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Ignition and Surface Temperatures of Double Base Propellants at Low Pressure: I. Thermocouple Measurements

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The autoignition, ignition, and surface temperatures of M-2 double base propellant were determined at low pressures. The autoignition temperature is defined to be equal to the initial temperature of the propellant at which the burning rate approaches infinity, which was found to be 145°C for M-2 propellant. The temperature at which a noticeable self-sustaining deflagration of the propellant first occurs (ignition temperature) was determined by imbedding a thermocouple at one end of the propellant specimen subject to a radiant flux. At 1 psia, the ignition temperature was found to be 214°C. The surface temperatures of the double-base propellant for steady-state burning were determined by imbedded thermocouple wires of various sizes, at pressures of 5, 10, and 15 psia. These thermocouple measurements were corrected by using a theoretical model of the thermocouple response characteristics. The correct surface temperature was determined by checking whether an assumed value predicted the experimentally measured bead temperatures of ½-, 1-, 2-, and 3-mil thermocouple wires. The emergence of the thermocouple bead from the solid into the gas phase was determined by high-speed motion pictures and is invariably associated with a plateau in the oscillograph recording of the temperature profile. The surface temperature predicted at these low pressures is about 300°C.

Nomenclature

A = surface area
B = $\rho C_p r/k$ of propellant
C_1, C_2, etc. = parameters defined in the text
C_p = specific heat of propellant at constant pressure
C_{pw} = specific heat of thermocouple wire at constant pressure
D = diameter of thermocouple bead
h = heat-transfer coefficient
k = thermal conductivity (of propellant, when used without a subscript)
L = length of thermocouple wire imbedded in propellant
P = length of thermocouple wire periphery
Q_{lead} = heat loss rate through thermocouple lead wires
r = burning rate
R = radius of thermocouple wire
t = time
T = temperature
V = volume
X,Y = coordinates defined in the text
ρ = density (of propellant, when used without a subscript)
α = angle between lead wire and axis of propellant

Subscripts

o = initial condition
B = thermocouple bead
p = propellant
s = burning surface
w = thermocouple wire

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Introduction

ONE of the important parameters that should be known in investigating the deflagration of solid propellants is the propellant temperature at various stages of burning. Without precisely determined values of the propellant temperatures as well as other parameters, the validity of a theoretical model cannot be tested. However, experimental results clarifying the role of these parameters for double base propellants are scarce. As a consequence, many of the multiparameter theoretical models are compared with only one or two limited experimental variables by varying the parameters to fit the limited experimental results, e.g., burning rate vs pressure. This paper is based on work done to isolate various parameters experimentally and to investigate their roles, and is confined to the propellant temperatures at various stages of deflagration.

Although the importance of the ignition, autoignition, and surface temperatures has been recognized, the experimentally measured values do not always agree, as Friedman[1] pointed out. Klein et al.,[2] Heller and Gordon,[3] Strittmater et al.,[4] and Sabadell et al.[5] measured the surface temperatures of double base propellants during steady-state deflagration at various ambient pressures. Powling and Smith[6] used an optical technique that is carefully studied in the companion article.[7] Figure 1 illustrates the extent of the scatter of the reported experimental results. The double base propellants used in these experiments were not the same in chemical composition and might have had different calorific values; nevertheless, the scatter of the surface temperatures measured is certainly more than what one would expect from such experiments, unless special additives were used that drastically affect the temperature. A review of the experimental data reveals that the variation between the experimental results can be attributed to the neglect of the thermocouple response characteristics and the erroneous identification of the burning surface in the experimentally determined temperature profiles.

Recently, there appeared two Russian papers by Zenin[8] and by Aleksandrov et al.[9] on the surface temperatures. Their experimental results were similar to part of the results presented in this paper, including the existence of the plateau in the temperature profile. However, their results were not corrected for the thermocouple response characteristics.

On the matter of ignition, one of the major difficulties has been in the definitions of ignition and autoignition. The appearance of flash or smoke, or the initiation of sustained burning, has been used as the ignition criterion. These criteria are not readily applicable, as it is difficult to assess the temperature associated with them. Furthermore, such criteria cannot distinguish between the minimum temperature at which the runaway reaction can take place because of its own heat of reaction under adiabatic conditions and the minimum temperature at which the runaway deflagration can occur under given operating conditions. These two temperatures need to be distinguished, as the former represents basic physical and chemical properties of the propellant, whereas the latter represents the general performance criterion. In this paper, "autoignition temperature" is used to imply the former and "ignition temperature" the latter.

Table 1 Physical properties and composition of M-2 propellant

Properties of M-2 double-base propellant	
Solid density	1.50–1.64 g/cc
Thermal conductivity	5.5×10^{-4} cal/cm sec°K
Specific heat	0.37 cal/g
Ratio of specific heats	1.191
Heat of explosion	1060 cal/g
Composition, wt %	
Nitrocellulose	76.65
Nitroglycerine	19.90
Graphite	0.30
Diphenylamine	0.65
Potassium nitrate	1.00
Barium nitrate	1.50
	100.00

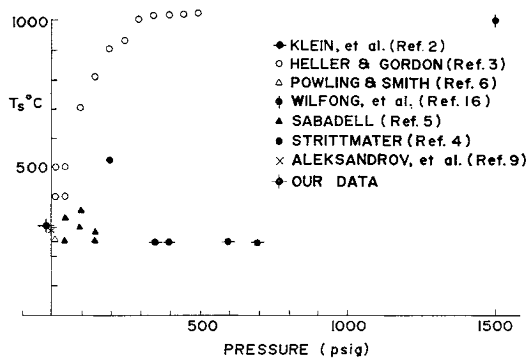

Fig. 1 Surface temperature vs pressure data collected from published literature.

This paper clearly establishes the autoignition temperature, the ignition temperature, and the surface temperature, primarily at low pressures. All these temperatures are measured by thermocouples. Since the thermocouples are used in a rapidly changing temperature field, the experimentally obtained data are analyzed by using a theoretical model for the thermocouple response and lead loss. The theoretical model is used to predict experimentally determined thermocouple bead temperatures for various sized thermocouples. M-2 propellant, whose physical properties and composition are given in Table 1, was used throughout the work.

Definitions and Outline of Experiments

The autoignition temperature will be defined as the temperature at which a propellant will deflagrate completely once an entire sample is uniformly brought to this temperature and kept adiabatically. In order to determine the autoignition temperature a series of experiments is performed in which the temperature of a strand of propellant is slowly raised to some predetermined value by passing a hot gas at that temperature around the propellant. Having reached this desired initial temperature, the propellant is ignited under constant pressure and the burning rate is recorded. As the initial temperature approaches the autoignition temperature, it is expected that the burning rate will approach infinity asymptotically. This asymptotic technique should be reliable, since it eliminates the possibility of accidental ignition near the autoignition temperature by a hot spot during heating. This technique assumes that as the initial temperature of the propellant is raised the chemical composition of the propellant does not change. The validity of this assumption is supported by the measurements of a differential scanning calorimeter, which show that there are neither exothermic nor endothermic reactions until the propellant temperature approaches the autoignition temperature.[10]

The ignition temperature will be defined as the surface temperature at which the propellant starts to deflagrate in given surrounding conditions. Therefore, the ignition temperature depends on the particular experimental conditions including the geometry of the propellant and heat flux. The ignition temperature is meaningful, however, in that it approximately defines the minimum surface temperature for self-deflagration under normally encountered ignition conditions. The ignition temperature is measured by imbedding a

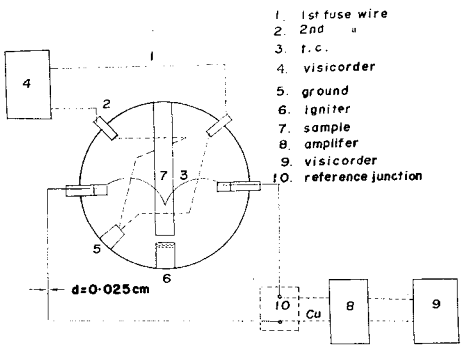

Fig. 2 Schematic representation of the combustion chamber and experimental arrangement.

thermocouple at the surface of the propellant and subjecting the surface to an external radiant flux.

The surface temperature will be defined as the temperature of the solid surface under steady-state burning conditions. That is, after the ignition temperature is reached, the burning rate and the surface temperature change until steady-state burning is attained. The surface temperature is measured experimentally by placing a thermocouple inside the propellant. The experimentally measured values are corrected for the thermocouple response and lead loss.

Experimental Arrangement and Procedure

Auto-Ignition Temperature Measurement

The general experimental arrangement is shown in Fig. 2. The chamber was made of stainless steel. The propellant was coated with Epon Adhesive 946 (0.003 cm thick) for planar burning. The coated propellant was cut into 7.00-cm lengths. Into each length, two No.-80-size holes for the indium burn wires and one No.-75-size hole for the thermocouple were drilled. The propellant then was placed in the test chamber and the proper electrical connections were made. The indium wire used was Indalloy Intermediate Solder No. 6, 30 gage, with melting point of 280–285°C. The thermocouple was made of 30 gage, single-strand enameled iron and constantan wires joined together by resistance welding.

The propellant then was raised slowly to some desired temperature by passing heated air around it, and the temperature history was recorded. Once the propellant reached the desired temperature, it was ignited by a platinum hot wire heater. The burning rate was determined from the melting times of the burn wires. The foregoing procedure was used with air as the surrounding atmosphere for the propellant. For the series of tests done with argon as the surrounding atmosphere the air was replaced by argon 2 min before the propellant was ignited.

Ignition Temperature Measurement

For the ignition temperature measurements, a 1-mil Alumel-Chromel thermocouple was imbedded at the end of the specimen to be subjected to a known radiant heat flux at 1 psia. At such a low pressure there is no flame, so the radiant heat flux to the propellant was constant for all practical purposes. The emf was amplified 10 times by a Honeywell Accudata 117 amplifier. The amplified signal caused a deflection of a Honeywell M600-350 galvanometer in the 1508 Visicorder. The radiation flux level was 0.5 cal/cm^2-sec at the propellant surface. The opposite end of the propellant sample was loaded with a spring so that the thermocouple was pressed against the propellant surface even after ignition. The ignition temperatures were independent of the tension applied to the thermocouple leads.

The chamber used for this series of experiments was different from that used in the preceding section, but for the purpose of acquiring the information presented in this paper they were essentially the same. This chamber allowed the burning rate to be recorded by high-speed photography and the external radiant flux to be applied to the propellant surface. During the experiments the temperature recording and the movie were synchronized by timing lights on the film strip and timing pulses on the Visicorder record. For a more detailed description, see Thompson and Suh.[11]

Surface Temperature Measurements

In these experiments the chamber used was the same as that used for the autoignition temperature, except when movies were taken to determine when the imbedded thermocouple emerged from the surface. The propellant was placed vertically in the chamber and was ignited at the lower end by the platinum wire heater. Leads were passed through the chamber walls for recording the burning rate and temperature, as shown in Fig. 2. A quartz window was placed at one end of the chamber so that the burning process could be observed. The position of the propellant was changed when the movie was taken, but the orientation of the propellant had no appreciable effect either on the burning rate or on the temperature profile. These experiments were run in argon.

Cylindrical propellant strands of 0.45-cm diam were cut into pieces 6.35 cm long, then one half of each strand was milled away (Fig. 3). A V-shaped thermocouple (platinum—platinum 10% rhodium thermocouple) was then imbedded in one half, and a similar piece was bonded to it with acetone and allowed to dry under pressure for at least one week at 45°C. The angle between the thermocouple leads was 40°. The diameter of the thermocouple bead junction is about three times that of the thermocouple wire. The orientation of the thermocouple is shown in Fig. 3. In order to have planar burning, the sides of the propellant were coated. After the coating had dried completely, two holes were drilled through the thermocouple-imbedded propellant for the indium burn wires. Experiments also were performed using Alumel-Chromel thermocouples in order to insure that thermocouples do not catalyze reactions. The results were the same, indicating there were no catalytic effects.

The strand was then placed in the combustion chamber filled with argon (Fig. 2). The thermocouple leads were passed through the chamber to an amplifier where the signal was amplified before entering the Honeywell Visicorder galvanometer (fluid-damped M 1650). By measuring the burning rate simultaneously, the emf vs time curve was converted to the temperature vs distance curve. The Visicorder paper speed was set at 80 in./sec so that the temperature change within a very small interval, say 0.001 sec, could be examined. This is the major difference between these experiments and those reported in the literature.

In order to identify the surface temperature of the burning propellant, a high-speed movie camera (Fastax 16 mm WF-14)

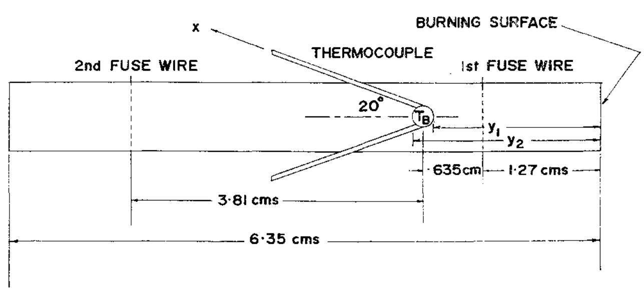

Fig. 3 Thermocouple position in the propellant.

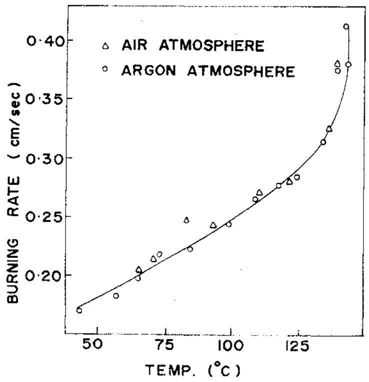

Fig. 4 Experimental results of burning rates at ambient pressure and various initial propellant temperatures in air and argon.

was used. The camera was specially focused so that the moment when the thermocouple bead junction emerged from the burning propellant could be identified. The timing of the camera was synchronized with that of the thermocouple output by feeding both 60 cps sine waves and 1000 cps square waves into the Visicorder and the camera. By examining the timing lights on the movie film, the corresponding temperature reading from the thermocouple output could be identified.

Experimental Results and Discussion

Autoignition Temperature

Figure 4 shows the experimental results for burning rate as a function of initial temperature. The experimental results obtained using both argon and air fall on the same line. The asymptotic temperature at which the burning rate approaches infinity is found to be 145°C. If the propellant is brought up to this temperature and isolated completely, its temperature will rise because of self heating and finally the propellant will deflagrate.

It is interesting to note in Fig. 5 that when the initial temperature vs the burning rate at 65, 115, and 165 psia is plotted, there is a common point at 15.5°C at which the pressure appears to have no effect on the burning rate. An extrapolation of the experimental results to initial temperatures below 15.5° indicates that the burning rate should decrease with increasing pressure. However, the burning rate data obtained at atmospheric (Fig. 4) and subatmospheric pressures (Suh and Clary[12]) do not intersect at this common point. In fact, these low-pressure curves are nearly parallel to each other. This comparison of the high-pressure data with the subatmospheric data indicates that the burning rate is controlled by different mechanisms in these two pressure regimes. It is interesting to note further that the common temperature is approximately equal to the self-extinction

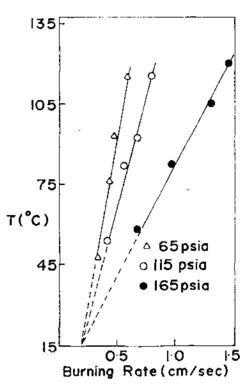

Fig. 5 Burning rate vs initial temperature of the propellant at various environmental pressures (argon).

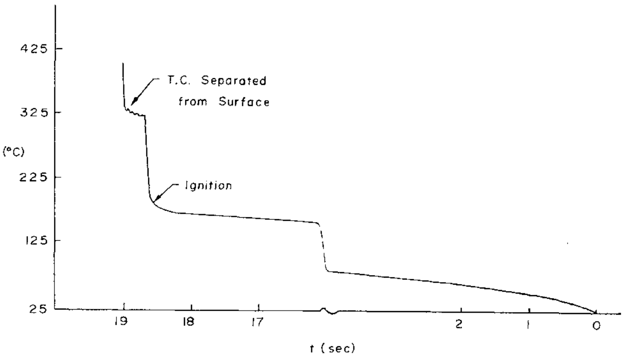

Fig. 6 Temperature history recorded by a thermocouple at the propellant surface under external radiant heat flux.

temperature reported by Suh and Clary.[12] Therefore, the common point may be thought of as the limiting temperature at which the heat-transfer rate to the solid from the gas is equal to the rate of the thermal energy required to sustain the steady-state burning.

Ignition Temperature

A typical temperature history recorded by the thermocouple at the surface is given in Fig. 6. The ignition temperature is indicated in the figure. If the propellant surface temperature reaches this value, runaway deflagration occurs. Under the test conditions the average ignition temperature of five runs was 213.6 ± 16°C, as determined from the recorded temperature profiles. As soon as the ignition flux is turned on, the temperature starts to rise slowly. At the instant ignition occurs, the temperature suddenly rises to a higher value and remains constant while the thermocouple remains pressed against the propellant surface. At the higher flux rates the ignition temperature appears lower than the above value. The determination of the ignition temperature can be somewhat arbitrary, since the beginning of the runaway deflagration is not well defined.

Surface Temperature

Data

A typical temperature-time record is shown in Fig. 7. The temperature curves are very reproducible up to the plateau and some of them are superimposable. After the plateau, however, the curves are not always reproducible, although most of them are similar. Sometimes the temperature drops down after the plateau and then rises again. The photographic records suggest two reasons for this. First, propellant particles stick on the thermocouple junction; second, in some experiments the deflagration oscillates across the surface. In the absence of these anomalies, these temperature profiles indicate that there are distinct gas-phase reactions that strongly depend on pressure (Thompson and Suh[13]).

Characteristics of plateau

If the heat input to the thermocouple junction is equal to the heat loss through the thermocouple leads, then there will

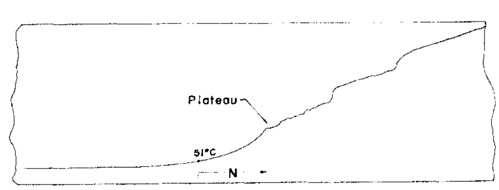

Fig. 7 Temperature profile measured using a thermocouple imbedded in the propellant.

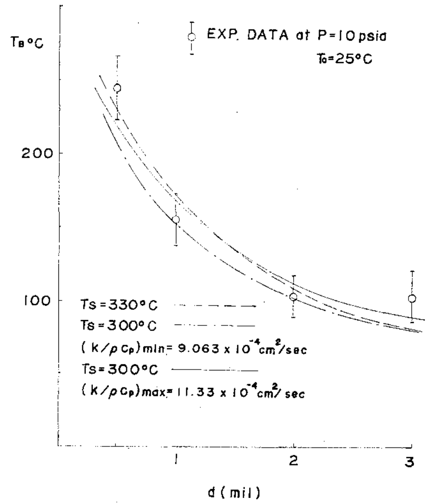

Fig. 8 Comparison of the experimentally and theoretically determined apparent surface temperature (bead temperatures) at 5 psia.

be a plateau on the temperature curve. When the thermocouple is entirely in the solid, the heat-transfer coefficient between the thermocouple and the propellant is uniform, so the temperature rises continuously as long as the surrounding temperature increases, provided the lead loss is less than heat input to the bead and wire. Similarly, if the thermocouple is entirely in the gas region, including a part of the lead wires, there cannot be a plateau. On the other hand, if the thermocouple junction is partially in the solid and partially in the gas, two different heat-transfer modes exist at the surface with two different heat-transfer coefficients, i.e., h_g in the gas and h_s in the solid. Generally, h_g is smaller than h_s; hence, the heat transfer into the junction may equal the heat loss through the leads and the plateau may result.

It is possible that there is an isothermal zone at the surface. However, this is not found to be the case. An isothermal zone near the surface of the propellant should result in a plateau for ½-mil thermocouples longer than or equal to that for 1-mil thermocouples. However, the opposite results were obtained. The plateau length depends upon the thermocouple bead size. The larger the bead size the longer the bead will be at the surface, which makes the plateau longer. Therefore, the isothermal reaction-zone hypothesis does not seem to be correct.

The length of the plateau is proportional to the thermocouple wire diameter (the larger the wire diameter the longer the plateau), but the temperatures indicated by the plateaus are inversely proportional to the thermocouple size. If we take 51°C as a reference point, then the plateau occurs at N second away from the reference point. N is found to be inversely proportional to the thermocouple size, as shown in Table 2.

On the basis of the experimental findings, it is concluded that the plateau temperature is the thermocouple bead temperature when it is at the surface. At the initial temperature of 25°C and ambient pressures of 5 psia, 10 psia, and 15 psia the surface temperatures indicated by ½-mil, 1-mil, 2-mil, and 3-mil thermocouples are 250°, 165°, 105°, and 100°C, respectively. That these are surface measurements is supported by a semilog plot of the temperature profile and high-speed photography. In the past, the point at which the experimental temperature profile departs from linearity was taken as the surface temperature. When this method is applied to our data, most of the experimental data yielded 280°C as the surface temperature for ½-m l thermocouples and 165°C for 1-mil thermocouples. These values are close to the plateau temperature. A high-speed photographic record was taken at 500–600 frames/sec. From careful observation of the movie and temperature profile, it was found that the appearance of the plateau region in the temperature profile coincides with the partial emergence of the thermocouple junction from the propellant surface.

Comparison with published data

Most of the results in the published literature report smooth temperature curves even in the gas region. Klein et al.,[2] Heller and Gordon,[3] and Sabadell et al.,[5] claimed that the temperature profiles were reproducible. Furthermore, none of them reported the existence of the plateau region in the temperature profiles.

In reviewing the published results of others, it was found that the temperature-recording speed was too low and that the amplification of the thermocouple output was small. Therefore, the details of the temperature profile could not be observed, thus losing much of the significant data. In the present experiments the thermocouple output was amplified approximately 250 times so that the galvanometer had a deflection sensitivity of 1 mv/in., and the paper speed was 80 in./sec.

Reliability of Thermocouple Measurements

There are uncertainties in the thermocouple measurements for the steady-state burning case because of the response delay of the thermocouple in the rapidly changing temperature field and the thermocouple lead loss. When the propellant is burning, there exists a steep temperature gradient near the burning surface. Therefore, as the thermocouple bead approaches the burning surface a large temperature difference across the thermocouple bead may exist in the propellant and a temperature gradient may also exist in the bead. The temperature gradient at the surface is found to be about

Table 2 Average experimental results[a]

Plateau temp, °C	Plateau length, 10^{-2} sec	N, 10^{-2} sec	Thermocouple size, mil	Burning rate, cm/sec	No. of experiments	Pressure, psia
237 ± 26.6	0.3 ± 0.05[b]	24 ± 3.16	½	0.124 ± 0.0049	8	
161.6 ± 14.8	0.8 ± 0.22	19 ± 1.73	1	0.115 ± 0.006	11	5
107 ± 12.1	1.9 ± 0.5	16.5 ± 3.16	2	0.108 ± 0.0057	5	
101 ± 9.1	1.8 ± 0.7	14 ± 2.24	3	0.124 ± 0.0064	7	
245 ± 22	0.36 ± 0.115	14.7 ± 2.83	½	0.144 ± 0.0046	7	
156 ± 18	0.5 ± 0.337	10.7 ± 1.68	1	0.147 ± 0.0067	9	10
103 ± 14.4	2.0 ± 0.83	11 ± 2.5	2	0.153 ± 0.0098	9	
103 ± 17.5	1.7 ± 0.89	11 ± 3.16	3	0.162 ± 0.0079	8	
248 ± 10	0.2 ± 0	17 ± 7.1	½	0.135 ± 0.001	3	
140 ± 15.1	1.1 ± 0.316	12 ± 3.08	1	0.15 ± 0.011	3	15

[a] T_o = 25°C; atmosphere: argon.
[b] Standard deviation.

$3.5°C/\mu$. For the $\frac{1}{2}$-mil thermocouple, the bead diameter is about 37.5μ and the temperature difference across the bead is about 130°C. However, the temperature gradient in the thermocouple bead may be relatively insignificant, since the thermal diffusivity of the thermocouple wire is about 300 times greater than that of the propellant, i.e., 0.25 cm²/sec vs 0.9×10^{-3} cm²/sec.

Thermocouple Analysis

Assume a thermocouple is imbedded in a deflagrating propellant with its bead at the center. The lead wires form a V shape and are straight. The coordinate axis is chosen such that the x axis is along one of the lead wires (Fig. 3). It will be assumed that the temperature distribution in the propellant is one-dimensional, being only a function of y, along the axis of the propellant. The burning surface is always perpendicular to the y axis and is planar. It is assumed that there is no radiant heat transfer. Thus, the governing equation for the temperature distribution in the lead wires is

$$(\partial^2 T_w/\partial x^2) + C_1(T_p - T_w) = C_2(\partial T_w/\partial t) \qquad (1)$$

where

$$C_1 = h\, P_w/k_w A_w,\ C_2 = \rho_w\, C_{pw}/k_w \qquad (2)$$

The boundary conditions are

$$\text{at } x = 0\text{: } T_w = T_B,\ \text{at } x = \infty\text{: } T_w = T_o \qquad (3)$$

The initial conditions are

$$\text{at } t = 0\text{: } T_w = T_o \qquad (4)$$

Since the thermocouple has a relatively high thermal diffusivity compared with the propellant, it is reasonable to assume that the temperature is uniform in the thermocouple bead. The temperature distribution in the propellant is exponential. Hence, heat transfer will take place between the bead and the propellant as well as between the propellant and the lead wires (Fig. 3). The governing equation becomes

$$\rho_w V_B C_{pw}\left(\frac{\partial T_B}{\partial t}\right) = \int_{A_{y_1}}^{A_{y_2}} h(T_p - T_b)dA_B - Q_{\text{lead}} \qquad (5)$$

If we assume the bead to be spherical, Eq. (5) becomes

$$C_3\left(\frac{\partial T_B}{\partial t}\right) = \int_{y_1}^{y_2} C_4(T_p - T_B)dy + 2C_5\left(\frac{\partial T_w}{\partial x}\right)_{x=0} \qquad (6)$$

where

$$C_3 = \rho_w V_B C_{pw},\ C_4 = \pi D h,\ C_5 = k_w A_w$$

The initial condition is

$$\text{at } t = 0,\ T_B = T_o \text{ if } y_1 \to \infty \qquad (7)$$

Here the density of the bead, ρ_w, and the specific heat of the bead, C_{pw}, are assumed to be the same as that of the thermocouple lead wires since Pt—Pt 10% Rh thermocouples were used. Also the physical properties ρ_w, C_{pw}, k_w are assumed to be equal for both of the lead wires.

The heat-transfer coefficient between the propellant and the thermocouple was given by Nydick[15] as

$$h = 2k/3R \ln(2L/R) + Bk(1 + e^{-BL\cos\alpha})/2 \qquad (8)$$

where R is the radius of the thermocouple wire, L is the length of the thermocouple wire imbedded in the propellant, α is the angle of the lead wire makes with the axis of the propellant, and B is $\rho C_p r/k$ given in Eq. (8).

The solution to Eqs. (1) and (6) is obtained numerically. The details of the numerical technique including the stability and the convergence of the numerical solution can be found in a separate article by Suh and Tsai.[14] The paper also discusses the effect of the various parameters.

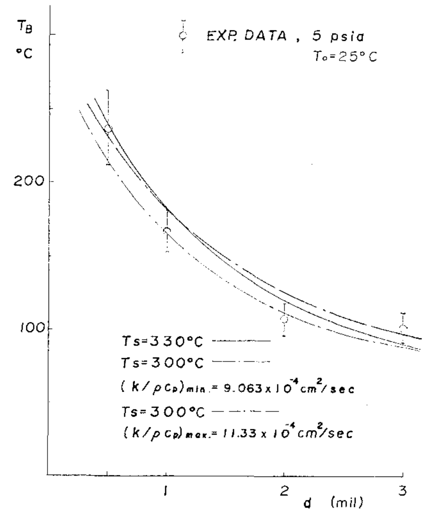

Fig. 9 Comparison of the experimentally and theoretically determined apparent surface temperature (bead temperatures) at 10 psia.

Discussion of Theoretical and Experimental Results

The experimentally determined apparent surface temperatures are compared with the theoretically predicted apparent surface temperatures, i.e., the bead temperatures, for 5 and 10 psia in Figs. 8 and 9. Since the physical properties of the propellant vary between the samples, the theoretically predicted results are given corresponding to the maximum and minimum values of the thermal diffusivity of the propellant. In all cases, the theoretical predictions lie within the spread of the experimental results. The surface temperatures assumed for the computations were 300 and 330°C. It was concluded that these values of surface temperature yielded optimum results. At 10 psia, these values are only slightly different from the values at 5 psia, due to the different burning rates.

Since the heat-transfer coefficients in the gas phase were chosen arbitrarily within a reasonable range, several values were used. An order-of-magnitude variation in the heat-transfer coefficient does not affect the experimental results significantly when the thermocouple bead size is small because of the relatively high heat-transfer coefficient in the solid compared with that in the gas. The effect, however, becomes quite significant for larger bead sizes, although a further decrease in the coefficient below the values indicated does not have much effect. Laminar flow around the thermocouple bead was assumed for the heat-transfer coefficient in the gas.

An important result of the theoretical analysis is that there exists an optimum ratio of the diameter of the bead of the thermocouple to the diameter of the lead wires. The optimum ratio, which is three, maximizes the response of the thermocouple. The main reason for this value of the optimum ratio is that the temperature change in the bead is proportional to the cube of the diameter of the bead, whereas the heat transfer to the bead is proportional to the square of the diameter of the bead. Of course, heat loss is also affected by the lead-wire diameter. Even with thermocouples with the optimum ratio, smaller diameter wires provide increased accuracy.

The theoretically predicted temperature profiles of the thermocouple bead corresponding to the maximum value of thermal diffusivity at $T_s = 300°C$ are plotted on the same figures as those for the experimental results, i.e., Figs. 10

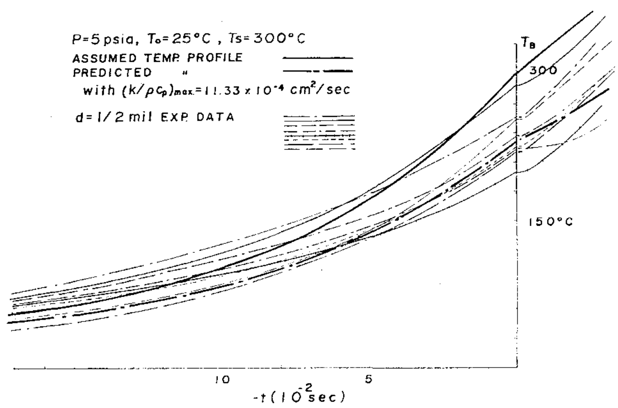

Fig. 10 Experimental and theoretical temperature profiles for ½-mil thermocouples at 5 psia.

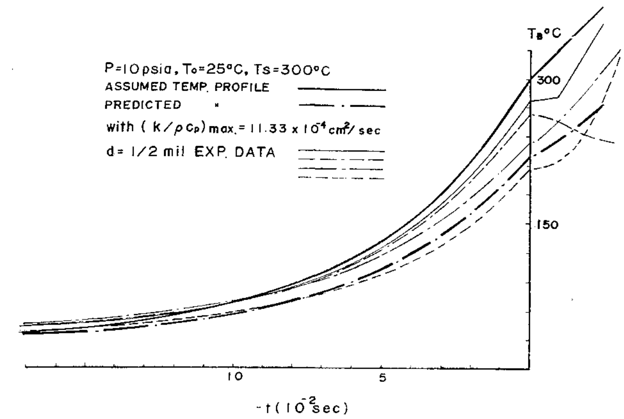

Fig. 11 Experimental and theoretical temperature profiles for ½-mil thermocouples at 10 psia.

and 11. Although the theoretical curves lie within the experimentally measured curves, it should be noted that in general the theoretically predicted bead temperatures are lower than the experimental results away from the burning surface. The slope of the experimental curves near the burning surface is close to that of the theoretically predicted curves. The most obvious reason for the deviation between the experimenta' and the theoretical results may be that the assumed theoretical, ideal temperature distribution in the propellant may be incorrect because of reactions in the solid, the foam zone, and the variations in physical properties as functions of temperature. If there indeed exists an isothermal foam zone, the plateau in the theoretically obtained temperature profile will be more significant, approaching the experimental results. Other contributing factors are clearly deviations of the physical dimensions and configurations of the actual thermocouples used from those assumed in the analysis. It should also be noted that both the experimental and the theoretical results do not show as flat a plateau when the thermocouple wire size is large as when it is small.

As shown in Figs. 10 and 11, the variation in the experimental results is so large that a precise prediction of the surface temperature is not possible. However, a surface temperature of 300°C gives the best correlation. The variation in the experimental results is inherent with the experimental system and therefore, unless a much more sophisticated control of all the experimental variables, such as the flow rate and chemical compositions, is employed, the precise determination of the surface temperature is difficult.

Discussion of the Results

The autoignition temperature, which depends only on chemical and physical properties, is 145°C for M-2 propellant. This temperature is the minimum surface temperature M-2 propellant can attain under any circumstance during steady-state burning. The measurements of heat of reaction of M-2 propellant reported by Kirby and Suh[10] with a differential scanning calorimeter indicate that near the autoignition temperature noticeable exothermic reactions begin to occur.

The ignition temperature under the experimental conditions employed is 214°C. As discussed in an earlier section, this temperature depends strongly on the environmental conditions and therefore is not very meaningful.

The steady-state surface temperature is about 300°C, according to the predictions of the theoretical model. The plot of this temperature, which is shown in Fig. 1, is somewhat lower than that of Heller and Gordon.[3] Since the thermocouple was spring-loaded against the propellant surface in their experiments, the thermocouples might have been exposed, at least partially, to the hot gas phase for prolonged periods of time, yielding high values. The present surface temperature is in the same range as Sabadell's[5] data, which are fairly independent of pressure. Again, it should be noted that these data were not corrected for the thermocouple response characteristics. In view of the fact that the thermocouple response has significant effects, especially when the burning rate is high, a direct comparison between the present results and uncorrected data of others is difficult. Our raw data obtained with ½-mil thermocouples agree well with the raw data of Aleksandrov et al.,[9] in spite of the fact that the location of the surface was determined relatively arbitrarily in their work. The surface temperature reported by Klein et al.[2] is low, probably because the temperature measured by them is not the surface temperature if there are reactions in the solid.

In view of these results, it may be thought that during the initial stage of the ignition process a pure thermal heating of the propellant by an externally supplied heat takes place until the autoignition temperature is reached. Then, the exothermic heat generated by the propellant and the externally supplied thermal energy bring the propellant to the ignition stage, finally leading to steady-state deflagration. Of course, the contribution of the exothermic reaction toward ignition is usually small due to the small magnitude of the exothermic heat released at the onset of the reaction. The heat conduction away from the surface, when the gradient is steep, also makes the contribution of the exothermic reaction small, since only a small volume of the propellant near the deflagrating surface is at temperatures higher than the autoignition temperature.

The plot of the initial temperature vs the burning rate, Fig. 6, shows a common point at 15.5°C at which the pressure appears to have no effect on the burning rate. The work by Suh and Clary[12] indicates that if the pressure is maintained at 5 to 165 psia and if the external supply of heat is completely cut off after ignition, M-2 propellant is self-extinguished when the initial temperature of the propellant is below 15.5°C. Aleksandrov et al.[9] found the temperature profile obtained at an initial propellant temperature below 0°C was quite different from those at higher initial temperatures. The anomaly observed by Aleksandrov et al.[9] may be related to the self-extinction observed by Suh and Clary. It should be also noted that Zenin[8] observed plateau in his surface temperature profile.

Conclusion

The minimum initial temperature of the propellant at which self-deflagration can occur, i.e., autoignition temperature, is determined to be 145°C. The ignition temperature under the conditions employed in this paper is 214°C and the surface temperature is found to be 300°C–315°C at 5 and 10 psia.

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