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SPECTROSCOPIC STUDIES
OF IMPLODING PLASMA RINGS
IN DETONABLE GAS MIXTURES

by
Kenneth M. Foreman
Maurice E. Levy

Plasma Propulsion Laboratory
REPUBLIC AVIATION CORPORATION
Farmingdale, L. I., New York

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ABSTRACT

Spectroscopic observations have been made of the flow field behind a cylindrical hypersonic wave front resulting from an imploding electrical discharge into an initially low pressure quiescent gas. Sub-microsecond resolved spectra were obtained by use of a quartz prism monochromator-photomultiplier detector system with photographic recording of an oscilloscope displayed output signal. Electrically produced plasma rings were established in detonable as well as non-reactive gases.

Strong indications were observed that exothermic reactions, as evidenced by H_2O emission, proceed in $\text{H}_2 + 1/2 \text{O}_2$ reactant mixtures within less than one microsecond under certain initial low pressure conditions. A reaction threshold at 1 mm Hg A. pressure was found for the particular reaction vessel used.

Time-integrated spectrometer photos support the conclusions reached with the transient spectra analysis equipment.

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SECTION I - INTRODUCTION

One of the objectives of current magnetogasdynamic research is the use of imploding electrical arc discharges in low pressure gas mixtures, i.e., on the order of 1 mm Hg A., to induce and maintain a chemical detonation (Reference 1). Although reaction rates are known to slow down at such low pressures, it has been hoped that high intensity illumination from the imploding plasma ring, as well as forward diffusion of high energy particles, would sensitize the reactants and initiate reactions in times of the order of less than one microsecond.

In detonations it is not sufficient for the intermediate (endothermic) reactions to begin. For mixtures of hydrogen and oxygen, for example, there must be the exothermic recombination step leading to the formation of the water molecule, H_2O , before the detonation can be considered developed and be useful as a source of energy release. The detection of the water molecule in most conventional combustion processes is achieved usually by IR absorption spectroscopy (Reference 2). However, at the low densities such as associated with the present research, absorption is so slight as to preclude detection, especially in the microsecond order of time of the event. Fortunately, investigators have reported (Reference 3) that several emission band heads in the visible spectrum exist under electrical discharge conditions which are attributed to electronic excitation modes of the H_2O molecule.

The report that follows describes the initial preliminary experiments designed to reveal, by submicrosecond-resolved as well as time-integrated spectroscopy, the nature of reactions and their emission spectra in imploding discharges primarily in a stoichiometric hydrogen-oxygen mixture.

SECTION II - DESCRIPTION OF EXPERIMENTAL APPARATUS

A. PHOTOMULTIPLIER RECORDING

A Leiss single prism monochromator was used in conjunction with a photomultiplier tube for observation of the emission radiation from the plasma and its flow field. Figure 1 is a schematic of the experimental apparatus and their relative positions. Figure 2 shows photographs of the equipment. The light transmitted through the quartz window covering slit A, on top of the cylindrical discharge chamber, is reflected by mirror C. The image of the slit A is formed perpendicular to the monochromator entrance slit S_E . This allows observation of a particular part of the plasma flow field, essentially the part where the slit S_E intersects the slit A image. The light received at slit S_E is then dispersed in the monochromator by a quartz prism (refracting angle 30° - 35° , height 50 mm, length of hypotenuse 75 mm). The beam that passes through the exit slit S_x falls on the photocathode of a 6467 DuMont photomultiplier tube whose output is displayed as a transient trace on a Tectronix 551 oscilloscope and recorded with a Polaroid camera. Figure 3 is a graph of the calibration of the monochromator instrument and Figure 4 is a plot of the relative calibration of the photomultiplier tube. This tube has an S-11 response, and, therefore, its useful range is between 3600Å and 5300Å, with peak response near 4400Å. The spectral range of the monochromator, using standard prisms, is 200 millimicrons to 20 microns. Table I lists the nominal resolution characteristics of the monochromator with a quartz prism as its dispersing element.

The photomultiplier detector was operated at a supply voltage of 1800 volts, and the output voltage was in the linear region of response, thus making it possible to compare relative intensities of emission lines after correcting for the spectral response of the photomultiplier and the transmission of the quartz prism. The photomultiplier circuit contained a 50 ohm output impedance to match the impedance of the coaxial line (the line was not terminated at the oscilloscope). Extraneous

electrical noise signals were virtually eliminated by shielded enclosures around the photomultiplier and its connecting cables and, additionally, by keeping it a long distance (about 4 feet) from the discharge device.

TABLE I. MONOCHROMATOR WITH QUARTZ PRISM

Wave length	Linear Dispersion	Field of View Slit width ($S_E = S_x$)		
		0.03 mm	0.05 mm	0.10 mm
3000Å	70 Å/mm	4.2Å	7.0Å	14.0Å
3500	110	6.6	11.0	22.0
4000	170	10.2	17.0	35.0
5000	400	24.0	40.0	80.0
6000	600	36.0	60.0	120.0

B. PHOTOGRAPHIC RECORDING

A Spencer spectrometer (made by the American Optical Co.) was used for photographing a wide range of the discharge spectrum. This instrument has a focal length of 162.5 mm and an aperture of 25.4 mm, giving a relative speed of $f/6.4$. The accessory camera supplied with this instrument was modified to accept a Polaroid film-holder and used to record the emission radiation. The spectra photographed were recorded on Polaroid ASA 3000 speed film. The positioning of the entrance slit of the Spencer spectrometer was identical to the relative positioning of the entrance slit of the Leiss monochromator as shown in Figure 1. The entrance slit of the Spencer spectrometer is not calibrated. Therefore, it is not known what exact size openings were used when photographing detonation spectra, although they were approximately on the order of 0.5 mm. It should be noted that despite the wide slit opening used, the $f/6.4$ speed of the instrument, and the ASA 3000 speed of the film, about 15 superimposed discharge images were necessary in order to obtain a good photograph for any particular set of conditions. The discharge lasted for a period of about 10 microseconds; some emission lines or bands, however, persisted for a longer period of time.

C. DETONATION TEST STAND

A detailed description of the detonation test stand is found in the Tenth Quarterly Progress Report (September 1961 - November 1961), AFOSR Contract AF49(638)-552, "Energy Release and Transformation," Republic Aviation Corporation Report PPL-TR-61-25, December 29, 1961, p. 10-14.

SECTION III - EXPERIMENTAL PROGRAM

The objective of the series of experiments was to determine whether an imploding plasma produced by a transient electrical discharge into an $\text{H}_2 + 1/2 \text{O}_2$ mixture could induce an exothermic chemical reaction under various conditions of pressure and stored electrical energy (the identification of H_2O bands being considered a reliable indicator of an exothermic reaction).

A rotating-mirror camera was used to obtain streak pictures of the moving discharge on black and white and color film in order to aid in guiding and interpreting the spectroscopic study (Reference 4).

The test procedure consisted of obtaining a spectrograph photo of a major portion of the discharge spectrum in $\text{H}_2 + 1/2 \text{O}_2$ mixture and identifying the emission lines and bands present. Lines or bands of interest subsequently were investigated by means of photomultiplier time-resolved observations. This data then, was related to the streak photographs.

The H_2O emission band-head at $5126\text{\AA}$ (Reference 3) was investigated with particular care. This band-head occurs at a part of the spectrum relatively clear of H_2 and O_2 emission and, therefore, can be isolated with comparative ease. In the systematic identification procedure for this particular band, discharges were induced in different non-reacting gases in order to find other possible contributions of emission near that wavelength. Discharges in He revealed a small contribution from metallic elements such as Fe, Ni and Cr, presumably from the stainless steel electrodes. Discharges in O_2 produced spectra of the metallic oxides such as FeO, NiO and CrO as well as the O_2 bands and lines. Pure hydrogen also was used for the discharges and its Balmer series lines and molecular bands were observed. At times the non-reacting mixture, $\text{He} + 1/2 \text{O}_2$, was substituted for $\text{H}_2 + 1/2 \text{O}_2$ as a further check on the correct identification of H_2O bands.

The dependence of certain spectra on pressure and voltage was studied, along with their time resolved characteristics. It was found possible to distinguish between two emission spectra occurring at nearly the same wavelength by means of the distinct behavior of their time resolved characteristics when initial pressure or voltage are varied. Precautions were exercised during these experiments to maintain the quality of the data. The discharge chamber was filled with fresh gas before every discharge and was evacuated to about 10 microns' pressure after each discharge preparatory for the next filling. The detonable mixtures were carefully mixed and not used for a period of time, usually of the order of weeks, in order to insure complete diffusion of the component gases into each other.

The reaction chamber was filled with the test gas to within $\pm 5\%$ of the desired pressure. Prior to initiating a discharge, the capacitor voltage was recorded to within $\pm 2\%$. Electrical discharge was initiated usually within one minute after the reaction chamber was filled.

Most of the discharge observations were made at about the mid-radius of the reaction chamber where the imploding arc was fully developed. Some observations were made near the periphery of the chamber, where the discharge is first formed.

All the dual-beam oscilloscope trace photos display, simultaneously, the photomultiplier output signal on the upper beam and the discharge current signal (integrated Rogowski coil output) on the lower beam. The current signal triggers the oscilloscope sweep.

SECTION IV - EXPERIMENTAL RESULTS

A number of general trends were observed during the test program and are reported below. Radiation in the vicinity of 5190Å was detected during discharges in pure hydrogen. No hydrogen atomic lines are catalogued there, but since the maximum intensity coincided with times of peak current, it represents, probably, molecular electronic excitation. Some of the radiation intensity also can be attributed to background continuum.

Figure 5 includes typical photographs of this type of emission. Figure 6 contains photographs of the H_γ , H_δ and H_ϵ emission lines of atomic hydrogen. The nature of the emission in these instances varies from that observed near 5190Å where only molecular excitation could be expected. The three atomic hydrogen Balmer lines display similar time resolved characteristics but the emission persists after the passage of the current, presumably representing a recombination relaxation process. Figures 7 and 8 show photographs of emission at wavelengths identified with the first and second negative bands of molecular oxygen ions. The discharge in helium gas produces excitation of the 5876Å line, as recorded in Figure 9. This emission also persists considerably longer than the current pulse. The 3889Å He line was not found excited. Other observations of helium discharges in regions where no helium lines may be expected indicate that for initial pressures less than 1 mm Hg A. electrode (metallic) elements produce significant background radiation. For pressure greater than about 1 mm Hg A., the metallic radiation appears suppressed.

The above observations were made at a position where the imploding discharge (luminous front) would have to travel at least to the mid-radius of the discharge chamber. When hydrogen or helium gas was used, the photomultiplier trace of the luminous front showed a dependence on initial capacitor voltage and gas pressure, whereas with oxygen the luminous front displayed no well-behaved relation to discharge voltage or gas pressure. This is an indication that the

plasma formed in the oxygen gas probably is irregular and asymmetric. Furthermore, with oxygen the luminous front did not always proceed across the point of observation, resulting in a poor overall reproducibility of emission intensity under otherwise identical initial conditions of discharge. When oxygen emission spectra were monitored in mixtures containing hydrogen or helium, a more reproducible condition existed, presumably as a result of the formation of more symmetric plasma cylinders (see Figure 10).

A comparison of spectra from discharges in helium, $\text{H}_2 + 1/2 \text{O}_2$ and hydrogen under similar test conditions is given by Figure 11. The discharge in He indicates that there is negligible metallic background emission in the 5100-5200Å bandwidth. There is a very small amount of emission by H_2 electronic bands. However, the emission obtained for discharges in $\text{H}_2 + 1/2 \text{O}_2$ is significantly large, strongly suggesting that the radiation is the H_2O band originating at the 5126Å band-head.

Further comparisons from data of Figure 12 tend to reinforce this view. Monitoring emission in the 5120-5220Å range resulting from electrical discharges in three different gas compositions, O_2 , $\text{H}_2 + 1/2 \text{O}_2$ and $\text{He} + 1/2 \text{O}_2$, but keeping the partial pressure of O_2 the same, there is significantly greater intensity from the $\text{H}_2 + 1/2 \text{O}_2$ mixture than the other two, which can be attributed also to the H_2O emission band. It should be noted further that in the band-width of 5190-5230Å shown by Figure 13, essentially the same characteristics are seen as in Figure 12, indicating that the H_2O band starting at 5126Å contributes significant intensity over some 80 to 100 Å of width, degraded toward the red.

Additional checks were made on the photomultiplier data by taking full spectrum photographs (time integrated) with the modified Spencer spectrograph as shown by Figure 14. A comparison between the $\text{H}_2 + 1/2 \text{O}_2$ spectrum and that of O_2 and H_2 alone shows that there are two bands in the $\text{H}_2 + 1/2 \text{O}_2$ mixture spectrum that cannot otherwise be identified with either H_2 or O_2 radiation systems. One is near 5126Å and the other at about 5690Å. The former one (5126Å) could be caused by H_2O emission; the one near 5690Å has not been satisfactorily identified.

The data discussed to this point was obtained with reactant mixtures at pressures between 1 and 3 mm Hg A. However, tests also were conducted with reactant pressures up to 11 mm Hg A. and down to 0.2 mm Hg A. With initial capacitor potential at 4500 volts and recording the radiation at $5210\text{\AA} \pm 40$, it was determined (see Figure 15) that electrical discharges into mixtures at 2 to 4 mm Hg A. pressure produced the largest peak-amplitude signal. For reactant pressure greater than 6 mm Hg., the peak-amplitude was reduced to about 70% of the 2-4 mm Hg. pressure range signal. At all pressures except at 3 mm Hg the peak-amplitude occurred at 2 microseconds after current initiation; for the exception it occurred at 2.6 microseconds.

Going to the lower reactant pressure range indicated the apparent existence of an excitation threshold at 1 mm Hg, because in no case was a significant radiation signal observed at lower pressures. This threshold, furthermore, appears to be very critical; a reduction of but .05 mm Hg initial pressure from this level reduced the photomultiplier signal trace to about 5% of its higher pressure peak-amplitude.

SECTION V - DISCUSSION

In evaluating the results of this test program two factors must be kept in mind. First, the data was found to be highly reproducible for reactant pressures of 1 mm Hg or greater and, second, the interpretation that exothermic reactions can proceed in sub-microsecond times at low pressures is dependent on the correctness of Schuler and Woeldike's (Reference 3) conclusions about excitation of H_2O at the 5126Å band-head.

The former fact shows that the gas mixtures were homogeneous and non-varying, from day to day and batch-to-batch, when the standardized preparation technique was used (by contrast, anomalous results accompanied use of mixtures only a few hours old). The process, furthermore, cannot be random and does not appear to change over a period of a large number of discharges.

The dependence on correctness of previously published work is an unhappy consequence of diagnostic equipment limitations. The only moderate resolution of the spectrometer, compounded with the limitation of photomultiplier spectral response range, makes detailed investigations of non-equilibrium chemical reactions somewhat impractical. However, the information reported here does represent significant exploratory data. No satisfactory pure collision-process form of reaction kinetics can be advanced for the prompt appearance of H_2O apparently revealed by the spectrum (Reference 5). It must, therefore, be assumed that electrically produced plasma radiation or energetic particles are responsible for accelerating the water molecule reaction. However, if the interpretation by the authors of Reference 3 is incorrect, and the radiation at 5126Å B.H. is of ionic excitation, then a possible explanation is the reaction:

which is endothermic (Reference 6). The oxygen ion was in abundant evidence

during the discharge (see Figure 14). On the other hand, one would expect the intensity of H_2O^+ , if this is the true reaction product, to vary proportional to the initial voltage (which is not borne out by the data of Figure 13) and there would be no reasonable explanation on these grounds for the apparent threshold at 1 mm Hg reactant pressure. Undoubtedly, capability of monitoring other radiation wavelengths attributed (Reference 3) to H_2O would have made possible a more conclusive investigation.

The reaction threshold, incidentally, may have an analogue in the pressure characteristics of the first explosion limit (Reference 7). Thus, the observed pressure dependence may represent quenching effects introduced by the plasma contaminants from the reaction chamber walls. If this is so, then chemical kinetic theory indicates that a larger reaction chamber would reduce the reaction threshold pressure, and suggests this direction for future work.

At 3 mm Hg initial pressure the stoichiometric hydrogen-oxygen mixture produces a peak signal of about 1.6 volts, in a wavelength band including the 5126Å band-head, for a peak discharge current approximately 72,000 amperes (initial capacitor voltage of 4500v) at 2 microseconds after current initiation (see Figure 11). The signal decays to essentially zero after 14 microseconds. Comparison with the 3500 volt condition shows that the photomultiplier peak-output signal is proportional to the peak current magnitude. Comparison of equal initial voltage conditions with lower reactant pressure at 1.5 mm Hg (see Figure 12 and 13) indicates that the monitored radiation signal amplitude is less than proportional to the initial pressure, and not proportional to initial voltage at the lower pressure. These facts tend to support the view that the presence of H_2O , indicated by radiation at 5126Å, results from a radiation-sensitized exothermic chemical reaction rather than purely an ion impact process because the reaction emission yield decreases with pressure but does not change similarly and consistently with electric arc current.

Streak camera pictures of imploding discharges in detonable mixtures (Reference 4) at initial conditions of 4500 volts and 1.0 mm Hg pressure indicate an initial wave velocity, based on light-front measurements, of between 12,000 and 25,000 fps. Thus the conclusions pertinent to the formation of the water

molecule in times of the order of a microsecond are under circumstances of a flow field behind a hypersonic traveling wave-front induced by a plasma accelerated by its self-induced magnetic field. Color streak pictures reveal that a thin leading edge portion of the wave is of red content (which is characteristic of hydrogen in the test apparatus) followed by a wide "white" region (Reference 4). This suggests that excited molecular and atomic (H_{α}) hydrogen predominates at the front, while many diverse reactions occur in the "white-light" region (see also Figure 14) including that at 5126Å.

SECTION VI - CONCLUSIONS

A preliminary spectrographic study of imploding arc discharges in a detonable gas mixture has indicated that an exothermic chemical reaction probably is produced in association with a hypersonic wave flow process. This is evidenced by emission radiation apparently characteristic of H_2O excitation which proceeds within the order of a microsecond after electric current initiation (plasma formation) and persists as long as 12 to 14 microseconds in the test apparatus used.

A reaction threshold is observed at a reactant pressure of 1 mm Hg. Below this pressure level, no H_2O produced radiation is observed, whereas a reproducible positive signal of appreciable magnitude is detected up to at least 11 mm Hg. (electrical parameters being held constant).

The chemical reaction mechanism has not been revealed because of spectral analysis equipment limitations which similarly restrict determination of gas temperature, H_2O concentration and time variation of product gas composition. However, it is believed that the rapid exothermic reaction is at least partially attributable to photochemical processes caused by radiative transfer processes from the intensely luminous plasma.

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FIG. 1 EQUIPMENT SCHEMATIC

a. Overall View of Detonation Test Stand with Spectroscopic Analysis Equipment in Place

b. View of Test Stand Seen from Monochromator Position

FIGURE 2. DETONATION RESEARCH TEST STAND

FIGURE 3 MONOCHROMATOR CALIBRATION

FIG. 4 CALIBRATION OF DUMONT 6467
PHOTOMULTIPLIER FOR 1800 VOLT
DYNODE SUPPLY

FIGURE 5 TIME-RESOLVED SPECTRAL CHARACTERISTICS
OF ELECTRICAL DISCHARGES IN HYDROGEN

UPPER TRACE - PHOTOMULTIPLIER OUTPUT
LOWER TRACE - DISCHARGE CURRENT

FIGURE 6 TIME-RESOLVED SPECTRA OF THREE BALMER SERIES LINES FOR ELECTRICAL DISCHARGES IN HYDROGEN

FIGURE 7 TIME-RESOLVED SPECTRAL CHARACTERISTICS
OF ELECTRICAL DISCHARGES IN OXYGEN

$\lambda = 4150 \text{ \AA} \pm 20 (O_2^+ \text{ 2nd Negative})$
 SLIT WIDTH—0.05 mm

INITIAL PRESSURE

INITIAL VOLTAGE

0.5 mm Hg

1.0 mm Hg

1000V

UPPER TRACE - PHOTOMULTIPLIER OUTPUT

3000V

LOWER TRACE - DISCHARGE CURRENT

2000V

FIGURE 8 TIME-RESOLVED SPECTRAL CHARACTERISTICS OF ELECTRICAL DISCHARGES IN OXYGEN

UPPER TRACE - PHOTOMULTIPLIER OUTPUT

LOWER TRACE - DISCHARGE CURRENT

FIGURE 9 TIME-RESOLVED SPECTRA CIRCA $\text{He } 5876\text{\AA}^0$
LINE RESULTING FROM ELECTRICAL
DISCHARGES IN HELIUM

INITIAL PRESSURE

1 mm Hg

INITIAL
VOLTAGE

4500V

UPPER TRACE - PHOTOMULTIPLIER OUTPUT

$\lambda = 3970 \text{ \AA} \pm 14$ (O_2^+ 2nd negative)

4500V

LOWER TRACE - DISCHARGE CURRENT

FIGURE 10 TIME-RESOLVED SPECTRA OF ELECTRICAL DISCHARGES IN A STOICHIOMETRIC HYDROGEN-OXYGEN MIXTURE

FIGURE 11 TIME RESOLVED SPECTRA OF ELECTRICAL DISCHARGES IN HELIUM, HYDROGEN AND $H_2 + \frac{1}{2} O_2$ MIXTURE

$$\lambda = 5170\text{\AA} \pm 50$$

INITIAL PRESSURE 0.5 mm Hg
GAS OXYGEN

UPPER TRACE - PHOTOMULTIPLIER OUTPUT
LOWER TRACE - CURRENT COIL

1.5 mm Hg
 $\text{H}_2 + 1/2 \text{O}_2$

1.5 mm Hg
 $\text{He} + 1/2 \text{O}_2$

FIGURE 12 TIME-RESOLVED SPECTRA OF ELECTRICAL DISCHARGES IN OXYGEN, $\text{He} + 1/2 \text{O}_2$, AND $\text{H}_2 + 1/2 \text{O}_2$ MIXTURES

$$\lambda = 5210\text{\AA} \pm 20$$

SLIT WIDTH — 0.05 mm

INITIAL PRESSURE 0.5 mm Hg

GAS O_2

UPPER TRACE - PHOTOMULTIPLIER OUTPUT
LOWER TRACE - DISCHARGE CURRENT

1.5 mm Hg
 $\text{H}_2 + 1/2 \text{O}_2$

1.5 mm Hg
 $\text{He} + 1/2 \text{O}_2$

FIGURE 13 TIME-RESOLVED SPECTRAL CHARACTERISTICS
OF ELECTRICAL DISCHARGES IN OXYGEN,
 $\text{He} + 1/2 \text{O}_2$, AND $\text{H}_2 + 1/2 \text{O}_2$ MIXTURES

$H_2 + 1/2 O_2$ Mixture

~

3 mm HgA Pressure

~

4500 Volts

15 EXPOSURES

~

4500 VOLTS

FIGURE 14. TIME INTEGRATED SPECTRA OF MULTIPLE ELECTRICAL DISCHARGES IN HELIUM, HYDROGEN, OXYGEN, AND $H_2 + 1/2 O_2$ MIXTURE.

P_i = INITIAL PRESSURE

SLIT WIDTH = 0.10 mm

$\lambda = 5210 \text{ \AA} \pm 40$

INITIAL VOLTAGE - 4500 V

FIGURE 15 TIME-RESOLVED SPECTRAL CHARACTERISTICS
OF ELECTRICAL DISCHARGES IN $\text{H}_2 + 1/2 \text{ O}_2$
MIXTURE - VARIATION WITH REACTANT PRESSURE

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