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# POLLUTANT CONTROL THROUGH STAGED COMBUSTION OF PULVERIZED COAL

## Phase I - Comprehensive Report

Interim Report for the
Period December 1975 - April 1976

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# ABSTRACT

A six lb/hr laboratory furnace was used to investigate the nitrogen oxide (NO) emissions resulting from the combustion of three types of pulverized bituminous coal (Colorado, Western Kentucky, Pittsburgh #8), a Montana Powder River subbituminous coal, and a coal char. Fuel NO, which was isolated by replacement of air with an appropriate synthetic oxidant, comprised more than seventy-five percent of the total NO emissions over a wide range of flame conditions. Fuel nitrogen oxidation was insensitive to temperature changes except at very high flame temperatures. Under equivalent combustion conditions, total NO emissions were only slightly dependent on coal composition, because fuel nitrogen conversion decreased with increasing nitrogen content. Coal char had fuel nitrogen conversions that, although appreciable, were lower than those of coal, and that were only slightly dependent on burner aerodynamics.

# ACKNOWLEDGEMENTS

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# 1. OBJECTIVE AND SCOPE

This report is a comprehensive account of the first of two phases of a research project the *overall* objective of which is to define the optimum application of staged combustion as a means of lowering Nitrogen Oxide emissions from pulverized coal firing. In order to achieve this overall goal, it is necessary to first investigate what factors are important in the formation of $NO_x$ under classical (unstaged) combustion conditions. The work described in this report focuses, therefore, on $NO_x$ emissions from self sustaining, pulverized fuel flames and addresses the following specific questions:

- What fraction of the total NO produced is the result of fuel nitrogen oxidation?
- What is the overall temperature dependence of the fuel NO mechanism?
- What is the effect of coal composition on fuel nitrogen conversion?
- What is the potential importance of char NO?

Future work will then consider those questions critical to the optimum application of two stage combustion:

- How rich should the primary zone be?
- How much heat should be extracted from the primary zone?
- Where and at what temperature should the second stage air be added?

In addition to answering the above critical questions, the proposed work provides technical insight into how the combustion/pyrolysis environment affects both the behavior of burning coal particles and the devolitalization of combustion products leaving the first (and second) stages. The results obtained can then be used to estimate the nature of problems associated with the utilization by combustion of high nitrogen Western coals, and the potential of solutions of such problems by combustion modifications.

# 2. SUMMARY AND CONCLUSIONS

The experimental investigation of $NO_x$ emissions from self sustaining pulverized fuel flames has been essentially completed. Baseline NO emission data on the effect of excess air, and other process variables are generally consistent in both magnitude and trend with those of other investigators. The results obtained from this facility should, therefore, have some general applicability to other pilot and full scale units, involving both front fired and tangentially fired configurations.

TOTAL NO emissions from pulverized coal flames increase with:

- Increased overall excess air
- Increased flame temperature
- Increased primary/secondary mixing
- Increased fuel nitrogen content

By replacing the combustion air with an $Ar/O_2/CO_2$ atmosphere it was possible to define the relative importance of the thermal and fuel NO mechanisms. The thermal NO emissions from pulverized coal combustion are of the same magnitude as thermal NO emissions from natural gas combustion *under similar combustion conditions*. In general THERMAL NO emissions:

- Increase markedly with increasing flame temperature
- Increase with increasing excess air
- Are not a strong function of coal composition

Fuel nitrogen oxidation is the principle source of NO emissions from pulverized coal combustion. In fact, fuel NO emissions account for at least seventy percent of the total NO emissions for all coals tested at all conditions. In general FUEL NO emissions:

- Increase markedly with increasing excess air.
- Are relatively insensitive to flame temperature over a broad range of practical interest.
- Increase slightly with increasing fuel nitrogen content. Under equivalent combustion conditions the percentage conversion, however, decreases as the nitrogen content increases.
- Are a function of composition parameters other than total bound nitrogen.
- Can be dramatically reduced by utilizing a fuel injector which slows the primary/secondary mixing.
- Increase markedly with substantial early mixing of the primary and secondary streams due to flame detachment (detachment means ignition is occurring at some point downstream of the fuel injector).

Combustion of coal char results in NO emissions which are low compared to coal and are essentially completely the result of fuel nitrogen oxidation. The data on the COED-FMC coal char suggests that CHAR NO:

- Has only a slight temperature dependence.
- Is essentially independent of fuel injector design (in strong contrast to total NO emissions from coal).
- Is approximately half the fuel NO produced from pulverized coal with equivalent bound nitrogen.

Since fuel NO emissions from coal are relatively dependent on flame fluid dynamics and char NO is not, fluid dynamics must play a key role in determining the conversion of volatile fuel nitrogen. For the volatiles, the local oxygen environment in the first few fractions of a second determine the percentage of fuel nitrogen converted to NO. Detached or lifted flames lead to very high volatile nitrogen conversions to NO, but do not affect char nitrogen conversion significantly. As far as pulverized coal is concerned, therefore, detached flames always lead to high NO emissions regardless of how flame detachment is achieved.

In terms of practical implications the results suggest that to obtain significant emission reductions with coal, control technology must markedly reduce conversion of fuel nitrogen to NO. This cannot be accomplished by reducing the flame temperature (as with flue gas recirculation), but burner design changes which tend to reduce the early mixing between the coal and secondary air will result in decreased NO emissions. (Excessive flame length and/or combustion instabilities must be avoided, however.)
Volatile NO appears amenable to abatement by combustion modifications but abatement of char NO may be extremely difficult. Thus unless the char/ volatile split can be altered, there may exist a lower limit on the emission level which can be achieved via combustion modifications.

# 3. COMBUSTION FACILITY

## 3.1 OVERVIEW

The combustion facility was designed to meet the following criteria:

- The furnace must contain the salient features of real combustion hardware, i.e. it must be capable of burning 75%-200 mesh coal in a swirling turbulent diffusion flame with inlet air velocities near 100 ft/sec and about 600°F preheat.

- It must be large enough to allow utilization of normal screw feeding technology to deliver the coal and to insure that the flame will be self sustaining. However, it must be small enough that synthetic oxidizer atmospheres can be supplied at reasonable cost using standard pressured cylinders.

- The facility must be flexible. In addition to a variety of pulverized coals, it should be suitable for studying the combustion of residual oils, solid (pulverized) wastes, low Btu gas, crude oil, char, oil shale, etc.

- It must be versatile so that only minor modifications are required to convert it from the classical tunnel chamber to one where the fluid dynamics are "well defined" so that the kinetically controlled $NO_x$ chemistry can be separated from the fluid dynamics.

- It must have variable wall temperature: hot walls to simulate the environment seen by an interior burner of a multi-burner array and cooled walls to study particle heating effects or simulate a package boiler system.

The design described in the following sections was developed from these criteria. In concept it is somewhat similar to that used by Beer and Thring (1960) to study pulverized fuel combustion rates.

## 3.2 FURNACE

The experimental furnace is illustrated in Figure 1. The vertical combustion chamber is 76" long and 6" in diameter inside. The overall outer

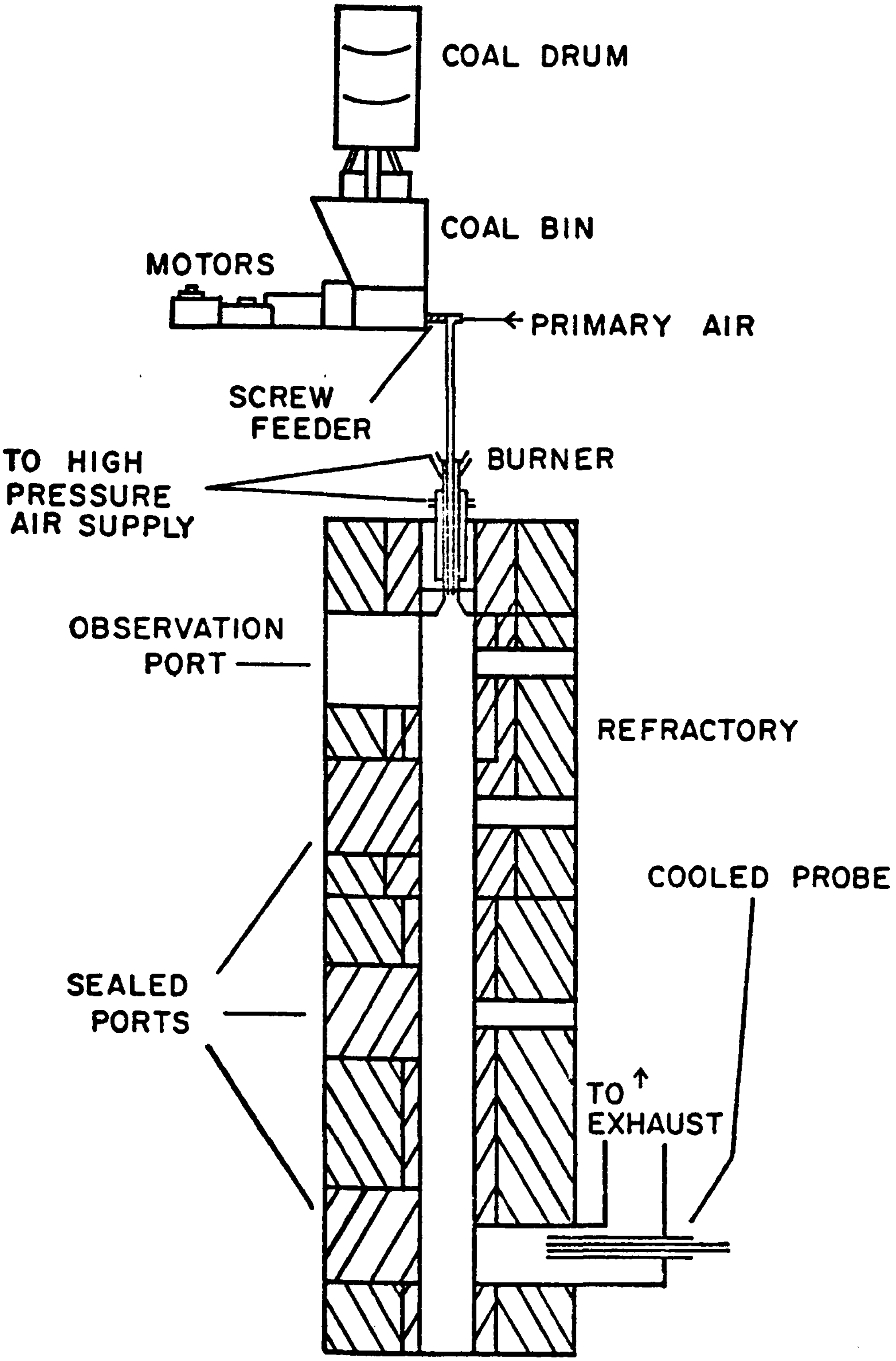

FIGURE 1. EXPERIMENTAL FURNACE

diameter is approximately 27". In the lower half of the furnace the walls consist of an outer steel shell, ¼" of roll board insulation, 8" of Harbison-Walker Lightweight 26 insulating castable (2600°F max. service temperature) and 2" of Harbison-Walker Castolast G 3200°F castable refractory. In the upper half of the furnace the walls consist of 6" of insulating castable and 4" of the high temperature Castolast G. This casting pattern was used so that the furnace would be capable of withstanding very hot combustion conditions (coal in argon/oxygen) and yet have minimal heat loss. As Figure 1 shows the upper portions of the inner refractory are removable so that a water cooled section of an alternate design can be installed. In future portions of the work the upper portion will be modified so that there is a well mixed zone followed by a plug flow zone.

At the full load firing rate of 85,000 Btu/hr (6.6 lbs coal/hr) the cylindrical combustion chamber provides a nominal residence time of approximately one second. This firing rate corresponds to a maximum combustion intensity of about 68,000 Btu/hr and is somewhat higher than originally planned. Initial experimentation revealed, however, that the flame was more stable and symmetric at this higher firing rate, probably because of a more uniform coal feed and a higher upper zone wall temperature.

There are four 6" wide x 10" long observation ports and three 2" diameter ports spaced down the length of the furnace for flame photography, visual observation, and optical wall temperature measurements.

Fuel and air enter the combustion chamber at the top via a water-cooled burner described in detail below. The combustion gases leave the furnace through a 6" diameter exhaust port and flow through approximately forty feet of air and water double pipe heat exchangers. After the exchangers the flue has a temperature of less than 300°F and is exhausted into a fume duct.

To start the system up initially an 18" Eclipse extended pilot is used. The pilot is positioned in a horizontal port in the refractory about 5" below the top of the furnace. It normally extends approximately half way through the refractory wall and is a long, internally spark ignited pilot burner. Upon ignition it produces a long horizontal flame directly across the outlet of the main burner. When the main gas is turned on it ignites immediately and the pilot system is turned off. Once the furnace walls are above approximately 1900°F the coal flame can be ignited directly via radiation and convection from the walls; in which case no pilot flame is required.

### 3.3 MULTIFUEL BURNER

The specially designed water-cooled burner is illustrated in Figure 2. It has separate axial and swirl air inlets and is similar to that used in previous studies by Pershing *et al.* (1975) The axial air enters through two ½" angled ports into the center pipe. Swirl air enters a vaned swirl chamber via two tangential ports 180 degrees opposed and passes through eight 0.100" curved swirl vanes as shown in Figure 3. The inside diameter of the burner itself is 1.38", however, several burner inserts were prepared so that the secondary air velocity (axial) could be maintained at 60 ft/sec for a variety of mass flows, air preheats, etc. (Thus when the inlet air temperature was increased from 530°R to 1060°R the burner throat area was also doubled to maintain a constant velocity.) The burner throat is water cooled and the exit

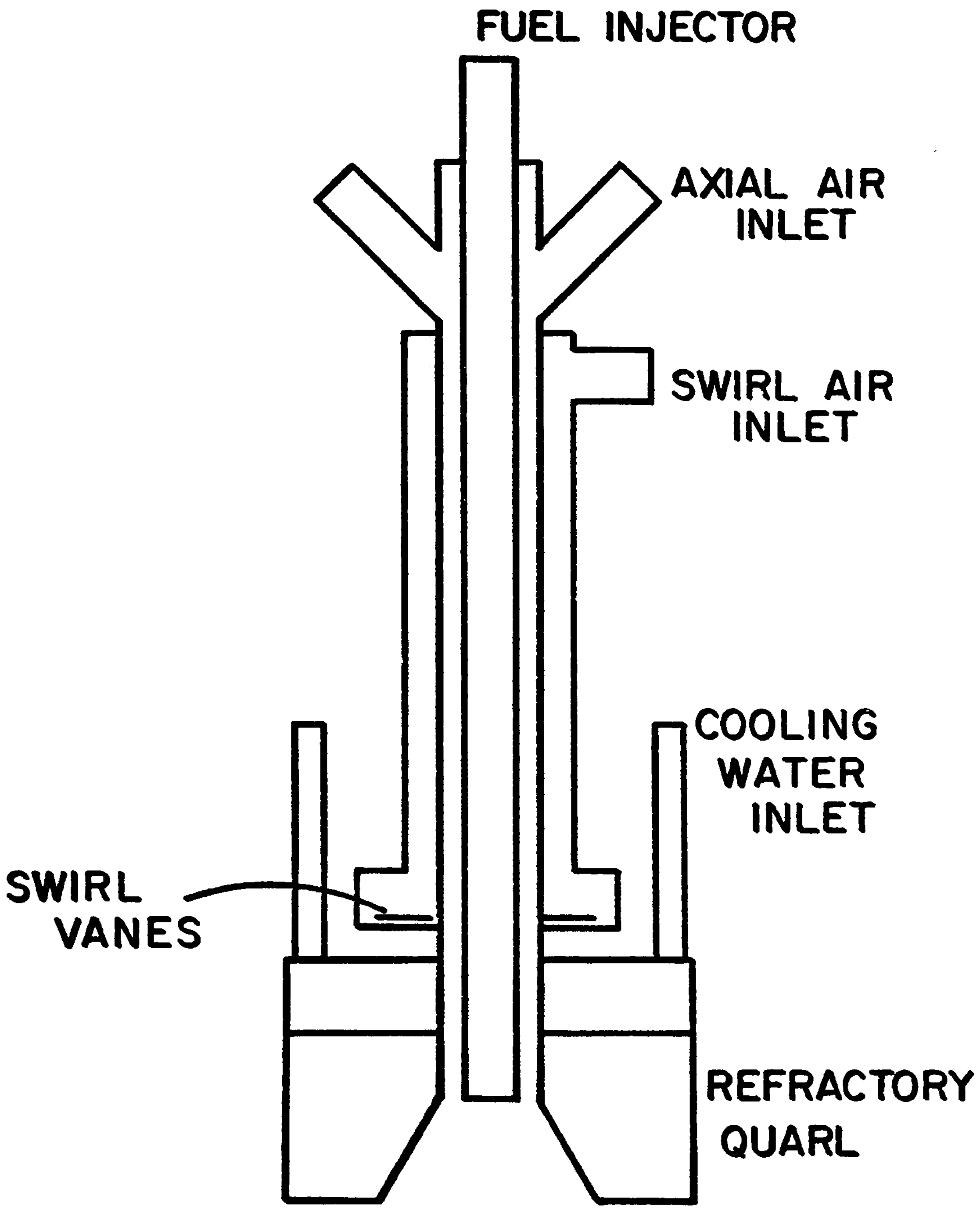

FIGURE 2. MULTI FUEL BURNER

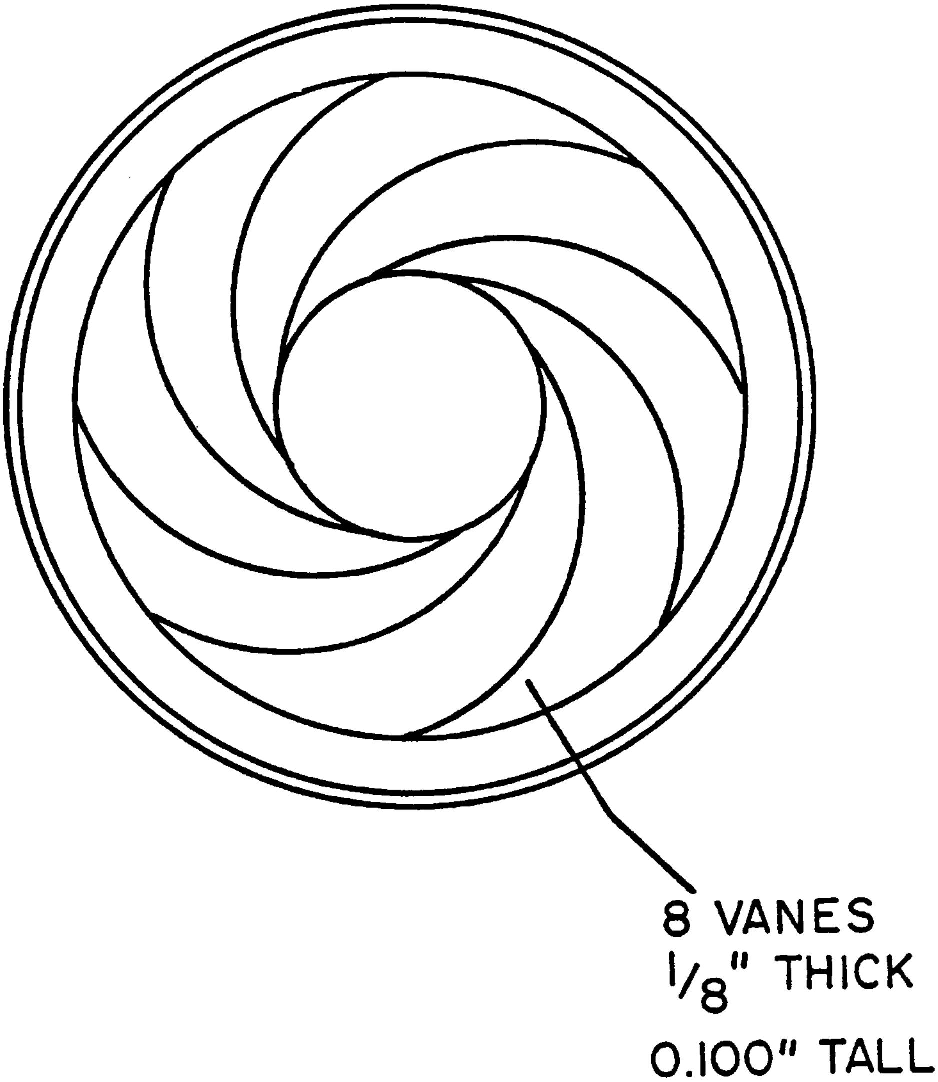

FIGURE 3. SWIRL VANES

is fitted with a 30 degree refractory (Castolast G) quarl that has an L/D ratio of 1. The top of the burner has a removable collar designed to accept a variety of fuel injectors for natural gas, fuel oils and pulverized coals.

## 3.4 FUEL INJECTORS

The fuel injectors used in this study are shown in Figure 4. Both injectors were fabricated from 3/4" stainless steel tubes with welded end plugs. The first contains three 11/64" holes angled to distribute the coal away from the axis of the furnace and is characterized as a rapid mixing injector because it produces a short bulbous flame. It was designed to be similar to the "coal spreader" system employed in many commercial systems. The second injector contains a single, 19/64" center hole with an area equal to that of the three holes in the divergent injector. It produces relatively slow mixing between the primary and secondary air streams and hence gives a long, very thin flame. The two injectors are thus somewhat representative of two different classes of coal combustion equipment - one with intense mixing common in wall fired units, the other with slow mixing common in tangentially fired units.

## 3.5 AIR SUPPLY SYSTEM

The air supply system for the furnace is shown in Figure 5. Under normal operating conditions a 100 psig air compressor provides the combustion air. After being filtered and partially dried, the air goes through two high volume regulators where the pressure is stepped down to approximately 30 psig. For special tests the air is enriched or replaced with varying amounts of carbon dioxide ($CO_2$), argon (Ar), and/or

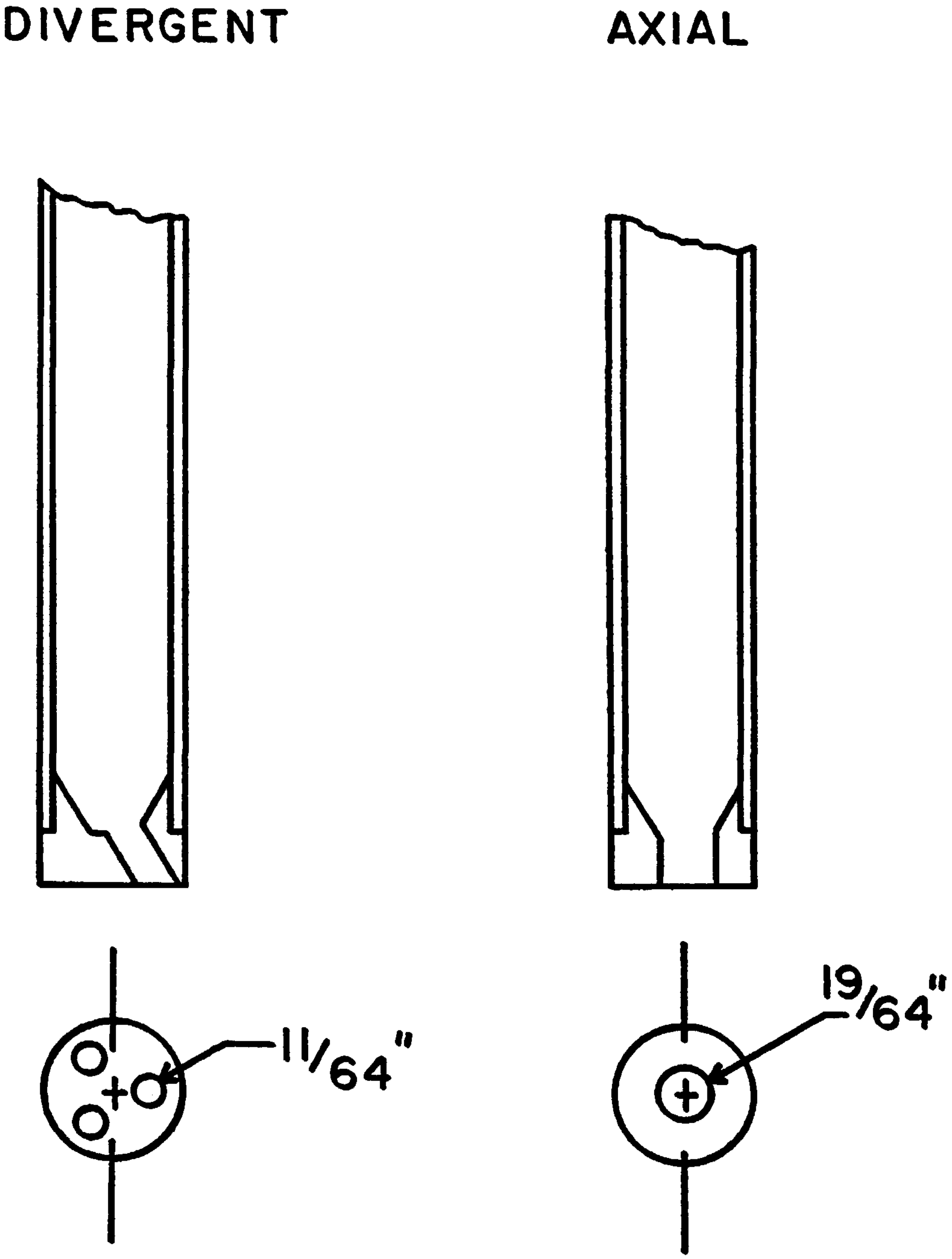

FIGURE 4. FUEL INJECTORS

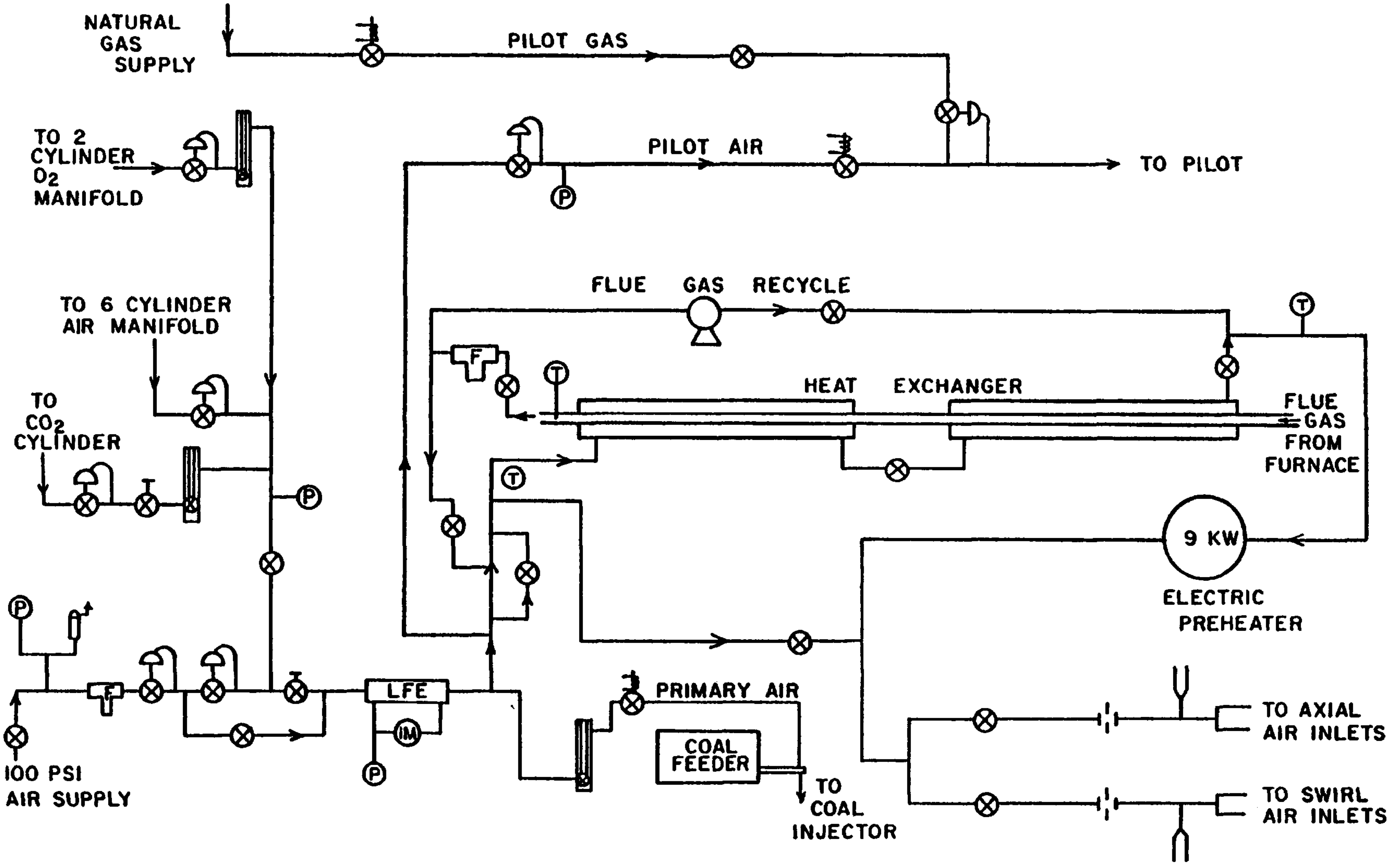

FIGURE 5. AIR SUPPLY SYSTEM

oxygen ($O_2$) all of which are supplied from 250 cubic feet high pressure cylinders. In each case there is an appropriate two-stage high volume regulator to step the pressure down to approximately 50 psig and a needle valve for fine control.

Once the pressure has been reduced the air (or artificial oxidizer mixture) goes through a 3/4" micro-needle valve for total flow control and a laminar flow element. The pressure drop across the laminar flow element is measured by a 10" $H_2O$ inclined manometer. In this way a relatively accurate measurement of the inlet oxidizer flow is obtained. Next the air is split into three separate streams: the pilot air, the primary air and the secondary air. On startup the pilot air goes through a regulator to step the pressure down to 10" $H_2O$ and then to an Eclipse blast pilot mixer where it is combined with the pilot gas stream. Once the main flame has been ignited, the pilot air is shut off.

The primary air stream is used to transport the coal from the screw feeder to the burner. The flow is controlled by a needle valve and is metered with a calibrated rotameter. The primary air system also contains a solenoid valve which closes automatically after a flame-out. At present the primary air is not preheated prior to the burner.

After the pilot and primary air streams are split off the remaining flow goes to the preheating system. Here the temperature is raised from 70°F to the desired level. This is accomplished by first passing it through the shell side of a double pipe heat exchanger. (The inner pipe contains the hot combustion gases from the furnace.) In this exchanger the air is preheated to approximately 300°F. The final preheating and temperature control is accomplished with a 9 Kw Chromalox circulation heater. The heater was constructed with a 304 stainless steel shell and incoloy elements so preheats up to 1000°F can be obtained.

When desired, filtered flue gas can be recycled from the outlet of the heat exchanger (~200°F) and added into the secondary air stream via an ejector prior to the heat exchanger or via a pump just prior to the electric preheater. In both cases the recirculated gases go through the electric preheater and enter the burner at the same temperature as the secondary air. Recirculation of flue gases is common industrial practice, and has been shown to be an effective $NO_x$ abatement measure for gas fired flames.

Once the secondary air has the desired temperature and composition it is split into axial and swirl air streams. The flow in each line is controlled with a high temperature globe valve and metered with a calibrated orifice. Two 0.036" exposed bead iron-constantan thermocouples just prior to the burner inlets are used both for measuring the temperature of each stream and for the input to the proportional temperature controller on the electric preheater.

## 3.6 FUEL DELIVERY SYSTEM

The coal delivery system was designed to be totally enclosed to minimize dust and safety problems. First the coal is loaded into small steel barrels (18" dia x 36" high) outside the facility. The barrels are then closed, brought inside and positioned above the feeder. After the appropriate connections are tightened, gate valves on the bottom of the barrels are opened and the coal flows into the feeder hopper.

The pulverized coal is metered with a twin-screw Acrison Model 105 feeder and the flow rate controlled with a mechanical variable speed drive. Flow problems were minimized by mounting the entire feeding system directly above the burner. Based on past experience and visual observation of the flow from the feeder, it was initially felt that coal pulsing was going to be a major problem. In an attempt to overcome this a variety of in-line mixing schemes were tried. Ultimately, however, the best approach proved to be a direct, vertical connection from the feeder to the fuel injector (no mixing device). Flow uniformity was further improved by operating the feeder at maximum rpm and by introducing the primary air as a high velocity air jet just opposite the screw outlet.

Natural gas is used for bringing the furnace up to temperature and maintaining thermal equilibrium when coal is not being fired. In order to obtain a stable lite-off on coal it is necessary that the wall temperature in the upper section of the furnace be above about 1900°F.

## 3.7 SAFETY/CONTROL SYSTEM

The furnace is equipped with an electrical interlock safety system to insure both safe startup and proper shut down in case of a variety of operational problems. The system was designed so that the facility could be operated without an attendant for long periods of time (nights, weekends, etc.) and thereby maintain thermal equilibrium. It monitors the outlet temperature and flow of the various cooling loops to insure system integrity. In the event of a poor or unstable flame for any reason it automatically shuts off all fuel flow. The system also monitors the inlet air and in the event of a compressor failure shuts down both the furnace and electric air preheater (to protect the incoloy elements from overheating.)

Before power is available for startup and operation the following electrical switches must be closed:

- Low pressure limit switch on the combustion air
- Remote shutdown toggle switch
- High temperature limit on the burner cooling water
- High temperature limit on the window cooling water
- Flow switch on the main cooling water
- Main power switch
- High temperature limit with sensor positioned above burner
- High temperature limit on flue cooling water

Once these switches are all closed (as indicated by a series of green lights on the control panel) startup can be initiated. The natural gas system consists of the following control components:

- Total gas solenoid valve (Maxon) which requires manual opening *after* it is activated electronically

- Pilot gas ball valve

- Pilot air solenoid valve-interlocked with the ignition transformer so that ignition cannot be attempted without pilot air flow.

- Main gas control valve

- Main gas solenoid valve

- Ignition switch and high voltage transformer.

The coal delivery system has a similar set of control components:

- Main coal feeder switch

- Primary air solenoid valve

- Coal feeder start relay and push-button

For both fuels, the systems are designed so that in the event of a shutdown for any reason, the operator must manually either reopen the gas valve or push the feeder start button again before the fuel flow will begin.

The coal, main gas, and pilot gas flames are all monitored by a Honeywell ultraviolet flame detector. In the event of a flame-out (or very poor flame) this flame detector shuts the system down. (During startup the UV sensor system is by-passed electronically.)

## 3.8 ANALYTICAL SYSTEM

The analytical system was designed so that continuous monitoring of NO, $NO_2$, CO, $CO_2$, $O_2$, and $SO_2$ could be achieved. Future work will involve, in addition, measurement of $NH_3$, XN, and solids loading and composition. Figure 6 shows a schematic of the sampling and analysis system. The flue gas is withdrawn from the stack through a 3/8" water-cooled stainless steel probe. During the initial shakedown testing the water-cooled stainless steel probe was compared with both cooled and uncooled quartz probes. No difference in the measured NO was noted, even with CO and unburned carbon present. It should be noted, however, that the flue gas has cooled to below 1000°F at the point of sampling and there is always at least 0.5% oxygen present in the sample.

Sample conditioning consists of a refrigerated dryer (water condenser), two glass wool filters, a 60µ stainless steel filter (50°F), a stainless/teflon sampling pump and a 7µ stainless filter. All sample lines are ¼" teflon and all fittings 316 stainless steel.

The analysis system consists of the following equipment:

- Beckman Model F3 Paramagnetic Oxygen Analyzer
- Beckman Model 864 Nondispersive Infrared CO Analyzer
- Beckman Model 864 Nondispersive Infrared $CO_2$ Analyzer
- Thermo Electron Model 40 Pulsed Fluorescent $SO_2$ Analyzer
- Beckman Model 715 Polarographic Oxygen Analyzer
- Thermo Electron Model 10AR Chemiluminescent NO-$NO_x$ Analyzer with Model 300 Molybdenum Converter.

All instruments are calibrated with zero and span gas at least every three hours.

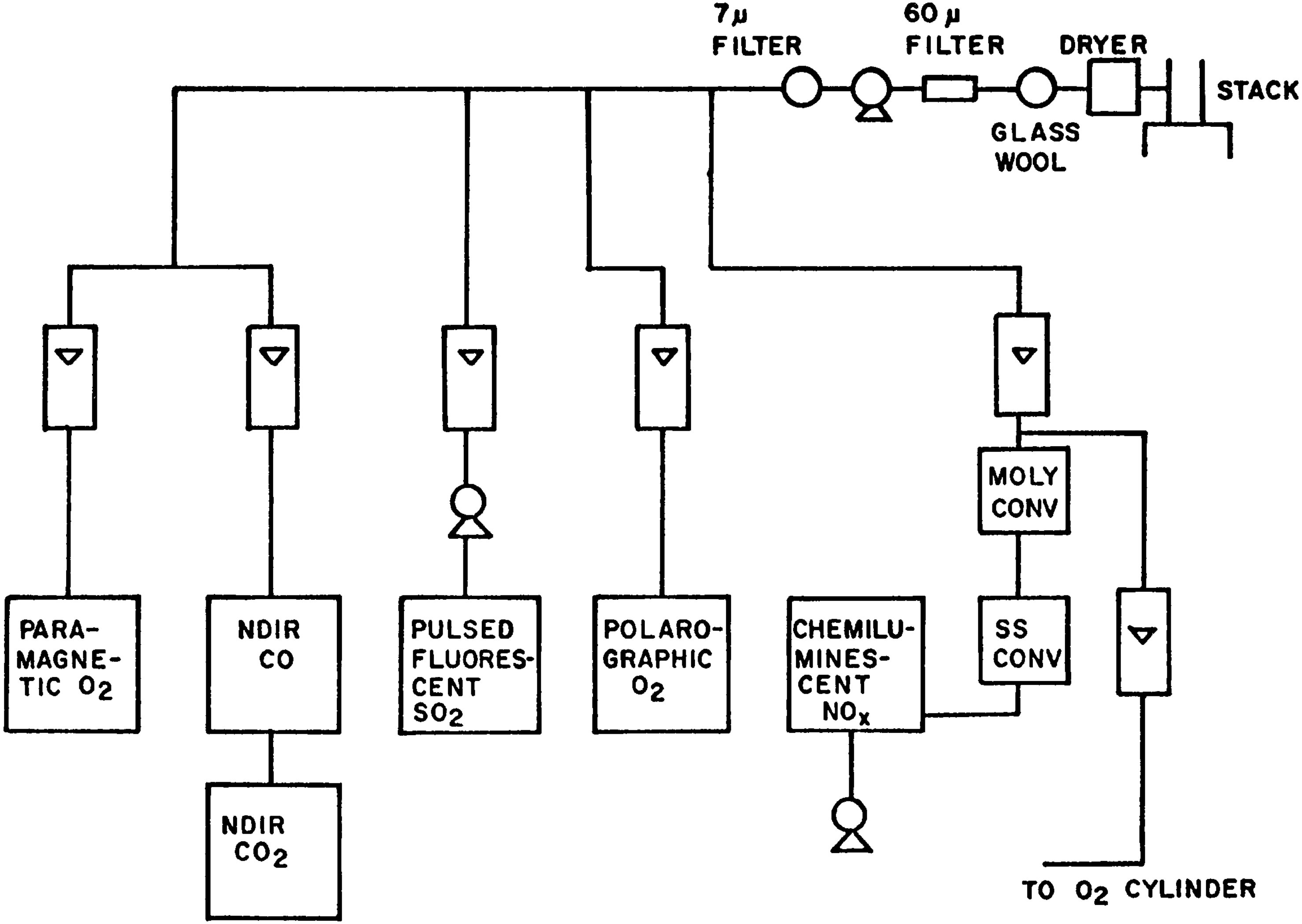

FIGURE 6. ANALYTICAL SYSTEM

Future efforts will involve design of a direct water spray quenched probe for sampling gases and particulate matter from hot furnace environments. Such a probe has been used successfully by researchers at the International Flame Research Foundation (Heap *et al.* 1973), the Massachusetts Institute of Technology (Bittner *et al.* 1975) and Brigham Young University (Horton *et al.* 1975).

# 4. EXPERIMENTAL RESULTS

## 4.1 INTRODUCTION

### 4.1.1. Definition of Terms

Before considering the actual experimental results, it is important to explicitly define the terminology used. Stoichiometric ratio (SR) is the ratio of the inlet oxygen to the amount of oxygen needed to completely burn the fuel to $CO_2$ and $H_2O$. Hence a stoichiometric ratio greater than 1.0 is fuel lean while one less than 1.0 is fuel rich. Excess air is the industrial term for defining the stoichiometric ratio; it is a measure of the amount of inlet oxygen which is in excess of that required for complete combustion and is equal to (SR-1.0)*100%.

In general all of the emission data, unless otherwise noted are reduced to stoichiometric conditions (STOICHI), i.e., they are corrected for dilution by excess combustion air. In particular the NO emission data (PPM NO, STOICHI) are presented as parts per million NO, by volume, dry, reduced to stoichiometric.

Primary air is that air used to transport the pulverized coal from the screw feeder to the furnace. As such, it is premixed with the coal prior to the burner and enters the combustion chamber through the fuel injector. Primary percentage and primary stoichiometry refer to the percent of the stoichiometric air requirement which is used as primary air.

Secondary air is that air not premixed with the coal prior to the burner. As previously described, it enters the combustion chamber through axial ports or tangential swirl vanes in the burner. The percent swirl is defined as the ratio of the volumetric flow of air introduced through the swirl vanes to the total secondary air volumetric flow rate (swirl plus axial) times one hundred percent.

*Air preheat* refers to the temperature of the secondary air. The primary air is not preheated prior to the burner.

The *secondary air velocity* is the axial velocity component of the *total* secondary air in the burner throat. Note that the axial velocity does not vary with the swirl percentage because it is based on the total seconary air going through the burner throat.

Flue gas recirculation (FGR) is define as:

$$\frac{\text{std. ft}^3\text{ recirculated}}{\text{std. ft}^3\text{ inlet air} + \text{std. ft}^3\text{ recirculated}} \times 100\%$$

and is only added to the secondary air.

Finally, inlet "air", secondary "air", etc. are used in a general sense to refer to the incoming oxidizer streams. At the baseline conditions, the oxidizer streams are truly air (21% $O_2$, 79% $N_2$) from the high pressure air compressor. However, as noted later, during certain tests, the inlet "air" was really a synthetic oxidizer containing $O_2$, $N_2$ or Ar, and perhaps $CO_2$, NO, or $NH_3$. *Oxygen enrichment* refers specifically to tests in which pure oxygen was added to the compressed room air to increase the inlet oxygen percentage. Ar/$O_2$/$CO_2$ *replacement* refers to tests in which the compressed room air was completely shut off and the furnace operated with a synthetic oxidizer containng argon, oxygen and perhaps carbon dioxide.

### 4.1.2 Fuel Analysis

The compositions of the four solid fuels used in this study are given in Table 1. The Colorado coal is the same coal used by Armento (1975). The Colorado, Pittsburgh #8, and Western Kentucky are all medium volatile bituminous coals while the Montana-Powder River Region coal is a subbituminous containing significant moisture. The coal char originated from the FMC-COED coal gasification process.

Figure 7 shows the particle size distribution of the fuels used in this study. Figure 8 is a cumulative size distribution plot and indicates that while all of the coals were pulverized to the normal industrial standard of approximately 75% - 200 mesh, the Colorado coal contained somewhat less fines than the others. The "pulverized char" was only 50% - 200 mesh but it did contain approximately as many fines as the coals. The unpulverized char (as received) was only 25% - 200 mesh.

TABLE 1. PULVERIZED FUEL COMPOSITIONS

| | COLORADO | PITTSBURGH #8 | WESTERN KENTUCKY | MONTANA-POWDER RIVER REGION | FMC COAL CHAR |
|---|---|---|---|---|---|
| ULTIMATE ANAYSIS (%,DRY) | | | | | |
| C | 73.1 | 77.2 | 73.0 | 67.2 | 72.8 |
| H | 5.1 | 5.2 | 5.0 | 4.4 | 0.9 |
| N | 1.16 | 1.19 | 1.40 | 1.10 | .99 |
| S | 1.1 | 2.6 | 3.1 | 0.9 | 3.5 |
| O | 9.7 | 5.9 | 9.3 | 14.0 | .7 |
| Ash | 9.8 | 7.9 | 8.2 | 11.7 | 21.2 |
| HEATING VALUE (BTU/LB,WET) | 12,400 | 13,700 | 12,450 | 8,900 | |
| PROXIMATE ANALYSIS (%,WET) | | | | | |
| VOLATILE | 38.9 | 37.0 | 36.1 | 30.5 | 3.6 |
| FIXED CARBON | 52.6 | 54.0 | 51.2 | 39.0 | 73.8 |
| MOISTURE | 3.3 | 1.2 | 4.8 | 21.2 | 1.8 |
| ASH | 8.9 | 7.8 | 7.8 | 9.2 | 20.8 |

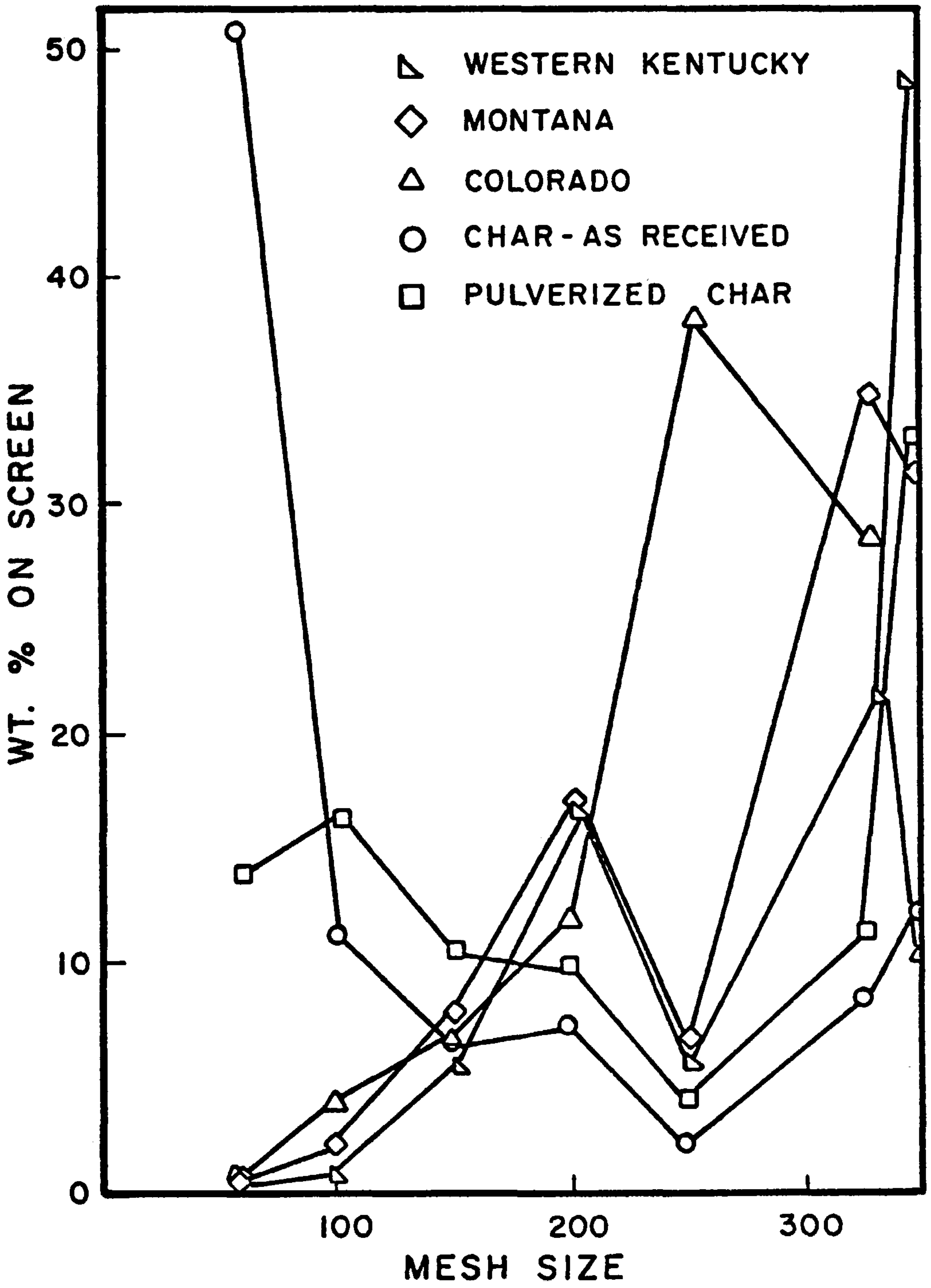

FIGURE 7. PARTICLE SIZE DISTRIBUTION

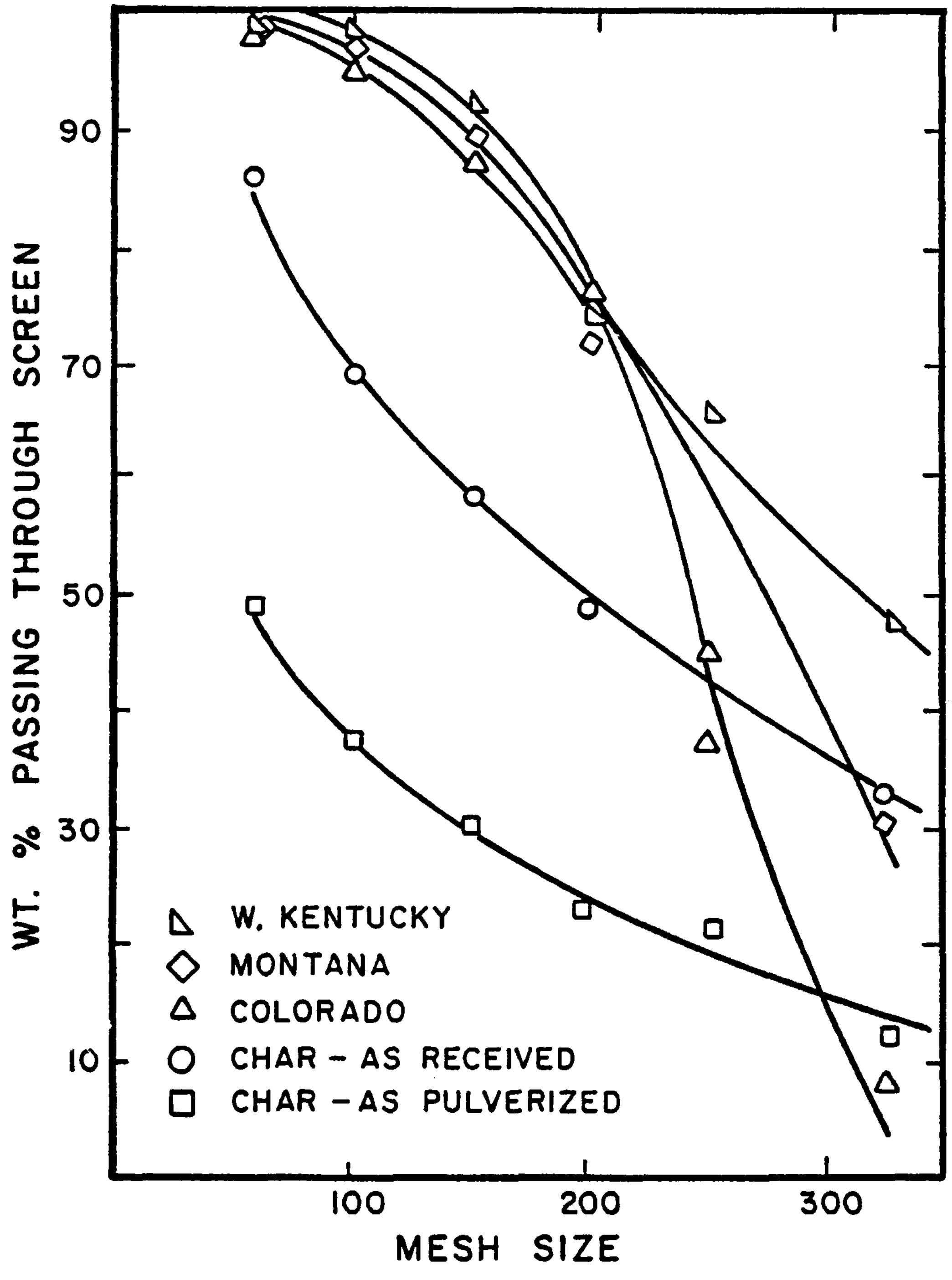

FIGURE 8. CUMULATIVE PARTICLE SIZE DISTRIBUTION

## 4.2 $NO_x$ CHEMISTRY

### 4.2.1 Oxygen Dependence - Total $NO_x$

Figure 9 shows the effect of excess air on NO emissions under "baseline" operating conditions for the Western Kentucky coal:

- full load - 5.9 lb coal/hr
- 44% swirl
- 650°F secondary air preheat
- 14% primary air
- 60 ft/sec secondary air velocity
- divergent coal injector

Ten sets of data were taken over a six month testing period to provide a measure of the reproducibility of the data. NO emissions increased with increasing excess air as previously reported by other investigators (Heap *et al.* 1973, Crawford *et al.* 1975, Armento 1975).

CO emissions were also measured during these tests and in general they were at or below the detection limit of the analyzer (400 ppm). The low CO levels and the good agreement obtained between the $O_2$ and $CO_2$ analysis and the measured fuel and air inputs indicate that the coal was being completely burned under the baseline conditions. Below about 7% excess air, however, the CO emissions start to increase and by 2% excess air there was approximately 0.5% CO in the flue.

$NO_2$ measurement were made at a limited number of conditions. Levels ranged from 5 to 25 ppm $NO_2$ (STOICHI) and were in general less than 5% of the total $NO_x$ emission.

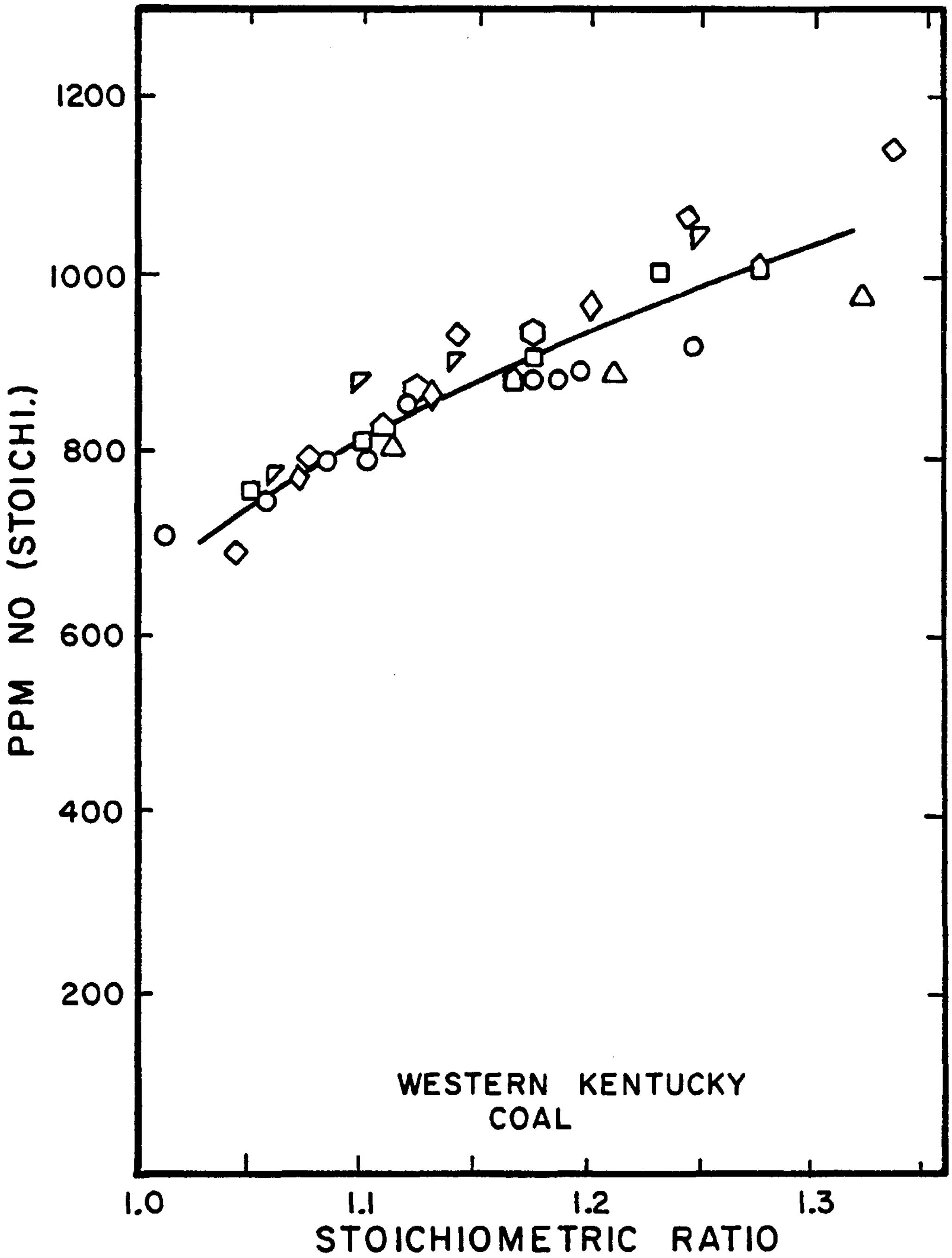

FIGURE 9. BASELINE DATA - EXCESS AIR (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR, 650°F PREHEAT)

Figure 10 shows the baseline data for the Colorado and Western Kentucky coals with the divergent injector plotted with field test results from nine wall-fired utility boilers. (Note that the NO emission data have been converted to a 3% $O_2$ basis since this is the usual point of reference for field testing results.) The divergent injector was designed to be similar to the "coal spreader" system employed in many wall-fired units. Figure 10 indicates that while there is considerable variation in actual field emission levels, the data reported herein on an 85,000 Btu/hr laboratory furnace with a divergent injector are consistent in both magnitude and trend with full scale data.

Figure 11 presents the baseline data for the Western Kentucky coal with the axial fuel injector along with the field test results (Crawford *et al.* 1974, 1975; Lachapelle, 1976) on six tangentially-fired field boilers. The axial fuel injector was designed to produce the relatively slow mixing between the primary air/coal stream and the secondary air stream which is characteristic of tangentially-fired units. As Figure 11 indicates, the data obtained in this study have the proper excess air dependence although the absolute emission levels are slightly higher than those obtained from most field units. Thus, it appears that the axial fuel injection system may provide a viable methodology for sub-scale simulation of $NO_x$ formation in a slowly mixed boiler.

In an attempt to further quantify the importance of inlet and local oxygen concentration, the effect of changes in oxygen concentration in the primary "air" was investigated. Figure 12 shows the results of testing where the primary oxygen was varied from 0% to 20% of the

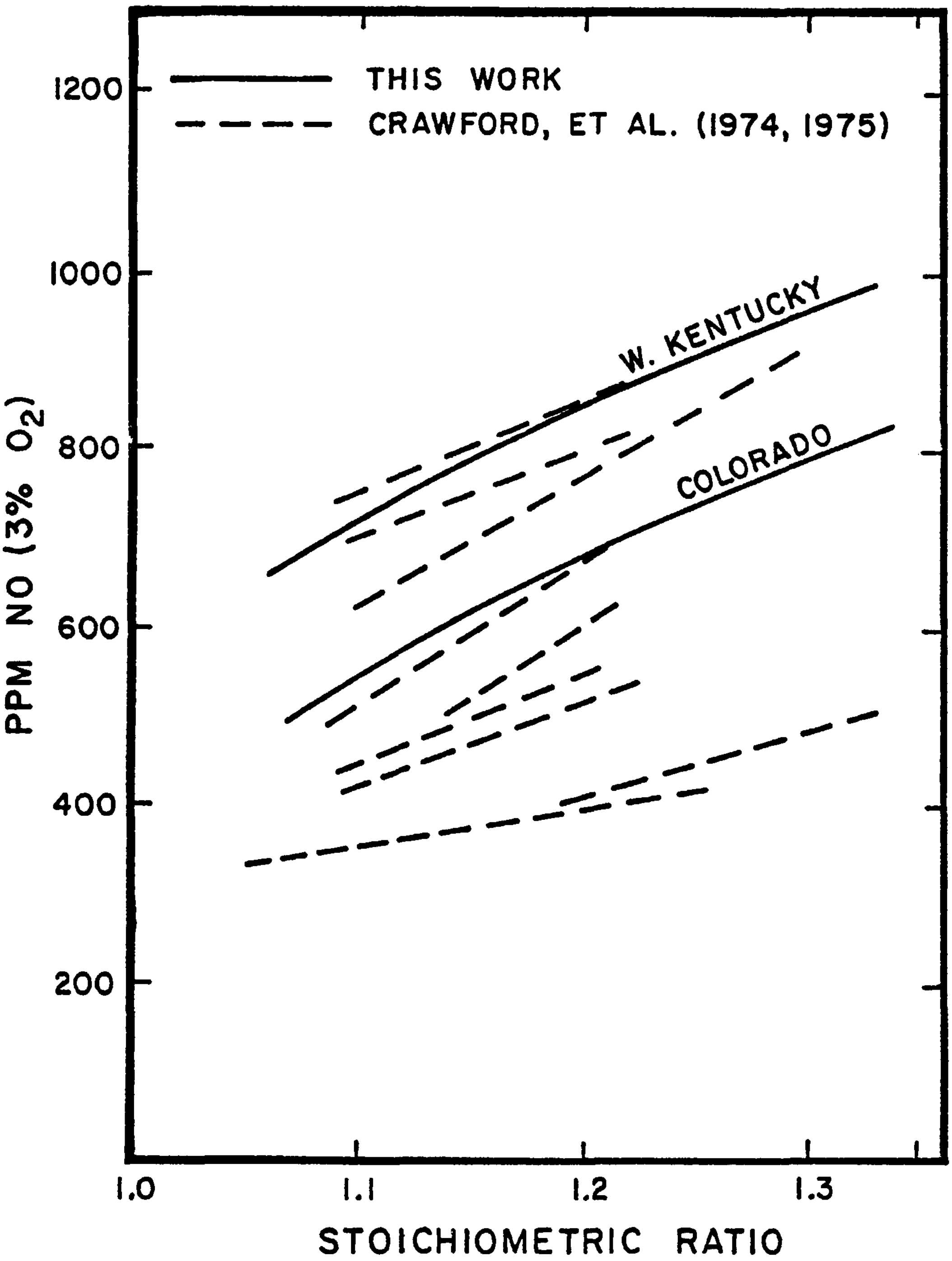

FIGURE 10. COMPARISON WITH FIELD DATA - WALL FIRING

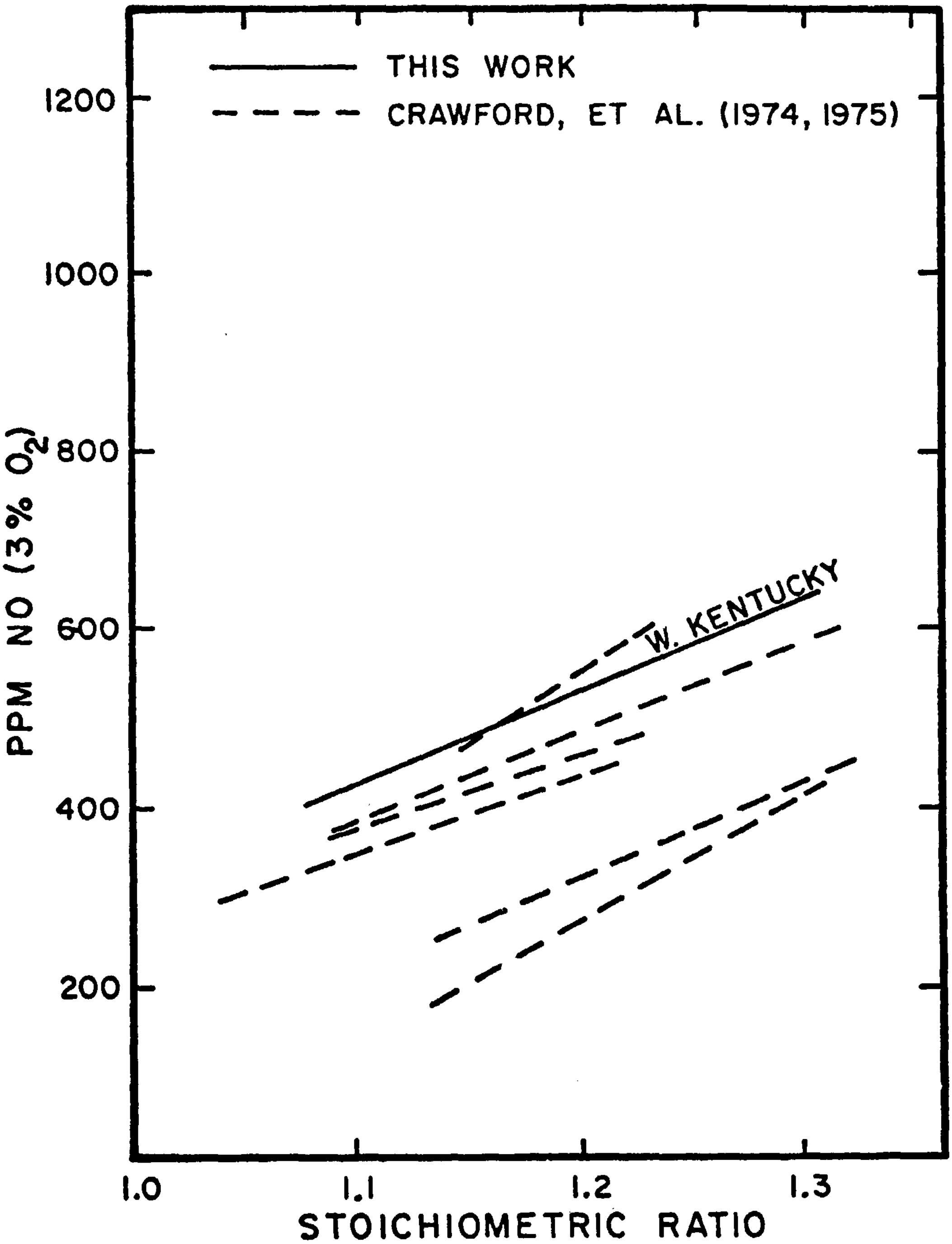

FIGURE 11. COMPARISON WITH FIELD DATA - TANGENTIAL FIRING

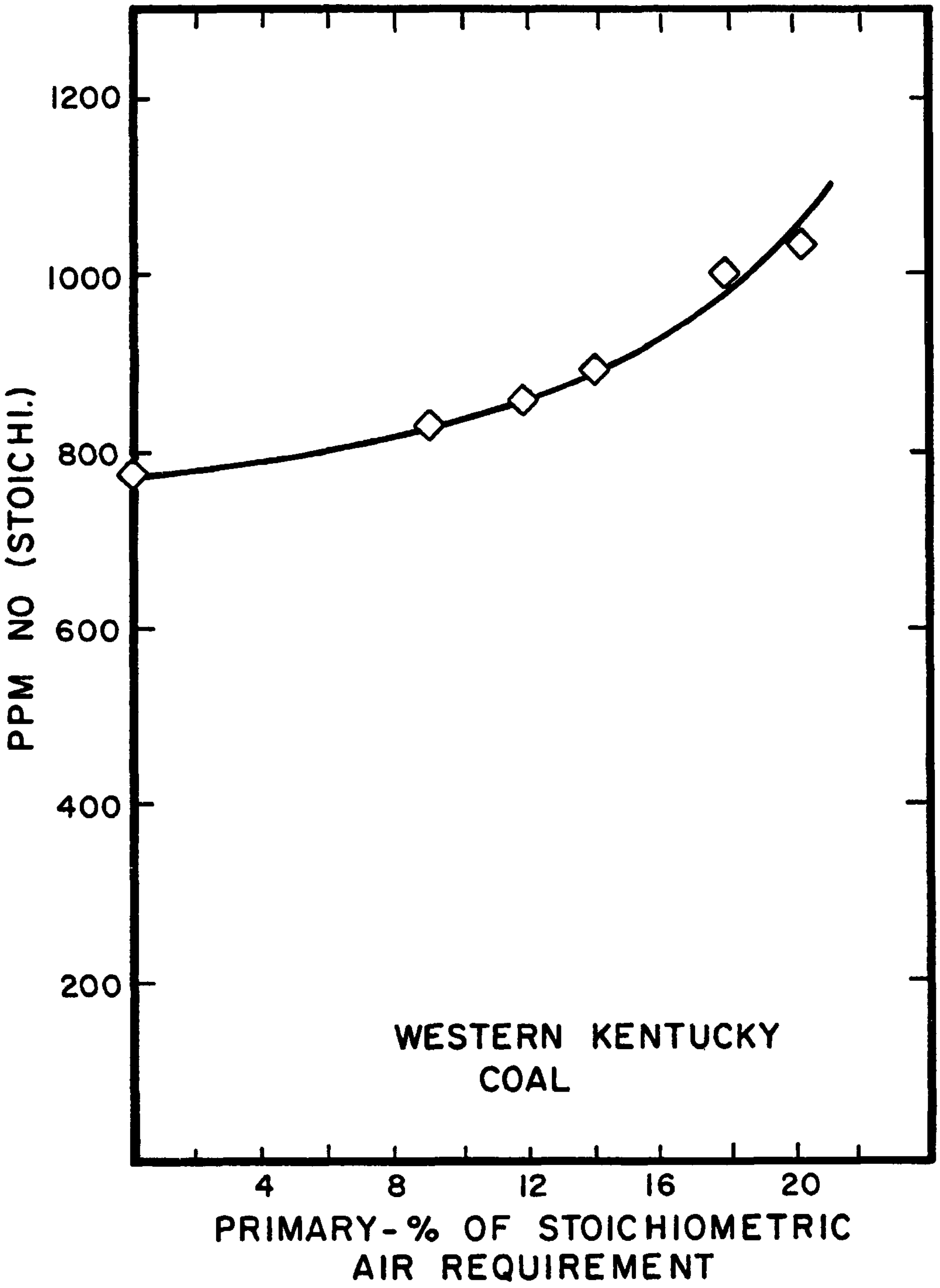

FIGURE 12. EFFECT OF INLET $O_2$ CONCENTRATION IN PRIMARY "AIR" AT A CONSTANT PRIMARY VELOCITY (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR, 15% EXCESS AIR, 620°F PREHEAT)

stoichiometric oxygen requirement by adding either $CO_2$ or pure $O_2$ to the primary stream. At the 0% condition the coal was being conveyed by pure $CO_2$. In all cases the volumetric flow (and hence primary jet velocity) was maintained constant 1.8 SCFM (62 ft/sec) to minimize changes in the flame fluid dynamics. The data indicate that with the divergent injector primary oxygen has little effect on total NO emissions until it is increased beyond approximately 15% of the stoichiometric requirement. The relatively small dependence may be the result of the divergent fuel injector design which causes the coal to come into rapid contact with the *secondary* air. In contrast to NO emissions, however, flame stability and ignition characteristics were significantly affected by inlet $O_2$ concentration changes in the primary air. This may indicate those very early volatiles necessary for ignition do not contain appreciable fuel N ultimately converted to NO, however, the subsequent volatiles, evolved during mixing with secondary air, do contain appreciable fuel N of which a portion is converted to NO. Further, the rate at which oxygen is supplied, through mixing, to these subsequent volatiles is of critical importance. The fluid dynamic aspects of these effects are discussed in Section 4.3.

In summary, the tests examining oxygen dependence indicated the following:

- It is possible to use a subscale experimental furnace to approximate the salient features of $NO_X$ formation during pulverized coal combustion in both wall-fired and tangentially-fired boilers.

- Increasing overall excess oxygen increases NO emissions from both types of combustion equipment.

- Below a certain level, further reduction of primary oxygen has little effect on NO emissions perhaps because the major portion of fuel nitrogen volatilized does so as it is mixed with the secondary air.

### 4.2.2 Importance of Fuel Nitrogen

It was once thought that nitrogen oxide emissions from residual oil and coal combustion were totally due to the fixation of atmospheric nitrogen. Martin (1972) and a variety of other investiagors have shown conclusively, however, that the conversion of nitrogen chemically bound in the fuel can be significant under many conditions. Sarofim *et al.* (1975), DeSoete (1973), Axworthy (1975), Sawyer (1975), and Merryman and Levy (1975) have studied the oxidation of typical nitrogenous compounds in laboratory flames and found the fuel NO conversion to be very sensitive to oxygen availability but only mildly dependent on temperature. Sternling and Wendt (1972) have considered the fate of chemically bound nitrogen during pulverized coal combustion and concluded that a major portion of the nitrogen may appear in the coal char. Recent experimental work by Pohl and Sarofim (1975) supports this conclusion and indicates that the division of the nitrogen between the char and volatiles is dependent upon particle heating rates. Pereira *et al.* (1974) found that in a laboratory fluid bed combustor most of the NO emissions resulted from oxidation of nitrogen in the coal and above 800°C the fuel NO was largely formed from the volatile combustion.

Thus while the potential of fuel NO is well established and considerable work is underway to elucidate the relevant mechanism(s), its absolute importance in boilers and furnaces is not yet know. In a recent definitive study Habelt and Howell (1976) calculated fuel nitrogen conversions based on a detailed experimental/theoretical analysis of thermal No formation in a full scale tangentially coal-fired steam generator. Their results indicate that percent conversions range from 0 to 30% depending upon the excess air. Based on a study of the influence of burner parameters on NO formation, Heap *et al.* (1973) postulated that fuel NO accounts for the major portion of the $NO_x$ formed during the combustion of pulverized coal. Pershing *et al.* (1975) later partially verified this hypothesis by burning pulverized coal in both air and argon/oxygen. This work was not absolutely definitive, however, because of the increased temperature associated with the argon/oxygen flames. A series of experiments were, therefore, undertaken to establish the absolute importance of fuel NO under practical combustion conditions.

First a methodology for determining fuel and thermal NO was established. By comparing the NO emissions at a particular set of operating conditions to those from the same fuel burning in an artificial atmosphere containing no $N_2$ it is in principle possible to establish the fuel and thermal NO contributions. In this study a synthetic oxidizer atmosphere containing 21% $O_2$, 18% $CO_2$ and the balance Ar was used because as Figure 13 indicates in addition to being free of $N_2$ it allows matching of theoretical flame temperature between the air and $Ar/O_2/CO_2$ cases. The $Ar/O_2/CO_2$ replacement method does, however, suffer from at least three potential weaknesses:

- Addition of $CO_2$ could have a chemical effect and hence change the NO kinetics.

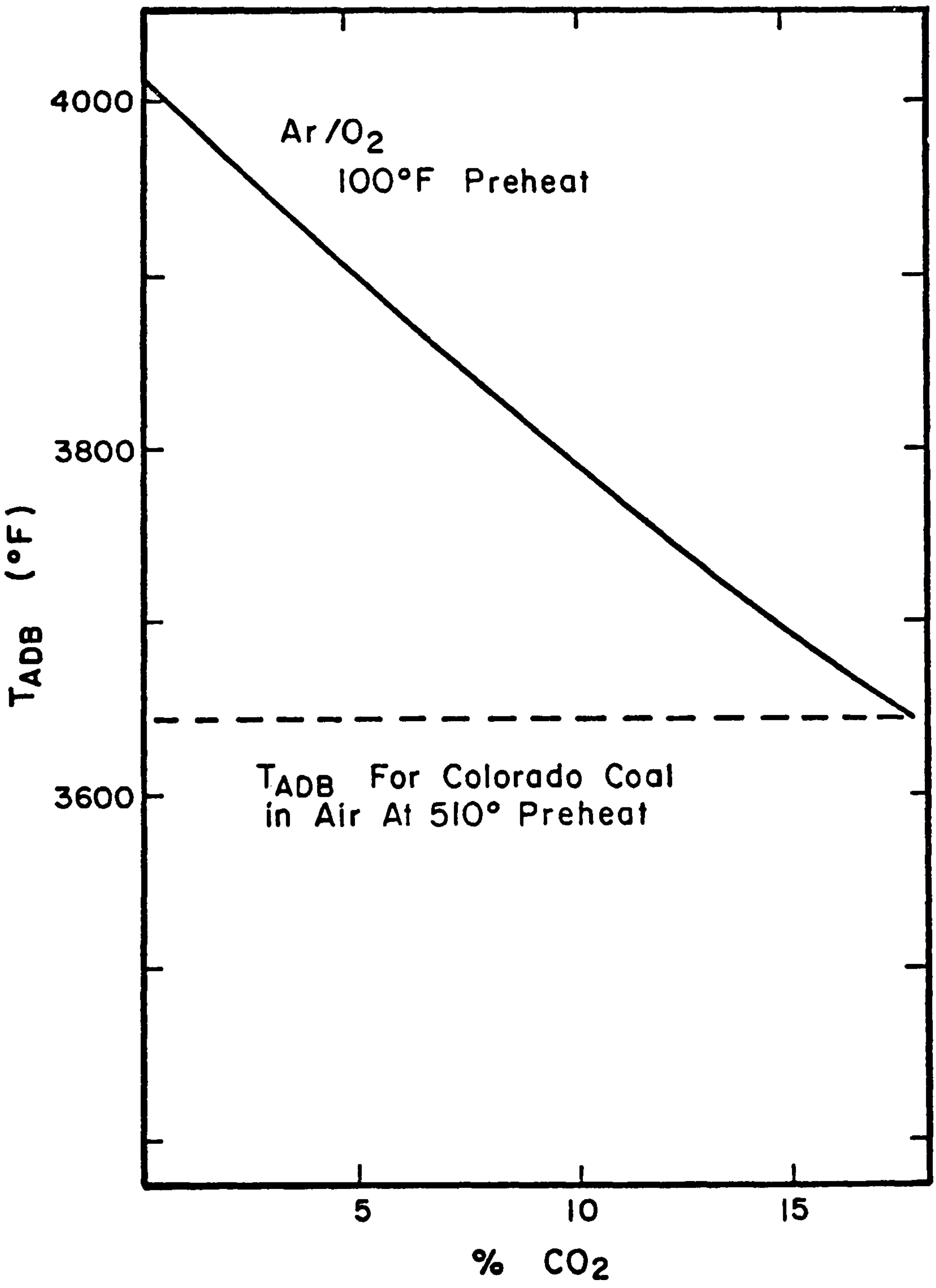

FIGURE 13. THEORETICAL FLAME TEMPERATURES FOR AIR AND $Ar/O_2/CO_2$ SYSTEMS

- Since the Ar, $O_2$ (and later $CO_2$) are commerical grade they could contain significant impurities (e.g. $N_2$ or NO) which would invalidate the results.

- If the furnace were not either leak tight or under positive pressure everywhere there could be significant $N_2$ present from the room air.

To evaluate the first of these potential problems a test series was conducted in which theoretical temperature was maintained with and without 18% $CO_2$ in the inlet air stream. Figure 14 shows these results and demonstrates that the presence of small amounts of $CO_2$ in the inlet air does not have any chemical effect on NO formation.

To evaluate the possibility of contamination of the Ar, $O_2$ and $CO_2$ and to demonstrate that there were no air leaks in the system, tests were run with 2-propanol and distillate oil. Under all conditions the emissions with $Ar/O_2/CO_2$ were less than 12 ppm, the bulk of which is probably due to the nearly complete oxidation of the small amount of fuel nitrogen in the distillate oil (Martin and Berkau, 1972). Thus it appears that $Ar/O_2/CO_2$ replacement is a valid methodology for determining fuel NO emissions.

Data on fuel and thermal NO emissions as a function of excess air are shown in Figure 15 for the Western Kentucky coal with both the divergent and axial fuel injectors. Thermal NO is defined as the difference between total NO and fuel NO, on the assumption that thermal fixation of atmospheric nitrogen does not inhibit fuel nitrogen conversion. The divergent injector data (650°F preheat, 45% swirl,

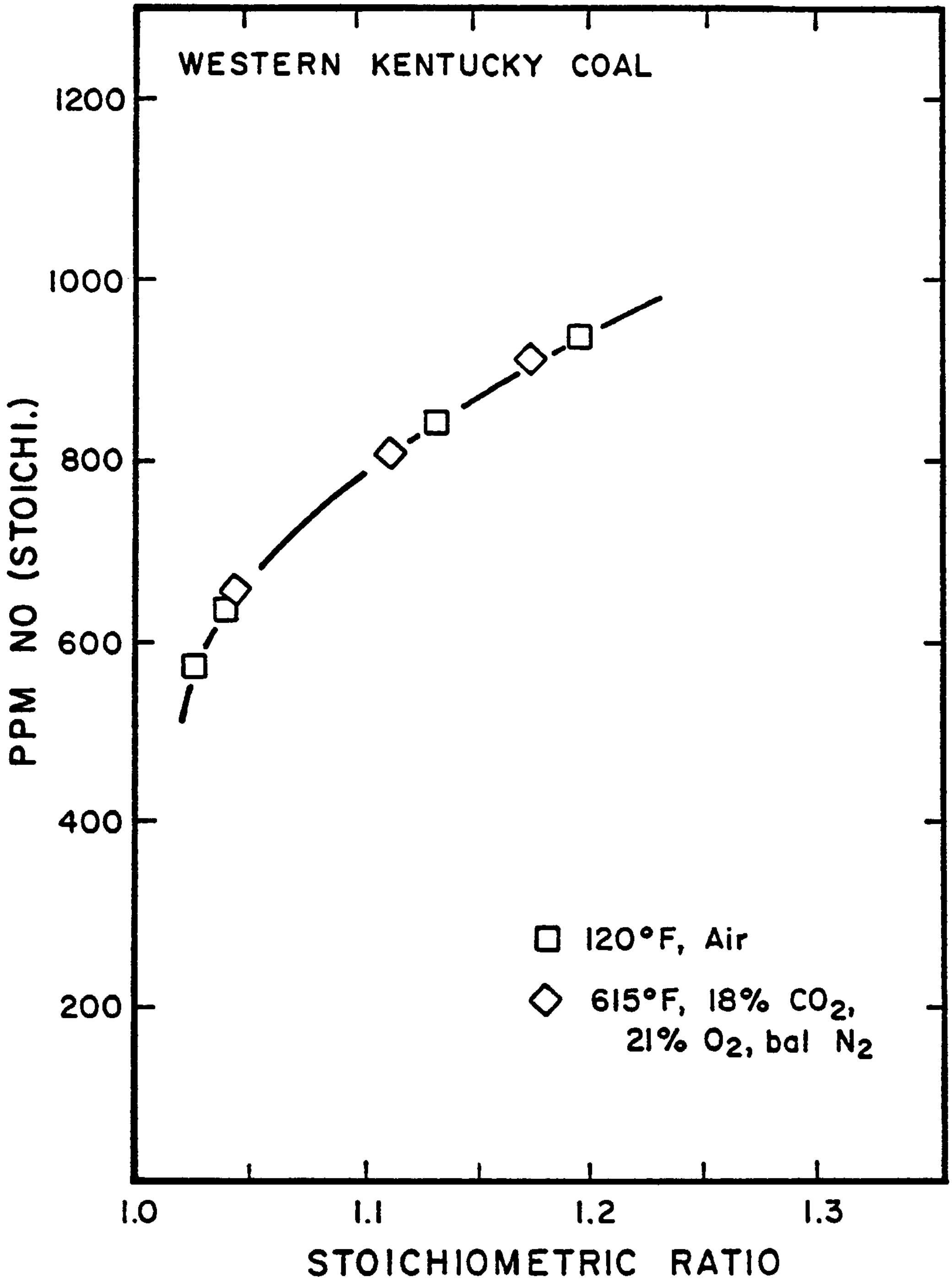

FIGURE 14. ADDITION OF $CO_2$ HAS NO CHEMICAL EFFECT ON TOTAL NO EMISSIONS $T_{ADB}$ CONSTANT (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR)

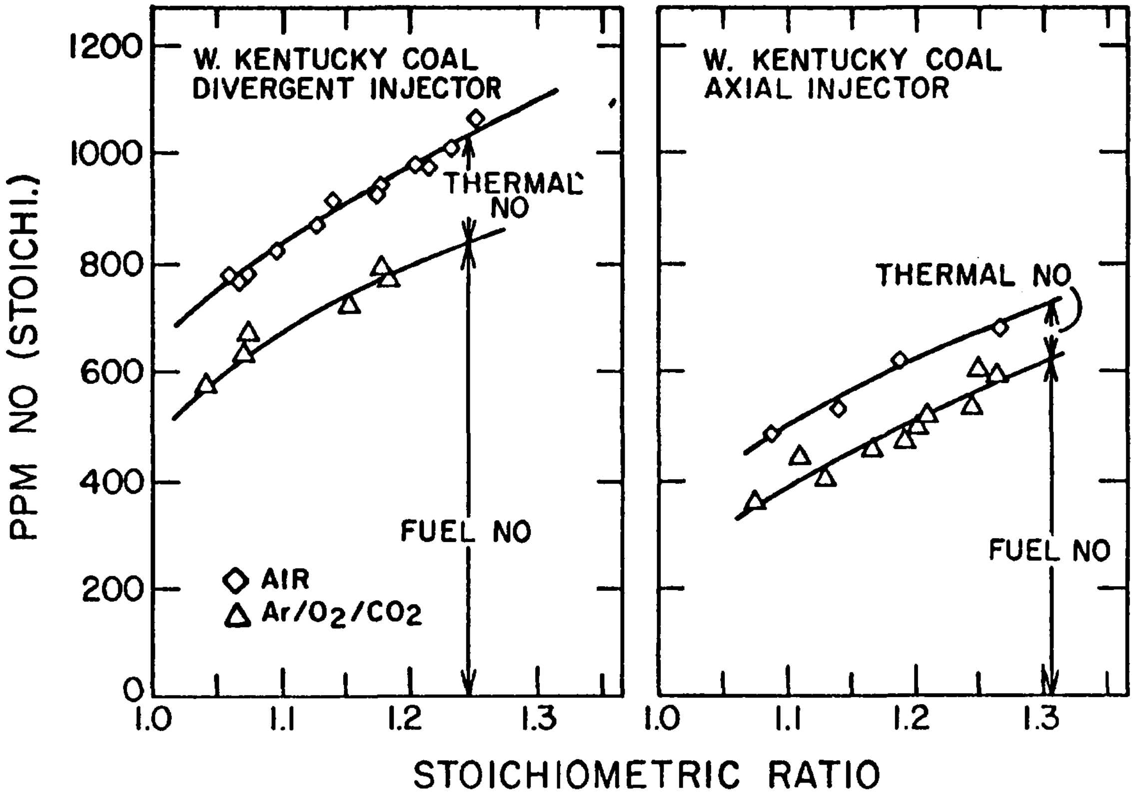

FIGURE 15. THERMAL AND FUEL NO EMISSIONS (WESTERN KENTUCKY COAL, 650°F PREHEAT)

14% primary air and 70 ft/sec throat velocity) clearly show that under these conditions over 80% of the total NO is the result of the oxidation of bound nitrogen in the fuel. Variations in primary air percentage, secondary air swirl and burner throat velocity did not change this finding, and under all conditions examined, fuel NO contributed at least 75% of the total NO emissions. (Pershing and Wendt, 1975)

Data from the single hole axial injector (650°F preheat, 45% swirl, 8% primary air) show that slow mixing significantly reduced total NO emissions. This is in agreement with pilot data (Heap *et al.* 1975) and field data on tangentially fired units (Crawford *et al.* 1975). However, it is clear that this dramatic reduction was due to a decrease in fuel NO emissions which again comprised approximately 80% of the total. Thus, although a change in mixing significantly altered total emission levels, the dominant NO producing mechanism in all cases was still through fuel nitrogen oxidation.

### 4.2.3 Coal Composition

The four pulverized coals tested during this investigation represent a cross-section of chemical and physical properties. Ultimate analyses are given in Table 1, Section 4.1.2. The Colorado, Pittsburgh and Western Kentucky are bituminous coals with heating values over 12,000 Btu/lb; the Montana coal is a subbituminous with a heating value of 8900 Btu/lb. The Colorado, Pittsburgh, and Montana, have statiscally identical fuel nitrogen contents of 1.3 ± 0.05 percent (dry, ash free) representative of many U.S. coals, while the Western Kentucky coal is a relatively high nitrogen coal (1.52 percent). Thus, in addition to considering the effect of total nitrogen percentage, this work focused on the effect of coal rank for coals with the same nitrogen content. It is to be expected that coal rank will effect the physical and chemical processes likely to occur during volatilization and combustion. The importance of total sulfur content was also considered; the Pittsburgh and Western Kentucky are high sulfur ($\geq$2.5 percent) eastern coals, while the Colarado and Montana are western coals with only approximately one percent sulfur.

Figure 16 summarizes the baseline total, fuel, and (by difference) thermal NO emission data for the divergent injector with all four coals. In each case, the fuel flow was maintained at approximately 6.2 lbs/hr, the primary air at 14 percent of stoichiometric, the secondary air swirl at 44 percent, and the air preheat at its maximum. The upper curve in each plot is data obtained while burning the coal with air (total NO emissions) and the lower curve is with $Ar/O_2/CO_2$ (fuel NO emissions). Over 75 percent of the total NO emissions are the result of fuel nitrogen oxidation for each of the four coals tested.

Figure 17 is a composite plot of the divergent injector data on the three bituminous coals at a constant set of operating conditions: 520°F secondary air preheat, SR = 1.15, 44 percent swirl, full load (80,600 ± 4,300 Btu/hr gross heat input), and 14 percent primary air. The data indicate that at these equivalent operating conditions, both the total and fuel NO emissions increase only slightly as fuel nitrogen increases. This is particularly surprising because the coals are known to have significantly different chemical and physical properties and because they exhibited different combustion characteristics. For example, the Pittsburgh coal is a caking coal; many of the particles melt upon heating and a fused carboneous residue and ash forms centispheres during

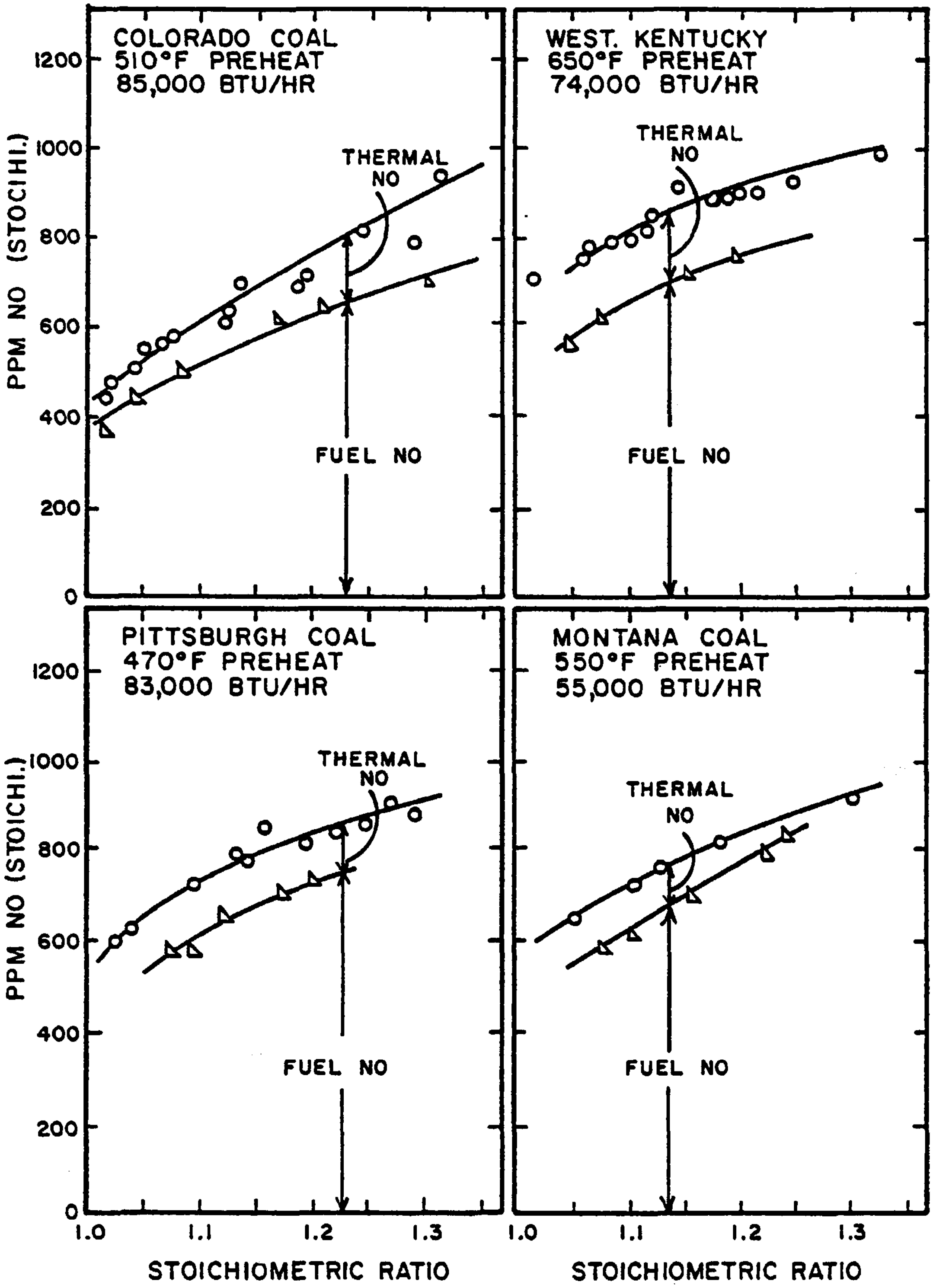

FIGURE 16. EFFECT OF COAL COMPOSITION ON THERMAL AND FUEL NO (ALL COALS, DIVERGENT INJECTOR)

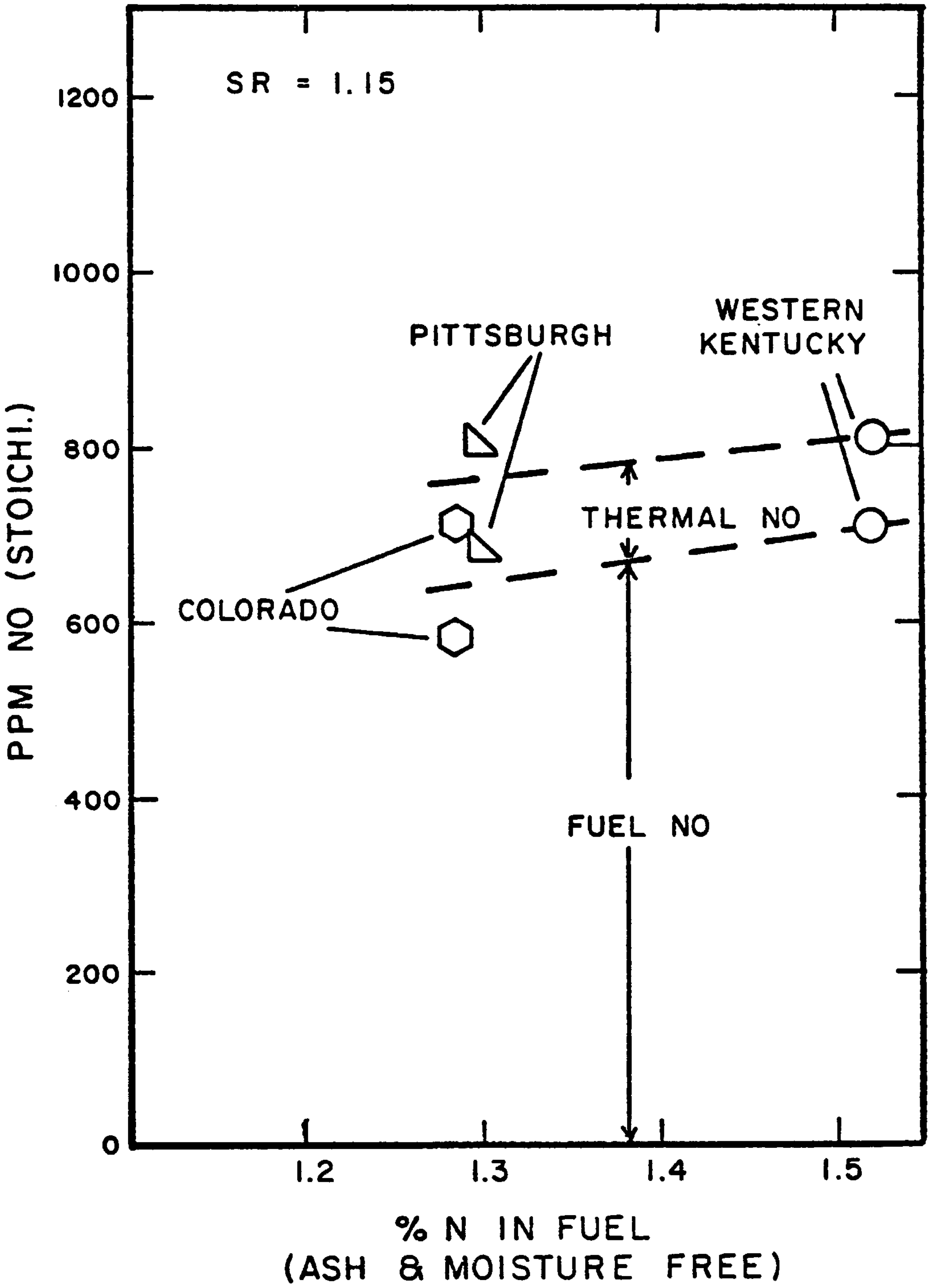

FIGURE 17. EFFECT OF FUEL NITROGEN CONTENT - BITUMINOUS COALS (DIVERGENT INJECTOR, 520°F PREHEAT, 15% EXCESS AIR)

combustion. The Pittsburgh coal flames were also generally less stable than the Colorado or Western Kentucky flames; i.e. the number of test conditions where a self-sustaining, attached flame could be achieved was much smaller for the Pittsburgh coal.

Figure 18 is a composite plot of the divergent injector data obtained with the Montana subbituminous coal compared to that from Western Kentucky coal. All of the data in Figure 18 were obtained with the divergent injector, a stoichiometric ratio of 1.15, approximately 550°F secondary air preheat, 14 percent primary stoichiometry, 44 percent swirl, and 75 percent load (55,000 Btu/hr) because this was the maximum heat input the coal feeder system could deliver with the Montana coal (due to the large amount of ash and moisture). The emissions again varied only slightly with increasing fuel nitrogen content, even though the combustion characteristics changed dramatically. The band of stable operating conditions was much reduced in the case of the Montana coal, perhaps because of its large moisture content (21 percent).

The lack of a first order effect by the composition parameters on total and fuel NO formation supports a recent observation by Blair *et al.* (1976) that volatile nitrogen evolution during particle pyrolysis is not a strong function of coal composition, even though the total mass volatilized varies widely from one coal to another. It is also consistent with the hypothesis of Flagan *et al.* (1974) that volatile fuel nitrogen conversion may be controlled by a rate constrained partial equilibrium and hence relatively independent of speciation.

Figure 19 is a composite plot of all the fuel NO data versus stoichiometric ratio. All four coals exhibited the same stoichiometry dependence

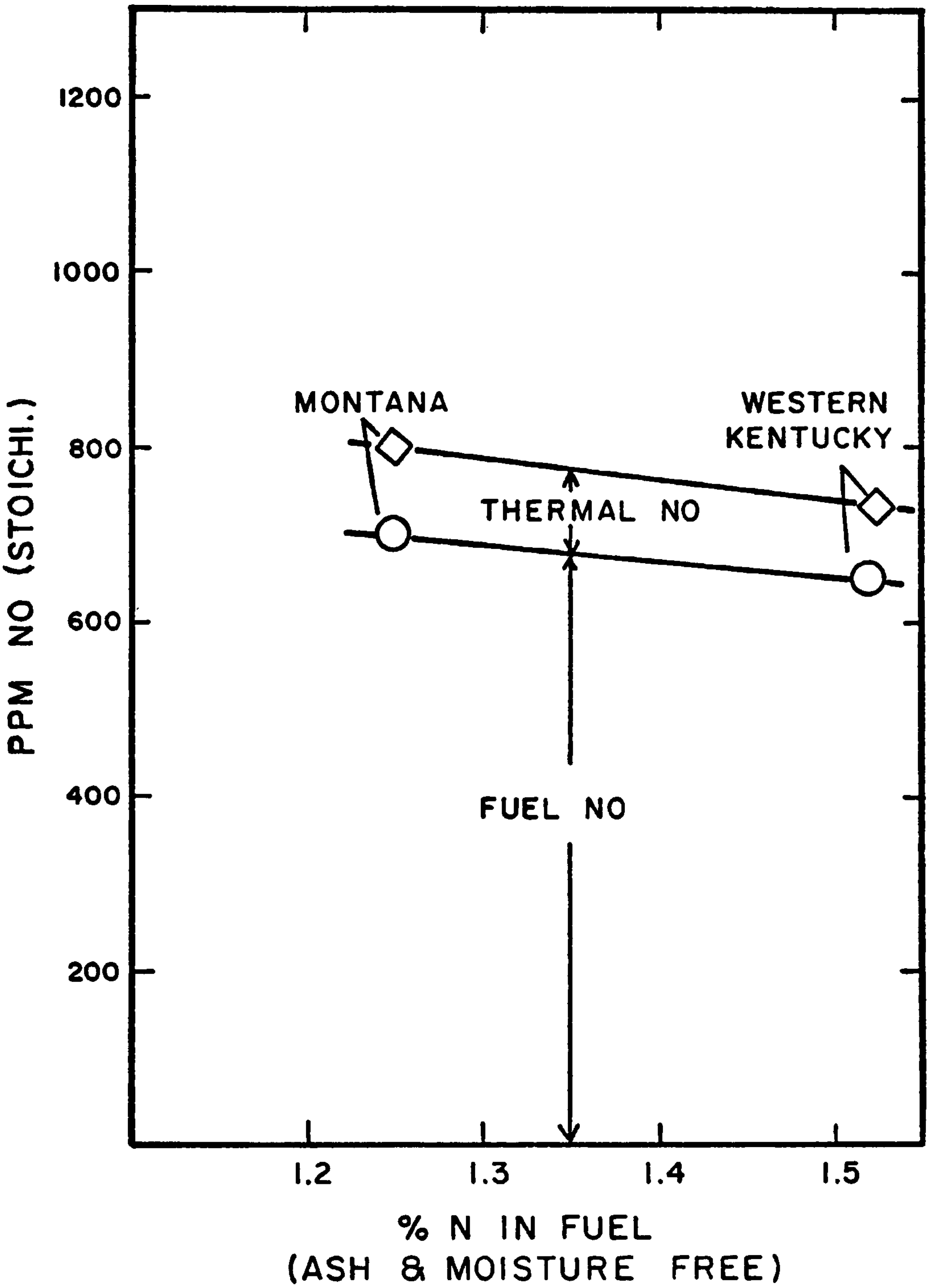

FIGURE 18. EFFECT OF COAL COMPOSITION AND RANK (DIVERGENT INJECTOR, 520°F PREHEAT, 15% EXCESS AIR)

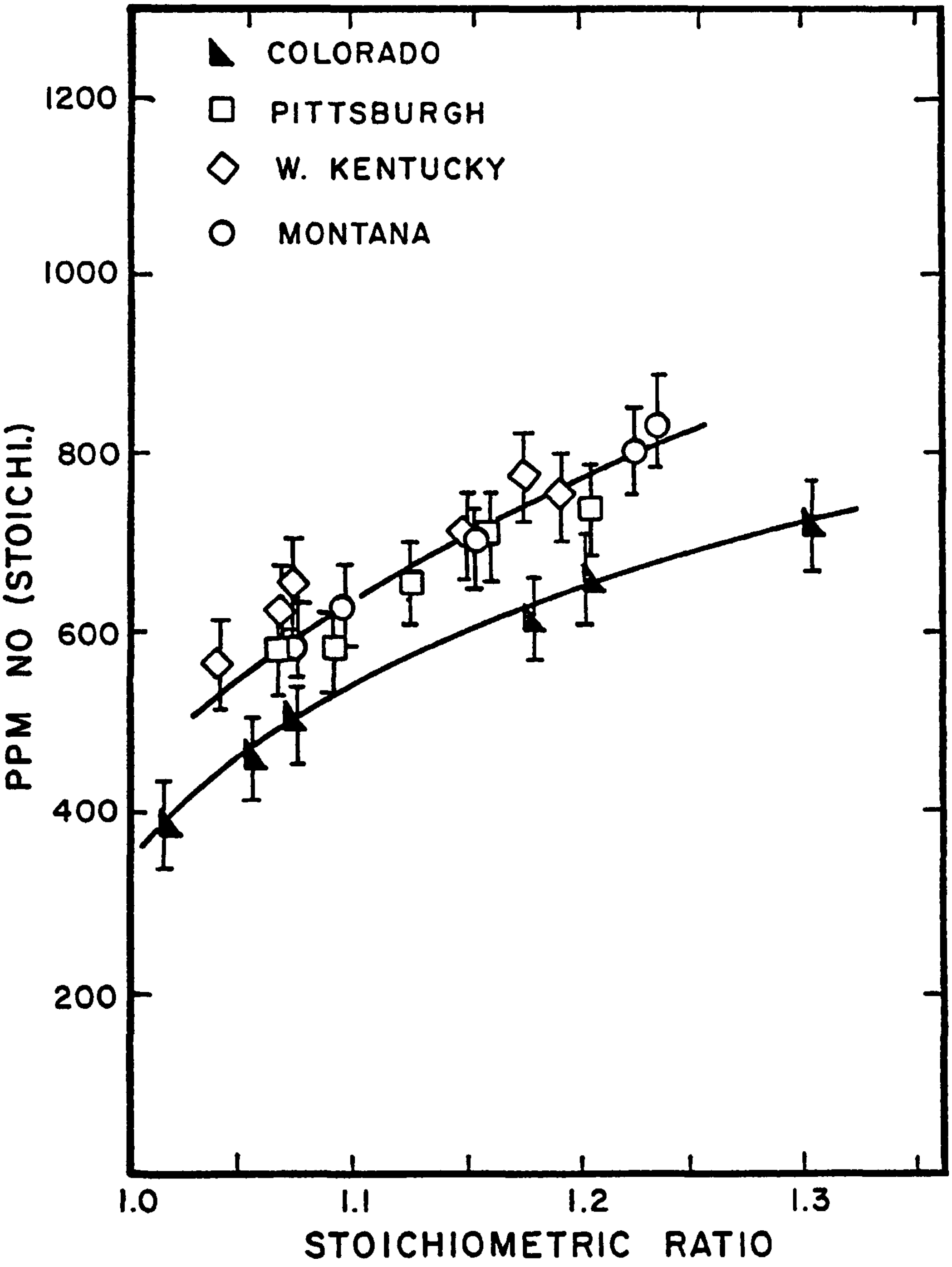

FIGURE 19. FUEL NO EMISSIONS VS EXCESS AIR (DIVERGENT INJECTOR)

and the actual fuel NO emission levels for the Pittsburgh, Western Kentucky and Montana coals were essentially identical measured either on a dry, corrected volumetric basis or as emission factors (0.82 ± 0.02 lbs $NO_2 10^6$ Btu at SR = 1.5). As the lower line in Figure 19 indicates, the Colorado coal gave slightly lower fuel NO emissions.

Figure 20 is a composite plot of the fuel nitrogen conversion (to NO) data for the divergent fuel injector with each of the four coals at SR = 1.15, 44 percent swirl, 520°F secondary air preheat and 14 percent primary stoichiometry. (The number associated with each point is the actual fuel NO emission in ppm (STOICHI) from which the conversion was calculated). As previously noted, the data indicate that none of the composition parameters has a first order effect on NO emissions.

As Figure 20 indicates the reason the fuel NO emissions do not change significantly as nitrogen content increases, is that the corresponding percentage conversion decreases simultaneously. (Note that to relate the fuel nitrogen conversion to the actual ppm emission level, one must consider not only the fuel nitrogen content, but also the total fuel composition, particularly the carbon/hydrogen/ash ratio). For the four coal tests, the baseline conversions ranged from twenty-three percent to twenty-eight percent and this is in good agreement with calculations from field results (Habelt and Howell, 1976; Dykema and Hall, 1975) and with recent definitive fundamental studies by Pohl and Sarofim (1975).

Figure 20 also shows two second order effects which should be noted. First, it indicates that fuel nitrogen conversion (and hence fuel NO) is dependent, albeit only slightly, on composition parameters other than

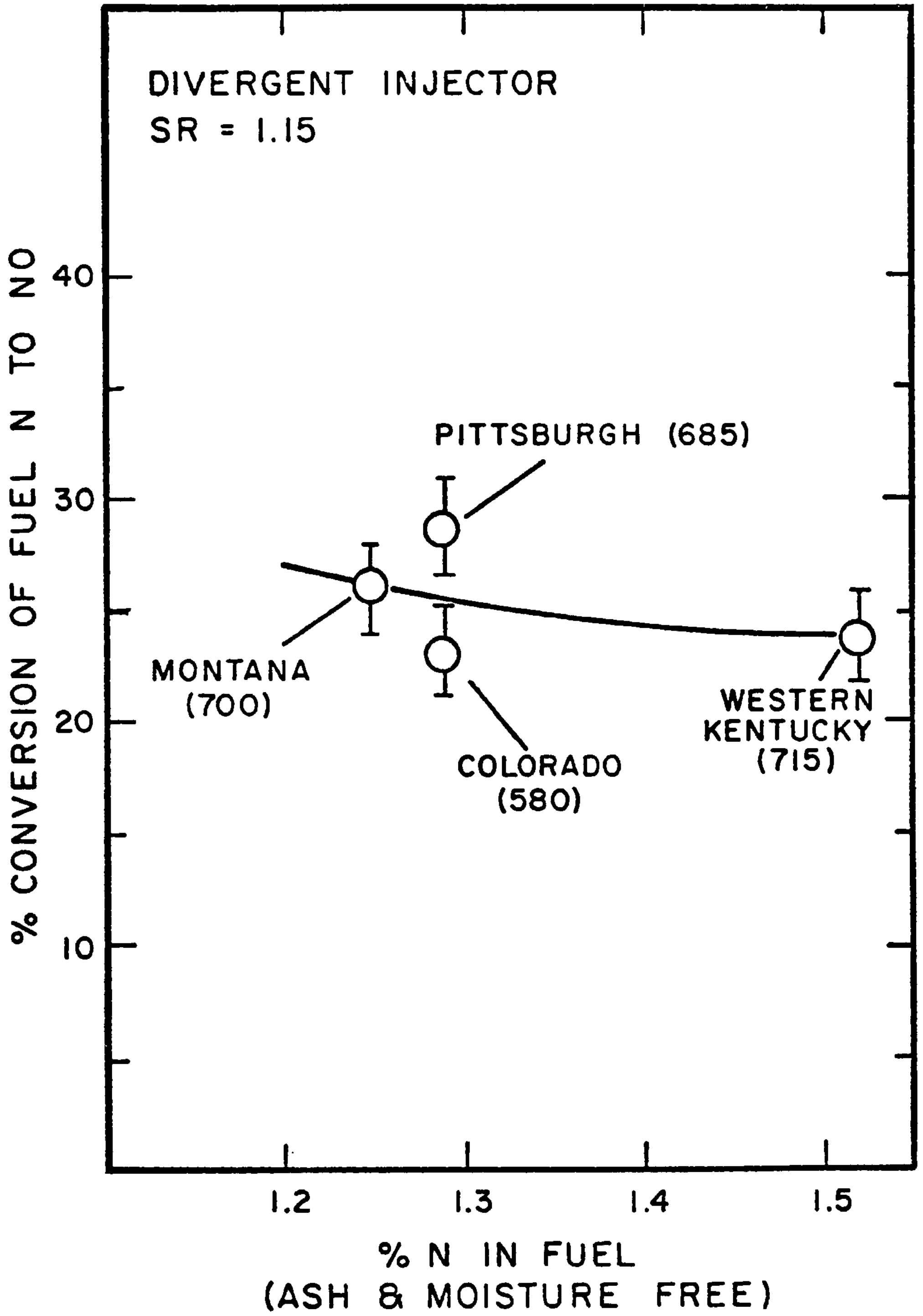

FIGURE 20. EFFECT OF COAL COMPOSITION ON FUEL NITROGEN CONVERSION (DIVERGENT INJECTOR, 15% EXCESS AIR)

total fuel nitrogen content. The Pittsburgh and Colorado are both bituminous coals with 1.29 percent fuel nitrogen. The difference in fuel nitrogen conversion (which is outside experimental error bounds) must, therefore, be attributed to composition parameters other than total nitrogen content and coal rank. The decreased conversion with the Colorado coal could be the result of its low sulfur content since sulfur oxidation has been shown to enhance fuel NO formation in oil flames (Wendt and Corley, 1976). It might also be the result of small differences in the nitrogen speciation, the physical behavior of the coals, or the particle size distribution.

Secondly, the Colorado and Montana are both Western, low-sulfur coals with nearly equal fuel nitrogen contents. Comparison of their conversions indicates that coal rank also appears to have a small influence on fuel NO emissions.

The experimental results from the four coals lead to the following general conclusions:

1. Fuel nitrogen oxidation is the primary NO formation mechanism in pulverized coal combustion regardless of chemical composition or rank of the coal.
2. Total and fuel NO emissions are only slightly dependent upon composition parameters. The wide variation in emissions from actual field units can not be attributed to differences in fuels.
3. Total and fuel NO emissions increase only slightly as the fuel nitrogen level increases because the percent conversion of fuel N to NO simultaneously decreases.

4. Composition parameters other than total nitrogen content (e.g. sulfur content, nitrogen speciation, etc.) may be of second order significance when comparing various coals.

5. Coal rank has a major effect on combustion characteristics but only a second order effect on either total or fuel NO emissions.

### 4.2.4 Temperature Dependence

Investigation of the temperature dependence of fuel NO is important because although it has been postulated (Flagan *et al.* 1974) that the temperature dependence of homogeneous oxidation of nitrogenous compounds is slight, these results cannot be directly extrapolated to coal combustion because both particle temperature and particle heating rate (Pohl and Sarofim, 1975) influence the fraction of fuel nitrogen that is devolatilized and the fraction that remains in the char (Lilley and Wendt, 1976) during combustion. Indeed variations of the physical behavior of coal due to differences in type and rank might also be expected to influence the fate of fuel nitrogen during coal volatilization.

A variety of experimental tests were conducted to investigate the overall temperature dependence of both the thermal and fuel NO formation mechanisms. Four separate methods of changing the local flame temperature were employed:

- Varying the secondary air preheat temperature

- Increasing the inert input by recirculating flue gas

- Altering the specific heat of the inlet air by replacing a portion of the $N_2$ (or Ar) with $CO_2$.

- Slightly enriching the inlet air with $O_2$

Burner sleeves were available to ensure that inlet velocities and, therefore, flow patterns could also be approximately matched, but our preliminary tests (Pershing and Wendt, 1975) indicated that they were not required since small variations in secondary air velocity had negligible effect.

Figure 21 shows the results of reducing the air preheat from 660°F to 110°F with the Western Kentucky coal. The NO emissions decreased by about 200 ppm to approximately the level previously determined to be the fuel NO (dotted line.) This type testing was not successful with any of the other coals due to combustion instabilities at the low preheat condition.

Figure 22 shows the results of recirculating various amounts of flue gas. Even with large quantities of flue gas recirculation it was not possible to reduce the total NO emissions significantly below the fuel NO level.

Figure 23 shows data taken during the combustion of the Colorado coal with varying amounts of $CO_2$ in the inlet air. The heat capacity of the inlet air was increased by replacing a portion of the nitrogen (di-atomic) with $CO_2$ (tri-atomic); in all cases the inlet oxygen was maintained at 21%. (Of all the temperature variation methods, replacement of $N_2$ by $CO_2$ is perhaps the best because it does not change either the mass or volumetric flow rate of the oxidizer significantly.) As Figure 23 indicates decreasing the flame temperature by increasing the oxidizer specific heat again reduces the NO emissions toward the fuel NO level.

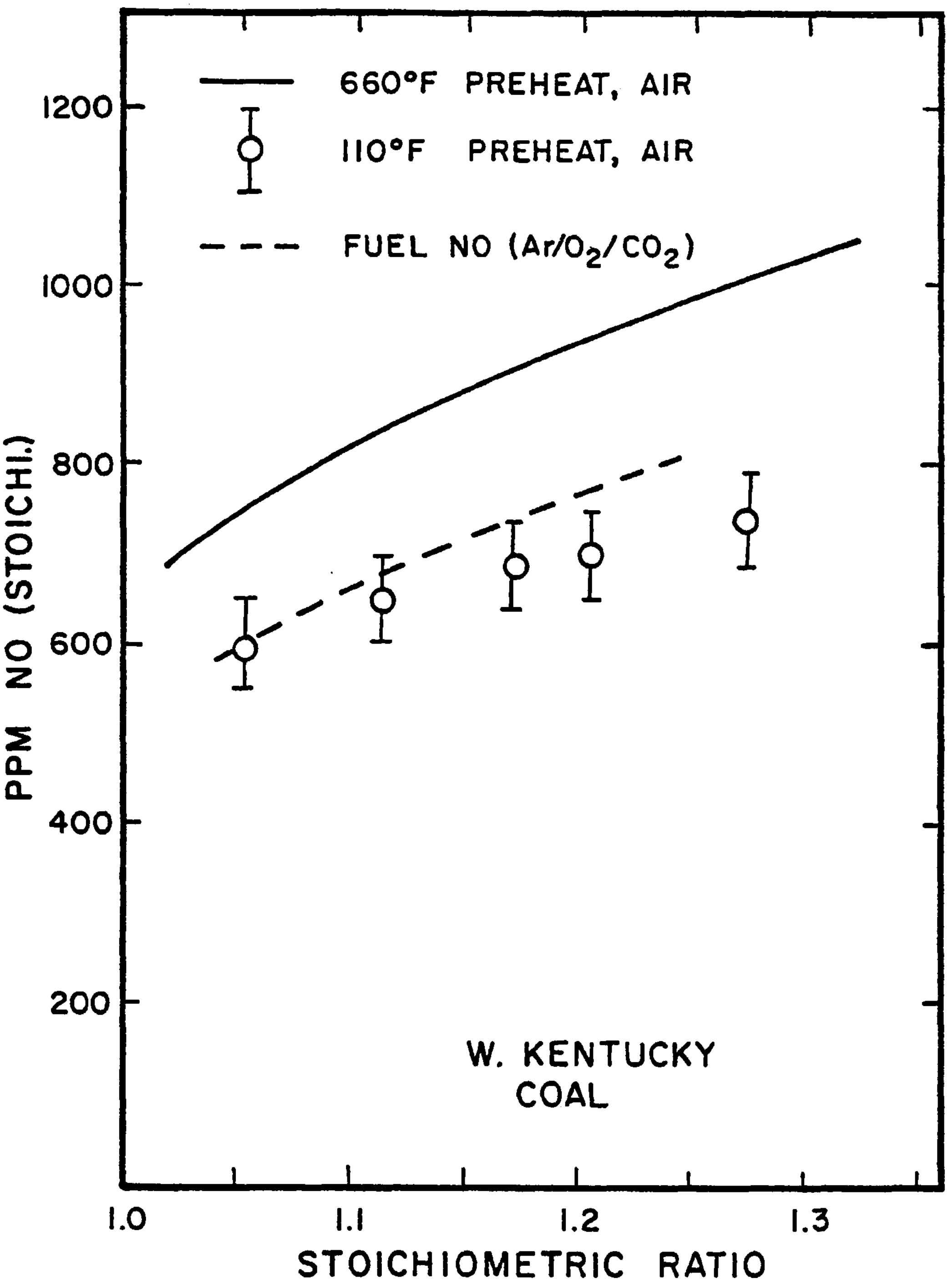

FIGURE 21. REDUCING SECONDARY AIR PREHEAT DECREASES TOTAL NO EMISSIONS (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR)

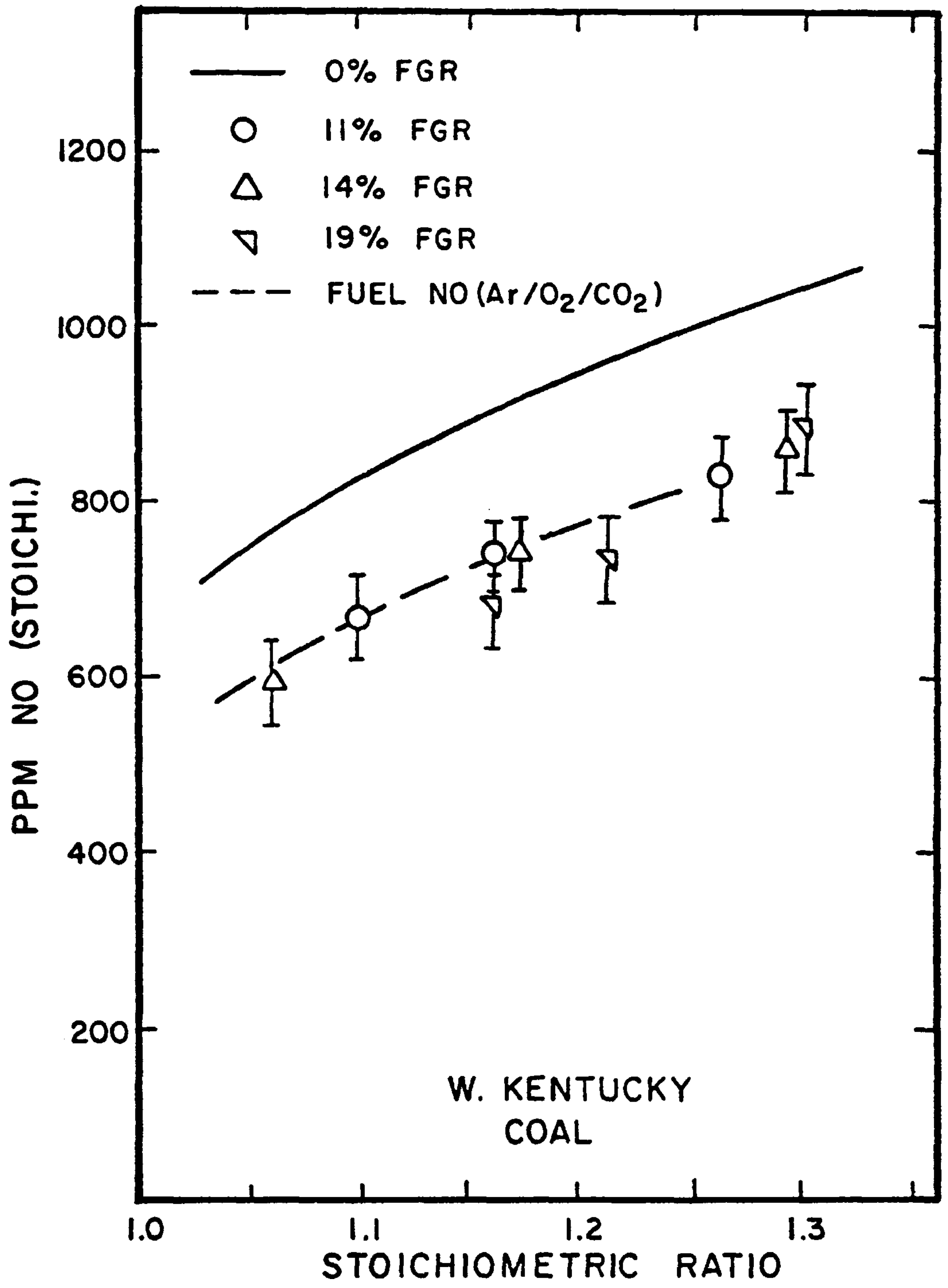

FIGURE 22. RECIRCULATING FLUE GAS DECREASES TOTAL NO EMISSIONS (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR, 700°F PREHEAT)

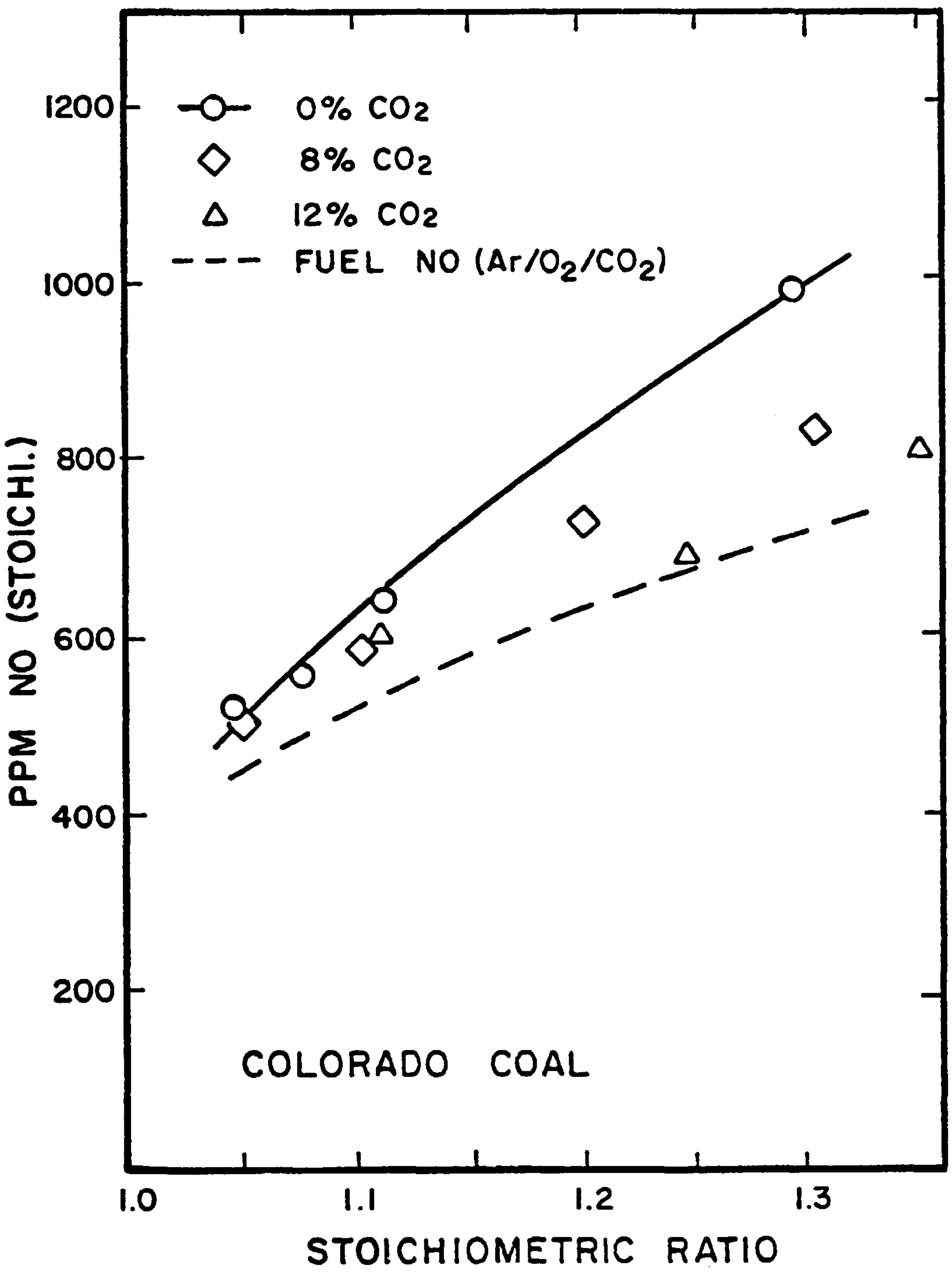

FIGURE 23. INCREASING THE HEAT CAPACITY OF THE OXIDIZER DECREASES TOTAL NO EMISSIONS (COLORADO COAL, DIVERGENT INJECTOR, 520°F PREHEAT)

The pulverized coal flames examined in this study were turbulent diffusion flames with large internal temperature gradients and turbulent fluctuations. Since they are, therefore, characterized by temperatures which are functions of both spatial position and time, the adiabatic flame temperature was chosen as the parameter correlating flame temperature. Clearly the actual peak temperatures were significantly lower. Results for the Western Kentucky and Colorado coals are shown in Figure 24, and are also typical of the other two coals. All the data shown are for 15% excess air and care was taken to minimize purely aerodynamic variations. The data is for the divergent injector and the numbers associated with the data points refer to Table 2 which describes how each condition was achieved.

Total NO emissions increased exponentially with theoretical flame temperature but fuel NO was remarkably insensitive to temperature over a wide range applicable to present combustion technology. For the Colorado coal, fuel NO emissions were essentially constant over a theoretical temperature range of 3600°F through 4100°F and a measured wall temperature range of 1850°F through 2150°F. Further, this insensitivity to temperature appears to be independent of the excess air level as shown in Figure 25. At very high temperatures, however, fuel NO emissions underwent a sudden increase, and this was observed to occur at a slightly different temperature for each coal shown here.

The Montana and Pittsburgh coals exhibited similar behavior in the medium temperature range, but did not allow investigation at the extremes due to combustion instabilities.

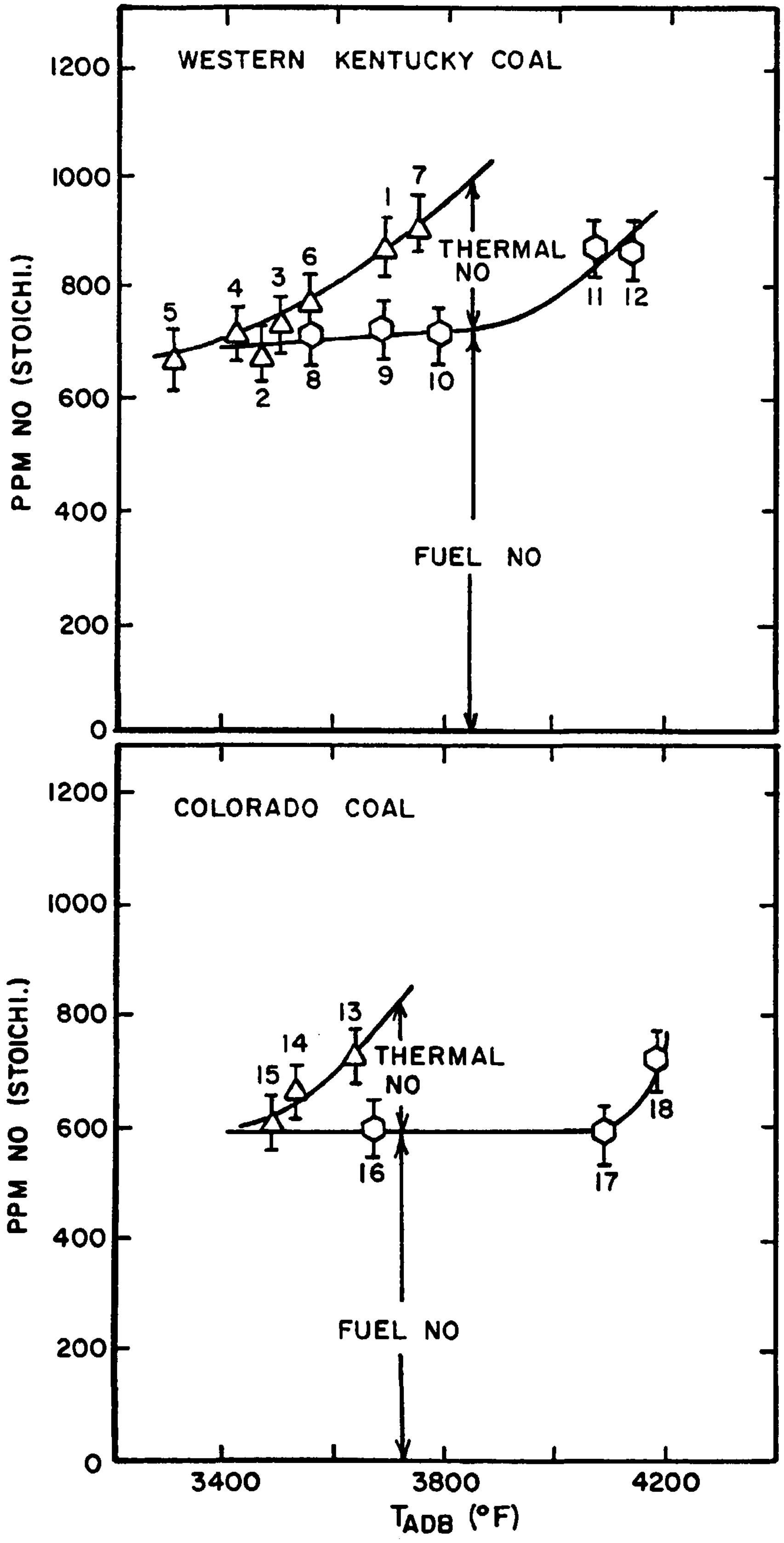

FIGURE 24. TEMPERATURE DPEENDENCE OF THERMAL AND FUEL NO (WESTERN KENTUCKY AND COLORADO COALS, 15% EXCESS AIR, DIVERGENT INJECTOR)

TABLE 2. EXPERIMENTAL CONDITIONS - FIGURE 24

| | |
|---|---|
| Western Kentucky Coal | |
| 1 | 666°F preheat (baseline) air |
| 2 | 110°F preheat air |
| 3 | 690°F preheat air with 10.6% FGR |
| 4 | 685°F preheat air with 14.1% FGR |
| 5 | 705°F preheat air with 19.0% FGR |
| 6 | 690°F preheat 21% $O_2$, 11.6% $CO_2$ in $N_2$ |
| 7 | 540°F preheat 22.6% $O_2$ in $N_2$ |
| 8 | 110°F preheat 19.3% $O_2$, 16.0% $CO_2$ in Ar |
| 9 | 110°F preheat 21.3% $O_2$, 18.7% $CO_2$ in Ar |
| 10 | 110°F preheat 21.4% $O_2$, 11.4% $CO_2$, in Ar |
| 11 | 475°F preheat 21% $O_2$ in Ar |
| 12 | 435°F preheat 23.0% $O_2$ in Ar |
| Colorado Coal | |
| 13 | 505°F preheat air |
| 14 | 515°F preheat 20.9% $O_2$, 7.7% $CO_2$ in $N_2$ |
| 15 | 530°F preheat 20.7% $O_2$, 11.8% $CO_2$ in $N_2$ |
| 16 | 215°F preheat 21% $O_2$, 18.7% $CO_2$ in Ar |
| 17 | 475°F preheat 21% $O_2$ in Ar |
| 18 | 481°F preheat 23.8% $O_2$ in Ar |

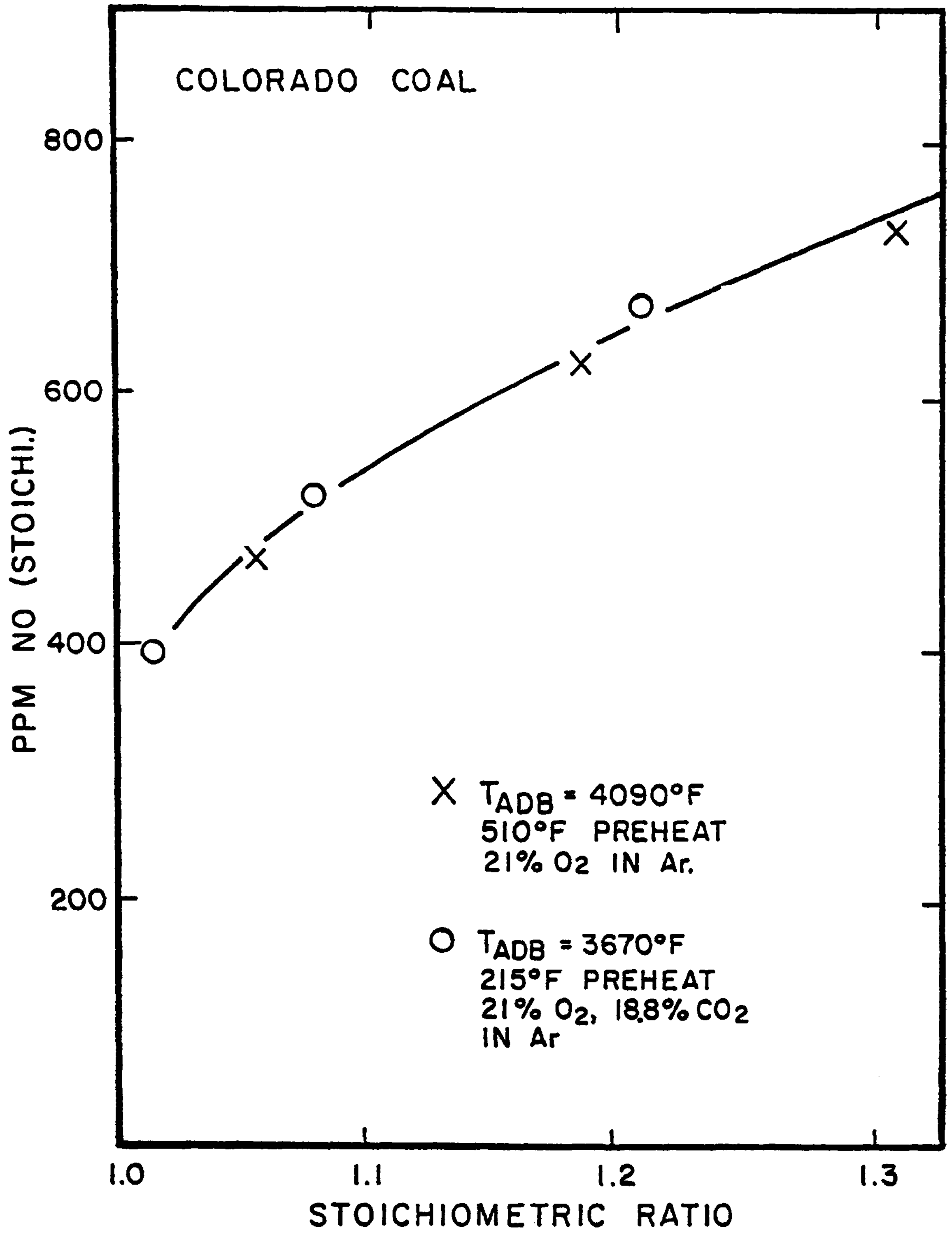

FIGURE 25. FUEL NO FORMATION IS ONLY WEAKLY TEMPERATURE DEPENDENT (COLORADO COAL, DIVERGENT INJECTOR)

At low temperatures the total NO emissions approached the (constant) fuel NO value and the thermal NO asymptotically approached zero. Furthermore, within the accuracy of the experiment, the emissions were not dependent on the method for reducing temperature. Therefore, so long as the fluid dynamics remain unchanged, flue gas recirculation (or any temperature reduction technique) will only decrease the thermal NO formation and is hence of somewhat limited value for pollutant ($NO_x$) control with pulverized coal. This had been shown previously by Armento (1975) on pilot scale systems and was recently demonstrated on a full scale wall-fired unit by Thompson (1976).

Figure 26 is a plot of the thermal NO measured for natural gas with that measured for the various coal flames. (Thermal NO was defined to be the difference between total NO and fuel NO as determined with $Ar/O_2/CO_2$.) The data indicate that the thermal NO values for coal are in line with those obtained for gas, in this combustor, for the same injectors and under similar aerodynamic and thermal conditions. This indicates that interactions between fuel and thermal NO are not of first order importance and it suggests that the controlling thermal mechanism may be similar to that in gas flames.

In summary, the temperature experimentation revealed that:

i) Fuel NO formation is relatively insensitive to flame temperature over a wide range of practical interest

ii) Thermal NO formation in coal flames behaves similarly to NO formation in natural gas flames under similar conditions, and

iii) All temperature reduction schemes have approximately the same small effect on total NO emissions in that only the thermal NO is reduced . Indeed it appears that without changes in aerodynamics and mixing, there is very little opportunity to lower

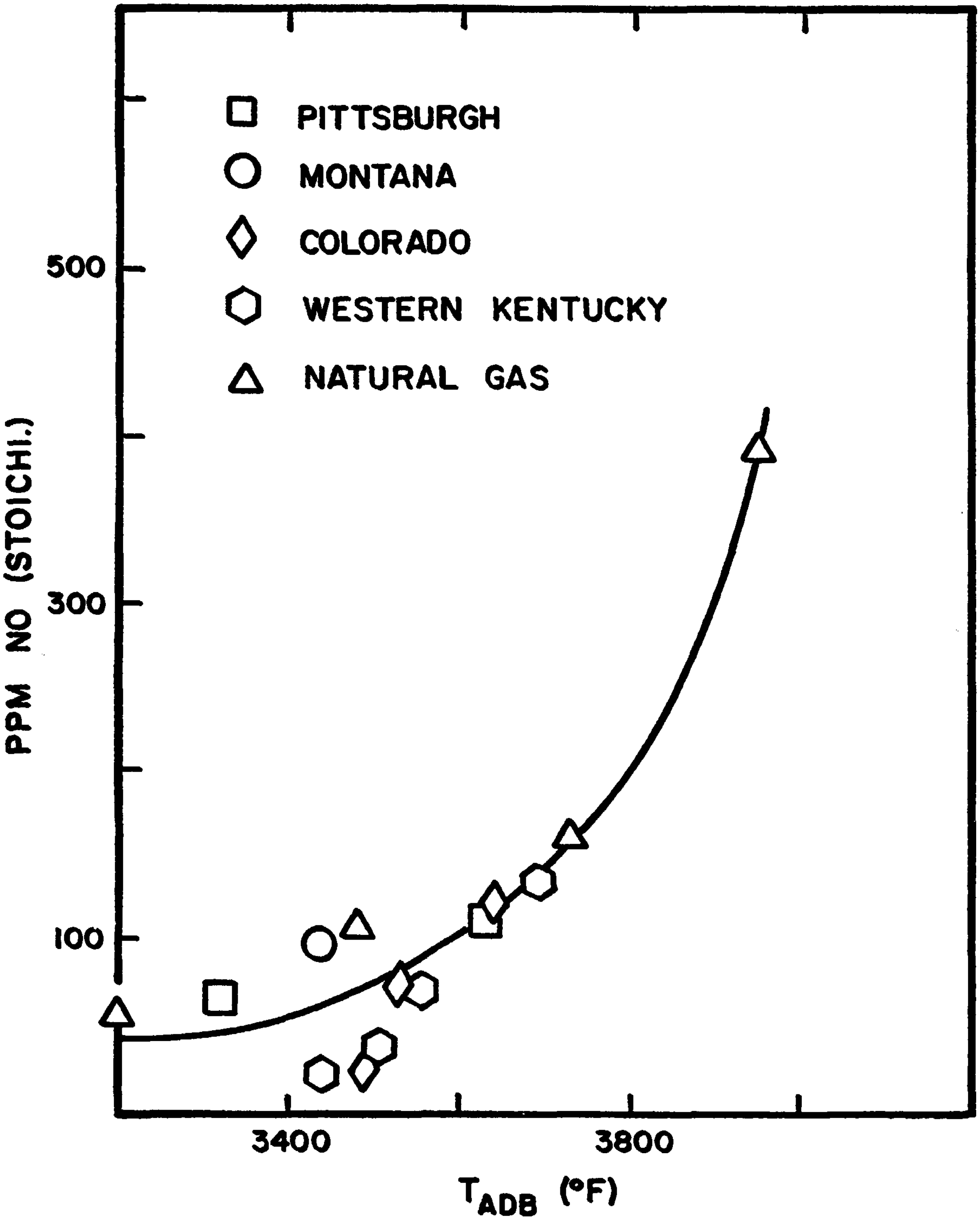

FIGURE 26. THERMAL NO EMISSIONS FROM COAL AND NATURAL GAS (DIVERGENT INJECTOR, 15% EXCESS AIR)

total NO emissions below the (constant) fuel NO level and still maintain stable flames. It would appear that particle heating rate becomes important only at very high flame (and wall) temperatures and presumably at high initial heat fluxes to the particle, whereupon an increase in fuel nitrogen conversion is observed.

### 4.2.5 Importance of Char Nitrogen

An FMC coal char was studied to establish its combustion/pollution characteristics and to provide input for estimating the importance of Char NO formation during pulverized coal combustion. Char was burned in two modes:

i) the flame mode, in which a turbulent diffusion flame was attached to the injector with the help of a small quantity of methane (21% of the total heat release) in the primary "air" and in which methane simulated nitrogen free volatiles;

ii) the reactor mode, in which pure char without methane, burned far from the injector and which simulated the char burnout regime of coal after all volatiles had been consumed and after significant mixing had taken place.

These two modes of char combustion spanned probable conditions during the char burnout regime of pulverized coal combustion, and helped determine the effect of mixing and of "shielding" by residual volatiles (Wendt and Sternling, 1974) on fuel nitrogen conversion to make char NO.

The data (corrected for dilution by methane combustion products) are shown in Figure 27. The influence of combustion mode is small

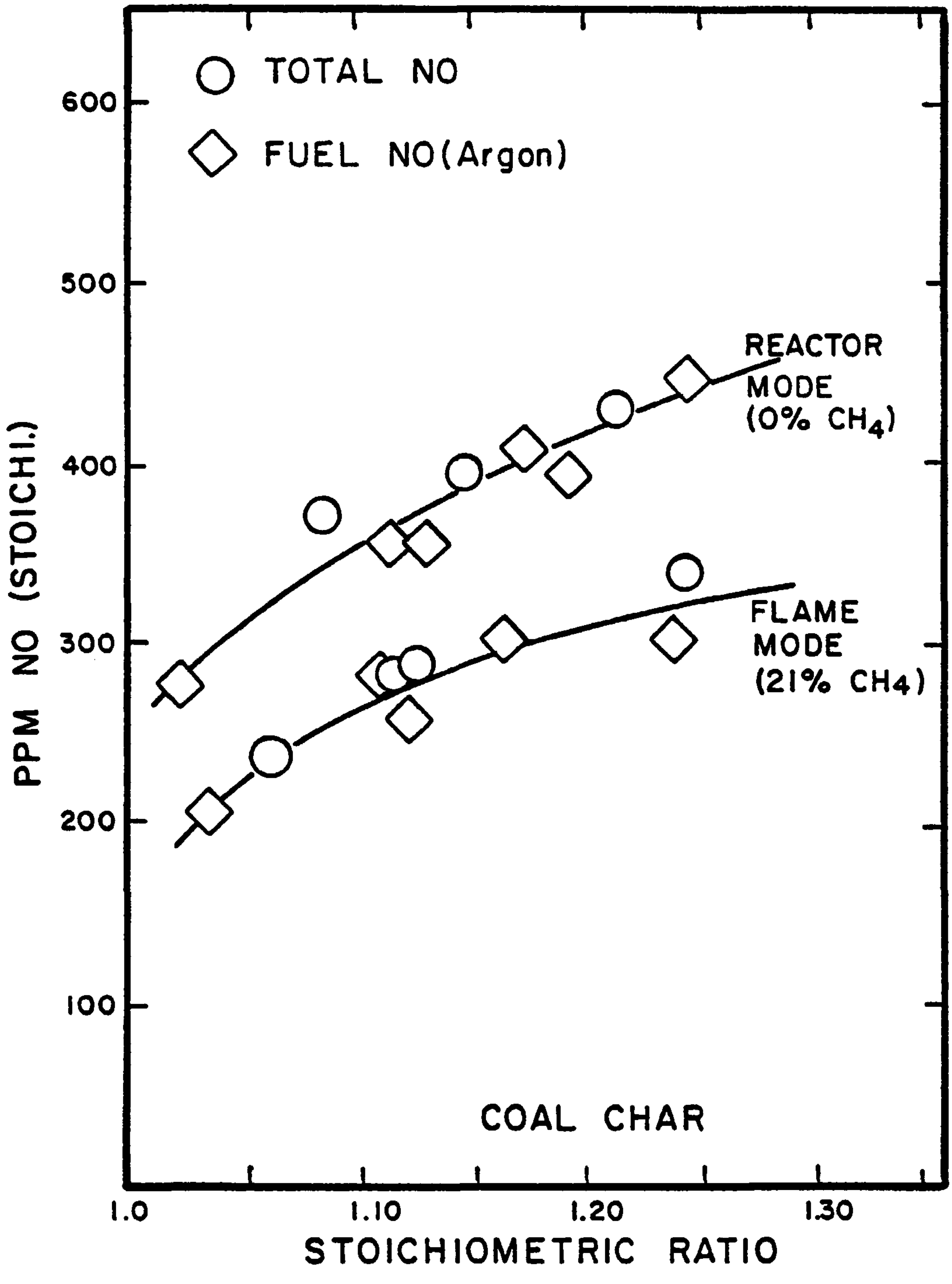

FIGURE 27. TOTAL NO AND FUEL NO EMISSIONS FROM COAL CHAR (AXIAL INJECTORS, 550°F PREHEAT)

(note expanded ordinate axis); reactor mode emissions are approximately 100 ppm higher than those in the flame mode. It is pertinent to note that when each of the other four coals was burned in the reactor mode, emissions increased by over 1000 ppm, indicating the importance of volatile nitrogen in coal. (See Section 4.3 of this report)

Figure 27 also indicates that essentially all the NO emissions are the result of fuel nitrogen oxidation; for both modes the data from combustion in argon/oxygen is coincident with that in air. Figure 28 presents data taken on fuel NO emissions at various combustion temperatures for both the reactor and flame mode. In char, as with coal, oxidation of chemically bound nitrogen is insensitive to combustion temperature.

Figure 29 shows the fuel nitrogen conversions in both modes of char combustion compared to that of the Pittsburgh coal which has approximately the same amount of fuel nitrogen. Char conversions are approximately half that of the corresponding coal and the conversion is somewhat less dependent upon total excess air.

Figure 30 summarizes all the conversion data for the four coals and the char (in the flame mode.) These data again illustrate the small char nitrogen conversions and the almost total insensitivity to injector design.

The char results have important implications as far as coal combustion is concerned, even though the actual char investigated

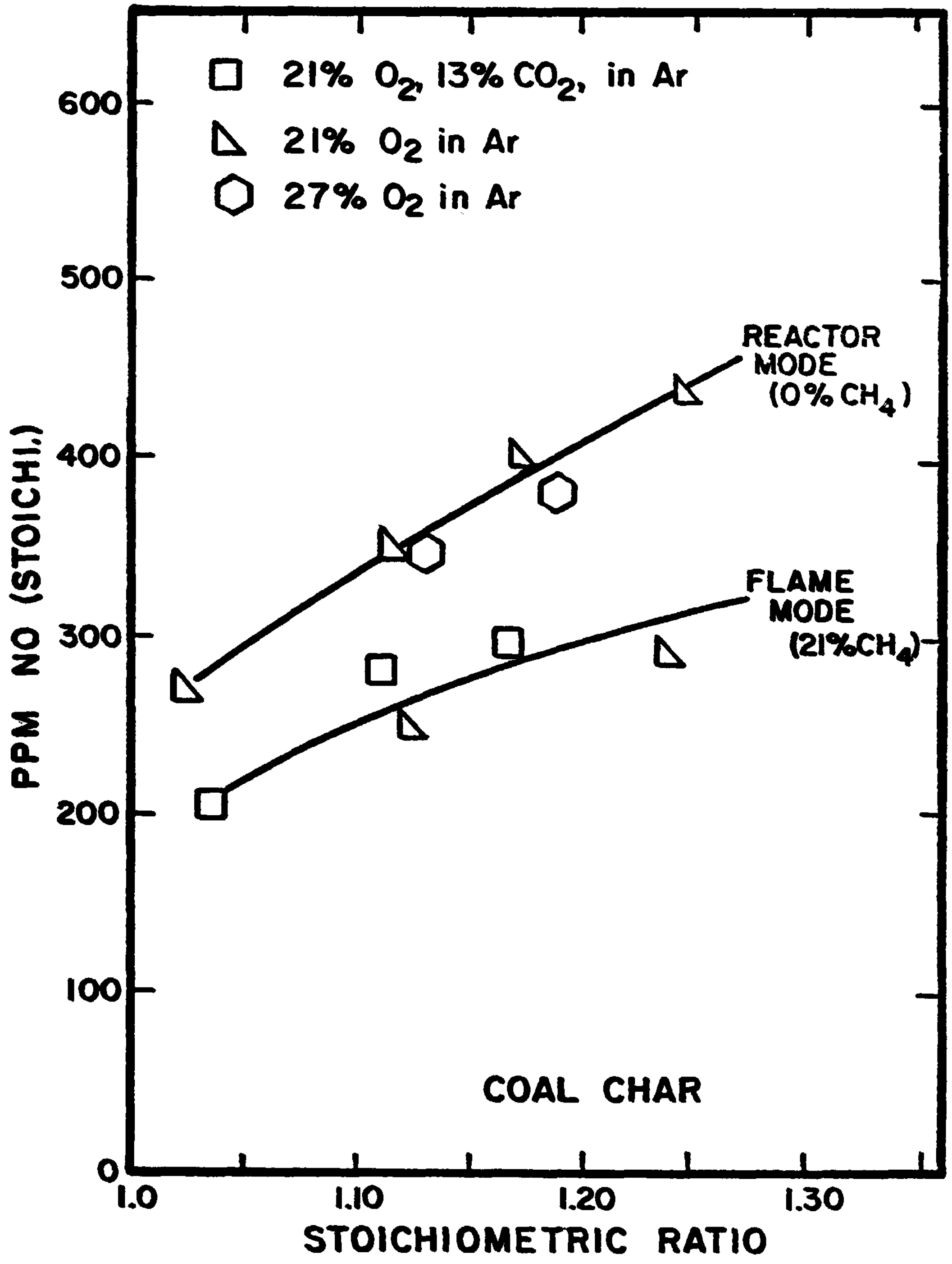

FIGURE 28. EFFECT OF COMBUSTION TEMPERATURE ON FUEL NO EMISSIONS FROM COAL CHAR (AXIAL INJECTORS 550°F PREHEAT)

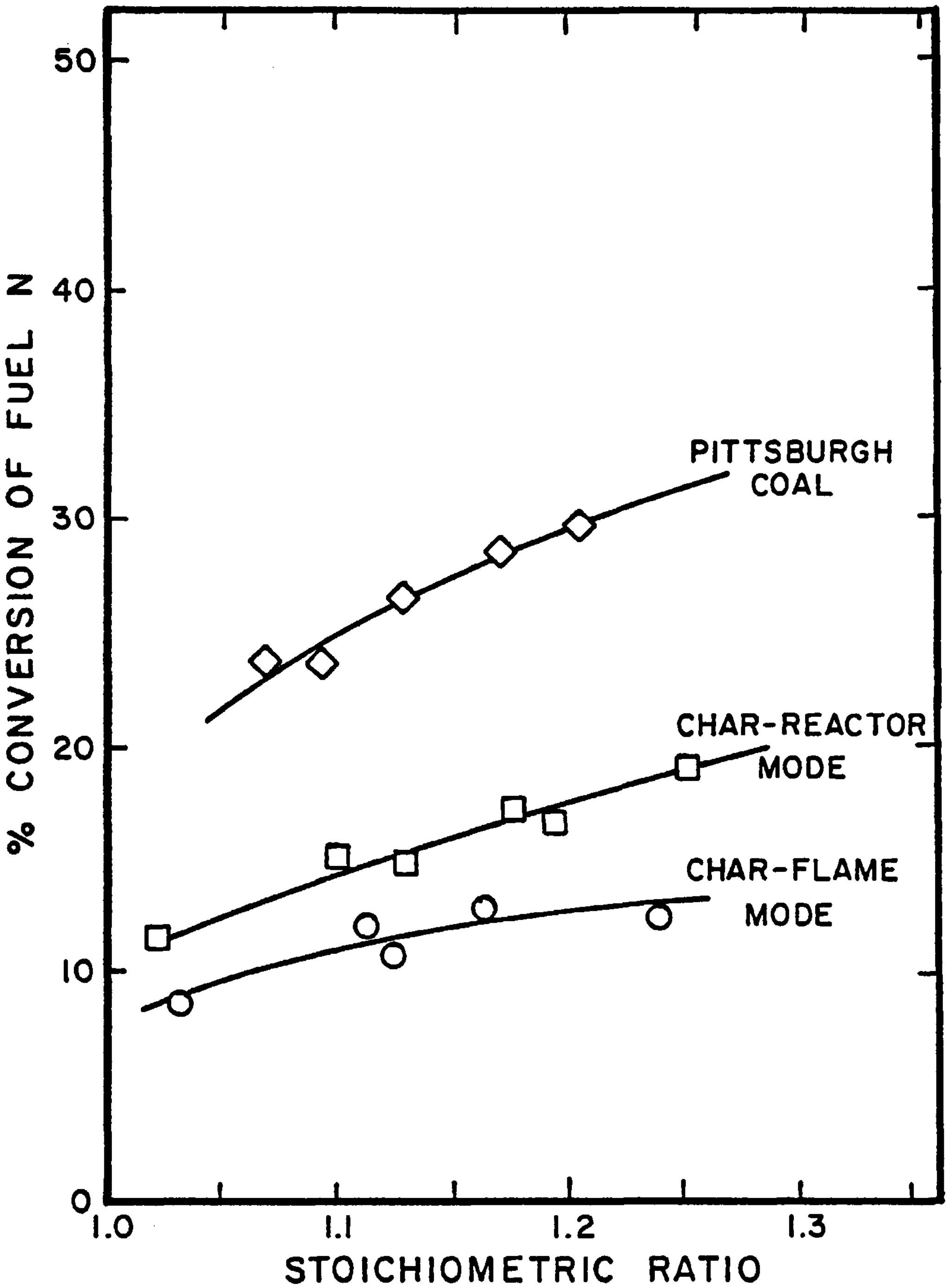

FIGURE 29. EXCESS AIR DEPENDENCE OF COAL AND CHAR

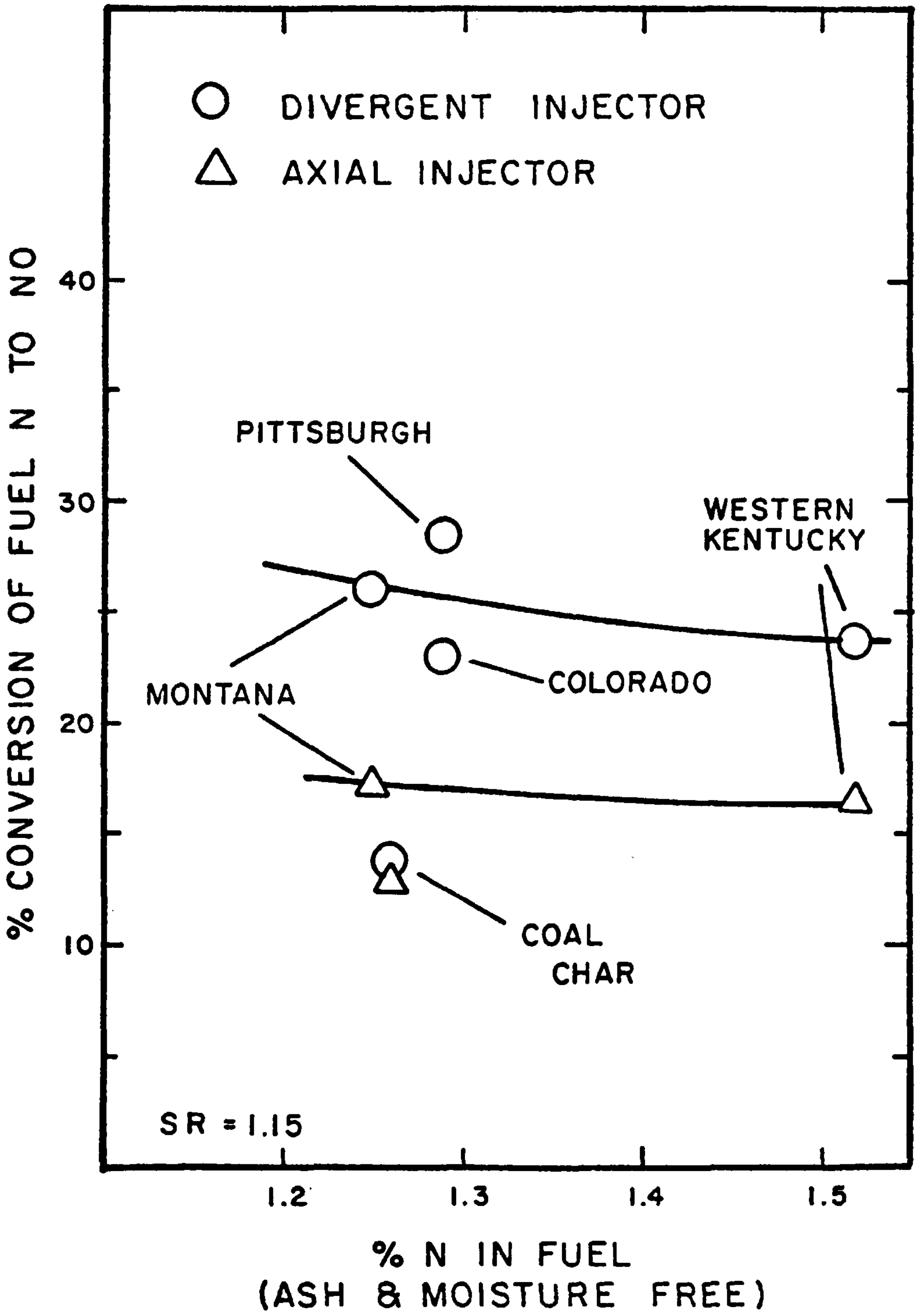

FIGURE 30. FUEL NITROGEN CONVERSION IN COAL AND CHAR FLAMES (15% EXCESS AIR)

was not the product of incomplete high temperature coal combustion. Of the nitrogen that goes to the char a sizeable fraction can be converted to NO at high temperatures, but this conversion is less than half of what would be expected from coal combustion. This finding is consistent with theory (Wendt and Schulze, 1976) and other data (Heap *et al*. 1975). In practical systems abatement of "char NO" by combustion modifications, may be difficult since unlike "volatile NO" it is relatively insensitive to changes in early mixing induced by injector design variations. This lends support to the theory that combustion modifications through aerodynamic changes primarily influence "volatile NO", although they may influence the fraction of fuel nitrogen remaining in char. This is consistent with the expectation that the time scale for conversion of volatile nitrogen is much shorter than the time scale for conversion of char nitrogen. The lack of temperature dependence of char NO is consistent with theories involving diffusion reaction interactions (Wendt and Schulze, 1976). Further work should entail the combustion of char derived from partial coal combustion, and of chars of various nitrogen contents. It will then be possible to ascertain the potential benefits of altering the volatile and char nitrogen ratio through combustion modifications.

## 4.3 FLUID DYNAMICS: THE ROLE OF EARLY MIXING

Results presented in the previous sections indicated that local oxygen content is a critical factor in determining the conversion of fuel nitrogen to NO in pulverized fuel flames. Local oxygen content can be changed both by variations of inlet and flue gas oxygen concentrations and by alterations in the mixing between the primary fuel stream and the secondary air. This section focuses on the general role of fluid dynamics as a means of changing local oxygen concentration. Changes in mixing patterns can be affected through

a) variations in fuel injector design
b) variations in primary and secondary air velocities
c) changes in swirl
d) changes in flame shape due to delayed particle ignition.

Of special interest is the role of early mixing which allows substantial secondary air to be in contact with fresh fuel nitrogen volatiles. However, a detailed discussion of turbulence and mixing is outside the scope of this work: rather, we attempt to develop general guidelines that address the problem, with a view to interpreting data and suggesting effective $NO_X$ control hardware configurations.

The role of injector design was alluded to in Section 4.2.1, in which it was shown that our divergent and axial injectors did simulate NO formation features representative of front fired and tangentially fired combustion units respectively. Field data showed that, on the average, front fired units had higher total NO emissions than tangentially fired, and our data suggested that this was due to increased fuel nitrogen conversion in wall-fired units.

Figure 31 summarizes the results on the role of fuel injector design. Both total and fuel NO emissions are shown as a function of stoichiometric ratio for both fuel injectors. Figure 31 shows that for both the Montana and Western Kentucky coals, the axial fuel injector, which has slow penetration of the secondary air stream into the primary air/fuel stream, leads to much lower fuel NO (and total NO) emissions than the three hole divergent injector, which causes rapid penetration. Decreasing early mixing by injector design leads to low fuel conversion to NO.

Mixing between the fuel and secondary air streams can also be altered by changes in primary jet momentum. Figure 32 presents the results of a series of experimental tests where the primary/secondary mixing was changed by varying the primary jet velocity for the axial injector. Both lines in Figure 32 refer to data taken at increasing primary air flow rates (while the total air and hence excess $O_2$ were held constant.) The upper line is data obtained in air while the lower line is data from combustion in $Ar/O_2/CO_2$. As the primary air flow rate is increased, the primary velocity increases and ultimately the flame lifts off the injector and stabilizes at some point downstream (due to the hot refractory walls.) For example with 14% primary air the flame was stabilized (luminous zone begins) approximately 9½" below the injector. This increases the mean particle heating time by at least one order of magnitude and allows for entrainment of considerable secondary air prior to combustion. Again the $Ar/O_2/CO_2$ data suggest that the large increases are due to increased fuel nitrogen conversion.

Figure 33 shows the effect of secondary air swirl on total NO emissions for the Pittsburgh and Western Kentucky coals. In both

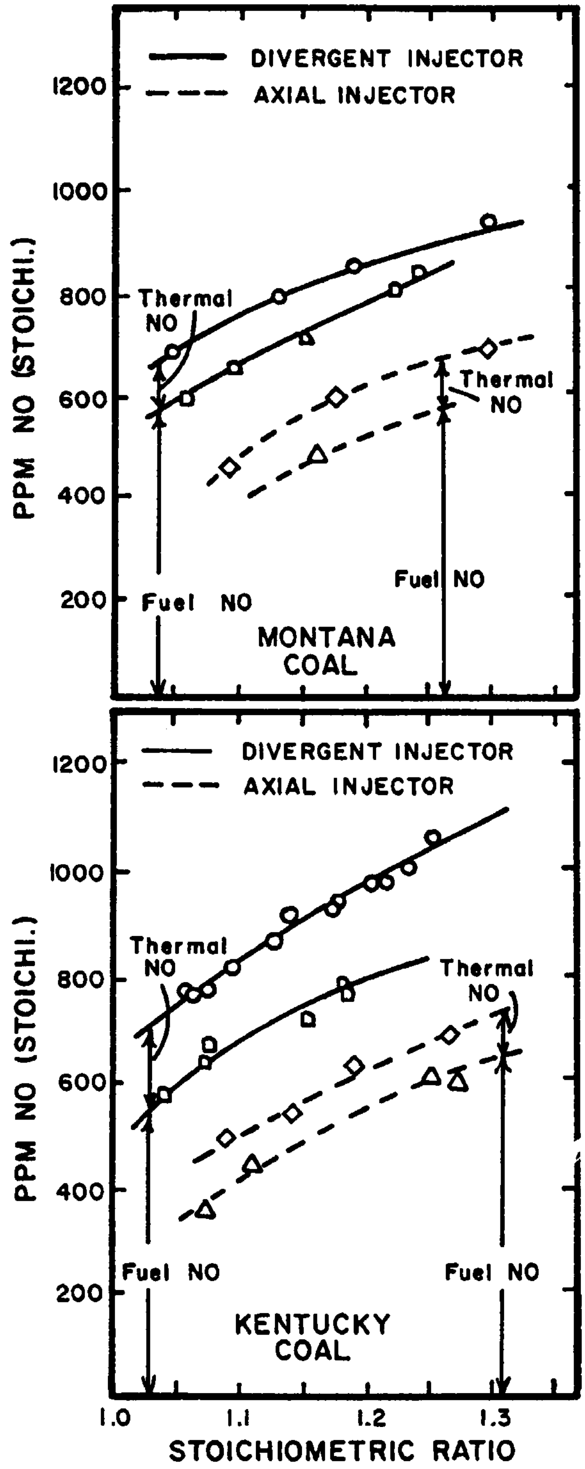

FIGURE 31. EFFECT OF FUEL INJECTOR DESIGN - THERMAL AND FUEL NO (MONTANA COAL, 550°F PREHEAT; WESTERN KENTUCKY COAL, 650°F PREHEAT)

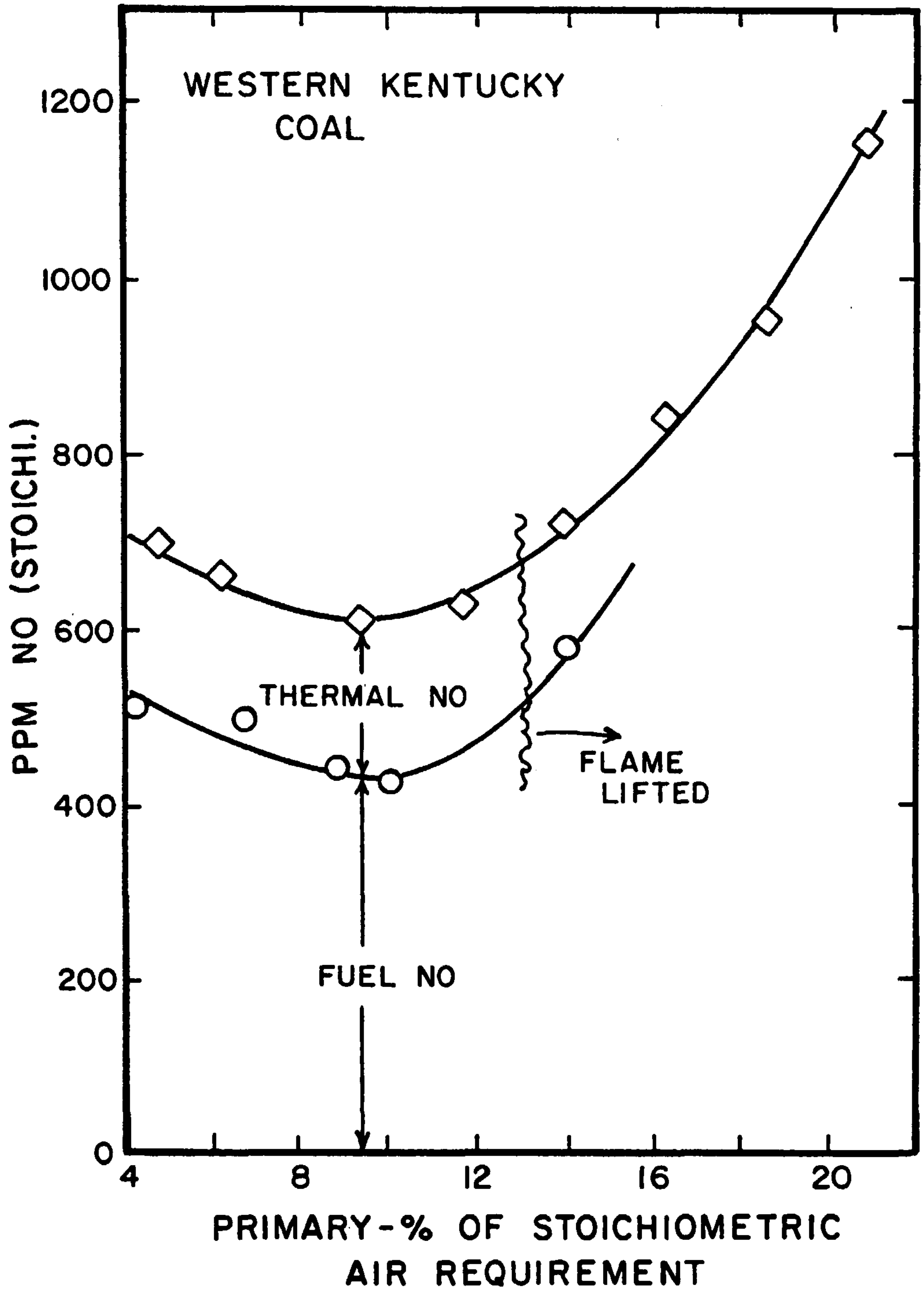

FIGURE 32. EFFECT OF PRIMARY AIR FLOW RATE (WESTERN KENTUCKY COAL, AXIAL INJECTOR, 600°F PREHEAT, 15% EXCESS AIR)

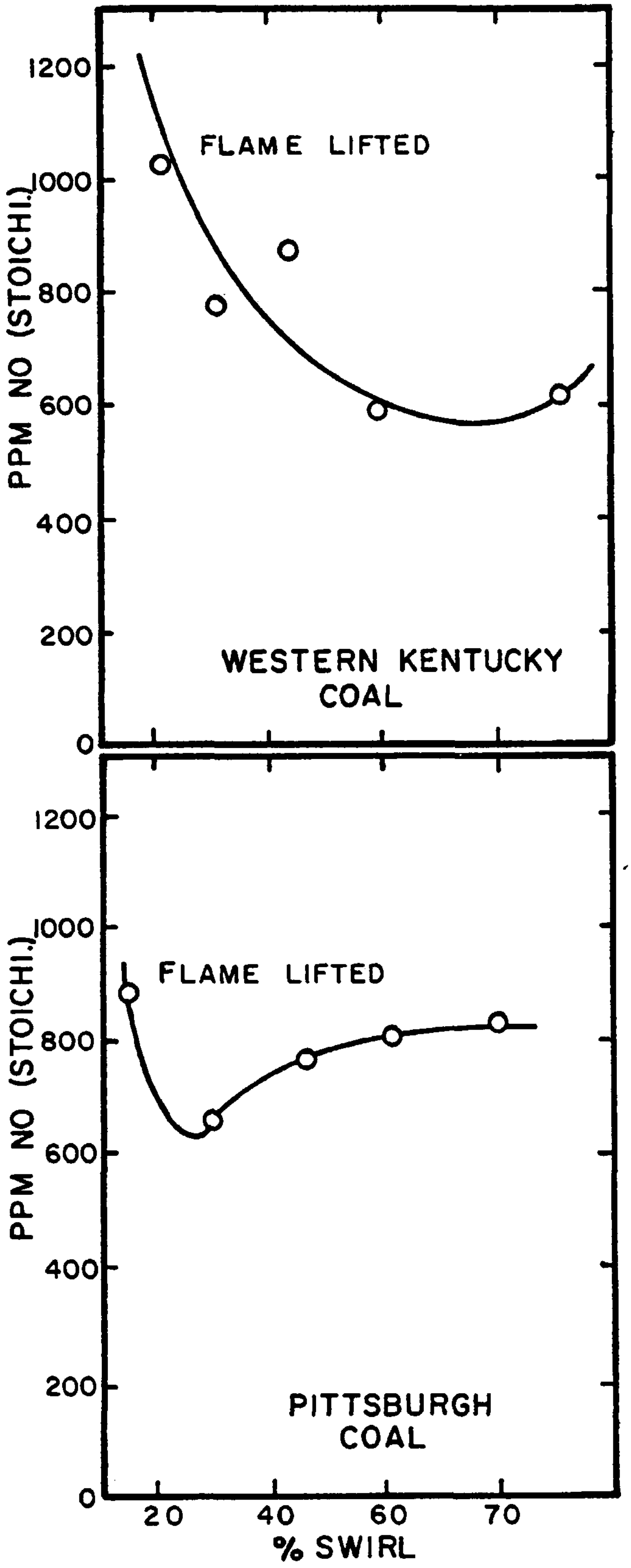

FIGURE 33. EFFECT OF SECONDARY AIR SWIRL (PITTSBURGH AND WESTERN KENTUCKY COALS, DIVERGENT INJECTOR, 15% EXCESS AIR)

cases dramatic increases in NO emissions were observed when the swirl was reduced to the point where the flame was no longer stable on the fuel injector. The NO increases can probably be attributed to increased entrainment prior to ignition and hence greater oxygen availability.

Figure 34 shows that reducing the secondary air preheat from 510°F to 155°F had a very unusual effect on the NO emissions from the Colorado coal. Above approximately 25% excess air the emissions decreased slightly as the temperature was lowered. This is consistent with the data reported in earlier sections on the other coals and with the data of Armento (1975) and Heap *et al.* (1975). At the lower excess air levels the 155°F preheat data is considerably higher than the baseline case and it was noted visually that the "flame" was completely detached from the burner in which case ignition took place at some distance down in the cylindrical chamber. The apparent increase in NO emissions is, therefore, probably due to a major change of local oxygen availability (because of mixing prior to ignition) rather than the decreased air preheat temperature.

Figure 35 shows total NO emissions as a function of excess air for both attached and lifted Western Kentucky coal flames. The data suggest a significant difference in the dependence on excess air.

In summary the data on combustion fluid dynamics suggests that any change which delays the ignition and allows considerable premixing of the coal and secondary air will markedly increase NO emissions.

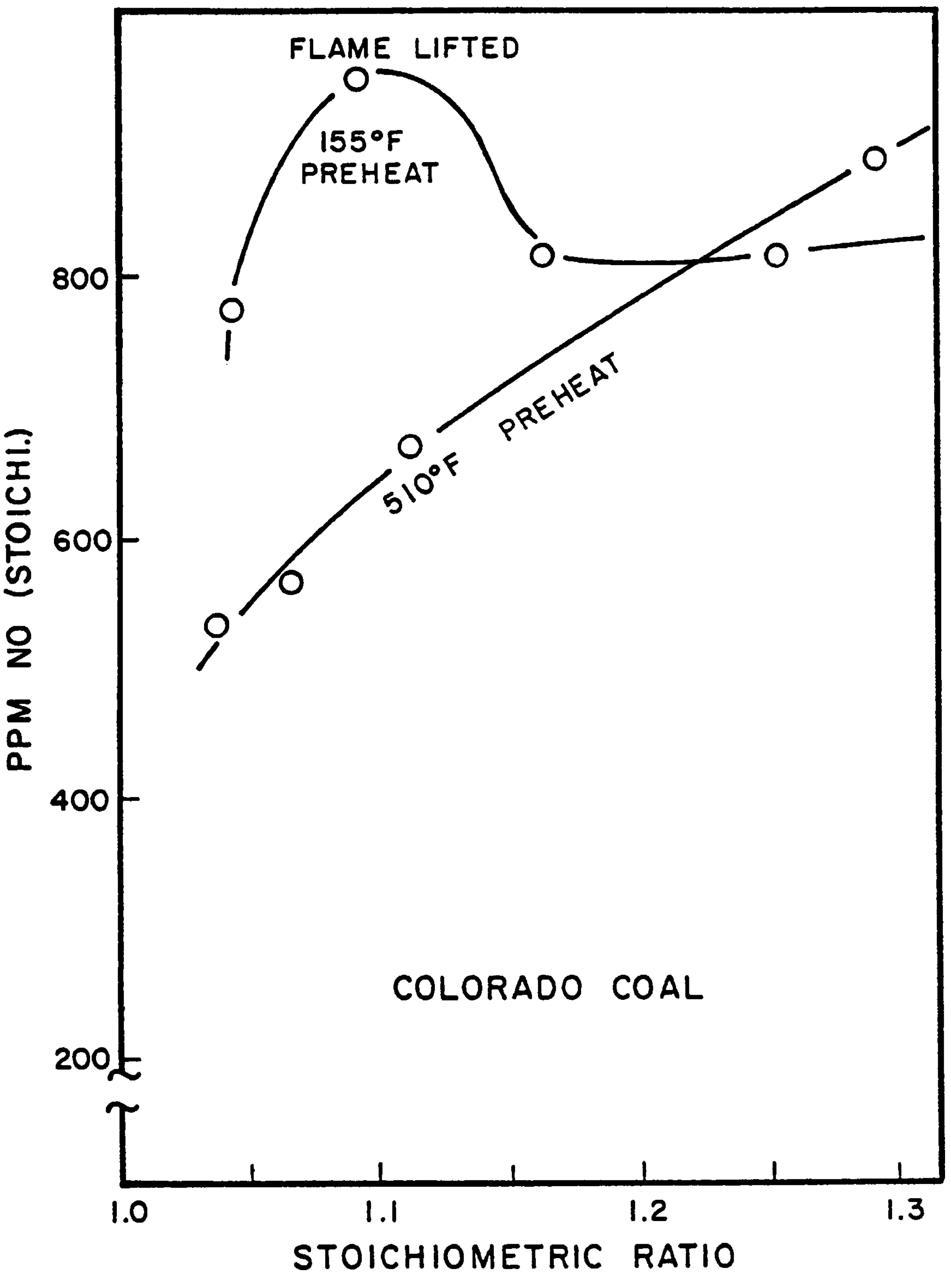

FIGURE 34. AERODYNAMIC EFFECT OF AIR PREHEAT (COLORADO COAL, DIVERGENT INJECTOR)

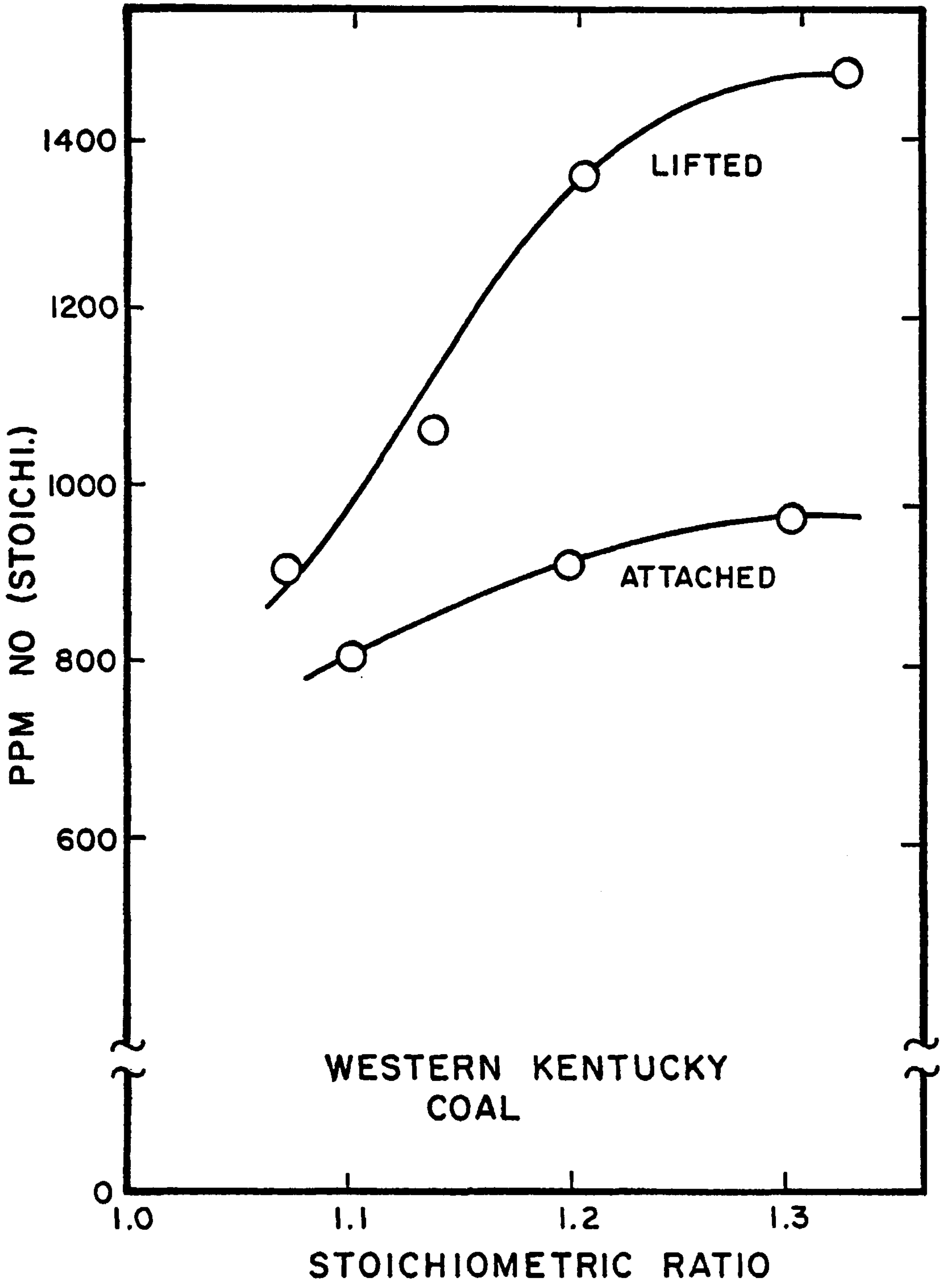

FIGURE 35. COMPARISON OF LIFTED AND ATTACHED FLAMES (WESTERN KENTUCKY COAL, DIVERGENT INJECTOR)

# REFERENCES

Armento, W.J., "Effects of Design and Operation Variables on $NO_x$ Formation in Coal Fired Furnaces". Babcock and Wilcox Company, Alliance, Ohio, Environmental Protection Technology Series Report EPA-650/2-74-002b (1975).

Axworthy, A.E., paper presented at the EPA Stationary Source Combustion Symposium, Atlanta (1975).

Bittner, J.D., R.A. Hites, J.B. Howard, P.H. Lin and G.P. Prado, paper presented at the EPA Stationary Source Combustion Symposium, Atlanta (1975).

Blair, D.W., J.O.L. Wendt, and W. Bartok, "On the Evolution of Nitrogen and Other Species During Controlled Pyrolysis of Coal", paper presented at the Sixteenth Symposium (International) on Combustion, Boston (1976)

Crawford, A.R., E.H. Manny, and W. Bartok, "Field Testing: Application of Combustion Modifications to Control $NO_x$ Emissions from Utility Boilers." Exxon Research and Engineering Company, Linden, New Jersey, Environmental Protection Technology Series Report EPA-650/2-74-066 (1974).

Crawford, A.R., E.H. Manny, M.W. Gregory, and W. Bartok, paper presented at the EPA Stationary Source Combustion Symp., Atlanta (1975).

DeSoete, G., Revue del Institut Francois du Petrole, 28 95 (1973).

Dykema, O.W. and R.E. Hall, paper presented at the EPA Stationary Source Combustion Symp., Atlanta (1975).

Flagan, R.C., S. Galant, and J.P. Appleton. Combustion and Flame, 22, 249 (1974).

Habelt, W.W. and B.M. Howell, paper presented at the EPRI $NO_x$ Control Technology Seminar, San Francisco (1976).

Heap, M.P., T.M. Lowes and R. Walmsley, Combustion Institute European Symposium 1973, ed. F.J. Weinberg, Academic Press, New York, p. 493 (1973).

Heap, M.P., T.J. Tyson, and T.M. Lowes, paper presented at the 68th Annual AIChE Meeting, Los Angeles (1975).

Horton, M.D., F.P. Goodson, and L.D. Smoot, paper presented at the 68th Annual AIChE Meeting, Los Angeles (1975).

Lachapelle, D.G., paper presented at the EPRI $NO_x$ Control Technology Seminar, San Francisco (1976).

Lilley, D.G. and J.O.L. Wendt "Modeling Pollutant Formation in Coal Combustion" Proceedings of the 1976 Heat Transfer and Fluid Mechanics Institute (Ed McKillop) Stanford University Press (to be published).

Martin, B.G. and E.E. Berkau, AICHE Symp. Ser. No. 126, 68, 45 (1972).

Merryman, E.L. And A. Levy, paper presented at the EPA Fund. Comb. Research Contractors Meeting, Menlo Park, California (1975).

Pereira, F.J., J.M. Beér, B. Gibbs, and A.B. Hedley, Fifteenth Symp. (Int) on Combustion, p. 1149, The Combustion Institute, (1974).

Pershing, D.W., G.B. Martin, and E.E. Berkau, AIChE Symp. Ser. No. 148, 71, 19 (1975).

Pershing, D.W. and J.O.L. Wendt "Pollutant Control through Staged Combustion of Pulverized Coal" U.S. ERDA Report No. 1817-1. Available through US ERDA Technical Information Center, Oak Ridge, Tenn. (1975).

Pohl, J.H. and A.F. Sarofim, paper presented at the EPA Stationary Source Combustion Symp., Atlanta (1975).

Sarofim, A.F., G.C. Williams, M. Modell and S.M. Slater, AIChE Symp. Ser. No. 148, 71, 51 (1975).

Sawyer, R.G., Paper presented at the EPA Fund. Comb. Research Contractors Meeting, Menlo Park, California (1975).

Sternling, D.V., and J.O.L. Wendt, Environmental Protection Tech. Ser. Report EPA-650/2-74-017 (1972).

Thompson, R.E., paper presented at EPRI $NO_x$ Control Technology Seminar, San Francisco (1976).

Wendt, J.O.L. and T.C. Corley, paper presented at the Third Annual EPA Fund. Comb. Research Contractors Meeting, Boston (1976)

Wendt, J.O.L. and C.V. Sternling, J. Air Poll. Contr. Assoc., 11, 1055 (1974).

Wendt, J.O.L. and O.E. Schulze, AIChE J., 22, 102 (1976)