Hot-Particle Ignition of Double-Base Propellants

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The effect of hot oxide particles from an igniter impinging on the surface of a double-base propellant has been simulated by means of heated wires having thermocouples attached. From observations of the rate of cooling and whether this led to ignition, energy relationships are established for a range of initial pressures and temperatures which confirm an ignition theory and give insight into the processes taking place. From measurements of the rate of heat transfer from a solid body to the propellant and a size analysis of the particulates produced by actual igniters, predictions have been made regarding the contribution of the oxide particles of various diameters to the total ignition process. The results suggest that, as a result of contributions from the particulate matter, there may be considerable variation in the ignition transient with varying ambient conditions.

Nomenclature

C	= specific heat
K	= thermal conductivity
q''	= heat per unit area
$\dot{q}''$	= heat flux per unit area
Q	= latent heat
t	= time
T	= temperature
u	= burning velocity
x	= distance normal to surface
α	= thermal diffusivity
ρ	= density

Subscripts

c	= condensed phase
ig	= ignition
s	= solid phase

I. Introduction

THE major research effort into the ignition of solid-propellant rockets in the recent past has concentrated on developing an understanding of the process that leads to ignition within the propellant itself. To this end various models for ignition have been proposed and compared with the experimental results obtained for ignition under relatively uncomplicated circumstances. Typically, ignition sources such as pure radiation, e.g., arc-image furnace or laser, and pure convection from a hot gas stream are considered. A comprehensive review of current ignition theories and concepts has been made by Price et al.[1]

While these works have led to an improvement in the understanding of the solid-propellant igniter process both for composite and double-base propellants, little information has emerged that is of direct help to the designer in the way of specifying an ignition for a particular duty in a rocket engine. For example, there is a gap between our knowledge of what total energy and flux are required for ignition of a propellant and what energy a given igniter composition and configuration will supply, and indeed whether the one will match the other. In making this assertion it must be recognized that the principal lack of information lies in the latter area.

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Indeed, measurements of the heat flux to the propellant surface under conditions representative of actual igniter operation have only recently been carried out by Allan, Bastress, and Smith[2] and Scagnetti and Crabol.[3] These workers have measured both convective and radiative heat fluxes in engines equipped with actual igniters, but a dummy propellant, and they have arrived at numbers relating to the heat flux which are, for the first time, of real value to the igniter designer.

However, a large contribution to the ingition of the propellant surface probably comes from the impact of hot-metal oxide particles, since the major proportion of the energy developed within the igniter is carried by particles either in the liquid or solid phase. Whereas Allan et al. make mention of the heat transfer to the propellant surface by virtue of particle impingement and condensation, and Scagnetti and Crabol accept that the major portion of the radiative heat flux originates from the cloud of solid oxide particles, neither investigator has considered or determined that proportion of the ignition stimulus which arises from actual particle impact. A more recent publication by Bastress and Niessen[4] on an experimental technique for simulating the presence of hot particles indicates they have recognized the problem, although no results of ignition tests in the presence of particulate matter have, as yet, been reported.

Heat conduction from an impacting particle is essentially transient, and hence it is unreasonable to consider the contribution to heat from this source if it is integrated over a time scale significantly longer than the cooling time of the particle. Thus, the heat transfer arising from the local impact of solid particles will supplement a more constant rate of heating provided by background radiation and convection.

The present research program was carried out in an endeavor to determine the extent to which hot particulate matter could contribute to the ignition process, and to simplify both the experimental and theoretical approach, only heat transfer from particles was considered.

If, as is the case in practice, more nearly steady-state contributions to the total heat transfer exist, then it should be a simple matter to sum these with the transient flux from the particular matter.

II. Theoretical Approach—Minimum Ignition Energy

The present studies were confined to the physically simpler (although chemically more complex) double-base propellants, and the theoretical treatment relies largely on the classical model proposed by Hicks.[5] The heat balance within the solid phase takes the form:

$$\rho c(\mathrm{d}T/\mathrm{d}t) = k(\mathrm{d}^2T/\mathrm{d}x^2) + \rho uc(\mathrm{d}T/\mathrm{d}x) \tag{1}$$

$$\dot{q}'' = -k(\mathrm{d}T/\mathrm{d}x) - \rho u Q \quad \text{at} \quad x=0 \tag{2}$$

where $\dot{q}''$ is the incident heat flux of an unspecified source. Replacing this by a term representing chemical heating, such as an Arrehnius reaction rate equation, an overall relationship for steady-state burning may be obtained. In the absence of established burning, $\dot{q}''$ may represent the transfer of energy during the ignition process, when the burning rate terms would be omitted.

During steady burning, the chemical reaction at the surface, itself initiated by heat transfer from the gaseous flame zone, results in a molten layer followed by a temperature gradient within the solid phase, the steepness of which will depend on the burning rate. During ignition the stimulus must provide at least sufficient energy to melt this thin layer of propellant and raise it to a temperature at which it will decompose. If the ignition energy is supplied at a sufficient rate, the temperature gradient in the solid phase will also be as steep as that which exists during steady-state burning, as postulated by Pantoflicek and Lebr,[6] and there will be a minimal diffusion of heat into the propellant. This is referred to as *minimal ignition*. If, on the other hand, the stimulus is applied over an extended period of time, significant penetration of heat within the solid phase will occur before the surface is raised to its self-decomposition temperature and the total amount of energy required for ignition will be greater. This will be referred to as *extended ignition*. Hence for minimal ignition we can write

$$(\mathrm{d}T/\mathrm{d}x)_s = (\rho c/k)(\mathrm{d}x/\mathrm{d}t)_s \Delta T_s \tag{3}$$

Now the rate of heat transfer from an impacting solid particle to a propellant surface is a complex process. As the propellant surface temperature rises and the particle surface temperature falls, heat flux will tend to decrease. The establishment of a temperature gradient within the particle itself may contribute to this decrease and prolong the period over which heat is transferred. Finally, the three-dimensional nature of the problem may introduce an additional nonlinearity in the temperature gradients within the solid phase.

For analytical purposes, the problem can be eased somewhat by considering either small high-temperature particles, such as are produced in a rocket engine igniter, or larger lower-temperature particles having a high thermal diffusivity compared with that of the propellant. With these restrictions the heat flux during the ignition phase may reasonably be considered to be constant. Furthermore, if the ignition time is short, the three-dimensional aspect of the problem may also be unimportant.

Apply Eqs. (1) and (2) for the nonregressive surface, the solution for the boundary conditions $\dot{q}=\dot{q}_s$ at $x=0$ and $t>0$ is

$$\dot{q}'' = \dot{q}_s'' \operatorname{erfc} x/2(\alpha t)^{1/2} \tag{4}$$

Thus

$$T = \frac{2\dot{q}_s''(\alpha t)^{1/2}}{k} \operatorname{ierfc} x/2(\alpha t)^{1/2} + T_0 \tag{5}$$

If the temperature at $x=0$ is T_s

$$\Delta T_s = T_s - T_0 = 2\dot{q}_s''(t/\pi k \rho c)^{1/2} \tag{6}$$

If the heat flux is constant, i.e., $q'' = \dot{q}''t$

$$\dot{q}'' q_s'' = (\dot{q}_s''/t)^2 = (\pi/4) k\rho c (\Delta T_S)^2 \tag{7}$$

but $\dot{q}_s'' = k\ (\mathrm{d}t/\mathrm{d}x) = \rho c\ (\mathrm{d}x/\mathrm{d}t)\ \Delta T$ (for a regressive surface) and $\mathrm{d}x/\mathrm{d}t = u$.

Assuming the heat flux for ignition corresponds to the heat flux transferred during steady-state burning, the ignition time may be found as

$$t = (\pi/4)\ k/\rho c u^2 \tag{8}$$

This will be referred to as $t_{\min}$, the minimum time for a stimulus to lead to ignition.

Since the energy absorbed in the solid phase will be

$$q'' = c\rho u \Delta T_s t \tag{9}$$

the total amount accumulated in this phase during minimum ignition will be given by

$$q_s'' = (\pi/4)\ (k/u)\Delta T_s \tag{10}$$

In addition, a quantity of heat will be stored in the condensed phase given by

$$q_c'' = \rho c x_c (T_s - T_0 + Q/c) \tag{11}$$

and thus the total minimum ignition energy will be

$$q_{ig}'' = q_s'' + q_c'' \tag{12}$$

Under conditions of minimal ignition, ignition will be accomplished when a certain quantity of heat, q_{ig}'' has been supplied to the propellant. If, on the other hand, the heat is supplied at a significantly lower rate than this, the heat soak into the solid phase will become appreciable and may even become dominant so far as the total heat input is concerned. Under such conditions of extended ignition, the contribution of heat to the condensed phase may be ignored and ignition will occur when a certain surface temperature, referred to as T_{ig} is attained.

The relationship between heat input and heat flux will then be represented by

$$\dot{q}'' = (\pi/4)\rho k c\ (T_{ig} - T_0)^2/q'' \tag{13}$$

and the slope of log q'' and log $\dot{q}''$ will be -1, instead of zero in the case of minimal ignition.

III. Experimental Investigation

To study directly the problem of propellant ignition by bringing particles of a representative size and temperature in sudden contact with the propellant surface was clearly impracticable. However, by substituting for the three-dimensional particle a two-dimensional wire, which could be electrically heated and have its temperature automatically recorded, a reasonably representative situation was created.

A diagrammatic form of the scheme that was adopted is shown in Fig. 1. With the solenoid energized as shown and the armature retracted, the wire is heated by means of a direct current. During heating no meaningful output was obtained from the thermocouple for the voltage drop along the wire far exceeds that of the thermal emf.

By de-energizing the solenoid, the heated wire moved forward to impact on the propellant surface and with the heating current now being interrupted a temperature-time history of the wire was obtained by recording the thermocouple output. In the event, one of three things happened. a) If the wire had ample energy a cooling curve followed by a rapid rise in temperature was recorded. The rate of temperature rise varied erratically as the now burning propellant receded from the surface of the wire. b) If just sufficient energy was transferred, the cooling curve was followed by a delay period before the temperature rise. c) If the energy was insufficient to cause ignition an interrupted cooling curve was obtained as the decomposing propellant fed energy back to the wire.

Table 1 Experimental data for 1 atm, 20°C

Wire diam. (cm)	Ignition delay (m-sec)	$\dot{q}''$ (cal/cm^{2}sec)	q'' (cal/cm^2)	t_{min} (m-sec)	Extrapolated ignition energy	ψ	Ignition energy (flat plate) (cal/cm^2)
0.013	85-180	20-49	2.7-4.9	93	2.6	0.41	1.1
0.038	160-820	5.4-11	1.0-6.7	93	2.1	0.63	1.3
0.061	100-900	5.9-10	1.7-8.1	93	1.7	0.73	1.3
0.094	160-500	3.3-9.5	1.1-3.3	93	1.6	0.79	1.3
0.120	210-480	3.8-11	1.7-2.6	93	1.6	0.86	1.4
0.16	320-890	2.3-2.6	2.1-2.6	93	1.6	0.90	1.4

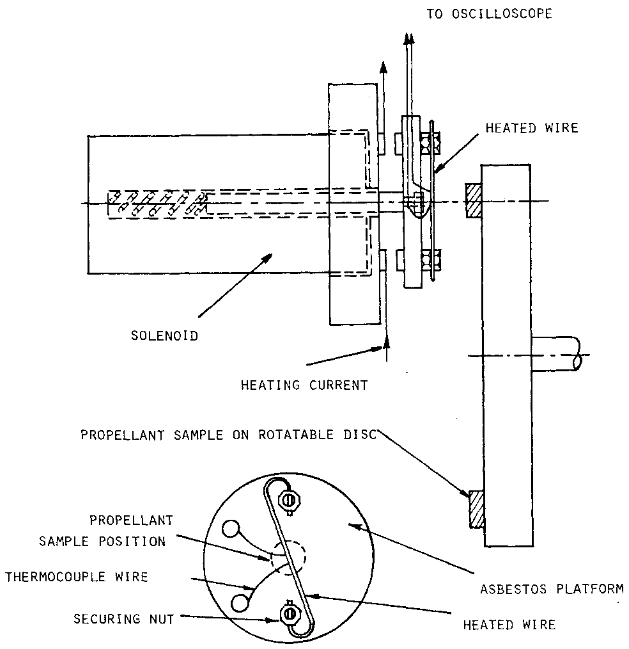

Fig. 1 Schematic arrangement.

Nickel/chromium alloy wires having diameters ranging from 0.163-0.013 cm were employed in the experimental program to which were attached Pt-Pt/Rh thermocouples. The thermocouples were never larger than 20% of the heated wire diameter in order to minimize the temperature drop at the point of attachment. However, the temperature depression was calculated for each wire/thermocouple combination and an appropriate correction applied.

Six propellant samples could be accommodated on the rotating disk and positioned to turn in the test position. The whole assembly was enclosed in a temperature-controlled pressure vessel equipped with an observation window. Tests were carried out in a nitrogen atmosphere at pressure ranging from 1-18 atm with initial propellant temperatures for 40°, 20°, 70°, and 100°C. The propellant employed was a standard platonized double-base composition, and the samples were taken from a single production batch.

IV. Analysis of Experiments

From the oscillograph records leading to ignition, the cooling curve was found to be substantially linear, so the heat flux transmitted to the propellant could be considered constant. To arrive at the actual heat transfered to the propellant the wire was assumed to be half embedded in the propellant sample. Although this might be regarded as perhaps the most questionable assumption of the experiments, inspection of "no ignition" results showed a burn mark across the propellant face having a width that, in most cases, was equal to the wire diameter. A correction for heat loss by natural convection and radiation from the half of the wire not in contact with the propellant was made by recording the natural

Table 2 Heat accumulated in the solid and condensed phases

Pressure (Atm)	T_0 (°C)	Ignition energy (cal/cm^2)	Phase energy (cal/cm^2)	Condensed phase energy (cal/cm^2)	Condensed phase thickness (cm × 10^3)
1	20	1.6	0.59	1.0	3.4
	70	1.1	0.29	0.81	3.2
	100	0.80	0.17	0.63	2.6
2	20	1.3	0.37	0.91	3.1
	70	0.93	0.18	0.75	2.8
	100	0.71	0.11	0.60	2.4
18	20	0.73	0.08	0.64	2.2
	70	0.55	0.04	0.51	1.9
	100	0.45	0.03	0.43	1.7
68	20	0.63	0.06	0.57	2.0

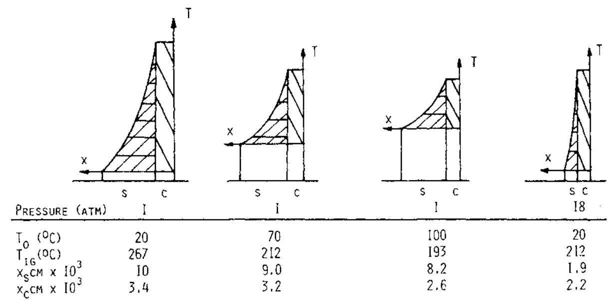

Fig. 2 Temperature in solid and condensed phases.

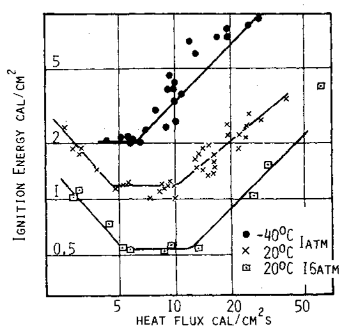

Fig. 3 Ignition energy and heat flux.

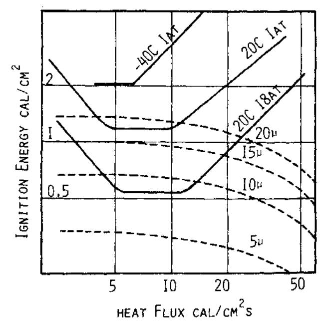

Fig. 4 Particle heating rates.

rate of cooling of the wire in the ambient atmosphere and deducting one-half of this amount.

A further correction ψ has to be applied on account of the fact that the temperature gradient within the propellant follows cylindrical coordinates as outlined in Ref. 6. For the larger diameter wires and for short heating times where the depth of penetration by the ignition stimulus is not great, this correction factor is close to unity. For smaller wires it is less than unity, and to correlate the results it is necessary to apply this correction in order to relate the heat flow to one-dimensional rather than two-dimensional coordinates.

Table 1 lists the data obtained for the seven wires with the propellant at atmospheric pressure and 20°C, and is typical of the results that were obtained at this and higher pressures. It will be noted that the time to ignition varies several-fold as does the rate of heating and the total heat input. Thus, many of the tests represented an extended rather than a minimal ignition.

Since the total ignition energy comprises the sum of the energies stored in the solid and condensed phases [Eq. (12)] and magnitude of the former for minimal ignition can be calculated from the burning rate by means of Eq. (10), the quantity of heat retained in the condensed phase can be arrived at by difference. Given an assumption of the propellant heat capacity and its latent heat of fusion, the thickness of the condensed phase can be calculated. Assuming these to be 0.357 cal/g°C and 100 cal/g, respectively, the values shown in Table 2 were calculated. It must be appreciated that although Q is a very approximate estimate, it is likely to remain constant and thus serves at least qualitatively to illustrate the trends. It will be observed that the reduction in igintion energy due to a pressure increase results from a reduction in the heat stored in the solid rather than the condensed phase, whereas for the case of a reduction in energy due to an increase in initial temperature the converse applies. This point is illustrated more clearly in Fig. 2, which depicts the calculated temperature profiles within the propellant that lead to ignition. The principal results obtained for the ignition energy and heating rate are shown in Fig. 3.

The ignition energy/heating rate curves does indeed approximate to a slope of -1 at low heating rates (corresponding to an extended ignition), and this is followed by a horizontal limb corresponding to near-minimal or minimal ignition conditions. As the heating rate is increased above that necessary to achieve minimal ignition, the curve assumes a slope of approximately $+1$, implying a constant ignition time, regardless of heat flux.

It has been postulated that, at these high heat fluxes, the physical processes (evaporating and boiling) effectively "outstrip" the chemical processes, i.e., reaction and combustion, and the source of heat feedback is thereby removed from the propellant surface. Certainly, in the experiments carried out at high heating rates a clearly visible "puff" of propellant vapour was produced on impact of the hot wire prior to ignition taking place. Furthermore, there is a reduction in the ignition energy both with increasing initial propellant temperature and pressure, as the theory predicts.

However, the absolute level of ignition energy is lower by a factor of about 2 than that found by other workers (e.g., Price et al.[7]) who have investigated this class of propellants. While the differing properties of the propellants might account for some of this discrepancy, it is unlikely to account for all of it.

An explanation may lie in the fact that the other investigators have employed either pure radiation or pure convection from heated inert gases to produce the ignition pulse. In both cases, the propellant vapor initially produced is isolated from further heating, for in the case of radiant heating the vapors are virtually transparent, and with convective heating the vapors are swept away and diluted by the inert gas stream. In the present tests the propellant vapor will remain in contact with the source of heat for a significant period of time and receive a further input of heat from the rear surface of the hot wire.

Table 3 Analysis of igniter particulates

Particle size range (μ)	Percentage of particles B-KNO$_X$	Mg-KNO$_3$
0- 4.9	84	52
5- 9.9	12½	33½
10-14.9	1½	7½
15-19.9	1	5
20-24.9	½	1½
25-29.9	1/10	1/3
30-34.9	...	1/3

V. Hot Particle Ignition

As well as establishing relationships between the ignition energy and heating rate of the solid-propellant surface, the cooling curves that were obtained enabled a heat-transfer coefficient to be established between the wire and the propellant surface during the period prior to ignition. With the finest wires employed (0.013-cm diam) an initial temperature of 1500°C was necessary to provide ignition, which was far in excess of the melting and boiling points of the propellant. Thus, the conditions at the interface from the point of view of vaporization and boiling were considered to be similar to those that might result from the impact of a particle perhaps double this temperature.

From the cooling curves obtained prior to ignition a heat-transfer coefficient was estimated which was found to vary from 0.043 to 0.061 cal/cm^2 -sec°C during the experiments, averaging at 0.05 cal/cm^2 -sec°C. Noteworthy was the fact that, even for the larger wires, when ignition took place from an initial temperature as low as 400°C, a similar value for the heat-transfer coefficient was found, suggesting that the heat-transfer mechanism was essentially the same as that for the higher wire temperatures. Such a coefficient might, therefore, legitimately be applied for the case of hot particles impacting on the propellant surface, which would enable the heating rate and total heat input from such particles to be calculated.

Now the heat flux per unit area is given by

$$\dot{q}'' = h(T_p - T_s) \tag{14}$$

and the total heat input per unit area for a semi-embedded spherical particle is

$$q'' = (d\rho C_p/3)\, T_{p0} - T_t \tag{15}$$

Assuming that the surface temperature remains constant relative to the particle temperature, and this will be so if the propellant is boiling, cooling curves can be calculated for particles according to their size. The question then arises as to the range of particulate sizes appropriate to real igniters. To determine this, a series of igniter tests was carried out in a dummy engine lined with cellulose acetate. The properties of the real propellant so far as capture of the hot particles was concerned, and by removal and inspection of the liner after a test the particulate size distribution could be determined.

The results shown in Table 3 indicate that, for a boron-potassium nitrate composition, by far the greatest mass of particulate matter lies in particles smaller than 5 μ. However, an appreciable fraction does occur in the 5-10 μ range, and even some up to 20 μ. For magnesium-potassium nitrate compositions there is a rather greater concentration in sizes about 5 μ.

Cooling curves for boron oxide from 2500K have been superimposed on the ignition energy curves in Fig. 4 for particle diameters ranging from 5 to 20 μ, thus enabling a comparison to be made between the energy required for ignition and that available from a particle of given size. If one makes the assumption that the whole of the energy delivered by the particle makes a useful contribution to the ignition process, i.e., that none is carried away by the propellant vapor, then it is clear that particles of 20 μ and above will alone lead to the ignition of the propellant at 20°C and atmospheric pressure, and that particles between 10 and 15 μ will achieve this if the pressure is at a realistic figure of about 7 atm.

Smaller particles, e.g., in the 5μ size range will not, of themselves, give rise to ignition. However, this is not to say they will not make a significant contribution to the process, for as Fig. 4 shows, they may contribute about one-third of the total energy requirement.

VI. Conclusions

The conclusion one may draw from this is that, if the combination of the igniter and the ambient state of the engine is such that an ample amount of ignition energy is available, then ignition will probably take place uniformly over the propellant surface from the quasi steady-state heat flux due to radiation and convection.

However, if these sources are marginal, for example for the same rocket ignited at a low ambient temperature, then ignition will occur at centers of particle impact, and then spread between these centers. In these circumstances the pressure-time history of the ignition process may be markedly different.

As a corollary, it might even prove profitable deliberately to introduce inert particles into the igniter composition in order to provide a consistent nongaseous source of heat, thus avoiding the over-pressure conditions that arise when ignition takes place rapidly as a result of the gaseous contribution from the igniter.

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