



Project Summary

Development of Criteria for Extension of Applicability of Low-Emission, High-Efficiency Coal Burners: Third Annual Report

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A brief summary of progress on this program since its initiation in 1977 is shown in Figure 1, along with the experimental efforts proposed for fiscal year 1981. The major thrust of the program in 1980 was in the small-scale efforts to define the impact of fuel properties and operating conditions on NO_x emissions. Initial studies were also undertaken to assess the viability of using dry sorbents for SO_x control. The progress of the program's pilot-scale studies has been hindered because the large water-tube simulator (LWS) and small water-tube simulator (SWS) facilities were not available after January 1980. Furnace deterioration necessitated complete rebuilding of these units before further testing could be accomplished. This rebuilding effort was completed in January 1981 and testing was commenced.

For the small-scale fuel studies, 28 coals covering all ranks were tested under a wide variety of conditions to ascertain the impact of coal properties on the fate of fuel nitrogen. Significant accomplishments in this part of the program include:

- Potential sources of NO_x during coal combustion are: (1) fixation of nitrogen in the combustion air; (2) conversion of fuel NO in the volatiles; and (3) conversion of fuel NO in the solid phase. Under typical conditions, oxidation of nitrogen chemi-

cally bound in the coal is the major source of NO_x emissions. The conversion of nitrogen which remains in the solid phase during devolatilization is inherently low. Hydrocarbon volatiles do, however, appear to enhance this conversion. The conversion of nitrogen evolved with the volatiles appears to be higher than that of char nitrogen, but significantly less than suggested by gas- and liquid-phase results. The solid phase is believed to play a major role in suppressing volatile nitrogen conversion.

- The bench-scale test results confirm the pilot-scale concept that decreasing the initial air/fuel ratio decreases fuel NO_x formation. The initial fuel-oxidant contacting rate appears to have little influence on heterogeneous nitrogen oxidation; however, it has a strong effect on volatile nitrogen oxidation. With premixed or rapidly mixed systems, fuel NO formation increases with increasing fuel nitrogen content; however, other fuel properties also significantly affect the fate of fuel-bound nitrogen during combustion. In particular, fuel nitrogen conversion is proportionally greater with coals containing a high fraction of volatile reactive nitrogen.

| <i>Program Objectives</i> | <i>Areas of study concentration Oct 1977 - Oct 1980</i> | <i>Key results</i> |
|---|---|---|
| <i>1. Expand fuel capability of distributed mixing burner (DMB)</i> | <ul style="list-style-type: none"> ● <i>Define impact of fuel properties and rank on NO_x</i> <ul style="list-style-type: none"> ● <i>Major effort in small-scale controlled experiments</i> ● <i>Limited pilot-scale efforts</i> | <ul style="list-style-type: none"> ● <i>Small scale</i> <ul style="list-style-type: none"> ● <i>Optimum staging condition is dependent on fuel properties</i> ● <i>No emissions are related to TFN as well as other fuel and process variables</i> ● <i>SO₂ measurement methods have been defined</i> ● <i>Pilot scale</i> <ul style="list-style-type: none"> ● <i>Impact of fuel properties on NO_x under typical burner combustion conditions more complex than simply fuel nitrogen content related</i> |
| <i>2. Explore additional concepts</i> | <ul style="list-style-type: none"> ● <i>No effort experimentally</i> | <ul style="list-style-type: none"> ● <i>Need for inboard tertiary air identified for retrofit boilers - aerodynamic staging</i> |
| <i>3. Burner configurations</i> | <ul style="list-style-type: none"> ● <i>Front wall fired</i> <ul style="list-style-type: none"> ● <i>Single burners</i> ● <i>Burner scale</i> ● <i>Multi-burners</i> | <ul style="list-style-type: none"> ● <i>Single burners</i> <ul style="list-style-type: none"> ● <i>Developed design data base for DMB (NO_x)</i> ● <i>Identified in-situ dry sorbent as having potential for combined NO_x/SO_x combustion modification control</i> ● <i>Multi-burners</i> <ul style="list-style-type: none"> ● <i>NO_x characteristics similar to single burners</i> ● <i>Emissions (NO_x) correlate with total heat input</i> ● <i>Tertiary air ports location critical to CO levels but insensitive to NO_x</i> ● <i>Scale</i> <ul style="list-style-type: none"> ● <i>Scaling by velocity results in higher NO emissions at higher heat input (might be related to furnace effects)</i> |
| <i>4. Compare DMB with commercial burners</i> | <ul style="list-style-type: none"> ● <i>Evaluation of commercial burners in LWS</i> | <ul style="list-style-type: none"> ● <i>General flame characteristics similar to field operation</i> ● <i>NO_x levels 50-100 ppm lower than field</i> ● <i>CO levels >> than field</i> ● <i>Stability and performance similar to field</i> |
| <i>5. Industry coordination</i> | <ul style="list-style-type: none"> ● <i>Setup broad participation in review panels</i> | <ul style="list-style-type: none"> ● <i>Held three technical panel meetings</i> ● <i>Held two technology transfer panel meetings</i> |
| <i>6. Support field application of DMB technology</i> | <ul style="list-style-type: none"> ● <i>Identified several field/commercial problem areas and limits on general acceptability of results</i> | <ul style="list-style-type: none"> ● <i>Retrofit of existing boilers with tertiary air ports not economical</i> ● <i>Need for flexibility in location of tertiary air ports</i> ● <i>More detailed design criteria required showing operating limits for DMB and performance data</i> ● <i>Need for testing with wide range of fuels</i> |

Figure 1. Program executive summary of key results.

- Detailed studies on the optimization of a staged combustion system suggest that the *stoichiometry producing minimum NO_x emissions is a function of both fuel composition and primary-zone conditions*. As first-stage stoichiometry is decreased, the NO formed in the first stage decreases, but other oxidizable gas nitrogen species increase as does nitrogen retention in the char.
- *The distribution of the total fixed nitrogen (TFM) species (NO, NH₃, HCN) leaving the first stage is strongly dependent on coal composition*. In general, the first stage NO percentage decreased significantly with decreasing coal rank from anthracite to lignite. However, the percentage of NH₃ grew with decreasing rank which may give greater NO emissions in the second stage. HCN was greater than NH₃ in all bituminous coals, but less than NH₃ with all subbituminous and lignite coals.
- The distribution of the first-stage fuel nitrogen emissions had a significant impact on the second-stage exhaust NO emissions. *The minimum second-stage NO emissions depend on competition between the first stage NO and increased gas and solid-phase nitrogen species*.
- During staged combustion, increasing the rate of heat extraction from the first stage, or fuel-rich zone, decreases the decay of TFN species, but dramatically decreases TFN conversion in the second stage. *Thus, first-stage heat extraction has the net effect of reducing exhaust NO emissions*.

This effort is now complete, and future small-scale studies will concentrate on the dry sorbent for SO_x control.

Previous effort suggested that careful control of the temperature/time/stoichiometry history in the burner zone was essential for effective utilization of a dry sorbent for removal of SO₂ in the furnace. Based on these results, a study was initiated to verify and develop the technical information to extend the results to an operating boiler system. The effort to date has concentrated on verifying the measurement techniques under both gas-phase and heterogeneous combustion conditions. The results suggest that:

- Uniform dispersion of limestone in the combustion zone is critical to obtaining high sorbent utilization.
- Proper control of temperature/time/stoichiometry can produce significant SO₂ removal from combustion products.
- Experimental measurement methods can be designed which provide sufficient accuracy for reasonable sulfur balances.

The major thrust of the experimental efforts during the next fiscal year is to provide critical data addressing problems and concerns expressed by manufacturers and users which could limit their acceptability of the DMB demonstration results and burner concept. These areas are: (1) applying the distributed-mixing concept to a burner without outboard air to meet retrofit application; (2) determining the impact of a wide range in coal characteristics on boiler operation, maintenance, and performance; and (3) defining limits with respect to performance and flame stability. These efforts will be closely coordinated with the companion EPA demonstration programs 68-02-3130 (utility boilers), 68-02-3127 (industrial boilers), and RFP DU 80-A162 (large utility boilers; under negotiation) to ensure that the most relevant pilot-scale experiments are conducted.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The EPA's overall program in research, development, and demonstration of low-NO_x wall-fired burners is shown in Figure 2. Since initiation of this effort in the early 1970s, EPA has recognized the importance of combustion modification through burner design as the most cost-effective NO_x control method. The Combustion Research Branch of EPA's IERL-RTP initiated the first study at the IFRF (68-02-0202) to better understand the influence of burner design on NO_x formation. The results of this study showed that fuel/air contacting, largely controlled by burner design, is the key to controlling NO_x emissions. This result then led to the definition and demonstration of EPA's first distributed-mixing burner (DMB) in 1972. Under two sepa-

rate contracts, the EPA then pursued the development of this concept. In the first study (68-02-1488), the basic burner design criteria were studied; units were scaled up to 100 x 10⁶ Btu/hr and multiple-burner interaction was investigated. The most significant result of this study was demonstration that under furnace-simulated conditions the system could operate at less than 0.2 lb NO_x/10⁶ Btu. As a logical extension of this effort, EPA then undertook this contract (68-02-2667) to extend the burner criteria to cover a wider range of fuels, as well as to study other burner configurations in addition to providing on-going support to EPA demonstration programs 68-02-3127, 68-02-3130, and RFP DU 80-A162. Consequently, contract 68-02-2667, in addition to its prime R/D objective, must also act as the bridge between the R/D studies and resolution of practical field application problem areas.

The Development of Criteria for Extension of Applicability of Low-Emission, High-Efficiency Coal Burners program was initiated in October 1977. The overall program is structured to meet six objectives:

1. Expand fuel capability of DMB to include fuels proposed for use by utilities.
2. Explore additional burner concepts.
3. Determine effects of burner configurations on performance/emissions characteristics.
4. Provide direct comparison between DMB and commercial burners.
5. Ensure appropriate technology transfer to manufacturers and users.
6. Provide test support for field demonstrations.

The program is divided into the seven tasks shown in Figure 3. Task 1—Program Definition—is designed to plan the three key elements of the program: (1) fuels selection; (2) experimental plan; and (3) measurements of protocol. The remainder of the program is structured to carry out these efforts. Tasks 2 through 5 are experimental efforts designed to progressively move from bench-scale process studies (Task 2—Fuel Screening Experiments) to single- (Task 3—Single-Burner Experiments) and multiple-burner configurations at pilot scale, which incorporate the distributed-mixing concept, and then to compare the operating and emissions characteristics of the DMB against commercial technology

(Task 5—Comparison to Current Technology). Throughout the program, results are continually reviewed with manufacturers and users to secure their advice and recommendations before the finalization of the testing program as part of Task 6—Industry Coordination. All of the information is then analyzed in Task 7—Data Analysis and Criteria Development—and DMB design guidelines are established.

Bench-Scale Studies to Assess the Impact of Coal Type on NO_x Formation

Twenty-eight different coals as well as several simulated coals (i.e., char/propane and char/propane/ammonia or

nitric oxide) were tested under excess air and staged conditions to study the effects of fuel properties on the fate of fuel-bound nitrogen during combustion. Detailed measurements of first-stage and exhaust species concentrations were performed for 14 different coals to investigate the parameters controlling a staged combustion process.

Results point to the following conclusions and implications.

Excess Air

NO formation during pulverized coal combustion can be attributed to the nitrogen in both the volatiles and the char. The conversion of nitrogen which remains in the solid phase during devolatilization is inherently low. The conver-

sion of nitrogen evolved with the volatiles appears to be higher than that of the char nitrogen. The presence of hydrocarbon volatiles, however, enhances the conversion of char nitrogen. On the other hand, the presence of solid phase is believed to suppress the conversion of volatile nitrogen. The initial fuel/air contacting rate controls the oxygen availability for both fuel nitrogen fractions. It appears that the volatile nitrogen conversion increases with increasing mixing rate, but the conversion of char nitrogen is less sensitive to change in the initial air distribution.

Thus, coals which evolve more volatile nitrogen are expected to produce higher NO emission under well mixed conditions. Data obtained under excess air condi-

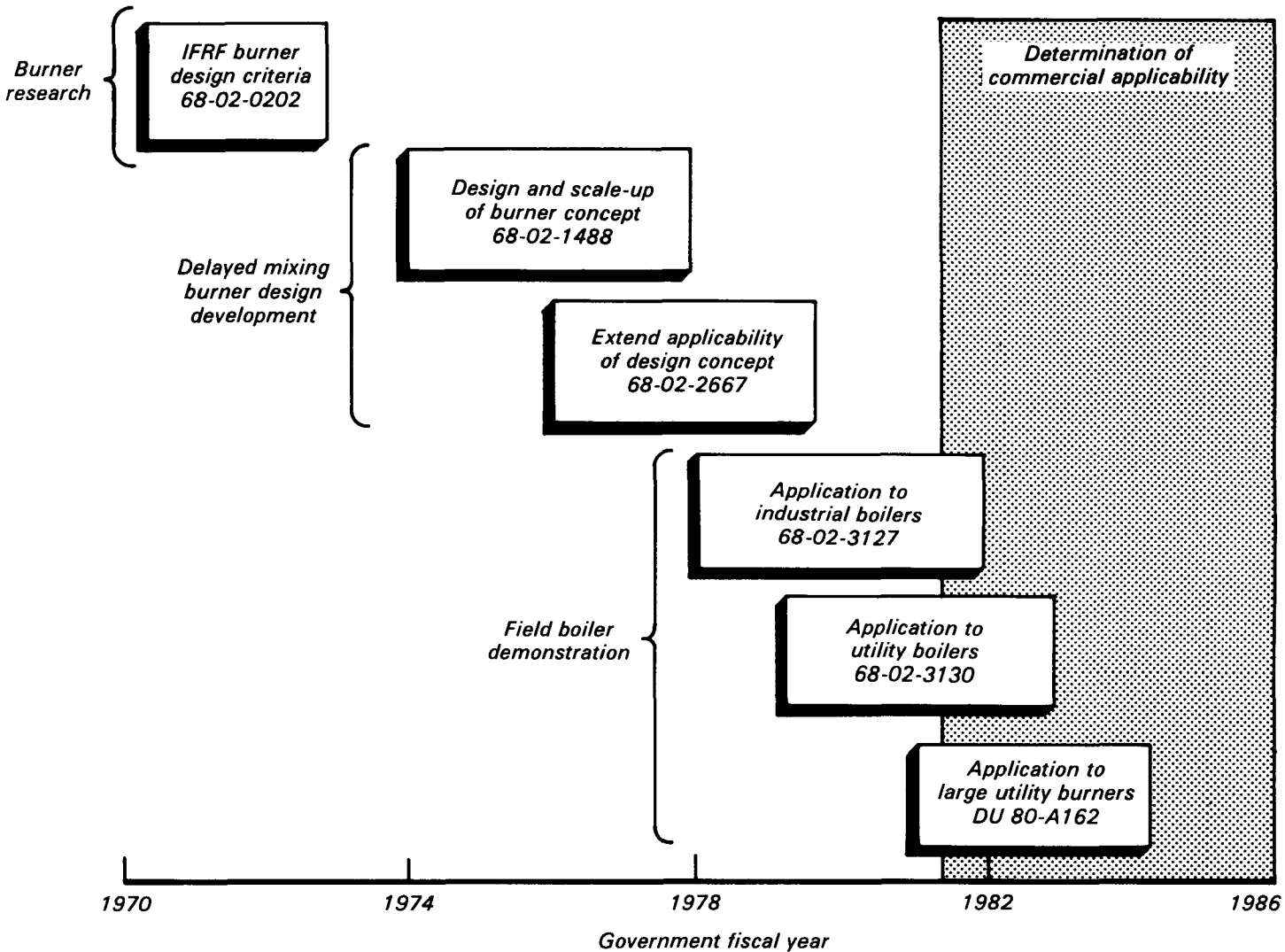


Figure 2. EPA low NO_x burner RD&D programs.

tions indicate that, with a premixed or rapidly mixed system, fuel NO formation increases with increasing fuel nitrogen content; however, other fuel properties also significantly affect the fate of coal nitrogen. Attempts to correlate these data (on the basis of the nitrogen lost in the ASTM volatile determination or with such basic fuel properties as percent nitrogen, percent volatiles, or coal rank) have been successful.

An experiment has been developed in another EPA-supported program to define the reactive volatile nitrogen fraction of coals. Six of the very different coals tested in the bench scale reactor were pyrolyzed under inert conditions at 1370 K, and the volatile nitrogen species which were converted to HCN were measured. Figure 4 shows that the data obtained in this quartz reactor pyrolysis experiment agree qualitatively with the experimental results obtained with the

premixed burner. The Savage lignite had the highest percentage conversion of fuel nitrogen to NO in the combustion experiments, and also the highest fractional volatile reactive nitrogen yield in the inert pyrolysis study. Likewise, the Beulah lignite had the lowest in both experiments. The slope of the correlation decreases with decreasing mixing rate (premixed or radial diffusion), and the axial diffusion data do not correlate well. This may be because the pyrolysis experiment does not characterize char nitrogen. Therefore, it can be concluded that, under excess air conditions, fuel nitrogen conversion is proportionally greater with coals containing a high fraction of volatile reactive nitrogen.

Staged Combustion

Detailed measurements of first stage and exhaust species concentrations suggest that a staged combustion sys-

tem must be optimized with respect to first-stage stoichiometry and residence time, fuel properties, and heat extraction rate. As first-stage stoichiometry is decreased, the NO formed in the first stage decreases, but other oxidizable gas-nitrogen species increase, as does nitrogen retention in the char. Total fixed nitrogen (TFN = NO + NH₃ + HCN) generally increases with increasing fuel nitrogen and correlates well with excess-air exhaust emissions. Increasing the residence time in the fuel-rich stage allows TFN species to decay toward low equilibrium values and thus reduces exhaust NO emissions.

The distribution of the TFN species leaving the first stage is strongly dependent on the coal composition. Of the 12 coals tested in detail, only two produced high HCN concentrations. Figure 5 shows the percent of input fuel nitrogen exiting the first stage as NO, NH₃, or

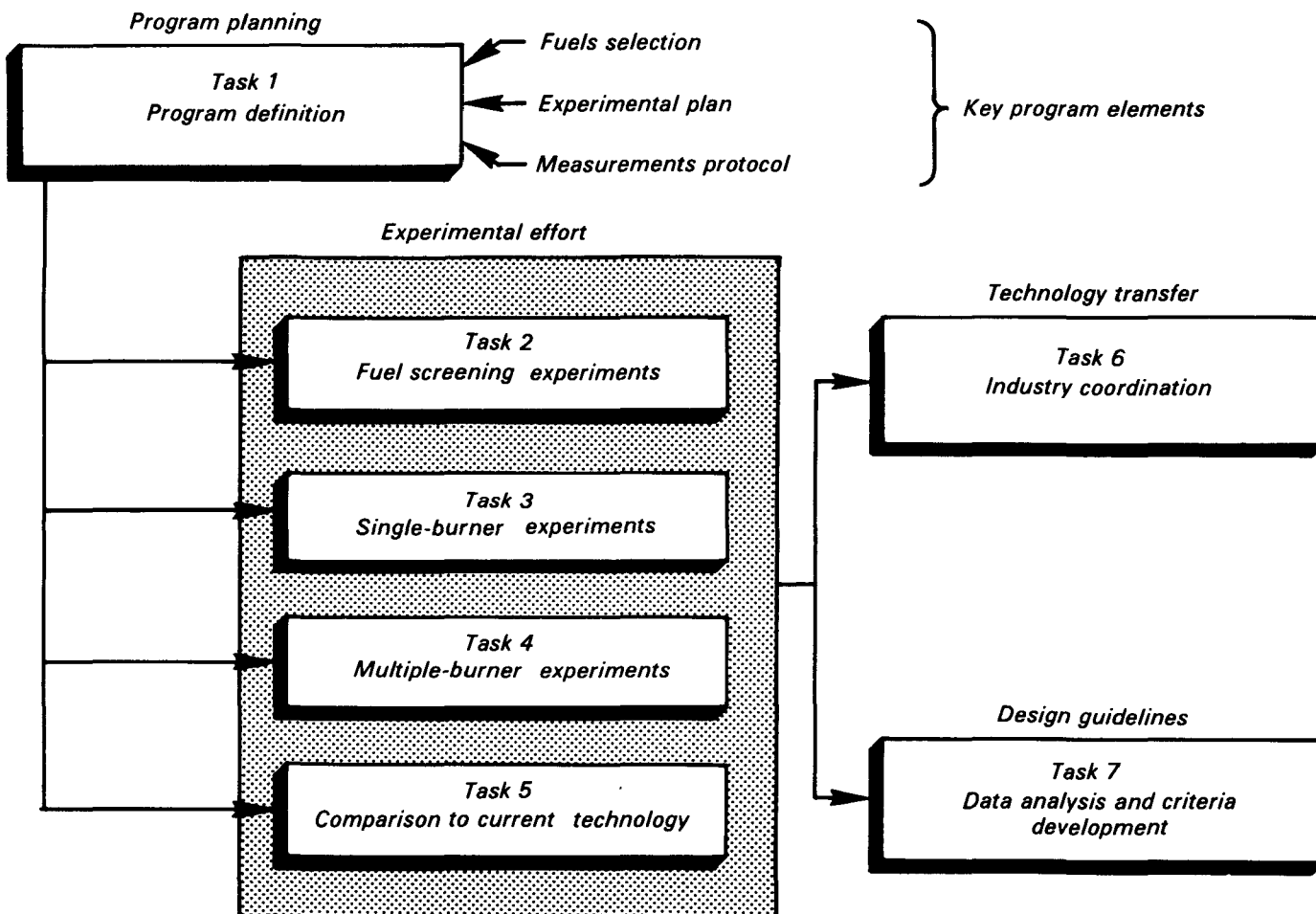


Figure 3. Program structure for development of criteria for extension of applicability of low-emission/high efficiency coal burners.

HCN at $SR_1 = 0.70$. The German bituminous coal (Saar) correlates well with American bituminous coals. The anthracite and the Utah char (classified as semianthracite according to the criteria outlined in ASTM Method D 388) produced relatively large amounts of first stage NO. This could be due to the increased oxygen partial pressure in the early stages of combustion. With low volatile fuels, the rate of consumption was reduced to decreased hydrocarbon evolution. For all the bituminous coals except the Indiana bituminous both NH_3 and HCN usually increased with increasing fuel volatility. HCN concentrations were higher than NH_3 for all the bituminous coals. However, for subbituminous and lignite coals' HCN concentrations were significantly less than NH_3 levels at all stoichiometries.

In general, NO at the exit of the first stage appears to decrease with decreasing coal rank from anthracite to lignite. Conversely, the relative amounts of NH_3 grow with decreasing rank. The percent of initial nitrogen leaving the first stage

as HCN was below 4 percent for most of the coals tested.

Second-stage TFN conversion decreases as the TFN distribution shifts in favor of HCN and NH_3 , and as the hydrocarbon content of the second-stage reactants increases. The percentage conversion of char nitrogen to NO in the second stage is low (< 20 percent). First-stage cooling decreases the TFN decay in the fuel-rich zone and, hence, increases the TFN and char nitrogen carry-over into the second stage. Reduced second-stage flame temperatures have little effect on solid-phase nitrogen conversion, but they dramatically decrease gas-phase TFN conversion due to a shift in controlling flame chemistry.

The exhaust NO emissions depend on first stage stoichiometry, on the amount and speciation of the TFN entering the second stage, and on the amount of char nitrogen. Thus the exhaust NO data can be correlated with a model of the form:

$$\text{NO exhaust} = X_1 (\text{NO}) + X_2 (\text{NH}_3 + \text{HCN}) + X_3 (C_N + \text{NO}_{\text{Th}})$$

where

- X_1 = conversion of NO,
- X_2 = conversion of NH_3 and HCN,
- X_3 = conversion of char nitrogen, and
- NO_{Th} = thermal NO formation

in the second stage flame. The TFN concentrations were correlated in ppm, dry 0% O_2 based on the detailed measurements at the end of the first stage. The char nitrogen (C_N) was used as equivalent ppm based on stoichiometric conversion of the remaining nitrogen.

As noted previously, NO conversion in the second stage flame did not appear to be a strong function of NO concentration; however, it was found to depend on first-stage stoichiometry (probably specifically on the hydrocarbon content of the second stage flame). Data from pairs of Utah char/ C_3H_8 / NH_3 experiments indicated that, as the SR was reduced from 0.9 to 0.6, NO conversion dropped from approximately 89 percent to 51 percent with no change in HCN or NH_3 concentrations. Therefore, NO conversion was modeled as:

$$X_1 = a \cdot SR$$

where a = correlation constant.

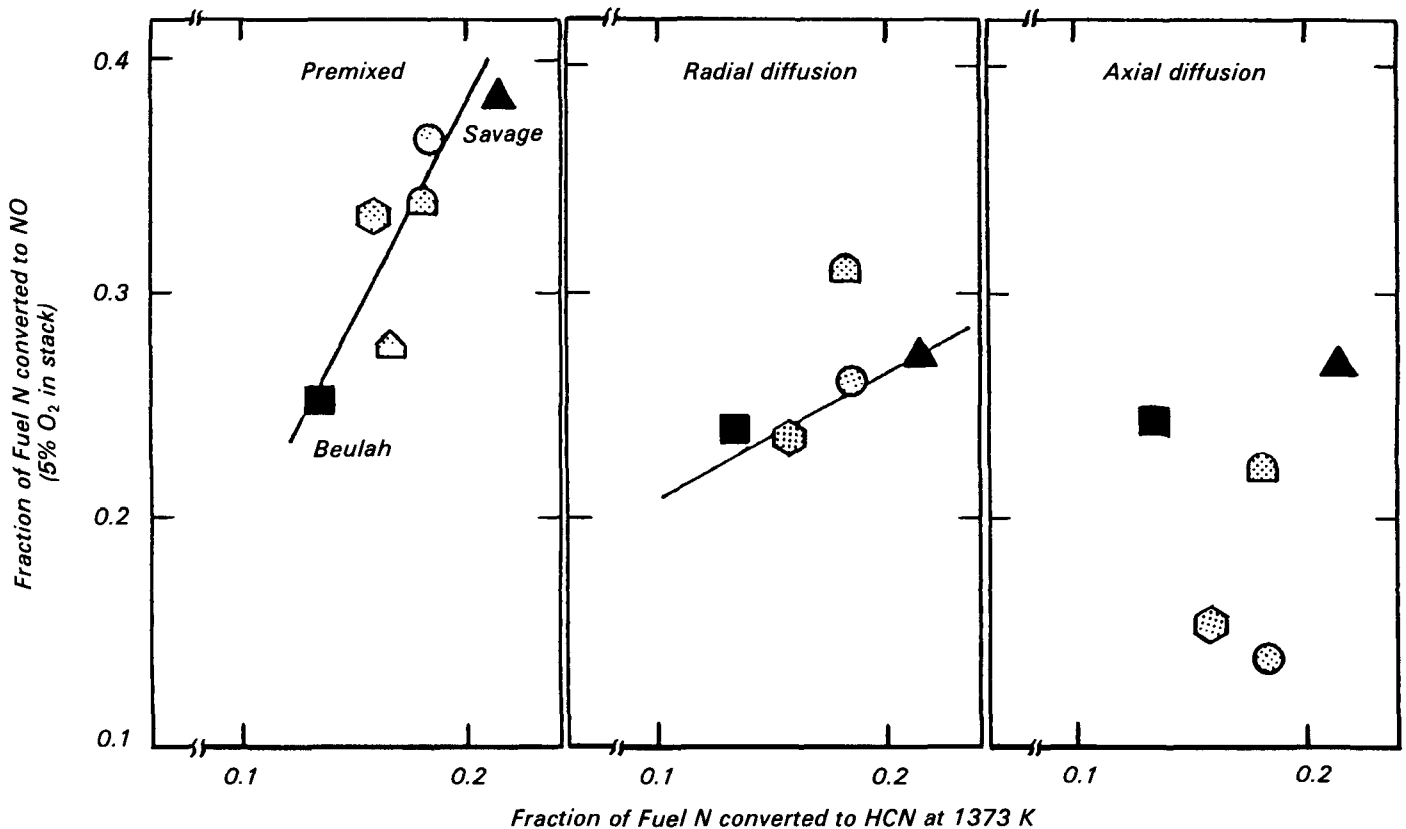


Figure 4. Fuel NO correlation.

Figure 6 indicates that the conversion of HCN and NH₃ decreases with increasing TFN content; hence, a reciprocal model was used for these conversions:

$$X_2 = \frac{1}{\beta + \gamma (\text{NH}_3 + \text{HCN})}$$

Char nitrogen conversion was assumed to be approximately constant: $X_3 = \delta$ as was the contribution of thermal NO formation, NO_{Th} .

A multivariable linear regression program was used to calculate the required parameters. The resulting correlation was:

$$\text{NO}_{\text{ex}} = 0.8(\text{SR})(\text{NO}) + \frac{1}{2 + 0.007(\text{NH}_3 + \text{HCN})} (\text{NH}_3 + \text{HCN}) + 0.2 C_N + 5.$$

Figure 6 shows the results of this correlation plotted against the measured ex-

haust NO and indicates that the agreement is reasonable considering the wide range of coals (lignite-bituminous) and conditions ($\text{SR}_1 = 0.45-0.90$). The overall correlation coefficient (r^2) was 0.93.

The equation suggests that first stage NO has a very significant impact on the ultimate exhaust NO concentration; even at $\text{SR} = 0.45$, the retention of NO is predicted to be 36 percent. NH₃ and HCN conversions are predicted to decrease from 50 percent at low concentrations to 18 percent at 500 ppm of HCN + NH₃. Char nitrogen conversion was estimated to be approximately 20 percent, and thermal NO was predicted to be relatively low in this system (5 ppm).

The above correlation, valid when the first stage operates without cooling, suggests that optimum staged combustion technology will minimize first stage NO formation and attempt to maximize the TFN decay in the first stage. Char NO formation in the second stage flame can also be significant if large amounts of nitrogen are allowed to remain in the solid phase; hence, this should be avoided. Results indicate that increasing the rate of heat extraction from a staged combustion system reduces NO emissions by a more complex reaction.

Bench-Scale Studies to Assess the Impact of Coal Type on SO_x Emissions and Sorbents for SO_x Emissions Reduction

This effort has only been recently initiated, and results should be considered preliminary. General conclusions that can be drawn from this effort are:

- Uniform dispersion of limestone in the combustion zone is critical to obtaining high sorbent utilization.
- Results suggest that proper control of temperature/time/stoichiometry can produce significant SO₂ removal from combustion products.
- Experimental measurement methods can be designed which provide sufficient accuracy for reasonable sulfur balances.

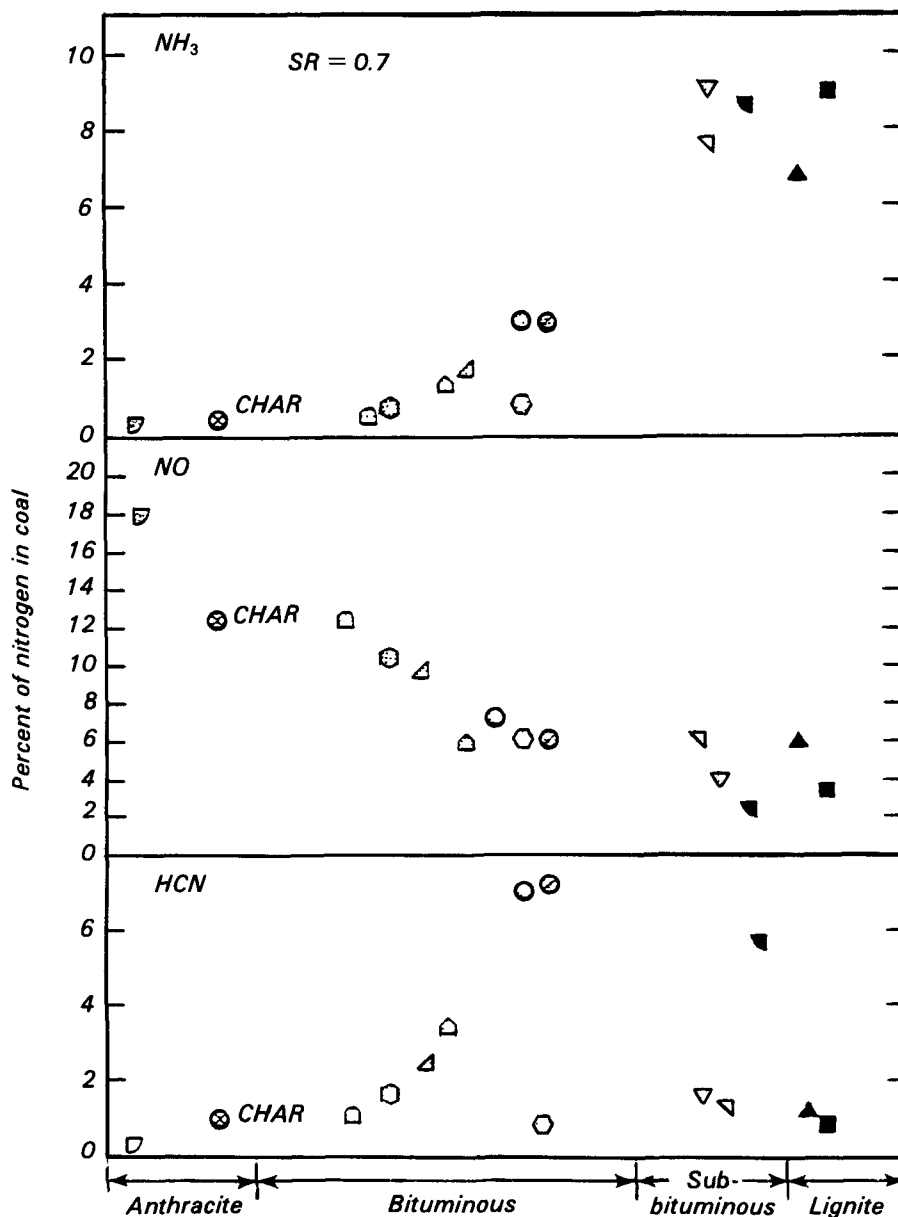


Figure 5 Influence of coal rank on gas-phase nitrogen species exiting first stage.

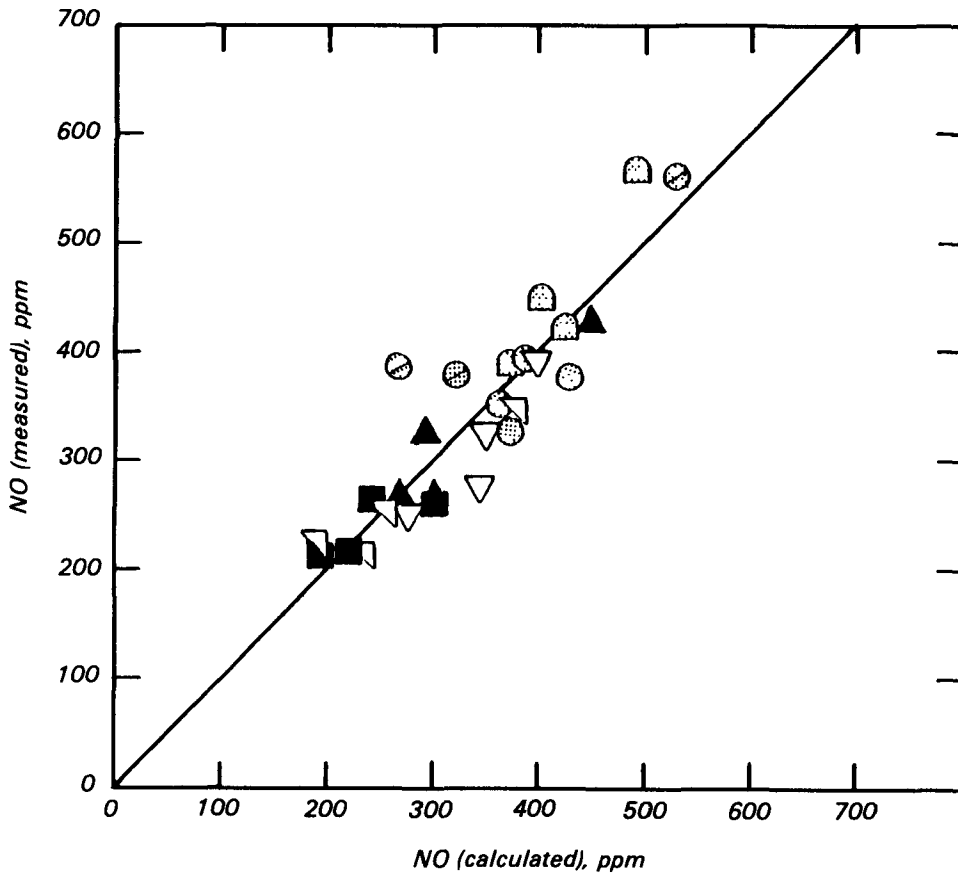


Figure 6. Exhaust NO can be calculated by $NO_{ex} = 0.8(SR)/(NO) + \frac{HCN + NH_3}{2 + 0.007 (HCN + NH_3)} + 0.2 (Char N) + 5.$

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The complete report, entitled "Development of Criteria for Extension of Applicability of Low-Emission, High-Efficiency Coal Burners: Third Annual Report," (Order No. PB 82-197 153; Cost: \$18.00, subject to change) will be available only from:

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