x conditions because it is possible that such conditions will alter the fate of volatile and semivolatile metal species and the emission of heavy hydrocarbons. Finally the University of Utah is responsible for the development of the improved NO_x and carbon burnout submodels.

2.0 Progress During Last Quarter

2.1 Introduction

Understanding the mechanisms of char-N oxidation and reduction is necessary for the accurate modeling of NO_x formation from coal combustion. This statement is especially true for combustors where low- NO_x combustion modification techniques have been applied because in such cases the fraction of total fuel- NO_x coming from the char is very high. This study has focused on obtaining experimental data that can be used to evaluate char-N oxidation and reduction mechanisms. The ultimate goal is to use this knowledge to improve the NO_x submodel in an existing computational fluid dynamics

(CFD) code. Several key experimental results were outlined in the report for the quarter ending 6/96. In order to clarify certain conclusions, more data were collected and analyzed during the quarter ending 9/96. Results from these new data as well as important results not included in the 6/96 report will be addressed here.

2.2 Char-N Experiments

Several mechanisms have been proposed for the heterogeneous reactions of char-N and NO_x in pulverized coal flames. One such mechanism is the reduction of gas phase NO_x to N_2 on carbon in the char. In order to determine the significance of this reaction to the overall reduction of NO_x , a series of tests were run using a high carbon, low ash, commercially available activated carbon known as Nuchar. The Nuchar is produced from coconut shells and has an extremely low nitrogen content (0.13 wt%). Because of this low nitrogen content, any competing NO_x reduction reactions (such as NO_x reacting with char-N to form N_2O) should be eliminated. The results obtained from burning the Nuchar with doped natural gas are shown in Figure 1. The y-axis in this figure represents the percent reduction of gas phase NO_x by the Nuchar. Because of experimental uncertainty, it was not known whether 100% or 75% of the nitrogen in the Nuchar formed NO_x , so both sets of data are shown here for comparison. The main conclusion from this figure is that NO_x reduction on carbon in the char is in the range of 6-13%, at least for the conditions of these experiments. This conclusion agrees with other researchers who have long thought that the solids loading in pulverized coal flames is not high enough for this reduction reaction to be significant relative to the total NO_x present. (Note, however, that this decrease is important relative to the amount of NO_x being generated by the char N is a typical experiment.)

If the reduction of gas phase NO_x to N_2 on a solid carbon surface isn't a significant contributor to the observed reduction of NO_x in gas+char flames, then what is? One possibility is that NO_x reduction must be occurring in the pores of the particle as the oxidized char-N diffuses out of the particle and into the gas phase. The importance of these intraparticle reactions was studied by burning the gas+char mixture in an artificial oxidant composed of Ar, CO_2 , and O_2 . In this system, any interfering reactions of gas phase NO_x with the char- NO_x are removed because there is no gas phase NO_x . Some data from these Ar/ CO_2 / O_2 experiments was presented in the 6/96 quarterly report. In Figure 1 of that report, the char-N to NO_x conversion was plotted as a function of char type. From this figure it was clear that intraparticle char-N to NO_x conversion is a function of char type with the lignite and subbituminous coal chars showing higher conversions than the bituminous coal chars.

In addition to coal type, other factors that may affect intraparticle char-N to NO_x conversion include oxygen concentration and flame temperature. Figure 2 illustrates the effect of oxygen concentration on char-N to NO_x conversion in Ar/ CO_2 / O_2 . These data were taken for two different Pittsburgh No. 8 chars. The char-N to NO_x conversion of both chars shows a strong dependence on oxygen concentration with conversion increasing by approximately 10% as oxygen concentration ranges from 2%-7%. Figure 3 shows the influence of flame temperature on NO_x formation from char-N. In this figure, data obtained with a Pittsburgh No. 8 char are plotted for two different mixtures of Ar/ CO_2 / O_2 . The "Heaters ON" and "Heaters OFF" designation refers the electric heaters which are used to preheat the combustion air. When the heaters are on, the temperature of the combustion air entering the burner is 530 K. When the heaters are off, the entering combustion air temperature is 300 K. It appears from Figure 3 that this temperature

difference does not affect NO_x production from the char. This result is surprising since chemical kinetics are highly dependent on temperature. These data would seem to indicate that something other than reaction kinetics is rate controlling.

2.3 Air Toxic Experiments

Coal combustion particle emissions in the range between 0.1 and 10 μm from the laboratory furnace have been measured. The mass distribution near the U-furnace exit, as measured using a dilution probe and Andersen cascade impactor, was reproducible with a typical standard deviation of 25% of the mean. The effect of alternative sampling methods was comparable to the run-to-run variation. The data are within the range of the measurements of particle mass or volume reported in the literature.

Scanning electron micrographs showed roughly spherical particles that are within the range expected from the impactor stage calibration. A quality assurance technique has been developed that can be used to check for oversize particle contamination in samples that are being prepared for elemental analysis. This experimental series successfully validated the equipment and methods which will be used the upcoming measurements of the effect of low- NO_x burners on fine particle emissions.

2.4 NO_x Modeling

The University of Utah pulverized coal combustion reactor represents the base case for the Nitrogen pollutant modeling effort. Experimental data obtained from the University of Utah combustion lab is used to compare experimental results to theoretical calculations. Glacier, a three dimensional combustion program developed by Professor Smith is used to converge the velocity, density, and temperature fields. The decoupled NO_x model is then used to calculate Nitrogen pollutant formation since low concentrations of Nitrogen pollutants do not appreciably affect the global velocity, density, and temperature fields.

Essential to accurate NO_x prediction is the correct correlation of Nitrogen release rates to the amount of Carbon reacted. Figure 4 represents a diagram of the three conceptualized algorithms. Algorithms (1) and (2) have been used extensively, while algorithm (3) is under development. Using algorithm (2), separate Nitrogen evolution rates are used during the devolatilization and oxidation process of coal combustion. These Nitrogen evolution rates are obtained from additional experiments which determine the Nitrogen/Carbon split for both coal and char. The advent of more complete data sets for the coal dependent functionality of Nitrogen release rates over both the devolatilization and oxidation regime [Sandia National Laboratory, 1995] has provided impetus to develop algorithm (3); incorporation of tracking Nitrogen release rates throughout the coal combustion process.

Figure 5 illustrates the comparison between experimental and theoretical results for NO_x concentrations in the University of Utah furnace using Pittsburgh #8 coal in a premixed burner. Figure 5 elucidates the fact that using the current model of Nitrogen pollutant formation, within the context of engineering accuracy, is satisfactory. Though other improvements in the NO_x model are warranted, we have now shifted our efforts to look at other primary effects of Nitrogen pollution formation such as the amount of residual carbon in fly ash. Though the Carbon burnout model is not contained within the NO_x model, the burnout model can directly influence the accuracy of NO_x prediction.

Our current Carbon burnout model does not include physical changes in the coal particle such as thermal annealing which dramatically affects the reactivity of the coal particle. The rate at which char oxidation proceeds dramatically affects the degree of carbon burnout. The group of Hurt *et al* (1996) has developed a model whereby variations of char density and combustion reactivity are quantified. The group has developed a "statistical model" which accounts for the heterogeneity of reactivity between particles with different "histories". The model of Hurt *et al* has not been incorporated into a global CFD code such as Glacier and efforts to incorporate it are under way. The model has only recently been furnished to our group.

It is our ongoing effort to improve the model of Nitrogen pollutant formation. We must realize; however, that the continual improvement of primary effects contained within the code Glacier is essential to physically based accurate NO_x predictions throughout the combustion chamber. Thus, the development of each separately would constitute a grave error; one model can not be overlooked during the improvement of the other. The two are concomitant.

3.0 Plans for Next Quarter

Work will continue in the analysis of all the experimental results with the goal of identifying the most important parameters for subsequent modeling. Char-N modeling will also continue with step one being the evaluation of the current NO_x submodel using experimental data. Most likely, the current NO_x model will have to be modified in order to accurately predict the results obtained in the char-N experiments.

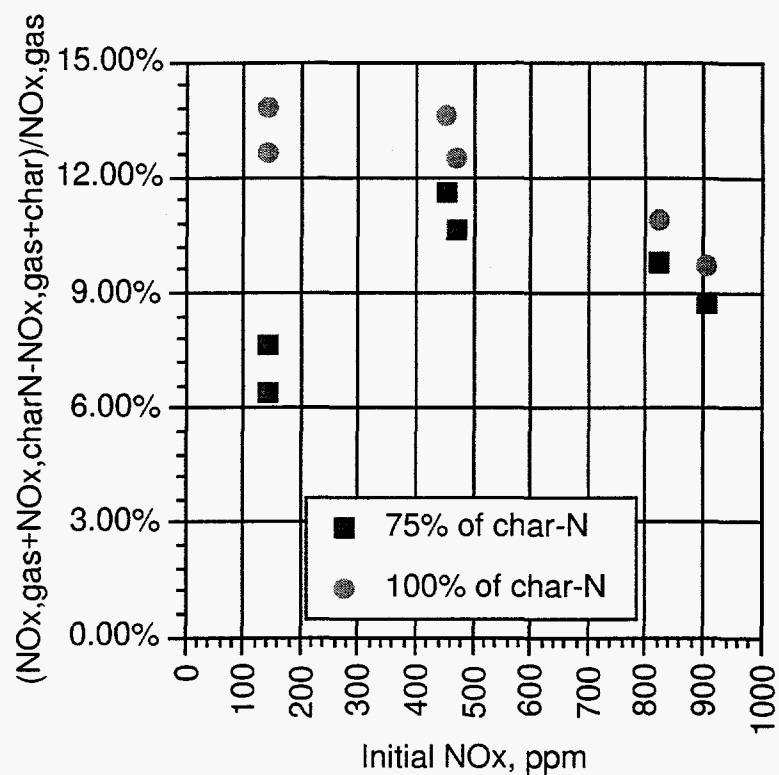


Figure 1
Gas firing rate=24.9 kW, Nuchar firing rate=4.4 kW, oxidant=air

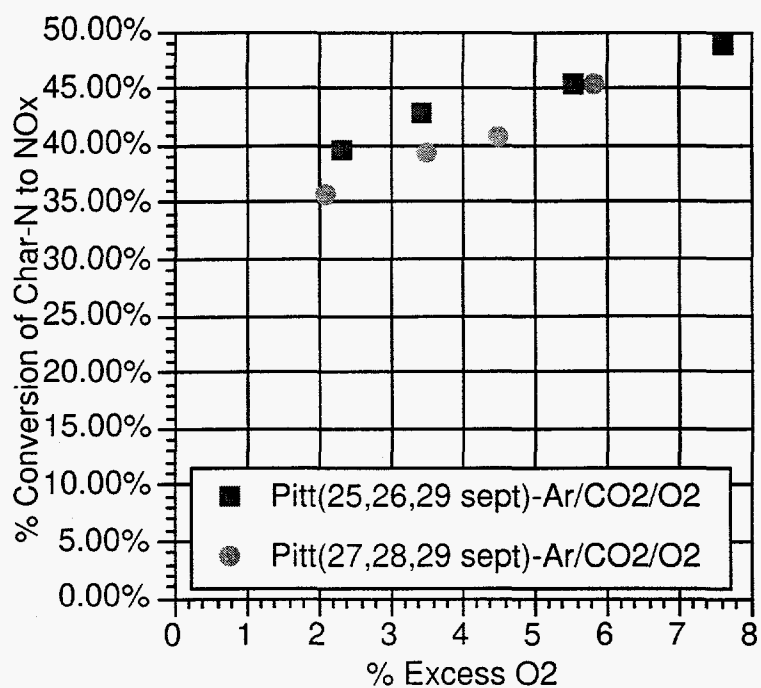


Figure 2
Gas firing rate=24.9 kW, char firing rate=4.4 kW, oxidant=64%Ar/15%CO₂/21%O₂

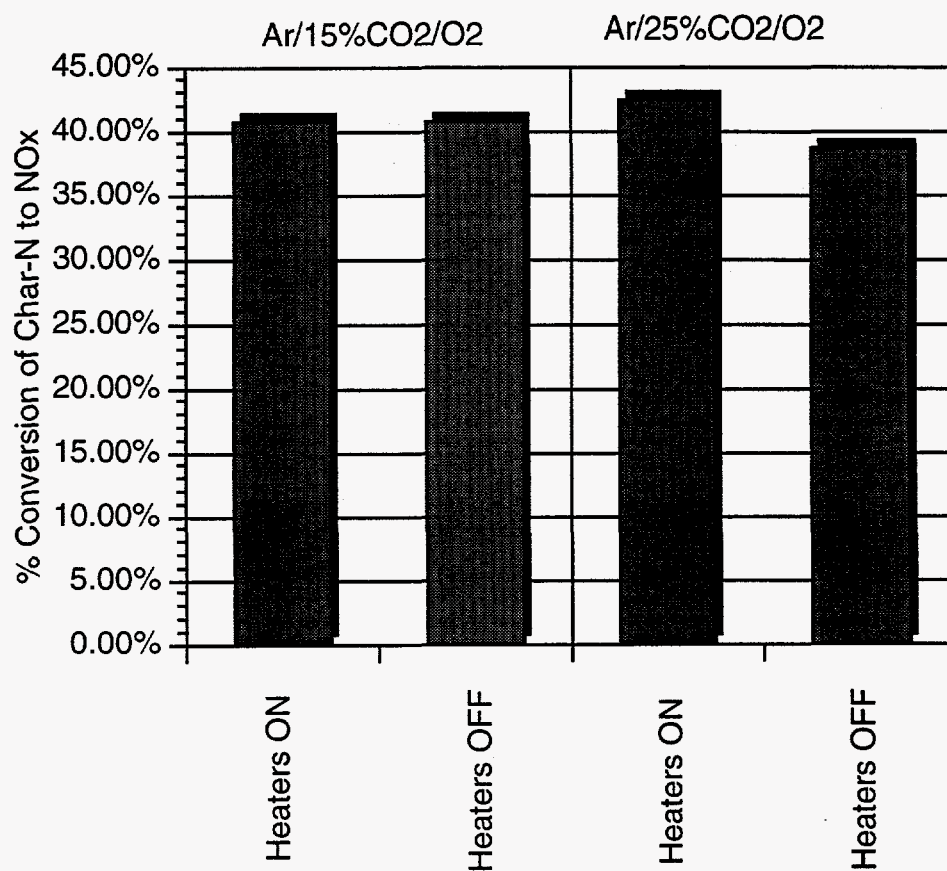


Figure 3
Gas firing rate=24.9 kW, char firing rate=4.4 kW
Oxidant=Ar/CO₂/O₂

Nitrogen Release Rates

Three generations of model algorithms

Algorithm (1) : N/C ratio fixed

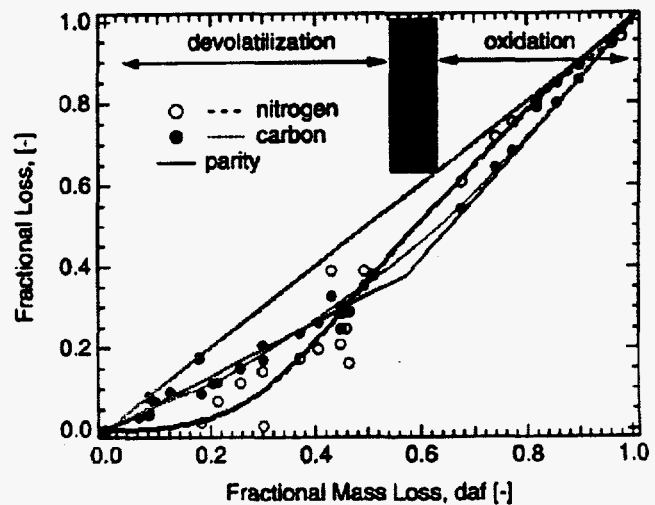
- most common means by which Nitrogen release rates assessed
- requires ultimate analysis of coal

Algorithm (2): N/C double-valued

- two separate Nitrogen evolution rates for devolatilization and oxidation
- requires additional experiments to determine split. —Data from ABB and Sandia National laboratories.

Algorithm (3) : N/C ratio tracked

- functionality of Nitrogen release rates used over both devolatilization and oxidation regime.
- Suite of 15 coal types— data from Sandia



Nitrogen release rates for Algorithms 1–3

Figure 4 Nitrogen release rate algorithms.

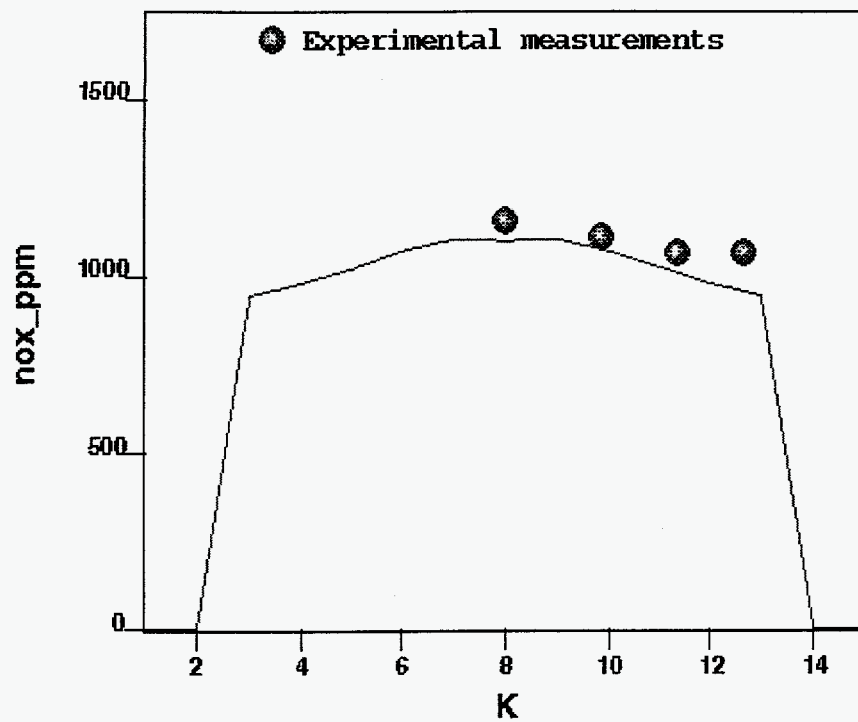


Figure 5 Experimental measurements compared to theoretical calculation for U of U case Small: Pittsburgh #8, premixed burner.