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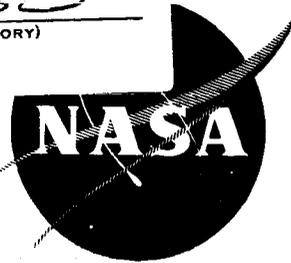
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INVESTIGATION OF THE FLAME STRUCTURE OF A THERMALLY UNSTABLE FUEL

BY

S. HERSH, B.R. LAWVER, R.J. HOFFMAN, AND B.P. BREEN

FINAL REPORT

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FOREWORD

This final report documents the work performed at the Dynamic Science Division of Marshall Industries under NASA Contract Number NAS7-442, relating to the description of the flame structure of thermally unstable fuels. At Dynamic Science, this work was assigned the project number SN-81 B & C. Two additional reports will be published under this NASA contract which will describe the work performed under Dynamic Science project number SN-81 A and SN-81 D.

All phases of this contract were monitored by Dr. Charles Feiler and Dr. Richard Priem of NASA-Lewis Research Center. At Dynamic Science, Dr. B. P. Breen was Program Manager of the total contract (NAS7-442). Mr. R. J. Hoffman acted as Technical Manager of the work reported in this report (SN-81 B & C). The analytical model (Appendix A) was developed by Mr. S. Hersh, Mr. B. R. Lawver designed and constructed the experimental equipment and the experiments were conducted by Mr. Hersh and Mr. Lawver. Dr. E. W. Nadig was responsible for the physical properties, Appendix B.

Thanks are expressed to Mr. John White, Mr. Wm. Irwin, and Mr. Frank Cummings for their assistance in constructing and operating the experimental equipment.

ABSTRACT

The results of an investigation of the hydrazine/nitrogen tetroxide droplet flame are reported. The results of the experimental work are the temperature and stable species concentration profiles. It is concluded that a hydrazine decomposition flame exists at the droplet surface causing a large temperature gradient which results in much higher burning rate than found for thermally stable fuels.

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SUMMARY

The flame structure of a 2500 micron hydrazine drop burning in nitrogen tetroxide was determined experimentally by measuring the temperature and specific concentrations at a plane 90° from the stagnation point. The hydrazine drop diameter was maintained constant by suspending it on the end of a water cooled hypodermic needle attached to a constant flow rate syringe. N_2O_4 was fed into the burner, establishing a thermal convective flow field around the drop. The flame was probed with a shielded 0.002 inch platinum-platinum/10% rhodium thermocouple as well as a concentration sampling probe. Both an "outer" and an "inner flame" were visually observable. The locations of these "flames" do not coincide with the high temperature points in the drop combustion zone, and therefore are not true flames but regions of strong visible radiation.

The experiments indicate that hydrazine decomposes very close to the liquid drop surface producing a steep temperature gradient which controls the burning rate. Although not proven conclusively, the zone between the drop and the inner flame appears to consist primarily of hydrazine decomposition products.

INTRODUCTION

An understanding of the processes by which a liquid fuel drop is vaporized and burned in the presence of an oxidizing media is essential to predicting the combustion process within a liquid rocket engine. In the course of an investigation of the rate of burning of a hydrazine drop in an atmosphere of N_2O_4/NO_2 , it was found that conventional methods of treating droplet burning did not give results in agreement with the experiment.

In an attempt to account for the difference in burning rate of an exothermic fuel drop (N_2H_4) and an endothermic drop (Heptane), experimental hydrazine decomposition rate data were applied to the prediction of combustion within the low velocity regions of a rocket engine (Ref. 1). While this approach showed promise of predicting correct burning rate and engine performance for the hydrazine family of fuels, it appeared appropriate, in view of the absence of similar studies, to investigate the two flame nature of hydrazine/nitrogen tetroxide burning in detail.

The objective of this work was thus to provide a description of the oxidation of thermally unstable rocket fuels so that model of burning rate within a combustion chamber could be developed.

SYMBOLS

- D_{ij} = the binary diffusion coefficients of i^{th} specie into each of the other species (cm^2/sec)
- H_i = the enthalpy of the i^{th} species at temperature T. (cal/mole)
- H_{fo} = the enthalpy of the fuel at its initial conditions (cal/mole)
- K = thermal conductivity ($\text{cal}/\text{cm sec } ^\circ\text{C}$)
- K_m = equilibrium constant for the m^{th} reaction
- l = index for atomic species (e.g. $l=1$ indicates the 1st atomic specie)
- P_i = partial pressure of the i^{th} specie ($P_i/P = x_i$)
- R = gas constant
- s = total number of atomic species
- W_f = the fuel burning rate in (moles/second)
- W_i = rate of transport of the i^{th} chemical species in (moles/second)
- x_i = the mole fraction of the i^{th} specie (moles i /moles $_{(\text{mix})}$)
- $\alpha_{i,l}$ = the number of l atoms in the i^{th} specie
- $\alpha_{f,l}$ = the number of l atoms in a fuel molecule.
- $\beta_{i,m}$ = stoichiometric coefficients for the reactants of the m^{th} reaction
- $\epsilon_{i,m}$ = stoichiometric coefficients for the products of the m^{th} reaction
- ν = total number of chemical species

DISCUSSION

Although semi-analytical models for the determination of the burning rate of thermally stable fuel drops have been developed (Ref. 2), an analytical determination of the burning rate of a thermally unstable drop, such as hydrazine has not been achieved.

Liquid hydrazine burning in NTO exhibits two visibly discrete "flames" (Figure 1). Initial studies ascribed the "inner flame" to the energy released by hydrazine decomposition, the "outer flame" being the region of oxidation of the decomposition products.

As part of a program to gain understanding of the processes underlying the hydrazine/NTO flame structure an analytical model which could be used to determine the N_2H_4 burning rate under several simplifying assumptions was formulated (Appendix A) and experimental studies of the flame properties were conducted.

The transport properties required by the analysis, consisting of both measured values and approximate formulas where no experimental data was available, are compiled in Appendix B. This information is of a general nature and is therefore of use for any analysis for which information of this type is necessary.

Two series of experimental tests were conducted. Both series utilize the same basic equipment, the difference being in the instrumentation used to measure the flame parameters. Temperature profiles from the ambient atmosphere to the drop surface were obtained using a thermocouple probe while individual specie concentrations in the hydrazine-nitrogen tetroxide flame were determined from a mass spectrometric analysis of samples withdrawn from the flame with a sampling probe.

Hydrazine was chosen for use in these tests because it is the basic ingredient in the most widely used thermally unstable fuel and the result would have the greatest practical application. Burning droplet tests were run with propellant grade hydrazine burning the oxidizers, nitrogen tetroxide, oxygen, air, and oxygen/nitrogen mixtures. Temperature profiles were measured with all of these oxidizers, however, concentration probing was done only for the nitrogen tetroxide oxidizer.

APPARATUS AND PROCEDURE

Objectives. - The purpose of these experiments was to measure the flame structure of a hydrazine droplet burning in nearly stagnant nitrogen tetroxide vapor. The flame structure was defined by measuring the temperature profile and chemical species concentration in the free convective flame surrounding a constant diameter hydrazine droplet. Tests were also conducted with hydrazine burning in oxygen/nitrogen mixtures to measure the effect of the oxidizer concentration on the flame structure.

These tests were conducted with propellant grade hydrazine and nitrogen tetroxide. The oxygen and nitrogen gases were supplied from standard gas cylinders.

The Burner. - The tests were conducted with the burner apparatus* shown in Figure 2. Basically the burner consists of a test section which permits observation of the combustion and the related propellant feed systems. The burner is designed such that either liquid fuel droplets or gaseous fuels can be burned in gaseous oxidizers. A flow schematic of the burner propellant feed system is shown in Figure 3.

The droplet flame is produced by suspending the fuel droplet from a water cooled hypodermic needle in the burner test section. The fuel is fed from a syringe into the suspension needle with a variable speed syringe drive. The droplet diameter can be maintained constant by proper adjustment of the syringe drive.

The fuel droplet suspension needle is shown in Figure 4. It is a 0.010" O.D. (0.005" I.D.) tube surrounded by two larger concentric tubes through which water is circulated. The water provides cooling of the exposed proportion of the small needle, to prevent boiling and thermal decomposition of the fuel within the needle. The oxidizing gases are fed into the burner through an injector tube located in the burner base plate. The oxidizer flow rates are controlled with calibrated sonic orifices, by regulating the orifice upstream pressures. Oxygen is fed to the burner orifice through a pressure regulator mounted on the supply bottle. The N_2O_4 vapor is generated by electrically heating the storage tanks and supply lines. The N_2O_4 vapor temperature and flowrate are controlled by regulating the heater current. The N_2O_4 flowrate is also measured with a calibrated sonic orifice.

*This burner was designed and built under Contract AF 04(611)-11616.

The burner test section has opposing pyrex windows for observing and photographing the flames. The combustion products are removed from the burner tube with an air driven ejector mounted to the burner top plate.

Temperature Probe. - The temperature probe is illustrated in Figure 5. This probe consists of 0.002" diameter platinum/platinum-10% rhodium thermocouple wires mounted in a small ceramic insulator encased in a stainless steel sheath. The steel sheath protects the exposed wires from the hot combustion gases when the probe is fully submerged into the flame, as is illustrated in Figure 7.

A 0.010" diameter platinum/platinum-10% rhodium thermocouple without the steel sheath was used for the initial probing of the N_2H_4 flame. These tests indicated that heat conducted along the thermocouple leads from the exposed portion of the wires caused erroneous temperature measurements, therefore, it was necessary to provide a steel sheath. In addition the response of the larger thermocouple was not satisfactory. To improve the response time an attempt was made to construct thermocouples with 0.001" diameter wires, however this proved to be too time consuming to be warranted. The response of the 0.002" diameter thermocouple is on the order of 40 milliseconds.

The thermocouples were calibrated by immersing the probe in compounds with known melting point temperatures. The calibration data points are shown in Figure 6 along with a plot of the standard calibration for thermocouples obtained from Reference 3. The good agreement between the observed and standard calibrations encouraged the use of the latter in the higher temperature range where calibration measurements were not made.

The thermocouple probe is traversed through the droplet flame with a variable speed electric motor drive mechanism as shown schematically in Figure 7. The thermocouple position is indicated on an oscilloscope by using the signal from a 5000 Ω linear potentiometer to drive the oscilloscope horizontal sweep. The thermocouple output voltage is applied to the oscilloscope vertical amplifier input, providing direct temperature versus distance profile throughout the flame region. The temperature profile is recorded by photographing the oscilloscope screen with a bezel mounted Poloroid camera.

Concentration Probe. - The concentration sampling technique described in Reference 4, was used to measure the chemical species concentrations in the N_2H_4 flames. The technique consists of kinetically freezing the reacting gases by drawing them through a very small supersonic nozzle into a vacuum environment. The rapid expansion quenches the reactions, thus preserving the stable chemical species for subsequent analysis.

The concentration sampling probe is a 6mm O.D. quartz tube with 1mm walls, drawn to a fine point at the sampling end to minimize the flame disturbance. The nozzle throat diameter is 0.002 inches in diameter which gives an expansion ratio of 6200/1. The inlet area ratio is 300/1. Based on a flame gas velocity of 6-10 ft/sec, the nozzle residence time is approximately 40 μ sec.

The sampling system is shown in Figure 8. The gas samples were collected in bottles and then analyzed on a Hitachi Perkin-Elmer mass spectrograph.

The mole fractions were calculated from the relationship

$$x_i = \frac{P_i}{P_T}$$

where

$$\begin{aligned} x_i &= \text{mole fraction of species } i \\ P_i &= \text{partial pressure of species } i \\ P_T &= \text{total pressure of sample} \end{aligned}$$

The partial pressures are determined from the magnitude of the mass peaks measured with the mass spectrograph. The partial pressures are found from the relationship

$$h_m = \sum_i^n K_i^m P_i$$

where

$$\begin{aligned} h_m &= \text{measured peak height of mass number } (m) \\ K_i &= \text{sensitivity of species } (i) \text{ to mass number } (m) \end{aligned}$$

There is one equation for each mass number and the problem then is the solution of a system of simultaneous equations. The number of equations is equal to the number of species involved. To simplify the data reduction, these equations were programmed on a CDC 3600 computer. The input consists of the measured mass peak heights of the species of interest and the sensitivity factors. The output is the species mole fractions.

Analysis of the mass peaks for the hydrazine/NTO flame indicated that the mass numbers of interest are,

<u>Mass Number</u>	<u>Species</u>
2	H ₂
17	NH ₃
18	H ₂ O
28	N ₂
29	N ₂ H ₄
30	NO
32	O ₂
44	N ₂ O
46	NO ₂

The mass number 29 was used for the hydrazine to distinguish it from oxygen, mass number 32. The hydrazine exhibits a 29 peak that is 43% of its 32 peak, also it exhibits a strong peak at mass number 31. The above mass numbers and species were used in determining the hydrazine/NTO flame structures.

Experimental Results

Temperature Profiles. - A typical temperature profile for a hydrazine-nitrogen tetroxide flame is shown in Figure 9. The temperature begins to rise before the "outer flame" is reached and rises to a peak between the "inner" and "outer flame." This peak temperature corresponds to a blue region which is visually observable but does not appear in the flame photographs. Attempts to use film with a greater spectral sensitivity in the blue region than the Polaroid color film normally used also failed to reproduce this region. The temperature decreases smoothly from its maximum, through the "inner flame," to a relatively high value close to the drop surface. It should be noted that the locations of the visible "flames" do not coincide with the high temperature points. Thus what have been denoted as the "inner" and "outer flames" are not true flames but regions of strong visible radiation.

The shape of this temperature profile, in particular the high temperature close to the drop surface would indicate that hydrazine decomposition had occurred at the droplet surface. To further examine this behavior, hydrazine droplets were burned in oxygen-nitrogen atmospheres at various O₂-N₂ concentrations. A hydrazine-air profile is shown in Figure 10. As can be seen, both the maximum and drop surface temperatures, are less than that obtained for the hydrazine-NTO system, (Figure 9).

The O_2-N_2 concentrations were varied over a small range by varying the pressures upstream of the sonic orifices. Figures 11a and b, are temperature profiles for hydrazine burning in oxygen-nitrogen atmospheres of different O_2-N_2 concentrations, Figure 11a being at a higher value of O_2-N_2 ratio. The temperature profile shown in Figure 12 was obtained by reducing the oxygen concentration until the only visual evidence of burning was a halo-type glow close to the drop surface (Figure 13). Under this condition the actual temperatures were lower than those for the other cases but the steep gradient at the drop surface is still evident. The change in distance scale should be noted. Also it should be noted that the temperature near the drop surface with N_2H_4 /air is not much higher than the maximum temperature of the halo flame.

To verify that the temperature profiles obtained for hydrazine were functions of the hydrazine itself and not related to the experimental procedures or equipment, the flame surrounding a heptane drop was also probed. Heptane was selected because of its endothermicity. Therefore, if the temperature gradient at the surface of the hydrazine drop is due to hydrazine decomposition, it would be expected that the profile obtained for the heptane drop would not exhibit this steep gradient. Figure 14a is the temperature profile obtained for heptane. As can be seen, the temperature rises to a peak and then decreases to the droplet boiling temperature. Figure 14b shows that two visible "flames" are also present when heptane is burned in nitrogen tetroxide. Other investigators, for example, Wharton, Miller, et al, (Ref. 5 and 6) have also observed a dual reaction zone in premixed n-butane- NO_2 flames. These results suggest that the two "visible flames" are associated with the oxidizer while two real flames are found only for hydrazine. The combustion of hydrazine with any oxidizer will be a two flame structure consisting of decomposition at the droplet surface and the oxidation of the decomposition products.

Concentration Profiles. - The concentration data taken at several positions in the flame are plotted versus reduced radius in Figure 15. It is noted that hydrazine was not found in measurable quantities. There is some question as to whether this is due to the nonexistence of hydrazine in the flame or to hydrazine decomposition in the sampling probe.

Figure 15 also indicates that the outer flame consists of the decomposition of NO_2 to NO and O_2 . The mole fractions of NO and O_2 decrease as the inner flame is approached.

The fact that the decrease in NO occurs in the region where the temperature reaches a maximum is in agreement with the results of Reference 5. That investigation demonstrated that the maximum temperature

in an n-butane- NO_2 flame was due to the energy release associated with the decomposition of NO . At the inner flame boundary the oxidizer concentrations drop sharply to very small amounts.

Several phenomena which relate to the accuracy of the sampling results were observed. It was found that ammonium nitrate was collected in the probe throat at the yellow inner flame boundary. This problem prevents an accurate description of the inner flame composition being made by direct sampling. It is not apparent whether the ammonium nitrate is formed in the probe or is precipitated from the flame. The formation of the ammonium nitrate in the probe would require very fast reaction rates at low temperature. Ammonium nitrate is a stable product of the room temperature hypergolic reaction between NH_3 and NO_2 , therefore, it is conceivable that it may also be an intermediate of the high temperature reaction.

The relative amounts of nitrogen were larger than they should have been, but this may be due to contamination from the probe purge system.

CONCLUSIONS

The facts that, all tests conducted with hydrazine indicate the presence of a steep temperature gradient near the drop, and that this property is associated only with hydrazine droplets, lead to the conclusion that hydrazine decomposition occurs at or very near the drop surface. This conclusion appears to be further verified by the concentration profiles obtained with the sampling probe. The failure to detect any hydrazine in the flame especially in the region near the drop surface, when viewed in the light of the temperature profiles, is a further indication of hydrazine decomposition at or close to the drop surface.

The results of these experiments indicate that at low Reynolds number the dominant process controlling the hydrazine burning rate is hydrazine decomposition, since this mechanism determines the temperature gradient at, and therefore the heat transfer to, the droplet surface. The existence of the decomposition flame at the drop surface causes hydrazine to have a significantly higher burning rate than thermally stable fuels such as heptane.

APPENDIX A

ANALYTICAL VAPOR PHASE DECOMPOSITION MODEL

Analytical Model

Derivation of Equations. - The analysis of the burning of a drop of hydrazine in a nitrogen tetroxide atmosphere has been formulated in a manner similar to that described by Coffin and Brokaw (Ref. 2), in which equilibrium chemistry was assumed. As in Reference 2, the problem is set up for a general fuel-oxidizer combination with the particular system under examination determining the necessary physical properties.

The equations describing the processes occurring in the region between the fuel drop surface and the ambient oxidizer atmosphere are:

Conservation of Mass

$$W_\ell = \sum_{i=1}^{\nu} \alpha_{i,\ell} W_i \quad \ell = 1, \dots, s$$

W_i = Rate of transport of the i^{th} chemical species in (moles/sec)

ℓ = Index for atomic species. (e.g. $\ell=1$ indicates the 1st atomic specie)

$\alpha_{i,\ell}$ = The number of ℓ atoms in the i^{th} specie

s = Total number of atomic species

ν = Total number of chemical species

This equation expresses the fact that the transport of the ℓ^{th} atomic species through any surface is equal to the sum of the contributions due to the transport of chemical species containing ℓ atoms. Since there is no sink in the system the net transport of oxidizer atoms is zero. However, there is a net production of fuel atoms since the drop is a fuel source. Therefore

$$W_\ell = \alpha_{f,\ell} W_f$$

W_f = the fuel burning rate in (moles/second) and $\alpha_{f,\ell}$ = the number of ℓ atoms in a fuel molecule.

or

$$\sum_{i=1}^{\nu} \alpha_{i,\ell} W_i = \alpha_{f,\ell} W_f \quad \ell = 1, \dots, s$$

which describes the conservation of atoms in the systems.

Energy Equation

$$W_f H_{fo} - \sum_{i=1}^{\nu} W_i H_i = -4\pi r^2 K \frac{dT}{dr}$$

H_i = The enthalpy of the i^{th} species at temperature T .

H_{fo} = The enthalpy of the fuel at its initial conditions

K = Thermal conductivity

Diffusion Equations

The equations describing the diffusion of the i^{th} species are obtained from the general diffusion equations (Eqn. 8-1-3 of Ref. 7) by neglecting external forces, pressure gradients, and thermal diffusion. In addition it is also assumed that each species i obeys the ideal gas law and that the binary diffusion coefficients of the i^{th} specie (D_{ij}) into each of the other species are equal, i.e. $D_{ij} = D_{ji}$. Under these conditions the diffusion equations can be put in the form (Ref. 2):

$$W_i = -4\pi r^2 \frac{P}{RT} \left(D_i \frac{dx_i}{dr} + \frac{x_i}{x_\nu} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} \right) + \frac{x_i}{x_\nu} W_\nu \quad i = 1, \dots, \nu-1$$

R = Gas constant

x_i = The mole fraction of the i^{th} specie $\left(\frac{\text{Moles } i}{\text{Moles (mix)}} \right)$

For a ν component gas there are $\nu-1$ independent transport rates.

Chemistry Equations

If the transport rates are much slower than the chemical rates the burning process can be considered to occur under equilibrium conditions. For a ν component system there are $\nu-s$ independent equilibrium relationships among the chemical species.

These relations are of the form

$$K_m = \frac{\prod_i^{\nu} p_i^{\epsilon_{i,m}}}{\prod_i^{\nu} p_i^{\beta_{i,m}}}$$

p_i = Partial pressure of the i^{th} specie $\left(\frac{p_i}{P} = x_i \right)$

K_m = Equilibrium constant for the m^{th} reaction.

$\epsilon_{i,m}$ = Stoichiometric coefficients for the products of the m^{th} reaction

$\beta_{i,m}$ = Stoichiometric coefficients for the reactants of the m^{th} reaction.

If the transport and chemical rates are comparable, the relationships between the gas components must be determined by finite rate chemistry.

Normalization Equation

$$\sum_{i=1}^{\nu} x_i = 1$$

That is, the sum of the mole fractions is unity.

In summary, there are s conservation of mass equations, one energy equation, $\nu-1$ diffusion equations, ν -s chemical equations, and a normalization equation for a total of $2\nu+1$ equations. There are $\nu+1$ unknown W 's (W_i $i=1, \dots, \nu$ and W_f) and ν unknown fractions (x_i $i=1, \dots, \nu$). Thus there are $2\nu+1$ unknowns and $2\nu+1$ equations and the problem is solvable in principle.

Since there are νW 's and $\nu-1$ diffusion equations one of the mass conservation equations must be used to determine the ν^{th} W . The conservation equations are

$$\sum_{i=1}^{\nu} \alpha_{i,\ell} W_i = \alpha_{f,\ell} W_f \quad \ell=1, \dots, s$$

Select ℓ for which $\alpha_{f,\ell} \neq 0$

Designate this value of ℓ as $\lambda 1$.

Then

$$\sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} W_i + \alpha_{\nu,\lambda 1} W_{\nu} = 0$$

or

$$W_{\nu} = - \frac{1}{\alpha_{\nu,\lambda 1}} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} W_i$$

Substituting for W_i from the diffusion equation and solving for W_{ν}

$$W_{\nu} = \frac{\frac{4\pi r^2 P}{RT} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \left(D_i \frac{dx_i}{dr} + \frac{x_i}{x_{\nu}} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} \right)}{\alpha_{\nu,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_{\nu}}}$$

Therefore

$$-W_i = 4\pi r^2 \frac{P}{RT} \left\{ \left(D_i \frac{dx_i}{dr} + \frac{x_i}{x_{\nu}} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} \right) - \frac{\frac{x_i}{x_{\nu}} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \left(D_i \frac{dx_i}{dr} + \frac{x_i}{x_{\nu}} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} \right)}{\alpha_{\nu,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_{\nu}}} \right\}$$

In order to eliminate all the W 's from the energy equation a second mass conservation equation is used to express W_f in terms of the W_i 's. Select ℓ for which $\alpha_{f,\ell} \neq 0$. Designate this value of ℓ as λ_2 . The $\ell = \lambda_2$ conservation equation gives

$$W_f = \frac{1}{\alpha_{f,\lambda_2}} \sum_{i=1}^{\nu} \alpha_{i,\lambda_2} W_i$$

Substituting into the energy equation gives

$$\frac{H_{fo}}{\alpha_{f,\lambda_2}} \sum_{i=1}^{\nu} \alpha_{i,\lambda_2} W_i - \sum_{i=1}^{\nu} W_i H_i = -4\pi r^2 K \frac{dT}{dr}$$

Rearranging

$$\sum_{i=1}^{\nu-1} \left(\frac{\alpha_{i,\lambda_2}}{\alpha_{f,\lambda_2}} H_{fo} - H_i \right) W_i + \left(\frac{\alpha_{\nu,\lambda_2}}{\alpha_{f,\lambda_2}} H_{fo} - H_{\nu} \right) W_{\nu} = -4\pi r^2 K \frac{dT}{dr} \quad (1)$$

The $S-2$ remaining conservation equations are

$$\sum_{i=1}^{\nu} \alpha_{i,\ell} = \frac{\alpha_{f,\ell}}{\alpha_{f,\lambda_2}} \sum_{i=1}^{\nu} \alpha_{i,\lambda_2} W_i \quad \begin{array}{l} \ell = 1, \dots, s \\ \ell \neq \lambda_1, \lambda_2 \end{array}$$

$$\sum_{i=1}^{\nu} \left(\alpha_{i,\ell} - \frac{\alpha_{f,\ell}}{\alpha_{f,\lambda_2}} \right) W_i = 0$$

$$\sum_{i=1}^{\nu-1} \left(\alpha_{i,\ell} - \frac{\alpha_{f,\ell}}{\alpha_{f,\lambda_2}} \alpha_{i,\lambda_2} \right) W_i + \left(\alpha_{\nu,\ell} - \frac{\alpha_{f,\ell}}{\alpha_{f,\lambda_2}} \alpha_{\nu,\lambda_2} \right) W_{\nu} = 0 \quad (2)$$

Equations (1) and (2) are of the form

$$\sum_{i=1}^{\nu-1} A_i W_i + B W_{\nu} = C \quad (3)$$

Examining the terms in the above equation separately gives for the first term

$$-\frac{RT}{4\pi r^2 P} \sum_{i=1}^{\nu-1} A_i W_i = \sum_{i=1}^{\nu-1} A_i D_i \frac{dx_i}{dr} + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} \left(\sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} \right) - \frac{1}{S} \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} D_i \frac{dx_i}{dr} - \frac{1}{S} \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr}$$

where

$$S = \alpha_{\nu,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu} .$$

This relation can be rearranged into the following form

$$-\frac{RT}{4\pi r^2 P} \sum_{i=1}^{\nu-1} A_i W_i = \sum_{i=1}^{\nu-1} \left\{ A_i - \frac{\alpha_{i,\lambda 1}}{S} \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} \left(1 - \frac{1}{S} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu} \right) \right\} D_i \frac{dx_i}{dr}$$

The second term is

$$\frac{RT}{4\pi r^2 P} B W_\nu = \frac{B}{S} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} D_i \frac{dx_i}{dr} + \frac{B}{S} \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu} \sum_{j=1}^{\nu-1} D_j \frac{dx_j}{dr} = \frac{B}{S} \sum_{i=1}^{\nu-1} \left(\alpha_{i,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu} \right) D_i \frac{dx_i}{dr}$$

Therefore Equation (3) becomes

$$\sum_{i=1}^{\nu-1} \left[\left\{ A_i + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} - (B + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu}) \left[\frac{\alpha_{i,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu}}{\alpha_{\nu,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu}} \right] \right\} D_i \frac{dx_i}{dr} \right] = -\frac{CRT}{4\pi r^2 P}$$

For Equation (1) $C = -4\pi r^2 K \frac{dT}{dr}$

For Equations (2) $C = 0$

Substituting into the above equation gives

$$\sum_{i=1}^{\nu-1} \left[\left\{ A_i + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu} - (B + \sum_{i=1}^{\nu-1} A_i \frac{x_i}{x_\nu}) \left[\frac{\alpha_{i,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu}}{\alpha_{\nu,\lambda 1} + \sum_{i=1}^{\nu-1} \alpha_{i,\lambda 1} \frac{x_i}{x_\nu}} \right] \right\} \delta_i \frac{dx_i}{dT} \right] = C' \quad (4)$$

where $\delta_i = \frac{D_i}{KRT}$

For Equation (1)

$$A_i = \left(\frac{\alpha_{i,\lambda_2}}{\alpha_{f,\lambda_2}} H_{fo} - H_i \right)$$

$$B = \left(\frac{\alpha_{\nu,\lambda_2}}{\alpha_{f,\lambda_2}} H_{fo} - H_i \right)$$

$$C' = \frac{1}{P}$$

For Equations (2)

$$A_i = \left(\alpha_{i,l} - \frac{\alpha_{f,l}}{\alpha_{f,\lambda_2}} \alpha_{i,\lambda_2} \right) \quad \begin{array}{l} l = 1, \dots, S \\ l \neq \lambda_1, \lambda_2 \end{array}$$

$$B = \left(\alpha_{\nu,l} - \frac{\alpha_{f,l}}{\alpha_{f,\lambda_2}} \alpha_{\nu,\lambda_2} \right)$$

$$C' = 0$$

The chemical equilibrium relations are

$$K_m = \frac{\prod_i^{\nu} p_i^{\epsilon_{i,m}}}{\prod_i^{\nu} p_i^{\beta_{i,m}}}$$

Taking the temperature derivative of this equation gives

$$\frac{d}{dT} \prod_i^{\nu} p_i^{\epsilon_{i,m}} = K_m \frac{d}{dT} \prod_i^{\nu} p_i^{\beta_{i,m}} + \prod_i^{\nu} p_i^{\beta_{i,m}} \frac{dK_m}{dT}$$

But $\frac{d}{dT} \prod_i^{\nu} p_i^{\epsilon_{i,m}} = \prod_j^{\nu} p_i^{\epsilon_{j,m}} \sum_{i=1}^{\nu} \epsilon_{i,m} p_i^{-1} \frac{dp_i}{dT}$

Substituting and rearranging yields

$$\sum_{i=1}^{\nu} \left\{ \left[\epsilon_{i,m} \prod_j^{\nu} p_j^{(\epsilon_{j,m} - \beta_{j,m})} - K_m \beta_{i,m} \right] \frac{1}{x_i} \frac{dx_i}{dT} \right\} = \frac{dK_m}{dT} \quad (5)$$

Where the relationship between the partial pressure and the mole fraction, $x_i = p_i/p$, has been used.

It should be noted that Equations (5) provide ν -S relations between the x_i 's. While these equations were derived for equilibrium chemistry and other chemical assumptions, e. g. finite rate kinetics, or arbitrary specification of x_i vs T, can be made, providing a set of ν -S independent relationships are obtained.

The normalization equation can also be differentiated to give

$$\sum_{i=1}^{\nu} \frac{dx_i}{dT} = 0 \quad (6)$$

Equations (4), (5), and (6) are respectively S-1, ν -S, 1, equations for a total of ν equations for the ν unknown x_i 's.

The above equations can be numerically integrated to give x_i vs T for all the species. The mass conservation equations can then be used to obtain the spatial specie distribution from the drop to the ambient atmosphere.

Since the experimental programs described above indicated that the hydrazine burning rate was controlled by decomposition close to the drop surface the usefulness and applicability of an analytical model of this type toward the prediction of the burning rate was uncertain and a computer code was not developed.

APPENDIX B

PHYSICAL PROPERTIES

For use in the analytical hydrazine decomposition model, enthalpies, diffusion coefficients, and thermal conductivities are needed for the following chemical species: H_2 , NH_3 , N_2 , H_2O , NO , NO_2 , O_2 , O , N , NH_4NO_3 , and N_2H_4 .

Enthalpies. - The enthalpies of H_2 , NH_3 , N_2 , H_2O , NO , NO_2 , O_2 , O , and N are given in Reference (8) from $0^\circ K$ to $6000^\circ K$. The enthalpies of N_2H_4 and NH_4NO_3 are not given but can be determined from the same NASA-Lewis program. However, apparently the program shows that N_2H_4 does not exist as such at any temperature since equilibrium calculations are involved.

Diffusion Coefficients. - Two suitable expressions are available for determining the coefficient of diffusion in a binary mixture.

Hirschfelder, Curtiss, and Bird, (Ref. 4) give the analytical expression

$$D_{12} = 0.0026280 \frac{\sqrt{T^3 (M_1 + M_2) / 2M_1 M_2}}{p \sigma_{12}^2 \Omega_{12}^{(1,1)*} (T_{12}^*)}$$

where

D_{12} = diffusion coefficient

p = pressure

$$\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2}$$

σ_1, σ_2 = molecular diameters of species 1 and 2

T = temperature

M_1, M_2 = molecular weights of species 1 and 2

$\Omega_{12}^{(1,1)*}$ = integral parameter, approximately equal to 1.

T_{12}^* = overall reduced temperature between the two species

Another suitable expression for estimating the diffusion coefficient between species 1 and 2 is the empirical Gilliland (Ref. 9) expression

$$D = 0.0043 \frac{T^{3/2}}{p(V_A^{1/3} + V_B^{1/3})^2} \sqrt{\frac{1}{M_1} + \frac{1}{M_2}}$$

where

D = diffusivity, cm²/sec

T = temperature, °K

p = pressure atm

M₁M₂ = molecular weights of gases

V_A, V_B = molecular volumes at the normal boiling points, cc/g-mole

The molecular volume can be calculated from the molecular weight and the density of the substance at the boiling point. If the density at the boiling point is unknown, the molecular volume can be calculated from the sum of the atomic volumes. The atomic volumes are given in Reference 10.

Thus, if the molecular diameters are given (some are given in Ref. 7) the more exact Hirschfelder, Curtiss, and Bird equation can be used. If the molecular diameters are unknown, the Gilliland equation can be used. Either equation will give a reasonable estimate of the diffusion coefficient in a binary mixture.

Since a binary gas mixture is assumed in use of the equations, in the actual multicomponent case the diffusion coefficient can be obtained from a mass weighted average of the properties of the remaining species into which the ith specie is diffusing. If the specie diffuses into a mixture containing a large amount of one component with respect to the others in the mixture (i.e. approaching an overall binary mixture between the specie and the remaining material) the calculation will be reasonably accurate. The error involved if the overall mixture (specie diffusing plus the remaining materials) is not a binary mixture is not known at this time. It should be mentioned, that in a binary gas mixture, the dependence of the diffusion coefficient on composition is only slight.

Thermal Conductivities. - Since only limited experimental thermal conductivity data is available, it is necessary to use a theoretical expression to determine the thermal conductivity.

The thermal conductivity of a substance can be reasonably estimated from an analytical expression given by Hirschfelder, Curtiss, and Bird, (Ref. 7).

$$k = 1989.1 \times 10^{-7} \frac{\sqrt{T/M}}{\sigma^2 \Omega^{(2,2)*}(T^*)} \left(\frac{4}{15} \frac{C_p}{R} + \frac{1}{3} \right)$$

where

- k = thermal conductivity
 T = temperature
 M = molecular weight
 σ = molecular diameter
 $\Omega^{(2,2)*}$ = integral parameter, approximately equal to 1.
 C_p = heat capacity
 R = universal gas constant
 T^* = reduced temperature

As used in the model, the thermal conductivity, k , is included because of a comparison with heat conduction. Therefore, k represents an overall constant value. In this analysis, the thermal conductivity will be determined from a mass weighted average of the thermal conductivities of the respective species. The closer one specie comes to being present in a large amount, the more accurate the use of the mass weighted average thermal conductivity will be.

Whenever possible experimentally determined values of the thermal conductivity of the substance should be used.

High temperature thermal conductivity data (3000-4700°F) is available in Reference 11. Figure 16 shows experimental results obtained from Reference 11 in addition to results from Reference 12 (indicated by the broken line).

The following low temperature experimental thermal conductivity (k) data is available for all of the species except O, N, NH_4NO_3 , and N_2H_4 .

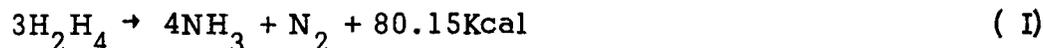
	Temperature,	$k \frac{\text{calories}}{\text{cm sec } ^\circ\text{C}}$	Reference
NH_3	- 57.6°C	3.82	13
	0	5.135	13
	100	7.09	13
N_2	-191.4	1.829	13
	- 78.4	4.305	13
	0	5.68	13
	100	7.18	13
20			

	<u>Temperature</u>	<u>k</u> $\frac{\text{calories}}{\text{cm sec}^\circ\text{C}}$	<u>Reference</u>
H ₂	-252.2°C	3.22	13
	- 78.4	30.65	13
	0	39.60	13
	100	49.94	13
H ₂ O(g)	46	4.580	13
	100	5.510	13
NO (g)	- 71.4	4.160	13
	0	5.55	13
NO ₂ (g)	55	8.88	13
O ₂	-191.4	1.721	13
	- 78.4	4.292	13
	0	5.70	13
	100	7.427	13
	80°K	1.701	14
	90	1.930	14
	100	2.159	14
	110	2.387	14
	120	2.614	14
	130	2.840	14
	140	3.064	14
	150	3.287	14
	160	3.508	14
	170	3.728	14
	180	3.946	14
	190	4.162	14
	200	4.375	14
	210	4.584	14
	220	4.790	14
	230	4.993	14
	240	5.194	14
	250	5.392	14
260	5.586	14	
270	5.780	14	
273.1	5.839	14	
280	5.970	14	
290	6.159	14	
293.1	6.218	14	
298.1	6.314	14	
300	6.350	14	

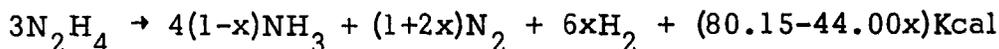
	<u>Temperature</u>	<u>k</u> $\frac{\text{calories}}{\text{cm sec}^\circ\text{C}}$	<u>Reference</u>
O ₂	310°K	6.547	14
	320	6.748	14
	330	6.954	14
	340	7.164	14
	350	7.378	14
	360	7.594	14
	370	7.812	14
	380	8.033	14

Thermal Decomposition of Hydrazine. - In connection with the model, information on the thermal decomposition of hydrazine is desired. Although there are discrepancies in the literature, the following information is considered significant.

Thomas (Ref. 15) shows that hydrazine decomposes according to two consecutive overall reactions at 20 atm total pressures



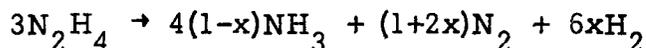
The overall decomposition reaction is



where x = the fraction of NH_3 decomposed.

Ammonia (NH_3) is only negligibly dissociated for temperatures below 400°K. As the temperature is raised above 400°K, NH_3 decomposes with the decomposition becoming complete at 800°K. Experimental results show that the decomposition reaction of NH_3 (II) is slower than reaction I. Hence, if the decomposition (I) occurs in the order of milliseconds, it then might be reasonable to assume that only part of the NH_3 could decompose.

Lucien (Ref. 16) carried out thermal decomposition studies of hydrazine in the ranges of 175° to 250° C and 300 to 430 psi. He found that the decomposition went according to



where $x = 0.017$ at 222° C and 300 psi

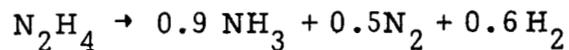
$= 0.034$ at 250° C and 430 psi

The calculated activation energy was about 73Kcal per mole which is greater by a factor of two than other reported values. The rate of decomposition decreased with pressure over the range studied.

Eberstein and Glassman (Ref.17) studied hydrazine decomposition. The rate constant for hydrazine was found to be

$$k = 10^{10.33} \exp(-36,170/RT) \text{sec}^{-1}$$

The stoichiometry observed was



The disagreements in the literature may indicate that the hydrazine decomposition mechanism changes with pressure and temperature.

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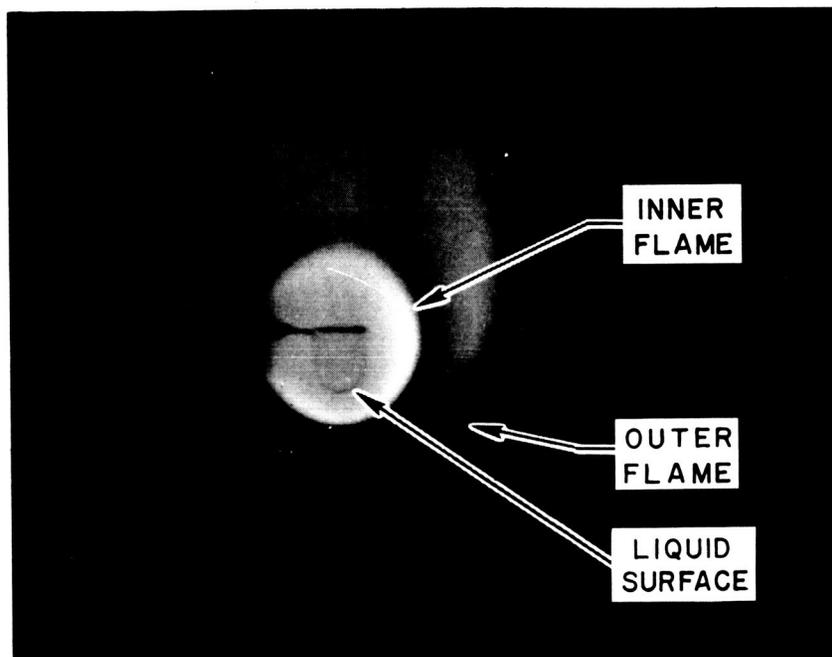


FIGURE 1. HYDRAZINE / NITROGEN TETROXIDE

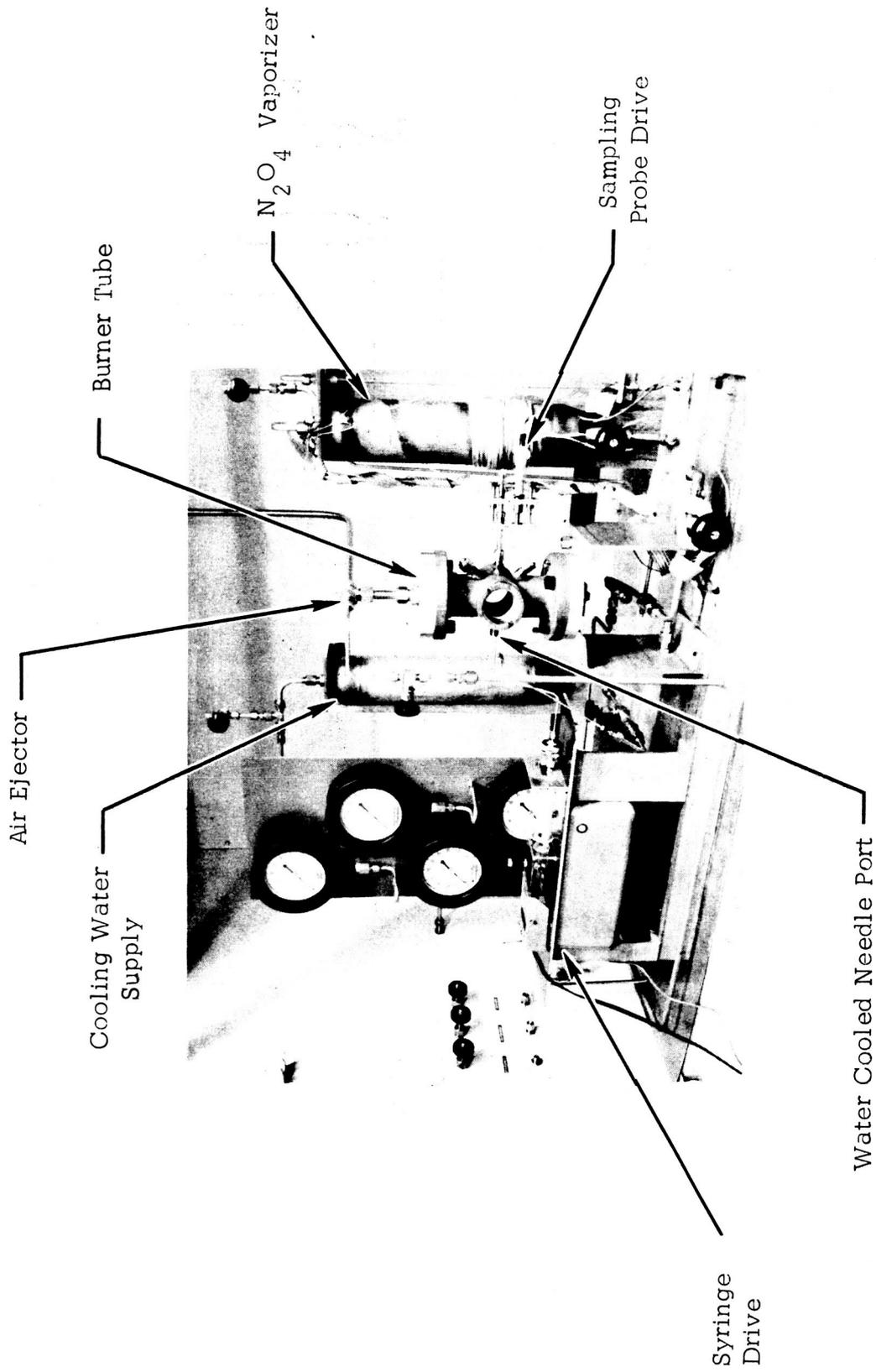


FIGURE 2. HYDRAZINE DROPLET BURNING SETUP.

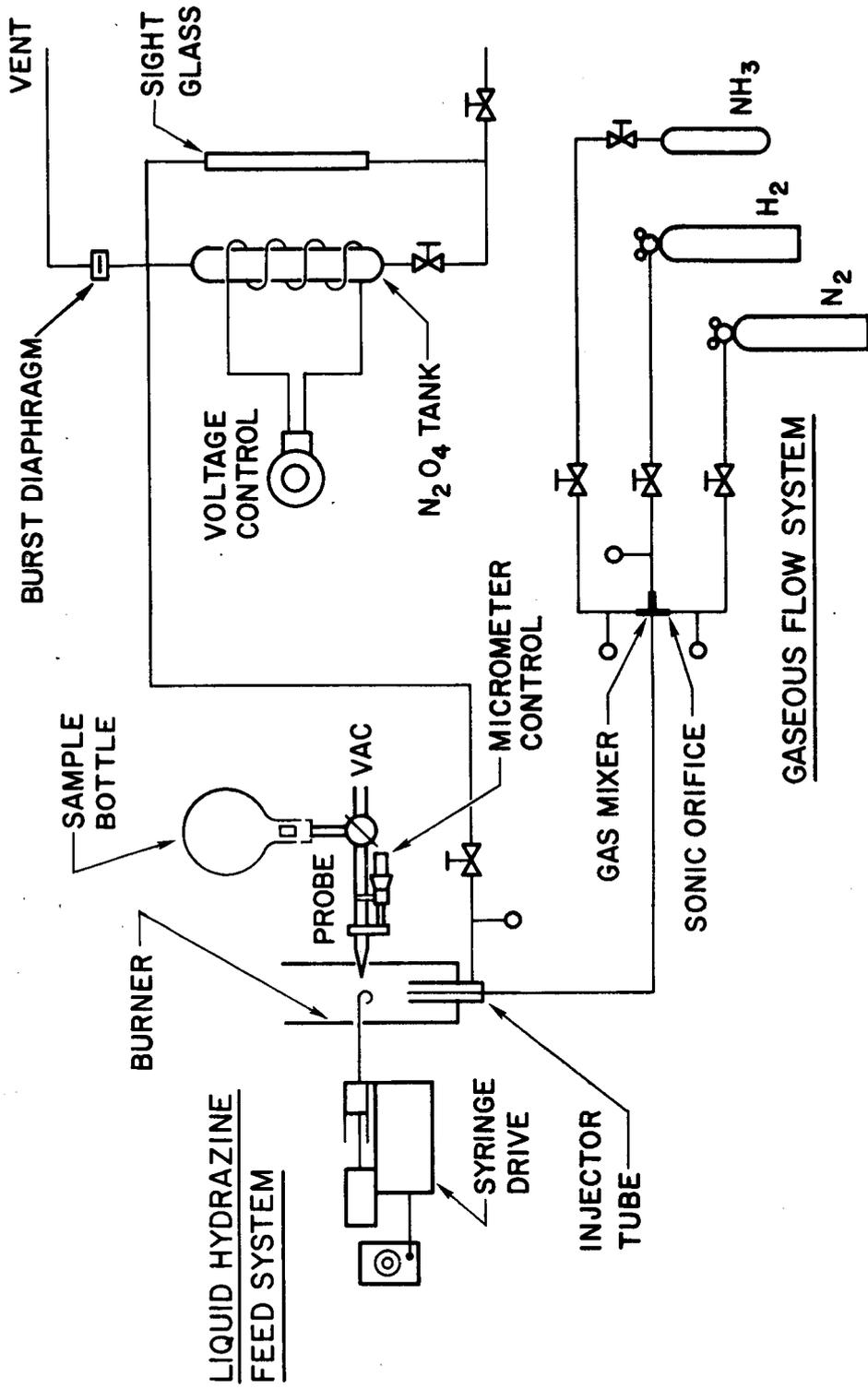


FIGURE 3. N_2H_4/N_2O_4 BURNER

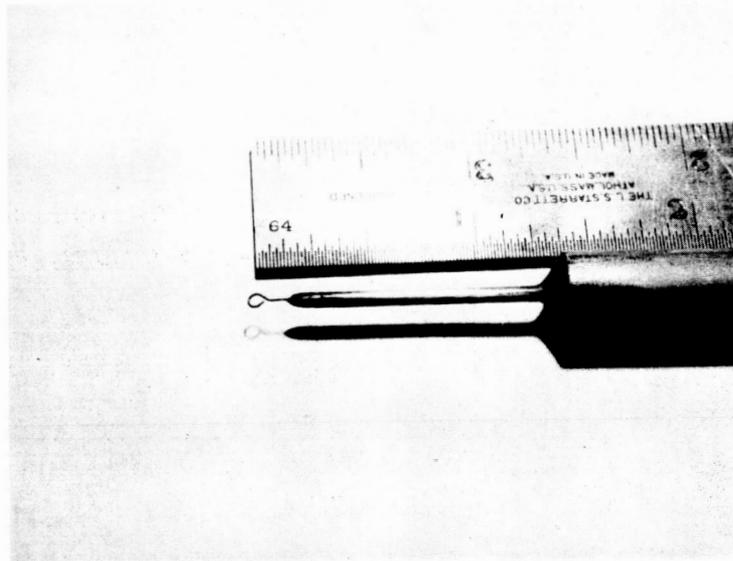
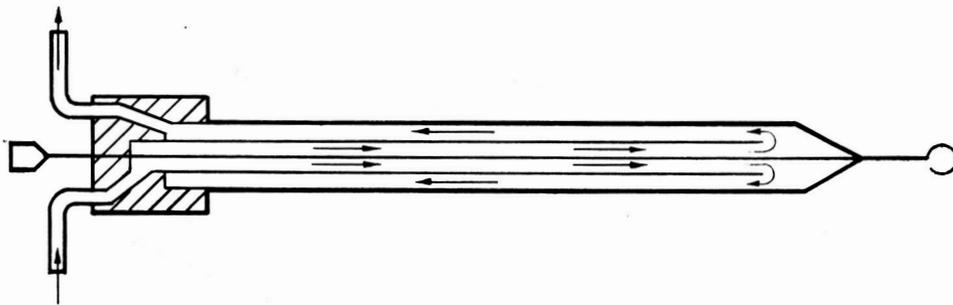
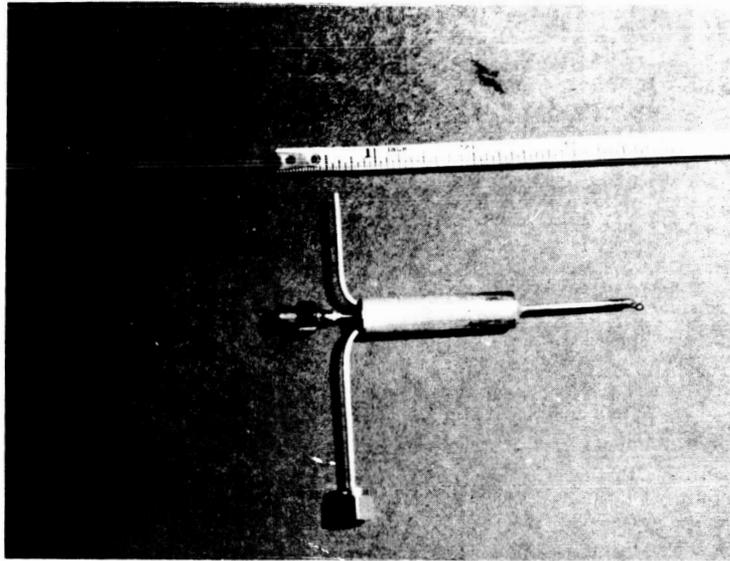


FIGURE 4. WATER COOLED NEEDLE

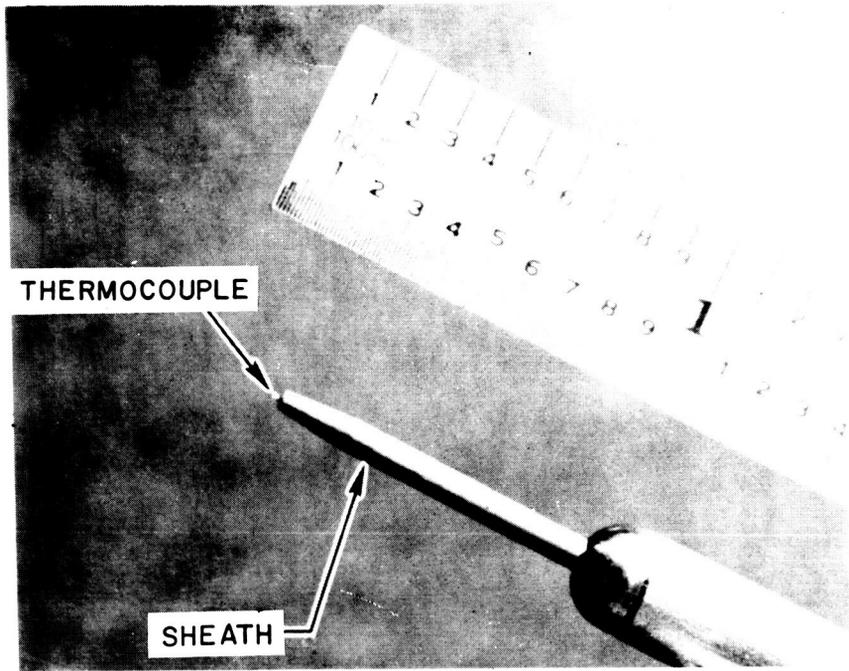


FIGURE 5. TEMPERATURE PROBE

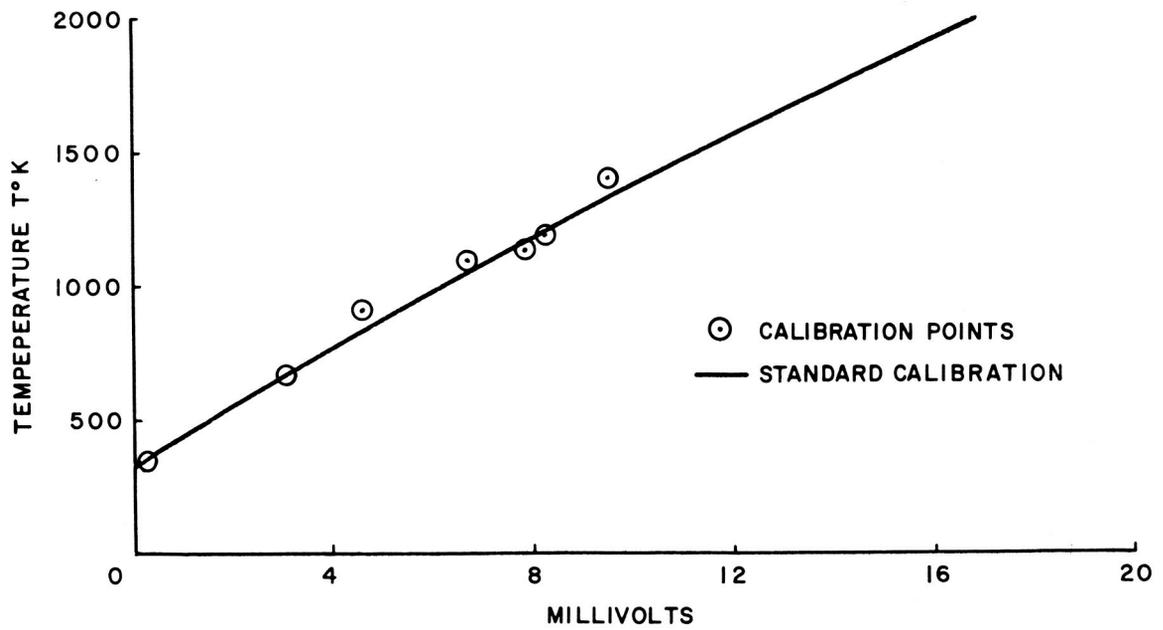


FIGURE 6. THERMOCOUPLE CALIBRATION

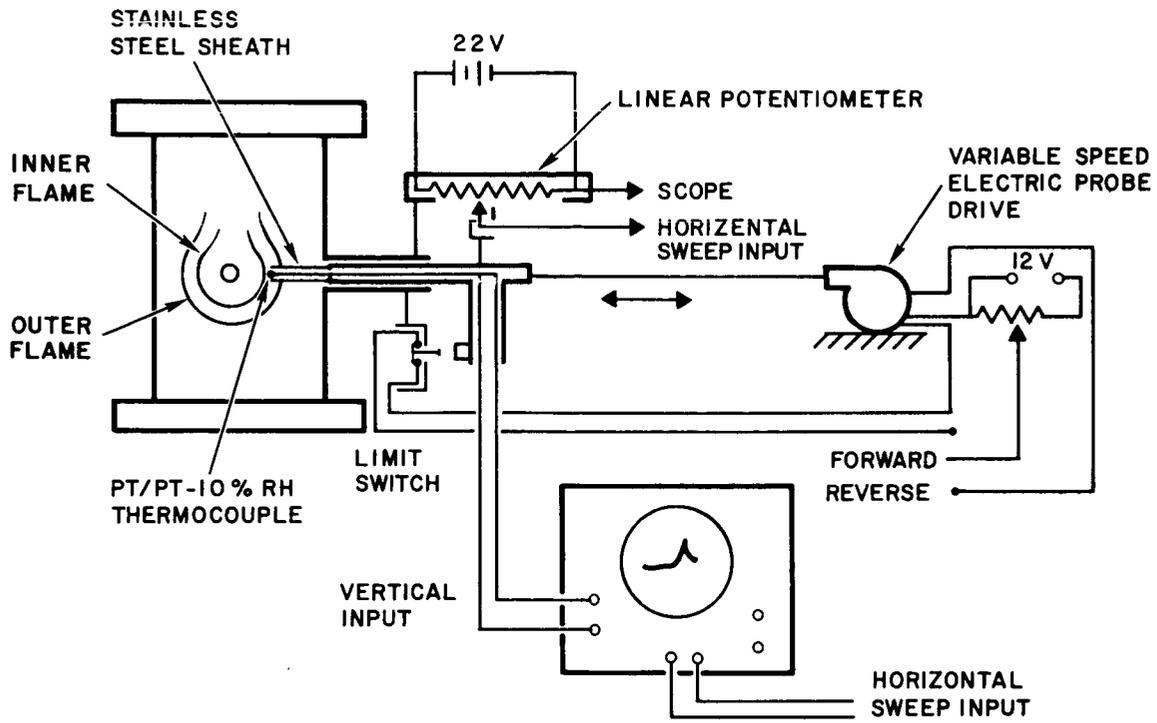


FIGURE 7. THERMOCOUPLE PROBE SETUP

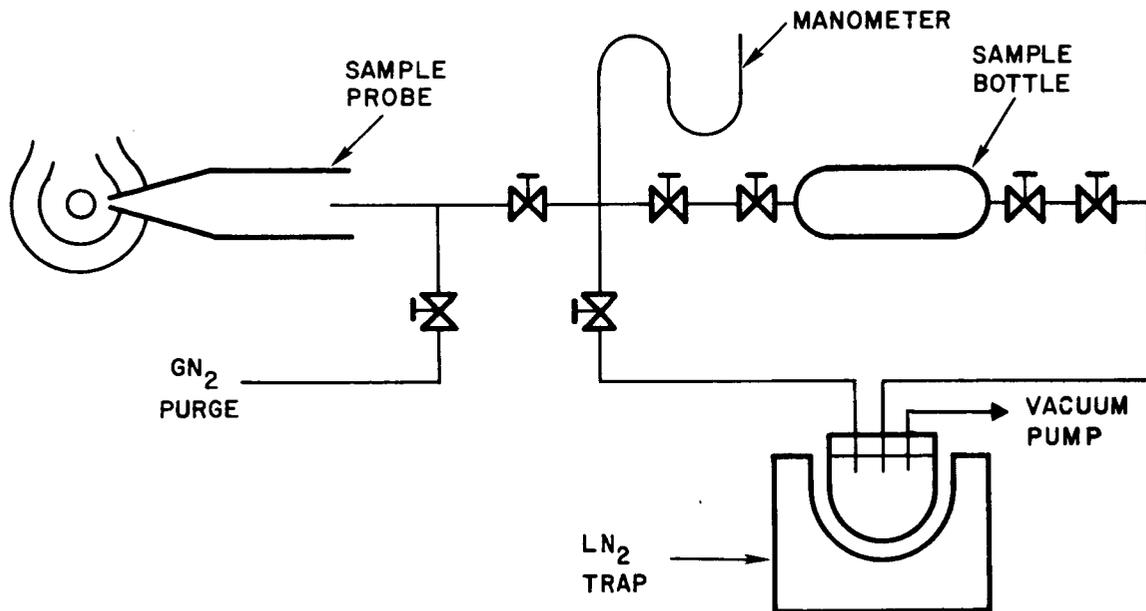


FIGURE 8. FLAME SAMPLING VACUUM SYSTEM

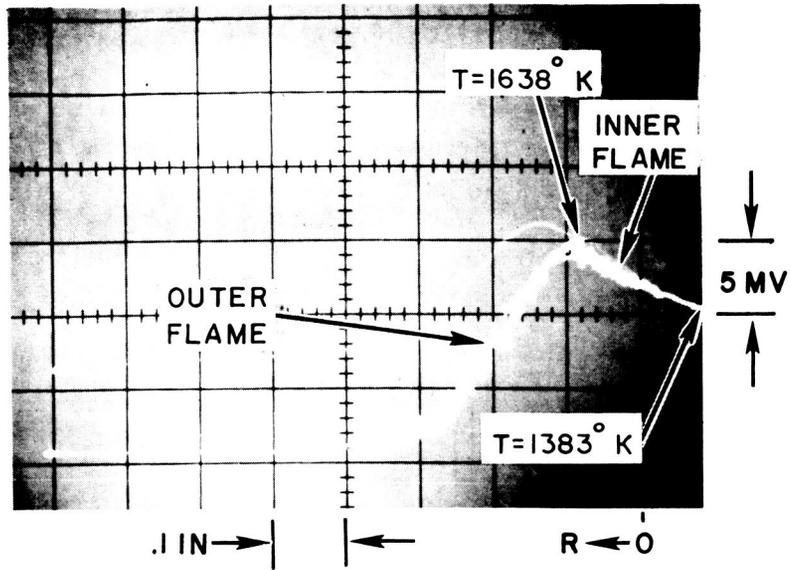


FIGURE 9. HYDRAZINE/NITROGEN TETROXIDE FLAME TEMPERATURE PROFILE

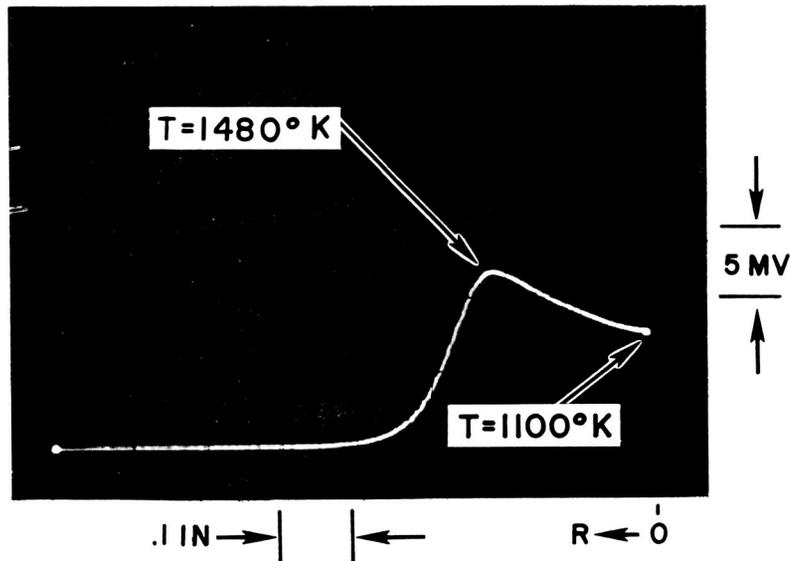


FIGURE 10. HYDRAZINE/AIR FLAME TEMPERATURE PROFILE

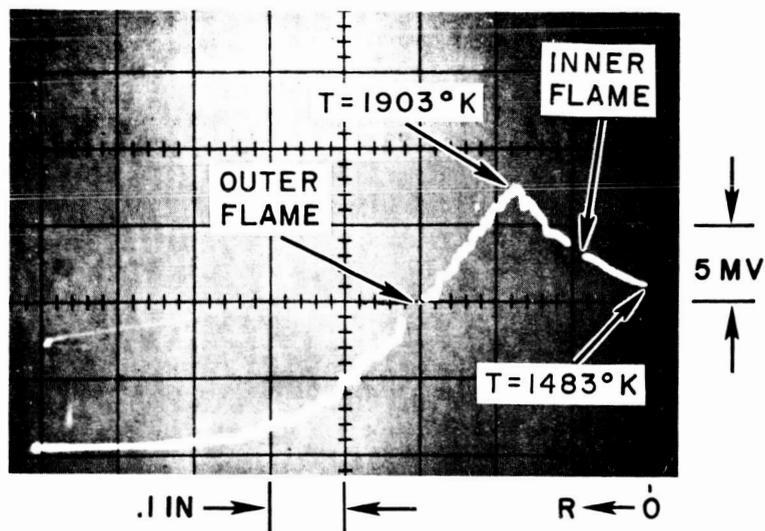


FIGURE 11a. HYDRAZINE - O_2/N_2 FLAME HIGH O_2/N_2 RATIO TEMPERATURE PROFILE

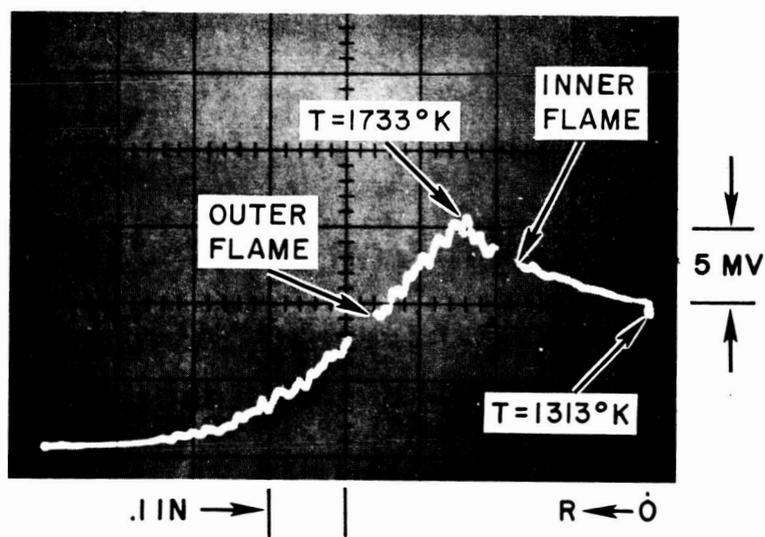


FIGURE 11b. HYDRAZINE - O_2/N_2 FLAME LOW O_2/N_2 RATIO TEMPERATURE PROFILE

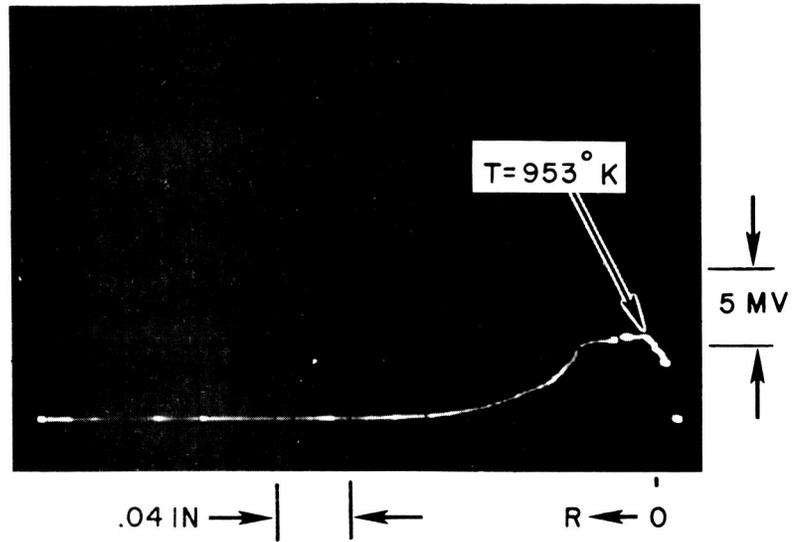


FIGURE 12. HYDRAZINE/LOW OXIDIZER CONCENTRATION TEMPERATURE PROFILE

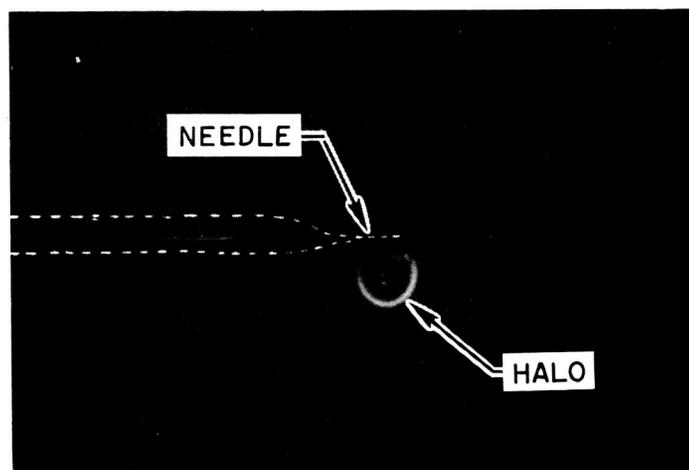


FIGURE 13. HYDRAZINE/LOW OXIDIZER CONCENTRATION HALO

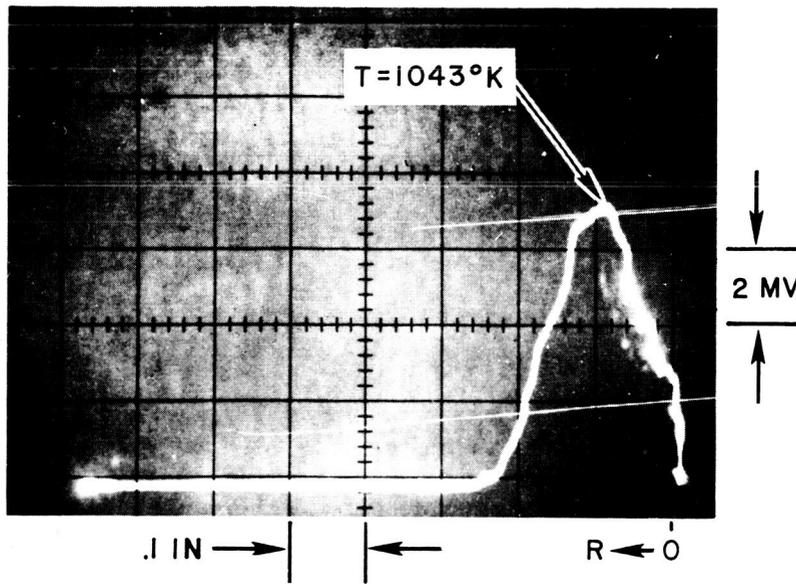


FIGURE 14a. TEMPERATURE PROFILE
HEPTANE/NTO FLAME



FIGURE 14b. DROPLET FLAME
HEPTANE/NTO FLAME

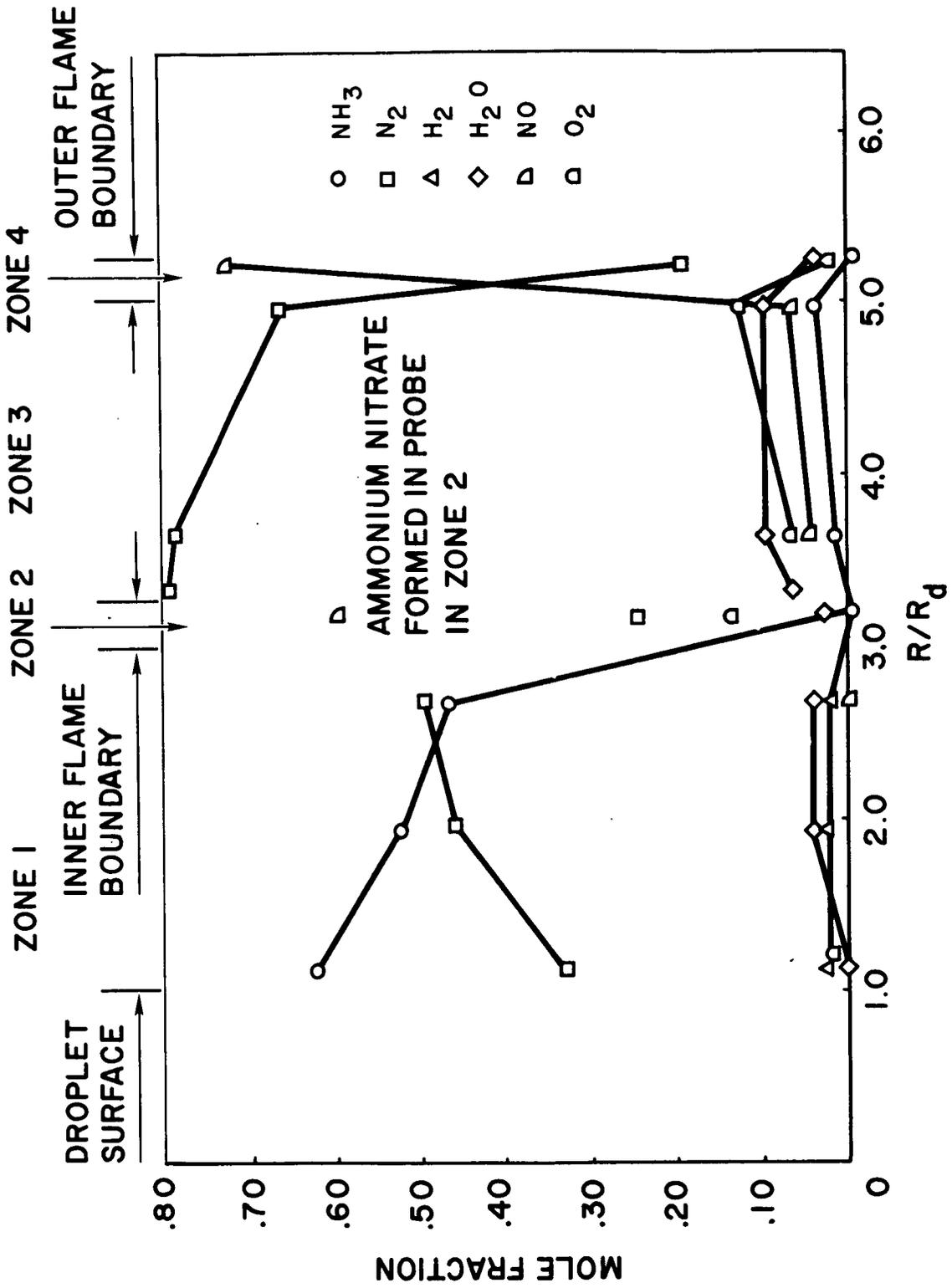


FIGURE 15. CONCENTRATION vs REDUCED RADIUS FOR N₂H₄/NO₂ FLAME

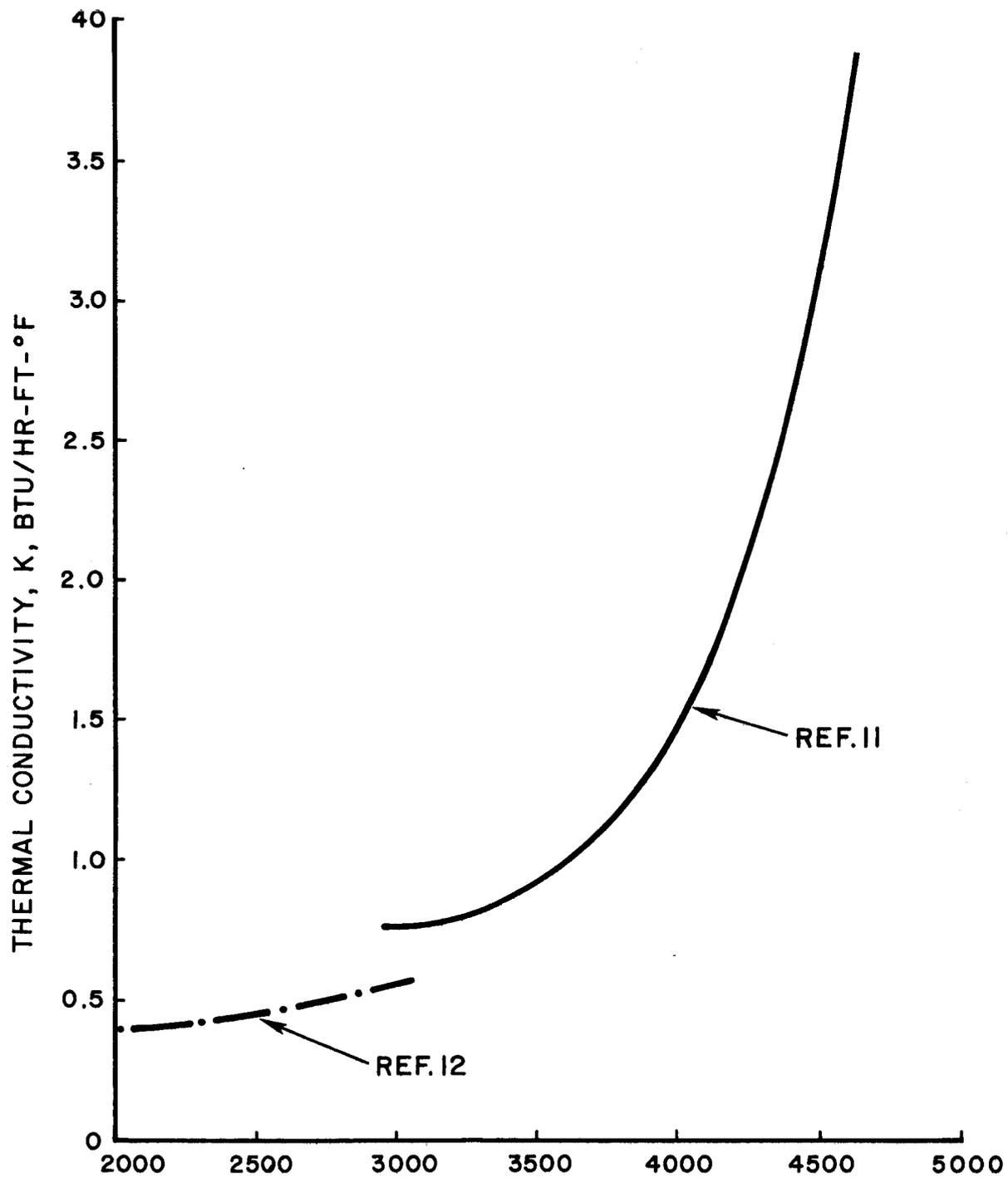


FIGURE 16. HYDROGEN THERMAL CONDUCTIVITY

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