EMISSION SPECTRA OF PROPANE-AIR FLAMES IRRADIATED

WITH A 1000-CURIE GOLD SOURCE

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The gold was flown to and from the Materials Testing Reactor by personnel of the Selfridge Air Force Base.

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ABSTRACT

The effect of nuclear radiation on propane-air flames at low pressures was investigated experimentally. The source of radiation was 30 grams of irradiated gold wire with an initial strength of 1000 curies. Both the preflame mixture and the flame zone were irradiated intensely. The number of ion pairs produced by beta radiation greatly exceeded that produced by gamma radiation. Emission spectra of flat flames were measured quantitatively at a series of elevations through the flame. The emission at 4315Å due to OH was increased up to 30 percent and the emission at 5165Å due to C₂ up to 150 percent by the radiation. The emission at 3063Å due to OH was not affected significantly. The maxima in the emission due to C₂ and OH were observed to occur at an earlier stage in the irradiated flames than the maxima in the emission due to CH.
INTRODUCTION

This paper reports one phase of an experimental investigation of
the effect of intense nuclear radiation upon flames. The rate of propagation
of irradiated bunsen flames has been reported previously.\textsuperscript{1}

Studies of combustion in ramjet burners and in low-pressure flames
have indicated that emission spectra provide more insight into the mechan-
ism of the combustion processes than do flame speed and stability measure-
ments.\textsuperscript{3,5,6} Accordingly, equipment was designed to obtain the spectral
emission at a series of elevations through a flat flame subjected to nuclear
radiation. The intensities of the \textsuperscript{C}_2, OH, and OH bands were measured
quantitatively.

The source of radiation for the emission studies was the same 30
grams of irradiated gold wire used in the flame-speed studies. The flame-
speed measurements were carried out at source intensities as high as 10,000
curies. The reported data on spectral emission were obtained after the gold
had decayed to 1,000 curies.

Data were obtained at a series of propane-air ratios and a series
of subatmospheric pressures as the source decayed.
EXPERIMENTAL EQUIPMENT

The equipment consisted of a premixed propane-air supply and metering system, a burner, a vacuum tank and exhaust system, shielding, a source of radiation, and an optical traversing system. All but the traversing system were described in the previous paper. A diagram of the system is shown in Figure 1 and a drawing of the burner and source assembly in Figure 2.

As indicated in Figure 1, a front-surfaced, aluminum-coated mirror was mounted in a 12-inch pipe, welded on the 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 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 on 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 was mounted to read elevations to a thousandth of an inch. The light emitted by the flame passed through the spectrograph slit, to the reflecting spectrograph grating, and then to the photographic film where the intensity at the various wavelengths was recorded.
The source consisted of about 30 grams of 99.95-percent gold wire, 0.005 inch in diameter. The gold was irradiated about four and one-half days in a high-flux section of the Materials Testing Reactor. The wire was wound in 1/16-inch coils before irradiation and was lightly compressed into the hollow cylinder shown in Figure 2 after irradiation. With this source location both the propane-air mixture and the flame were irradiated intensely.

From neutron flux calculations and subsequent measurements, the strength of the gold source on removal from the Materials Testing Reactor was estimated to be 9600 curies due to Au-198 and 5670 curies due to Au-199 for a total of 15,270 curies. The strength at the time of the various spectral emission experiments was computed on the basis of half lives of 2.69 days for Au-198 and 3.15 days for Au-199. The results are summarized in Table I.

**TABLE I. STRENGTH OF SOURCE**

<table>
<thead>
<tr>
<th>Condition</th>
<th>Hours</th>
<th>Au-198</th>
<th>Au-199</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Removal from pile</td>
<td>0</td>
<td>9600</td>
<td>5670</td>
<td>15,270</td>
</tr>
<tr>
<td>Experiment No. 1</td>
<td>268</td>
<td>538</td>
<td>484</td>
<td>1,022</td>
</tr>
<tr>
<td>Experiment No. 2</td>
<td>362</td>
<td>198</td>
<td>205</td>
<td>403</td>
</tr>
<tr>
<td>Experiment No. 3</td>
<td>429</td>
<td>96</td>
<td>111</td>
<td>207</td>
</tr>
<tr>
<td>Experiment No. 4</td>
<td>595</td>
<td>16</td>
<td>24</td>
<td>40</td>
</tr>
<tr>
<td>Experiment No. 5</td>
<td>772</td>
<td>2.4</td>
<td>4.8</td>
<td>7.2</td>
</tr>
</tbody>
</table>

The production of ion pairs in the gas phase in the cylinder and in the flame front would appear to be the most significant phenomenon resulting from the gold source. The disintegration scheme of Au-198 and Au-199

*Phillips Petroleum Corporation, Idaho Falls, Idaho.*
and the estimated rate of production of ion pairs per cubic centimeter per curie in these two regions were presented previously.\textsuperscript{1}
EXPERIMENTAL PROCEDURE

Premixed propane and air was passed from storage tanks through a metering system into the burner. The propane was 99.5 percent pure. The composition of the propane-air mixtures was established from blending pressures and was checked by analysis.

All runs were made with a linear velocity of about 1.0 ft/sec at the burner head. At this velocity a flat flame floating above the burner screen, as shown in Figure 2, was obtained over the entire range of operating pressures and propane-air ratios. At somewhat higher velocities and low pressures the flame formed a bunsen cone and at somewhat lower velocities and high pressures it burned on the screen. As a further condition emission from the flame had to be sufficiently intense to produce a satisfactory spectrographic negative in 30 minutes. The flame was essentially a disc about one inch in diameter and of the order of 0.05 inch thick. The exact thickness and distance above the burner screen varied with pressure and propane-air ratio. This thin vertical dimension of the flame was traversed by the spectrograph. The temperature of the propane-air mixture leaving the burner head was not measured due to failure of a thermocouple, but was undoubtedly within a few degrees of the ambient temperature.

The pressure and flow rate were first established. The spectrograph film pack was loaded with Royal Pan* film and the slit adjusted to the desired elevation. The room was darkened, the mask removed from the film pack, and the shutter was opened for thirty minutes. The time of exposure was limited by darkening of the film by scattered gamma radiation. The shutter was closed, a new strip of the film was shifted into position, the slit elevation was adjusted, and the shutter was opened again.

Exposures were made at eight different elevations through the flame zone. This procedure was carried out for tank pressures of 6, 8, 10, and 14 in. Hg abs at a propane-to-air mass ratio of 0.08 and then at propane-to-air mass ratios of 0.070, 0.0637, and 0.060 at a tank pressure of 14 in. Hg abs. The experiment was repeated on succeeding days at lower source strengths.

The spectrographic films were developed under carefully controlled conditions to permit a quantitative comparison of the density of exposure at the various wavelengths. They were developed at constant temperatures, agitation, and time in 15 gallons of DK-50* developer, diluted one to one with water. The same batch of developer was used for all films to guard against changing developer strength. The quantity used was so great that it can be assumed its strength did not change appreciably with use.

After development, the films were scanned with a microphotometer and the density of exposure was recorded as a function of wavelength. A typical microphotometer tracing is shown in Figure 3.
EXPERIMENTAL RESULTS

A. Emission at 4315°A

The strongest emission from the flame occurred at 4315°A due to OH. The relative intensity at this wavelength was read from the microphotometer traces such as Figure 3 and is plotted versus elevation through the flat flame at various pressures and source strengths in Figure 4. The propane-air mass ratio for this set of data was 0.08. The maximum spectral intensities with respect to elevation were read from Figure 4 and plotted versus source strength in Figure 5. The data for different propane-air ratios at a tank pressure of 14 in. Hg abs are presented in Figure 6. The maximum spectral intensities with respect to elevation obtained from Figure 6 are plotted versus source strength in Figure 7.

B. Emission at 5165°A and 3063°A

The most intense emission due to C₂ occurred at 5165°A and the most intense emission due to OH at 3063°A. The same procedure was followed for these spectral lines as for the OH line at 4315°A. However, in some cases the emission was so weak that quantitative information could not be obtained from the microphotometer tracings.

Spectral intensities at both 5165°A and 3063°A are plotted versus elevation for a propane-air mass ratio of 0.08 in Figure 8. The maxima are plotted versus source strength in Figure 9, with pressure as a parameter. The data did not permit construction of a plot such as Figure 7, showing the effect of propane-air ratio.
DISCUSSION

The intensity of emission is observed in Figures 4, 6, and 8 to be almost symmetrical about the elevation at which the maximum emission occurs, indicating that the maximum rate of reaction occurs midway between initiation and completion. The maxima decrease and shift away from the burner as the pressure is decreased. Figure 6 indicates a decrease in intensity of CH as the propane-air ratio is decreased, which is to be expected since the concentration of total carbon decreases. The elevation of the maxima does not appear to be much of a function of propane-air ratio.

The scatter in the abstracted points in Figures 5 and 9, and particularly in Figure 7, is due in part to the difficulty in determining the maxima in the otherwise well-defined curves in Figures 4, 6, and 8. Some improvement might have been obtained if the raw data were first fitted by empirical equations of appropriate form but this refinement was not believed necessary. From Figures 5 and 9, it is apparent that the maxima at 4315°A due to CH and at 5165°A due to C₂ increase with source strength, while the maxima at 3063°A due to OH are relatively constant. The effect of the radiation appears to increase with pressure as would be expected since the probability of beta interaction is greater at greater densities.

Despite the scatter in the data in Figure 7, the effect of irradiation is clearly greatest at high propane-air ratios. This too is to be expected since the probability of effective beta interaction is greater with higher concentrations of fuel molecules and radicals.

In order to compare the emission due to CH, C₂, and OH, the data for emission at 4315°A, 5165°A, and 3063°A are replotted versus elevation at a propane-air ratio of 0.08 and a tank pressure of 14 in.Hg abs in Figure 10. The maxima are observed to occur at different elevations. The
elevations at which the maxima occur are plotted versus source strength for a propane-air ratio of 0.08 and pressures of 8 and 14 in. Hg abs in Figure 11. Contrary to expectations, the maxima due to C\textsubscript{2} and OH occur at about the same elevation and the maxima due to CH occur at a higher elevation. This suggests the possibility that carbon monoxide is formed in flames by the reaction

\[
\text{C}_2 + \text{OH} \rightarrow \text{CH} + \text{CO}.
\]
CONCLUSIONS

Intense irradiation of propane-air mixtures and flames increases significantly the emission due to CH and $C_2$. The emission due to OH is not greatly affected. The effect of the radiation increases with pressure and the propane-air ratio. The maxima in the emission due to $C_2$ and OH occur at an earlier stage in irradiated flames than the maxima in the emission due to CH.
REFERENCES


2. Emmons, A. H., Private Communication.


4. Lewis, W. B., Private Communication.


Figure 5

Tank Pressure:
- 14 in. Hg abs
- 10 in. Hg abs
- 8 in. Hg abs
- 6 in. Hg abs

Maximum Relative Spectral Intensity - (Zero-Clear Film) vs. Source Strength - Ciernes
Figure 11