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Technical Note

COMBUSTION STUDIES WITH A 12000-CURIE GOLD SOURCE

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By

Stuart W. Churchill
Alexander Weir, Jr.
Leroy F. Ornella
Roy L. Gealer
Robert E. Kelley
Martin E. Gluckstein

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PREFACE

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Beside the authors of this report, the following personnel of the University of Michigan contributed to the research work.

Mr. A. H. Emmons of the School of Public Health helped plan the shielding and irradiation, assisted in monitoring the source during the experimental work, and calculated the rate of irradiation of the flame and fuel-air mixture.

Mr. C. C. Palmiter and Mr. W. R. Dunbar of the School of Public Health directed the transfer of the source from the Materials Testing Reactor to the laboratory and monitored the source during the experiment.

Dr. J. H. Enns of the Department of Physics helped realign the spectrograph.

Mr. T. B. Khammash of the Aircraft Propulsion Laboratory assisted in the calculation of rotational temperature.

Mr. M. P. Moyle of the Aircraft Propulsion Laboratory assisted in installing the source and in analyzing the fuel-air mixture.

Mr. R. E. Cullen of the Aircraft Propulsion Laboratory assisted in the installation of the source.

Mr. E. T. Howard of the Aircraft Propulsion Laboratory assisted in the experimental work.

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Dr. W. B. Lewis of the Atomic Energy Division, Phillips Petroleum Co., helped ascertain the strength of the irradiation of the gold.

ABSTRACT

The effect of nuclear radiation from Au-198 and Au-199 on propane-air flames at low pressures was investigated experimentally. The source had an initial strength of 15,270 curies. Both the pre-flame mixture and the flame zone were irradiated. The number of ion pairs produced by beta radiation greatly exceeded that produced by gamma radiation. The rate of propagation of bunsen flames was increased up to 50 per cent by irradiation. The emission due to CH, C_2, and OH was measured at a series of elevations through a flat flame, and CH rotational temperatures were computed. The emission at 4315 Å due to CH was increased up to 33 per cent, and the emission at 5165 Å due to C_2 was increased up to 150 per cent with the source at a level of 1500 curies. The emission due to OH was not affected significantly. Increases in CH rotational temperature up to 100 per cent were calculated for a source level of 1500 curies. The maxima in the emission due to C_2 and OH were observed to occur at an earlier stage than the maximum in emission due to CH in the irradiated flames.

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12. Abstract: The effect of nuclear radiation from Au-198 and Au-199 on propane-air flames at low pressures was investigated experimentally. The source had an initial strength of 15,270 curies. Both the pre-flame mixture and the flame zone were irradiated. The number of ion pairs produced by beta radiation greatly exceeded that produced by gamma radiation. The rate of propagation of bunsen flames was increased up to 50 per cent by irradiation. The emission due to CH, C_2, and OH was measured at a series of elevations through a flat flame, and CH rotational temperatures were computed. The emission at 4315 Å due to CH was increased up to 33 per cent, and the emission at 5165 Å due to C_2 was increased up to 150 per cent with the source at a level of 1500 curies. The emission due to OH was not affected significantly. Increases in CH rotational temperature up to 100 per cent were calculated for a source level of 1500 curies. The maxima in the emission due to C_2 and OH were observed to occur at an earlier stage than the maximum in emission due to CH in the irradiated flames.

PART I. INTRODUCTION

In the course of investigations of the utilization of gross fission products at the University of Michigan, flames were irradiated with gold and palladium sources. These investigations, which were carried out at low intensities, indicated some effect upon flame stability (1) and no effect upon flame speed (2, 3).

The objective of this investigation was to study the effect of more intensive radiation upon flames. Previous studies on ramjet burners and low-pressure flames indicated that the emission spectra of the flame provide more information on the mechanism of the combustion process than flame speed and stability measurements (4, 5). Accordingly, equipment was designed to obtain a spectral traverse of a low-pressure flat flame subjected to nuclear radiation. The intensity of the C_2, CH, and OH bands was measured, and CH-rotational temperatures were computed as a function of distance through the flame at several levels of radiation. During the course of the experiment itself, it was found possible to obtain flame-speed measurements as well as the spectral data.

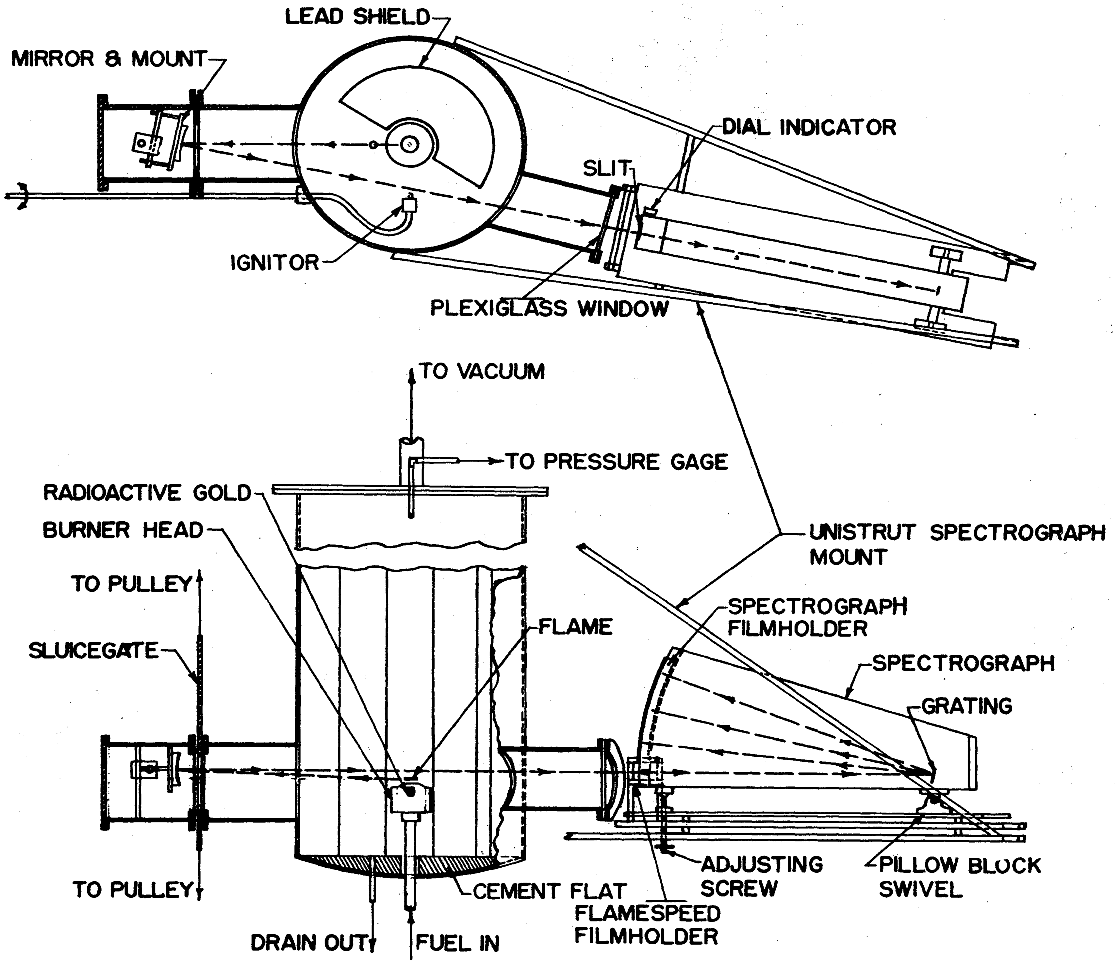
The source of radiation was approximately 30 grams of gold wire with an intensity of about 15,000 curies at the beginning of the experimental work. A series of measurements was made as the source decayed to 0.0008 of its initial intensity.

PART II. EXPERIMENTAL EQUIPMENT

The equipment consisted of a premixed propane-air supply and metering system, a burner designed to produce a flat flame, a vacuum tank and system, radiation shielding, an optical traversing system, and the source of radiation. A diagram of the system is shown in Figure 1.

A. BURNER ASSEMBLY

The burner and source assembly are shown in Figure 2. The burner head was a converging nozzle with a 100-mesh copper screen stretched across the 1-inch I.D. opening. With such a screen, a flat flame can be obtained



NOTE: LIGHT PATH INDICATED BY ——→——→——

Fig. 1. Experimental equipment

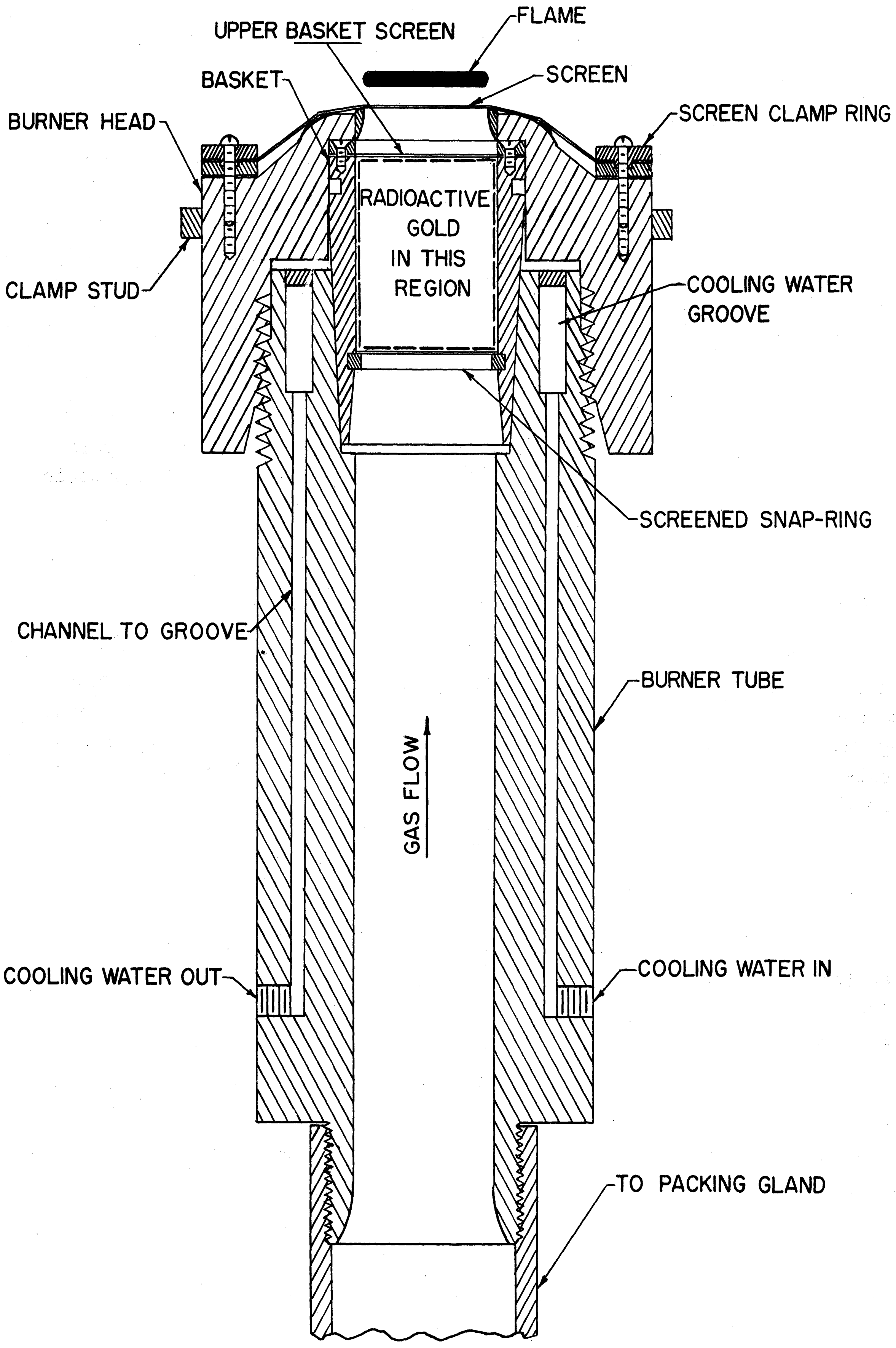


Fig. 2. Burner assembly

at pressures up to one-half an atmosphere as compared to about one-tenth of an atmosphere without a screen. At higher pressures, the familiar bunsen cone is obtained.

As indicated in Figure 2, the source was contained in a tapered hollow cylinder covered at the top and bottom with 100-mesh copper screen. The screen at the bottom was mounted in a ring which snapped into a groove inside of the cylinder or basket. The source was prepared from 0.005-inch diameter 99.95 per cent pure gold wire. Before irradiation, the wire was wound into a number of 1/16-inch-diameter helices. After irradiation, these helices were inserted in the basket and compressed by installation of the bottom screen.

Cooling water was circulated through an annular space in the top of the 8-inch-long burner tube. The basket was supported by the tapered inner wall of the tube, and the burner head was threaded onto the outside of the tube.

B. FUEL-AIR SUPPLY

Five mixtures of air and propane were prepared and stored in cylinders. The cylinders were buried at some distance from the test stand for reasons of safety. The gas from the cylinders was passed through a reducing valve and a rotameter to measure the rate of flow. The rate was controlled by a hand-operated needle valve. The fuel-air mixture then passed through a flame trap and into the burner tube.

C. VACUUM SYSTEM

The burner assembly was located inside a tank 2 1/2 feet in diameter and 9 feet high. The removable tank cover was fastened with swing bolts and sealed with a lead core o-ring. The exhaust gas from the flame passed through a 2-inch flexible metal hose leading from the cover of the tank to a filter box. The filter box contained a Cambridge absolute filter to remove entrained particles which might be radioactive. The exhaust gas then passed through a Nash Hytor vacuum pump (using water as a working fluid) to the atmosphere. The pressure in the tank was controlled by bleeding air into pump suction.

The vacuum tank also contained a pressure tap, a lamp for illumination, and a spark plug for ignition, mounted on a swinging arm and controllable from outside the tank.

D. OPTICAL SYSTEM

As indicated in Figure 1, a front surface aluminum-coated convex mirror was mounted in 12-inch pipe welded on a side of the tank. The mirror reflected light emitted by the flame out through a plastic window in the tank wall and focused the image of the flame on the slit of the spectrograph. Dry air was blown on the mirror and window to prevent fogging. A slide valve was installed in the 12-inch pipe to keep the mirror dry when the tank was filled with water.

The spectrograph was a Cenco student model with a plastic replica grating. It was mounted directly on the tank to prevent relative motion between the spectrograph and flame image. The construction of the mount is shown in Figure 1. The back of the spectrograph was pivoted on an axle set in a pair of pillow blocks fastened to a bed plate. The front of the spectrograph rested on an adjusting screw fastened to the same bed plate. Thus, the position of the horizontal slit could be adjusted up and down to traverse the flame image vertically. A dial indicator gage, reading in thousandths of an inch, was mounted in such a way that the elevation could be recorded.

Thus light emitted by the flame passed from the flame to the mirror, through the window, through the spectrograph slit, to the reflecting spectrograph grating, and then to the photographic film where the intensities of the various wavelengths were recorded.

A frame made to hold a 4 x 6-inch film holder was mounted immediately in front of the spectrograph slit in such a position that the image from the mirror focused on the film. Photographs obtained in this manner were used for flame-speed measurements.

E. SHIELDING

Considerable shielding was necessary to protect personnel from gamma radiation during the installation of the gold source and during the experimental work. It was also necessary to shield photographic film during a four-hour period in the spectrograph.

A hemi-cylindrical ring of lead 7 inches thick, weighing 2500 pounds, shielded one side of the burner, as shown in Figure 1. Four hundred and fifty 100-pound bags of Portland cement were stacked around the perimeter of the tank. Portland cement was found to be the cheapest bagged material on a mass basis available. About 500 pounds of lead sheet were installed at strategic positions, particularly around the spectrograph.

During the installation of the source and burner head, the vacuum tank was filled with water. The water provided trnasparent shielding and permitted direct observation and manipulations from the top of the tank.

PART III. SOURCE

A. SELECTION

Gold was selected as a source material because the half-life obtained upon irradiation was more convenient for the experimental work than the half-life of other available sources of beta radiation. As indicated in the disintegration scheme in Figure 3, the half-life of Au-198 is 2.69 days while that of Au-199 is 3.15 days (6). This average half-life of about 3 days allowed a 24-hour experimental run to be completed with only a 20 per cent decrease in activity. The gamma radiation which is emitted from both Au-198 and Au-199 is, of course, somewhat disadvantageous because of shielding problems and the possible effect of the gamma radiation upon combustion.

B. PREPARATION

The source was prepared form 99.95 per cent gold wire, 0.005 inch in diameter. The wire was wound helically on a 1/16-inch rod into coils approximately 12 inches long. The coils were then rolled into two small loose balls of 15 grams each. Each ball was then placed in an aluminum cylinder containing a helium atmosphere. Figure 4 is a drawing of the cylinder. These aluminum cylinders were irradiated for 4 1/2 days in a high-flux section of the Materials Testing Reactor, Arco, Idaho.

C. TRANSFER

After irradiation, the gold wire was transferred in the Materials Test Reactor canal from the aluminum cylinders to the basket shown in Figure 2 and the loading string was attached. The basket was then placed in the shipping container shown in Figure 5. The container was flown from Idaho Falls, Idaho, to Willow Run Airport by the Air Force.

The container was unloaded from the plane and after removal of the cover bolts was lowered into the tank which had previously been filled with water. Figure 6 shows this operation during a rehearsal. With the container resting on the top of the lead hemisphere, the loading string attached to the basket was strung through an eye suspended above the tank. The lid of the shipping container was then removed. The basket was next pulled out of the container with the string and, with the aid of a mirror, was lowered into the bottom of the tank. After removal of the shipping container, the basket was lifted into the burner tube with the string. The string was cut and the burner head screwed into position using extension tools. Figure 7 shows a rehearsal of this operation. The cover of the tank

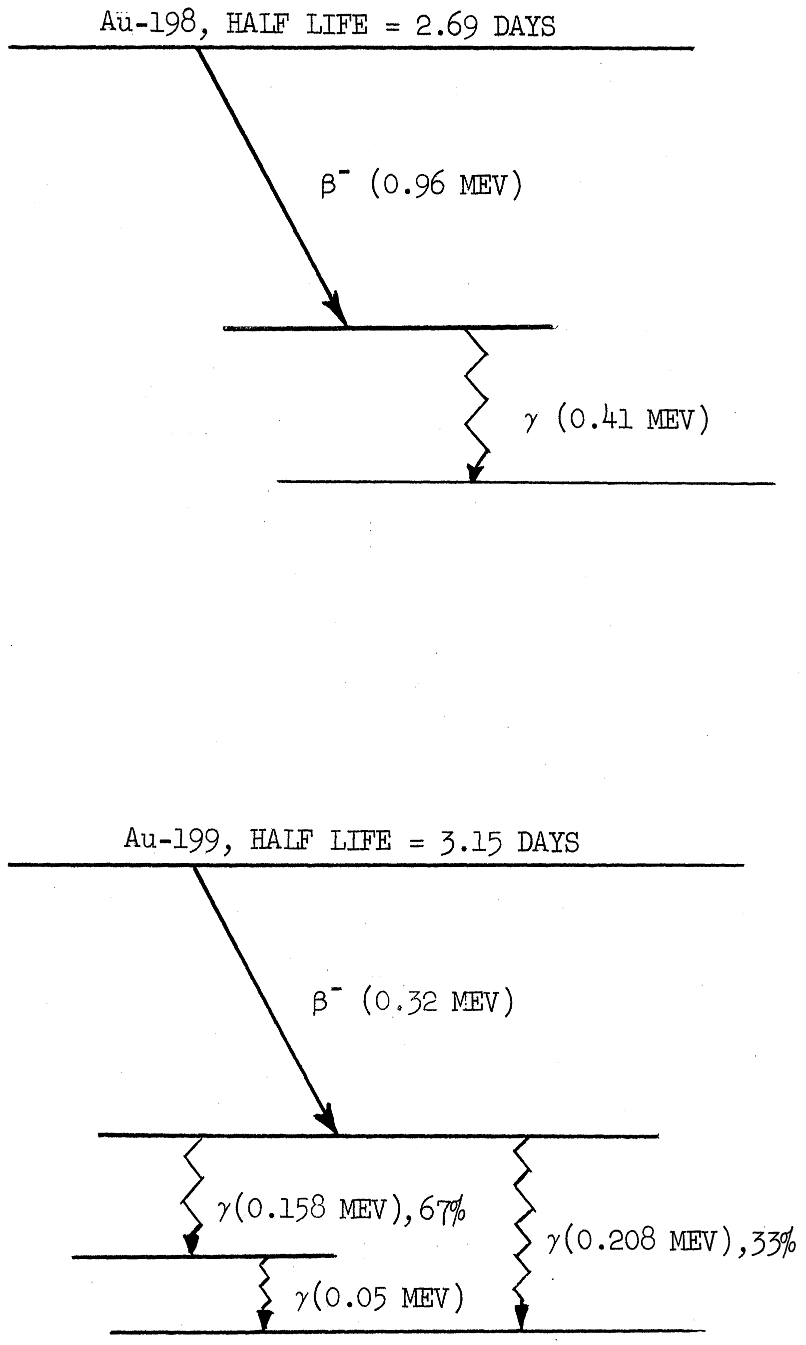


Fig. 3. Disintegration Schemes of Au-198 and Au-199

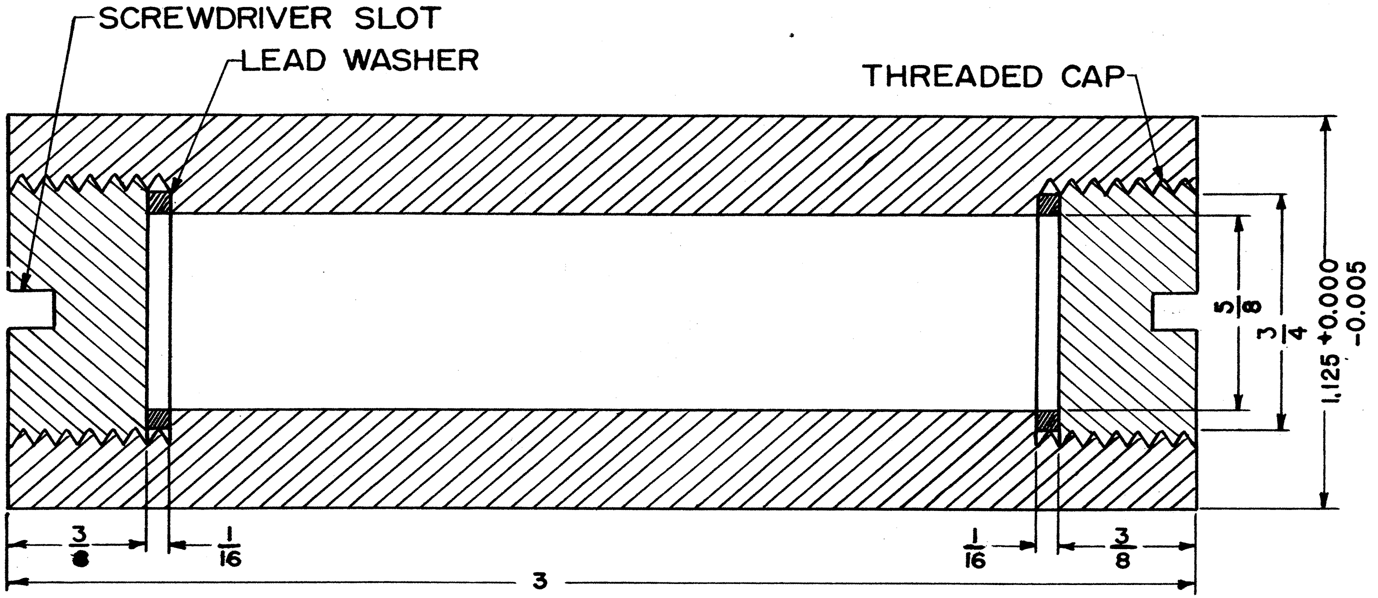
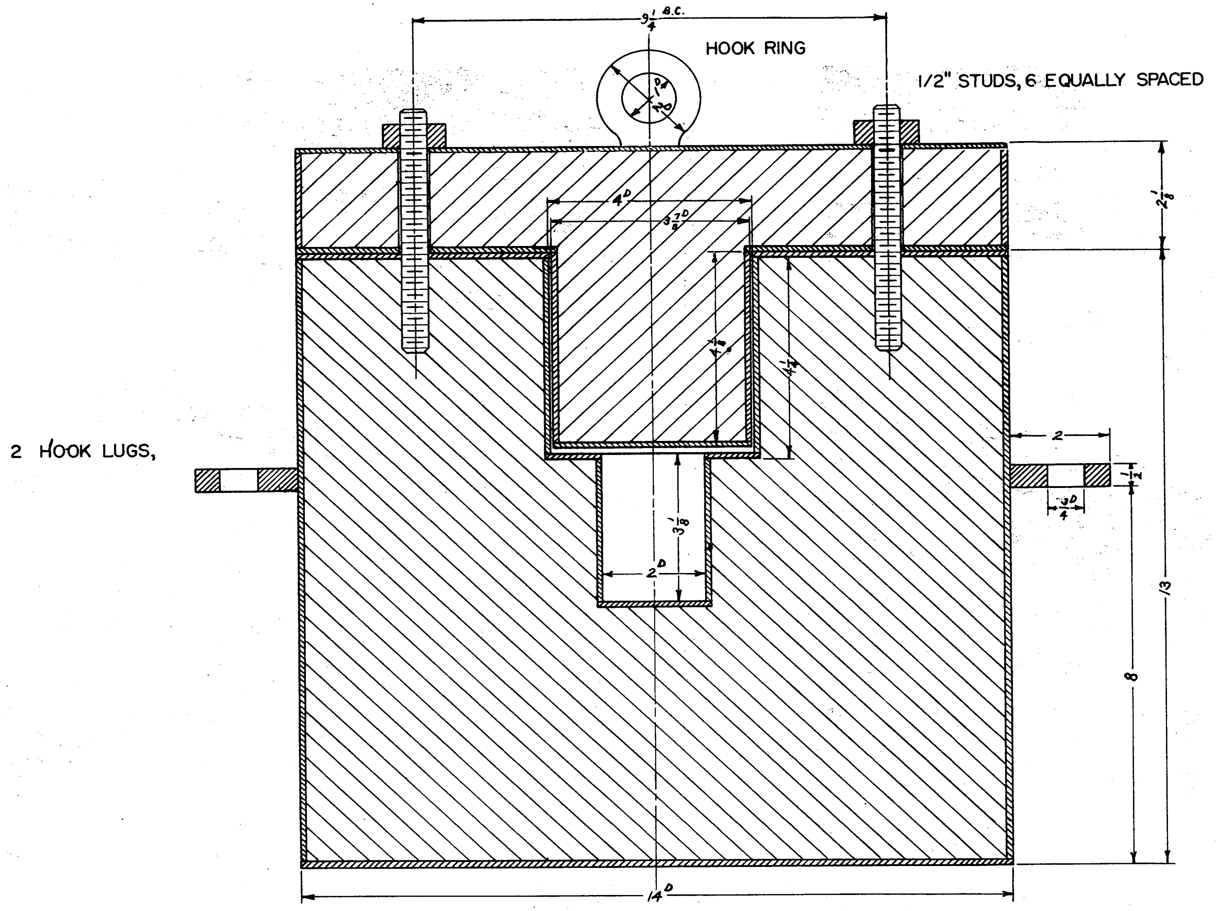


Fig. 4. Aluminum cylinder for retention of gold in Materials Testing Reactor



NOTE: CONTAINER MADE OF CAST LEAD
ENCASED IN 1/8" SHEET STEEL

Fig. 5. Shipping container

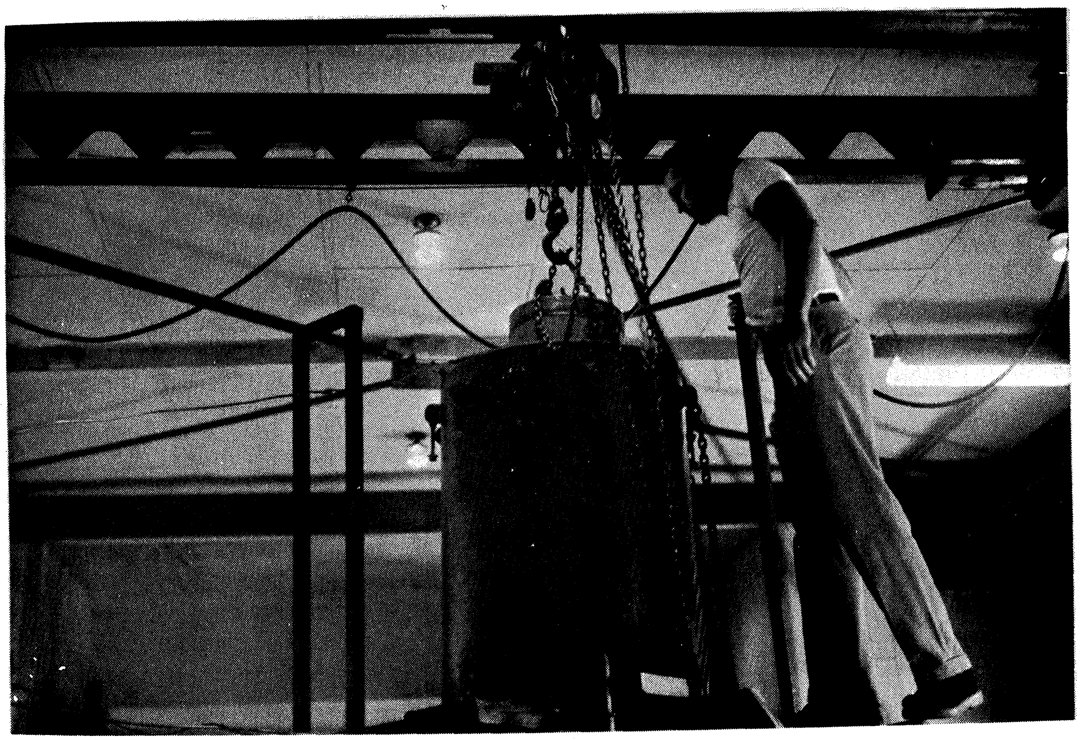


Fig. 6. Lowering the shipping container into tank (rehearsal)

Fig. 7. Installing the burner head (rehearsal)

was bolted down and the water in the tank was forced out under air pressure. The slide valve protecting the mirror was opened and air was introduced in several places for drying purposes.

The entire transfer operation was carefully monitored. As a result of considerable rehearsal, all these operations were carried out with dispatch and with an exposure of less than 100 milliroentgens for any individual.

After removal of the water from the tank, intensities of about 800 milliroentgens per hour were encountered in the region of the spectrograph. This high rate was due to unanticipated scatter through the optical port and from the ceiling. Original plans called for eight twenty-minute exposures on each spectrographic film and for each individual to work in this area about 10 hours per week. Since 30 milliroentgens on the film and 300 milliroentgens per week for personnel were permissible, it was apparent that considerable additional shielding was necessary in the region of the spectrograph. One hundred fifty 100-pound bags of cement and 500 pounds of lead were added to the shielding before any experimental work was begun.

D. ACTIVITY

As noted previously, as strong a source of beta radiation as possible was considered desirable in order to bound any possible effects of beta radiation upon the fuel-air mixture and the flame. It appeared that as much as 25,000 curies could be handled safely in the proposed experimental program. Theoretically, 25,000 curies could be obtained from 30 grams of gold irradiated in a high-flux region of the Materials Testing Reactor (7).

The gold wire was actually irradiated for a slightly shorter period than requested. On the basis of neutron flux calculations, the activity of the gold on removal from the pile at 11 a.m. EST, July 22, 1955, was reported by W. B. Lewis (8) to be:

Au-198	9,600 curies
Au-199	5,670 curies
Total	15,270 curies

Experimental measurements of the radiation from the gold source were made through the water in the tank during installation. These data were compared with data taken under similar conditions with a known Co-60 source. The activity of the gold computed from these measurements agreed within 20 per cent with the above values. After completion of the combustion experiments, the gold was removed from the burner and the gamma spectrum was measured. The ratio of Au-198 to Au-199 determined from the gamma spectrum and extrapolated back to removal from the reactor was also in reasonable agreement with the above values. Accordingly, the above values were used in subsequent calculations.

Based on half lives of 2.69 days for Au-198 and 3.15 days for Au-199 the curie values of the source as a function of time are represented by the equations

$$C_{Au-198} = 7740\ e^{-0.00537t} \tag{1}$$

and

$$C_{Au-199} = 3930\ e^{-0.00917t} \tag{2}$$

where t = time in hours measured from the hour of the first experiment at 3 a.m. EST, July 24, 1955.

The periods of the various combustion experiments are noted on a plot of the relative intensities in Figure 8.

The number of ion pairs produced in the gas phase in various regions would appear to be the most significant phenomenon resulting from the gold source. The decay scheme in Figure 3 was used to compute the number of ion pairs per cubic centimeter produced by the source within the cylinder containing the source and in the region of the flame itself.

Suitable corrections were made for scattering and absorption by the gold wire, the cylinder wall, and the retaining screens. The results are summarized in Table I.

TABLE I

ION PAIRS PRODUCED BY GOLD SOURCE

	Ion pairs/(cc)(sec)(curie of Au-198 or Au-199)		
	Beta Production	Gamma Production	Total Production
		In Flame Front	
From Au-198	5.38×10^9	2.07×10^9	7.45×10^9
From Au-199	8.00×10^4	1.03×10^9	1.03×10^9
		In Cylinder Containing Source	
From Au-198	2.42×10^{13}	3.16×10^9	2.42×10^{13}
From Au-199	2.68×10^{12}	1.68×10^9	2.68×10^{12}

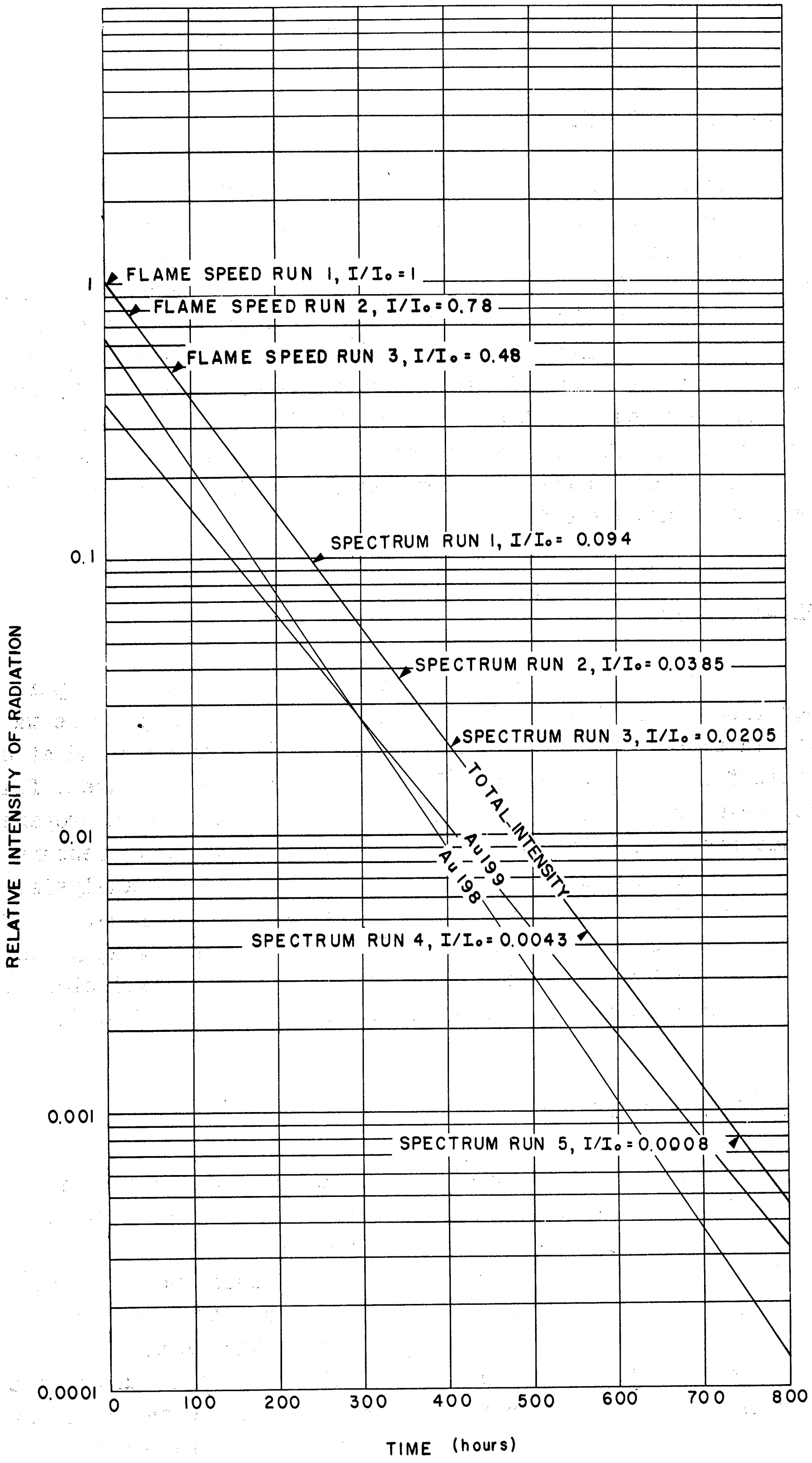


Fig. 8. Source strength during experimental program

PART IV. EXPERIMENTAL PROCEDURE

A. TIME OF EXPERIMENTS

Figure 8 shows the relative intensity of radiation during the periods in which the spectrographic traverses and the flame speed measurements were performed. As was previously stated, supplemental lead shielding was placed on the spectrograph assembly after installation of the gold. This necessitated realignment of the spectrograph, since the flame, mirror image, slit, and grating (see Figure 1) must be in the same plane. The realignment procedure involved repetitive adjustments, each followed by a thirty-minute exposure of the photographic film at various locations within the spectrograph. This necessary but time-consuming process resulted in the time delay apparent in Figure 8 between the initial flame speed and initial spectrographic measurements.

B. FUEL-AIR RATIO

The fuel used in the experiments was 99.5 per cent pure propane obtained from the Mathieson Chemical Corporation. The propane was premixed with air in eight 3-cu ft cylinders buried 5 feet underground at a distance of 50 feet from the laboratory. The cylinders were evacuated, filled with propane, re-evacuated to 2 in. Hg, and finally filled with propane to a predetermined pressure. Air was then added until the total pressure was 225 psia. After a four-hour period, samples were taken for analysis. Samples were not obtained of the air-fuel mixtures used in the first two flame-speed runs and the composition was computed from the mixing procedure. Samples of subsequent mixtures were analyzed by an Orsat-explosion technique. The calculated compositions and analyses are given in Table II.

C. FLAME-SPEED EXPERIMENTS

The vacuum was adjusted until the desired pressure existed within the tank. The rate of flow of the fuel-air mixture was adjusted until a bunsen flame existed. A 4 x 6-inch filmholder was placed in the filmholder frame. No shutter was used. The room was darkened and the mask was removed from the filmholder and quickly replaced. With contact Process Ortho film, this procedure yielded a satisfactory image of the flame. A typical photograph is shown in Figure 9. The rate of flow was then changed and the procedure repeated. Four different rates of flow were established at each pressure. When the rate of flow was reduced, the area of the flame front decreased but the flame speed remained relatively constant. The flame speed would be independent of flow rate if a heat sink (burner head) were not lo-

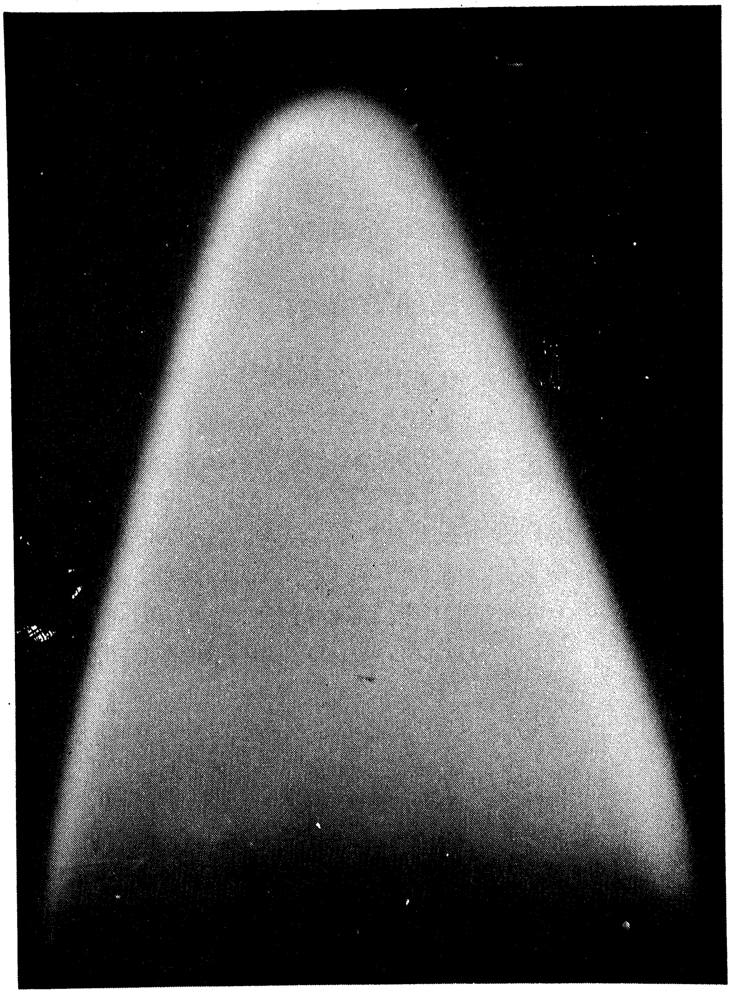


Fig. 9. Typical photograph of irradiated bunsen flame

Table II

COMPOSITION OF PROPANE-AIR MIXTURES

Run	Pressure in. Hg	Propane-to-Air Ratio lb/lb
Flame-Speed Run No. 1	10	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
	8	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
	6	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
	4	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
Flame-Speed Run No. 2	8	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
	6	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
	4	0.08*, 0.07*, 0.0637*, 0.06*, 0.055*
Flame-Speed Run (Cold)	10	0.0917-0.0939, 0.0748-0.0758, 0.0710-0.0777 0.067-0.067, 0.0603-0.0642
	8	0.0917-0.0939, 0.0748-0.0758, 0.0710-0.0777 0.067-0.067, 0.0603-0.0642
	6	0.0917-0.0939, 0.0748-0.0758, 0.0710-0.0777 0.067-0.067, 0.0603-0.0642
	4	0.0917-0.0939, 0.0748-0.0758, 0.0710-0.0777 0.067-0.067, 0.0603-0.0642
Spectral Run No. 1	14	0.0802-0.0883, 0.0709-0.072, 0.0645-0.0682 0.0677-0.0679
	10	0.0802-0.0883
	8	0.0802-0.0883
	6	0.0802-0.0883
Spectral Run No. 2	14	0.0747-0.0818, 0.0613-0.0629, 0.0637* 0.0621-0.0641
	10	0.0747-0.0818
	8	0.0747-0.0818
	6	0.0747-0.0818

---

*Composition computed from mixing procedure. For all other mixtures, the range of compositions obtained from the Orsat-explosion analyses are given.

TABLE II (cont.)

COMPOSITION OF PROPANE-AIR MIXTURES

Run	Pressure in. Hg	Propane-to-Air Ratio lb/lb
Spectral Run No. 3	14	0.0772-0.0805, 0.0691-0.0705, 0.0596-0.0629 0.0574-0.0726
	10	0.0772-0.0805
	8	0.0772-0.0805
	6	0.0772-0.0805
Spectral Run No. 4	14	0.0834-0.0852, 0.0716-0.0788, 0.0637-0.0667 0.0523-0.0640
	10	0.0834-0.0852
	8	0.0834-0.0852
	6	0.0834-0.0852
Spectral Run No. 5	14	0.0829-0.0874, 0.0813-0.0845, 0.0704-0.0706 0.0648-0.0668
	10	0.0829-0.0874
	8	0.0829-0.0874
	6	0.0829-0.0874

cated near the flame. The flame speeds obtained at the four rates of flow were averaged, and only the average was considered in the correlations.

The pressure in the tank was changed and the entire procedure repeated at the same fuel-air ratio. Then a different fuel-air ratio was introduced and the procedure repeated again at absolute pressures of 4, 6, 8, and 10 in. Hg. On succeeding days, at lower intensities of radiation, the entire procedure was repeated.

D. SPECTROGRAPHIC TRAVERSES

The desired pressure was established within the tank, and the rate of flow was adjusted until a flat flame floating above the tight screen with a very thin gap between the screen and the zone of burning was obtained. The flame was essentially a disc about 1 inch in diameter and of the order of 0.05 inch thick. The exact thickness and distance above the screen varied with the pressure in the tank and the rate of flow. This thin vertical dimension of the flame was traversed by the spectrograph. The spectrograph film pack was loaded with Royal Pan film and the slit adjusted to the desired elevation. The room was darkened, the mask was removed from the film pack, and the shutter was opened for thirty minutes. The shutter was closed, and a new strip of the film was shifted into position. The slit elevation was adjusted to the elevation in the flame at which the next exposure was to be taken, and the shutter was again opened for thirty minutes. This procedure was repeated for about eight different elevations through the flame. A maximum of fourteen exposures per film was available.

The pressure in the tank was then adjusted to a new value and the rate of flow adjusted so that the volumetric rate of flow at the pressure of the tank remained constant, i.e., the linear velocity of gas at the burner head was kept constant. The rate of flow for the first run was chosen so that the flame did not become a bunsen flame at the lower pressures nor burn under the screen at the higher pressures established for subsequent runs. As a further condition, the flame had to be sufficiently intense to produce a satisfactory spectrographic negative in 30 minutes.

The procedure was repeated at several pressures for one fuel-air ratio. Then the tank pressure was held constant, and the procedure repeated for several fuel-air ratios. On succeeding days, at lower intensities of radiation, the entire procedure was repeated.

The spectrographic films were developed under constant conditions to ensure a valid comparison of the density of exposure at the various wavelengths. They were developed in 15 gallons of DK-50 developer, diluted 1 to 1 with water, at constant temperature, agitation, and developing time. The same batch of developer was used for all of the films to guard against changing developer strength. The quantity of developer used was so great that it was assumed that its strength did not change appreciable with use.

After developing, the films were fed through a microphotometer which scanned and recorded the density of exposure as a function of the wavelength. A sample microphotometer tracing is shown in Figure 10.

PART V. RESULTS

A. FLAME-SPEED EXPERIMENTS

Using the photographs of the bunsen flames, the circumferential areas of the flame surfaces were computed from enlargements of the negatives. The flame speeds were computed from the relation

$$v_f = Q/A_f \tag{3}$$

where v_f = flame speed (rate of propagation of flame normal to flame front), ft/sec

Q = volumetric rate of flow of air-fuel mixture leaving the burner head, cu ft/sec

A_f = $2\pi L \Sigma R_n$ = surface area of flame, sq ft

L = length of increment along perimeter of major vertical cross section of flame

R_n = mean radius of element of flame surface

Figure 11 is a schematic diagram illustrating the method of flame-area measurement. Equation (3) assumes that the density of the material leaving the burner head is equal to the density of the material entering the flame front, which is the usual assumption made in computing flame speeds by the Guoy method.

The average flame speeds computed from the photographs at four different rates of flow are plotted versus nominal fuel-air ratio in Figure 12 for tank pressures of 4, 6, 8, and 10 in. Hg abs. As previously stated, analyses were not obtained for the fuel-air ratios used in the first two flame-speed experiments (at relative intensities of radiation of $I/I_o = 1$ and $I/I_o = 0.78$). The shift in the maxima of the curves for the second flame-speed experiment ($I/I_o = 0.78$) can undoubtedly be attributed to an incorrect value for the computed fuel-air ratio rather than to the change in intensity of radiation. The curves for $I/I_o = 0$ in Figure 12 were obtained after the source of radiation was removed from the system. The compositions of the mixtures used in all but the first two experiments were obtained by analysis.

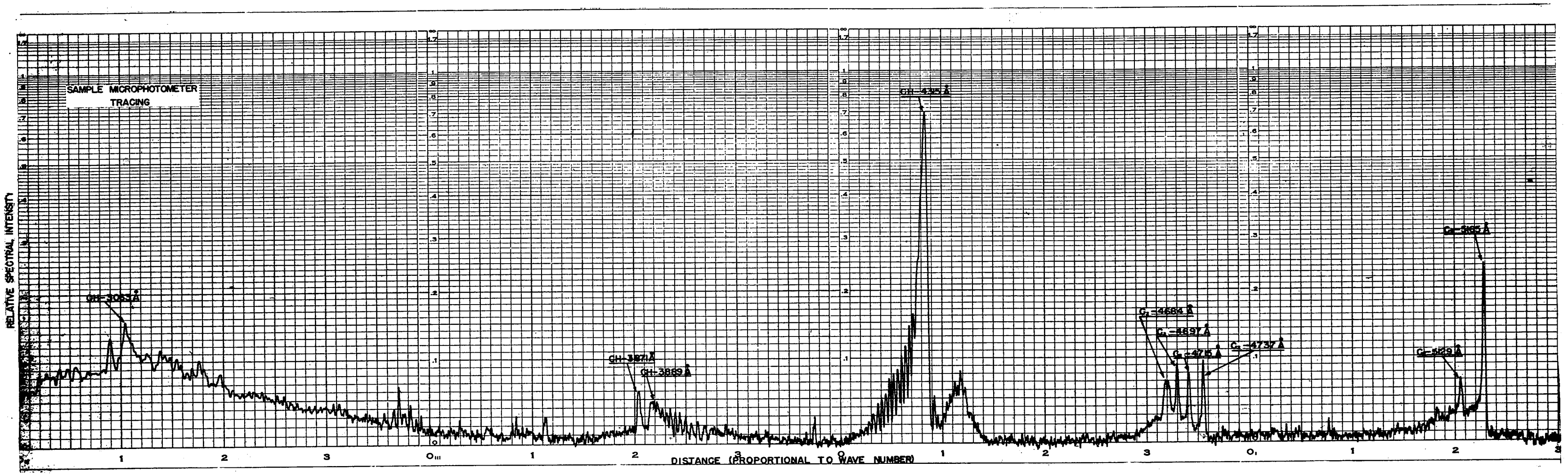


Fig. 10. Sample microphotometer tracing

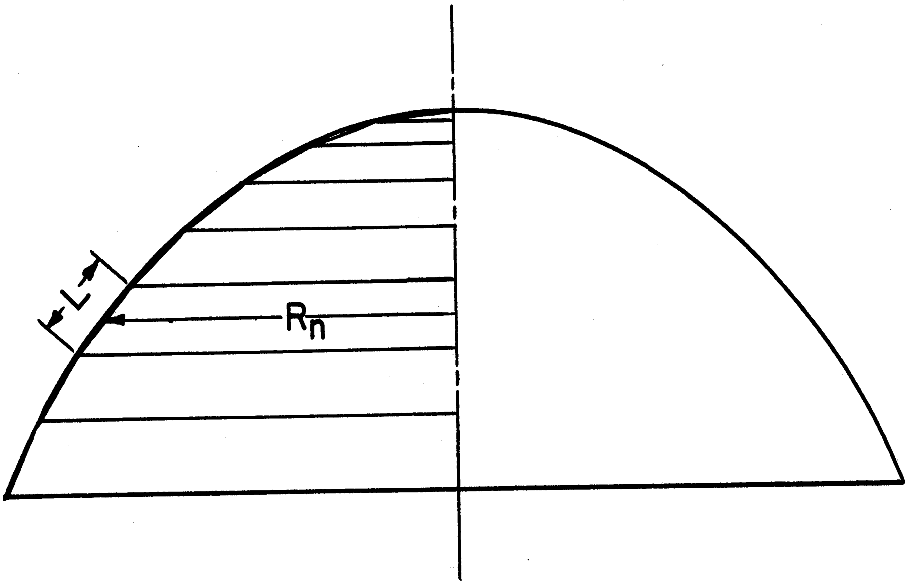


Fig. 11. Area measurement for flame-speed calculations

Figure 8 indicates that a third flame-speed experiment was performed at a relative intensity of radiation of 0.48. The results of this experiment are not plotted in Figure 12 but were discarded because of deformation of the screen and gas leakage between screen and burner head. The deformation of the screen occurred because the system was deliberately operated (after the second flame-speed experiment) so that burning occurred on the screen. This was necessary in order to obtain a maximum emission of light during realignment of the spectrograph. A new screen was inserted before spectrographic experiments were made, and again before the flame-speed experiment at $I/I_o = 0$. In the latter experiment, the container and gold source were placed with an identical container and an equivalent amount of copper wire.

The data presented in Figure 12 indicate that an increase in intensity of radiation yields an increase in flame speed. The maxima in the flame-speed curves were plotted versus relative intensity of radiation in Figure 13a, since it was assumed that all the maxima occurred at the same fuel-air ratio. Figure 13b shows the effect of pressure on flame speed at three intensities of radiation. The decrease in flame speed with decreasing pressure below a certain pressure (6 in. Hg) was previously shown by Cullen (9, 10) to be due to the action of the burner head as a heat sink.

B. SPECTRAL INTENSITIES

CH at 4315 Å

As indicated previously, traces of the flame were made at five different intensities of radiation, varying first pressure and then the fuel-air ratio at each intensity. The strongest emission due to CH occurred at 4315 Å. The relative intensity of light emitted at this wavelength is plotted versus elevation through the flat flame at various pressures in Figure 14. Separate plots are presented for relative intensities of radiation of 0.094, 0.0043, and 0.0008. The average fuel-air ratio in these experiments was 0.085. The maximum spectral intensities with respect to elevation through the flame obtained from Figure 14 are then plotted versus relative intensity of radiation for the various pressures in Figure 15.

The spectral intensity at 4315 Å is plotted versus elevation through the flat flame at various fuel-air ratios and intensities of radiation at a constant pressure of 14 in. Hg abs. in Figure 16. The maximum spectral intensities with respect to elevation through the flat flame obtained from Figure 16 are then plotted versus fuel-air ratio at the various intensities in Figure 17. The fuel-air ratios are plotted as a range corresponding to the results obtained from several chemical analyses of the same sample. Figure 17 is cross-plotted in Figure 18 to yield a curve of maximum spectral intensity versus relative intensity of radiation at various fuel-air ratios.

C_2 at 5165 Å and OH at 3063 Å

The most intense spectral line due to C_2 occurred at 5165 Å, and the most intense spectral line due to OH occurred at 3063 Å. The same procedure was followed for these spectral lines as for the CH at 4315 Å. However, in some cases, the intensities of C_2 and OH were so low that no information could be gained from the microphotometer traces. Spectral intensities of C_2 at 5165 Å and OH at 3063 are plotted versus elevation at an average fuel-air ratio of 0.085 and various pressures and relative intensities of radiation in Figure 19.

Maximum spectral intensities of C_2 at 5165 Å with respect to elevation are plotted versus relative intensity of radiation for an average fuel-air ratio of 0.085 and pressures of 14, 10, and 8 in. Hg in Figure 20. The maximum spectral intensities of OH at 3063 Å are also shown for pressures of 14 and 10 in.Hg.

The emission due to C_2 and particularly OH was much less than from CH. These low intensities of light, coupled with the wide variations in fuel-air ratio in the experiments at 14 in. Hg abs. prevent any meaningful presentation of the C_2 and OH intensities in the form used for CH in Figure 18.

PART VI. DISCUSSION

A. FLAME SPEEDS

The experimental data in Figure 13 clearly indicate the marked increase in flame speed caused by irradiation of the flame. The flame speed at a relative intensity of radiation of 1 corresponding to an absolute intensity of 11, 670 curies is at least 50 per cent greater than the flame speed of an unirradiated flame. The negligible effect of beta radiation on flame speed reported by Morrison, Cullen, and Weir was based on experiments performed with a 6.6-curie source (2) and again with a 36-curie source (3). Figure 13 indicates that a source considerably stronger than 36 curies is required to produce a significant increase in flame speed.

The increase in flame speed may be attributed to the energy imparted to the reacting molecules by beta particles, since Table I indicates that the number of ion pairs produced by gamma radiation is negligible in comparison to those produced by beta radiation.

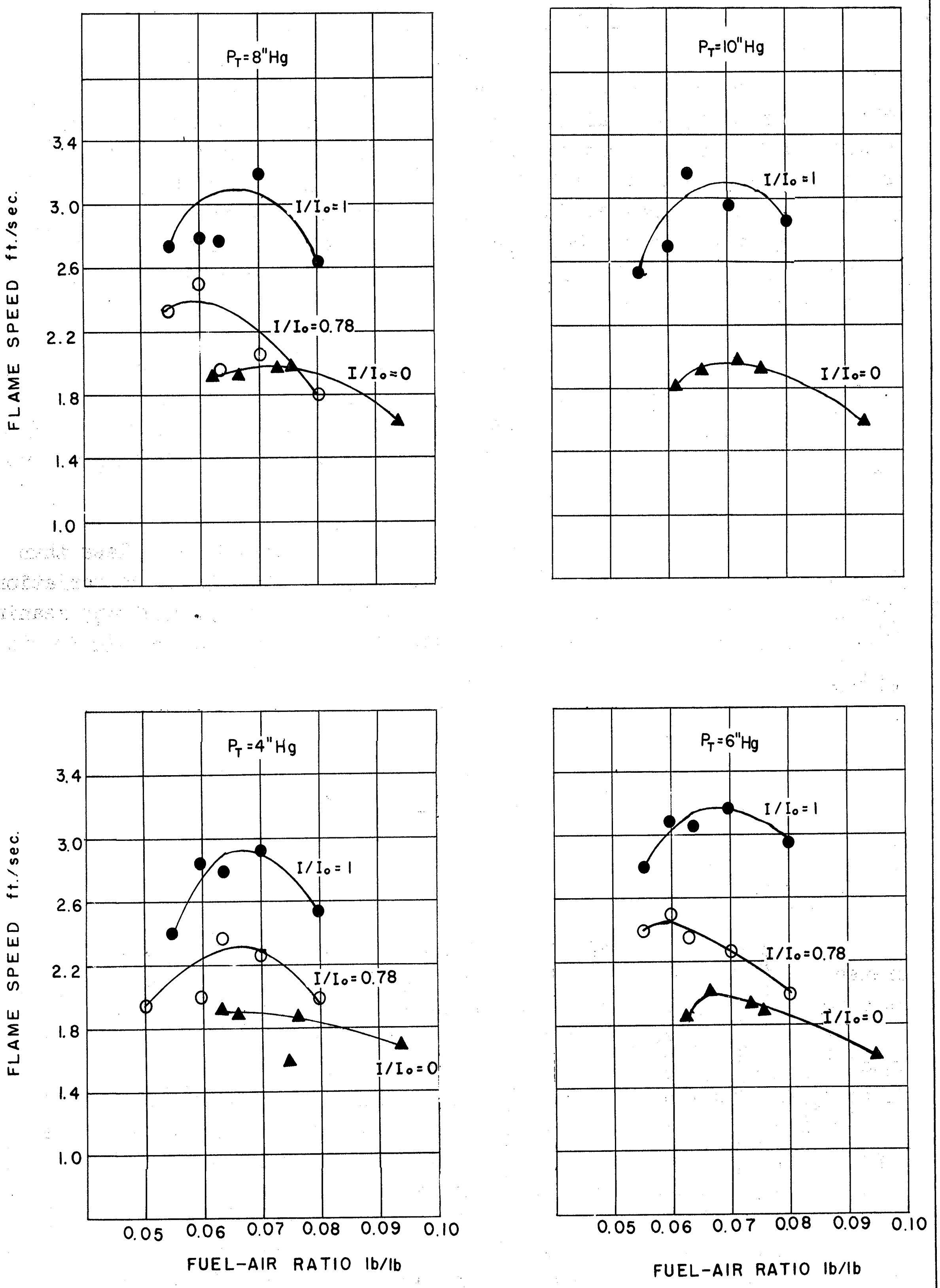


Fig. 12. Effect of fuel-air ratio on flame speed of irradiated flames

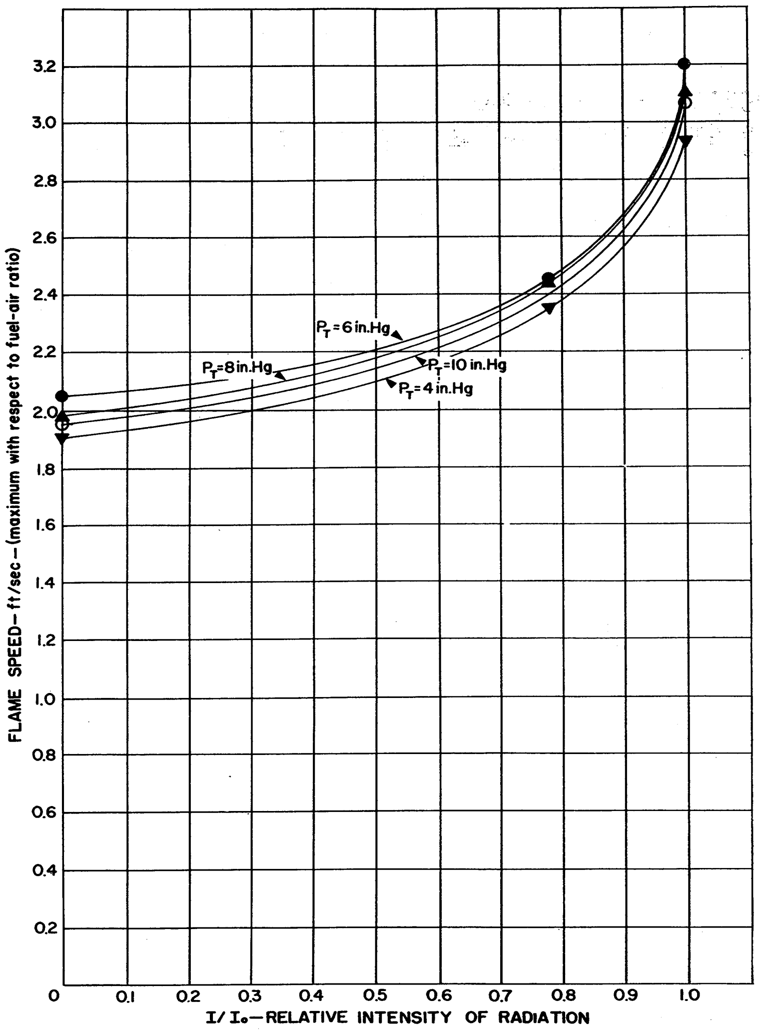


Fig. 13a. The effect of radiation on flame speed

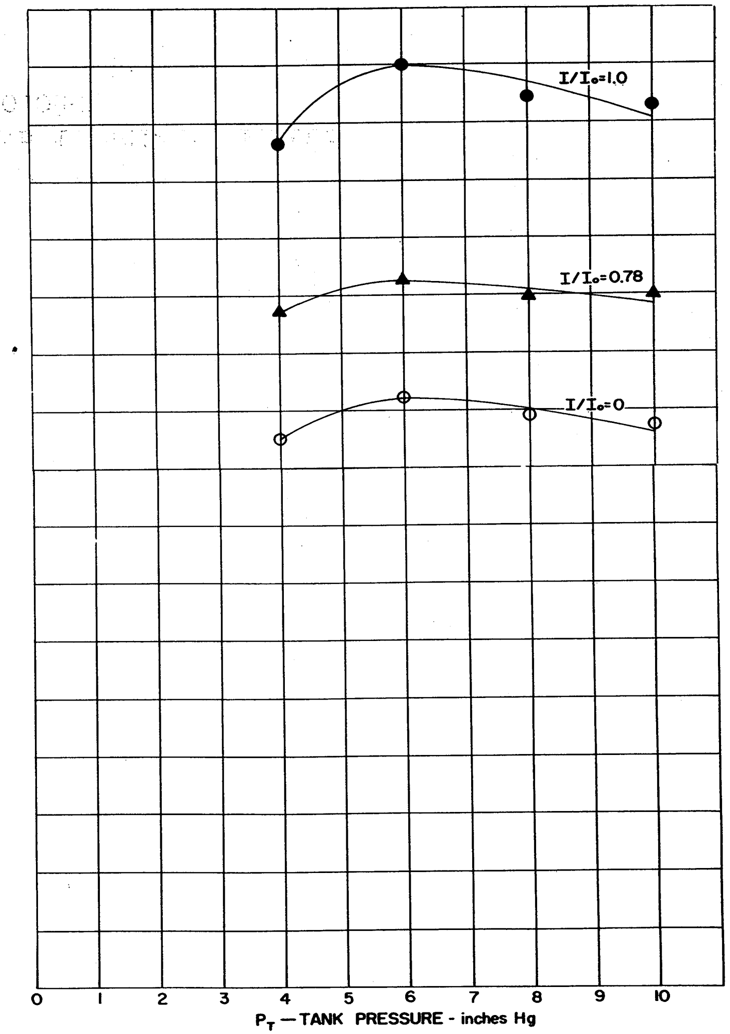


Fig. 13b. The effect of pressure on flame speed of irradiated flames

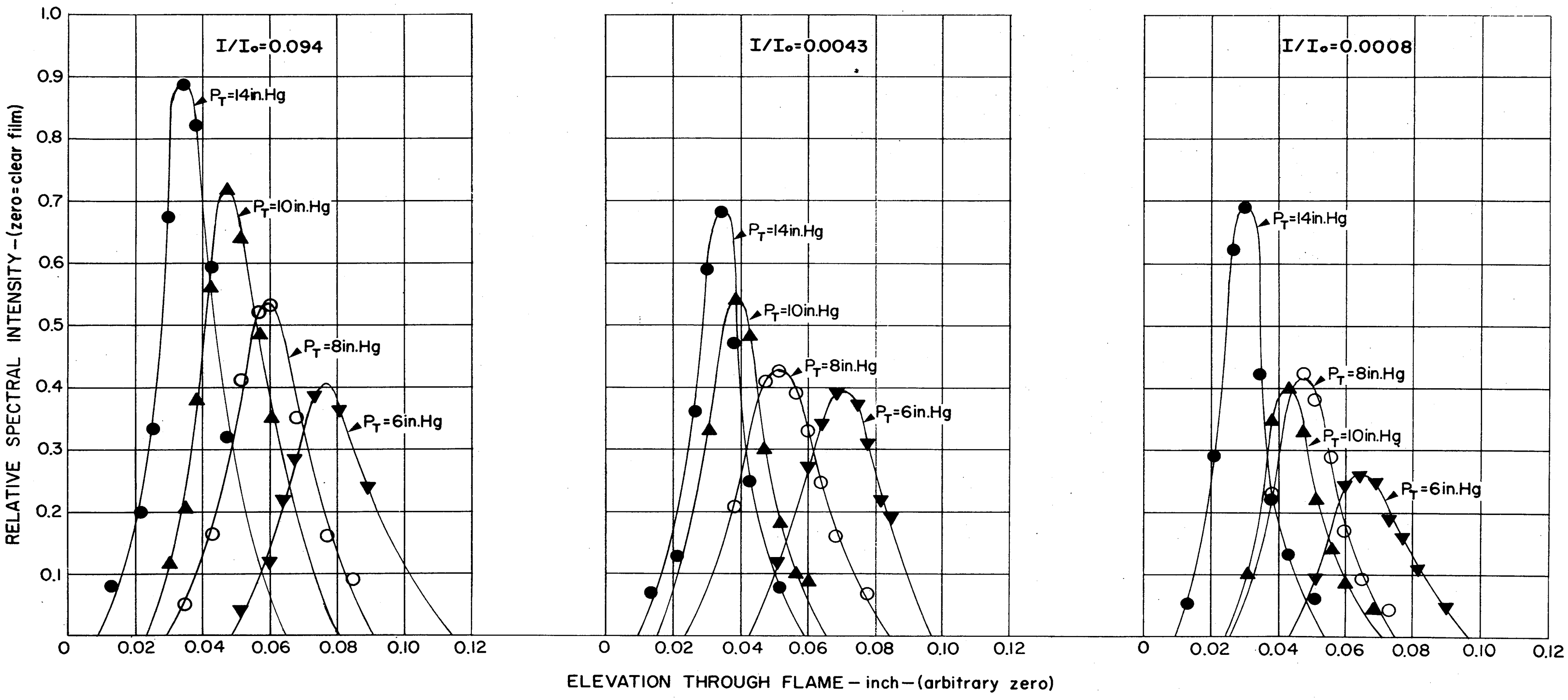


Fig. 14. Emission at 4315 Å within irradiated flames fuel/air = 0.0802 - 0.0883

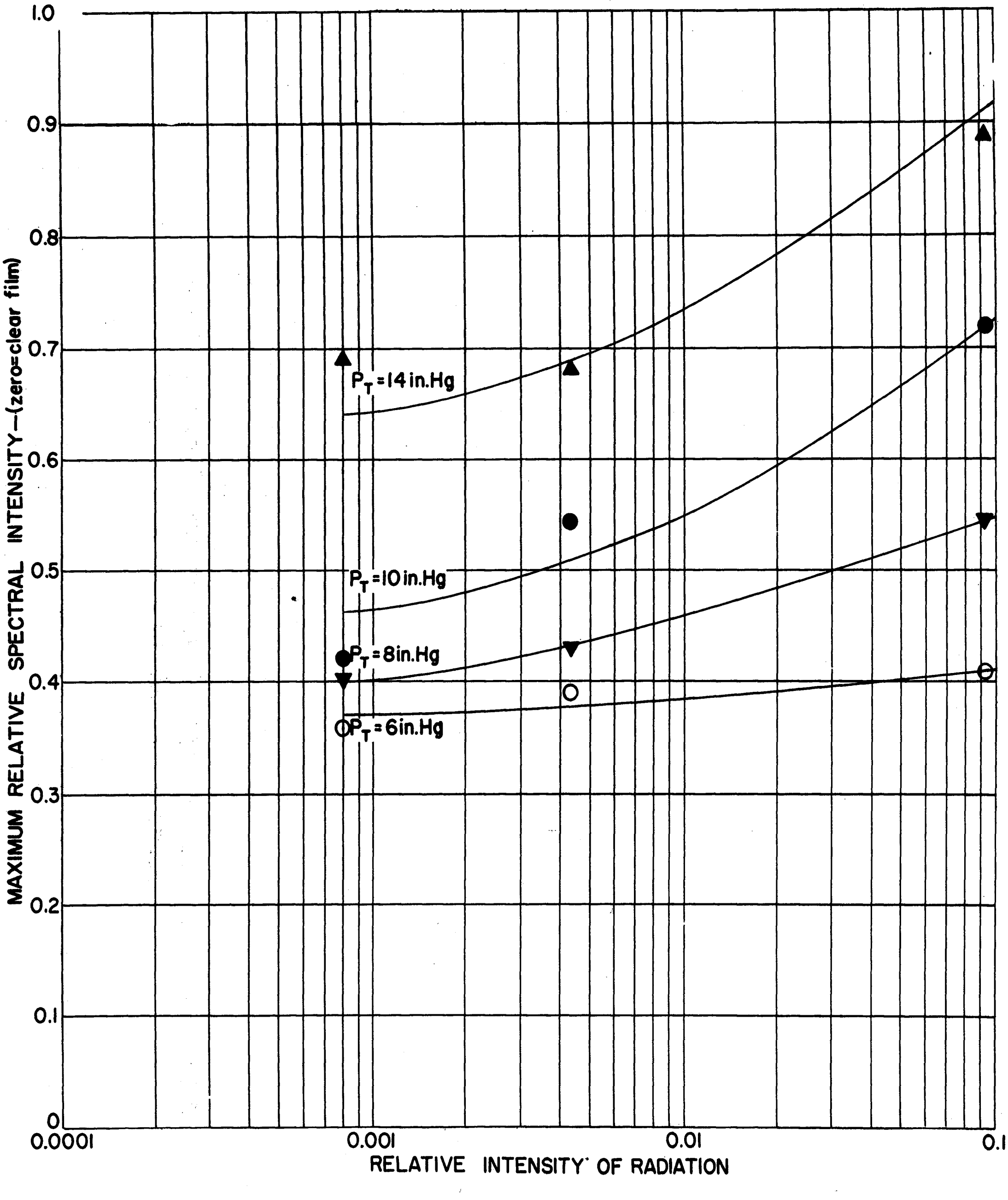


Fig. 15. The effect of radiation on maximum emission at 4315 Å
fuel/air = 0.0802 - 0.0883

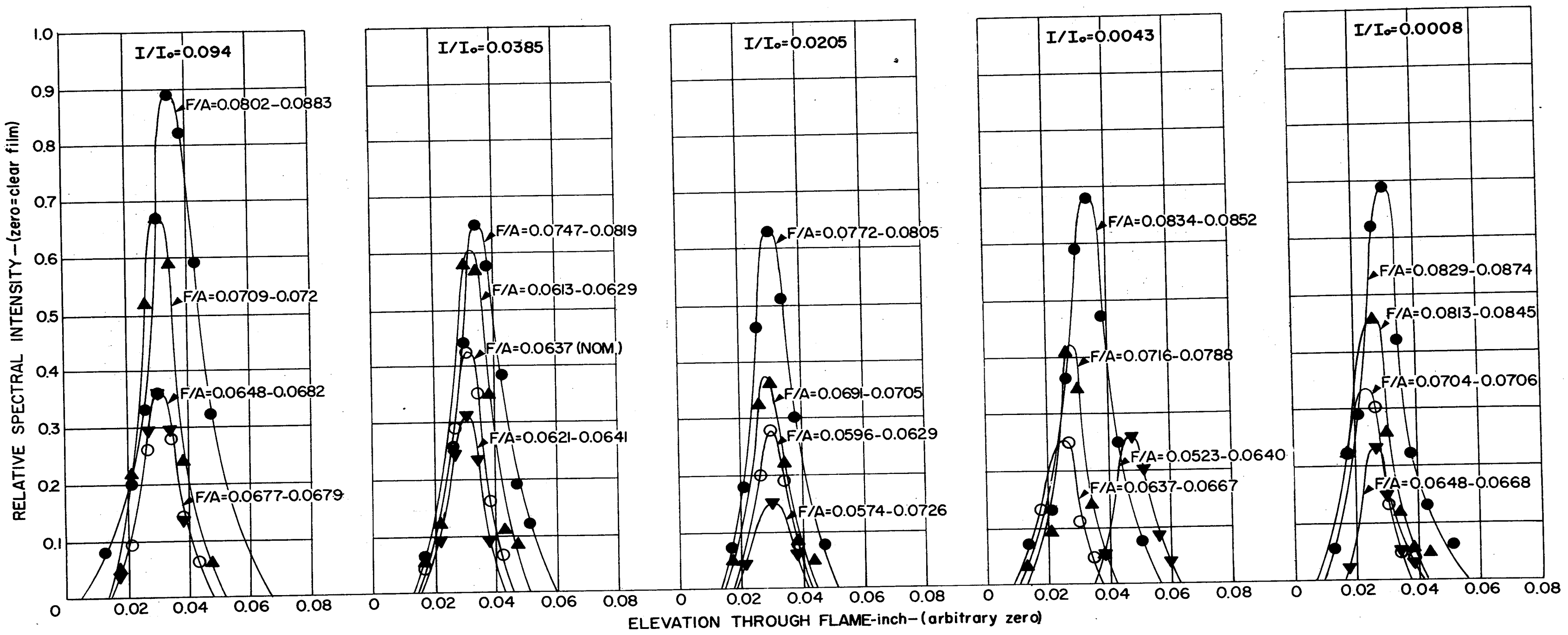


Fig. 16. Emission at 4315 Å within irradiated flames pressure = 14 in. Hg

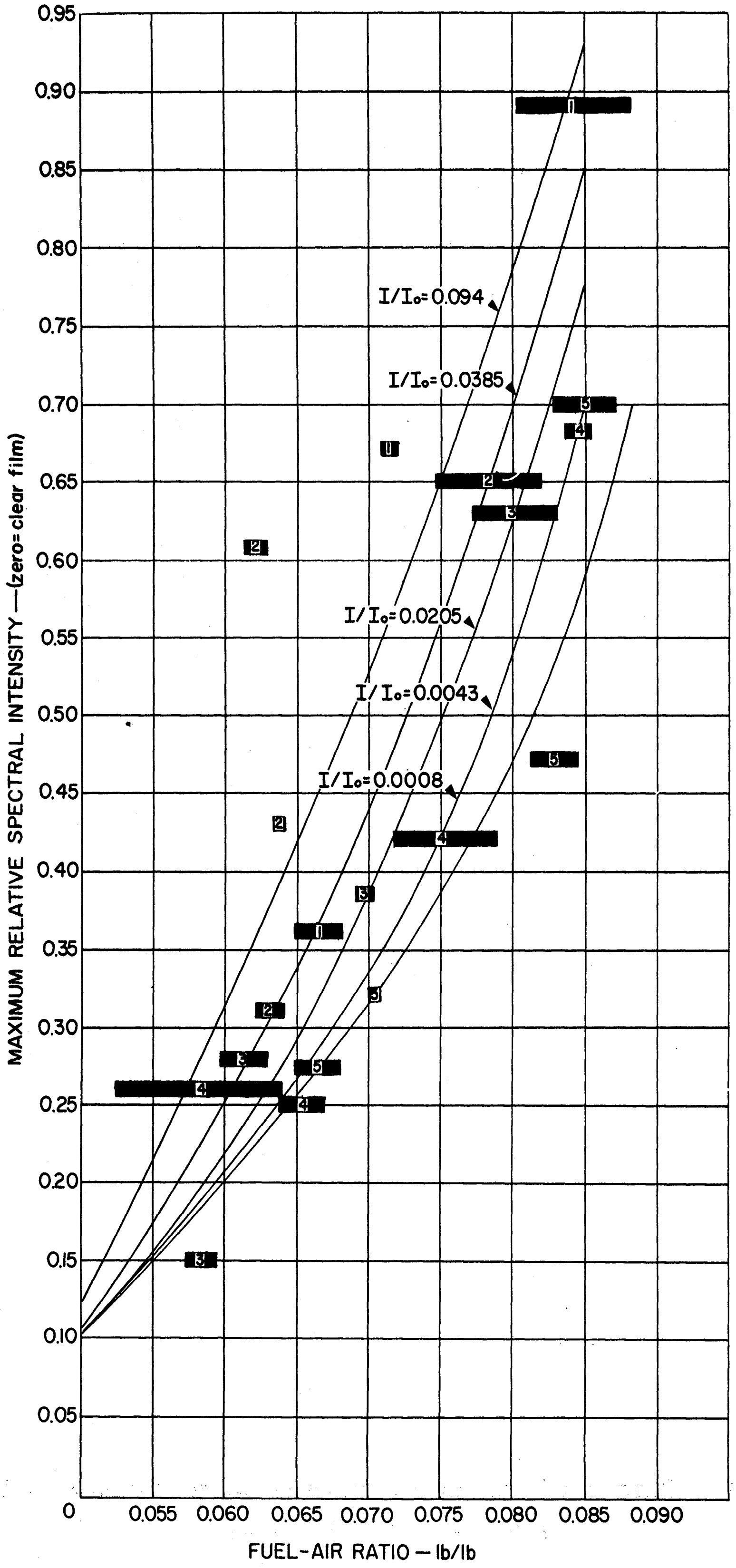


Fig. 17. The effect of fuel-air ratio on maximum emission at 4315 Å in irradiated flames pressure = 14 in. Hg

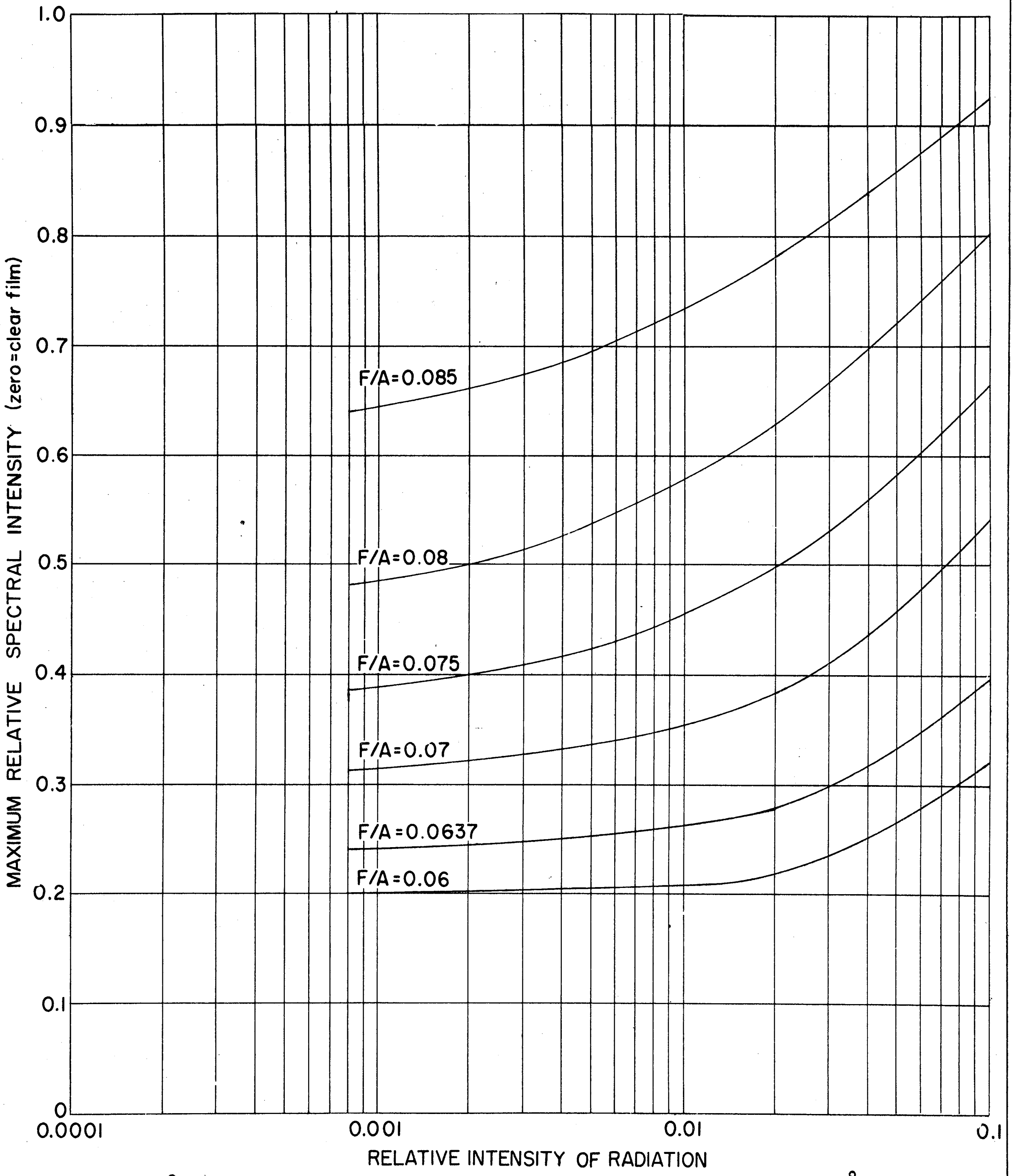


Fig. 18. Effect of radiation on maximum emission at 4315 Å
pressure = 14 in. Hg

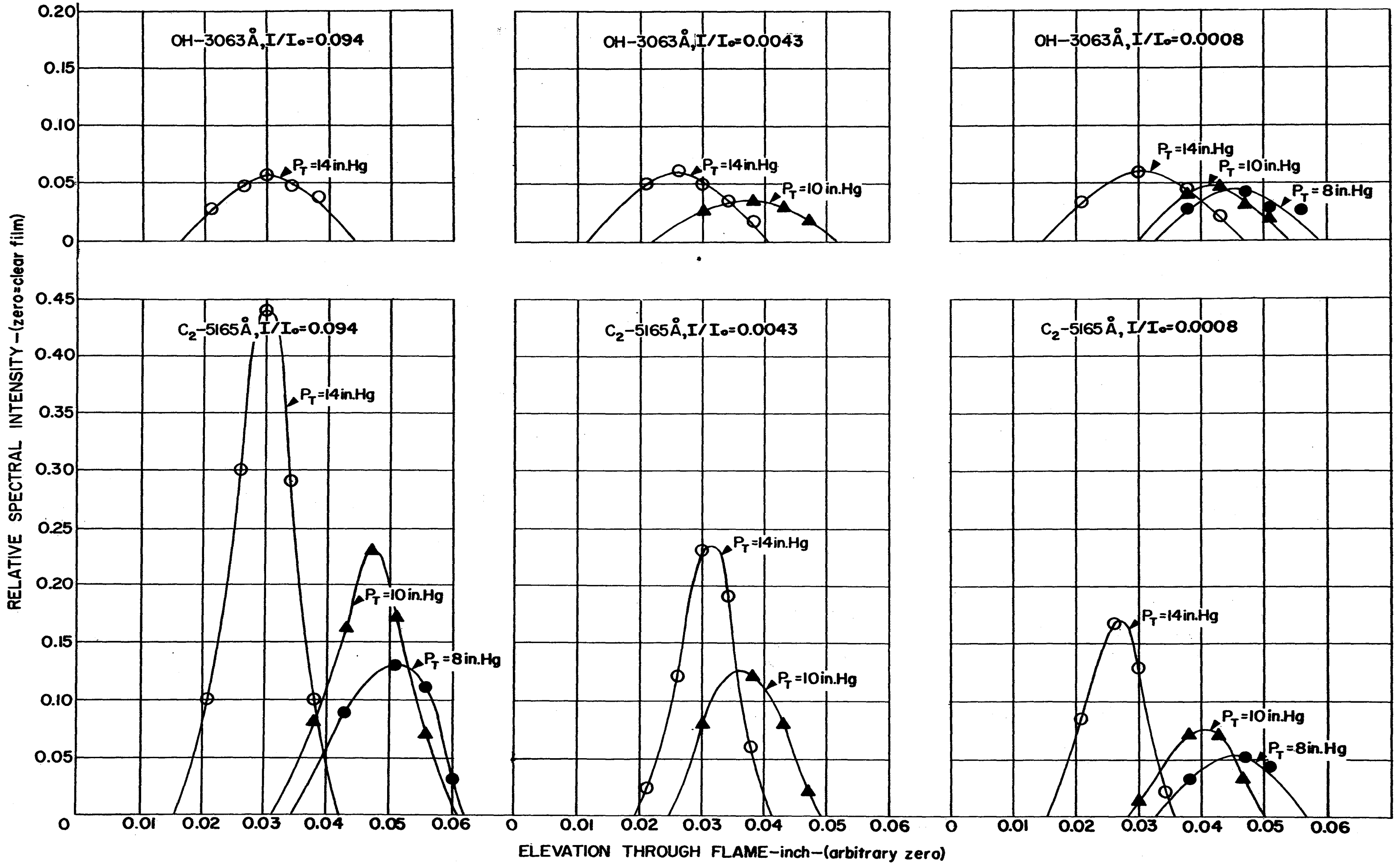


Fig. 19. Emission at 5165 Å and 3063 Å within irradiated flames fuel/air = 0.0802 - 0.0883

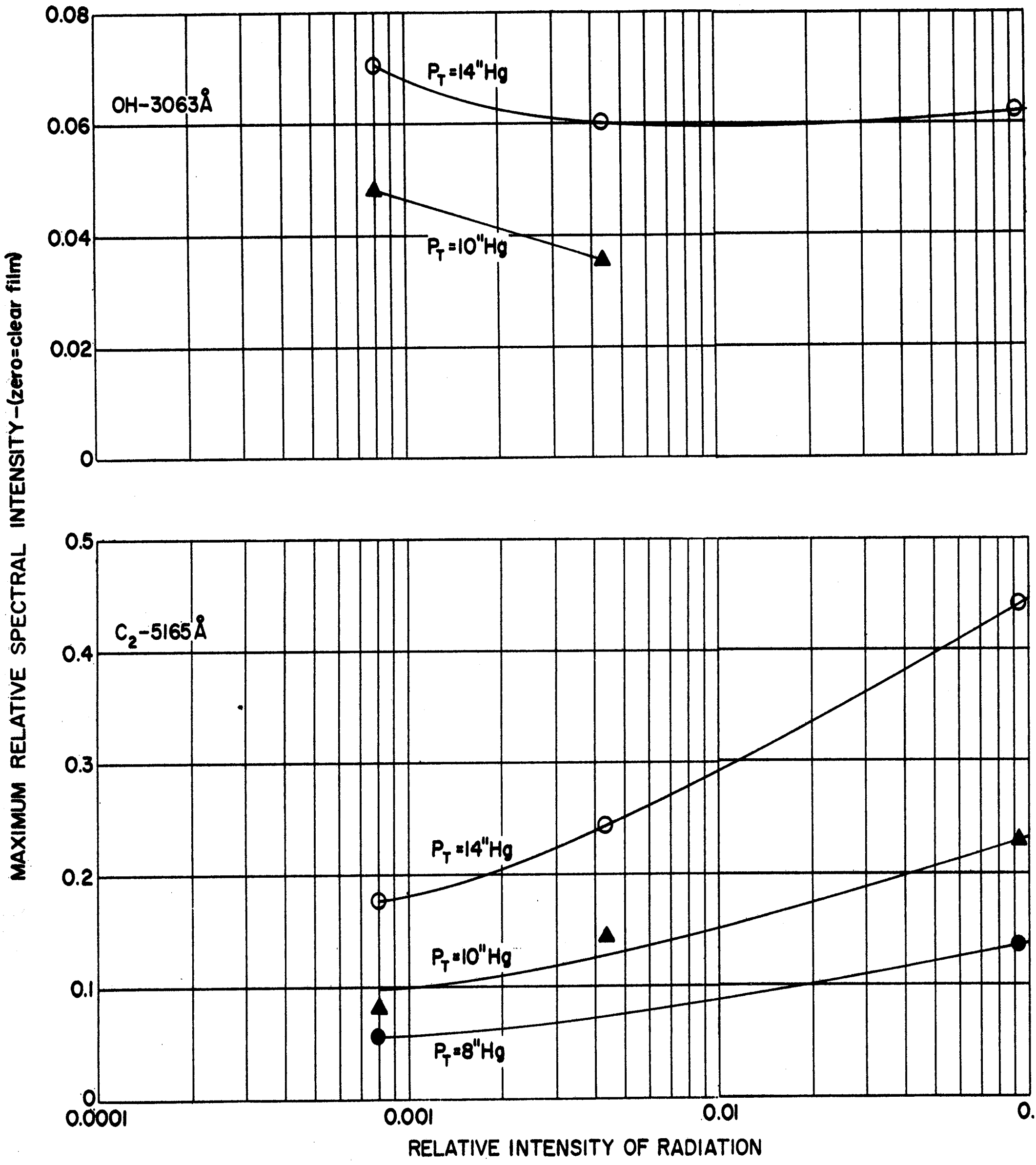


Fig. 20. The effect of radiation on maximum emission at 5165 Å and 3063 Å
fuel/air = 0.0802 - 0.0883

B. CH ROTATIONAL TEMPERATURES

The rotational temperature of the CH molecule may be computed from the spectroscopic data previously presented. The temperature is computed from the microphotometer tracings of the experimental spectrograms. A typical example of such a tracing showing the P, Q, and R branches is presented in Figure 21. The procedure for computation of the rotational temperature is outlined below.

Procedure for Computing Rotational Temperature of CH Molecules

The frequency of the distinct band heads were obtained from Pearse and Gaydon (11). The wave numbers, $\nu = 1/\lambda$, of these band heads were then determined and plotted against distance on the microphotometer plot using the 4315 Å line as ν_o. The resulting curve is shown in Figure 22. The nonlinearity of the correlation indicates that the frequency on the spectrograms is not a linear function of distance even though a grating spectrograph was used. This probably is due to the use of the plastic replica grating. However, in the region of interest for this calculation (the region surrounding the 4315 Å line) the curve can be represented by

$$\nu = 23,170 - 18.9\ s \tag{4}$$

where ν = wave number
s = distance from ν_o on microphotometer plot.

Equation (4) can be combined with the expression for the wave number in terms of the rotational quantum number K, as follows:

$$\nu = 23,170 - 18.9\ s = 23,170 + 29K + 0.10\ K^2 \tag{5}$$

where K is the rotational quantum number and the constants 29 and 0.10 are functions of the moment of inertia of the CH molecule. The quantum number K is a positive integer and varies by a unity from one state to the next consecutive one. K need only be determined for the first distinct peak in the R region of the bank, since the quantum number of each of the subsequent peaks is a unit higher than the preceeding peak. The distance s from ν_o for each of these peaks was computed and is tabulated below:

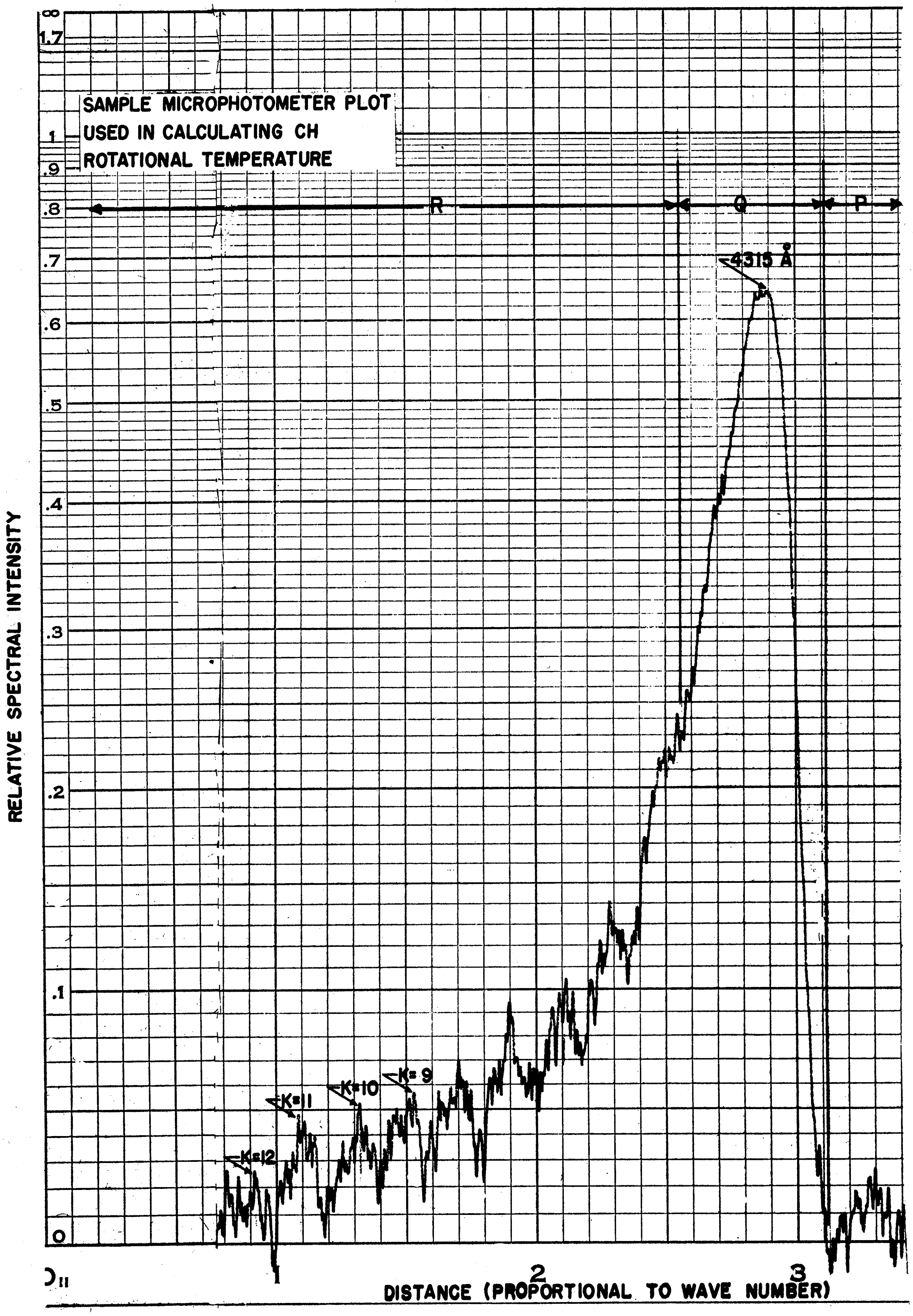


Fig. 21. P, Q, and R branches of a microphotometer tracing

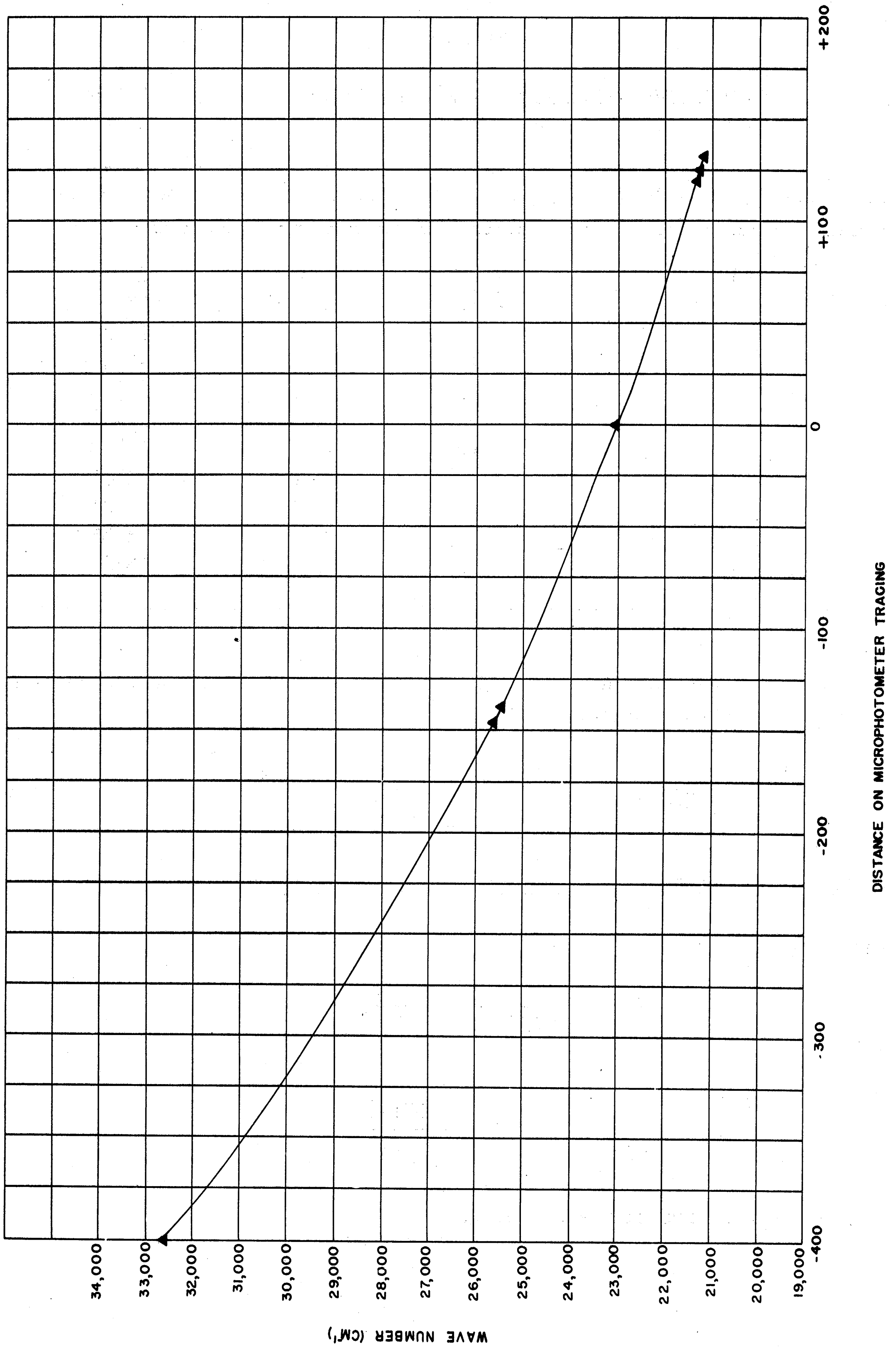


Fig. 22. Microphotometer calibration

TABLE III

POSITION AND WAVE NUMBER OF PEAKS IN MICROPHOTOMETER TRACING

Rotational Quantum Number K	Distance s	Wave Length λ, Å	Wave Number ν, 1/cm.
3	- 4.6	4300	23256
4	- 6.2	4294	23286
5	- 7.8	4289	23317
6	- 9.4	4283	23348
7	-11.0	4278	23378
8	-12.6	4272	23408
9	-14.2	4267	23438
10	-15.8	4261	23469
11	-17.5	4255	23501
12	-19.2	4249	23533
13	-20.8	4244	23563

The relative spectral intensity I(K) at any peak is related to the quantum number K as shown in Equation (6).

$$I(K) = \left(\frac{K^2}{2K-1} + \frac{K(K-2)}{2K-3}\right) e^{-BK(K+1)\frac{h_c}{kT}} \qquad (6)$$

where $B = 14.5\ \mathrm{Cm}^{-1}$ for CH molecule and $h_c/k = 1.43$.

Defining F(K) as

$$F(K) = \frac{K^2}{2K-1} + \frac{K(K-2)}{2K-3}$$

Equation (6) becomes

$$I(K) = F(K)\, e^{-\frac{B\,K(K+1)h_c}{kT}} \quad . \qquad (7)$$

F(K) and K(K+1) are tabulated below:

TABLE IV

QUANTUM FUNCTIONS

K	F(K)	K(K+1)
3	.800	12
4	.686	20
5	.634	30
6	.607	42
7	.587	56
8	.574	75
9	.565	90
10	.557	110
11	.551	132
12	.547	156
13	.544	182

For a given spectrographic exposure, F(K)/I(K) was obtained and $\ln\left[F(K)/I(K)\right]$ was plotted against K(K+1). The slope of this curve was obtained and equated to Bh_c/kT, i.e.,

$$\text{slope} = m = \frac{14.5 \times 1.45}{T}$$

where T = degrees Kelvin

in order to obtain a value of the rotational temperature of CH for that particular spectrographic exposure. A typical plot of $\ln\left[F(K)/I(K)\right]$ versus K(K+1) is shown in Figure 23. The data for other such plots are tabulated in the Appendix.

The plot is not linear at the low quantum numbers but becomes very close to a straight line at the higher quantum numbers. For this reason only quantum numbers above 8 were used in determining the slope by least squares. Gaydon and Wolfhard (12)found a similar nonlinearity in determining rotational temperatures of OH molecules in low-pressure flames.

Computed CH Rotational Temperatures

The CH rotational temperature is plotted versus distance through the flame in Figure 24. The maximum temperatures were then plotted versus

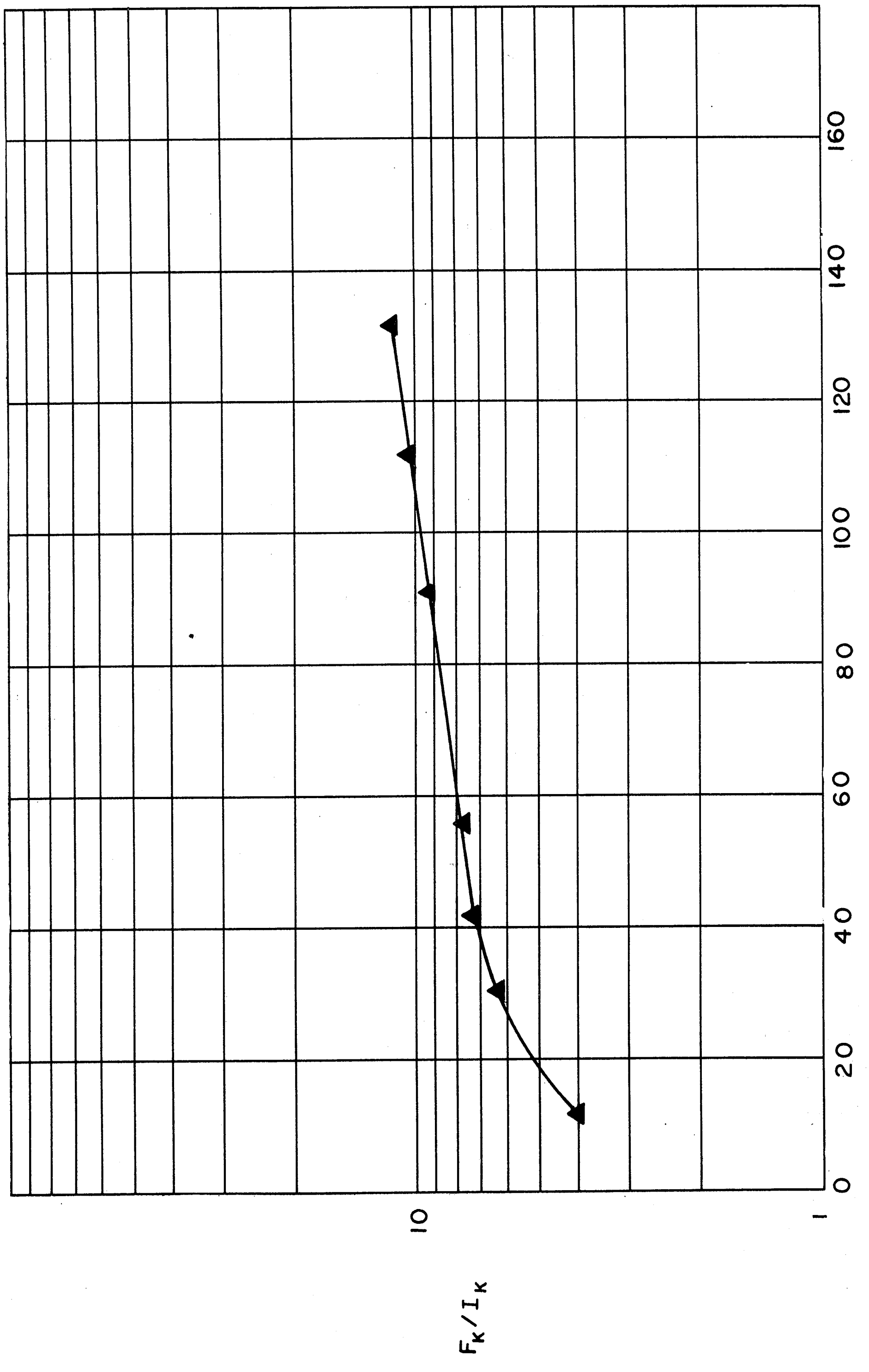


Fig. 23. Spectral intensity as a function of quantum number

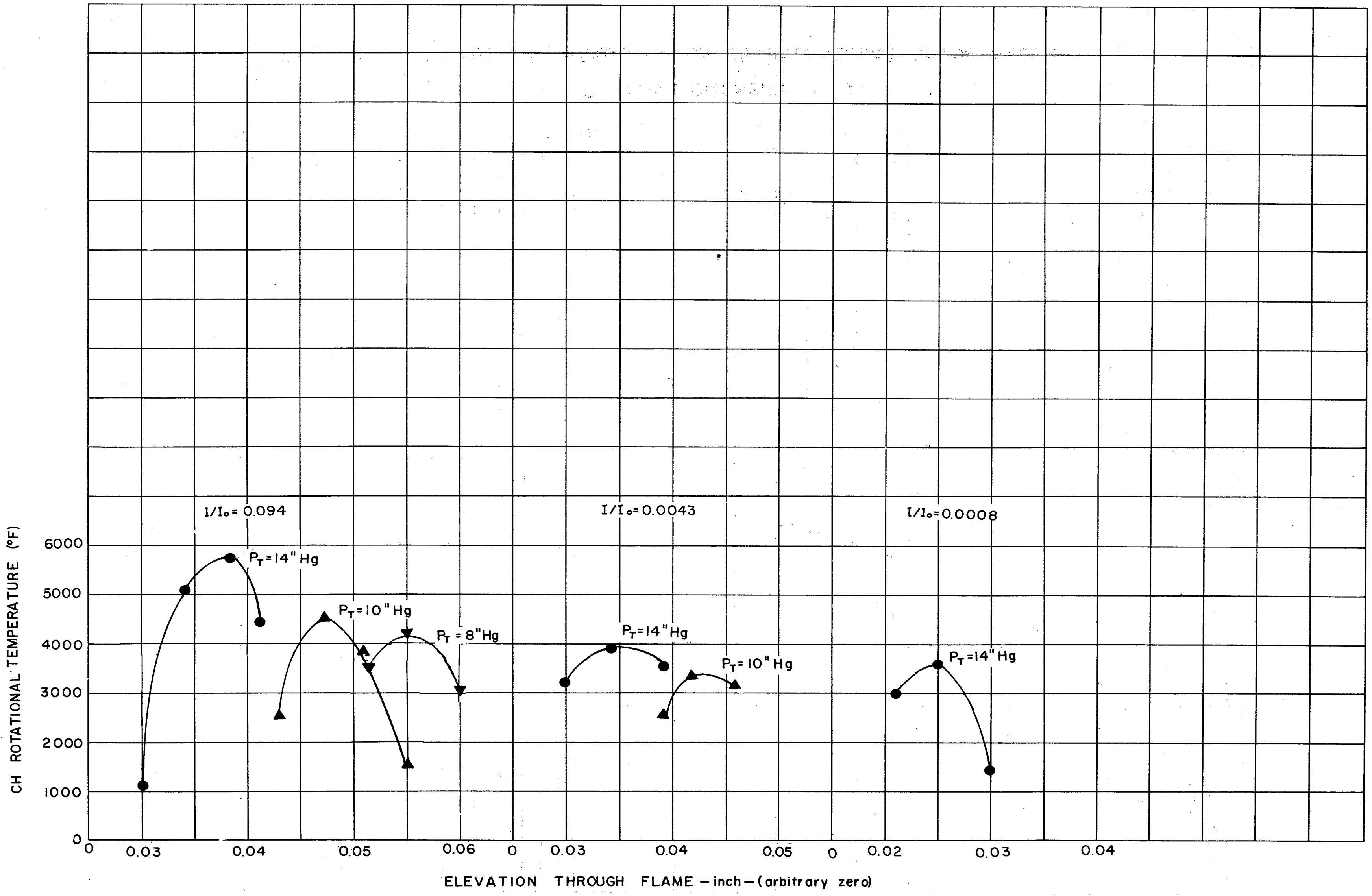


Fig. 24. CH rotational temperature within an irradiated flame

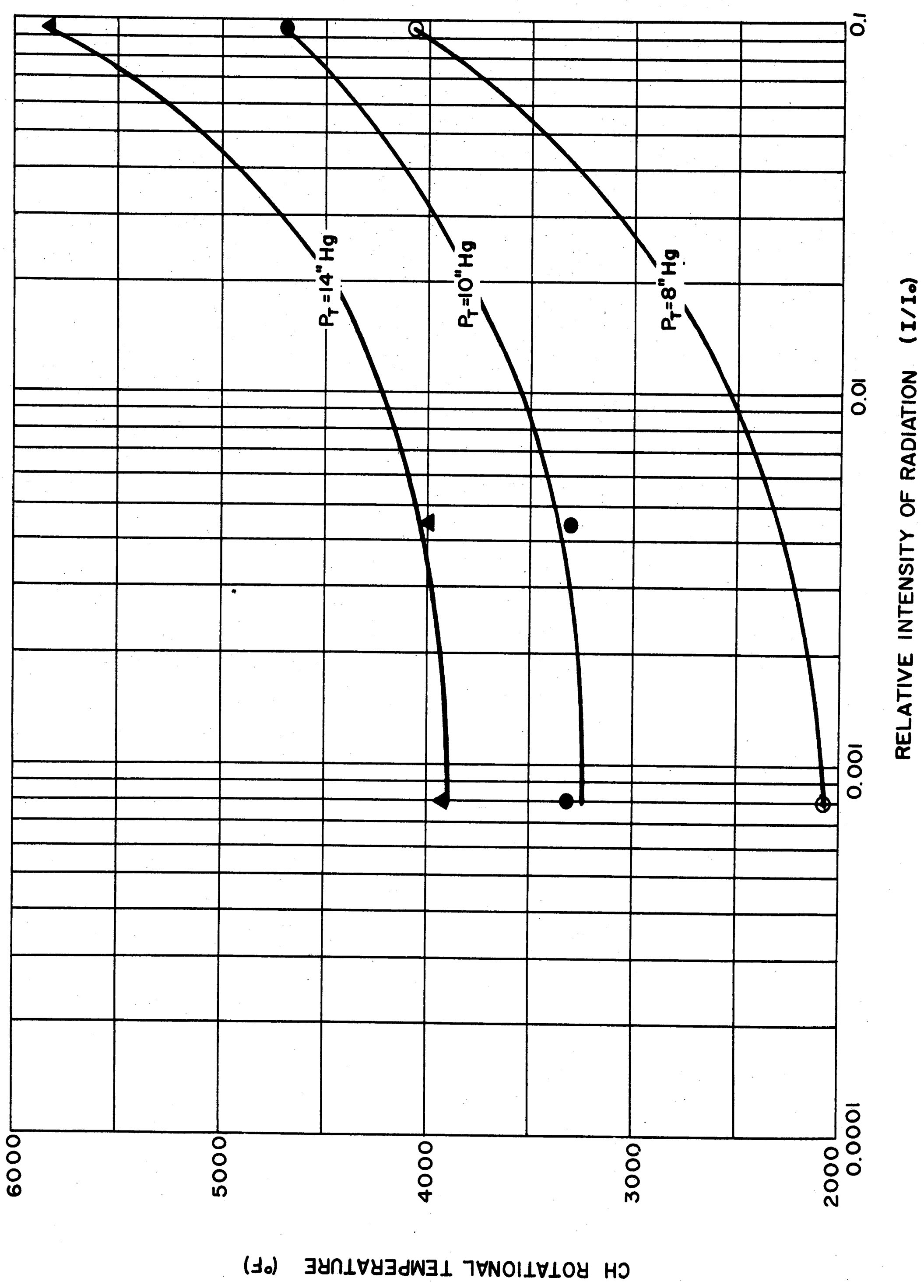


Fig. 25. Effect of radiation on CH rotational temperature

relative intensity of radiation at several pressures, as shown in Figure 25. The temperature was not plotted as a function of fuel-air ratio because of inability to read the relative spectral intensity, I(K), with sufficient accuracy. This was also true of the data obtained at pressures of 6 and 8 in. Hg at a relative intensity of radiation of 0.0008. The values of I(K) are particularly difficult to obtain when the intensity of the light emitted in the 4315 Å band is low.

In Figure 25 it can be seen that the rotational temperature increases with an increase in relative intensity of radiation. This suggests that the CH molecules are more energetic at higher intensities of radiation. This should result in a more rapid rate of reaction with oxygen and perhaps explains the increase in flame speed. At a pressure of 14 in. Hg abs., the CH rotational temperature at the optimum location in the flame was increased 47 per cent by increase in intensity of radiation from 12 curies to 1500 curies. At a pressure of 10 in. Hg abs., the CH rotational temperature is increased by 42 per cent, while at a pressure of 8 in. an increase of 95 per cent was observed for the same increase in radiation. Quantitative significance should not be attached to these percentages due to the limited data.

C. SPECTRAL INTENSITIES

It is seen from Figures 14, 16, and 19 that the intensities of the three bands are almost symmetrical about the elevation at which the individual maxima occur. This indicates that the maximum rate of reaction occurs midway between the initiation of reaction and the end of reaction, and seems to have no other significance. However, Figures 14 and 19 do show a definite decrease in the intensity with decrease in pressure, indicating a decrease in the quantities of CH, C_2, and OH, respectively, with decrease in pressure; and a shift of the maximum with pressure, indicating that at lower pressures the reaction occurs farther away from the screen. Figure 16 indicates a decrease in the intensity of CH with decrease in fuel-air ratio, which is to be expected because of the decrease in the quantity of fuel present. The elevation at which the maximum intensity of the CH band occurs does not appear to be much of a function of the fuel-air ratio.

It can be seen in Figures 15 and 20 that the maximum intensities due to CH and C_2 increase with radiation and also with pressure. Another interesting fact is also illustrated; the slopes of the curves are greater at the higher pressures. This means that the radiation has a greater effect on emission at higher pressures than at lower pressures, which is to be expected since the greater density of gas at the higher pressures results in a greater probability of beta-ray capture. At 6 in. Hg, the radiation has very little effect on the spectral intensity. The percentage increase in intensity due to CH at 1500 curies over that at 12 curies ranges from about

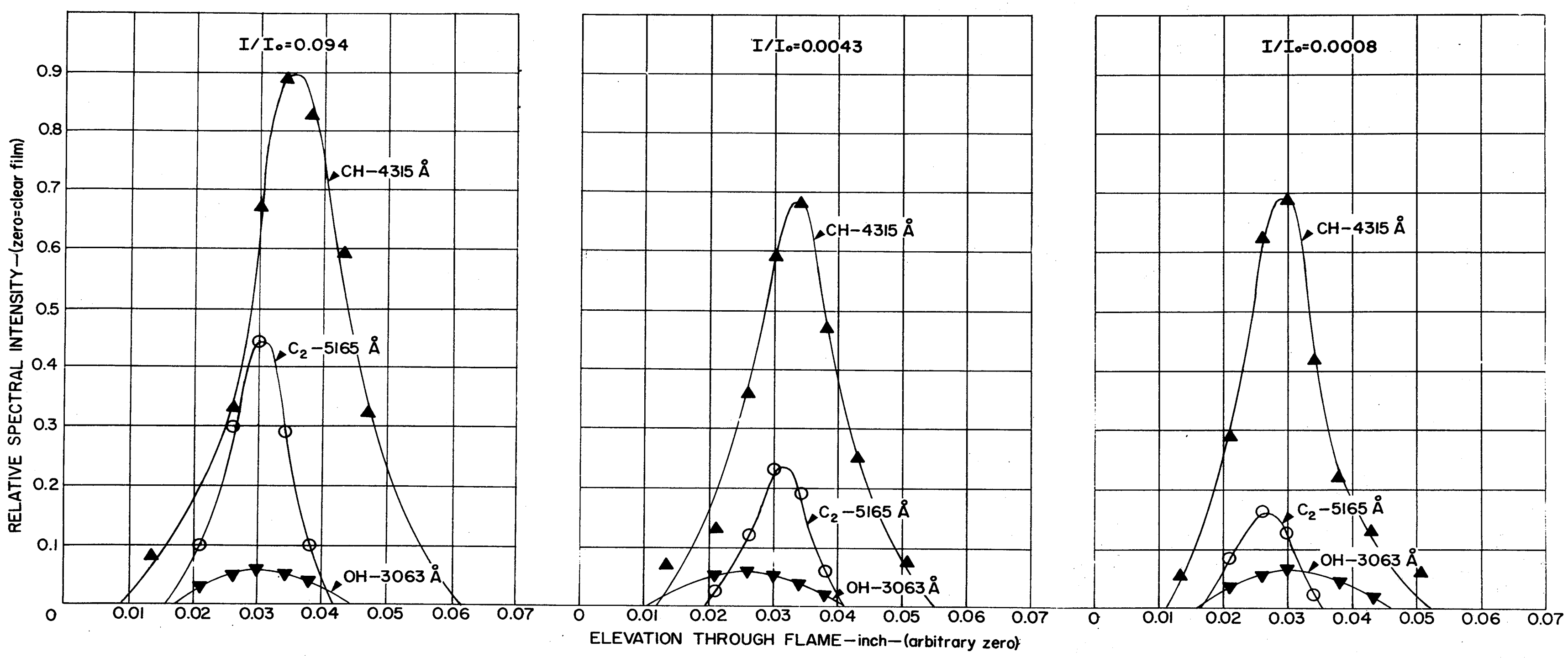


Fig. 26. Comparison of emission at 4315 Å, 5165 Å, and 3063 Å within irradiated flames pressure = 14 in. Hg, fuel/air = 0.0802 - 0.0883

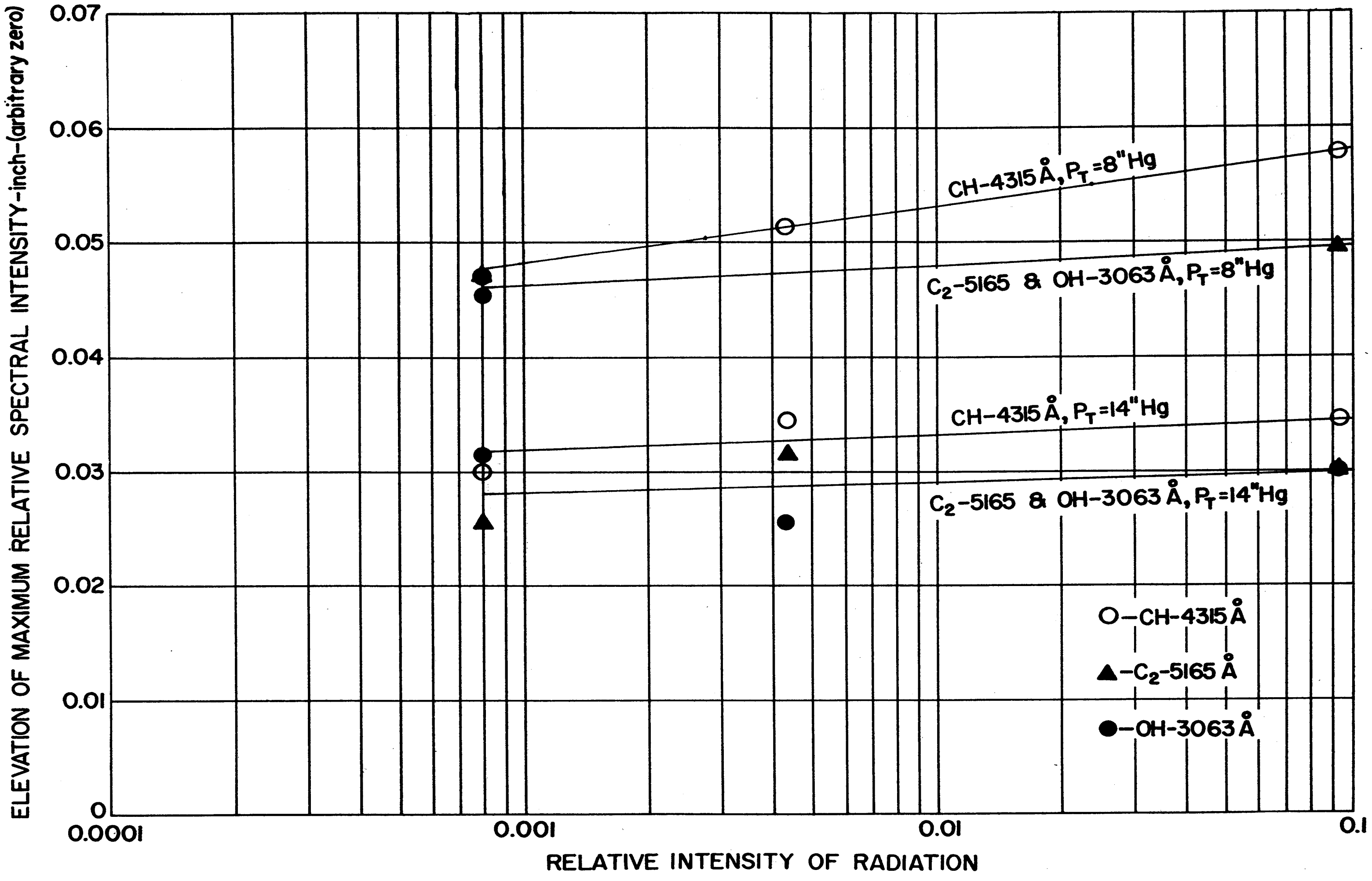


Fig. 27. Effect of radiation on location of maximum emission at 4315 Å, 5165 Å, and 3063 Å

14 per cent at 6 in. Hg to about 33 per cent at 14 in. Hg. The corresponding increase in intensity due to C_2 ranges from about 145 per cent at 8 in. Hg to about 152 per cent at 14 in. Hg.

Figure 17 illustrates more clearly than Figure 16 that the quantity of CH present decreases very abruptly with decrease in fuel-air ratio. Figure 18, which is a cross plot of Figure 17, shows the increase in the intensity due to CH with an increase in radiant intensity at various fuel-air ratios. The slopes indicate a greater effect due to radiation at higher fuel-air ratios than at lower ratios. This is to be expected since more fuel molecules or radicals are present to capture beta rays. The per cent increase in spectral intensity at 1500 curies over that at 12 curies ranges from about 57 per cent at a fuel-air ratio of 0.06 to about 68 per cent at a fuel-air ratio of 0.08.

In order to compare the data for the three different species, the spectral intensities at 4315 Å, 5165 Å, and 3063 Å were plotted versus elevation through the flat flame at a constant pressure of 14 in. Hg abs. and a nearly constant fuel-air ratio of 0.085 for three relative intensities of radiation (0.094, 0.0043, and 0.0008). These plots are shown in Figure 26.

The maxima in the spectral intensities occur at different elevations for the CH, C_2, and OH. The elevation in the flame at which the maxima occurred are plotted versus relative intensity of radiation at various pressures and constant fuel-air ratio in Figure 27. Contrary to expectations, the maxima in the spectral intensities are shifted away from the burner head by the beta radiation. Furthermore, the maximum intensities due to C_2 and OH occur at the same elevation, followed by the maximum intensity due to CH at a higher elevation within the flame. This lends support to the theory that carbon monoxide is formed in flames from the reaction

$$C_2 + OH \rightleftarrows CH + CO$$

PART VII. SUMMARY AND CONCLUSIONS

The influence of nuclear radiation from Au-198 and Au-199 on propane air flames at low pressure was investigated experimentally. The intensity of light emission by CH, C_2, and OH molecules was measured, and the rotational temperature of the CH molecule was computed as a function of distance through the flame. Flame speeds were also measured. The following effects were observed, presumably due to beta rather than gamma radiation.

1. Rates of flame propagation determined from photographs of bunsen flames were increased up to 50 per cent by nuclear radiation from the 12000-curie source.

2. The intensity of radiation at 4315 Å due to CH was increased by 14 per cent at 6 in. Hg abs. and up to 33 per cent at 14 in. Hg abs. by nuclear radiation at a level of 1500 curies.

3. The intensity of radiation at 5165 Å due to C_2 was increased up to 150 per cent by nuclear radiation at a level of 1500 curies.

4. The intensity of radiation at 3063 Å due to OH was not affected significantly by nuclear radiation at levels up to 1500 curies.

5. Increases in the CH-rotational temperature ranging from 50 to 100 per cent were calculated from a limited amount of experimental data obtained at source strengths up to 1500 curies.

6. Under the influence of radiation, the maxima in the emission due to C_2 and OH were observed to occur at an earlier stage of the flame than the maximum of emission due to CH. This lends support to the theory that carbon monoxide is produced in flames by the reaction

$$C_2 + OH = CH + CO$$

APPENDIX

TABLE V

FUNCTIONS FOR CALCULATION OF CH-ROTATIONAL TEMPERATURES

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.094, F/A = 0.0802 - 0.0883, Pressure = 14 psia			
Elevation = 0.030 in.			
9	0.073	7.74	90
10	0.062	8.99	110
11	0.043	12.82	132
12	0.046	12.10	156
13	0.024	22.61	186
T = 1185°K			
Elevation = 0.034 in.			
9	0.124	4.55	90
10	0.095	5.87	110
11	0.083	6.64	132
12	0.072	7.59	156
13	0.062	8.75	186
T = 3077°K			
Elevation = 0.038 in.			
9	0.110	5.13	90
10	0.098	5.69	110
11	0.078	7.07	132
12	0.066	8.28	156
13	0.061	8.90	182
T = 3379°K			

TABLE V (cont.)

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.094, F/A = 0.0802 - 0.0883, Pressure = 14 psia			
Elevation = 0.043 in.			
9	0.057	9.91	90
10	0.046	12.11	110
11	0.039	14.14	132
12	0.028	19.52	156
13	0.028	19.38	182
T = 2669°K			
I/I_o = 0.094, F/A = 0.0802 - 0.0883, Pressure = 10 psia			
Elevation = 0.043 in.			
9	0.054	10.46	90
10	0.041	13.59	110
11	0.034	16.22	132
12	0.024	22.78	156
T = 1659°K			
Elevation = 0.047 in.			
9	0.076	7.43	90
10	0.061	9.14	110
11	0.050	11.03	132
12	0.048	11.39	156
T = 2841°K			
Elevation = 0.051 in.			
9	0.058	9.74	90
10	0.052	10.72	110
11	0.040	13.79	132
12	0.020	27.73	156
T = 2406°K			

TABLE V (cont.)

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.094, F/A = 0.0802 - 0.0883, Pressure = 10 psia			
Elevation = 0.056 in.			
9	0.047	12.07	90
10	0.040	13.93	110
11	0.025	22.06	132
12	0.015	36.44	156
T = 1057°K			
I/I_o = 0.094; F/A = 0.0802 - 0.0883, Pressure = 8 psia			
Elevation = 0.051 in.			
9	0.033	17.11	90
10	0.031	17.98	110
11	0.022	24.85	132
12	0.020	27.33	156
T = 2351°K			
Elevation = 0.056 in.			
9	0.052	10.86	90
10	0.047	11.86	110
11	0.042	13.13	132
12	0.036	15.18	156
13	0.023	23.59	182
T = 2610°K			
Elevation = 0.060 in.			
9	0.062	9.11	90
10	0.059	9.45	110
11	0.031	17.79	132
12	0.024	22.78	156
13	0.024	22.61	186
T = 1994°K			

TABLE V (cont.)

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.0043, F/A = 0.0834 - 0.0852, Pressure = 14 psia			
Elevation = 0.030 in.			
9	0.070	8.07	90
10	0.052	10.7	110
11	0.041	13.4	132
12	0.038	14.4	156
T = 2103°K			
Elevation = 0.034 in.			
9	0.10	5.65	90
10	0.073	7.63	110
11	0.059	9.35	132
12	0.059	9.26	156
T = 2448°K			
I/I_o = 0.0043, F/A = 0.0834 - 0.0852, Pressure = 10 psia			
Elevation = 0.038 in.			
9	0.032	17.6	90
10	0.025	22.3	110
11	0.019	29.0	132
T = 1714°K			
Elevation = 0.043 in.			
9	0.036	15.7	90
10	0.028	19.9	110
11	0.024	23.0	132
12	0.020	27.3	156
T = 2230°K			

TABLE V (cont.)

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.0043, F/A = 0.0834 - 0.0852, Pressure = 10 psia			
Elevation = 0.047 in.			
9	0.024	23.5	90
10	0.026	21.4	110
11	0.019	29.0	132
12	0.014	39.0	156
T = 2153°K			
I/I_o = 0.0008, F/A = 0.0829 - 0.0874, Pressure = 14 psia			
Elevation = 0.038 in.			
9	0.040	14.1	90
10	0.030	18.6	110
11	0.026	21.2	132
12	0.021	26.0	156
T = 1957°K			
Elevation = 0.043 in.			
9	0.051	11.1	90
10	0.037	15.1	110
11	0.032	17.2	132
12	0.028	19.5	156
T = 2400°K			
Elevation = 0.047 in.			
9	0.042	13.4	90
10	0.030	18.6	110
11	0.026	21.2	132
12	0.028	19.5	156
T = 1000°K			

TABLE V (concl.)

K	I(K)	I(K)/F(K)	K(K+1)
I/I_o = 0.0008, F/A = 0.0829 - 0.0874, Pressure = 10 psia			
Elevation = 0.030 in.			
9	0.10	5.6	90
10	0.08	7.0	110
11	0.065	8.5	132
12	0.060	9.1	156
T = 2250°K			
I/I_o = 0.0008, F/A = 0.0829 - 0.0874, Pressure = 8 psia			
Elevation = 0.030 in.			
9	0.026	21.7	90
10	0.021	26.5	110
11	0.015	36.8	132
12	0.011	49.7	156
T = 1450°K			

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