STUDIES OF OH, CO, CH, AND C₂ RADIATION FROM LAMINAR AND TURBULENT PROPANE-AIR AND ETHYLENE-AIR FLAMES

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Summary

OH, CO, CH, and C₂ radiations from propane-air and ethylene-air flames were isolated with a monochromator, and the OH, CH, and C₂ emitters were purified by subtracting the unwanted CO background. The changes in intensity with changing equivalence ratio were determined for each emitter. The ratio of the C₂ and the CH radiation intensities was found to be a good index of the equivalence ratio of the flame, but the C₂-CH ratio was good only for the fuel for which it was calibrated.

Radiation ratios were measured in different regions of laminar and turbulent ethylene-air flames. No changes were found in laminar flames. Turbulent open flames burning on tubes were found to become leaner near the base, but in the turbulent brush they appeared to be burning at the metered equivalence ratio. No appreciable mixing with secondary air was detected in the turbulent brush, even at 125 feet per second, the maximum average gas-flow velocity obtainable.

The amount of radiation given off by the flame per unit rate of fuel consumption decreased with increasing average gas-flow velocities and Reynolds numbers. This decrease could not be accounted for by admixing of the secondary air with the turbulent flame brush.

Introduction

In recent years attention has been focused on theories of turbulent-flame structure and reaction mechanism. Experimental verification of these theories has been difficult to obtain because of the high temperatures, high gas velocities, and rapid reaction rates existing in turbulent flames. Probing techniques have been tried, but they are fraught with experimental pitfalls. Because of the generally unsatisfactory nature of the probing techniques in their current state of development, other methods of studying flame structure and reaction mechanism have
been considered. One of the most promising of these is the study of the radiation emitted by a flame.

The radiation from small, nonluminous propane-air flames seated on bench-scale open-tube burners has been measured with a photomultiplier tube through blue and yellow filters (ref. 1). At a given equivalence ratio, the radiation intensity was found to increase linearly with fuel-flow rate in both the laminar and the low-intensity turbulent flames. When the laminar flame changed to a turbulent brush flame at the higher gas-flow velocities, no discontinuity was observed in the relation between radiation intensity and fuel flow. In addition, the ratios of the intensities measured through the two filters were found to be a function of the equivalence ratio.

The filters used in the work of reference 1 were wide-band-pass elements that transmitted band spectra from more than one diatomic emitter. In addition, a background continuum of CO radiation was transmitted by the filters. Recently, a number of papers dealing with flame radiation have appeared (refs. 2 to 10), some of which contain descriptions of improved measuring techniques. Reference 2, for example, describes the use of a quartz spectrograph to isolate the fine-line-band spectra of the OH, CH, and C2 diatomic emitters. Reference 4 makes use of narrow-band-pass interference filters to isolate CH and C2 radiation. This same reference also describes the use of the ratio of C2 to CH radiation as an index of the changing equivalence ratio of a flame burning in a duct. Reference 9 describes the use of a spectrophotometer with wide-slit settings to isolate portions of the band spectra of the OH, CO, CH, and C2 diatomic emitters. This reference also describes a method for subtracting CO background radiation from OH, CH, and C2 band spectra so that the pure radiation from single emitters can be compared.

An extension of the type of work reported in reference 1 is contained in reference 6. Here, narrow-band-pass interference filters were used to isolate CO, CH, and C2 radiation regions in the spectra of laminar and turbulent propane-air flames. (No attempt was made to subtract the CO background radiation from the CH and C2 emission, as was done in ref. 9.) These studies used the emitter intensity per unit fuel-flow rate as an index of reaction rate and the ratio of C2 and CH radiation intensities as an index of reaction mechanism.

The presence of CO background radiation in the CH and C2 intensities does not permit the unambiguous determination of either relative changes in the intensity or changes in the ratio of CH and C2 emitter radiation. These determinations should properly be made using pure emitter radiations. However, up to the present time, no studies of flame radiation such as those described in references 1 and 6 have been made with pure diatomic emitter radiation.
Because of the apparent interest in using radiation techniques to study the turbulent-flame reaction rate, reaction mechanism, and flame structure, the methods described in references 1 and 9 were combined, and the improved technique was used to continue the study of laminar and turbulent propane-air and ethylene-air flames seated on bench-scale burners. Some of the work reported in reference 6 was repeated to determine what differences in results, if any, occur when pure radiations and radiation ratios are used. The present report also contains suggestions for extending this improved technique to other types of flame study.

APPARATUS AND PROCEDURE

Apparatus for Measuring Flame Radiation

A grating monochromator was used to isolate radiation from various regions of the flame spectrum. The radiation was detected by a photomultiplier tube placed at the exit slit of the monochromator, and the radiation intensity was read on a microammeter. All intensity measurements were in terms of the photomultiplier output current in microamperes. No attempt was made to convert the relative intensities to absolute values.

Two types of radiation measurements were made: total intensities from the whole flame, and local intensities in different parts of the flame. To measure changes in total flame radiation, the monochromator was placed in position 1, as shown in figure 1. Light from the flame struck a 45°-front-surface mirror and reflected into the monochromator. This light was unfocused radiant flux from the flame as viewed from above at a distance of 6 feet. The flame thus served as a virtual point source as outlined in reference 1.

The monochromator was fitted with a collimator containing an end slit for close-up measurements of radiation in different regions of the flame. The monochromator was placed in position 2, relative to the flame, as shown in figure 1. The end slit in the collimator was made parallel to the front slit of the monochromator. The monochromator was then oriented so that the slit system was horizontal, at right angles to the flame cone. The slits were made 20 millimeters long so that radiation from the complete diameter of the flame at its widest point could enter the monochromator. The slit width was set to accept radiation from a cross-sectional slice of flame about 1.0 millimeter high. To scan the flame, the monochromator was fixed, but the burner tube was moved up and down to bring different regions of the flame opposite the collimator slit.

This method of flame scanning differs from the methods used in references 6 and 9. In reference 9, the spectrometer scanned through the
flame front along a streamline to the limits of the outer cone. In reference 6, a small circular aperture was used to measure the relative intensity of a single emitter up the central axis of a two-dimensional flame. In the present work, the ratio of C₂ and CH radiations at different heights in the flame cone or brush was determined. No measurement was made of the relative intensity of single emitters up the flame cone. Likewise, no measurement was made of the distribution of emitter intensities within the thickness of the flame reaction zone itself. Measurements of this type have already been made (refs. 6 and 8).

Correcting for CO Background Radiation

Figure 2 shows how peaks of diatomic emitter radiation are superimposed on a continuum of CO radiation. (This figure differs from a similar figure in ref. 9 because of the different types of spectrometers and detectors used in the two studies.) Flame-intensity traces like those in figure 2 were made for flames of different equivalence ratios. These traces were examined for the best between-peak band-free area in which to measure pure CO radiation. A wavelength setting of 4500 angstroms was selected as a CO radiation check point in reference 9. However, the results of the present surveys indicate that a CO check point at 4120 angstroms is less likely to be contaminated by the band spectra of the other emitters when wide slits are used.

Once a pure CO point was chosen, radiation curves for the inner and the outer flame cones were normalized at 4120 angstroms and compared. As shown in figure 2, the troughs between peaks of the inner-cone spectrum are identical with the continuum of the outer cone, once the OH radiation has been passed. This equivalence suggested that, as a first approximation, the background-radiation continuum was all due to CO. As a check, a rich CO-air flame spectrum was normalized at 4120 angstroms and superimposed on the propane-air flame curves; it proved to be identical to the pure CO portion of the outer-cone radiation. This CO curve was compared with the outer-cone spectra of propane-air flames for a variety of equivalence ratios. The curve was also compared with the troughs between the peaks of the total flame spectrum at several equivalence ratios. In all cases, the CO points of the normalized curves coincided almost exactly with the CO spectrum. Thus, a single CO spectrum curve was assumed to be valid as a calibration curve for hydrocarbon-air flames over a range of equivalence ratios from 0.6 to 1.5. (The same result was found for butane-air flames, as reported in ref. 9.)

In order to calibrate the CO continuum out of the flame spectrum, a plot of intensity against wavelength of the CO-air flame was made for the same slit sizes used with the propane-air flame. The curve was normalized by adjusting the vertical intensity scale until the intensity at 4120 angstroms was equal to 1.00. The intensities at 3100, 4290, and
5130 angstroms were read off the curve, to be used as correction factors for CO under the OH, CH, and C2 radiation peaks, respectively, measured at the same wavelengths.

Spectral Intensity Measurements with Photomultiplier Tubes

Unfocused radiation measured at a distance from a flame is so weak that sensitive photomultipliers must be used to get an adequate signal. Photomultiplier tubes have an essentially linear response to intensity variations at any given wavelength. However, they are sensitive to slight changes in supply voltage. The tubes themselves often have undesirable fatigue characteristics if they are operated with excessive output current. (These problems are discussed in ref. 6.)

In this work, a voltage-regulated power supply was used to reduce supply-voltage effects on the photomultiplier tube. Excessive fatigue was avoided by operating the tube at outputs less than 5.0 microamperes. For some experiments the tube was prefatigued by exposing it continuously to a light source giving a 1.0-microampere output to the microammeter. The tube was kept fatigued, when not in use, by the same source. The flame itself was usually used as a source; otherwise a small electric light was used. Inasmuch as the tube fatigued rapidly at first, then leveled off, and recovered slowly, the prefatiguing gave more consistent results when relative readings were being made with the tube. Even so, the sensitivity varied slightly from day to day with the result that total readings taken on nominally the same flame were not identical on different days. Therefore, a method of internal calibration was used each day, with the smallest flame serving as a standard.

Fuel and Air Supply

C.p. propane and surgical-grade ethylene were used as fuels. Air was obtained from a filtered, 100-pound-pressure, laboratory air line. Both fuels and air were metered with jeweled critical-flow orifices mounted in tubing fittings for convenience in examination and cleaning. The orifice jewels were cleaned with redistilled acetone before each use.

Because of the large range of fuel and air flows used, a total of six orifices was required. The individual orifices were calibrated to a reproducibility of better than 1.0 percent. An over-all accuracy of 1.0 to 2.0 percent was calculated for the total flow range.
Propane-Air and Ethylene-Air Burners

Three different burners were used for generating laminar and turbulent flames. Most of the work on propane-air flames was done with a water-jacketed and piloted straight-tube burner (fig. 3(a)). The diameter of the inner tube was 1.012 centimeters. The pilot mixture was bled from the main fuel-air mixture through a small bypass controlled by a needle valve. All the ethylene-air work was done on the same type of burner, but the inner-tube diameter was only 0.635 centimeter.

Propane-air flames were also studied with a second type of burner, shown in figure 3(b). The fuel-air mixture was injected through two impinging circular annuli into the central cavity of a water-cooled doughnut-shaped burner. The mixture burned without admixture of secondary air and exhausted through two side ports.

Range of Experimentation

<table>
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<th>Propane-air</th>
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<td>Burner tube diameter, cm</td>
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<td>Leanest equivalence ratio</td>
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<td>Richest equivalence ratio</td>
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<tr>
<td>Highest average gas-flow velocity, ft/sec</td>
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<td>Highest Reynolds number</td>
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RESULTS AND DISCUSSION

Variation of Emitter Intensity with Equivalence Ratio

Flames were viewed by the monochromator at a constant distance for all total-flame radiation-intensity measurements. Thus, variation in the photomultiplier output current could be directly related to changes in the total intensity of a given emitter, provided a constant slit size and wavelength setting were used. The vertical scales of figures 4 to 6 are in photomultiplier output current per cubic centimeter of fuel burned per second. Since each emitter was measured at a different wavelength, and thus with a different photomultiplier-tube sensitivity, the
different emitter curves for the same fuel cannot be compared on an absolute basis. However, relative changes in the intensity of a single emitter or changes in the ratio of intensities of two emitters are valid determinations. Such comparisons form the basis for the results described in this report.

As shown in figures 5 and 6, peak intensities occur at different equivalence ratios for each emitter. CH and C₂ vary markedly with equivalence ratio, whereas CO varies only slightly with equivalence ratio. Below an equivalence ratio of 0.8, C₂ radiation almost disappears. The sensitivity of the intensity of C₂ radiation to equivalence ratio is most obvious in figure 4(d). Here, a 10-percent change in equivalence ratio, from 1.0 to 1.1, represents a 100-percent change in intensity.

For meaningful measurements of changes in the intensity of single emitters, it would be desirable to meter mixtures of fuel and air to a composition accuracy of 0.1 percent. This is 10 times more accurate than the results usually obtained with orifice metering systems. Such accuracies would entail difficulties in flow calibration. At the high flow rates involved, it is also unrealistic to premix the gases in a large storage vessel prior to burning them. Thus, accurate gas metering systems for flame-radiation studies still present a problem.

The ratios of the intensities also change with equivalence ratio, as can be deduced from the manner in which the radiation curves in figures 5 and 6 peak at different equivalence ratios. With four emitters, 12 different ratios can be taken, half of which are the reciprocals of the others. Of the six different emitter ratios, only the two containing C₂ and CH come exclusively from the inner-cone reaction zone. Of these two ratios, C₂/CH plots best.

The ratios of C₂ and CH radiation intensities determined from figures 5 and 6 are shown in figure 7 as a function of equivalence ratio for propane-air and ethylene-air flames. The curves represent C₂-CH radiation ratios independent of varying amounts of CO. If varying amounts of CO were present, which might occur if only a portion of a flame were examined, radiation ratios would fluctuate about the curves, even though the metered equivalence ratio remained constant. The desirability of subtracting the CO radiation is obvious.

The C₂-CH ratios for ethylene-air flames do not decrease at rich equivalence ratios as do the ratios for propane-air flames. Even at leaner equivalence ratios, the C₂-CH ratios for the flames of the two different fuels, although similar, do not coincide exactly. These differences emphasize the necessity of calibrating with the fuel to be used in any radiation study. The prospect of a universal radiation "equivalence-ratio meter" does not appear too promising.
The sensitivity of a single emitter to equivalence ratio is an advantage in using radiation ratios as an index of equivalence ratio. As shown in figure 7, radiation ratios in the rich region are very sensitive to changes in equivalence ratio. For ethylene-air flames at an equivalence ratio of 1.3, for example, a change of ±1.0 percent in equivalence ratio represents a change of ±5.0 percent in the intensity ratio. Thus, being able to determine intensity ratios within 1.0 percent can mean determining equivalence ratios to an accuracy of 0.2 percent. The radiation-ratio principle could therefore be used to detect very small changes in equivalence ratio in flames.

Calibration of the radiation characteristics of flames was done with laminar flames burning on open tubes. The question arises as to how pertinent these data are to flames burning in enclosed burners. The results of experimentation with a doughnut-shaped burner are also shown in figure 7. Within experimental limits, the radiation ratios for the open and the enclosed flames are the same for identical equivalence ratios.

From an examination of the radiation characteristics of flames, it appears that meaningful radiation measurements require precise control of the equivalence ratios of the metered gases. The ratio of C₂ to CH radiation is a sensitive index of the fuel-air ratio of the flame; however, radiation-ratio calibrations should be made with the fuel being studied. C₂-CH ratios become insensitive to equivalence-ratio changes at equivalence ratios less than 1.0. Both open and enclosed flames exhibit the same radiation ratios at identical equivalence ratios.

Variation of Radiation Equivalence Ratio in Flame Column

The C₂-CH ratio should be an indication of local equivalence ratios in a flame as well as of over-all flame equivalence ratio. Laminar and turbulent ethylene-air flames were scanned from base to tip with the collimated slit system. Figure 8 shows how the equivalence ratios varied up the length of the flame column. The first result of interest was the manner in which the portion of the flame just above the burner exhibited a progressively lower equivalence ratio as the average flow velocity in the burner increased. The percentage decrease seemed to be more related to average flow velocity than to equivalence ratio. The most logical explanation is that this point in the flame was the region of maximum secondary-air admixture. This decrease in equivalence ratio might have been more extreme than the curves indicated but was integrated out by the 1.0-millimeter width of the scanning slit. The return to richer equivalence ratios at the extreme base was caused by the presence of the flame from the bypass pilot.
The second interesting fact was observed higher up in the brush of the flame. Here, the radiation ratio returned to the value equal to the original metered fuel-air ratio. Apparently, no appreciable mixing with secondary air occurred in the upper brush of the turbulent flames. Since the experimental error in metering was about 1.0 percent, the occasional 1.0-percent excursions in the plots of figure 8 are not considered significant. The data near the tip are also erratic, probably because the flame tip was unstable and flickered in and out of this region. One might assume that the slight decrease in equivalence ratio in the upper brush of the rich turbulent flames was caused by mixing. However, stoichiometric flames at much higher average flow velocities did not exhibit this effect. The apparent decrease in equivalence ratio might have been real, or slight differences from the calibration of laminar-flame radiation might have existed in the turbulent flames.

The measurement of local radiation-intensity ratios just described is an example of the use of flame radiation to examine the detailed structure of flames. It might be worthwhile to make similar measurements on enclosed flames burning in a homogeneous reactor of the type described in reference 11. The details of flame-surface structure for the type of flames described in references 7 and 8 might also be investigated by these radiation methods. Transient equivalence-ratio measurements could be made by having two exit slits on the monochromator - one for CH and one for C2 radiation. The radiation could be detected by two photomultiplier tubes and displayed simultaneously on a dual-beam oscilloscope. It might also be desirable to display the radiation ratio on a single-beam instrument. Similar work has been recently reported in reference 12.

Variation of Emitter Intensity with Fuel-Flow Rate and Reynolds Number

The average gas-flow velocity of the propane-air open burner did not reach very high values before blowoff occurred. Therefore, propane-air flames were considered unsatisfactory for studies at high gas velocities in open tubes. The use of a hydrogen pilot was considered but rejected because of its unknown effect on the hydrocarbon-flame radiation. With ethylene-air flames, gas-flow velocities as high as 125 feet per second were obtained for stoichiometric mixtures.

As shown in figure 4(d), C2 radiation in ethylene-air flames is very sensitive to small changes in equivalence ratio. Slight errors in fuel and air metering resulted in C2 data with too much scatter to be plotted accurately. Inasmuch as the total flame was being examined, the CO radiation, which is relatively insensitive to equivalence-ratio fluctuations, was used to monitor CH radiation. The results are shown in figure 9.
The most noticeable characteristic of figure 9 is the break that occurs between Reynolds numbers of 6000 and 8000. The curves seem to be made up of two approximately straight-line portions, each of which represents an almost linear change of intensity with increasing gas-flow velocity. Figure 10 is a replott of the top graph in figure 9 in terms of the emitter intensity per cubic centimeter of fuel burned per second. It closely resembles figure 14 of reference 6. The breaks in the curves occur within about the same average flow-velocity region in both studies. The percentage difference between the low and the high gas velocity portions of the CH and CO curves in this report and in reference 6 is also about the same. Thus, determinations of this type with CH and CO radiation, with the CH intensity uncorrected for the CO background, give results as do determinations with pure radiation. The absolute values of radiation intensity are naturally not the same. However, the results of the two studies are probably similar because changes in the relative intensity of radiation from the total flame were measured, and CO and CH vary to about the same degree.

An actual decrease in equivalence ratio due to secondary-air intermixing at high gas-flow rates would result in lower unit intensities for CH radiation. It would thus seem logical to attribute the change in slope or decrease in intensity per unit fuel flow in figure 9 to mixing phenomena. Several factors make such an assumption highly questionable. Figure 6 indicates that CH is more sensitive than CO radiation to equivalence-ratio changes by an order of magnitude. Yet, in figure 9, the extent of the break in the CH and CO curves is about the same. Furthermore, in figure 9 for an equivalence ratio of 1.1 where CO radiation should show no appreciable change with small changes in equivalence ratio, the break in the curve is as decided as it is for the other equivalence ratios. Also, mixing does not appear to occur in the major portion of the turbulent flame brush for near-stoichiometric flames; the C2-CR radiation ratios of the flames in figure 8 indicated an equivalence ratio almost identical with the metered value. One might suspect that the reduction in total intensity is related to apparent mixing in the portion of the flame just above the burner lip. This would suffice as an explanation for some decrease in CH and C2 radiation but would not explain the decrease in CO radiation. It is possible that in turbulent flames the reaction rate decreases, with an accompanying decrease in temperature and radiation. This decrease might be very small because radiation intensity is very sensitive to temperature. However, flame-radiation studies up to the present time have not been able to answer this question.

Flame-radiation methods might possibly be extended to determine relative reaction rates in localized regions of both laminar and turbulent flames. Space heating rates of open turbulent flames, for example, have previously been determined by measurements on photographic negatives, with uncertain results (refs. 13 and 14). The scanning technique
described in the present paper can be used to determine the relative intensities of the \( \text{OH} \), \( \text{CO} \), \( \text{CH} \), and \( \text{C}_2 \) emitters in different equivalent flame volumes up the column of the flame. Figures 125 and 126 of reference 6 show such plots for turbulent propane-air flames. The intensity can be related to the amount of fuel being burned in the volume by proper calibration curves (such as appear in fig. 9). The local intensity values can be monitored by the radiation ratio technique shown in figure 7, with correction data obtained from figures 5 and 6. The size, shape, and thickness of the turbulent brush and the spatial concentration of emitters in the cross section of the brush can be determined by direct scanning across the flame with a collimated-slit monochromator. Curves of this type are shown in figures 122 and 123 of reference 6. The traces could be interpreted by methods outlined in reference 10.

**SUMMARY OF RESULTS**

The relative amounts of \( \text{OH} \), \( \text{CO} \), \( \text{CH} \), and \( \text{C}_2 \) radiation from propane-air and ethylene-air flames were isolated and compared at different equivalence ratios of the combustible mixtures. The following results were obtained:

1. The intensities of \( \text{CH} \) and \( \text{C}_2 \) radiation from the flames were found to be very sensitive to small changes in equivalence ratio.

2. The \( \text{C}_2-\text{CH} \) ratios were found to be an accurate index of the initial metered equivalence ratio of the combustible gases. This relation held for both open and enclosed flames.

3. When localized radiation measurements were made up the columns of laminar and turbulent open flames, the radiation equivalence ratio was found to decrease near the bases of turbulent flames. The greater the average flow velocity in turbulent flames, the greater became the apparent decrease in equivalence ratio at the bases of the flames. However, no appreciable change from the metered equivalence ratio was observed in the radiation equivalence ratios of the major upper portion of turbulent flames burning at stoichiometric equivalence ratios and average gas-flow velocities as high as 125 feet per second. Turbulent flames at a metered equivalence ratio of 1.3, on the other hand, showed an apparent 1- to 2-percent decrease in the radiation equivalence ratio of the upper flame brush.

4. At gas-flow velocities greater than 50 feet per second, all emitters in turbulent ethylene-air flames exhibited a decrease in the relative radiation intensities per unit fuel-flow rate. This decrease in radiation intensity could not be accounted for by changes in relative
radiation-intensity ratio, which might occur from the mixing of air from the surrounding atmosphere with the combustible gases.

Lewis Flight Propulsion Laboratory
National Advisory Committee for Aeronautics
Cleveland, Ohio, February 13, 1958

REFERENCES


Figure 1. - Monochromator and associated components used to study total and local flame radiation.
Pilot fuel and air mixture in Hater out

Viewing direction, total radiation

Pilot fuel and air mixture in

Exhaust products out

Water in

Water out

Open tube:
Propane-air, 1.012 cm I.D.
Ethylene-air, 0.635 cm I.D.

(a) Water-cooled and piloted open-tube burner for both propane-air and ethylene-air flames.

(b) Water-cooled doughnut-shaped burner for propane-air flames.

Figure 3. - Diagrams of burners used in flame-radiation studies.
Figure 4. - Calibration curves for OH, CO, CH, and C₂ radiation intensity as a function of equivalence ratio in ethylene-air flames.
Figure 5. - Averaged OH, CO, CH, and C₂ radiation intensity as a function of equivalence ratio in propane-air flames.
Figure 6. - Averaged OH, CO, CH, and C₂ radiation intensity as a function of equivalence ratio in ethylene-air flames.
Figure 7. - Ratio of $C_2$ and OH radiation intensity as a function of equivalence ratio for propane-air and ethylene-air flames.
Figure 8. - Variation in equivalence ratio up the columns of turbulent ethylene-air flames. All photographs to scale.
Figure 9. - \( \text{CH and CO radiation intensity as a function of fuel flow and Reynolds number for ethylene-air flames.} \)
Figure 10. CH and CO radiation intensity per cubic centimeter of fuel burned per second in ethylene-air flames as a function of fuel flow and average gas-flow velocity. Propane-air data from figures 59 and 90 of reference 6 were adjusted in intensity to equal ethylene-air data at average flow velocity of 20 feet per second. All equivalence ratios equal 1.0.