

RADIATION AND BURNING VELOCITY STUDIES  
OF PROPANE-OXYGEN-NITROGEN MIXTURES

Thesis by

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## ABSTRACT

In several recent papers, the authors have assumed that the spectral intensity ratio of suitable emission lines (e.g.,  $C_2/CH$ ) from laminar flames was dependent only on the equivalence ratio for a particular fuel-oxidizer system. The objective of this study was to determine the effect of various amounts of inert diluent on the spectral intensity ratio of a propane-oxygen-nitrogen system. Experiments were conducted with inert diluent contents of 50, 60, 70, and 80 percent nitrogen (by mass) and with equivalence ratios of 0.6, 0.8, 1.0, 1.2, 1.4, and 1.6. The spectral intensity ratio was found to be a function of the percentage of inert diluent as well as a function of the equivalence ratio. Since the flame temperatures are dependent on the percentage of inert diluent, it appears likely that the spectral intensity ratio is a function of temperature.

The angle method was used to obtain burning velocity data. The relative changes of burning velocity with change of composition were deduced from the experimental data.

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## I. Introduction

In several recently published papers, the authors have used the ratios of apparent peak intensities of two rotational lines belonging to different emitters (e.g., CH and  $C_2$ ) as a measure of the mixture ratio in hydrocarbon flames. The procedure involves first the determination of the spectral intensity as a function of equivalence ratio (actual moles of fuel per mole of oxygen divided by the moles of fuel per mole of oxygen for the stoichiometric mixture ratio) using a known fuel-air input to a laminar flame. Then, using a calibration curve obtained in this manner for a given fuel-oxidizer system, the spectral intensity ratios are employed, in turn, to determine local equivalence ratios in turbulent flames, reverse-jet flames, etc.

Clark<sup>(1)</sup> found the intensity ratios of  $C_2$  to CH bands to be a good index of the equivalence ratios of flames for a given fuel. He measured intensity ratios in different regions of laminar and turbulent ethylene-air flames and found that the intensity ratios were constant in the laminar flames.

Clark and Bittker<sup>(2)</sup> found that the radiation per unit area of a laminar flame depends only on the equivalence ratio. They proposed to use this result for the measurement of burning velocities.

John and Summerfield<sup>(3)</sup> have shown that the relative changes of  $C_2$  and CH intensity for a turbulent flame (propane-air) enclosed in a duct are nearly equal, and hence turbulence did not alter appreciably the correlation of the spectral intensity ratio with equivalence ratio. In

several studies dealing with flame stabilization the spectral intensity ratio method has been used. (4, 5, 6)

Fuhs<sup>(6)</sup> has used the result that the apparent intensity (assuming that the population temperature and the effective temperature in the formula for the line radiancy are equal) from optically thin gases, observed with a spectroscope having a slit width large compared to the line width, is proportional to the ratio of the population of emitters in the excited states.

The radiation from  $C_2$  and CH in flames is probably not of thermal origin but may be the results of chemical reactions which produce molecules in excited states (chemiluminescence). In laminar flames the  $C_2$  and CH radiation occurs predominantly in the inner cone; it is unlikely that self-absorption affects the measurements of intensity for  $C_2$  and CH emitters. (7, 8)

In the work using the spectral intensity method discussed earlier, the investigators have stated or assumed, without convincing proof, that the spectral intensity ratio is a function only of the local equivalence ratio for a given fuel-oxidizer system. The prime objective of the studies undertaken by the author was to see whether this assumption is indeed correct. It was decided to determine the effect of various amounts of inert diluent on the spectral intensity ratio as a function of equivalence ratio. A propane-oxygen-nitrogen system was used. Chemically pure propane was selected because it had been used in some of the studies mentioned earlier in this discussion. By varying the amount of inert diluent, the adiabatic flame temperature is changed appreciably; hence

our results should provide some information concerning the effect of temperature on the spectral intensity method.

The laminar burning velocities for propane-oxygen-nitrogen mixtures were also measured in order to serve as an additional check for comparing our results with those of other investigators. The burning velocity is defined as the normal velocity of the flame front with respect to the initial combustible mixture. <sup>(9)</sup> The angle method and the area method are commonly used to determine the burning velocity of stationary laminar flames on cylindrical burners. The burning velocity, as determined by the area method, is equal to the volume flow rate of the mixture divided by the area of the inner cone surface of the flame. The area method gives a burning velocity which is averaged over the total flame surface. Since a considerable fraction of the total surface of the cone lies close to the burner rim, cooling undoubtedly lowers the burning velocity; on the other hand, the gas near the tip is heated from all sides of the cone, thus increasing the burning velocity in this region. <sup>(10)</sup>

The angle method consists of drawing tangents to the straight portion of the inner cone (the region where the tip and base effects are negligible). The laminar burning velocity is found by using the relation  $V_b = V_m \sin \alpha$ , <sup>(10)</sup> where  $V_b$  equals the burning velocity,  $V_m$  is the average velocity in the burner tube, and  $\alpha$  is the half angle of the inner cone (obtained from the tangent data). The accuracy of this method depends on how closely the following assumptions apply to the flame studied: <sup>(9)</sup> (1) The divergence of the gas stream from the axial direction

between the burner port and burning surface is negligible. (2) The nature of the flame reactions in the straight portion of the cone are nearly constant and are representative of the reaction under study. (3) The flow is laminar. (4) The straight portion of the cone includes the stream tube where the velocity is the average velocity passing through the burner (0.707 times the burner radius). (5) The surface of maximum temperature gradient is parallel to the region defined by the onset of luminosity.

Many variations of the above methods have been used. It is actually debatable as to which method (if either) is superior. Comprehensive surveys and extensive references to the published literature on this subject may be found in the books by Lewis and von Elbe,<sup>(11)</sup> Gaydon and Wolfhard,<sup>(12)</sup> and Jost<sup>(13)</sup> as well as in surveys by Poorman,<sup>(9)</sup> Linnett,<sup>(10)</sup> and Flock.<sup>(14)</sup>

In the studies discussed in this report the angle method was used because the apparatus needed was simple and inexpensive, and the results obtainable are satisfactory for showing the relative changes of burning velocities with changes of composition.

## II. Apparatus and Experimental Procedure

The apparatus which was assembled to obtain laminar flames of gas mixtures of known equivalence ratio and percentage inert diluent content is shown schematically in Figure 1. Also shown is the optical system used in obtaining information needed to determine laminar burning velocities. The gases were supplied to the system from standard commercial high-pressure tanks. Chemically pure propane, oil pumped nitrogen, and

extra dry grade oxygen were the gases used in all experiments. The gas pressure was reduced by a high-pressure regulator and an accurate low-pressure regulator connected in series in each line. Pressure gauges, sensitive to about (1/16) psig, were attached downstream of each low pressure regulator.

Fischer and Porter precision bore metering tubes with ball-type floats were used to meter the gases. These rotameters deliver within  $\pm 2\%$  of their calibrated flow, except near the limits of their respective operating ranges. The flow rate of each gas was controlled by a needle valve placed downstream of the rotameter in each line.

Each gas line was tapped into the upstream end of a mixing chamber, which consisted of a steel pipe about 2.5 inches in diameter and 24 inches long, capped at each end. The chamber was filled with small tubular glass beads to promote mixing and also to make the chamber an effective flashback arrester. It was concluded that good mixing was achieved since the observed flames were steady and reproducible. Copper tubing was used to connect the outlet from the downstream end of the mixing chamber to the burner.

The burners used for the studies were hard copper tubing with inside diameters of 1.012, 0.704, and 0.386 centimeters. The burners of various inside diameters were needed to achieve laminar flow and reasonable flow rates, and to keep the quenching effect of the burner to a minimum for the large number of mixtures of gases used during the studies.

An objective lense system from a motion picture camera was used

to magnify and project the image of the inner cone of the flame onto a ground glass screen. A plane mirror was used in the system to place the image on a horizontal surface for ease in tracing the outline of the inner cone image. A magnification of about 4:1 was used. The system was aligned and focused by placing an illuminated square grid network in the plane of the flame cone. Then the system was adjusted to obtain a clear magnified image of identical vertical and horizontal proportions as the grid network on the ground glass screen. Originally a double-convex lens was used in the system, but pincushion distortion was found to be severe. With the final system it is estimated that errors in optical alignment could not contribute an error of more than 1% to the measured value of the burning velocity.

Figure 2 shows schematically the experimental equipment that was used to obtain the data needed to compute the spectral intensity ratios. A Jarrell Ash spectroscope with an f number of 6.5 and a linear dispersion of  $18 \text{ \AA}/\text{mm}$  was used. An RCA 1P28 photomultiplier tube (glass) located at the exit slit of the spectroscope served as a detector. The spectra were recorded by a Leeds and Northrup Speedomax Recorder. The voltage input to the photomultiplier tube was maintained constant and relatively low throughout all of the experiments. By using this method, a lower noise level was achieved and, furthermore, the output current of the photomultiplier was kept low, thus helping to avoid the difficulties associated with fatigue of the photomultiplier tube. <sup>(15)</sup> The output of the photomultiplier tube was amplified and then fed to the recorder unit. Various amplifications were found to change a given spectral intensity

ratio by less than 2%; this discrepancy was smaller than the lack of reproducibility (5%) observed when identical tests were conducted on different days.

An image of the inner cone was formed on the slit of the spectro- scope by suitable adjustment of the condensing lense. The slit was masked to prevent radiation from the tip and base of the inner cone from entering the spectroscope. Also the slit was located so that radiation from the edge (or leg) of the cone did not enter the spectroscope, because it was found the spectral intensity ratio from the edge increased, re- mained the same, or decreased, depending on the specific fuel-oxygen- nitrogen mixture. This effect may be understood in terms of the relative positions of the radicals in the reaction zone. (16, 17, 18) Spectral intensity ratios determined from any part of the straight portion of the inner cone (excluding the cone edge) were the same for identical fuel- oxygen-nitrogen input to the flame. In all tests the axis of the inner cone image was placed on the slit and a slit width of several hundred microns was used. The spectral intensity ratio was found to be independent of the monochromator scanning speed, for all practical purposes. A scanning rate of 50 Å/min was used to get sufficient line detail and still not make the time required to get the desired spectral regions pro- hibitively long.

The following rotational lines were selected for the computation of spectral intensity ratios:

<u>Wavelength, Å</u>	<u>Transitions</u>	<u>Emitter</u>
5165	$^3\Pi \rightarrow ^3\Pi(0, 0)$	C <sub>2</sub>
4737	$^3\Pi \rightarrow ^3\Pi(1, 0)$	C <sub>2</sub>
4324	$^2\Delta \rightarrow ^2\Pi(0, 0)$	CH
4312	$^2\Delta \rightarrow ^2\Pi(0, 0)$	CH

These lines were selected because they are well isolated; furthermore, the spectral intensity ratios of several combinations of lines could be compared. For a given equivalence ratio, the spectral intensity ratios based on the stronger lines (5165 Å, 4312 Å) were reproducible to within about 5 percent when using the same rotameters.

All tests were conducted in a darkened room, and a flat black screen was used behind the flame. These precautions were taken to reduce scattered light and radiation reflection, stop air movement from disturbing the flame, and make the image of the flame on the ground glass screen distinct. The temperature of the room was at all times between 71°F and 75°F while the experiments were in progress.

The procedure followed in the tests involved first the calculation of the necessary flow rates for each gas to obtain a gas mixture of a desired equivalence ratio and a desired percentage of nitrogen (by mass). It was very trying to obtain a total flow rate of a desired gas mixture such that the flame neither blew off nor flashed back, and had a suitable inner cone. We considered the inner cone suitable when the straight portion of the cone was large as compared to the base and tip sections (this was usually a height/radius ratio of greater than 5 or 6). Flames from gas mixtures

of low equivalence ratios and large percentage of nitrogen were very sensitive to blow-off and flashback (a very narrow range of flows could be used), while flames from rich gas mixtures containing large amounts of nitrogen are not very steady (air currents caused slight movements). When a suitable flame was obtained, the radiation entering the slit was scanned with the spectroscope. A wavelength interval on each side of a desired line was taken so that one could determine the magnitude of background continuum (see Figure 3). The background radiation was always subtracted in measurements of line intensities, thus insuring that only radiation from the desired emitters was used in the calculation of the intensity ratio. This scanning operation was repeated so that one could determine the average height of the lines and determine the amount of variation. Usually the variation was less than 2% of the total line height.

Tangents to the image of the inner cone of the flame (see Figure 4 for a typical image) were drawn on tracing paper. These tangents were drawn parallel to the upstream side of the bright image. Consecutive measurements of the angle between the tangents for the same flame showed a reproducibility of about five percent. This variation would affect the burning velocity reproducibility by the same amount. Attempts to reproduce the burning velocity data on different days resulted in variations as large as 10% in some cases.

### III. Experimental Results and Discussion

The results of the experimental studies are presented in Figures 5 through 12. In Figures 5 through 11 the spectral intensity ratio (the intensity of a  $C_2$  line divided by the intensity of a CH line) is plotted against

the equivalence ratio of the mixture (the actual fuel-oxygen ratio divided by the stoichiometric fuel-oxygen ratio). Curves have been drawn through the points obtained from mixtures containing the same percentage of nitrogen (by mass). Figures 5 through 8 are believed to be more accurate than Figures 9, 10, and 11 since in the former the lines (5165 Å, 4312 Å) used to determine the spectral intensity ratio are stronger and, therefore, the signal to noise levels were much larger. It was found experimentally that the intensity ratios of the 5165 Å to the 4312 Å lines were reproducible to within about 5 percent on different days, even with different total flow rates, as long as the same diameter burner and the same combination of flowmeters were used. If different combinations of flowmeters were used, the reproducibility was not as good in general. This fact emphasizes the need to use flowmeters with the proper flow range. In this work the same combination of flowmeters usually proved satisfactory for obtaining all the needed information for mixtures of a fixed percentage nitrogen content and for a certain burner tube. The reproducibility of the ratios obtained from the weaker lines was found to be poorer.

Figures 5, 6, 9, and 10 show that the maximum intensity ratio occurs at a larger equivalence ratio as one decreases the percentage nitrogen content in the gas mixture. Also the maximum intensity ratio of the stronger lines tends to be larger with decreasing inert diluent content while, to a lesser extent, the opposite is observed to be the case for the maximum intensity ratio based on the weaker lines. The curves seem to become flatter with increasing diluent content. Thus it is obvious that,

in general, the intensity ratio is a more sensitive function of the equivalence ratio for mixtures containing smaller amounts of nitrogen.

Smaller diameter burner tubes were needed to prevent the flow from becoming turbulent as mixtures containing lower percentages of nitrogen (faster burning) were used. Figures 7 and 11 show the changes in spectral intensity ratio with different burner tubes. The curves for the 0.386 cm burner (60% nitrogen) are somewhat higher than the curves for the 0.704 cm burner (60% nitrogen). In fact, the curves of 60% nitrogen for the 0.386 cm burner are quite similar to the curves of 70% nitrogen for the 0.704 cm burner. These changes of intensity ratios with change of burners were probably due to increased quenching with the smaller tube.

The curves on Figure 8 show that quenching had very little effect when the 0.704 cm tube and 1.012 cm tube were used. The relatively large scatter of the points for the curve obtained with a 1.012 cm tube was due to changes of the initial mixture temperatures of about 11 to 14°F. The larger spectral intensity ratios were found on days when the room temperature was approximately 82 to 85°F, instead of the normal 71 to 75°F room temperature.

Wohl and Welty<sup>(19)</sup> have studied radiation intensity as a function of position in reaction zones for butane-air flames. They found that the decay of C<sub>2</sub>-radiation in a lean flame was very rapid as compared to CH-radiation decay. As they used progressively richer mixtures, the decay rate of C<sub>2</sub>-radiation began to approach that of CH radiation. On the basis of these results, they concluded that the reaction between C<sub>2</sub>

and  $O_2$  was especially fast. Possibly the shift of the maximums of the curves in Figures 5, 6, 9, and 10 to richer mixtures with decreasing percentages of nitrogen in our results was partially due to the more intimate contact of the  $C_2$  with  $O_2$ , thus promoting fast removal of  $C_2$  from the reaction zone for mixtures of lower equivalence ratios.

Thomas<sup>(20)</sup> measured the effect of preheating on the  $C_2$  and CH emission from ethylene-air flames of fixed equivalence ratio. His results showed that the ratios of  $C_2$  to CH intensities were functions of the initial mixture temperature. Since preheating and inert diluent content both change the final flame temperature, it is believed that the changes of our curves with amount of inert diluent are largely determined by changes in temperature.

Clark<sup>(1, 2)</sup> found curves of spectral intensity ratio as a function of equivalence ratio for propane-air flames that are very similar to the curves we found for mixtures containing 70% nitrogen (by mass). Clark stated that leveling off at equivalence ratios of approximately 1.3 might be due to the surrounding air mixing with the richer flames, thus causing the actual equivalence ratios to remain near 1.3. The curves on Figures 5 and 9 tend to prove the above observation to be false, as the curves for 75% and 80% nitrogen leveled off at equivalence ratios where there was a surplus of oxygen.

Clark<sup>(1)</sup> also found that the intensity ratios were constant in different regions of laminar flames. We found changes at the tip, the base, and at the edges of the cone during preliminary tests. We believe that Clark did not find changes because he used a much larger slit width

than we did. We minimized this change with position from our results by always looking at a portion of the central axis not containing parts of the flame tip or flame base of the inner cone.

In Figure 12 the laminar burning velocity (in cm/sec) is plotted as a function of the percentage of inert diluent (nitrogen) by mass. The large scatter of the points obtained from richer mixtures was primarily due to the slight unsteadiness of very rich flames, which made the procedure of drawing tangents to the inner cone inaccurate. Burning velocity data were not taken for lean mixture flames, since in some cases the flame tended to lift off the burner slightly and the inner cone became flatter. When this happened, the diameter of the base of the cone was larger than the burner diameter, and the angle method was no longer useful. It was impossible to get stable flames from any mixtures containing 90% nitrogen. The burning velocities of mixtures containing 80% nitrogen were the same for all practical purposes. Hence, it is believed that the mixtures containing 80% nitrogen are very near the flammability limit.

The use of the angle method for determining burning velocities of relatively slow burning hydrocarbon flames probably fails to satisfy the requirement that the divergence of the gases from the axial direction between the port and the burning surface be negligible. This and the above mentioned difficulties seem to indicate that it is very difficult to obtain accurate burning velocities of slow burning hydrocarbon flames over a wide range of equivalence ratios by either the angle method or the total area method. We can conclude that the method used was inaccurate

for absolute determination of burning velocities, but was satisfactory for showing the relative change of burning velocities with composition.

#### IV. Conclusions

The spectral intensity ratios of  $C_2$  and CH lines from laminar flames were found to be dependent on the percentage of inert diluent and the equivalence ratio of the gas mixtures. The chemical reaction rates producing CH and  $C_2$  are known to be functions of the amount of inert diluent. Our results show that the spectral intensity ratios of CH to  $C_2$  are not unique functions of the mixture ratio but rather of the chemiluminescent processes producing the emitting species. Hence the use of the spectral intensity ratio method for measuring local equivalence ratios in turbulent flames, reverse-jet flames, etc. is questionable.

The burning velocities of nearly stoichiometric mixtures were found to be a more sensitive function of the amount of inert diluent than for rich mixtures.

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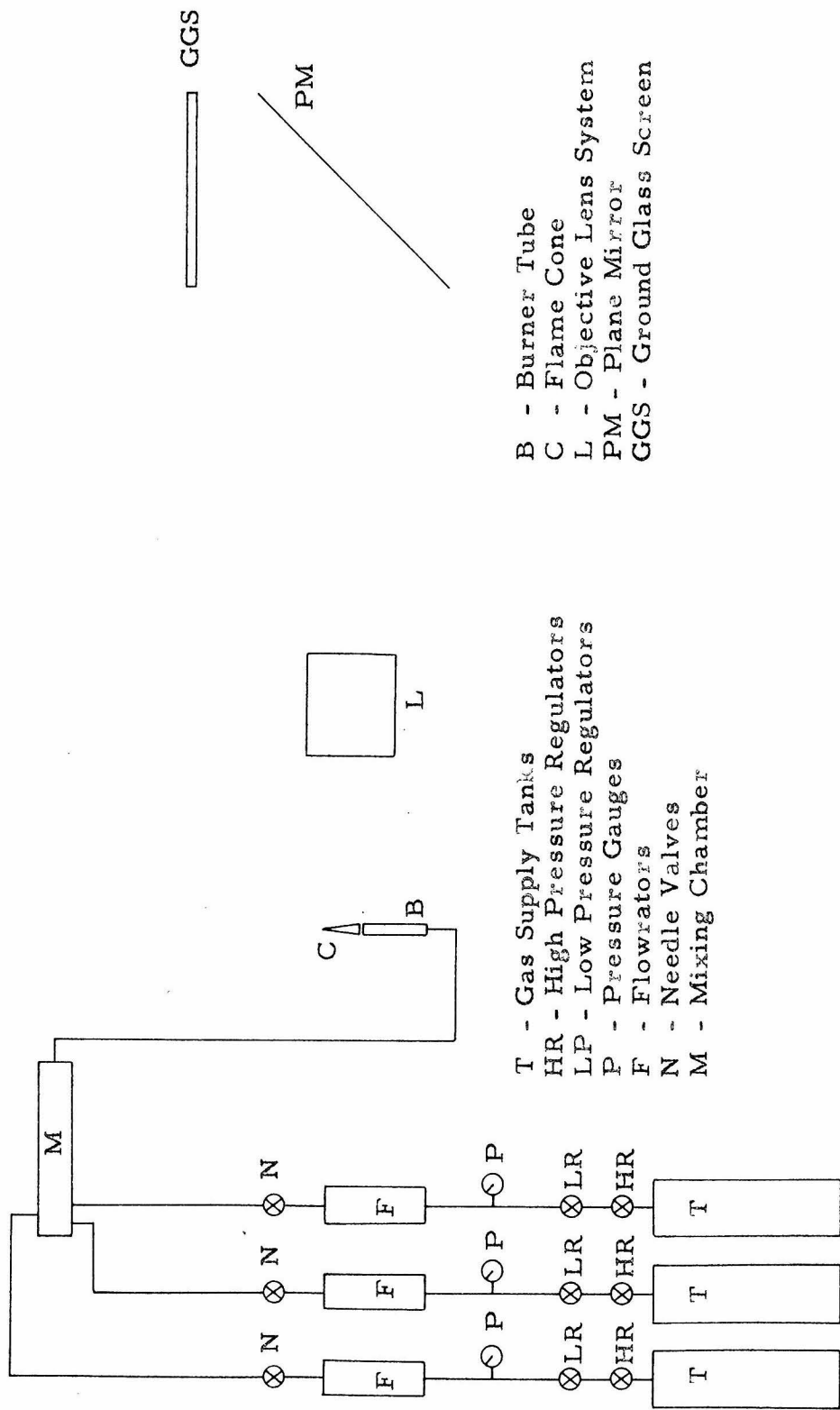


Figure 1. Schematic Diagram of the Apparatus Used to Obtain Desired Gas Mixtures and Laminar Burning Velocity Data.

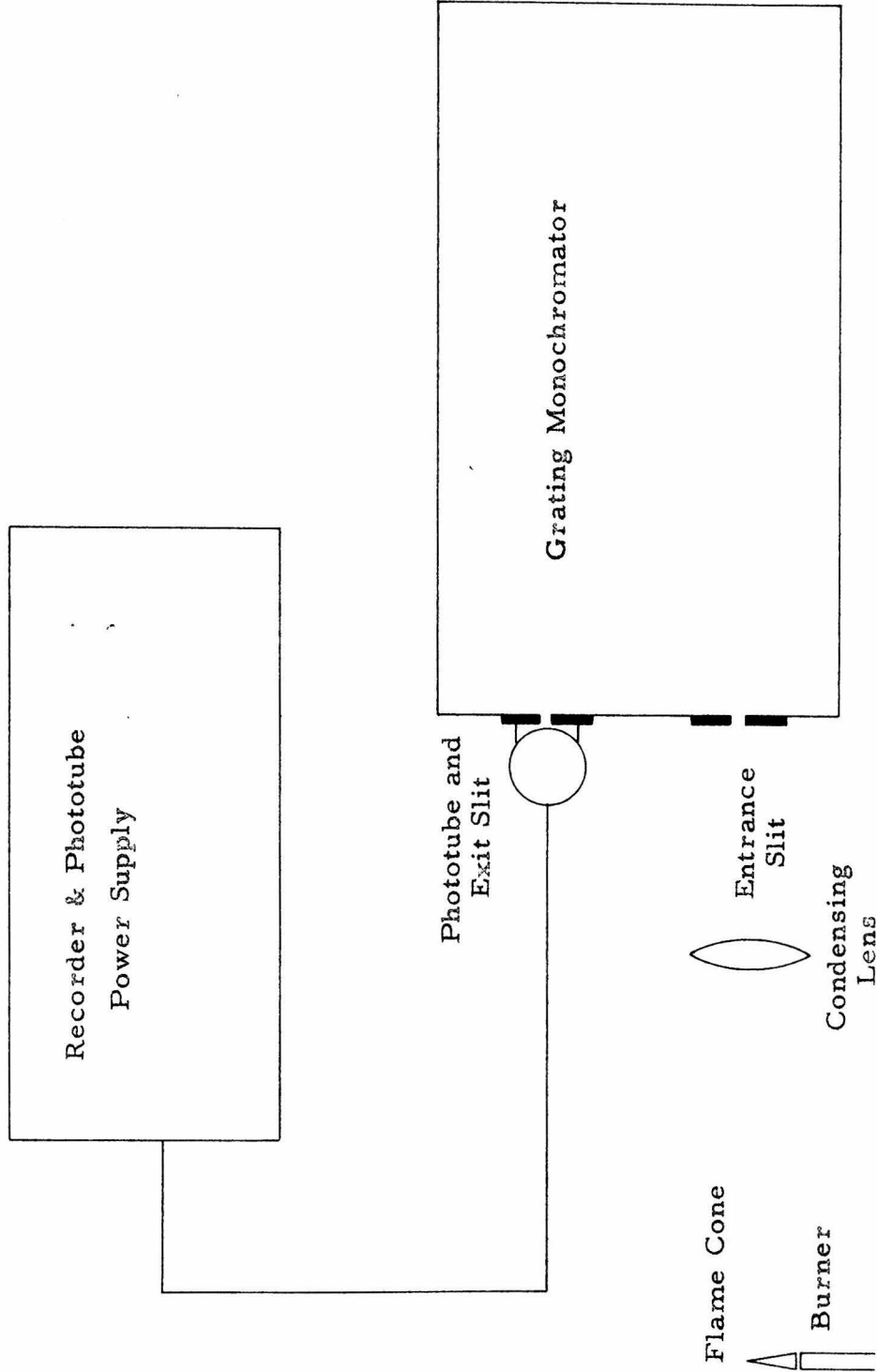


Figure 2. Schematic Diagram of the Apparatus Used to Obtain Data for Computation of the Spectral Intensity Ratios.

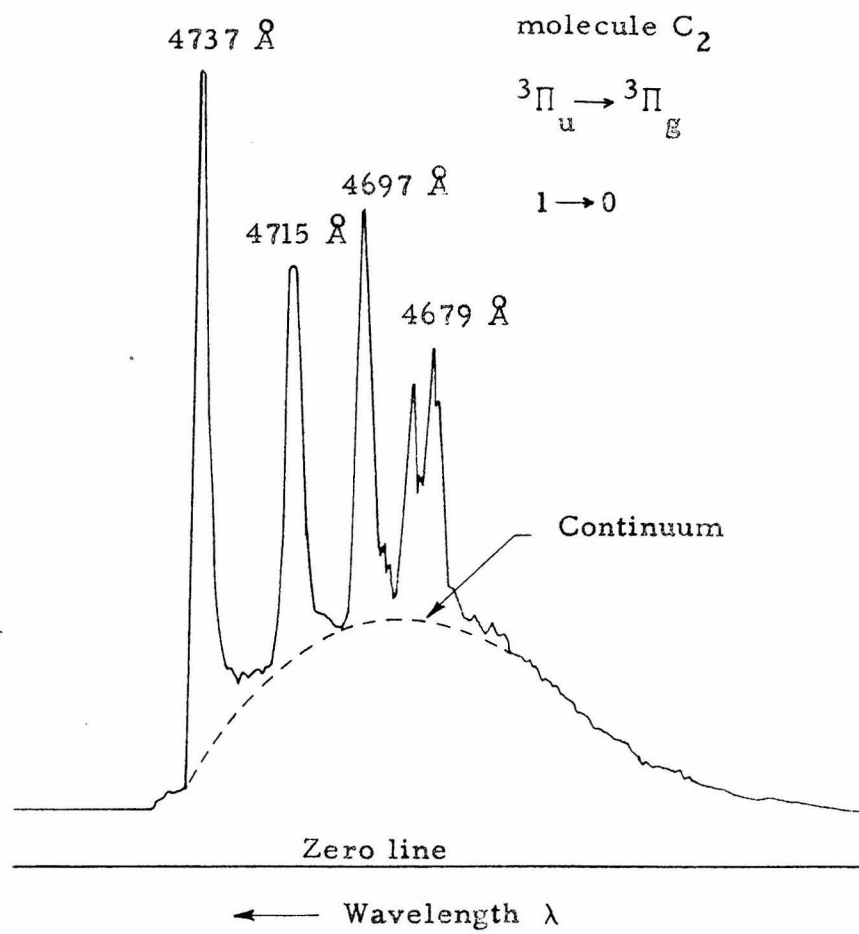


Figure 3. Apparent Emission Intensity as a Function of Wavelength.

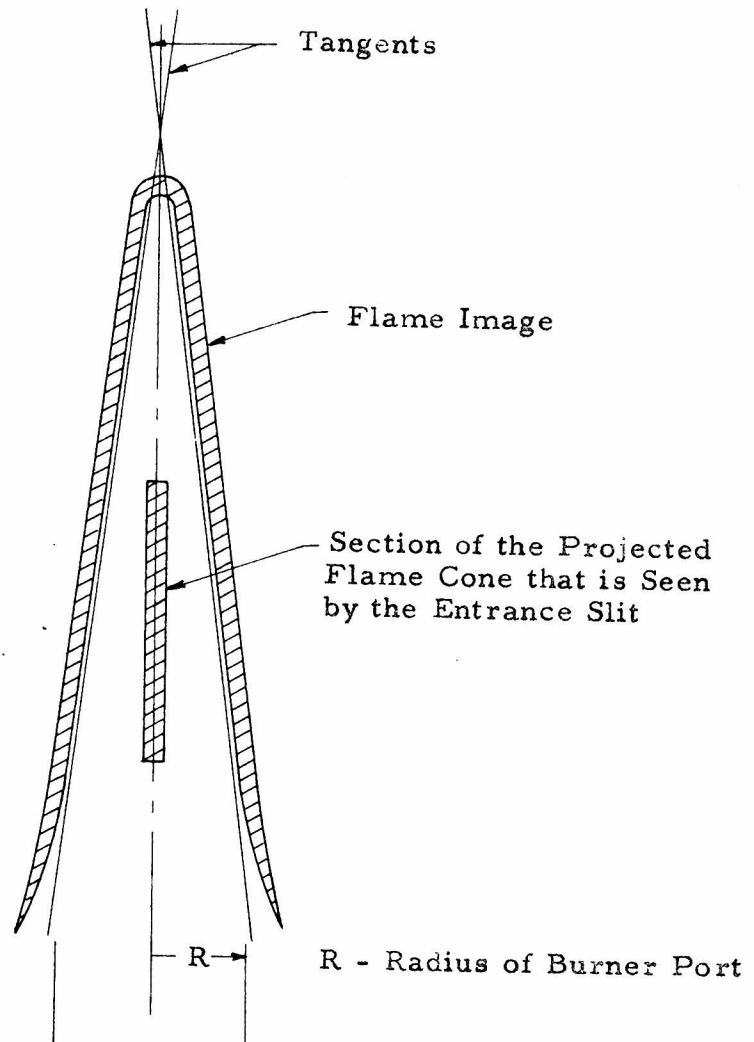


Figure 4. Diagram of a Typical Inner Cone Image

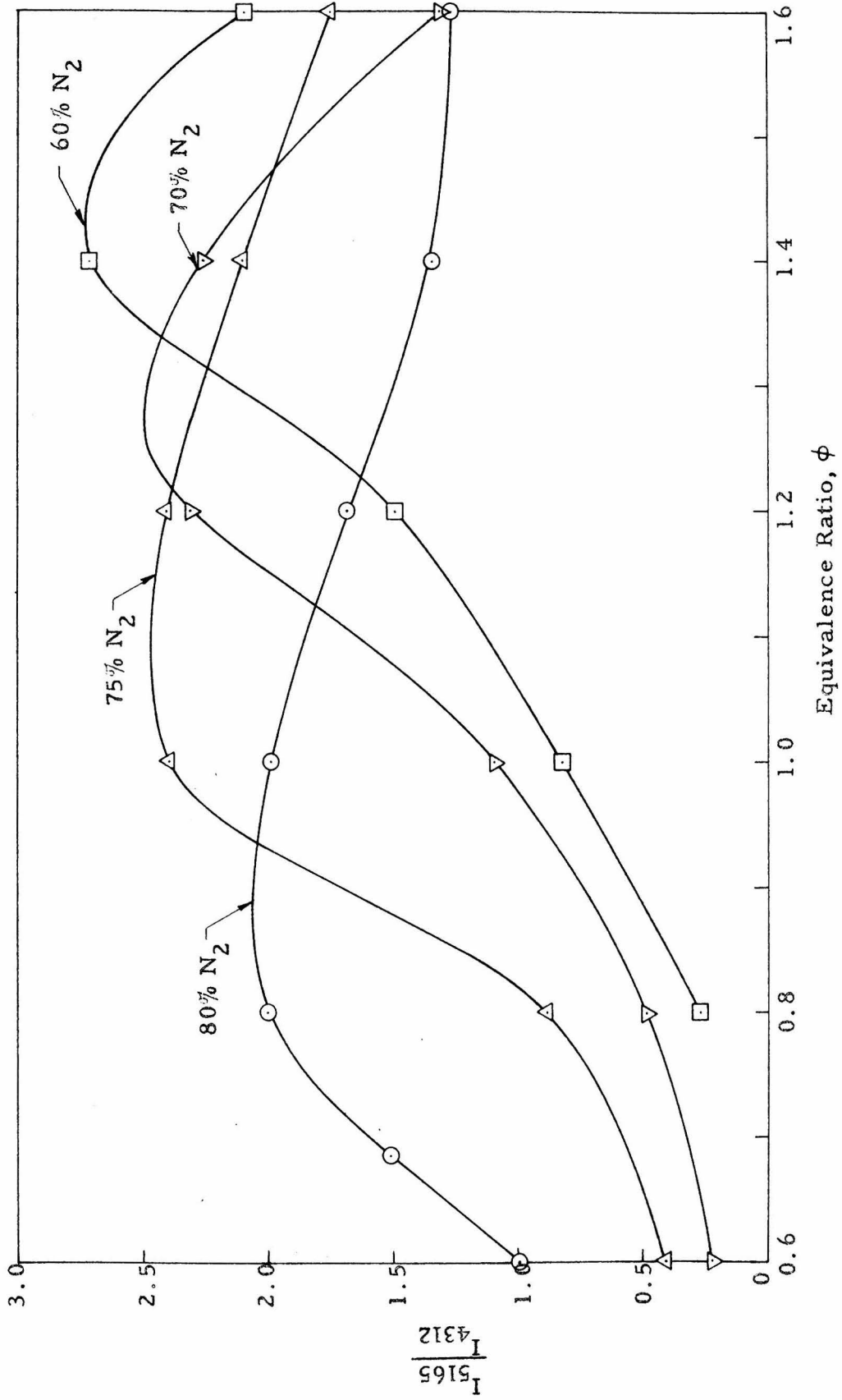


Figure 5. Spectral Intensity Ratio ( $I_{5165}/I_{4312}$ ) as a Function of Equivalence Ratio for Various Nitrogen Contents (by Mass). 0.704 cm I.D. Burner.

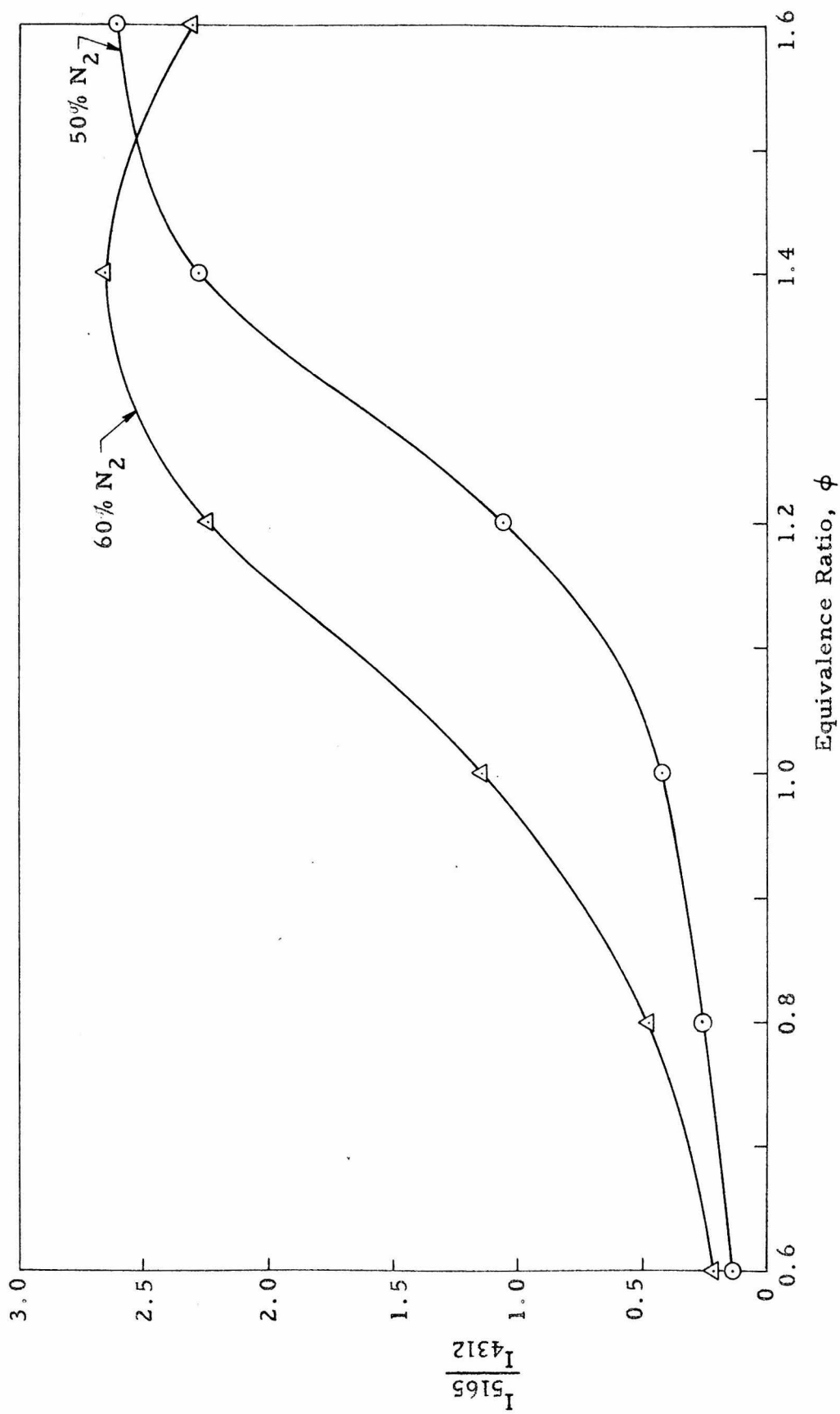


Figure 6. Spectral Intensity Ratio ( $I_{5165}/I_{4312}$ ) as a Function of Equivalence Ratio for Various Nitrogen Contents (by Mass). 0.386 cm I.D. Burner.

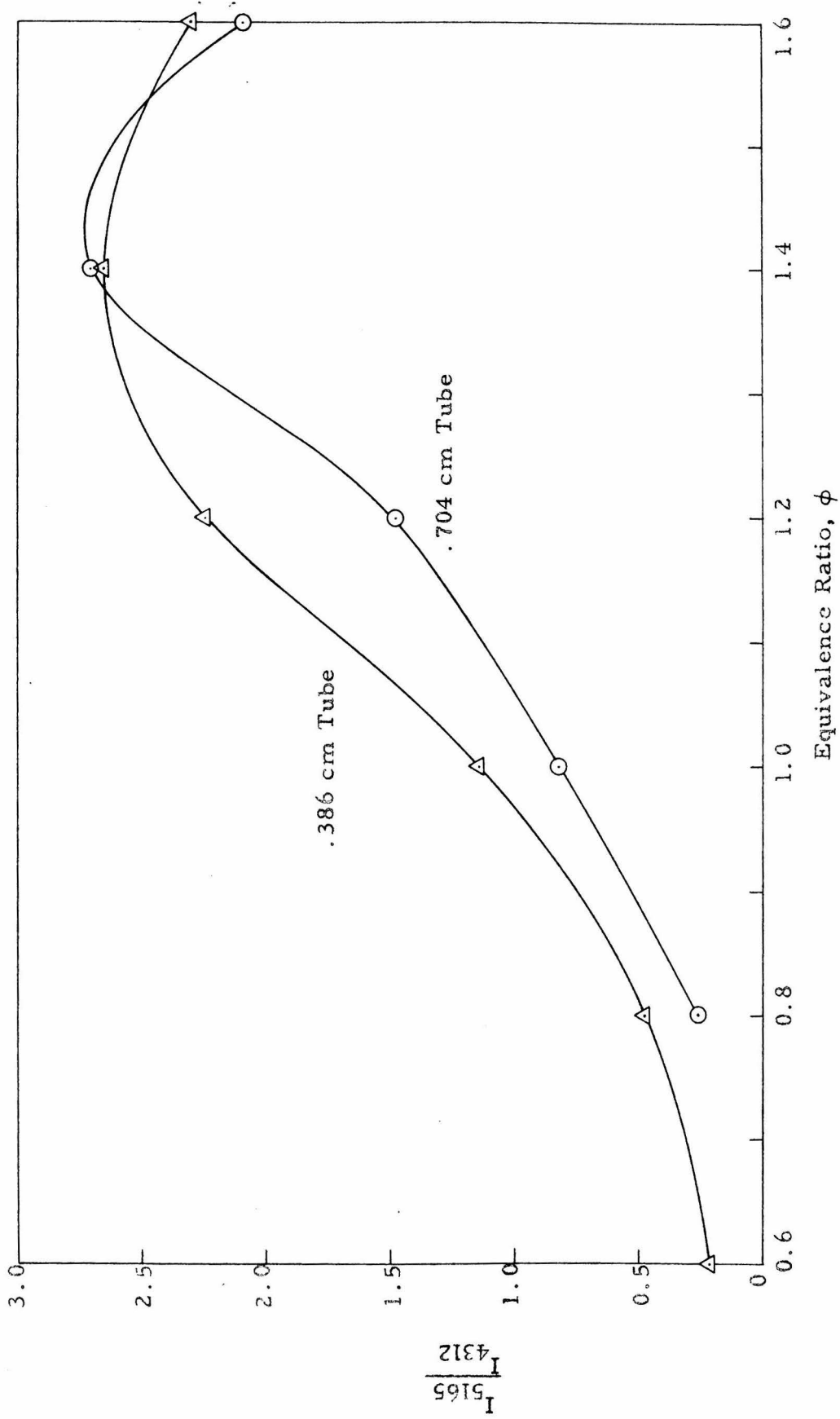


Figure 7. Spectral Intensity Ratio ( $I_{5165}/I_{4312}$ ) as a Function of Equivalence Ratio for 0.386 cm and 0.704 cm I.D. Burners. Nitrogen Content in Mixtures: 60% by Mass.

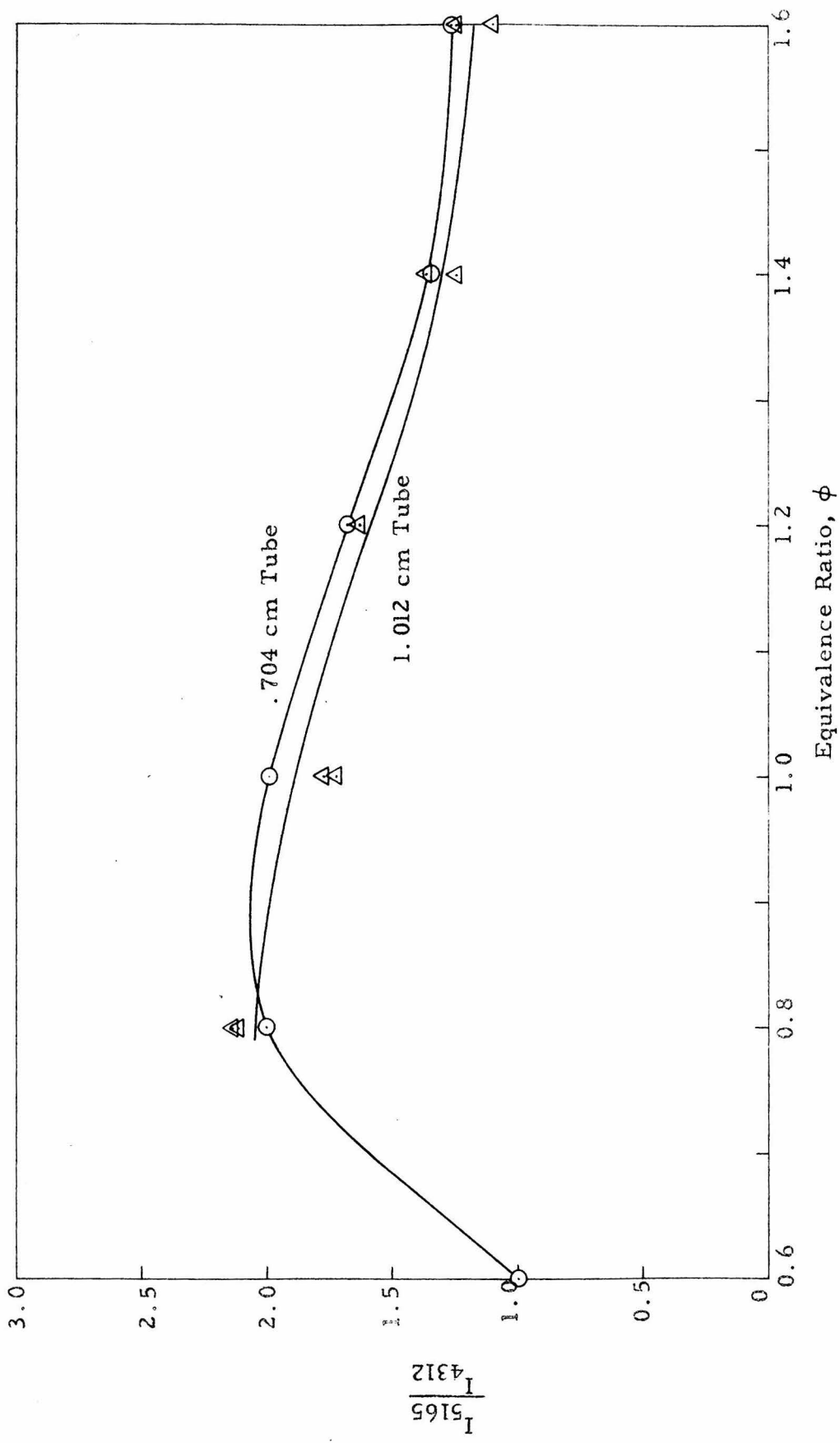


Figure 8. Spectral Intensity Ratio ( $I_{5165}/I_{4312}$ ) as a Function of Equivalence Ratio for 0.704 cm I.D. Burner and 1.012 cm I.D. Burner. Nitrogen Content in Mixtures: 80% by Mass.

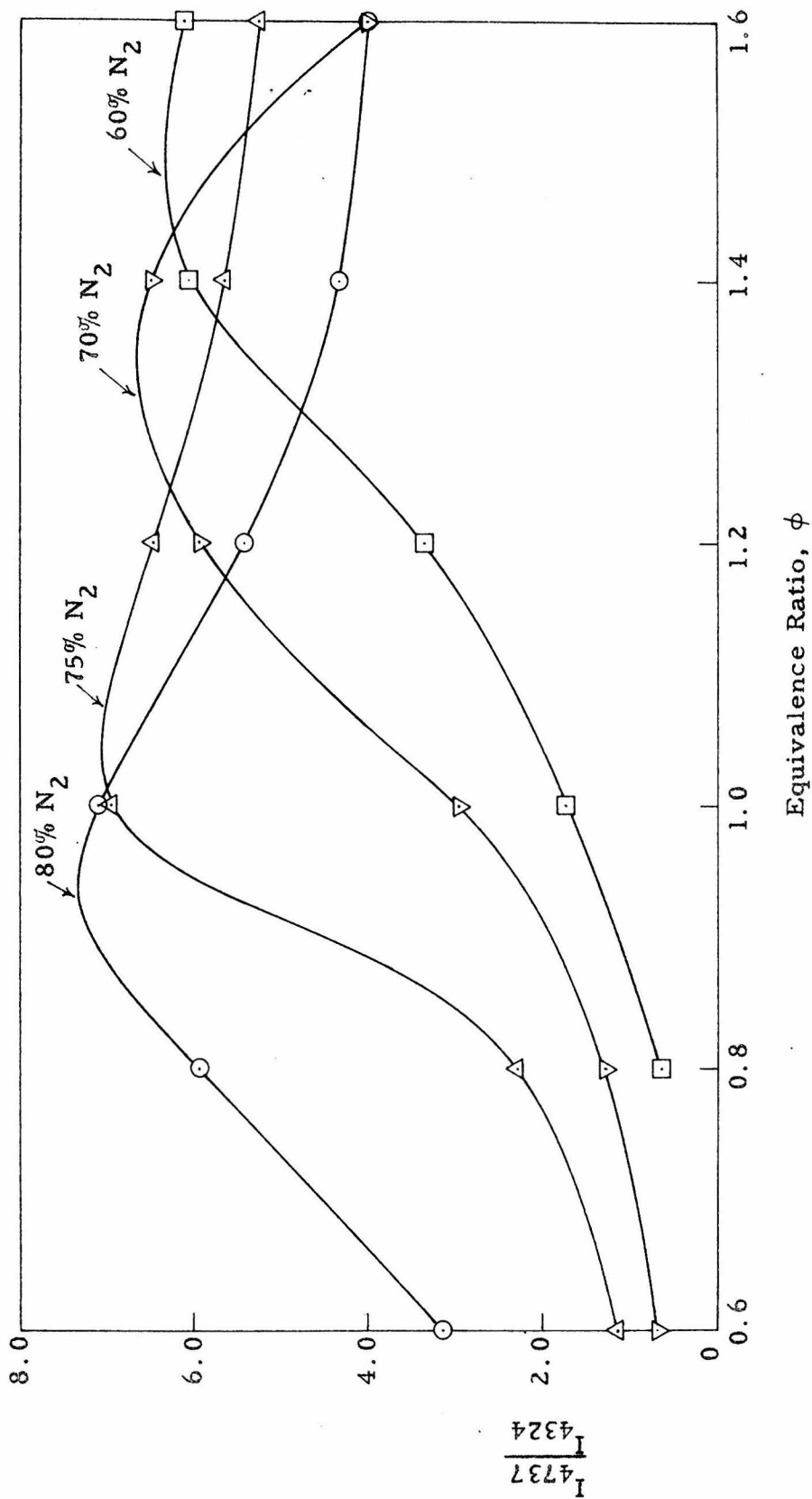


Figure 9. Spectral Intensity Ratio ( $I_{4737}/I_{4324}$ ) as a Function of Equivalence Ratio for Various Nitrogen Contents (by Mass). 0.704 cm I.D. Burner.

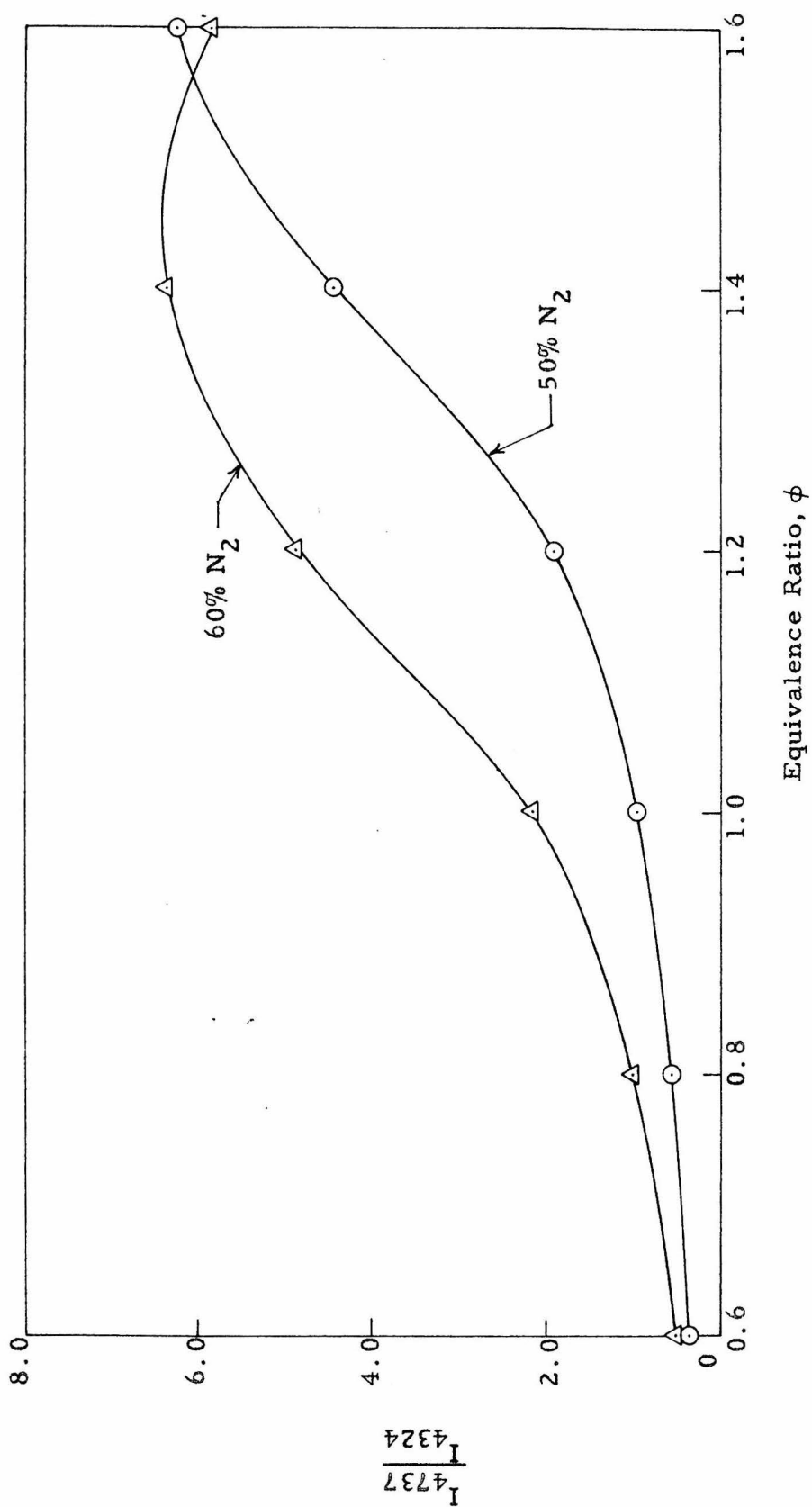


Figure 10. Spectral Intensity Ratio ( $I_{4737}/I_{4324}$ ) as a Function of Equivalence Ratio for Various Nitrogen Contents (by Mass). 0.386 cm I.D. Burner.

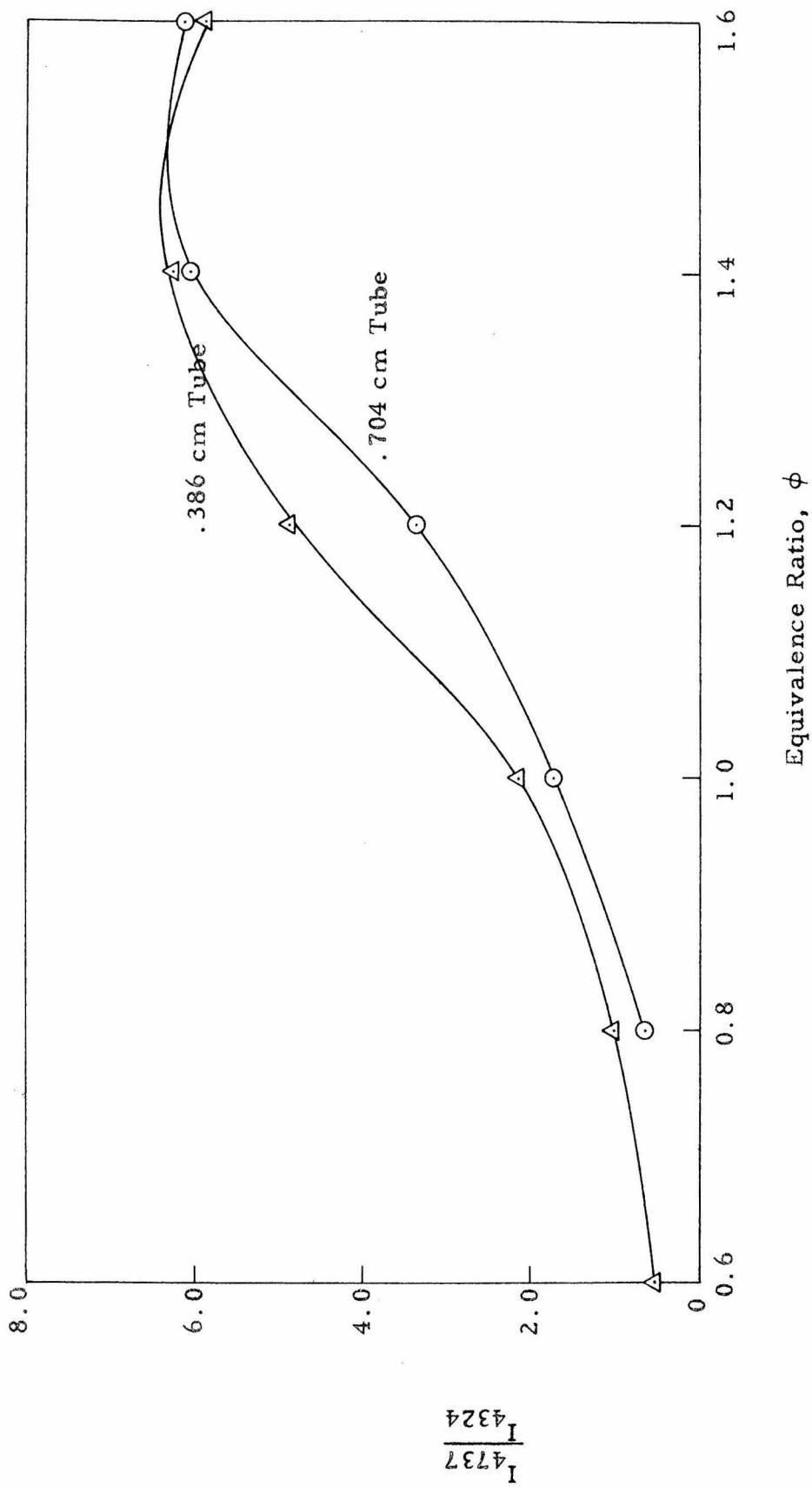


Figure 11. Spectral Intensity Ratio ( $I_{4737}/I_{4324}$ ) as a Function of Equivalence Ratio for 0.386 cm and 0.704 cm I.D. Burner. Nitrogen Content in Mixtures: 60% by Mass.

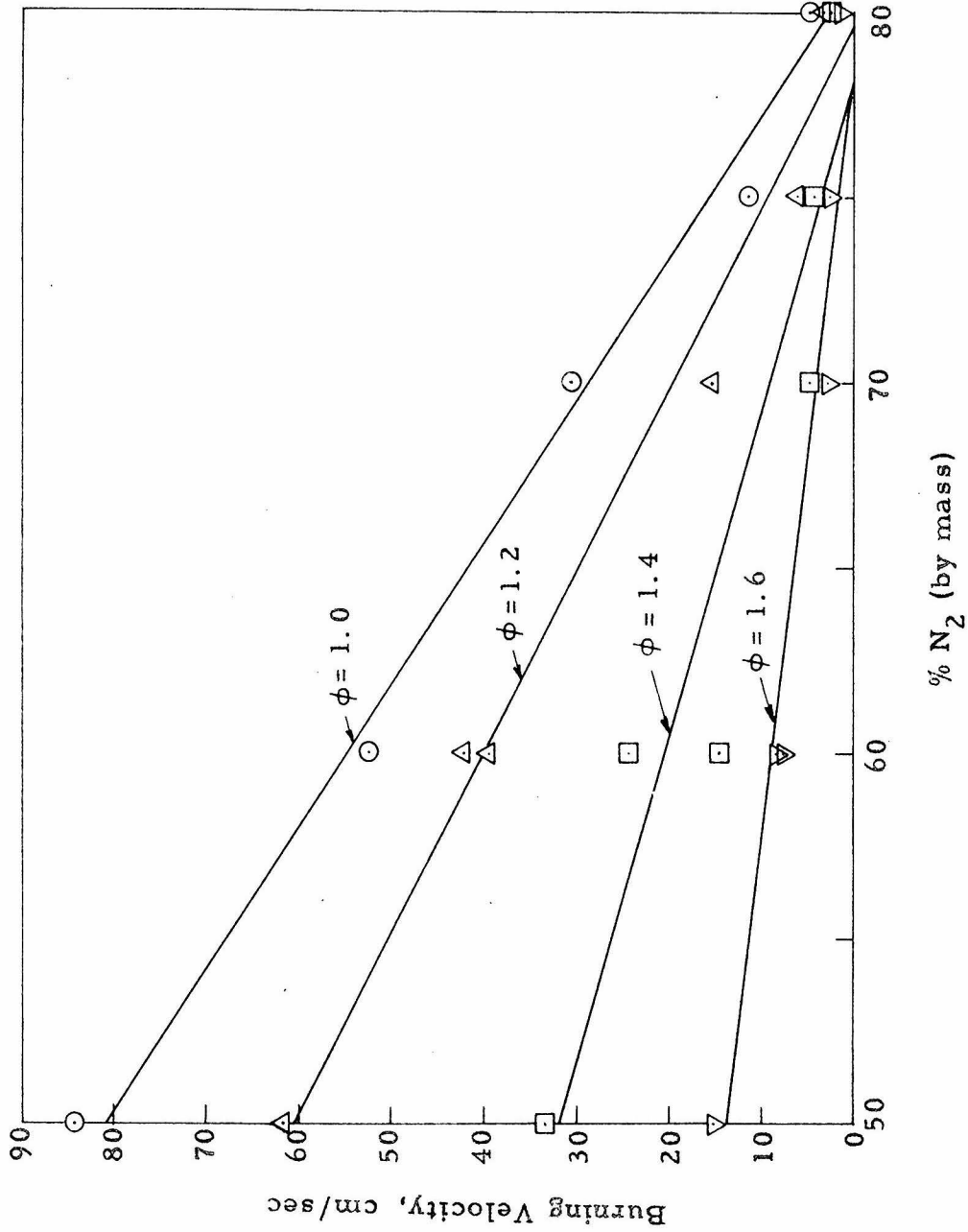


Figure 12. Laminar Burning Velocity as a Function of Nitrogen Content (% by Mass) for Various Equivalence Ratios.