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RADIATION PROCESSES RELATED TO OXYGEN-HYDROGEN COMBUSTION AT HIGH PRESSURES

by Marshall C. Burrows

Lewis Research Center

Cleveland, Ohio

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By Marshall C. Burrows

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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SUMMARY

Radiation related to the combustion of liquid-oxygen jets in a gaseous hydrogen atmosphere was studied by measuring the intensities of the hydroxyl radical (OH), oxygen (O_2), and water (H_2O) radiation in a combustion chamber equipped with windows. Overall oxidant-fuel weight ratio was varied from 1 to 10 at a chamber pressure of 24 atmospheres. Maximum intensities from the radiating species were obtained near stoichiometric mixture ratios. Gas temperatures derived from the measured color temperature of a tungsten plate in the gas stream agreed with theoretical temperatures of the products. Gas radiation appeared to be thermally excited and in local equilibrium at all locations within the combustor.

Radiation measurements made in the upstream region of a stable combustor showed that intensities varied little with oxidant-fuel weight ratio. Measured intensities initially increased in the direction of gas flow because of the increased extent of reaction and then decreased when diluted with excess reactant or cooled by heat transfer to the walls. Ultraviolet and total radiation data showed that the reaction proceeded at approximately a constant rate until the limiting reactant was consumed. The distance required to complete the reaction varied from 1.5 to 12 inches for a variation in oxidant-fuel weight ratio of 1 to 9.

During combustion with pressure and velocity oscillations along the axis of the combustion chamber, average intensities of the radiating gases showed that the axial distance required to react the liquid oxygen (O_2) and hydrogen (H_2) was reduced. The distance required to complete the reaction in this case varied from 1 to 6 inches for an oxidant-fuel weight-ratio variation of 1 to 9.

The extent of reaction as determined by radiation measurements in the two combustors was compared to the predicted trends where turbulent mixing or vaporization was considered to control the reaction.

INTRODUCTION

Various analytical models of the combustion processes in liquids and gases

have considered the role of vaporization rate (ref. 1), turbulent mixing rate (ref. 2), and chemical reaction rate (ref. 3) in efforts to determine the rate-controlling processes of various reactants. An experimental method of measuring these rates would be a valuable tool for the analysis of stable and unstable combustion processes.

In this paper, the overall processes controlling the combustion of liquid oxygen and gaseous hydrogen were related to radiation from several molecular species along the axis of a high-pressure combustor. Previous work has established the thermal origin of the radiation (refs. 1 and 5 and unpublished NASA data obtained by Marshall Burrows and Ronald Razner) and therefore makes it possible to relate radiation intensities to the reaction process, and equilibrium products and heat losses to the combustor walls. Data are presented for stable combustion at constant pressure and unstable combustion with a maximum pressure oscillation of 30 percent. Thermal and chemical equilibrium in the gases are discussed, and extents of reaction in the two combustors as measured by radiation intensities are compared to the calculated trends for several combustion models.

PROCEDURE

A copper combustor was constructed in sections to permit the location of a section with windows at various axial distances from the injector (fig. 1).

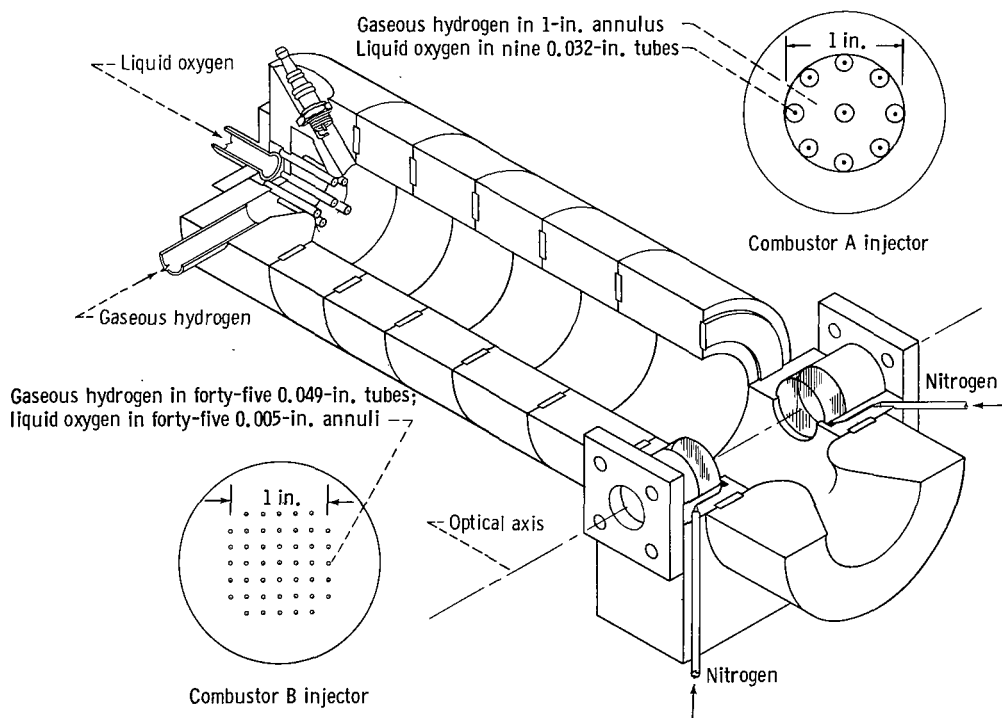
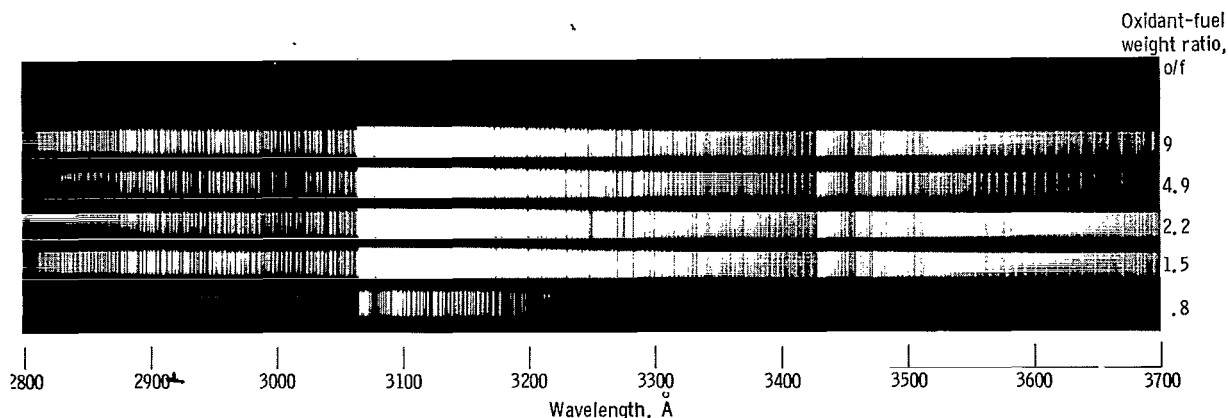


Figure 1. - Gaseous-hydrogen - liquid-oxygen combustor. Distance from injector to optical axis variable from 0.8 to 21 inches; chamber diameter, 2 inches; nozzle diameter, 0.60 inch. Injector A, nine 0.032-inch liquid-oxygen jets parallel to low-velocity hydrogen flow. Injector B, forty-five 0.049-inch high-velocity hydrogen jets surrounded by 0.005-inch liquid-oxygen annuli.

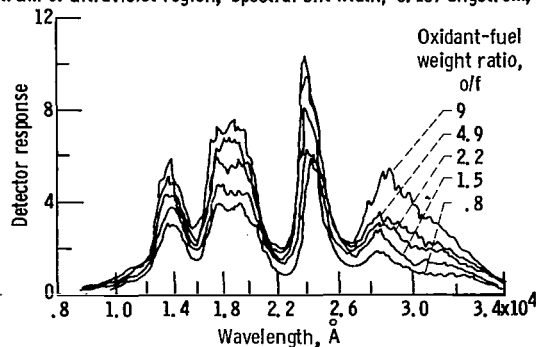
High-velocity nitrogen parallel to the inner surfaces of the windows prevented condensation and kept the optical path free of absorbing gases. Stable combustion was provided by nine 0.032-inch liquid-oxygen jets injected axially with low-velocity hydrogen gas, hereinafter referred to as combustor A. Consistently unstable combustion was provided by 45 high-velocity gaseous-hydrogen jets surrounded by liquid oxygen in 0.005-inch annuli, hereinafter referred to as combustor B. Both combustors were operated over a range of oxygen and hydrogen flows to obtain oxidant-fuel weight-ratio variations (o/f) from 1 to 10 ± 0.1 . Calculated oxygen velocities varied from 60 to 100 feet per second in combustor A and from 35 to 60 feet per second in combustor B. Data are presented for an average chamber pressure of 24 atmospheres.

Emission spectra from the combustor window were recorded photographically in the ultraviolet and visible spectral region on a 1.5-meter grating spectrograph. With a spectral slit width of 0.109 angstroms, exposures of 3 seconds were sufficient to expose type 1-0 Kodak film. Infrared spectra were obtained by a monochromator equipped with a calcium fluoride prism and an indium antimonide detector. Spectral slit width was approximately 1000 angstroms. The spectral region from 6,000 to 35,000 angstroms was scanned in approximately 6 seconds, and detector outputs were recorded as a function of wavelength.

Ultraviolet radiation passed through a 0.050-inch restriction and quartz



(a) Photographic spectrum of ultraviolet region; spectral slit width, 0.109 angstrom; Kodak type 1-0 film.



(b) Spectrogram of infrared region; indium antimonide detector; spectral slit width, 1000 angstroms; calcium fluoride and mirror optics.

Figure 2. - Spectral radiation from combustor gases as function of wavelength.

window to a photometer equipped with an ultraviolet filter in front of a photomultiplier (S-5 response). Spectral response of the unit extended from 2400 to 4000 angstroms with a broad maximum at 3000 to 3200 angstroms. The photomultiplier was operated within its linear range, and its output was monitored by an X-Y plotter from which average intensities were measured during each run. Precision of the measurements was approximately ± 10 percent. Runs were duplicated at least once to insure reproducibility of the data.

Total radiation was transmitted through the 1-inch window port to a pyrometer with parabolic mirrors that focused the radiation on a thermocouple. Spectral response of the pyrometer was limited from 2,000 to 35,000 angstroms by the quartz combustor windows. Run duration was at least 5 seconds to allow the pyrometer sufficient time to respond to the radiation. Maximum pyrometer readings during each run were reproducible within ± 10 percent.

Output voltages from the pyrometer were directly proportional to the total radiation intensities. Calibration with a tungsten lamp showed that the voltage e varied according to the relation $e = \alpha T^4$,³³ where α is a calibration

constant equal to 4.48×10^{-18} volts per $^{\circ}\text{K}$ and T is the color temperature in $^{\circ}\text{K}$.

RESULTS

Spectra

Spectra of the gases within the combustor (fig. 2) show that predominant radiation in the ultraviolet and visible spectral region is due to the hydroxyl radical (OH) with the maximum intensities concentrated around 3100 angstroms. Weak spectral lines between 3200 and 4500 angstroms are largely due to molecular oxygen radiation (Schumann-Runge bands). The infrared spectra show the characteristic water-vapor bands centered near 14,000, 19,000, and 27,000 angstroms. The OH radical also has emission bands near these wavelengths, but they are masked by the stronger water (H_2O) bands and, therefore, cannot be distinguished.

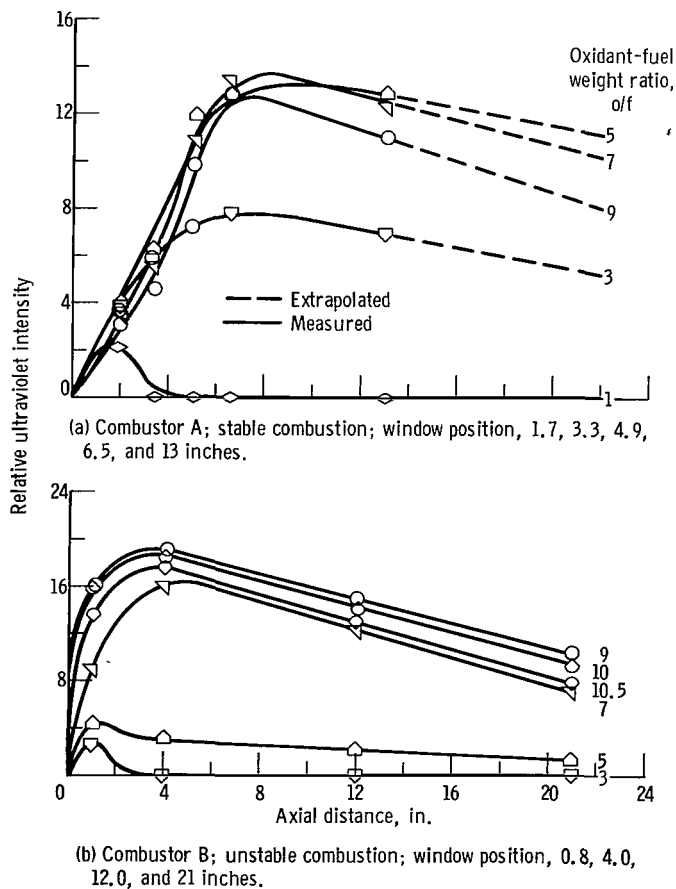


Figure 3. - Relative ultraviolet intensities as function of axial distance from injector.

Ultraviolet Radiation

A photometer was used

to measure the ultraviolet radiation emitted by the gases in the combustor. Average ultraviolet radiation intensities were measured as a function of axial distance from the injector in combustors A and B (fig. 3). Overall oxidant-fuel weight ratios were varied from 1 to 9 in combustor A and from 3 to 10.5 in combustor B. Radiation near the injector in combustor A increased at approximately a constant rate with axial distance. Maximum ultraviolet radiation intensities occurred approximately 1.5 inches from the injector at an o/f of 1 and 8 inches downstream at an o/f of 7. Data were not obtained from combustor A at axial distances greater than 13 inches from the injector. Radiation thereafter was extrapolated as a straight dashed line for each o/f. Apparent decreases in radiation after the maxima occurred at rates that varied from 1.4 to 2.8 percent per inch of combustor length.

Combustor B was operated at the same average pressure as combustor A, but with a high-frequency pressure oscillation with peak-to-peak amplitudes that varied from 5 to 30 percent of the average pressure. Intensities measured by the photometer showed that average radiation increased at very high rates near the injector of combustor B with the rate increasing with overall mixture ratio to a maximum at an o/f of 9. Maximum ultraviolet intensities occurred at approximately one-half the distance from the injector as compared to the distance required during stable combustion (figs. 3(a) and (b)). The decrease in radiation intensities further downstream was larger during unstable combustion, the rate varying from 3.5 to 4.5 percent per inch of combustor length.

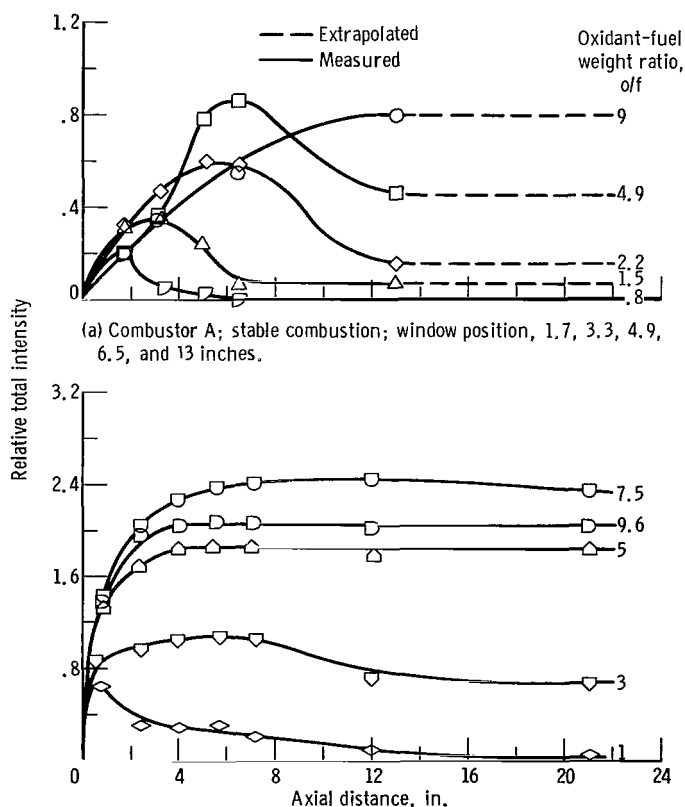


Figure 4. - Relative total intensities as function of axial distance from injector.

Total Radiation

A total radiation pyrometer was used to measure the total radiance from the gases within the chamber. Total radiation intensities varied with axial distance from the injector, as shown in figure 4, for combustors A and B.

Total radiation near the injector of combustor A increased at approximately a constant rate with distance, which is similar to the behavior of the ultraviolet

radiation. Maximum total radiation occurred at approximately 1.5 inches from the injector for an o/f of 0.8, 6 inches for an o/f of 4.9, and 12 inches for an o/f of 9. The radiation at distances greater than 13 inches appeared to be constant and was extrapolated as a dashed line.

Total radiation intensities in combustor B increased at very high rates near the injector. Maximum total radiation occurred at approximately 0.8 inch from the injector for an o/f of 1 and increased to 6 inches for an o/f of 7.5. At downstream distances greater than 12 inches, measured radiation intensities remained nearly constant.

Gas Temperature

Gas temperatures were experimentally derived as a function of the overall mixture ratio, 12 inches from the injector in combustor A. An oxidized tungsten plate with a high surface emittance (ref. 6 and unpublished NASA data by C. Liebert) was placed in the center of the combustor, and the pyrometer was focused on its surface through the combustor window. Total radiation from the plate is shown (curve A) in figure 5 as a function of o/f. When the equation relating the pyrometer voltage to the color temperature of the tungsten is used, the radiation data show that the apparent plate temperature increases from $1200^{\circ}\pm 50^{\circ}$ K at an o/f of 1 to $3240^{\circ}\pm 100^{\circ}$ K at an o/f of 9. These temperatures are somewhat lower than the calculated temperatures for equilibrium gas mixtures (refs. 7 to 9) that varied from 1250° K at an o/f of 1 to a maximum of 3470° K at an o/f of 8. Plate radiation, corrected for plate emittance, window absorption, and heat-transfer losses, would give plate temperatures quite close to calculated temperatures for equilibrium gas mixtures. The average temperature of the gases heating the tungsten plate, therefore, was substantially that of the calculated temperature; this agreement indicates that the reaction was completed 12 inches from the injector. The total gas radiation at 12 inches from the injector is plotted as curves B and C in figure 5 for combustors A and B, respectively. Intensities are lower than those for the

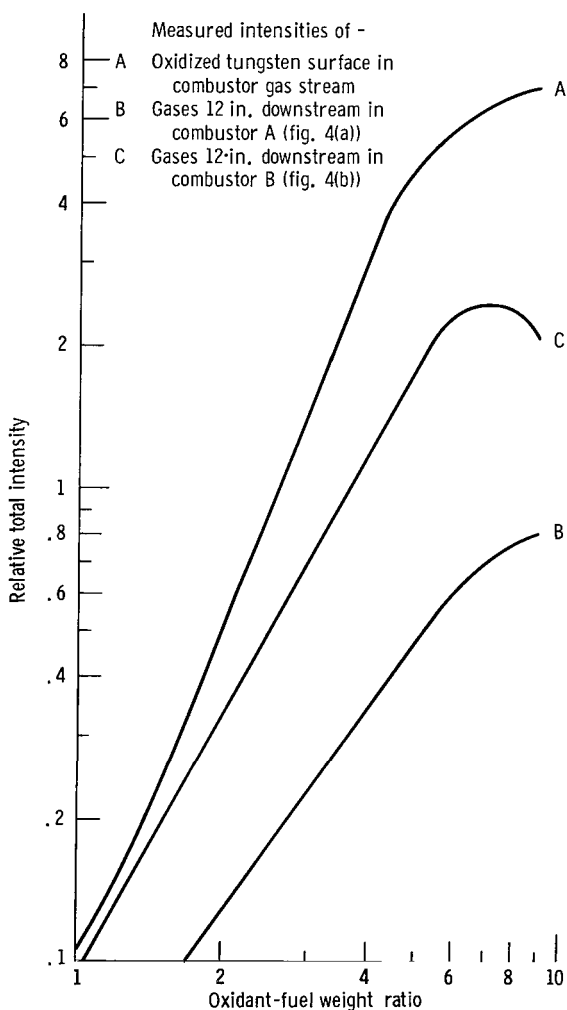


Figure 5. - Relative total intensities as function of oxidant-fuel weight ratio.

tungsten plate.

DISCUSSION

Thermal Equilibrium

The spectrograms of emitted radiation from combustors A and B indicate that the principal ultraviolet emission was from the OH radical and that the infrared radiation was primarily from H₂O. The ultraviolet photometer was assumed to measure average intensities of the OH radical emission centered at 3100 angstroms, and the total radiation pyrometer was assumed to measure average intensities of the H₂O emission centered at 24,000 angstroms.

A decrease in ultraviolet radiation intensities was shown at large axial distances from the injector in figure 3. Since previous work has shown that the OH radical is thermally excited in the combustor (ref. 4 and unpublished NASA data obtained by Marshall Burrows and Ronald Razner), the decrease in radiation intensities was assumed to be due to the decreasing temperature of the gases caused by heat transfer to the chamber walls. Heat-transfer rates to the walls can be estimated from the calculated temperature decrease and from the reduced enthalpy of the gases. The decrease in gas temperature corresponding to a decrease in the radiation intensity was calculated from Planck's radiation law. A wavelength of 3100 angstroms, initial gas temperature of 3470° K, and constant gas emissivity were assumed. An average radiation decrease in combustor A of 2.0 percent per inch results in a temperature decrease of 6° K. The corresponding decrease in the enthalpy of the gases results in a calculated heat-transfer rate of 4 Btu per square inch per second to the walls. This value is close to the measured rate of 4 to 5 Btu per square inch per second in a similar combustor (ref. 10). Ultraviolet radiation in combustor B decreased at approximately twice the rate measured in combustor A; the decrease indicates higher heat transfer to the walls during unstable combustion.

In a well-mixed adiabatic system, thermally excited ultraviolet intensities would rise to limiting values for each o/f and thereafter remain constant with increasing axial distance. If the limiting value of radiation is assumed to be reached when the reaction is completed, the maximum intensities in the two combustors indicate the approximate end of reaction. The axial distance required to complete the reaction in combustor A (fig. 3, p. 4) varied from 1.5 to 8 inches for a variation in oxidant-fuel weight ratio from 1 to 9. By contrast, the reaction was completed in approximately one-half this distance during unstable combustion in combustor B.

Infrared radiation is much less sensitive to a small temperature change in the gases, so heat-transfer effects on these radiation intensities are small. For example, it was previously shown that a 20-percent decrease in ultraviolet radiation at 3100 angstroms could be caused by only a 60° K decrease in temperature. This same temperature decrease would decrease H₂O radiation at 24,000 angstroms by only 3.3 percent. This effect is shown in figure 4(b)(p. 5) where the total radiation intensities at distances greater than 12 inches from the injector are nearly constant. The small variations that do

occur at low o/f's are probably due to slower overall mixing with large quantities of excess hydrogen. For the infrared data, the reaction was again assumed to be completed at maximum total intensities. In combustor A, the distance required for complete reaction varied from 1.5 to 12 inches for an o/f variation from 1 to 9. In combustor B, this distance was reduced to vary from 0.8 to 6 inches. The agreement between the infrared and ultraviolet radiation data indicates probable thermal equilibrium in the combustors.

Chemical Equilibrium

The previously indicated close agreement between the plate temperatures and the equilibrium gas temperatures in combustor A indicates probable chemical equilibrium of the products. A further check on both chemical and thermal equilibrium of the products can be made, based on chemical equilibrium temperatures and concentrations, by comparing the observed radiation to the calculated radiation.

Calculated OH intensities of the (0,0) band centered near 3100 angstroms vary with temperature according to the relation (ref. 11)

$$I_{\Delta\lambda} = \left(1 - e^{-K_{\Delta\lambda} P_{OH} L T_o / T_g}\right) \frac{C_1}{\lambda^5 (e^{C_2 / \lambda T_g} - 1)}$$

where

$I_{\Delta\lambda}$ band intensity of OH

$K_{\Delta\lambda}$ integrated absorption coefficient of OH band, assumed equal to 0.89 cm⁻¹ atm⁻¹ from reference 11

P_{OH} equilibrium concentration of OH in atmospheres (refs. 7 to 9)

L path length of 5 cm

T_o / T_g correction for temperatures different from T_o equal to 2877° K

C_1, C_2 constants in Planck's radiation law

λ effective wavelength of 3100 angstroms

T_g equilibrium gas temperature (refs. 7 to 9)

Calculated OH concentrations, temperatures, emissivities, and intensities are tabulated in the following table for mixture ratios between 1 and 10, an integrated absorption coefficient of the OH band of 0.89 per centimeter per atmosphere, a temperature (T_o) of 2877° K, and a path length of 5 centimeters:

Oxidant-fuel weight ratio, o/f	Equilibrium OH concentration, P_{OH} , atm	Equilibrium gas temperature, T_g , $^{\circ}K$	$K_{\Delta\lambda} P_{OH} L (T_o/T_g)$	Emissivity of OH band, $\epsilon_{\Delta\lambda}$	Relative blackbody radiance, $I_{\Delta\lambda}$	$\epsilon_{\Delta\lambda} I_{\Delta\lambda}$
1	-----	1260	-----	-----	-----	-----
2	0.0002	2030	0.001	0.001	0.05	-----
3	.022	2630	.11	.11	.9	0.01
4	.104	3030	.44	.36	9.3	3.3
6	1.176	3410	4.41	.988	51.1	51.0
8	2.116	3470	7.80	.9996	64.6	64.6
10	2.382	3420	8.94	.9999	53.1	53.1

These calculated intensities vary with o/f as shown in curve B in figure 6. If the OH emissivity is unity (exponent containing $K_{\Delta\lambda}$ is very large) at all mixture ratios, radiation intensities vary only with the temperature at each o/f as shown by curve A in figure 6. The hydroxyl radical in thermal and chemical equilibrium within the combustors is expected to vary with o/f in a manner similar to the calculated trends. Measured ultraviolet intensities 12 inches from the injector in combustors A and B were corrected for the heat-transfer losses mentioned previously, normalized to the calculated intensity at an o/f of 10, and plotted as curves C and D, respectively, in figure 6. The slopes of the calculated curves compare favorably with the curve of combustor B. Combustor A radiation does not conform to the trends in calculated intensities, especially since those intensities were much higher than the intensities calculated at the lower o/f ratios. Since the average gas temperatures in this combustor were close to the calculated equilibrium temperatures, the observed variation must be due to insufficient uniformity in the gases. The presence of local high temperature zones at low overall mixture ratios will increase the average OH intensities to abnormal values. This increase can be illustrated by averaging the intensities obtained from a sinusoidal variation in o/f. Varying the local o/f ± 20 percent above and below a value of 4 (3.2 to 4.8), increases the average OH intensity 1.95 times the intensity for a gas at an o/f of 4.

The lack of homogeneous gases in the stable combustor A appears to be due to the lack of adequate mixing. In combustor B mixing is accomplished by the high velocities associated with the axial pressure oscillation.

Intensity of water-vapor emission in the infrared region depends on the integrated intensities of bands at 11,000, 14,000, 19,000, and 27,000 angstroms (ref. 11). When the total radiation data at 12 inches from the injector in combustors A and B are compared to the radiation intensities of the tungsten plate, total emissivities of the gases are considerably below the plate emissivity (fig. 5). If the gases are at the calculated temperature of equilibrium products, the apparent emissivity of the gases in combustor A varied from 0.15 to 0.075. Apparent emissivities of combustor B products varied from 0.7 to 0.25.

The data that are available on water-vapor radiation (ref. 11) indicate that the gas emissivities in the combustors are less than 0.15. Combustor B

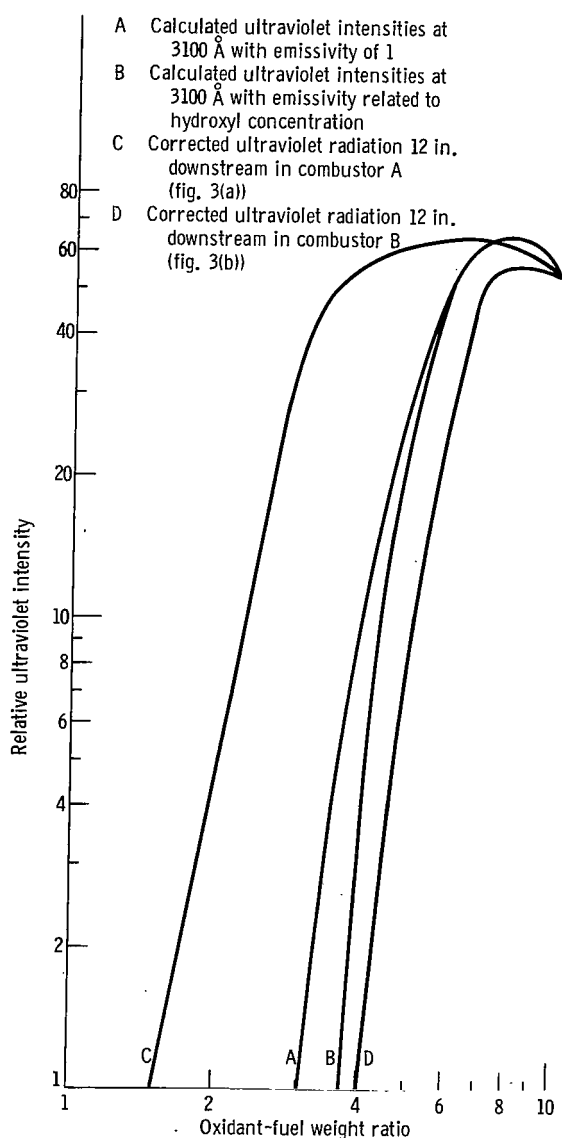


Figure 6. - Relative ultraviolet intensities as function of oxidant-fuel weight ratio.

metric mixture. Optical depth of the radiating gases increases directly with increased reaction, and hence the radiation intensity increases in direct proportion to the extent of reaction. After the reaction is complete (all of the O_2 or H_2 reacted), the excess reactant continues to mix with the products to form a homogeneous mixture that approaches the calculated equilibrium composition, temperature, and radiation intensity. During unstable combustion, the preceding processes occur in the same order but within a much shorter distance in the combustor.

gases, therefore, emit higher than expected intensities. Radiation from the chamber wall and quartz window may have contributed to at least part of the high radiation observed. An additional difference between the measured plate radiation and gas radiation can be due to a slight change in the solid angle observed by the pyrometer.

The temperature effect on radiation intensities is much lower in the infrared region, and the H_2O concentration increases much slower than OH concentration with o/f. The anomalous behavior of OH radiation in combustor A (fig. 6) does not appear in the infrared data in figure 5 (p. 6).

Extent of Reaction

The processes that make up the mechanisms of oxygen vaporization, mixing, reaction, and establishment of equilibrium products can be outlined on the basis of the preceding data. During stable combustion, OH and H_2O intensities near the injector are nearly independent of the overall oxidant-fuel weight ratio. Temperatures near the injector are also independent of the overall mixture ratio (unpublished NASA data obtained by Marshall Burrows and Ronald Razner). It is believed, therefore, that the reaction takes place in mantles around the liquid-oxygen jets at temperatures that approach those of a stoichio-

Turbulent mixing (refs. 2, 12, and unpublished NASA data obtained from

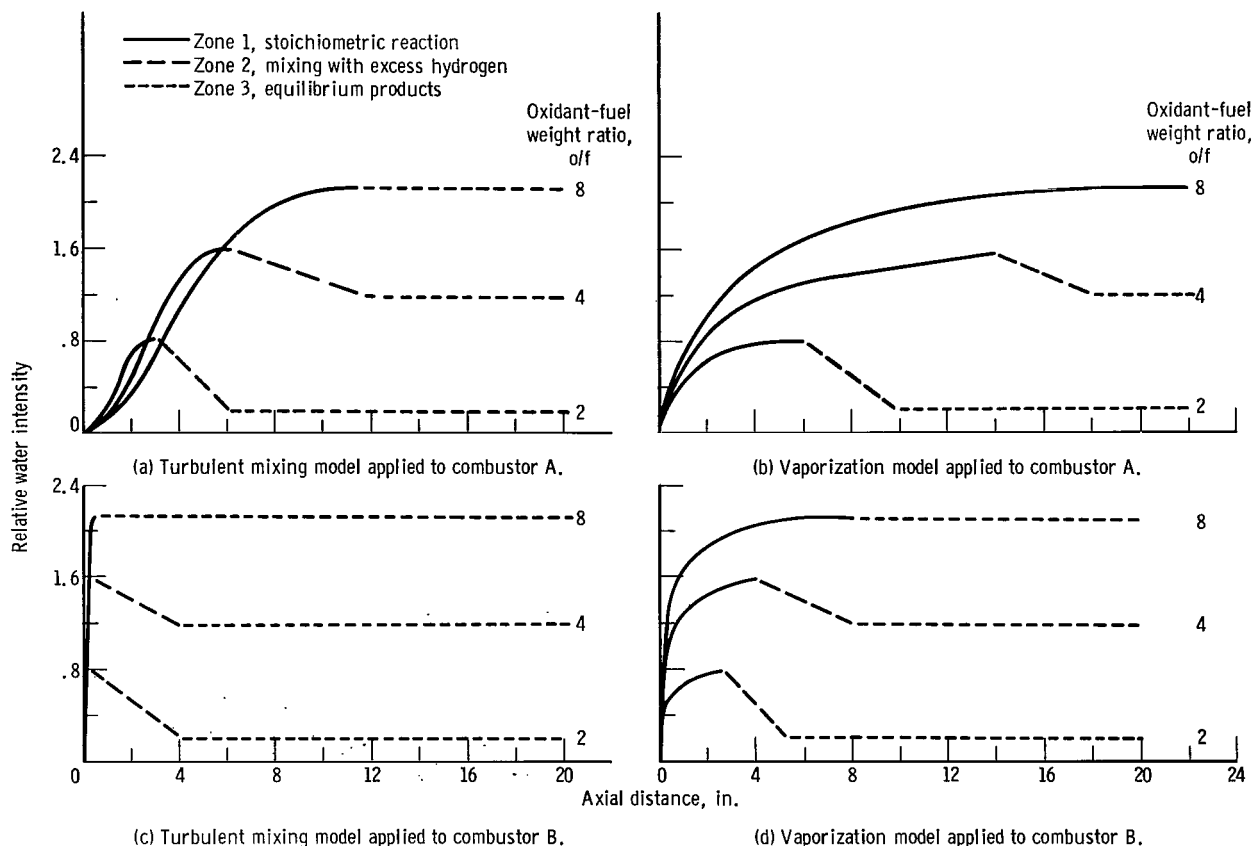


Figure 7. - Relative water intensities at 24,000 angstroms as function of axial distance from injector.

Martin Hersch) and vaporization (ref. 1) of the reactants have both been considered to have considerable influence on the extent of reaction in a combustor. The turbulent mixing model assumes that the diffusion of one reactant into the other is limited by the local turbulence intensity and the distance between the injection elements. The vaporization model considers reactant vaporization (oxygen in the present case) to control the extent of reaction.

The two models were used to predict the behavior of H_2O radiation at 24,000 angstroms in combustors A and B as a function of axial distance as shown in figure 7. The techniques used to calculate the extent of reaction by turbulent mixing are given in references 2, 12, and unpublished NASA data obtained from Martin Hersch; vaporization calculations are given in reference 1. The combustor was arbitrarily divided into the following three zones: (1) stoichiometric reaction, (2) mixing with excess hydrogen, and (3) uniform products. Radiation intensities in zone (1) varied with the concentration of H_2O in proportion to the extent of reaction. This zone was controlled by turbulent mixing or vaporization and is shown as a solid line. Radiation in zone (3) was proportional to the H_2O concentration at the temperature of equilibrium products (refs. 7 to 9), while the mixing behavior of zone (2) radiation was assumed. The latter zones are not important to this discussion and are shown as dotted and dashed lines, respectively.

Combustor parameters that influenced the turbulent mixing model were primarily the injector element spacing and the local turbulence intensity. These parameters differed by at least 20 times between combustors A and B and caused the reaction by turbulent mixing to be completed in less than 1 inch in combustor B (fig. 7(c)).

The vaporization of liquid oxygen was controlled largely by the median drop size of oxygen produced by the injector (ref. 1). Hence, the reaction at a mixture ratio of 8 was completed in 22 inches in combustor A (fig. 7(b)) and in 6 inches in combustor B (fig. 7(d)).

When the trends indicated by the calculated radiation in figure 7 are compared to the total measured intensities in figure 4 (p. 5), it appears that the extent of reaction is controlled by turbulent mixing in combustor A, and the reaction in combustor B is limited by vaporization. While refinements are desirable in the assumptions made in these calculations, the indicated trends show the merit of comparing radiation measurements with these or similar analytical models for determining the extent of reaction for various reactants and combustor configurations.

SUMMARY OF RESULTS

From this investigation of radiation processes related to oxygen-hydrogen combustion at high temperatures the following results were obtained:

(1) Hydroxyl (OH) and water (H_2O) molecules appear to be in local thermal and chemical equilibrium at all locations with the chamber of a liquid oxygen-gaseous hydrogen combustor.

(2) The required distance to react liquid oxygen with hydrogen was deduced from radiation data for a stable and unstable combustor. The reaction was completed in less than half the distance for unstable combustion.

(3) Ultraviolet intensities are seriously affected by the cooling of combustor gases through heat transfer to the walls and by the lack of uniformity in the gases. Errors in infrared intensities are possible because of the radiation emitted by the hot surfaces in the combustor.

(4) Rate processes related to the combustion of liquid oxygen and hydrogen, based on a constant temperature reaction with subsequent mixing of the products, are inferred from the data. Measured intensities were compared to calculated radiation that was assumed to be controlled by turbulent mixing or vaporization. The reaction in the stable combustor appeared to be controlled by turbulent mixing. Vaporization apparently limited the reaction in the unstable combustor.

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