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Burning Behavior of Composite Propellants with Fast-Burning Inclusions

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Introduction

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ARIOUS applications of propellants make it necessary to modify the propellants to increase the burning rate. For the most part, this problem can be solved by the use of various combustion catalysts. ¹⁻⁵ However, there comes a point where all burning rate catalyst possibilities have been exhausted, but the desired burning rate level has yet to be reached. In this case, one may employ various additives⁶⁻⁸ that possess a burning rate of their own several times superior to that of the starting propellant composition. The combustion mechanism of such compositions has not been fully considered, though an increase in the burning rate was assumed to be due to a cratering effect in the burning propellant caused by extremely fast decomposition of particles of the additive.⁶ Thus far it is not clear, however, how the particle size, burning rate, and content of additive entered into a propellant composition can influence the burning rate and character of $r_b(p)$ dependence. In this connection, the effect on the burning rate of entering fast-burning energetic materials (FBEM) into ammonium perchlorate-polymeric binder propellant formulations has been studied.

Experimental

Lead salt of 2,4,6-trinitro-meta-cresol (LTNC), the combustion of which has been studied previously,9 was used as the fast-burning additive. It is a stable compound with ignition temperature of 250 K and demonstrates a burning rate 21.6 cm/s at 10 MPa, that is, almost

10 times faster than conventional composite propellants. Grains of LTNC were prepared by granulating tablets of the substance pressed at 500 MPa to the density of 2.34 g/cm³ followed by sieving.

A window constant-pressure bomb of 1.5-liter volume pressurized with nitrogen was used to measure the burning rate of compositions in the pressure range 0.1–40 MPa. The behavior and velocity of burning were registered using a slit camera. In preparing the strands, uncured propellant compositions containing LTNC grains were put into transparent acrylic tubes 7 or 12 mm i.d. and 30-50 mm height.

The delay time of ignition of the propellant layer immediately under an FBEM grain was determined using strands separated into two parts with a pressed FBEM tablet approximately 1 mm thick and 7 mm diameter. The slit camera was used to record the front of flame propagation through the propellant-FBEM tablet-propellant strand section that allowed the delay of ignition of the lower propellant layer to be derived from the record.

Results and Discussion

Incorporation of LTNC of fine particle size (less than 10 μ m) into propellant, which burnt at 2.2 cm/s at 10 MPa, failed to enhance the burning rate (Fig. 1). Quite different observations have been made when the FBEM particle size was changed. Figure 1 shows that an increase in the LTNC particle size to 500 μ m lead to a progressive enhancement of the burning rate of the propellant composition containing 15% LTNC grains. On further increasing the particle size, the burning rate of the composition remained practically unchanged.

Previous experiments on critical combustion diameter d_c showed that d_c of LTNC was equal to 15 μ m, and there was no change in the LTNC burning rate within the 100–1000 μ m diameter range. It follows, therefore, that the observed decrease in the composition burning rate as LTNC particle size decreases from 500 to 100 μ m (see Fig. 1) cannot be attributed to the influence of the critical combustion diameter.

To explain the results observed, let us consider a simple combustion model of a propellant formulation containing FBEM grains. The FBEM grains are assumed to be of spherical form, to have diameter d, to have the burning rate of their own r_{FBEM} superior to that of the baseline propellant, and to be evenly distributed in the propellant bulk, as shown in Fig. 2. In the case of the one-dimensional model, the time of combustion of a unit volume is a sum of combustion times of the propellant layers above and under an FBEM particle and the particle itself. Also one may suggest the occurrence of some delay in ignition of the propellant layer underneath the FBEM particle due

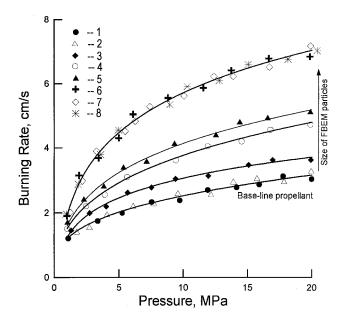


Fig. 1 Burning rate vs pressure for a composite propellant containing 15% LTNC grains at different LTNC particle sizes: 1, baseline propellent; 2, less than 10 θ m; 3, less than 100 θ m; 4, 200–315 θ m; 5, $315-400 \ \theta \text{ m}$; 6, 400-630 $\theta \text{ m}$; 7, 630-800 $\theta \text{ m}$; and 8, 800-1000 $\theta \text{ m}$.

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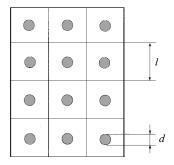
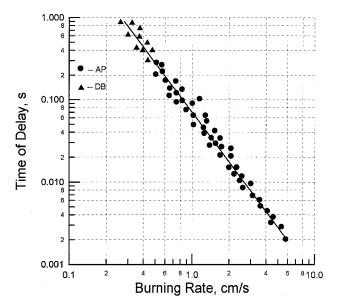


Fig. 2 Schematic of distribution of FBEM grains in a propellant composition, where l is unit volume size and d is FBEM grain size



 $Fig. \ 3 \quad Ignition delay time \ as \ a function of the burning \ rate \ of \ AP-based \ composite \ propellants \ and \ double-base \ propellants.$

to a big difference in burn rates of the FBEM and the propellant. Thus, the time of combustion of a unit volume can be defined by

$$\tau = l/W = (l - d)/r_p + d/r_{\text{FBEM}} + \tau_d \tag{1}$$

where l is the unit volume size, d is the diameter of the FBEM particle, r_p is the burning rate of baseline propellant, τ_d is the time of delay in ignition of the propellant layer underneath the FBEM particle, and W is the overall burning rate of the composition. If the designation for volume fraction of FBEM grains in the propellant composition is $\alpha = \pi d^3/6l^3$ and the ratio of $r_p/r_{\rm FBEM}$ is represented by z, then W can be written as

$$W = r_p / \left[1 - (1 - z - \tau_d r_p / d) \cdot \sqrt[3]{(6/\pi)}\alpha\right]$$
 (2)

It follows from Eq. (2) that $W=r_p$ when the burning rate of FBEM approaches to that of the baseline propellant, that is, at $z \to 1$ and $\tau_d \to 0$.

Ignition delay time τ_d has been measured for various propellant compositions burning at 1.8–6.6 cm/s at 10 MPa. FBEMs of different chemical nature were introduced into the compositions; they had their own burning rates of 22–100 cm/s at 10 MPa. Experiments carried out in the pressure range of 1–20 MPa have shown that τ_d depends only on the burning rate of the starting propellant and is independent of its particular composition as well as the FBEM chemical nature (Fig. 3). Experimental data for both ammonium perchlorate-(AP-) based composite propellants and conventional double-base propellants fall on a straight line, which gives an expression for τ_d as

$$\tau_d = k / r_n^2 \tag{3}$$

where $k = 7 \times 10^{-2}$ cm²/s. The substitution of τ_d into Eq. (2) yields the final equation for the burning rate of a propellant formulation containing FBEM grains:

$$W = r_p / \left[1 - (1 - z - k/dr_p) \cdot \sqrt[3]{(6/\pi)} \alpha \right]$$
 (4)

Calculations of burning rates from Eq. (4) for a composition with $r_p = 2.2$ cm/s and comprising 15% LTNC of different particle size give results well below experimental data when $k = 7 \times 10^{-2}$ cm²/s is used (Fig. 4). [Note that in Figs. 4 and 5 curves were calculated from Eq. (4) for different values of proportionality constant k.] If τ_d is not taken into considerationat all, that is, assuming $k/dr_p = 0$, the calculated values of W prove to be higher than experimental ones and independent of the FBEM grain size. However, as seen from Figs. 2 and 4, in practice the particle size has a profound

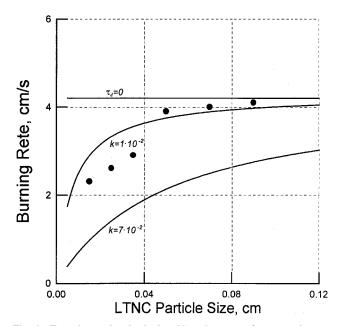


Fig. 4 Experimental and calculated burning rates of a composite propellant containing 15% LTNC grains at 4 MPa as a function of LTNC particle size.

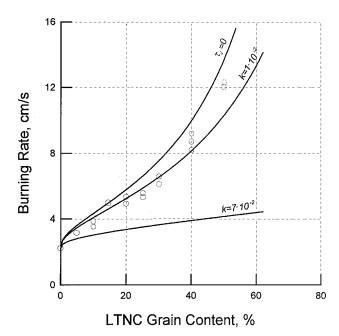


Fig. 5 Experimental and calculated burning rates of a composite propellant containing LTNC of $800-\theta$ m particle size at 10 MPa as a function of LTNC content.

impact on the burning rate in the region of small sizes. Experimental data may be adequately approximated by Eq. (4) if one assumes $k = 1 \times 10^{-2}$ cm²/s.

Proportionality constant k is the product of thermal diffusivity and some coefficient that depends on the shape and state of the propellant surface to be ignited. The value of 7×10^{-2} cm²/s has been obtained for a particular case of the parallel-sided FBEM pellets with the smooth contact surfaces. In the real case, after burning up an FBEM particle, a crater is left in the propellant bulk, which replicates the initial particle shape and has a rough surface. This facilitates the ignition of the adjacent propellant layer, resulting in a lower value of k than one obtained in the experiments with parallel-sided FBEM pellets.

Figure 5 demonstrates a fair agreement between experimental and calculated (for $k = 1 \times 10^{-2}$ cm²/s) burning rate data for combustion of a propellant composition with different contents of FBEM grains in the formulation.

Using Eq. (4) and assuming that the additive will influence the burning rate only if the time of combustion of FBEM particle plus ignition delay time is less than the time of burning of a propellant layer with thickness d, that is, when $d/r_{\rm FBEM} + \tau_d < d/r_p$, one may derive an expression for the criterion for FBEM performance in the propellant composition:

$$K_p = \frac{dr_p(1-z)}{1 \times 10^{-2}} > 1 \tag{5}$$

When the criterion $K_p \le 1$, an FBEM additive will not increase the burning rate of the baseline propellant, however large the FBEM burning rate and content in the composition. For example, an FBEM additive with 200–300 μ m particle size and 20 cm/s burning rate is unable to increase the burning rate of a propellant formulation that burns itself at 0.3–0.5 cm/s. The lower the burning rate is of the baseline propellant, the more difficult it is to accelerate by using FBEM inclusions. The effect of incorporation of 15% LTNC of 250- μ m particle size, calculated from Eq. (4), is about a 1.8 times burning-rate increase for a baseline propellant burning at 2.5 cm/s, whereas only a 10% increase is achievable if a baseline propellant burns at 0.5 cm/s.

Conclusions

Application of FBEMs as additives to AP-based composite propellants has been shown to increase the burning rate. A simple model for propellant formulations containing FBEM inclusions has been proposed that satisfactorily describes the experimental results. This model takes account the FBEM particle size, its content, and burning rate, as well as some delay in ignition of the propellant layer adjacent to the FBEM particles. Under the tested operating conditions, the ignition delay time has been experimentally measured to be independent of the type of propellant used, but is determined by a propellant burning-rate value at a particular pressure, thus providing important evidence in support of the model. The criterion for FBEM performance in the propellant has been derived to provide the possibility of estimating FBEM effect on the propellant burning rate.

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