A CONTROL STRATEGY FOR NO$_X$ FROM FUEL-NITROGEN

by

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ABSTRACT

Use of fuels with a high nitrogen content is hindered by the ease
with which they form NO_x. Under fuel lean operation, typically 80-100
percent of the fuel-nitrogen is converted to NO_x. For a characteristic
synthetic fuel with a nitrogen concentration of 1 - 2 percent this repre-
sents the dominant NO_x source. The conversion of fuel-nitrogen to NO_x is
insensitive to flame temperature under lean conditions so dilution with
an inert gas is ineffective, leaving only two stage combustion as a con-
trol technique.

Data from an experimental burner simulating the rich stage of a
staged combustor indicates that it is the summation of fixed-nitrogen-
species which is the important "measuring stick" in evaluating a flame's
emission potential. Raising the flame temperature by preheating the in-
let air will cause a reduction in the summation of fixed-nitrogen-species
in rich flames. The reduction only occurs over a narrow range of equiva-
lence ratios and only for flame conditions with low concentrations of un-
burned hydrocarbons at burner exhaust. This requires that the fuel and
air must be intensely mixed at the entrance of the burner. Any flame
which resulted in soot formation heavy enough to turn the flame bright
yellow yielded high concentrations of unburned hydrocarbons and HCN. For
rich flames this resulted in a high summation of fixed-nitrogen-species
because HCN was the dominant nitrogen containing species of those flames.
Although the summation of fixed-nitrogen-species could be reduced by
raising the inlet air temperature, it is shown to still be constrained by
both mixing and kinetic constraints. The measured concentrations of the
fixed-nitrogen-species from the turbulent diffusion flame studied here,
are higher than those of premixed laminar flames of similar conditions
which in turn are higher than equilibrium values.

1. INTRODUCTION

For combustors operating lean, NO_x from oxidation of fuel nitrogen alone can be close to, or exceed, the emission standards even before the inclusion of the contribution of NO_x formed by the oxidation of atmospheric nitrogen. Combustion of shale oils would yield emissions in excess of existing standards, unless the nitrogen content of the oils was reduced by processing.

The anticipated increase in the use of fuels derived from coal and shales provides motivation for developing combustion processes that yield low NO_x levels when fired with fuels having high nitrogen content.

2. IDENTIFICATION OF FACTORS EFFECTING NO_x FORMATION

2.1 NO_x from Two Sources

The combustion of fuels containing nitrogen gives rise to two sources of NO_x, that from the oxidized molecular nitrogen which is present in the combustion air (thermal-NO_x), and that from the nitrogen in the fuel (fuel-NO_x). Because the problem of fuel-NO_x has only recently become of interest, it is not very well understood. Furthermore, the control strategies for NO_x in use today are based on control of thermal rather than both thermal and fuel NO_x[1]. It has been shown experimentally[2] that reduction of flame temperature, which is effective in control of thermal NO_x, is not very effective for controlling the NO_x formed from fuel-nitrogen.

Studies by Fenimore[3] and Sarofim et al.[4] show the effect of different flame conditions on the yield of NO_x from the fuel nitrogen. Using these studies as a guide, it is felt that the control of NO_x from nitrogen containing fuels would be obtained with two stage combustion.

2.2 Equilibrium vs. Measured Concentrations

Equilibrium concentrations of NO_x in rich combustion products are low. At an equivalence ratio of 1.5 for a typical fuel (kerosene with 1% nitrogen by mass) the equilibrium concentration of HCN, NH_i (i = 0,1,2,3) and NO together account for one half of one percent of the fuel-nitrogen. The remaining 99.5% has been converted to N_2.

In the flame studies of Fenimore and Haynes[5,6,7] the concentration of the summation of HCN, NH_i and NO (hereafter referred to as the summation of fixed-nitrogen-species, or fixed-N) were found to be two to three orders of magnitude above the equilibrium values. There must be a rate limiting constraint governing the reaction kinetics. Therefore by raising the flame temperature it might be possible to overcome the limitation and drive the products closer to equilibrium, thus reducing NO.

2.3 Turbulent Diffusion Flames

Up to this point the identification of factors affecting NO_x formation has been based on kinetic considerations only. In a practical combustor the flame will be a turbulent diffusion flame and not the premixed laminar flame. As a result there will be an additional parameter the mixing rate which will affect the NO_x emissions.

In a turbulent diffusion flame air and fuel enter the burner separately. They then mix to form a combustible mixture. It has been shown that the rate at which this mixing takes place has a direct influence on the resultant burner emissions[8,9]. Turbulent diffusion flames of the same equivalence ratio but with different mixing rates can have drastically different NO_x emissions[10].

This brings to four the number of parameters which are expected to influence the NO_x emissions from a practical combustor burning nitrogen containing fuels. They are:

1) The fuel-air equivalence ratio
2) The amount of nitrogen in the fuel
3) The inlet air temperature
4) The mixing intensity of the air and fuel in the burner

3. EXPERIMENTAL

3.1 The Burner

An experimental burner to study the rich stage of a staged combustor was built and is shown in Figure 1. It is basically a cylindrical burner in which the combustion air is supplied at atmospheric pressure at one end through a 45-degree blade angle swirler. Liquid fuel was supplied through an air assist atomizer on the burner axis. The atomizer nozzle air-pressure drop was controlled independently of the fuel flow rate. The fuel and air induction system upstream of the swirler vanes is symmetric and contoured to minimize the propagation of flow assymmetries into the burner. Upstream of the burner is an 85 kW air preheater capable of heating the primary combustion air from room temperature (300°K) to 733°K.

Independent control of the parameters effecting NO_x emissions is achieved by varying the fuel flow, changing the nitrogen content of the fuel, using the air preheater and changing the atomizing pressure of the air assist atomizer. For the first three listed, it is obvious how they relate to their respective control parameters. How the atomizing pressure effects the air fuel mixing has been explained elsewhere[11,12].

Downstream of the burner is an afterburner section where dilution air can be added to the combustion products of the burner. The purpose

of this section was strictly safety in the work reported here. Attempts to sample in the afterburner section proved inconclusive because the dilution air was poorly mixed with the combustion products.

Exhaust samples were drawn continuously into a heated teflon sample line from a water cooled, quartz tipped probe. Care was taken to maintain the probe and sample line temperature above the dew point of the exhaust sample, which was estimated to be 52°C. The exhaust sample rate was held constant at 2.8 liters/min and passed through a filter of 0.5 μm porosity. The exhaust samples are therefore time mean readings.

The sampling probe and all fittings in the sampling line were teflon coated to prevent the exhaust gases from coming into contact with the stainless steel walls of the probe or the brass walls of the fittings. This was done to prevent the hydrogen in the rich combustion products from dissociating on the hot metal walls of the sampling system and re-acting with the NO to form water and possibly ammonia[13,14]. The same problem can occur in the NO_2 to NO converters of chemiluminescent NO_x analyzers[15,16]. For this reason all data for rich flames is measured as NO (there will be very little NO_2 in rich flames).

The species; O_2, CO, CO_2, NO, HCN and NH_3 were monitored. All species except HCN and NH_3, which were measured by wet chemical techniques, were continuously monitored with standard commercially available instruments.

3.2 Fuels Used

One of the major assumptions made in this work was that real synthetic fuels which would have a nitrogen content on the order of 1% could be simulated by a base fuel plus a nitrogen doping compound. Except for the

nitrogen and sulfur contents, it appears that kerosene and a synthetic fuel with the same API gravity and boiling range would have the same burning characteristics.

The nitrogen content of the fuel was simulated with the addition of pyridine which is one of the forms in which nitrogen is found in the synthetic fuels.

3.3 Burner Characterization

The burner was run with constant total air flow in all experiments. The primary air flow, which passes through the swirler vanes, was adjusted so that the total of the atomizing air flow and primary air flow was equal to 56.7 kg/hr. At the highest atomizing pressure (446 kPa (50 psig)) the atomizing air flow accounted for 14 percent of the total flow.

It should be emphasized at this point that all exhaust samples are for the rich stage of a staged combustor. When statements are made in regard to the exit of the burner they are synonymous to statements regarding the exit from the rich stage of a staged combustor.

4. RESULTS

4.1 Experimental Matrix

Baseline data was taken over a wide range of operating conditions to isolate particular operating conditions for more detailed investigation. The initial experimental matrix covered the following range of variables:

$$1.0 < \bar{\phi} < 2.0$$

$$300°K < T_{in} < 733°K$$

$$204 \text{ kPa } (15 \text{ psig}) < \Delta P < 446 \text{ kPa } (50 \text{ psig})$$

$$\%N = 0, 1, 5$$

Additive type: Pyridine, dodecylamine

For each operating condition either exit $(\frac{x}{D} = 5.0)$ or axial exhaust samples were taken. The data taking procedure involved drawing exhaust samples from six to ten different radial positions at each axial probe location. These readings were then averaged on an area basis.

4.2 Baseline Experiments

Figure 2 shows a compilation of data for kerosene and kerosene doped with pyridine to 1% N by mass at a constant atomizing pressure of 274 kPa (25 psig) over the range of equivalence ratios $1.0 < \phi < 2.0$ and inlet air temperatures of $300°K < T_{in} < 733°K$. For this compilation neither ammonia nor hydrogen cyanide were measured. The only nitrogen containing species that was monitored was NO. Note that in all cases increasing the inlet air temperature increases the NO concentration at burner exhaust. This increase is observed at all equivalence ratios whether or not there has been nitrogen added to the fuel. It is observed that at equivalence ratios greater than 1.3 the NO concentrations are super equilibrium. An estimate of the percent of conversion of fuel-N to NO can be made by subtracting the undoped measurement from the doped one and comparing it to the 100 percent conversion line of fuel-N to NO. When this was done it was found that for equivalence ratios in excess of 1.3 the percent of fuel-N which can be accounted for by the NO measurements is less than five percent and remains approximately constant with increasing inlet air temperature. Clearly it is important to know the form of the remaining fuel-N. If the remaining fuel-N were in other fixed-nitrogen-species

such as NH_3 or HCN, it would very rapidly oxidize to NO when brought into the presence of additional oxygen, as would be the case in an afterburner or secondary stage of the combustor. However, if the balance of the fuel-N were N_2 the addition of secondary air would result in little or no additional NO formation and the present data would be representative of the NO emissions of a two stage combustor operating at an overall stoichiometry of unity or less.

4.3 Fixed-N-Species Distribution

Table 1 shows the distribution of fixed-nitrogen-species at three different equivalence ratios for burner flames of kerosene with 1% nitrogen, at high atomizing pressure with room temperature inlet air. There is a very drastic change in the distribution in going from an equivalence ratio of 1.3 to 1.5. At an equivalence ratio of 1.3 the majority of the fixed-nitrogen is NO, however at 1.5 and 1.7 the majority of the fixed-nitrogen is HCN. By comparing the total of the fixed-nitrogen-species, an indication of the control potential of a particular operating point is obtained. For these conditions concluding that an equivalence ratio of 1.5, based on NO measurements alone, is the optimal operating point is totally erroneous.

In the baseline data it was also observed that raising the inlet air temperature caused an increase in the NO emissions. With this knowledge further investigation of the equivalence ratio 1.3 at higher inlet air temperature is superfluous. However, the effect of inlet air temperature on the fixed-nitrogen distributions at equivalence ratios of 1.5 and 1.7 is of interest and cannot be surmised apriori.

4.4 Operation at Equivalence Ratio $\phi = 1.5$

Now a general operating condition for the burner has been attained, more detail about the behavior of the different species for ϕ greater than 1.3 is desired. Figures 3, 4, and 5 show the fixed-nitrogen-species for the burner operating on kerosene with a nitrogen concentration of 1% by mass (added as pyridine) at an equivalence ratio of 1.5. The unburned hydrocarbons which were not shown in Table 1 have also been displayed.

Figure 3 shows the exit concentrations of the different species at different inlet air temperatures. Notice that increasing the temperature decreases the summation of fixed-nitrogen-species. Even though the NO increases with temperature its effect on the summation of nitrogen species is more than offset by the decrease in HCN and NH_3.

Figure 4 shows axial profiles of the different species for combustion with 600°K inlet air. All of the species concentrations except NH_3 decay as the combustion products move down the burner, unburned hydrocarbons and HCN showing the strongest decay. The decay of HCN and NO is greater than the production of NH_3 so the summation of fixed-nitrogen species also decays as the gases move down the burner. This behavior is similar to behavior observed by Fenimore[5], Haynes[6,7] and most recently Taylor[19] in laminar flat flame investigations. In the laminar flames however there are no hydrocarbons. The appearance of unburned hydrocarbons and their subsequent behavior is specific to the turbulent diffusion flame.

The effect of atomizing pressure on the different species concentrations at burner exit, $\frac{x}{D} = 5.0$, with an inlet air temperature of 600°K and equivalence ratio of 1.5 is shown in Figure 5. There is no counterpart

to this data in the premixed laminar flames. Note that in changing the atomizing pressure from 274 kPa to 446 kPa the HCN concentration changes from approximately ninety percent of the nitrogen added to five percent. The unburned hydrocarbons also show a decrease of over an order of magnitude. The NO and NH_3 also show changes with atomizing pressure but their impact on the summation of fixed-nitrogen-species is secondary compared with that caused by the change in the HCN concentrations. Also shown in Figure 5 is a curve labelled luminosity. This curve is a measure of the brightness of the flame at the different atomizing pressures and is believed to be an important flame characteristic in terms of its correlation with the summation of fixed-nitrogen-species in a flame. A more detailed discussion of the flame luminosity and the resultant emissions is presented shortly.

5. DISCUSSION

5.1 Flame Characteristics

During the course of the measurements it was observed that the visual properties of the flame changed drastically between certain burner operating conditions. In particular, flames which resulted in a low summation of fixed nitrogen species at the burner exhaust ($\phi = 1.5$, $\Delta P = 446$ kPa, $T_{in} = 600°K$) were blue and of low luminosity. Those which had a high summation of fixed nitrogen species ($\phi = 1.5$, $\Delta P = 446$ kPa, $T_{in} = 300°K$) were a very bright luminous yellow. This suggests that the appearance of the flame might be used as a criteria to evaluate its emission potential.

To document the change in appearance of the flame for different burner operating conditions, photographs were taken over the same range of

variables for which data was taken. So, now a picture of the flame can be shown for each burner operating condition studied.

Shown in Figure 6 is a series of pictures for different atomizing pressures and equivalence ratios for an inlet air temperature of 600°K. The picture in the top right hand corner; ΔP = 446 kPa, ϕ = 1.5, corresponds to the low summation of fixed-nitrogen-species. The number τA which appears under each flame photograph is the product of the shutter speed and aperature setting of the camera lens that were required to get pictures of constant exposure. This will be a number which is inversely proportional to the luminosity of the flame.

In terms of chemical species, the appearance of the luminous yellow flame is a strong indication of the presence of soot in the flame. In equilibrium calculations[20], solid carbon, soot, shows a threshold value type of behavior similar to that shown in Figure 6. Experimentally, Gaydon[21] observed what he called "the threshold value for the appearance of soot in a flame" which was a function of both equivalence ratio and flame temperature. If the luminosity of the flame is taken as a measure of the concentration of the soot in the flame, then $(\tau A)^{-1}$ can be super-imposed on the species concentration graphs as if it were a concentration measurement itself. Furthermore one might argue that since the appearance of soot in a flame is preceded by hydrocarbon formation that there will be a correspondence between the unburned hydrocarbon and soot concentra-tions. That is, the behavior of the hydrocarbon concentration will parallel the behavior of the luminosity of the flame. Finally it is ob-served that in the data presented in Section 4 there is a parallelism in

the movement between the unburned hydrocarbons and the hydrogen cyanide, so HCN should also parallel the luminosity curve.

The luminosity, $(\tau A)^{-1}$, of the flame has been superimposed on Figure 5. In Figure 5, based on the shape of the luminosity curve, an alternative shape to the HCN and HC curves would be a plateau at the high levels of HCN and HC between atomizing pressures of 204 and 274 kPa, then a very rapid decrease parallel to the luminosity curve to a lower plateau at an atomizing pressure of 446 kPa. Because the summation of fixed nitrogen species is basically controlled by the HCN concentration in this case, it too would exhibit this behavior of a sharp decline between a plateau of approximately 100% of the fuel-nitrogen to a plateau of 22% of the total fuel-nitrogen. Thus the transition from the luminous yellow flame to the low luminosity blue flame would represent an equally sharp transition from a condition of high fixed-nitrogen to low fixed-nitrogen.

5.2 Kinetic Considerations

Correlations between soot, unburned hydrocarbons and nitrogen containing species have not been extensively studied because the laminar flat flames (which are usually used for kinetic studies) have all been hydrocarbon and soot free. In this experiment sufficient data was taken to demonstrate that the nitrogen containing species observed in the rich sooting flames were attributable to the fuel-nitrogen and not the molecular nitrogen from the combustion air.

5.3 The Role of Mixing

Coincident with the need for an understanding of the important kinetics is the need for the understanding of the fluid mechanics in the

burner and its coupling with the chemical kinetics. Data has been presented which shows the effect of atomizing pressure on the burner emissions. How the atomizing pressure effects the fluid mechanics in the burner has been analyzed and quantized by other investigations[8,9].

The turbulent diffusion flame can be thought of as a collection of gas pockets or eddies which interact through both diffusion and convection. The turbulent mixing intensity is a measure of the frequency with which these interactions take place. Each of these eddies or gas packets will have a characteristic equivalence ratio. They will range from pure air (primary combustion air) to very fuel rich (eddies of the fuel spray which will be practically all fuel). By varying the atomizing air pressure the mixing intensity and equivalence ratio distribution among these eddies is changed.

The concept of a distribution of gas packets of varying equivalence ratio which are mixing and reacting simultaneously necessitates the consideration of two characteristic times, a mixing time and a chemical reaction time. If the mixing time is much larger than the chemical time the individual gas packets will approach their equilibrium concentrations between mixing, and the combustion is said to be mixing controlled. If the chemical time is much larger than the mixing time the combustion will be kinetically or rate limited. As is often the case, there will be one or more rate limiting reactions. In this case the species which are not rate limited will equilibrate themselves according to the equivalence ratio of the gas packet and the changing concentration of those species which are rate limited. They will therefore be both mixing and rate controlled.

If the reaction of a product species is mixing controlled the effect of mixing will be to generate a weighted average of the species over the equivalence ratio distribution of the fluid eddies. Plotting the species concentration of the turbulent diffusion flame adjacent to a premixed flame of similar operating conditions clearly shows the averaging effect that a distribution of equivalence ratios has on the species concentration. This was done for the data and is shown in Figure 7.

The data is plotted on a single time axis to exhibit the effect of mixing in stretching the time scale. For the laminar case a time of 2 msec represents the time at which the gases leave the flame front and commence their post flame reactions. In the turbulent flame it is approximately 15 msec before the gases leave the recirculation zone (which can be thought of as the flame front or reaction zone) and enter the post flame reaction zone. In the turbulent flame every species concentration is larger than the respective concentration in the premixed case because of the distribution of equivalence ratios, and its averaging effect.

5.4 Combined Non-Equilibrium and Mixing Effects

Figure 7 indicated that the reduction of the summation of fixed-nitrogen-species in a turbulent diffusion flame with low unburned hydrocarbon concentrations was mixing controlled. That it is also kinetically controlled can be seen by comparing the final fixed-nitrogen-species concentration in a premixed laminar flame with the equilibrium concentrations. The resultant HCN concentrations are often super equilibrium by two orders of magnitude. The extent of non-equilibrium effects for the turbulent diffusion flame can be seen in Figure 8, which is the summation

of fixed-nitrogen-species measured from the burner superimposed on the
equilibrium graph of the same species under the same conditions. The
fact that the minimum of the fixed-nitrogen occurs at two different
equivalence ratios for the equilibrium and burner profiles is probably
due to operational limitations of the burner. Being unable to operate
the burner at an equivalence ratio of 1.7 and obtain low unburned hydro-
carbons resulted in the high summation of fixed-nitrogen observed.

Another point of interest in Figure 8 is the point of intersection
between the 100 percent conversion line of fuel-N to fixed-N and the
equilibrium summation of fixed-nitrogen species. This occurs at an
equivalence ratio of 3.0. It suggests that there will be a rich limit
theoretically as well as practically. If equilibrium concentrations were
attainable and there were no practical constraints on the combustor; the
optimal operating equivalence ratio would be 2.45.

6. SUMMARY

Figure 9 represents a summary of the fixed-nitrogen-species data.
Each data point represents the summation of the fixed-nitrogen-species at
that operating condition normalized by the total concentration of fuel-
nitrogen at the same condition. It is observed that with room tempera-
ture inlet air, increasing the equivalence ratio causes an increase in
the fraction of fixed-nitrogen-species at the burner exit. However, by
raising the inlet air temperature a minimum in the summation of fixed-
nitrogen is observed. While operating rich, raising the flame temperature
seems to attenuate the kinetic constraints occurring in the system and
reduce the summation of fixed-nitrogen-species. This inturn will result

in a decrease in the NO concentration at the exit of a second stage of a staged combustor. From the figure it is also apparent that the occurrence of the minimum is confined to a relatively narrow operating range of the burner. It is likely that the location of this minimum will be combustor dependent. Axial profiles of the burner under different operating conditions demonstrated that at equivalence ratios greater than 1.5, HCN is the dominent species leaving the recirculation zone of the flame.

Review of the data led to a correlation between the appearance of the flame, the unburned hydrocarbons, and the HCN concentration. Blue, low luminosity flames had low concentrations of unburned hydrocarbons and HCN. For $\phi = 1.5$ where HCN was the dominate fixed-nitrogen-species this meant a low summation of fixed-nitrogen at burner exit. Luminous yellow flames had high concentrations of hydrocarbons, HCN, and summation of fixed-nitrogen-species. Attributing the flame luminosity to soot implies a possible link between the soot loading of the burner, the unburned hydrocarbon concentration, and the HCN concentration; for rich flames ($\phi = 1.5, 1.7$) this would also indicate the summation of fixed-nitrogen-species.
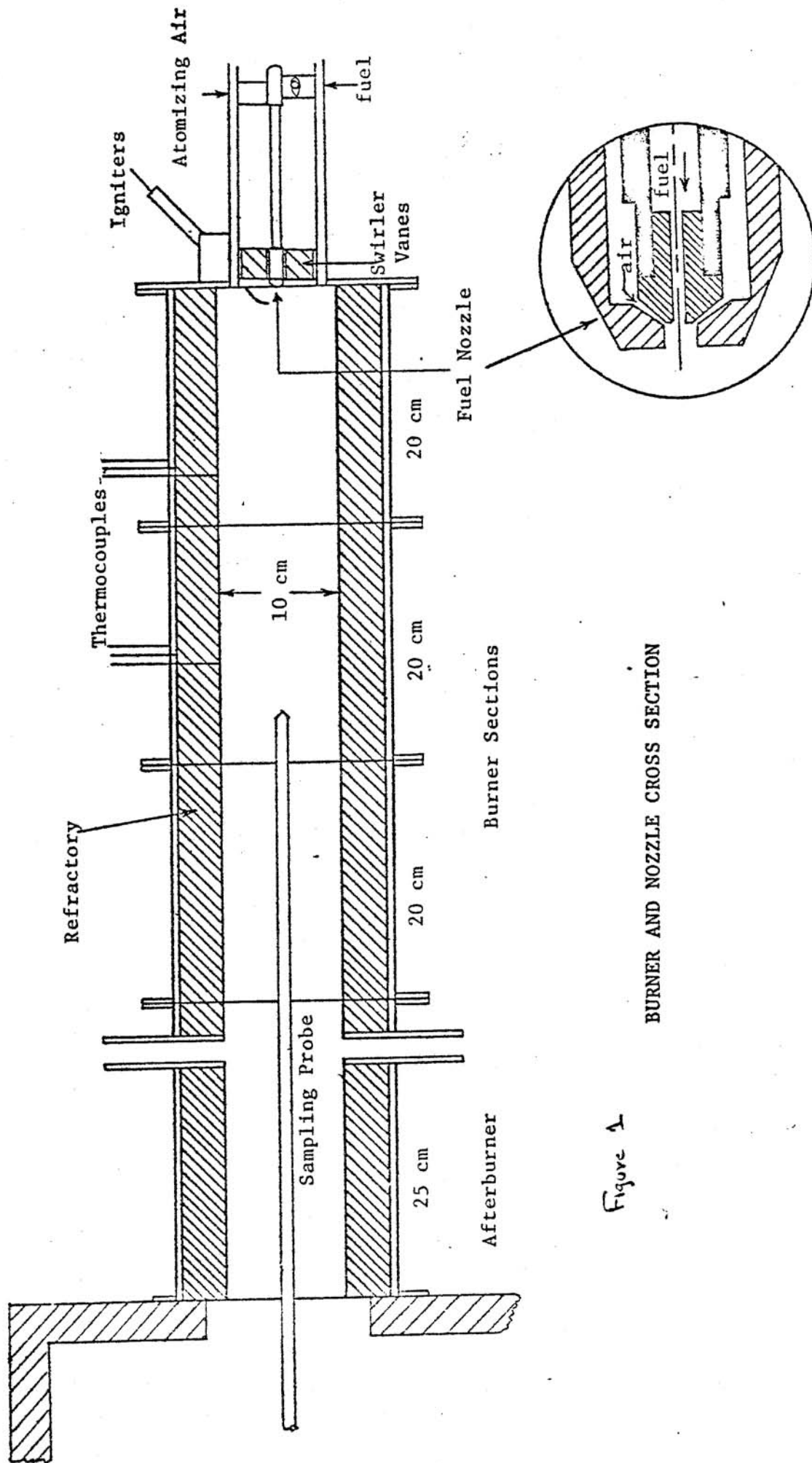
The inability to obtain a low luminosity blue flame at equivalence ratios greater than 1.5 could be the reason that the summation of fixed-nitrogen-species were minimized over such a narrow range of equivalence ratios. At equivalence ratios of 1.7 and above the flame remained luminous yellow for all atomizing pressures and inlet air temperatures. So, the narrow range of equivalence ratios at which the summation of fixed-nitrogen can be reduced could be due to the operational range of the burner and not a limitation on the control strategy.

Finally, it was observed that even though raising the inlet air temperature from room temperature to 600°K reduced the summation of fixed-nitrogen-species at $\phi = 1.5$, the resultant summation was still super equilibrium by one and a half orders of magnitude. The concentration of the fixed-nitrogen-species at burner exit is therefore controlled by both mixing and kinetic constraints.

One parameter which was not investigated was the effect of combustion gas residence times longer than the approximate 50 msec which were spent in the present combustor.

ACKNOWLEDGEMENT

Igniters

Atomizing Air

fuel

Swirler
Vanes

Fuel Nozzle

air

fuel

Thermocouples

10 cm

20 cm

20 cm

Refractory

Burner Sections

20 cm

Sampling Probe

25 cm

Afterburner

Figure 1

BURNER AND NOZZLE CROSS SECTION
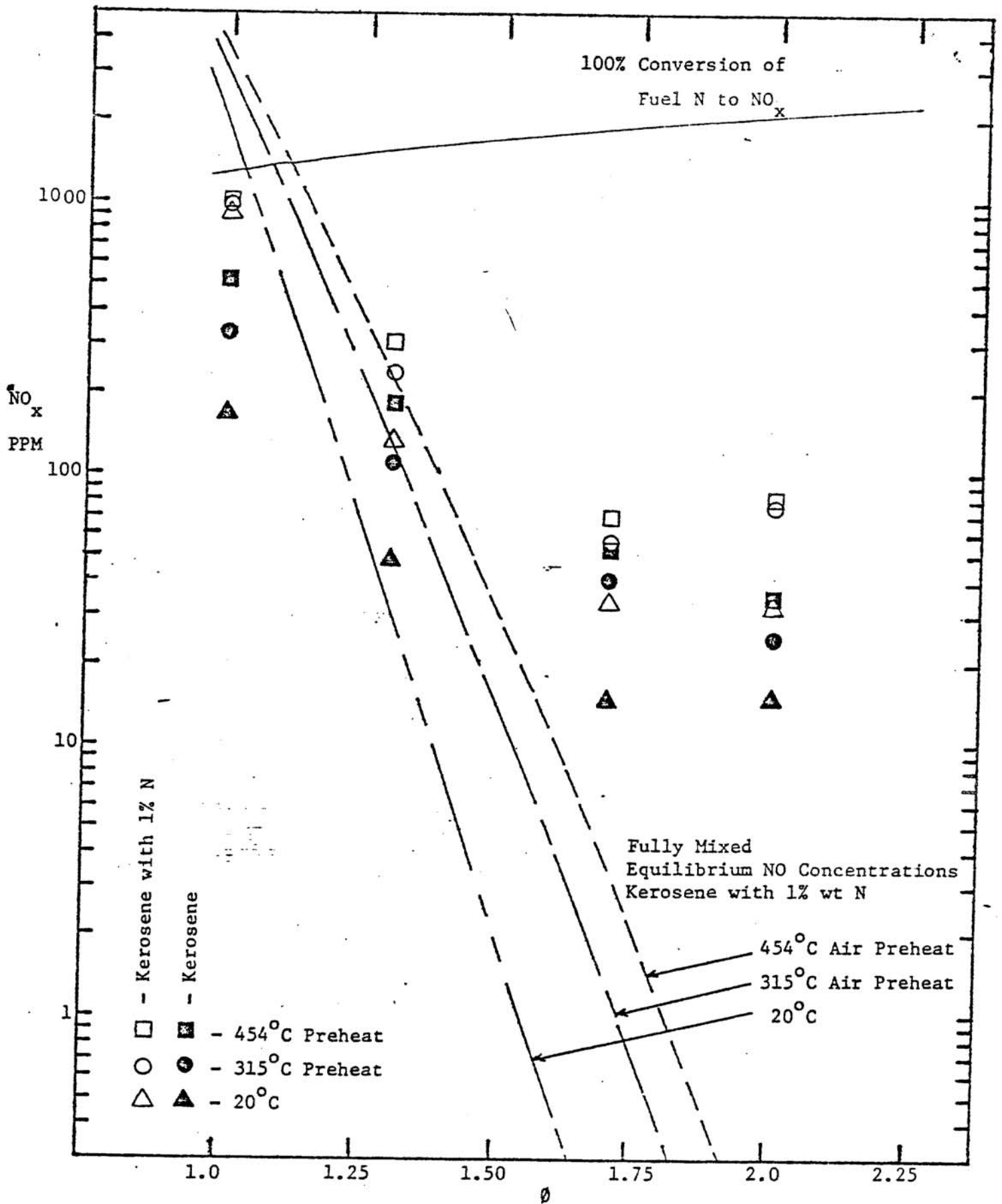
Figure 2. Exhaust NO_x Concentrations for Kerosene and Kerosene with 1% Nitrogen, 5 Diameters from the Nozzle, 1.7 atm Atomizing Pressure

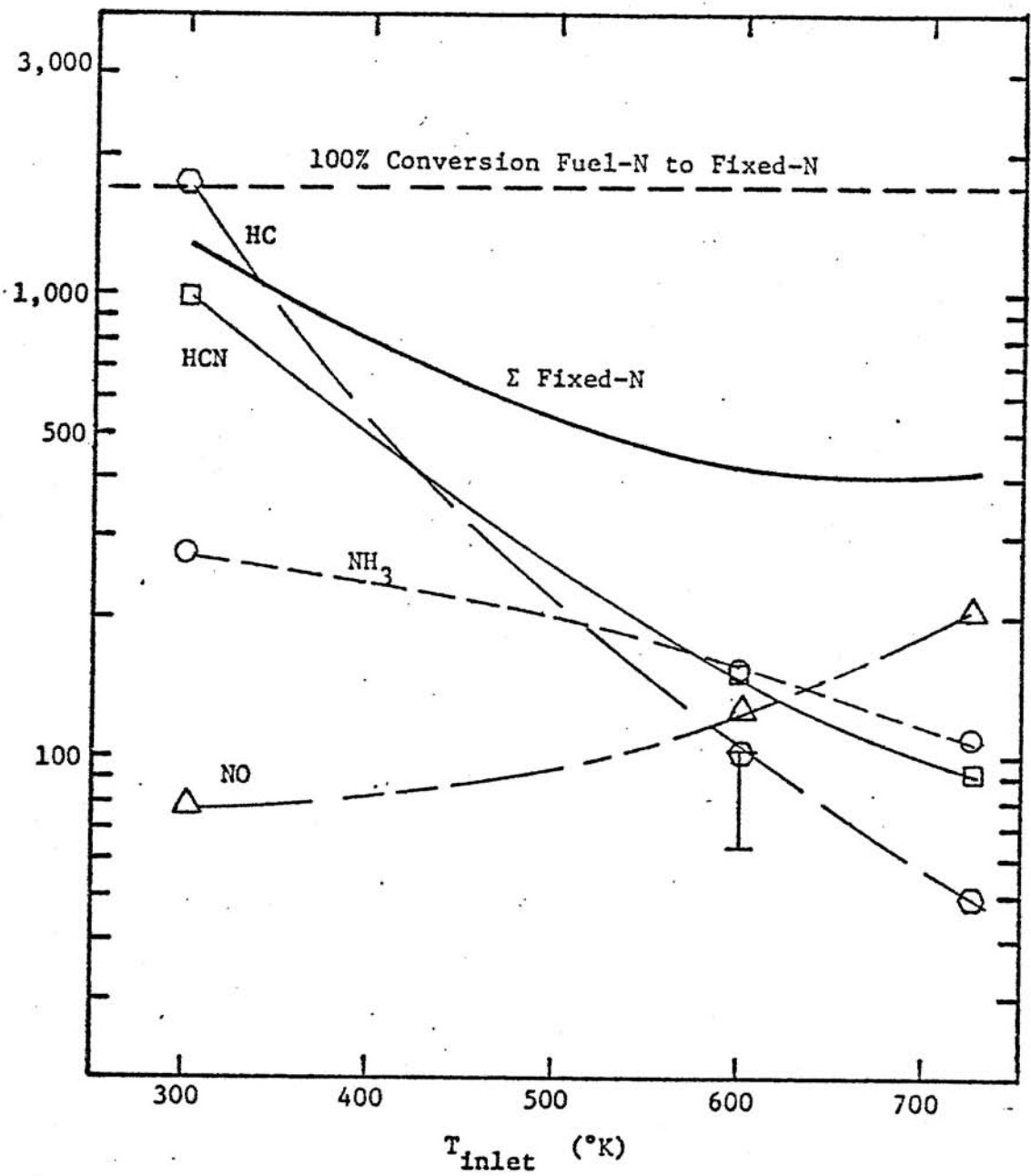Figure 3   Effect of Temperature on Species Concentration: kerosene
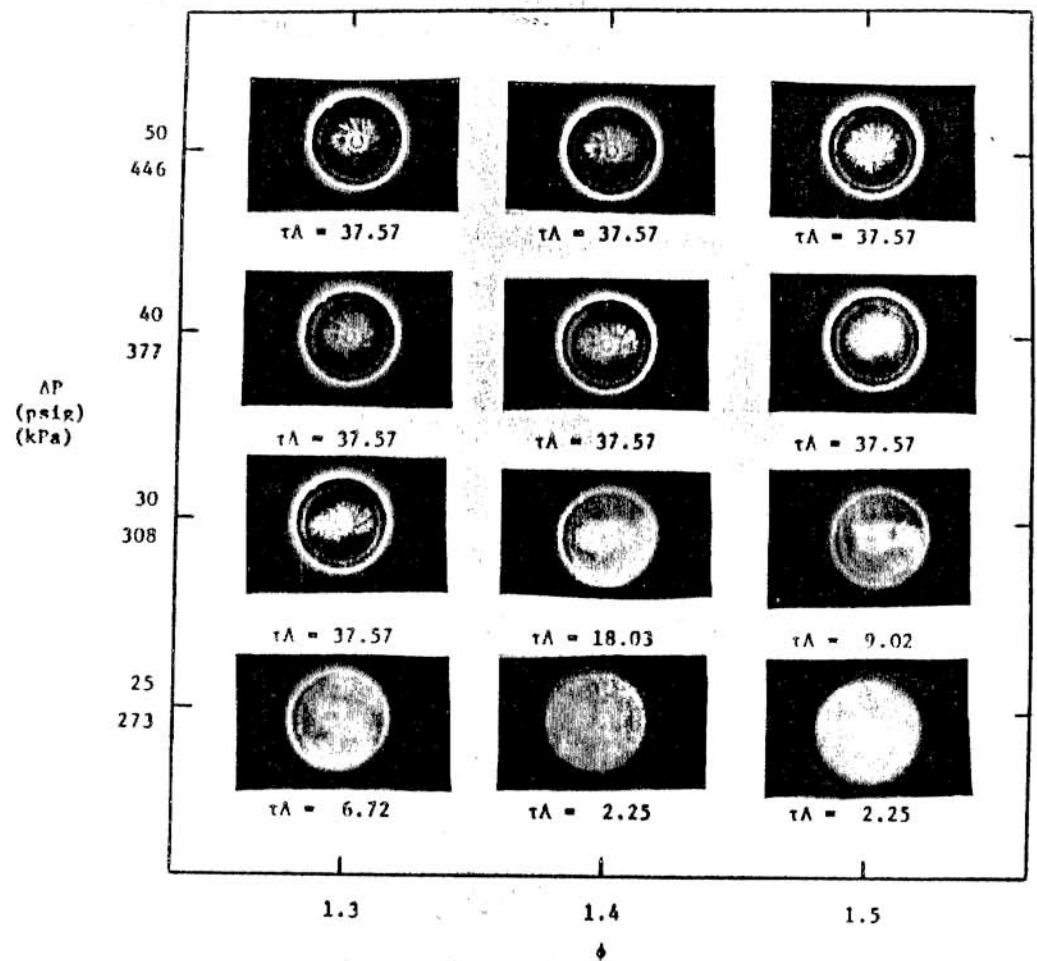with pyridine to 1% N by mass, ΔP = 446 kPa, X/D = 5.0
ϕ = 1.5
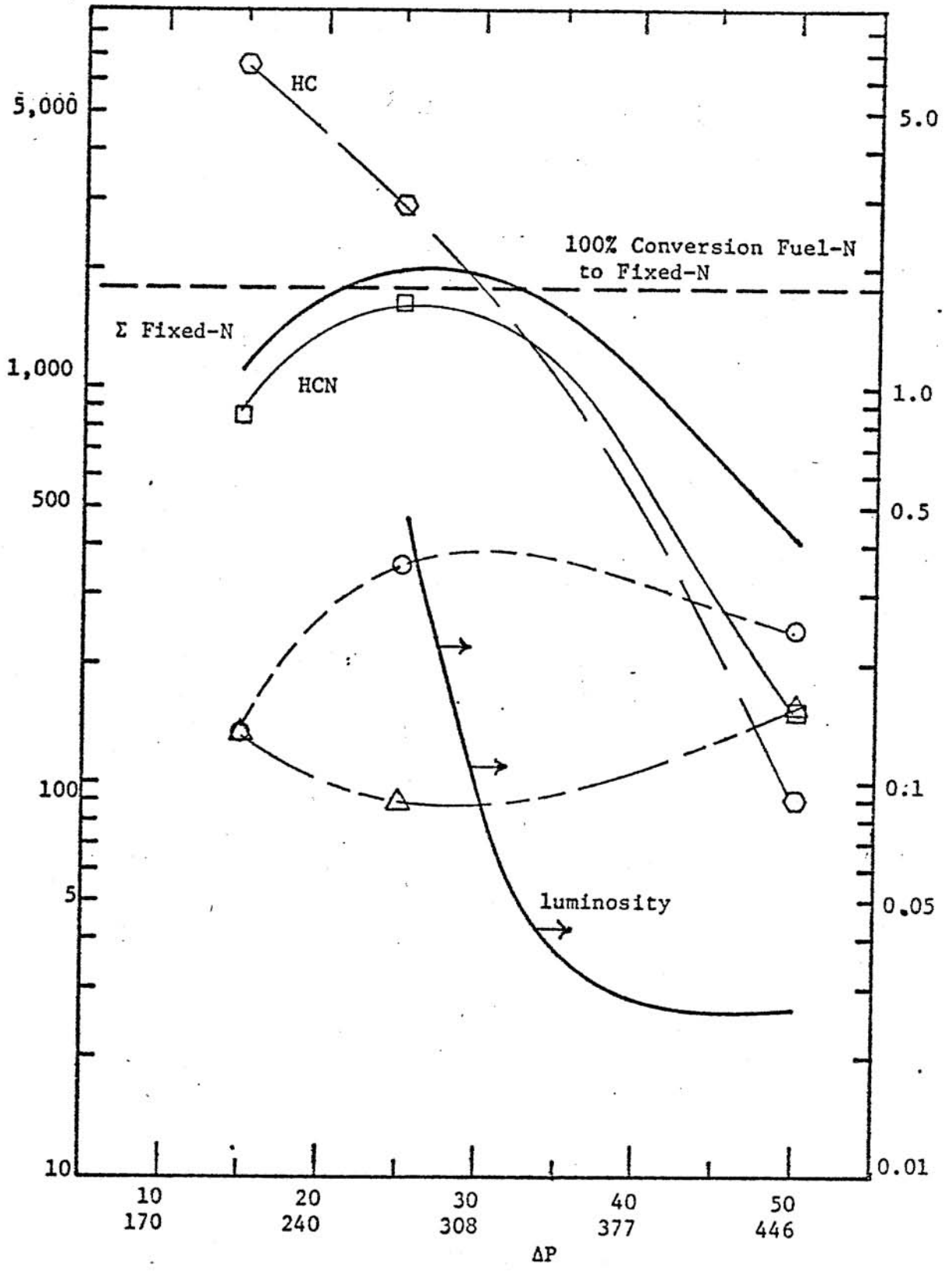
Figure 5

Effect of Mixing on Fixed Nitrogen Species: kerosene with
1% N as pyridine, $\phi = 1.5$, $T_{in} = 600°K$, $X/D = 5.0$

Flame Photographs: τA = shutter speed x aperture area, kerosene, T_{inlet} = 600°K.
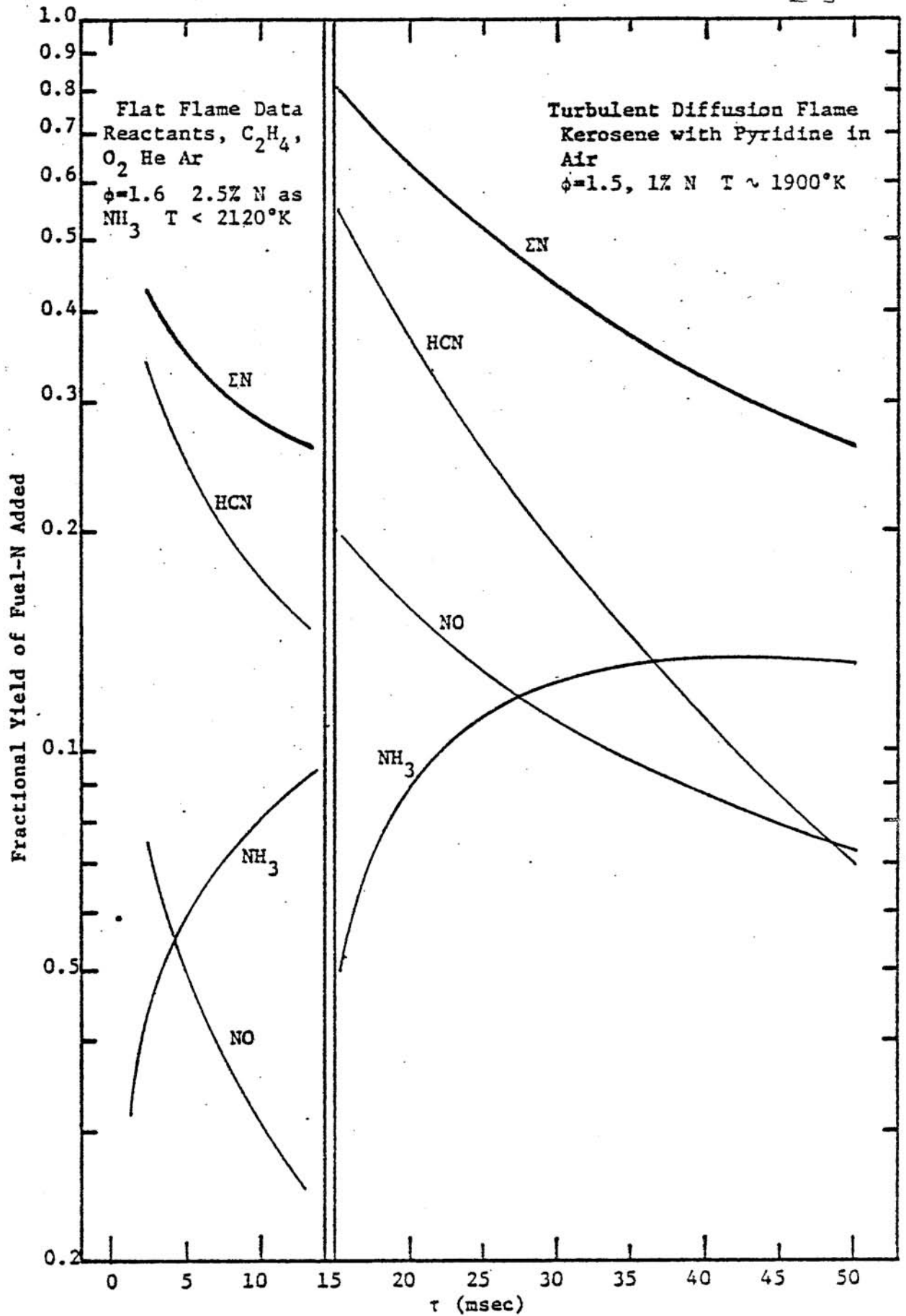
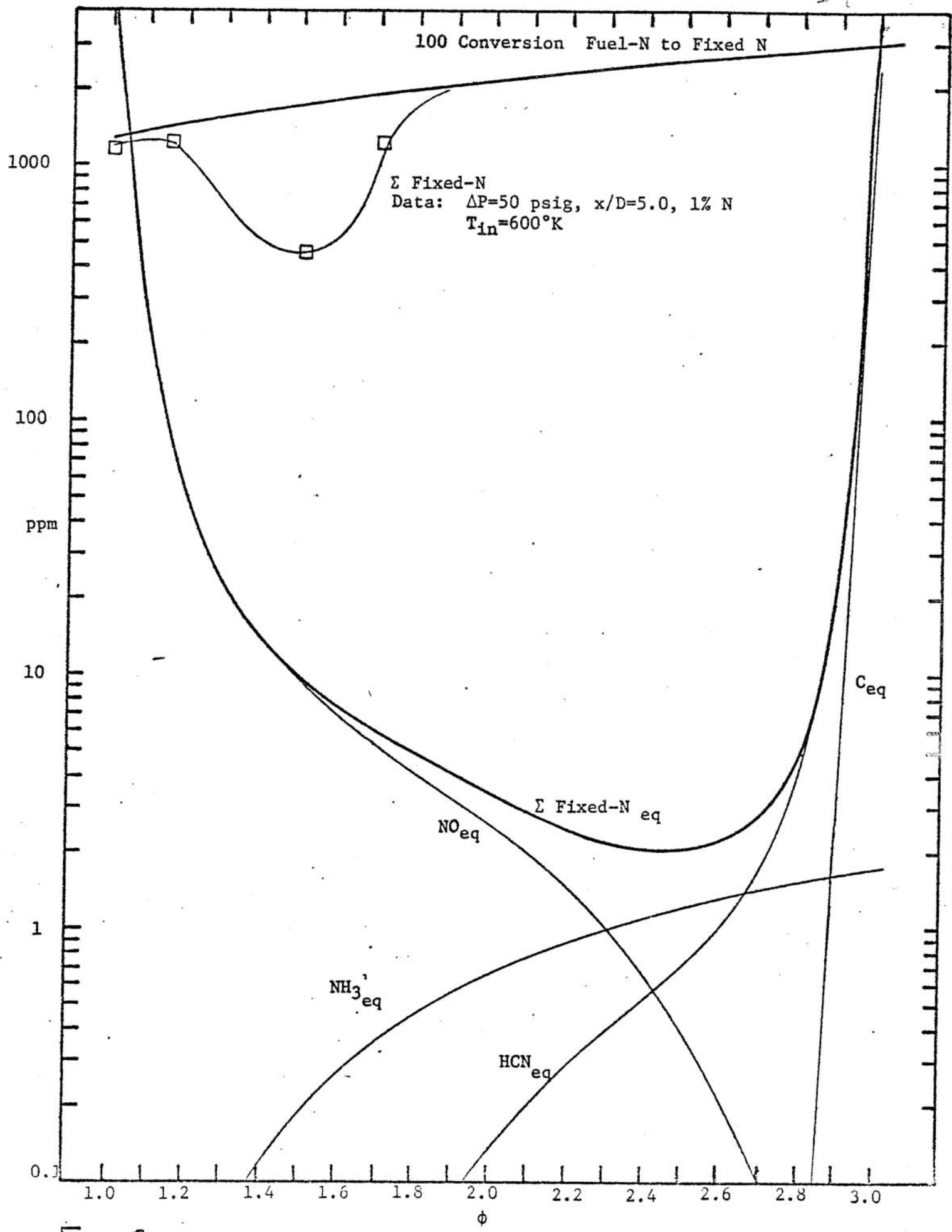Figure 7   Joint Plot, Laminar and Turbulent Diffusion Flame Data.
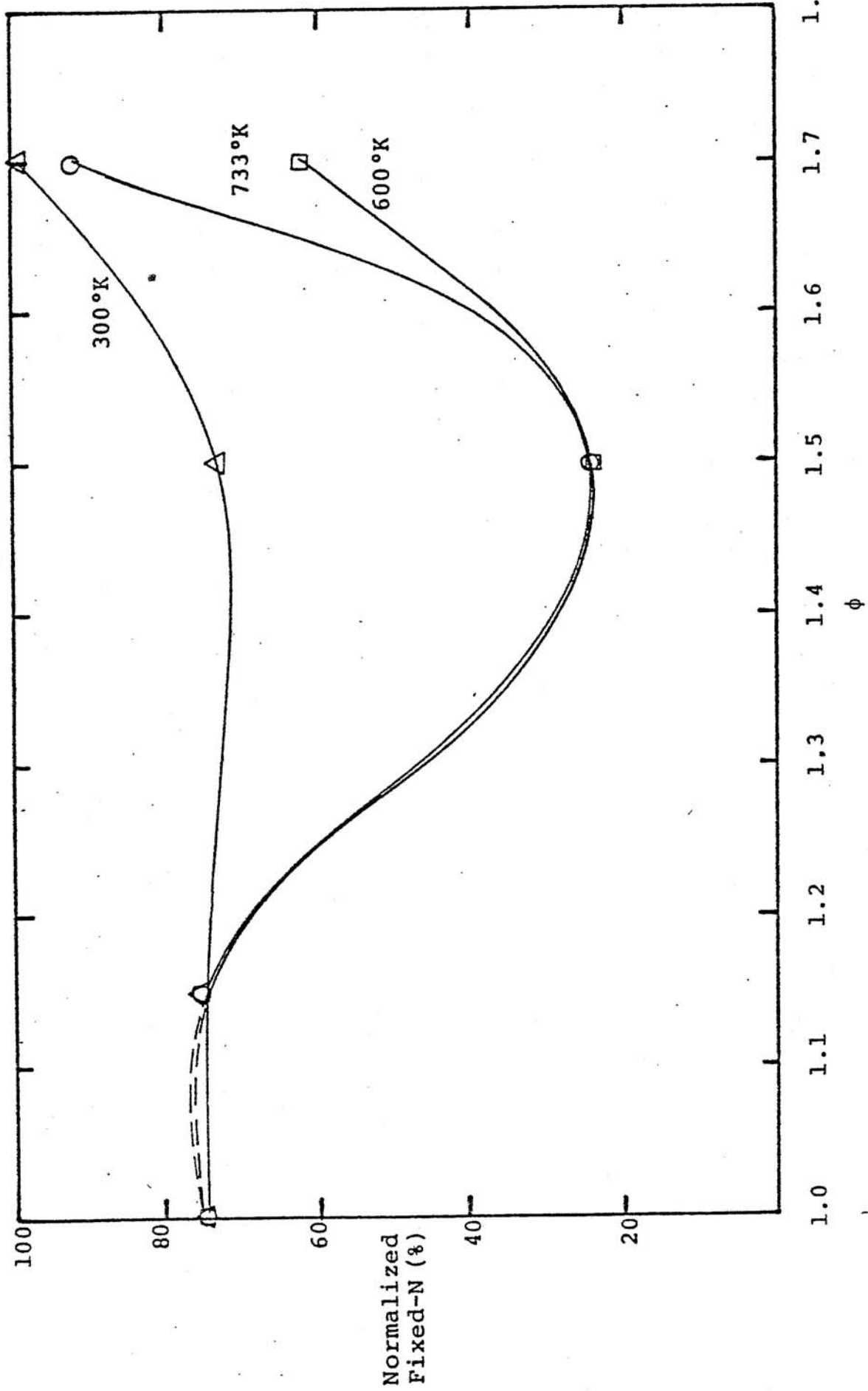
Figure 8

Effect of Equivalence Ratio on Fixed-N Species

Figure 9   Effect of Equivalence Ratio and Temperature on Fractional Yield of Fixed-N Species:

$\Delta P = 446$ kPa, X/D = 5.0

ϕ	NO (PPM)	HCN (PPM)	NH$_3$ (PPM)	TOTAL
1.3	1200	~ 20	~ 18	1238
1.5	30	2000	218	2248
1.7	50	2780	175	3005

Table 1   EFFECT OF EQUIVALENCE RATIO ON FIXED NITROGEN SPECIES DISTRIBUTION

KEROSENE DOPED WITH PYRIDINE TO 1% N BY WEIGHT; INLET AIR TEMPERATURE, 300°K; ATOMIZING PRESSURE, 446 KPA (50 PSIG); DISTANCE DOWNSTREAM, X/D = 5.0

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