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### Combustion in Heterogeneous Media, Solid and Hybrid Rocket Engines

#### Theme

The paper describes the results of measurements made in the flame zone above the burning surface of a composite solid propellant. The results are compared to other similar experimental results as well as to current theoretical results.

#### Content

Temperature measurements in the gaseous reaction zone close to the surface of a burning composite solid (polysulfide-AP) have been made using a modified line reversal pyrometer.

In conjunction with the pyrometer, a servo-controlled feedshaft was employed to drive the strand of propellant toward the temperature measurement region at the same rate at which the strand burned. The results of the temperature measurements showed that the gaseous reaction zone cannot be presented by a one-dimensional temperature profile. It was also found that the gaseous reaction zone extends up to a distance of approximately 1 mm from the surface for pressures from 1 to about 15 atmospheres. It is concluded that the gaseous reaction zone alone does not supply sufficient energy to the surface for sustaining the controlling surface reactions.

## Composite Propellant Combustion

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Temperature measurements in the gaseous reaction zone close to the surface of a burning composite solid propellant have been made using a modified line reversal pyrometer. In conjunction with the pyrometer, a servo-controlled feedshaft was employed to drive the strand of propellant toward the temperature measurement region at the same rate at which the strand burned. The results of the temperature measurement showed that the gaseous reaction zone cannot be represented by a one-dimensional temperature profile. It was also found that the gaseous reaction zone extends up to a distance of approximately 1 mm from the surface for pressures from 1 to about 15 atmospheres. It is concluded that the gaseous reaction zone alone does not supply sufficient energy to the surface for sustaining the controlling surface reactions.

### Introduction

ONE of the most widely debated issues regarding the steady-state combustion of composite solid propellants concerns the contribution of the heat generated in the gaseous reaction zone to the total heat necessary to sustain the controlling surface reactions. Indeed, the flame structure above the burning surface is not well understood and has been approximated as follows: a) occurring adjacent to the burning surface such that almost all of the energy required by surface decomposition reactions originates from the gas phase reactions<sup>1</sup>; b) occurring at an intermediate distance from the burning surface whereby only a fraction of the heat necessary to sustain surface decomposition reactions comes from the gas phase reactions<sup>2-4</sup>; and c) occurring at a distance from the burning surface great enough that the surface decomposition

reactions must depend on surface and subsurface exothermic reactions for their sustaining energy.<sup>5,6</sup>

The prime reason for the confusion is the lack of agreement in the results of experiments which are designed to determine the structure of the flame zone. Results have been obtained which show that the adiabatic flame temperature of the gas phase reaction occurs very close to the burning surface,<sup>7</sup> while other results<sup>8,9</sup> have shown that the reaction zone is two orders of magnitude larger than that reported in Refs. 1 and 2.

As a result of the discrepancies of the previous experimental work and the importance of determining the contribution of the energy feedback from the gas phase to the propellant surface, it was deemed important to attempt to reconcile the differences reported in the literature.

### Experimental Study

#### Approach

In all of the previous experimental approaches,<sup>7-9</sup> the procedure followed has consisted of fixing the temperature measurement point at a location below the future burning surface of the propellant sample. After the propellant is ignited, the surface burns past the measurement point such that a single temperature scan of the gaseous reaction zone is obtained.

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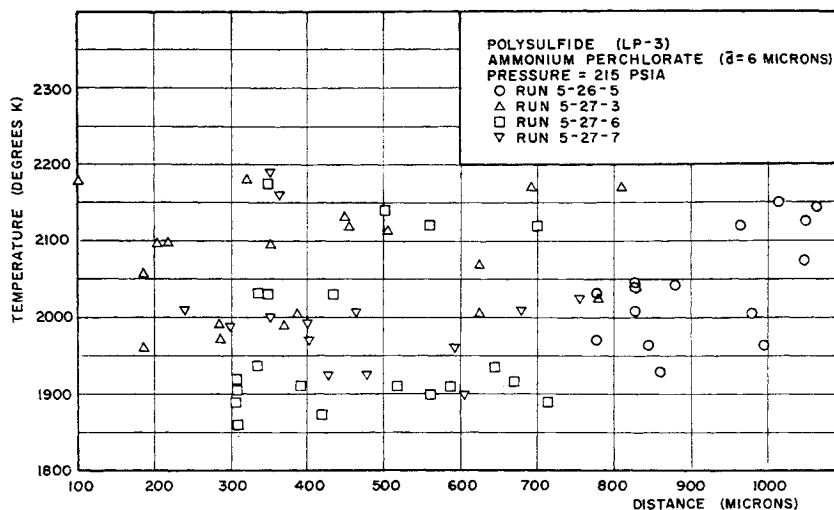


Fig. 1 Flame temperature measurement with servomotor on.

If the reaction zone is of an inhomogeneous nature, this experimental approach could lead to erroneous results in the determination of a temperature profile.

Thus, a temperature measurement system was developed<sup>10</sup> which measured the temperature over a fixed region in the flame for a controlled length of time. The system consisted of a modified line reversal pyrometer and a servo-controlled propellant feedshaft. The servomechanism served two purposes. The first was to drive the propellant strand toward the temperature measurement region at a rate equal to the propellant burning rate, enabling the examination of a zone in the flame for a controlled length of time. The second was to locate the burning surface with respect to the temperature measurement zone at any given time.

The burning surface was detected with a visible light beam position detection system.<sup>10</sup> In that system, a tungsten lamp was used to supply a beam of light over the burning surface of the propellant strand. The fraction of light passing by the strand was detected by a photomultiplier tube. As a result of the optical design, the output from the photomultiplier tube was proportional to the propellant position with respect to the temperature measurement region. To permit discrimination between the radiation from the tungsten lamp and the luminosity of the combustion zone, the light beam emanating from the lamp was chopped at a low audio frequency (450 cps) before passing over the burning surface.

The temperature measurement was accomplished by a modified line reversal pyrometer which was capable of a temperature measurement once every 2 msec. The D lines of sodium were used as the reversing lines and a calibrated tungsten strip lamp was employed as the comparison source. For a more complete description of the temperature measuring scheme see Ref. 10.

## Results

The propellant examined was a nonmetalized polysulfide-ammonium perchlorate propellant. Two unimodal oxidizer grind propellants were used in the study, a fine grind of mean diameter equal to 6  $\mu$  and a coarse grind of mean diameter equal to 50  $\mu$ . Each propellant contained 0.5% by weight of NaCl powder (mean diameter <5  $\mu$ ) to enhance sodium D line emission.

The results of the temperature measurements over the pressure range of from 1 to about 15 atm showed that the reaction zone is very inhomogeneous. In particular, the temperature measurements showed that no average, one-dimensional temperature profile exists above the burning surface of the propellant at any of the pressures examined. The temperature measurements of both the fine and coarse oxidizer grind propellants over the scanning range of the instrument

(up to 1000  $\mu$  above the surface) showed temperatures varying from the lower limit of accurate temperature measurement (1800°K) to the adiabatic flame temperature of the polysulfide propellant (about 2200°K).

A plot of the temperature measured for a fine oxidizer grind propellant burning at a pressure of 215 psia is shown in Fig. 1. Here it can be seen from the results of four separate runs that no specific distance above the surface exists for which a high temperature is favored.

The accuracy of the distance measurements was about  $\pm 15\mu$ , and that for the temperature measurement was about  $\pm 60^\circ\text{K}$ .<sup>10</sup> Neither of these will account for the scatter shown in Fig. 1.

In light of the discovery that the temperature measurement system did not yield a temperature profile as found in previous research programs,<sup>7-9</sup> an attempt was made to measure profiles in the same manner that had been employed in those investigations. That is, the servomotor was turned off and the burning surface was allowed to burn by the temperature measurement region. Results of this type are shown in Fig. 2. Here the temperatures were measured as a function of time and a distance for each temperature was calculated. The results show that a profile could be inferred where the maximum temperature is attained at a distance of about 300  $\mu$ . As a result, it appears that the gaseous reactions between the fuel and oxidizer are occurring both very close to the surface and at distances of the order of 1000  $\mu$  or more. At this point it may be helpful to make a rough estimate of the thickness of the reaction zone required in order that a significant energy transfer from that zone to the surface exists. Friedman<sup>11</sup> assumed that a heat flux of approximately 400 cal/cm<sup>2</sup>-sec was necessary to vaporize the surface of a typical composite solid propellant burning at a rate of 1 cm/sec. By equating the heat flux to  $\lambda(\Delta T/\Delta X)$ , where  $\lambda$  is the gas conductivity, and assuming  $\lambda = 0.0002$  cal/cm-sec/°C and  $\Delta T = 2000^\circ\text{C}$ , Friedman found that the height of the flame above the surface must be about 10  $\mu$ . It can be seen, then, that the gaseous reaction zone is quite small for the case of complete energy transfer from the gas phase to the surface.

Another interesting result of the experiments was that of finding more frequent temperature fluctuations in the gases at distances of about 500  $\mu$  above the surface of the coarse oxidizer grind propellant when compared to the fluctuations at about the same distance for the fine oxidizer grind propellant. The indication here is that the gaseous reactions between the fuel and oxidizer are occurring farther from the surface for the coarse oxidizer grind propellant. As a result, the energy transfer from the gaseous phase reactions to the surface should be less for the coarse oxidizer grind propellant. This decrease in energy transfer from the gas phase for the larger oxidizer

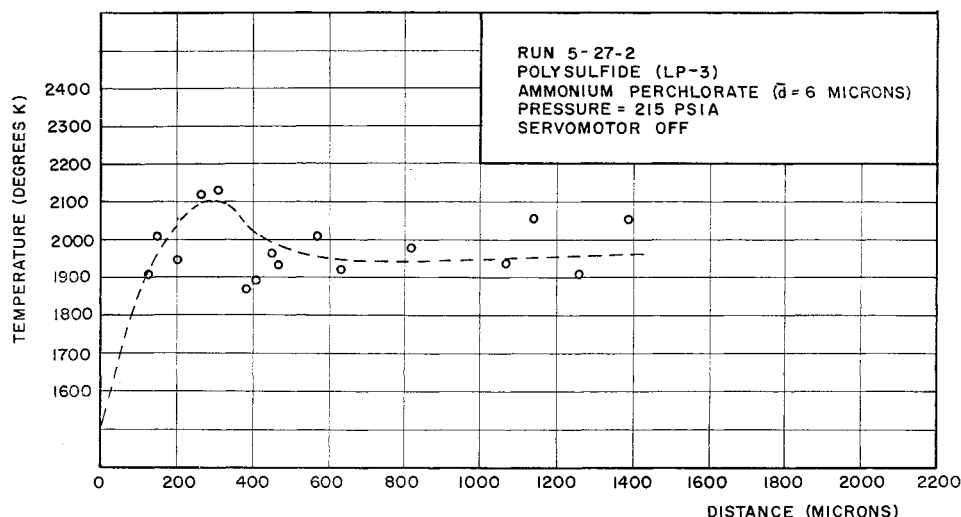


Fig. 2 Temperature profile measurements with servo-motor off.

provides an explanation for lower propellant burning rate with increasing oxidizer particle size.

It should be noted that although results for measurements within  $100\ \mu$  of the surface are considered here, it is recognized that some error in distance and temperature measurements prevails at these small distances. These errors result from partial masking of the tungsten lamp radiation passing over the propellant surface and propellant surface irregularities.

### Discussion

An important result that arises from the postulated inhomogeneous and extended gaseous reaction zone structure is the key to understanding the discrepancies of previous investigations.<sup>7-9,12</sup> If the reaction zone is of an inhomogeneous nature where the inhomogeneities could be of the order of hundreds of microns, the experimental approaches of previous attempts to measure temperature profiles could lead to erroneous results. In all of the past experimental approaches, the procedure followed has consisted of fixing the temperature measurement point (or region) at a point below the surface of the propellant sample. The propellant is ignited and the surface then burns past the measurement point such that a single temperature scan of the gaseous reaction zone is obtained. Since the gaseous reactions are occurring at various points throughout the zone traversed by the temperature measurement point and the locations of these reactions are changing constantly as the surface regresses away from the temperature measurement point, it is conceivable that almost any shape of temperature profile is possible above the burning surface. Thus, single scan temperature measurements can lead to serious errors in the study of the combustion zone structure.

From Sutherland's temperature measurement,<sup>7</sup> the following results are noted. First, the adiabatic flame temperature was reached at a distance of  $100\ \mu$  from the surface. The second is that the flame zone was microscopically inhomogeneous with inhomogeneities of the order of at least  $10\ \mu$ , a situation which was inferred from the rippling of the signal when the radiation from the flame was recorded.

The technique for measuring the temperatures as employed by Sutherland was basically the same as that employed in this investigation.<sup>10</sup> The major difference was the employment of a single temperature scan technique in Sutherland's experiments instead of a surface controlling technique as employed in the present experimental approach. It would be expected that the experimental results of these programs agree. Upon comparison of the results, it appears that agreement occurs in the existence of high temperatures close to the surface as well as the occurrence of temperature fluctuations above the surface.

However, the conclusions drawn from the experimental results are not in agreement. In Sutherland's work, the conclu-

sion is made that the results lend support to a gaseous flame adjacent to the burning surface that reduces the need of energy manifested from surface and subsurface reactions to sustain the over all combustion process. It is evident from the previous comments pertaining to errors in measurements less than  $100\ \mu$  from the propellant surface and gas phase inhomogeneities, that such a conclusion is unfounded if it is based upon results obtained from a single temperature scan experiment. If Sutherland had introduced a means of measuring the temperature over a fixed region with respect to the burning surface as done in this experimental approach, the presence of temperature fluctuations over that zone would have been established. Thus, it is proposed that the conclusion of a steep temperature gradient at the surface would not have been made.

The results of Penzias<sup>8</sup> showed an extended reaction zone which substantiate the conclusions of the controlled surface position experiment. However, this was again a single scan technique, and it is emphasized that the possibility of error is present in locating the distance where the maximum temperature occurs.

An interesting result included in Penzias' work is an observed temperature fluctuation measured at a wavelength of  $3.30\ \mu$ . Thus, the results of Penzias at this wavelength are also consistent with the conclusion that the gaseous reaction zone is of a very inhomogeneous nature.

The results of Sabadell's thermocouple experiments<sup>9</sup> are again based on a single temperature scan of the gaseous reaction zone. The results of the study are again consistent with the postulated extensive gaseous reaction region. In fact, Sabadell suggested the existence of such a reaction zone to explain the very slow rise in temperature experienced by the thermocouple after emerging from the burning surface. The absence of temperature fluctuations from the thermocouple is not surprising due to the thermal lag present in the thermocouple output.

Finally, the results of spectroscopic studies<sup>12</sup> of the flame in which sites of CN radiation were measured confirm the results established in this investigation. The results showed that the reaction zone extended to distances of about 1 to 2 mm above the surface. In regard to fluctuations in the reaction zone, the spectrographic technique is not suitable for drawing meaningful results. The radiation is recorded on a photographic plate which integrates the total radiation from the flame over the time the strand burns by the detecting slit. Thus, the output is not known for very short time intervals.

### Conclusions

The conclusion, then, is that the gaseous reaction zone of a composite propellant is not confined to a very thin layer adjacent to the propellant surface such that the gaseous reaction

between the fuel and oxidizer is the sole source of energy for sustaining the controlling surface reactions. Also, it has been found that previous attempts to measure the heat-transfer characteristics of the gaseous phase by means of single temperature scans of that zone have led to erroneous conclusions in some cases.

In regard to the over all combustion process of composite propellants, the results of this investigation indicate that heat generation at or below the propellant surface may be necessary in order to supplement the energy transfer from the gaseous reaction zone to the propellant surface. However, the finding of greater temperature fluctuations in the gases above the surface of the coarse oxidizer grind propellant when compared to the fine oxidizer grind propellant indicates that the gaseous reactions between fuel and oxidizer must not be ignored completely. Furthermore, it is felt that inhomogeneities in the gaseous reaction zone are sufficiently large to preclude the validity of a one-dimensional temperature profile to represent heat-transfer processes.

In light of the findings of this research program, it is concluded that one-dimensional theoretical and experimental approaches to the steady-state combustion of a composite solid propellant will never supply the answers to the more complicated areas of nonsteady combustion.<sup>13</sup> In particular, it appears that studies in the gaseous phase must now be directed towards understanding reactions that occur in a zone which is at least 1000  $\mu$  thick and contains inhomogeneities of the order of several hundred microns; as a result, a three-dimensional approach appears necessary to represent the combustion process.

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