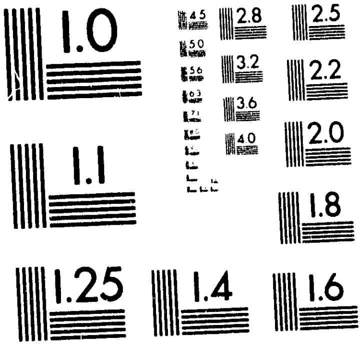
1.0
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1 of 1

P-7. Figure 1
Report Returned to Contractor
A-1

Glenn

Date: 23 February 1993

Contract Title: Industrial Pulverized Coal Low NO_x Burner: Phase 1

Contract Number: DE-AC22-~~9~~292PC~~92152~~ 92151

Contract Period: 1 April 1992 to 31 March 1993

Contractor Name: Arthur D. Little, Inc.
20 Acorn Park
Cambridge, MA 02140

1.0 Introduction

Objective. The objective of Phase I of the "Industrial Pulverized Coal Low NO_x Burner" Program is to develop a novel low NO_x, pulverized coal burner, which offers near-term commercialization potential, uses preheated combustion air of up to 1000°F, and which can be applied to high-temperature industrial heating furnaces, chemical process furnaces, fired heaters, and boilers. The program team is led by Arthur D. Little, Inc., and includes the Massachusetts Institute of Technology (MIT) and Hauck Manufacturing Company.

Concept Overview. In the low NO_x coal burner concept, the flue gas is recycled to the burner by jet pump action provided by the momentum of the primary air flow. The recycled flue gas is used to convey the pulverized coal to the jet pump where mixing with the primary air takes place. Ignition occurs downstream of the jet mixing section.

The recycled flue gas is at high temperature. When the pulverized coal is entrained, it is heated by conduction from the flue gas. The coal is pyrolyzed to a large extent before being mixed with the primary air. These pyrolysis products are the source of energy for the downstream flame.

In this process, the fuel nitrogen associated with pyrolysis products can be converted to molecular nitrogen in the pyrolysis flame if the oxygen is held to sub-stoichiometric concentrations based upon the burning species (pyrolysis products and some char). Pyrolysis products combustion is believed to be the primary source of NO_x emissions in coal combustors.

2.0 Progress Summary

First Quarter Summary. During the first quarter of the program (April 1992 to June 1992), the program team developed the overall program management plan; began a market survey to identify coals suitable for modeling the low NO_x burner design and performance, as well as for use in Phase II burner tests; and defined the preliminary burner design specifications, sized the prototype burner, and produced the first concept schematic.

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Second Quarter Summary. During the second quarter (July 1992 to September 1992), the program team completed the study of industrial coal usage and sources; refined the preliminary burner design and confirmed it as the basis for computer modeling; and started the definition of the modeling work scope.

Third Quarter Summary. This report is for the third quarter (October 1992 to December 1992). In the previous quarterly report it was reported that during this period, the program team would:

- Complete the lift-line design of the burner based on the pyrolysis of the model;
- Define the mixing and burning zones of the burner based on its kinetic model; and
- Establish NO_x predictions for the rich burning zone of the burner.

In fact, these tasks were deferred, due to delay in the development of the kinetic model to the fourth quarter. Instead work continued on advancing the fluid flow modeling at MIT and the chemical kinetics modeling at Arthur D. Little. The planned third quarter activities will be conducted in the fourth quarter.

3.0 Fluid Flow Modeling

The MIT modeling study focussed on Pittsburgh #8 30 μm-coal particles. Full text of the MIT Modeling Study is enclosed as Appendix A. The study suggests the following:

- The minimum desirable temperature for the pyrolysis of Pittsburgh #8 in the burner lift-line is 1,470°F (800°C) (Figure 6, Appendix A);
- The 1,470°F pyrolysis temperature can be achieved when the recirculating flue gas, entering the lift-line, is at a temperature of at least 2,010°F (1,100°C), and its mass flow rate is at least three times greater than the feed rate of the coal (Figure 3, Appendix A);
- The 30 μm coal particle entrainment period is predicted to be 2 ms (Figure 4, Appendix A);
- The coal particle will be heated to thermal equilibrium in 10 ms (Figure 5, Appendix A); and
- The 30 μm coal particle pyrolysis is nearly complete after 15 ms (Figure 6, Appendix A).

In addition, the MIT study has found that of the total fuel-bound Nitrogen, approximately 70% may remain in the char after pyrolysis. However, the conversion of this Nitrogen to NO_x can be greatly reduced, if both the pyrolysis products and the char are burnt in fuel-rich mode ($W_{air}/W_{coal} \approx 3.25$), and at high temperature ($\geq$ 2,730°F). Under these conditions, the percent-burnout of char is also significantly reduced (Figure 12, Appendix A). The fuel-rich reaction rate is estimated at 20 s^{-1} (vol-reactants/sec/vol-space).

4.0 Chemical Kinetics Modeling

During this period, Arthur D. Little started the chemical kinetics modeling of:

- Coal volatiles in the burner lift-line, and
- Mixture of volatiles, recirculated flue gas, and primary combustion air in the fuel-rich regions of the burner.

Arthur D. Little is using the latest version of CHEMKIN routines from Sandia National Laboratories in this analysis. The objective of the effort is to determine the degree of conversion of the fixed Nitrogen species resulting from coal devolatilization in the lift-line, to molecular Nitrogen, before entering the lean burnout zone in the furnace.

5.0 Burner Design

Burner design will be finalized when the MIT and Arthur D. Little modeling studies have been completed. The lift-line will be sized to support a predicted (15 ms) residence time for the coal particle and to provide a particle lift-velocity of 20 to 30 afps.

Air staging will be arranged to provide a primary air mass flow of approximately 3 1/4 x coal mass flow. Flue gas recirculation mass will be approximately 3 x coal mass.

Burner fuel-rich zone design will be compared with CHEMKIN recommendations when these become available. Burner fuel-lean zone design will be developed after the results of CHEMKIN and FLUENT modeling are completed during the next quarter.

6.0 Plans for the Next Reporting Period

The initial modeling results are helping to establish the optimum temperature parameters for the pyrolysis of coal in a flue-gas life-line (1,400°-1,500°F) and the fuel equivalence ratio parameters for the fuel-rich zone combustion of coal. It is uncertain, however, whether these conditions and the 1,000°F combustion air will create a sufficiently hot fuel-rich zone to ensure adequate conversion of fuel-N to N_2, instead of to NO_x.

Questions to be resolved during the fourth quarter, while completing the modeling studies, will include options such as:

- Increase air preheat?
- Draw flue-gas from the hottest area of the furnace?
- Reduce coal particle size to increase reactivity?
- Promote oxygen-coal reaction in the lift-line to raise the temperature of pyrolysis products.

One or a combination of the above options will raise the temperature of reactants, and thus, of the fuel-rich zone products. Thus, the fourth quarter modeling results will be the most critical for developing the burner design.

Appendix A

MIT Modeling Study
on
Industrial Pulverized Coal
Low-NO_x Burner

DRAFT

Industrial Pulverized Coal
Low-NO$_x$ Burner

Interim Report
January 1993

Modeling Study

Submitted by:

The Energy Laboratory and Department of Chemical Engineering
Massachusetts Institute of Technology
Cambridge, MA 02139

DRAFT

ADL/PETC Coal Combustion
Interim Report
Modeling Study

Introduction

The purpose of this modeling study is to evaluate the proposed concept of a Low-NO_x coal combustor shown in the sketch of Figure 1. In this configuration, flue gas is recycled to the burner by jet pump action provided by the momentum of the primary air flow. The recycled flue gas is used to convey the pulverized coal to the jet pump where mixing with the primary air takes place. Ignition takes place downstream of the jet mixing section.

The recycled flue gas is at high temperature and when the pulverized coal is entrained it is heated by conduction from the flue gas. The coal is pyrolyzed a large extent before being mixed with the primary air. These pyrolysis products are the source of energy for the downstream flame.

In this process, fuel nitrogen associated with pyrolysis products can be converted to molecular nitrogen in the pyrolysis flame if the oxygen is held to sub-stoichiometric concentrations based upon the burning species (pyrolysis products and some char). Pyrolysis products combustion is thought to be the primary source of NO_x emissions in coal combustors.

Coal Heating

The coal selected for this study is Pittsburgh #8, ground to 30 μm mean diameter. Pyrolysis of Pittsburgh #8 coal has been studied by various researchers[1-3]. The data of Suuberg et al., is presented in Figure 2 and is typical of data of other researchers. Two aspects of this data are of particular interest: pyrolysis becomes strong at around 800°C, the dominant pyrolysis products are tars and liquids. The data of Suuberg et al.,[1] also provides the information that pyrolysis of this coal is energetically neutral.

With the minimum desired temperature of 800°C in mind, we calculate the pyrolysis (mixed) temperature for 100°C coal and various temperatures of flue gas as a function of flue gas to coal mass flow rate in the lift line. The results are shown in Figure 3. From these results we see that a flue gas temperature of 1100°C is desirable and a flue gas to coal mass flow rates of at least 3 is required at this temperature.

The time scale for entrainment of the particles is found from the momentum equation:

$$m_p \frac{du_p}{dt} = \frac{C_D}{2} \rho(u-u_p)|u-u_p| \pi r_p^2 \tag{1}$$

where the drag coefficient,

$$C_D = 24/R_e + 4/R_e^{1/2} + 0.4 \tag{2}$$

The Reynolds Number

$$Re = 2\rho \frac{|u-u_p| r_p}{\mu} < 10, \tag{3}$$

so that

$$\frac{du_p}{dt} = \frac{9\mu}{r_p^2 \rho_p} (u-u_p) \tag{4}$$

where $\rho_p \simeq 1.4$ g.cm^{-3} is the coal particle density and $\mu \simeq 0.3 \times 10^{-3}$ g.cm^{-1} s^{-1} is the viscosity at intermediate temperature. The solution to the equation for $r_p = 15$ μm,

$$\frac{du_p}{dt} = 860(u-u_p) \tag{5}$$

is readily found when u, the gas velocity is held constant. This result is presented in Figure 4 as the decay of the velocity difference between the particles and gas as a function of gas flow time. The 95% equilibration time is seen to be 2 ms for the 30 μm diameter mean particles size and scales as the square of the particle size. This is then not a limiting process as the gas velocity is only 20 f/s so that the distance is small. Further we note that the drag of equation 5 is many times gravitational force.

The temperature equilibration is found from conservation of energy as

$$m_p\, Cp_p \frac{dT_p}{dt} = \frac{\mu 2\pi r_p}{0.7} \left(2+\sqrt{R_e}\right) Cp\, (T-T_p) \tag{6}$$

with the coal specific heat, $Cp_p = 1.6\ J.g^{-1}{}^\circ C^{-1}$

and the gas specific heat, $Cp = 1.2\ J.g^{-1}{}^\circ C^{-1}$

The Reynolds number is low because of the small velocity difference so that we can write for $r_p = 15\ \mu m$,

$$\frac{dT_p}{dt} = 300\ (T-T_p) \tag{7}$$

This equation is readily solved and the decay of particle to gas temperature difference is shown in Figure 5.

The solution assumes that the gas temperature is not affected and that the particle size does not change. These are both approximations as the particle mass loading is expected to be fairly high and the swelling could double the particle size. The swelling actually increases the heating rate as the density drops as the cube of the radius while at constant density the heating shows as the inverse square of the radius. The change is gas temperature primarily effects the final temperature and not the time scale. The thermal equilibration time is seen to be 10 ms for the mean particle size so that gas heating of the particles is not deemed an issue.

Coal Pyrolysis

Coal particle pyrolysis time is assessed by use of a proprietary computer code obtained from Advanced Fuel Research[4]. Calculations were performed using the temperature profile of

Figure 5 with an initial coal temperature of 100°C and a final coal temperature of 800°C. The results are shown in Figure 6. The coal is Pittsburgh #8 with 1.4% moisture and 11.3% mineral matter. The results of Figure 6 are on a dry-ash-free basis and show the mass fraction of the coal evolved as gas and tar. We see that pyrolysis is nearly complete after 15 ms, and that about 45% total mass loss is formed and that three quarters of the mass loss is in tar. This is in agreement with experimental data[1-3].

Of additional interest is the evolution of the elemental composition of the char, tar and gas. These are shown in Figures 7, 8 and 9 respectively and indicate that in particular, the nitrogen concentration is enhanced in the char, the nitrogen concentration in the tar is about equal to that in the original coal. The nitrogen concentration in the gas phase is significantly depressed.

There is general agreement that coal nitrogen is bound in two forms[5-7], with the large tar source structures and with the char structure. These nitrogen atoms are released at two very different rates; the former with the tar, the latter much more slowly (see Figure 10)[5]. The amount of nitrogen left behind in the char has been measured and is given for Pittsburgh #8 in the right hand side of Figure 11[8]. This indicates as predicted that about 70% of the nitrogen will remain in the char. The nitrogen distribution in the gas/tar will change as the tar molecules "crack" with time[9].

Bench Scale Studies

Bench scale studies have been performed at MIT in the past which are quite relevant to the current study. These studies included entrained flow reactor testing and were performed by Pohl and Sarofim[8] and by Song et al.[10].

Figure 12 shows results from Pohl and Sarofim[8] including Pittsburgh #8 coal reacting in a flow of oxygen in helium at various fuel equivalence ratio's. Residence times vary from

200-1000 ms. We notice from the data that, although char burnout is significantly reduced at fuel equivalence ratio's greater than unity, the conversion of fuel nitrogen is dramatically reduced, particularly at a fuel equivalence ratio greater than two.

When the coal is pyrolyzed separately and the resultant char then oxidized the resultant behavior is shown in Figure 13. Here we see that at a char fuel equivalence ratio slightly greater than unity, conversion to NO_x is reduced.

The speculation then is that to minimize NO_x formation from fuel nitrogen that pyrolysis products should be burned fuel rich based upon their stoichiometry alone ($\leq$ 40% of coal) and that the resultant char also be burned fuel rich.

If the reaction can be made to occur at a high temperature, the combustion of the pyrolysis products will occur while some char combustion takes place, so that the air staging can be reduced and still maintain a fuel rich environment for the pyrolysis products combustion. This behavior can be seen to a limited extent in the data of Figure 14 and 15, taken from the work of Song et al.[10.] The gas phase distribution from the combustion of Figure 14 is given in Table 1 for the very fuel rich conditions. These higher temperature data are inconclusive, however, since the coal utilized is a lignite with an oxygen content in excess of 15 wt%.

Oxidation/Ignition

For the purpose of these studies we have assumed that we wish to perform combustion of the pyrolysis products in a fuel rich environment. We assume there will be some char reaction so that if we design for stoichiometric combustion of the pyrolysis products, that the reaction will be fuel rich. Excess reaction of the char will be a bonus. In this way we determine the minimum for the primary air flow and the jet pump requirements.

We include in the stoichiometric calculation, the oxygen in the flue gas which may vary from 0 to 5%. The primary air flow is then given in Figure 16 as a function of the flue gas flow. From this we expect values of air to coal flow of 3 at flue gas to coal of 3.25.

We have performed a mixture (including char) temperature calculation to determine the pre combustion and post combustion conditions assuming no heat loss. The results for no dissociation are shown in Figure 17. The appropriate premixed temperature is 625° and the expected reacted temperature 2200°C. The adiabatic temperature with dissociation is given in Figure 18 as a function of air to coal ratio and yields 2100°C. If we assume 40% loss of chemical energy this yields 1500°C flame temperature.

Data correlated by Lewis and Simons[11-13] would predict an intrinsic reaction rate of $20 s^{-1}$ at 20% O_2 for a similar char (Figure 19). Pyrolysis combustion times measured by Shaw et al.[14] give combustion times of microseconds as would be predicted for large hydrocarbons[15]. Diffusion times are very much like the heating times of 10 ms discussed above, but the pyrolysis products should be well dispersed in the transport process. Mixing of the primary air in the jet is probably limiting. We may, thus, expect a premixed flame behavior with very little reaction of the char in the flame itself.

Future Work

Future work will include an evaluation of the flowfield in the jet mixing region and possible flame holder geometries. *Chemkin* calculations may still be performed but may lead to misleading conclusions because of the absence of char reactions. Flow field modeling with *Fluent* seems more appropriate to evaluate the "flame holder" concepts and the jet mixing of both primary and secondary air.

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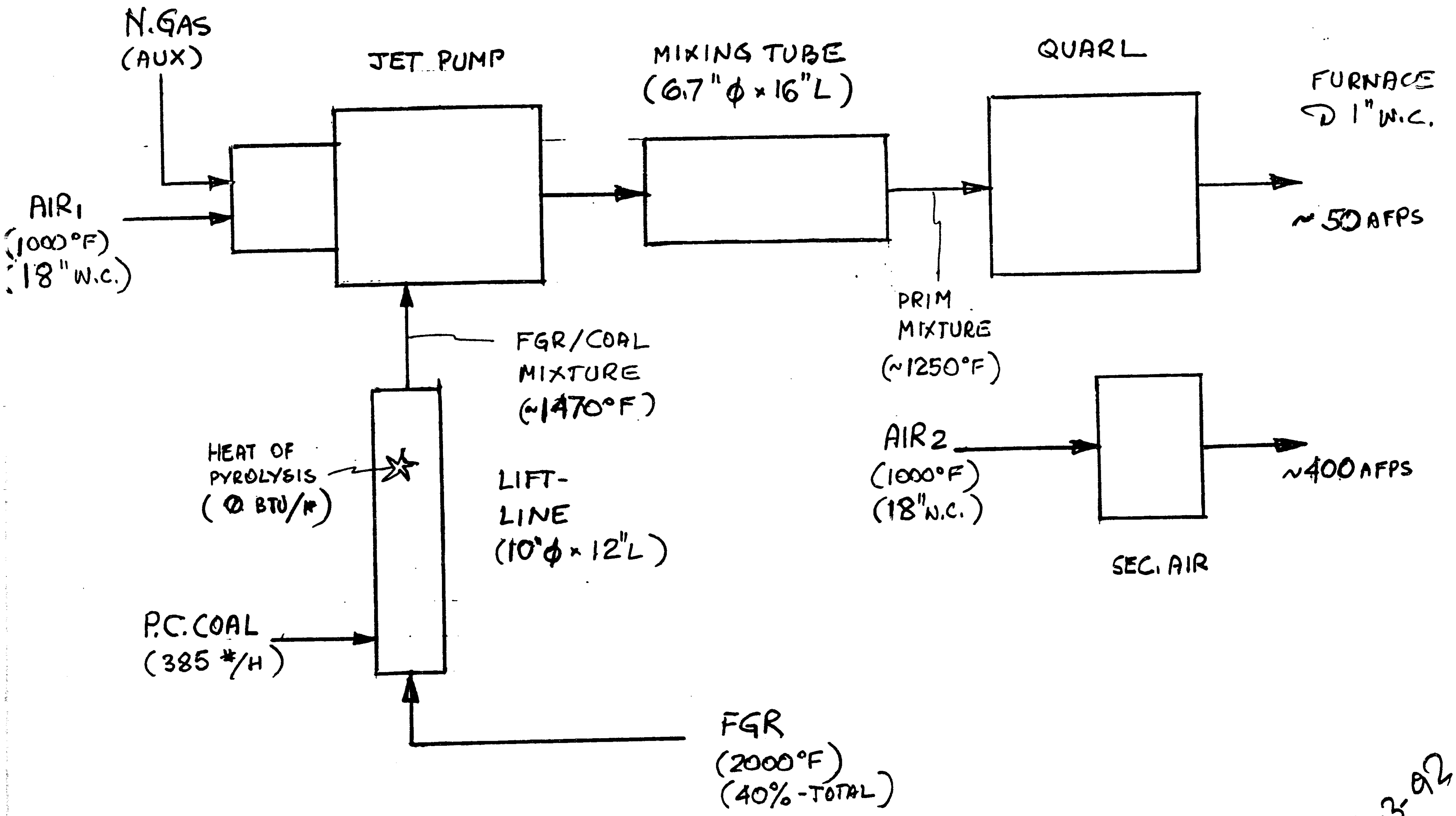

LOW NO_x P.C. BURNER — PROCESS SCHEMATIC

FIGURE 1.

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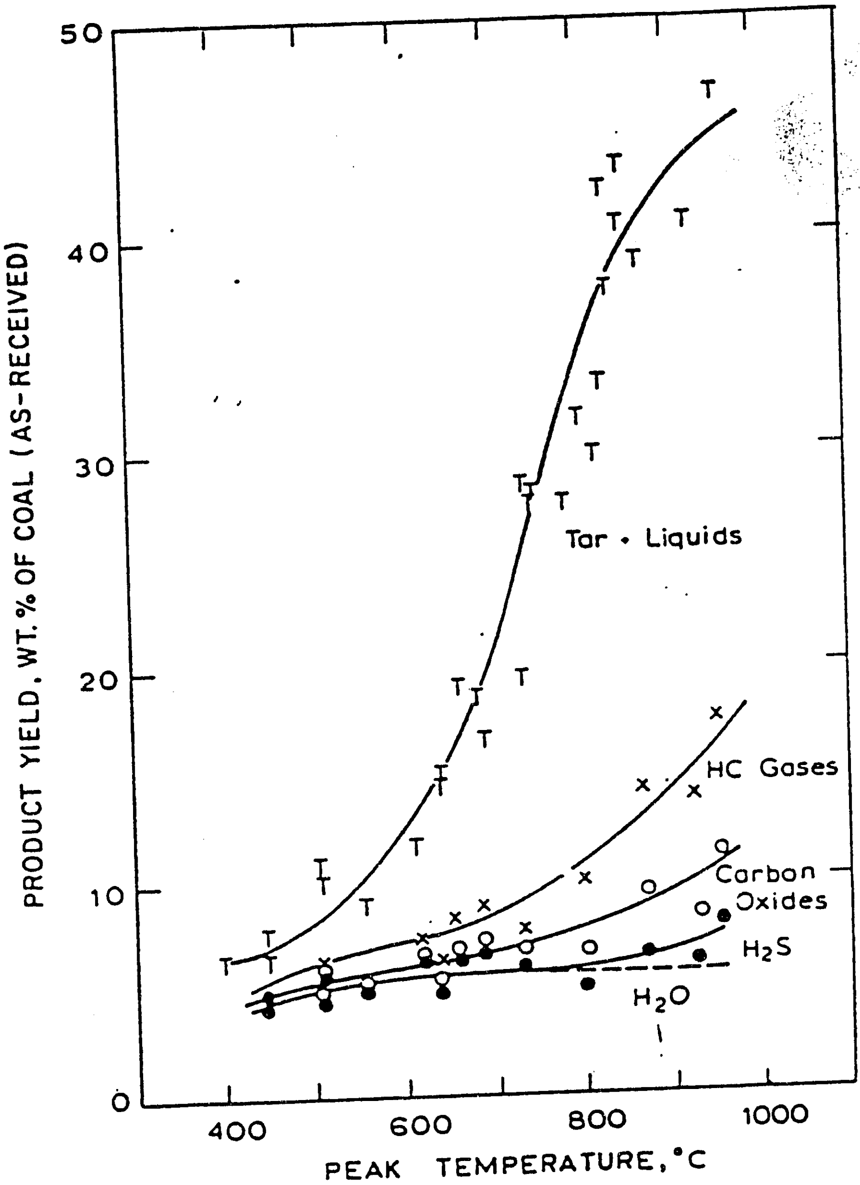

Figure 2.[1] Pyrolysis product distributions from bituminous coal heated to different peak temperatures [(●) H_2O and H_2S; (O) H_2O, H_2S, CO, and CO_2; (x) H_2O, H_2S, CO, CO_2 and all hydrocarbon gases; (T) total weight loss, i.e., H_2O, H_2S, CO, CO_2, all HC gases, tar and liquids. Pressure = 1 atm (helium). Heating rate = 1000°C/s].

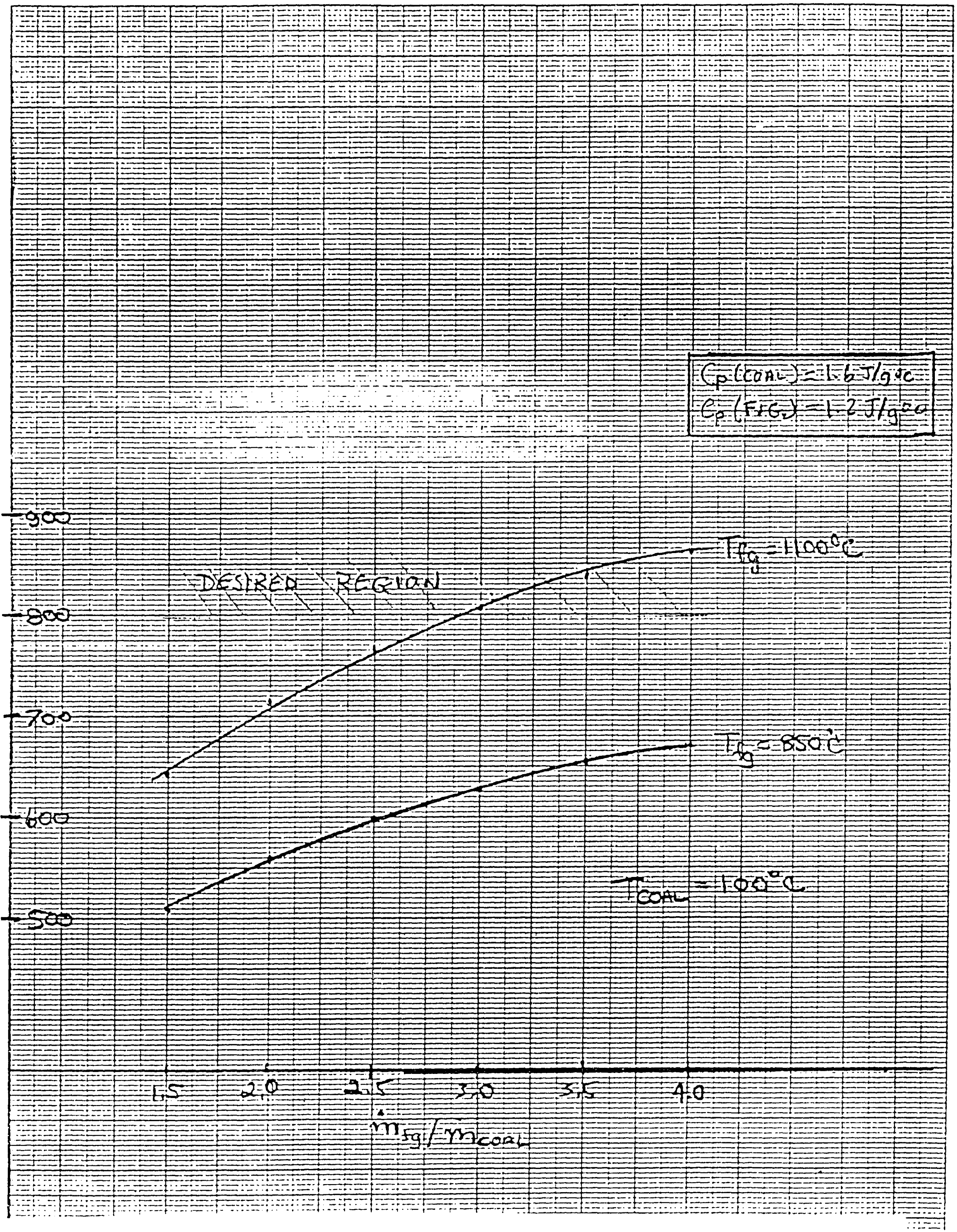

Figure 3. Coal Pyrolysis Temperature from Mixing Coal and Flue Gas

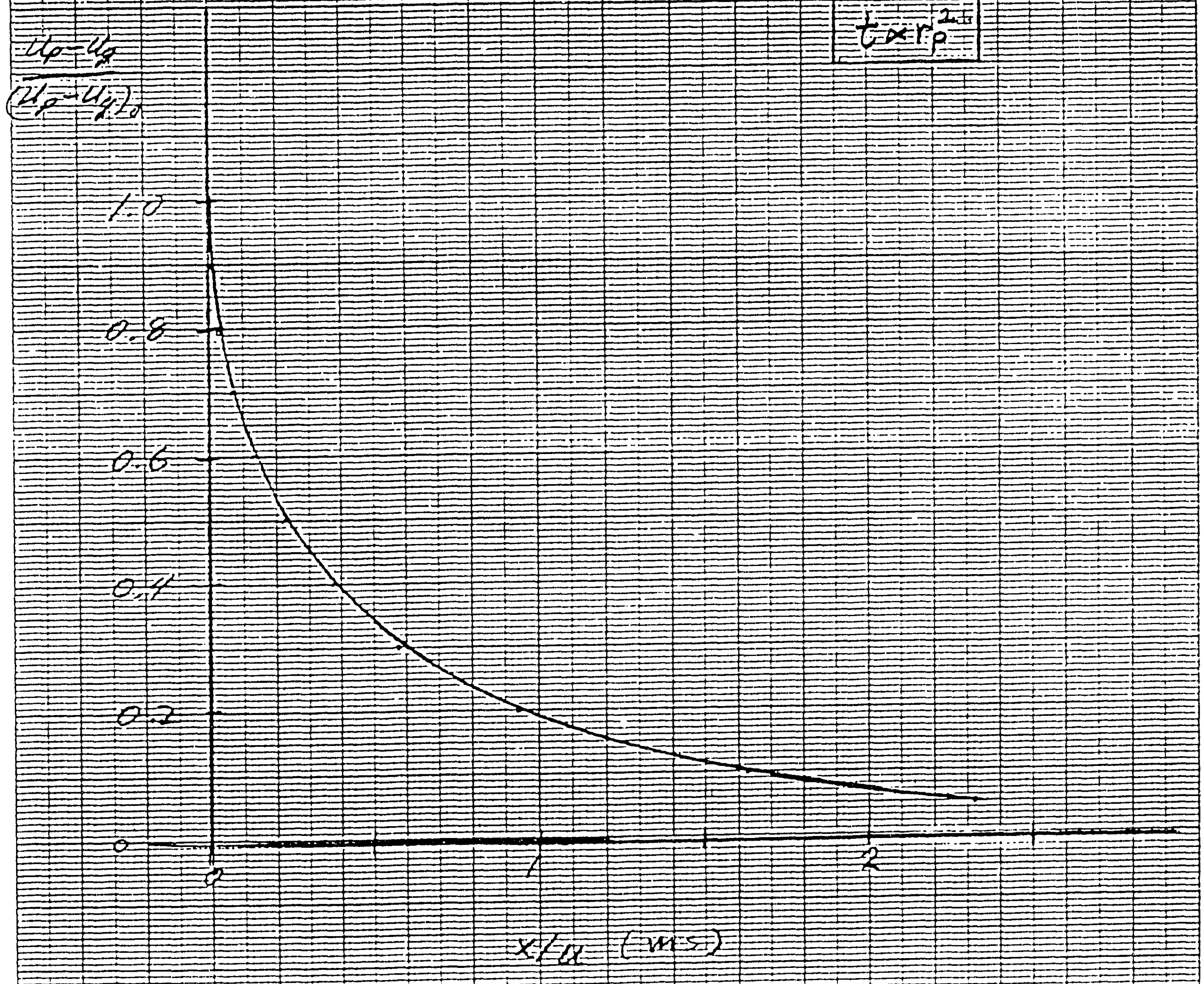

Figure 4. 30 μ Particle Entrainment into Gas Stream

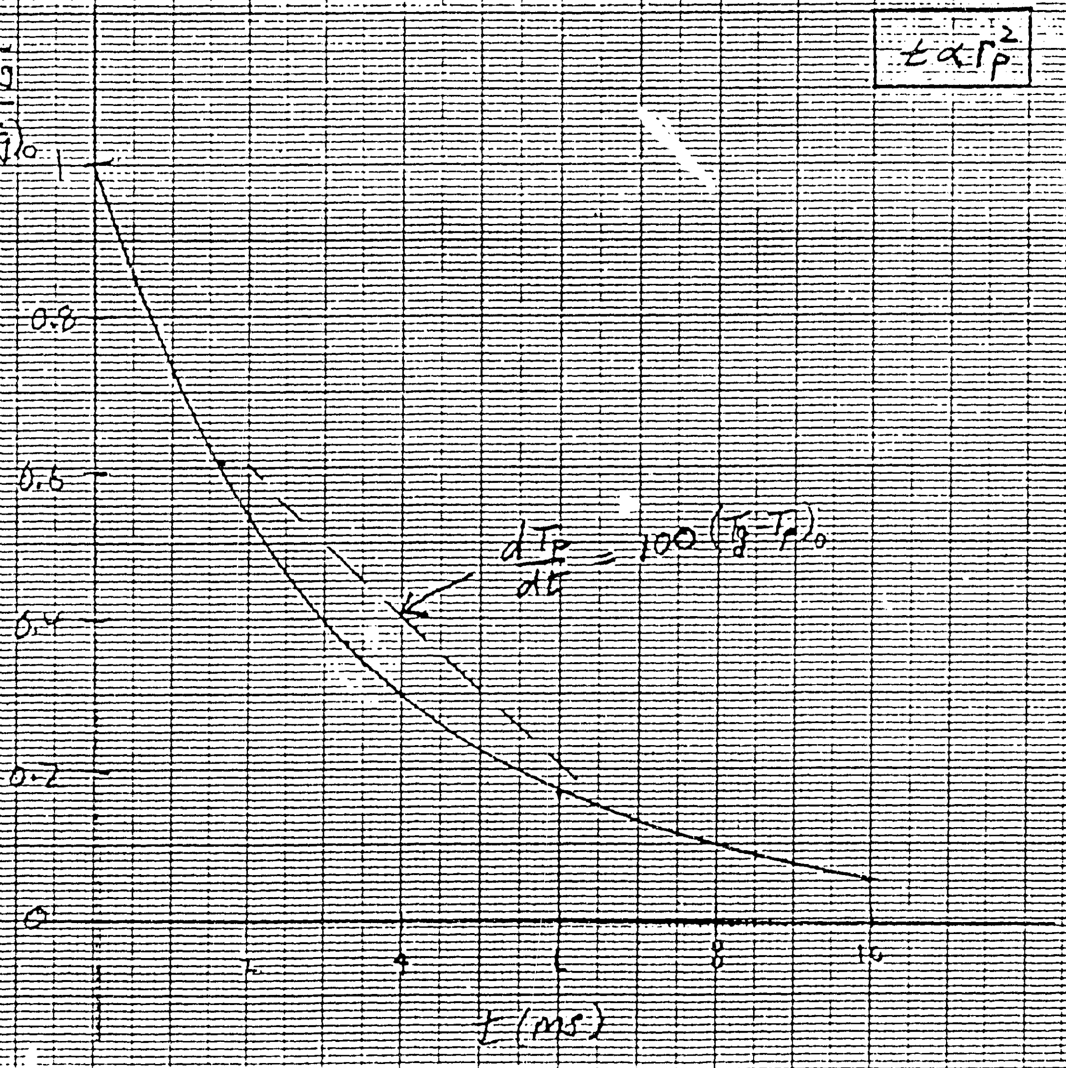

Figure 5. Temperature Equilibration of 30 μm Diameter Coal Particle in Heated Gas

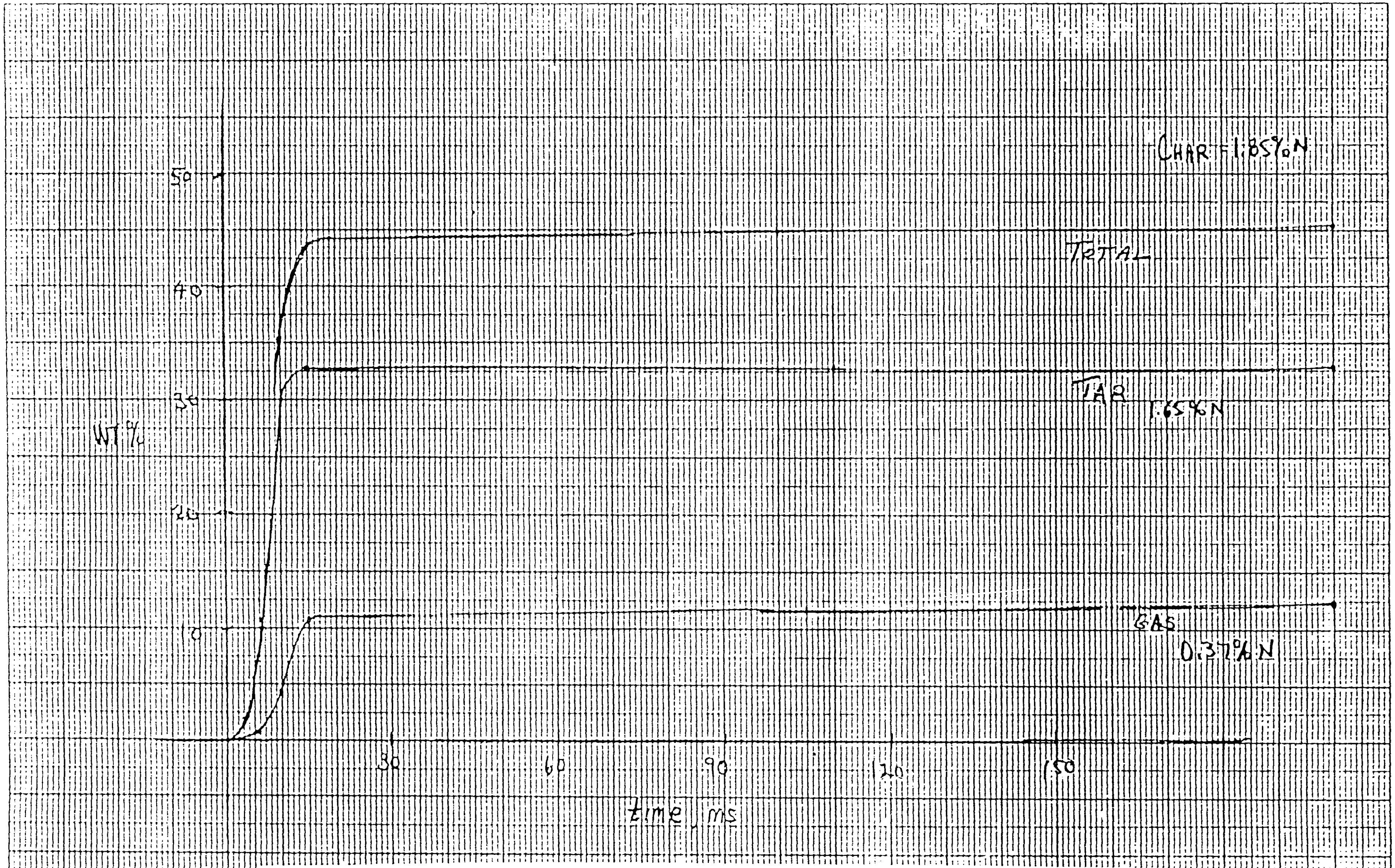

Figure 6. AFR Pyrolysis Model Results
Pyrolysis: 800°C Final Temperature Coal = 1.6% N (DAF)

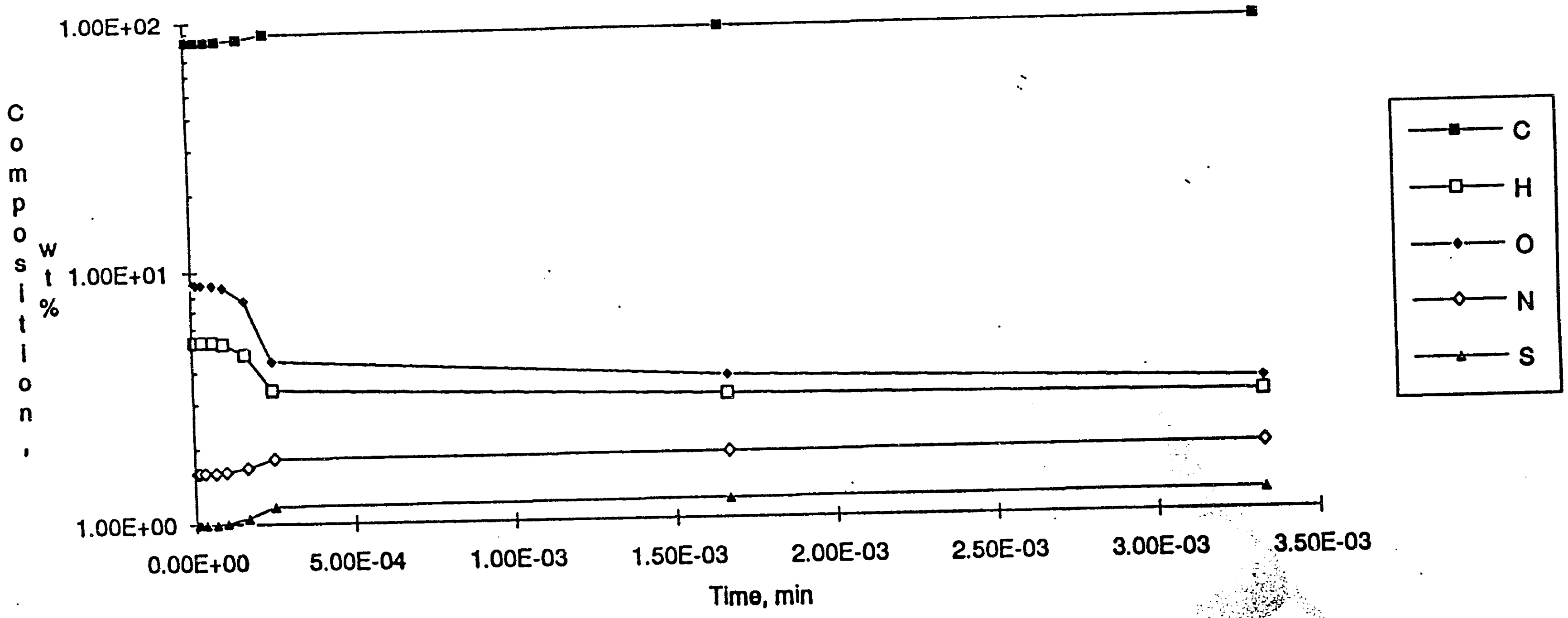

Figure 7. Char Composition

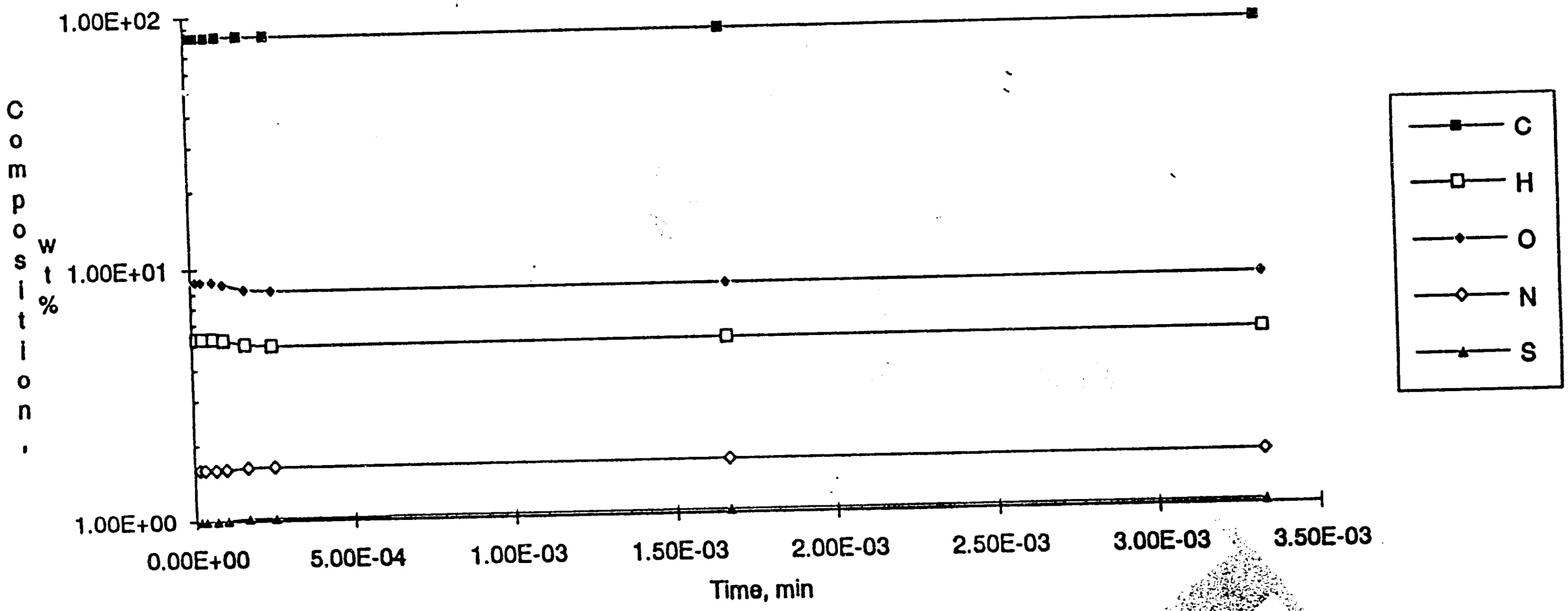

Figure 8. Tar Composition

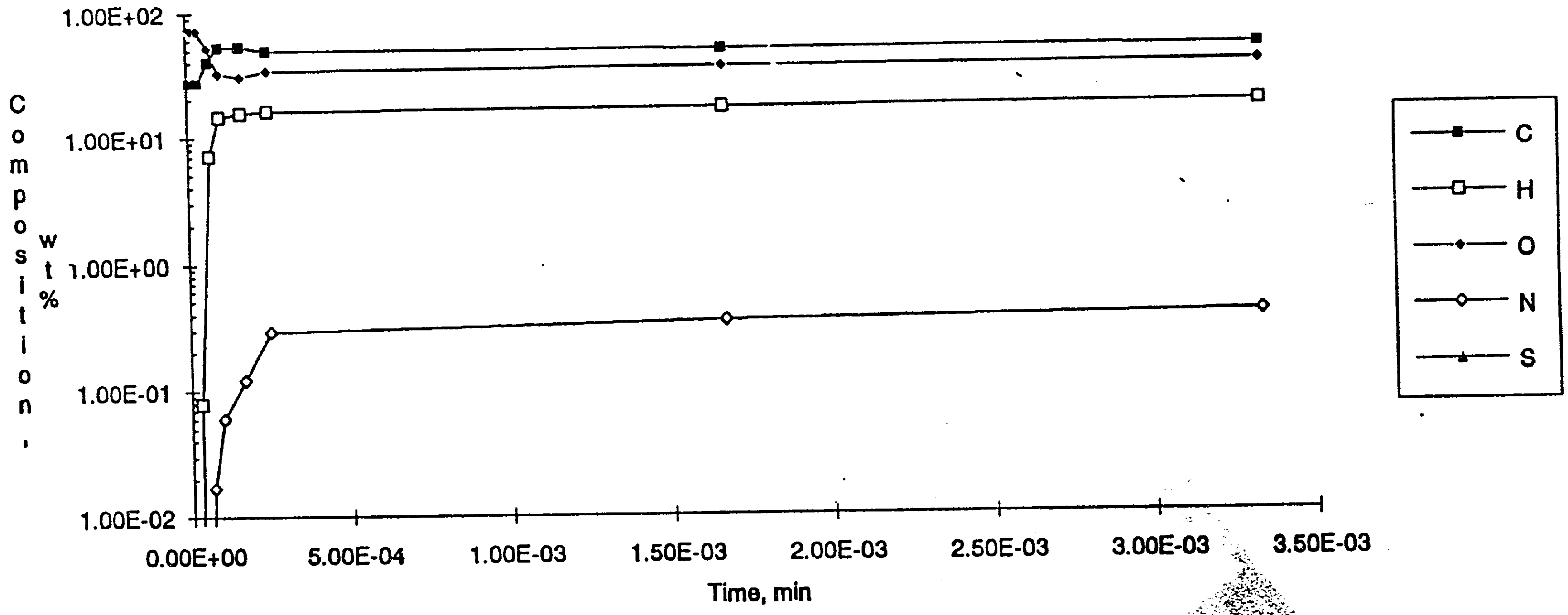

Figure 9. Gas Composition

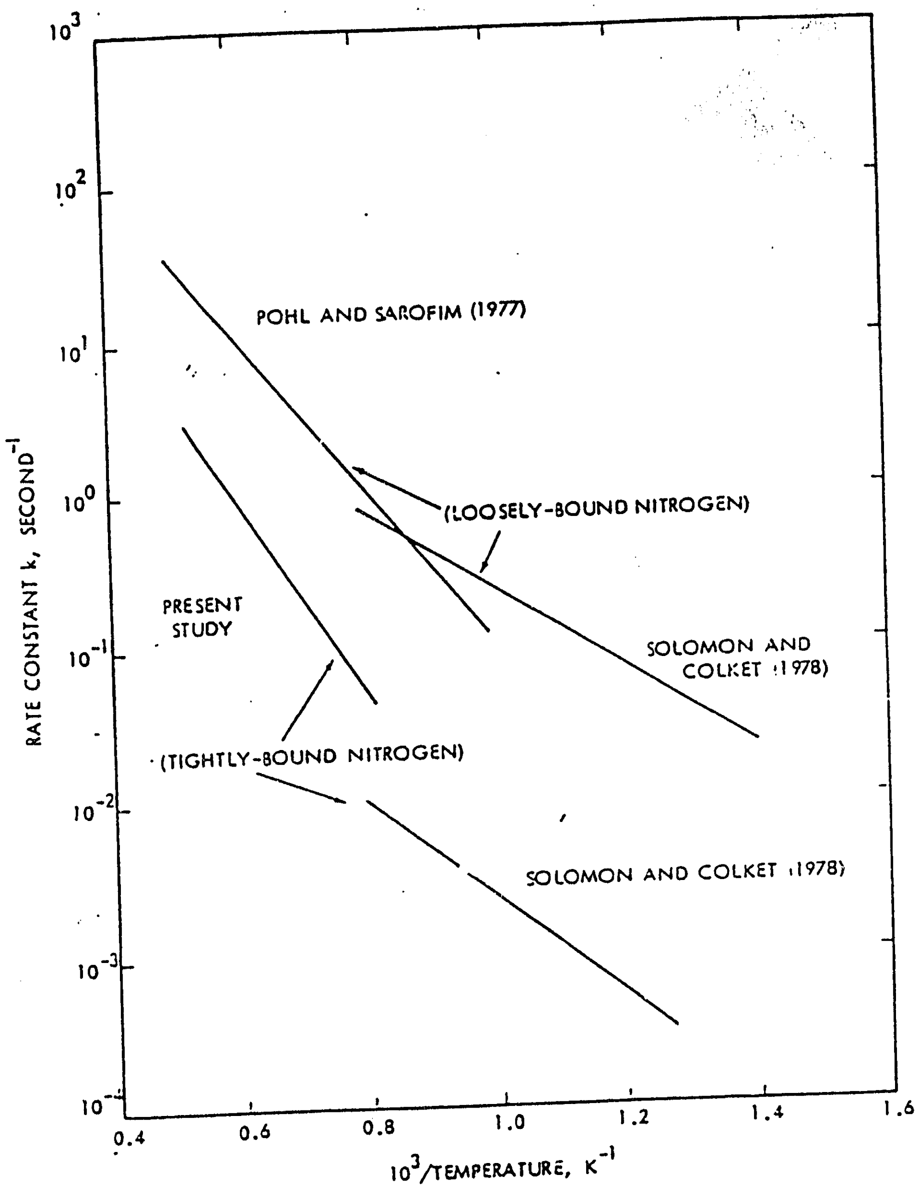

Figure 10.5 Comparison of First-order Devolatilization Rate Constants of Fuel Bound Nitrogen.

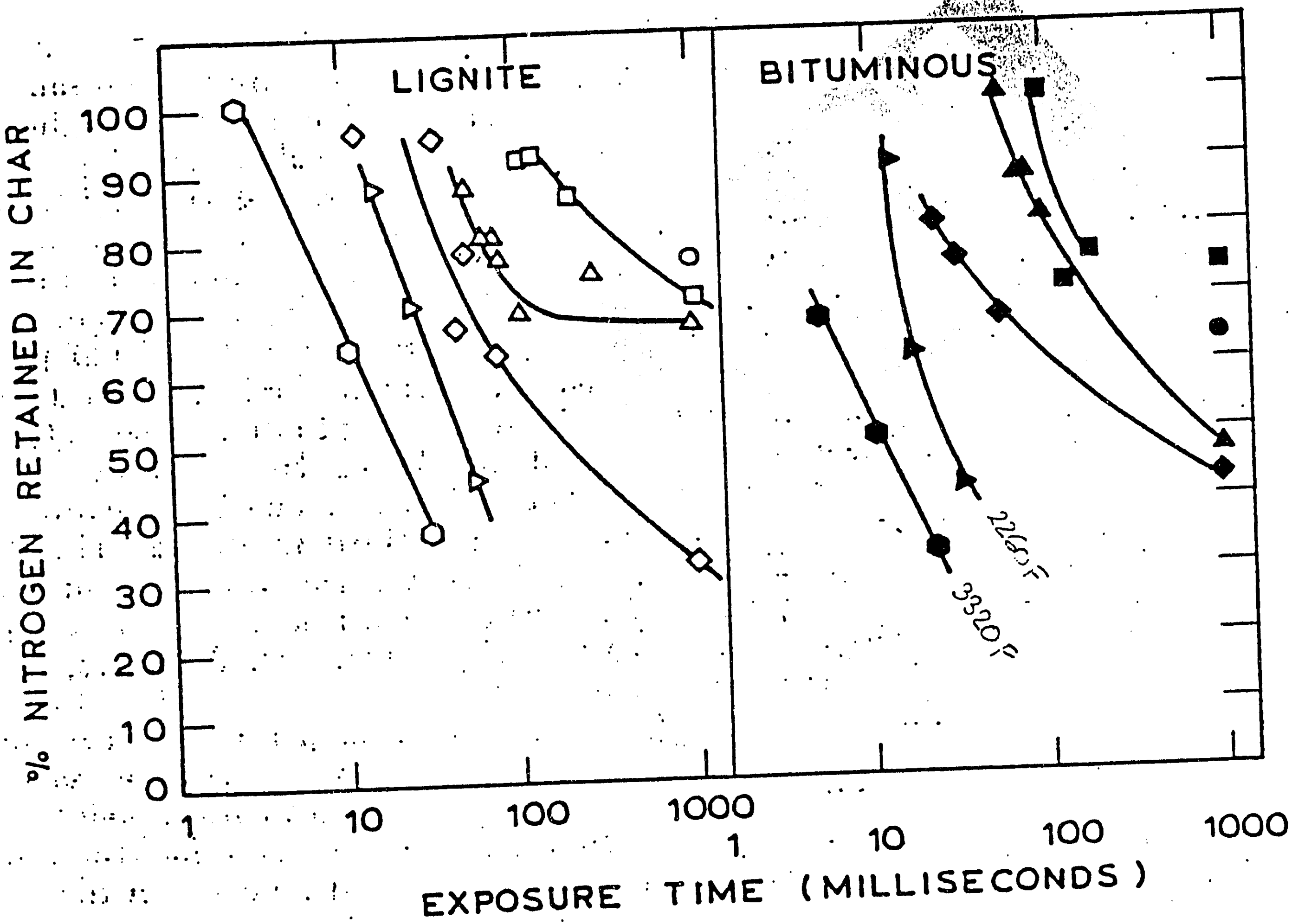

Figure 11.[8] Retention of Nitrogen in Char Versus Time Furnace Temperatures: O-1000°K, □-1260°K,△-1510°K◇-1740°K, ▷-1960°K, ○-2100°K

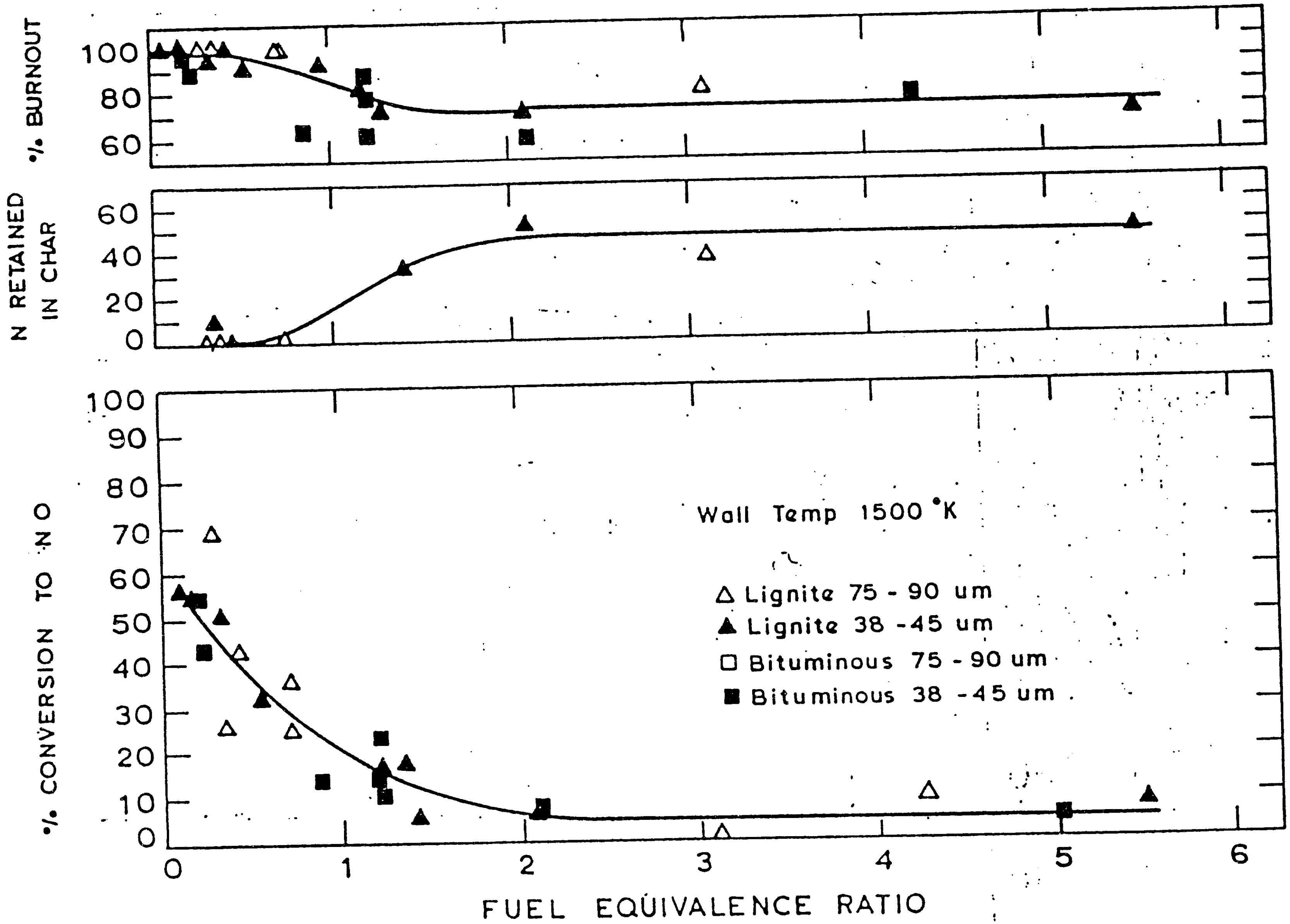

Figure 12.[8] Conversion of Coal Nitrogen to Nitric Oxide, Percent Carbon Burnout, and Percent of Coal Nitrogen

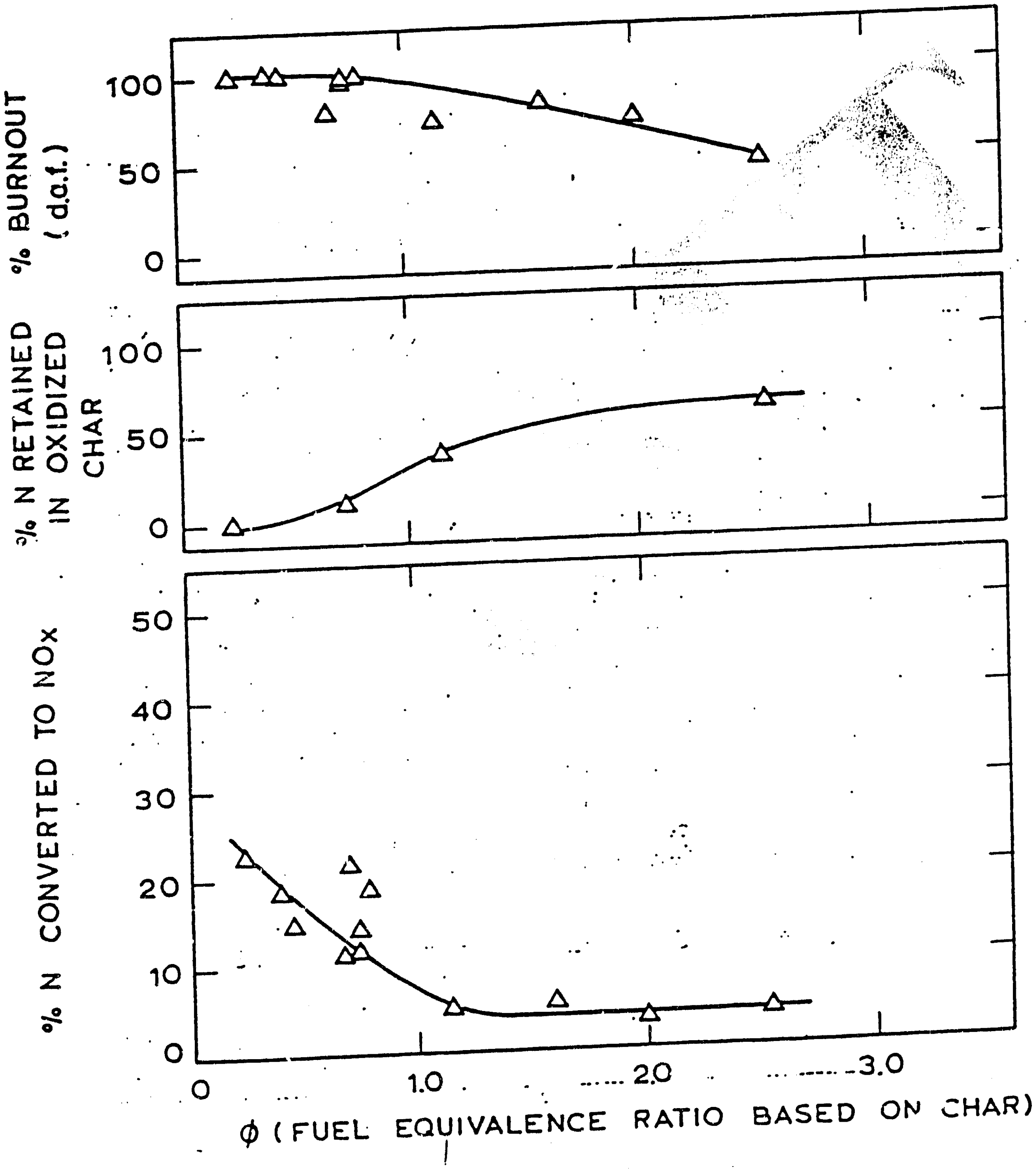

Figure 13.[8] Conversion of char nitrogen to nitric oxide, percent char burnout, and percent of char nitrogen retained in Unburned char. Char was produced at 1500°K.

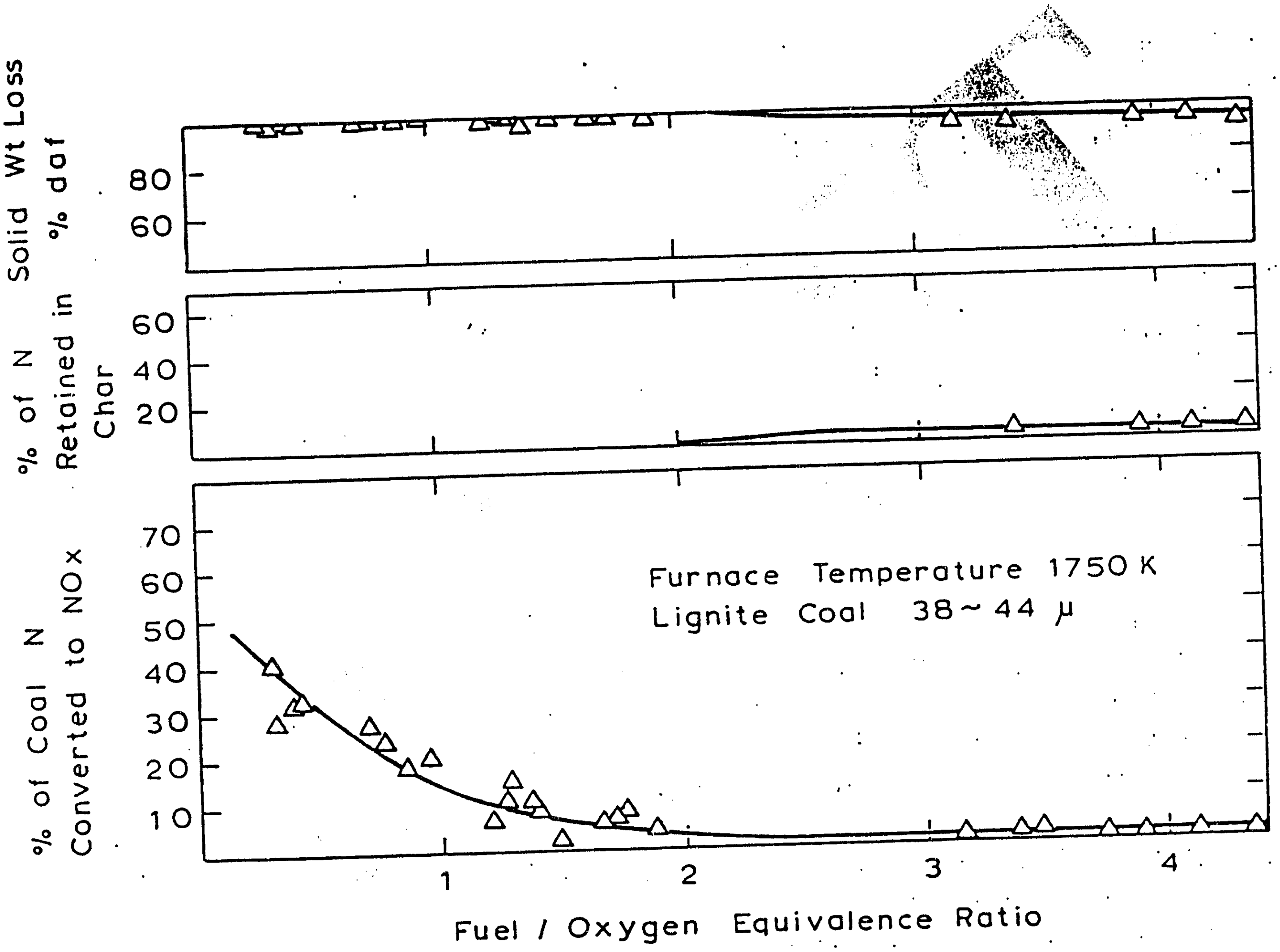

Figure 14.[10] Fate of coal-nitrogen during oxidation: conversion to nitric oxide (bottom); retention by unburned char (middle); and combustion efficiency (top). Montana lignite at 1750 K.

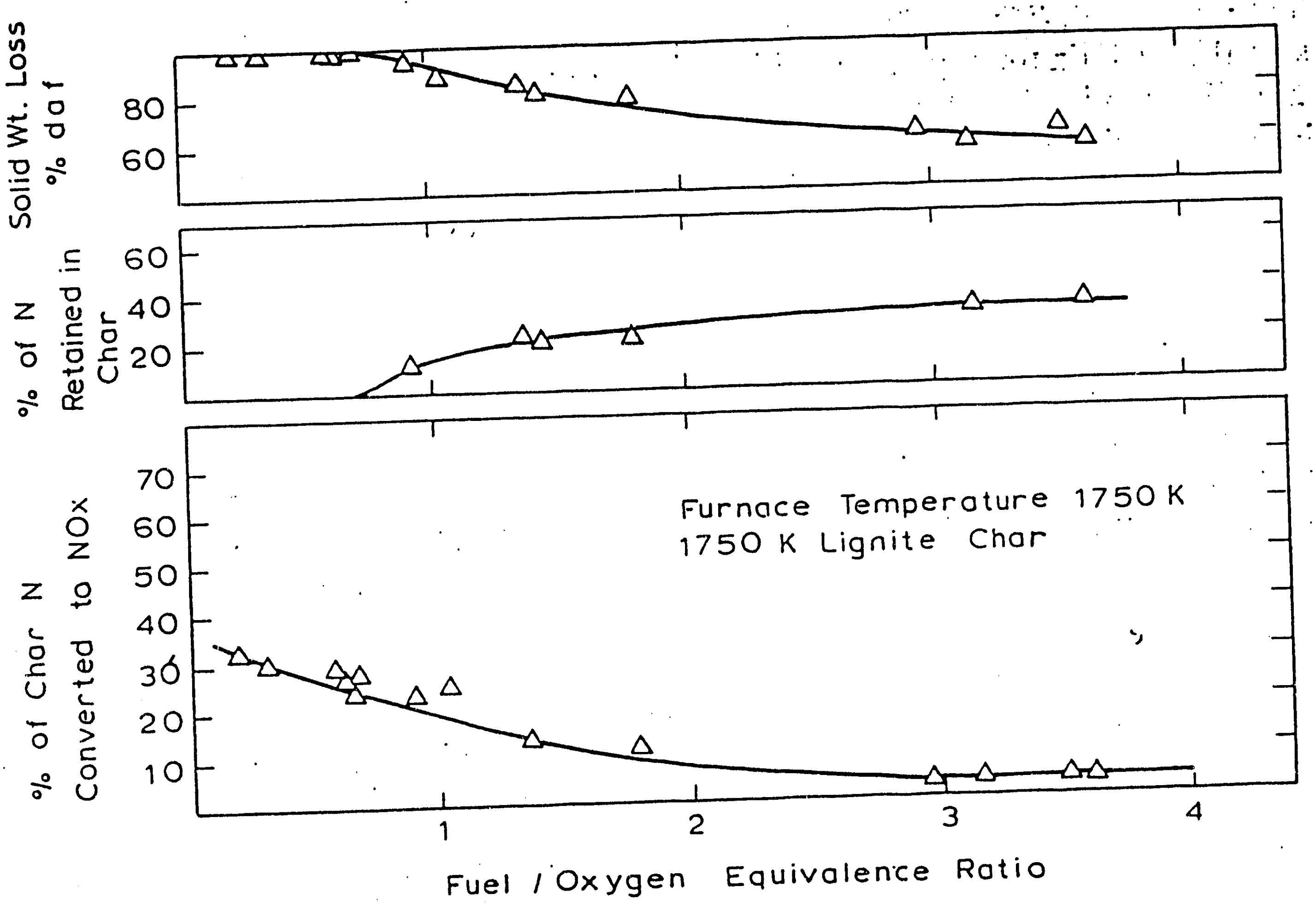

Figure 15[10] Fate of char-nitrogen during oxidation: conversion to nitric oxide (bottom); retention by unburned char (middle); and combustion efficiency (top). Char from Montana lignite pyrolyzed and then oxidated at 1750 K,

	Wt % of the original nitrogen in lignite
A. Nitrogen distribution	
N_2	71~75
HCN	6~ 8
NH_3	Trace
NO_x	1~ 3
N in char	3~ 5
Total	81~91

	Wt % of the original carbon in lignite
B. Carbon distribution	
CO	72~75
CO_2	11~16
CH_4	1~ 2
Other CH	Not measured
TAR	Not measured
C in char	4~ 5
Total	88~98

Table I[10] Nitrogen and Carbon distribution during combustion of lignite at $T = 1750$ K and $\phi = 3{\sim}4$

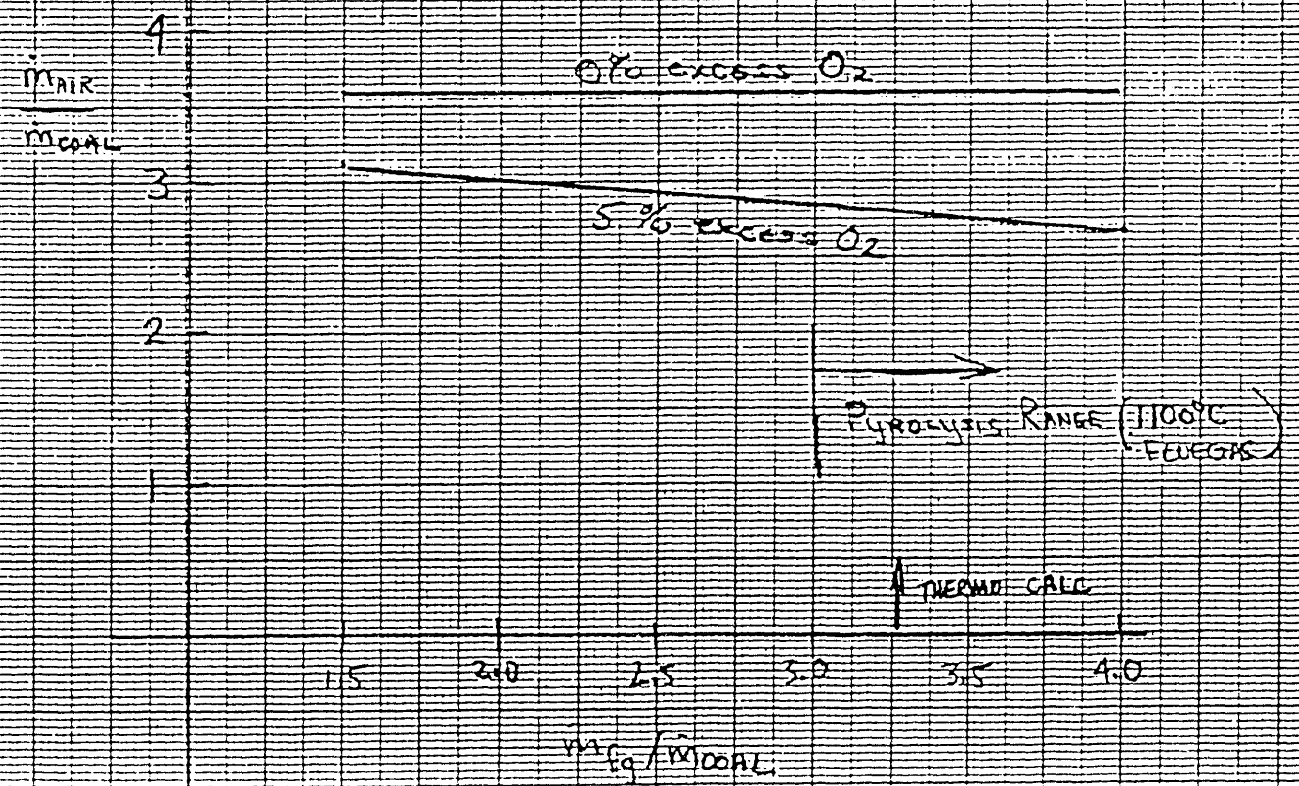

Figure 16. Air Requirements for Stoichiometric Combustion of Pyrolysis Products

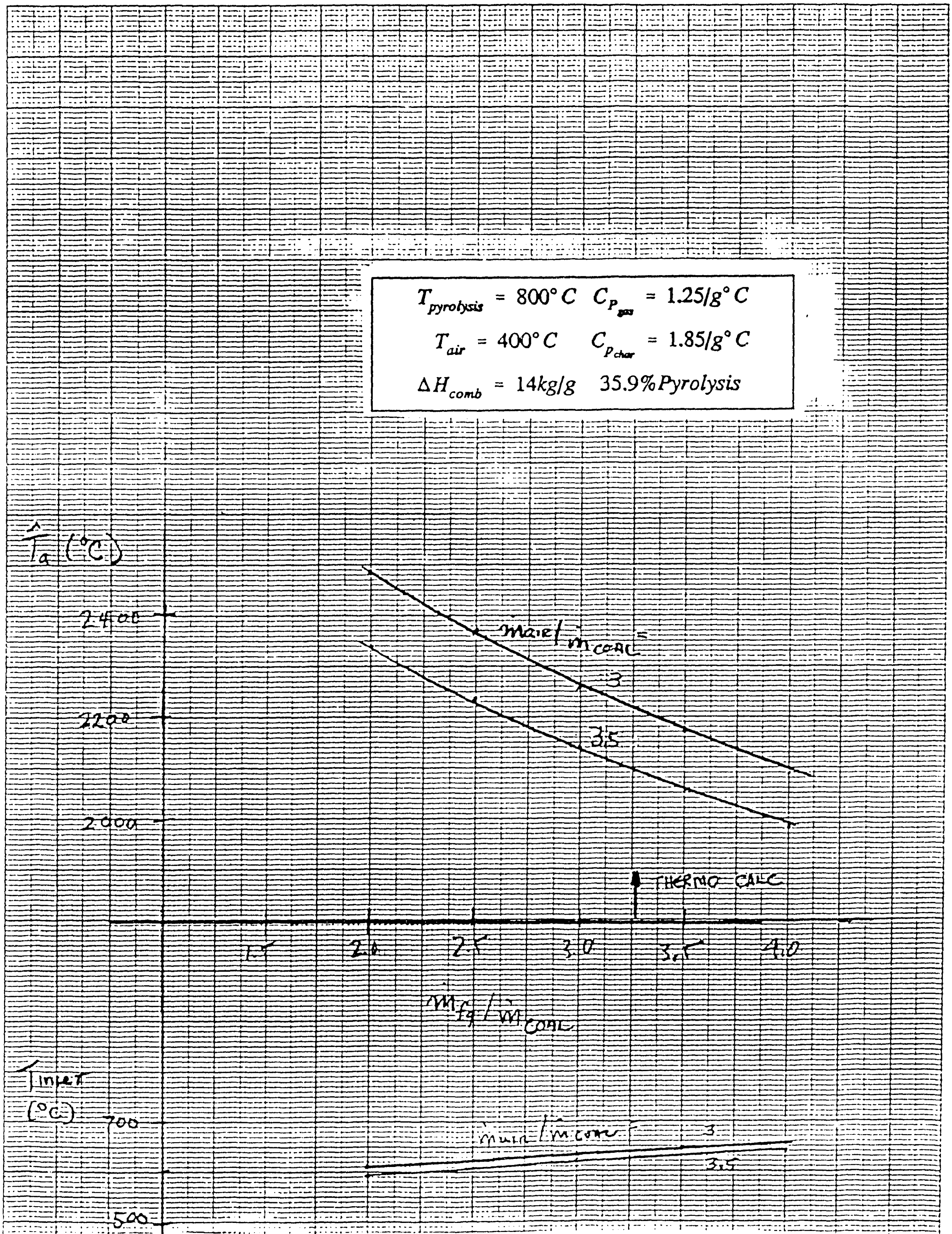

Figure 17.   Maximum Adiabatic Flame Temperature

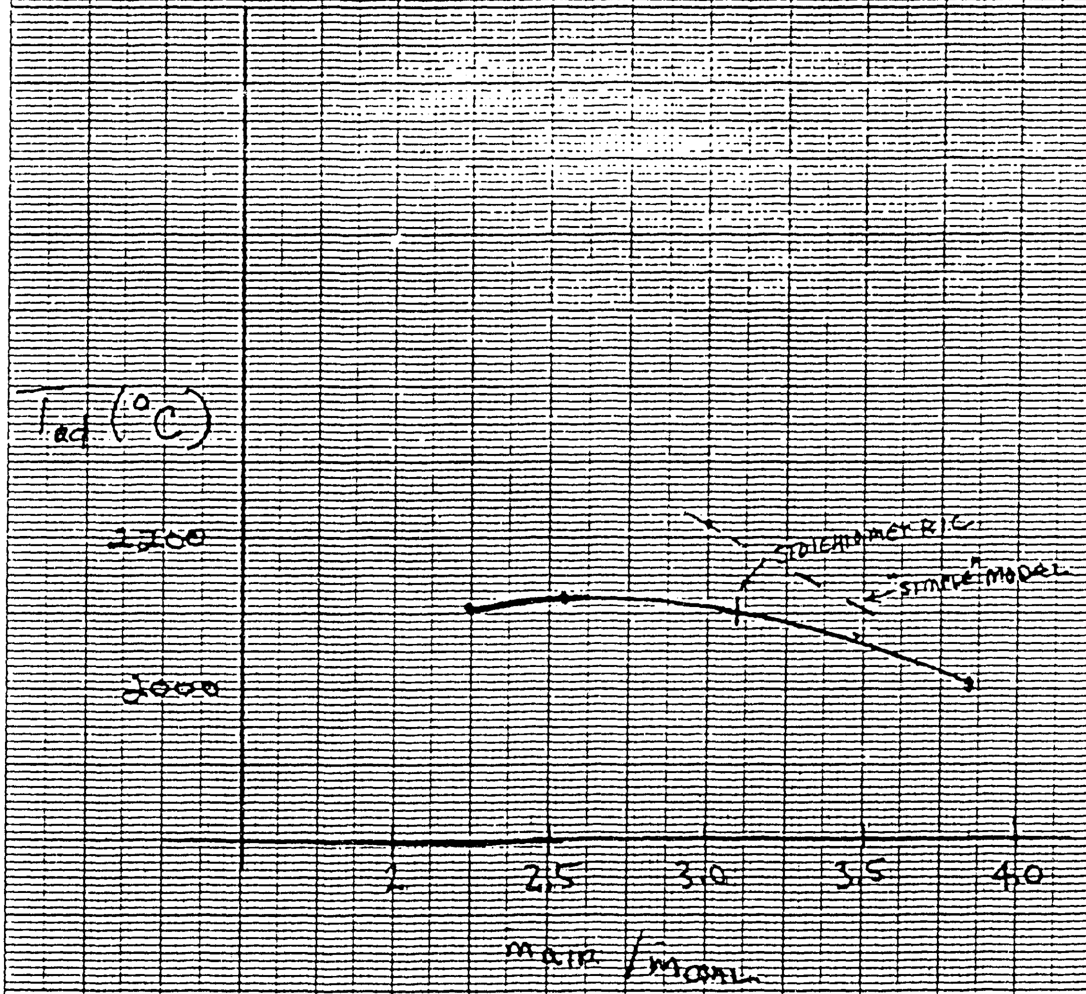

Figure 18. Adiabatic Flame Temperature 5% O_2 in Flue Gas Recirculated $\dot{M}_f/\dot{M}_{coal}$ = 3.26

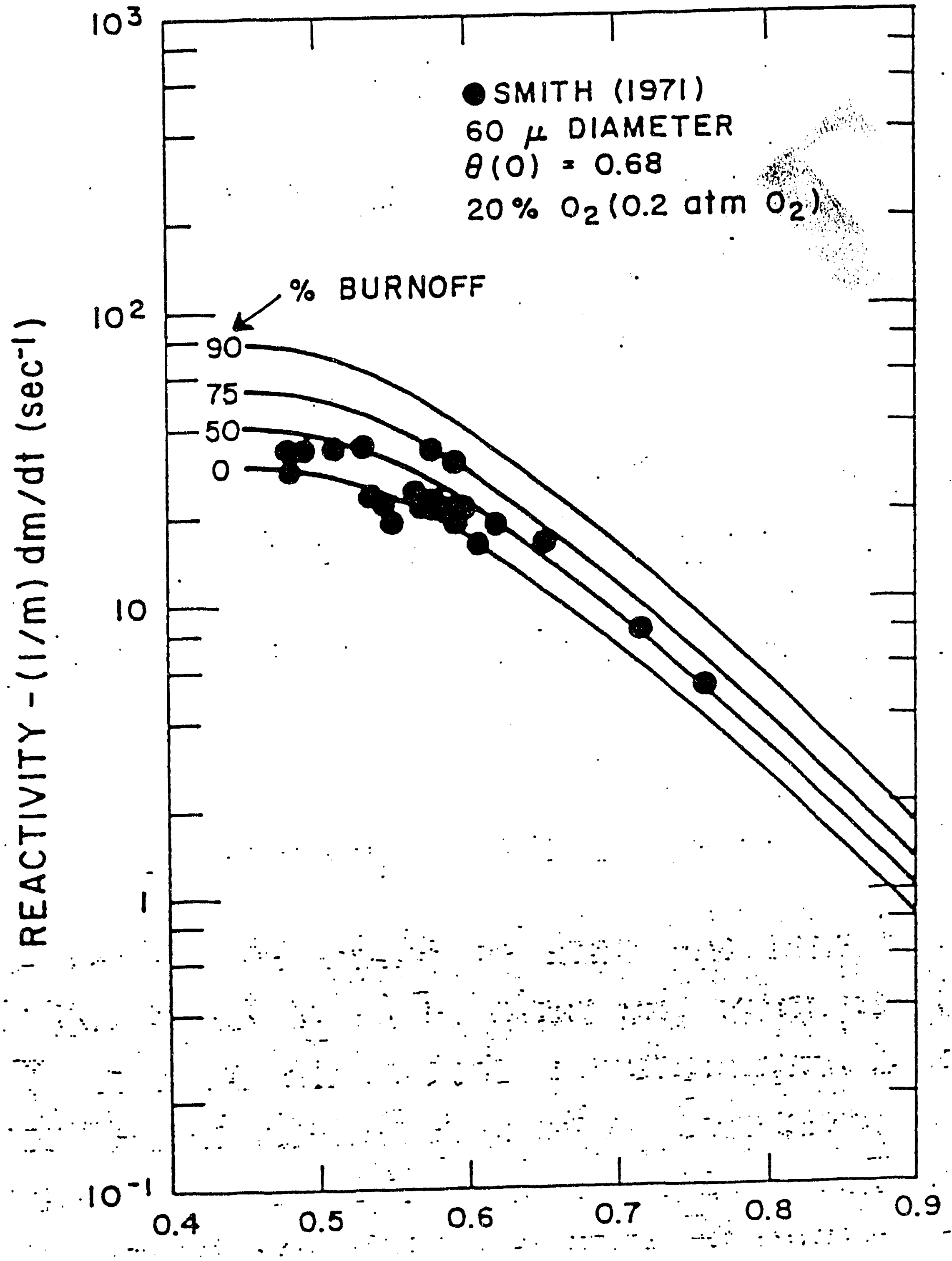

Figure 19.[11] Comparison with Oxidation Data for Bituminous Char.

A-1

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MAY 26 1993

April 2, 1993

Mr. Robert J. Fisher
Assistant Chief
Department of Energy
DOE Chicago Field Office
9800 South Cass Avenue
Argonne, IL 60439

Subject: Contract No. DE-AC22-92PC92151

ADL Reference: 41244

Dear Mr. Fisher:

As you requested with your 17 March 1993 letter we have removed the proprietary markings (RESTRICTED DOCUMENT) from the Draft Progress Report and are resubmitting the report herewith.

If you have any questions, please feel free to call me at (617) 498-5646.

Very truly yours,

ARTHUR D. LITTLE, INC.

Matthew J. Ring
Senior Contracting Officer

MJR/vd

Enclosure

cc: Dona Sheehan, PETC, Bldg. 921, w/o encl.
Clifford Smith, PETC, Bldg. 922,w/o encl.
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41244

A. Syska

Department of Energy
DOE Chicago Field Office
9800 South Cass Avenue
Argonne, Illinois 60439

March 17, 1993

Arthur D. Little, Inc.
MAR 19 1993
Govt. Contracting Office

Mr. Matthew J. Ring
Senior Contracting Officer
Arthur D. Little, Inc.
Acorn Park
Cambridge, MA 02140-2390

Dear Mr. Ring:

SUBJECT: CONTRACT NO. DE-AC22-92PC92151, DRAFT PROGRESS REPORT INDUSTRIAL PULVERIZED COAL LOW NOx BURNER: PHASE 1 - APRIL 1992- MARCH 1993.

We do not agree with the proprietary markings on the above-identified report and are returning it to you. The report includes proprietary markings (RESTRICTED DOCUMENT) in Figure 1, that are not in accordance with the Data provisions of the subject contract.

By copy of this letter we are advising the below-listed PETC employees to return to you all copies of the original report, or at least the material marked proprietary.

Please contact the DOE Contracting Officer's Representative and Mr. Hugh Glenn of this office at (708) 252-2178 to discuss how the marked information is to be treated.

Sincerely,

Robert J. Fisher

Robert J. Fisher
Assistant Chief
Office of Intellectual
Property Counsel

Arthur D. Little, Inc
MAR 24 1993
Govt. Contracting Office

Enclosure:
As stated

cc: Dona Sheehan, PETC, Bldg. 921, w/o encl.
Clifford Smith, PETC, Bldg. 922, w/o encl.
Office of Technology Transfer, PETC, Bldg. 94, w/o encl.
Curtis McBride, PETC, Bldg. 922, w/o encl.

DATE FILMED

10/19/93

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