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INERT PLUG FORMATION IN THE DDT OF GRANULAR ENERGETIC MATERIALS *

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A mechanism is proposed to explain the "plugs" that have been observed in deflagration-to-detonation transition (DDT) of granular explosives. Numerical simulations are performed that demonstrate the proposed mechanism. Observed trends are reproduced.

INTRODUCTION

Experiments on the deflagration-to-detonation transition (DDT) of granular explosives, confined in thick walled steel tubes, are referred to as "tube tests." In these tests, an upwards moving piston drives a compaction wave into the granular explosive. After an ignition delay, the explosive near the piston reacts vigorously which further pressurizes the bed leading to the formation of an inert plug at some distance from the piston. Full detonation initiates near the top of the plug, and the plug is slowly consumed by reaction. Figure 1 illustrates this process. Even with the detailed experimental evidence available (*e.g.* 1,2), the exact nature and role of the observed plug remains unclear. In fact, there are indications that plugs may not always form (2), with higher burning rate explosives being less likely to exhibit plugs than lower burning rate materials (3).

Previous DDT models have produced high density regions; however, these plugs are quickly consumed from below by a vigorous reactive wave below that rapidly evolves to full detonation (5,6). In this paper, a scenario for plug formation is demonstrated. The essence of the idea is that the plug forms when there is no longer adequate intergranular pore space to yield significant gas-phase reactions (*cf.* Chapt. 3 of (2)). In relatively slow compaction processes these gas-phase reactions are needed to provide conductive feedback of energy to the solid to sustain vigorous pyrolysis. In very rapid compaction processes, energy dissipated to particle "surfaces" (*i.e.*, friction, plastic

work, *etc.*) is sufficient to yield vigorous pyrolysis of the solid without significant conductive feedback from the gas phase. This latter process initiates rapid reaction at the top of the plug, leading to full detonation (see Fig. 1).

FIGURE 1. Distance-time calculation of solid volume fraction, ϕ_s , of a DDT event in a 1D tube test. The region denoted by $R_k < R_{k0}$ is where gas-phase ignition has been inhibited.

MODEL

Relatively simple modifications are made to the basic framework of previous models instead of prescribing a new model. The basic governing equations adopted are from (7) and are listed in (8) in these proceedings (see Eqs. 1-6). A Jones-Wilkins-Lee (JWL) equation of state (EOS) is assumed for the gas and a Hayes EOS is assumed for the solid (see Eqs. 8-11 of (8)). The source term adopted for the compaction equation is $\mathcal{F} = (\phi_s \phi_g / \mu_c)(\max(P_s, \beta) - \max(P_g, \beta))$

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where μ_c is the compaction viscosity, P is pressure, and ϕ is volume fraction (7). The subscripts "s" and "g" refer to solid and gas, respectively. The configuration pressure, β , is modeled using $\min[-\max((\phi_s - \phi_{s0}), 0)\tau \ln(\phi_g)/\phi_g, \beta_{max}]$ where τ is a stress coefficient (7). The volumetric heat transfer coefficient, $\mathcal{H}$, is assumed to have the form, $\mathcal{H} = (3k_H\phi_s)/(a_p^2)$ where a_p is an effective particle radius, $a_p = a_{p0}\min(1, (\phi_s/\phi_{s0})^{1/3})$. Vigorous reaction significantly affects interphase heat transfer. To crudely account for this, $k_H \equiv H(I - 0.5)k_1 + (1 - H(I - 0.5))k_2$ where I is the induction time variable ($0 \leq I \leq 1$), H is the Heaviside step function, and k_1 & k_2 are constants. The source term assumed for the energy equation is $\mathcal{E} = -P_g\mathcal{F} - \mathcal{H}(T_{int} - T_g)$. The first term corresponds to depositing all compaction work with the solid (7), and the second is Newton's law for heating the solid where T_{int} is the interface temperature.

Heating of the Solid

We need an estimate for T_{int} to be used in the heat transfer potential and as an ignition criterion. The solid energy equation can be written as,

$$\frac{De_s}{Dt} = \frac{P_s}{\rho_s^2} \frac{D\rho_s}{Dt} + \frac{(P_s - P_g)\mathcal{F}}{(\phi_s\rho_s)} + \frac{\mathcal{H}(T_g - T_{int})}{(\phi_s\rho_s)}. \quad (1)$$

The terms on the right hand side of Eq. (1) correspond to heating (increase in internal energy) via compression, compaction (frictional, plastic work, etc.), and heat transfer processes, respectively. This heating is not uniform; "hot spots" form which act as reaction centers. As a first approximation we assume compaction heating is deposited at the particle interface (in addition to convective heat transfer), and compressive heating is uniformly distributed. We employ integral methods (e.g., (9)) to obtain an expression for the interface temperature. The temperature profile in the particle is assumed to be of the form $(T(r) - T_c)/(T_{int} - T_c) = (r/a_{p0})^m$, where T_c is the centerline temperature and m is a constant. We obtain the bulk temperature, T_s , in terms of T_c and T_{int} by integrating the assumed temperature profile to yield $T_s = (mT_c +$

$3T_{int})/(m + 3)$. Before significant reaction occurs, the boundary condition at the interface of a spherical particle is then $k_s(\partial T/\partial r)_{r=a_p} = (a_p/3\phi_s)[\mathcal{H}(T_g - T_{int}) + (P_s - P_g)\mathcal{F}]$. Applying this boundary condition to eliminate T_c gives

$$T_{int} = \frac{(m + 3)T_s + 3BiT_g}{m + 3Bi + 3} + \frac{a_{p0}a_p(P_s - P_g)\mathcal{F}}{3\phi_s k_s(m + 3Bi + 3)}, \quad (2)$$

where $Bi \equiv ha_{p0}/3k_s = k_H a_{p0}/3a_p k_s$. When vigorous reaction begins, this expression is no longer appropriate because reaction was neglected in its derivation. Therefore, T_{int} is not allowed to exceed 600 K.

Effect of Pore Size on Ignition

Ignition and deflagration of energetic materials are greatly influenced by the gas-phase zone (pre-heat and reaction zone), often called the stand-off distance, L_g . The pore size, L_p , in a damaged explosive is estimated to vary from about 0.2 to 15 μm for porosities of 0.01 to 0.4, assuming an effective particle size of 35 μm . The stand-off distance of HMX is estimated to vary from about 0.5 to 50 μm for pressures of 100 to 0.1 MPa, respectively (e.g. (11)). Therefore, conditions can exist where gas-phase ignition would be inhibited by the pore size ($L_p < L_g$); as well as, the case where $L_p > L_g$ (see Fig. 2) (cf. Chapt. 3 of (2)).

To investigate the effects of pore size on ignition and burning we consider an additional ignition constraint for gas-phase driven combustion to occur, $L_p/L_g > \text{const.}$ L_g in the simplest approximation scales with $k_g/(\rho_s r_b C_{vg})$ (e.g. (11)) where k_g , ρ_s , and C_{vg} are the conductivity of the gas, density of the solid and heat capacity of the gas, respectively. In steady gas-phase driven combustion $r_b \sim k_p(P_g/D_p)^{b_n}$ where b_n is a constant and D_p is a scaling constant, so $L_g \sim k_g/(\rho_s k_p(P_g/D_p)^{b_n} C_{vg})$. A measure of the pore size is $L_p \sim \sqrt{\kappa}$ where κ is the permeability. Assuming the correlation for permeability adopted by Baer & Nunziato (BN) (10), we find $L_p \sim a_p \phi_g^{2.25}$. Thus, $L_p/L_g \sim a_p \phi_g^{2.25}(\rho_s k_p(P_g/D_p)^{b_n} C_{vg})/k_g$. Lumping nearly constant parameters into a specified threshold parameter, R_{k0} , we require for gas-

Table 1. Default parameters for $\phi_{s0} = 0.73$.

$\Delta H_{det} = 7.796 \times 10^6 \text{ J/kg}$	$C_{vg} = 2380 \text{ J/kg/K}$
$\Delta H_{pyr} = 2.3 \times 10^6 \text{ J/kg}$	$C_{vs} = 1500 \text{ J/kg/K}$
$t_3 = 4.97 \times 10^5 \text{ J/kg}$	$\tau = 1.2 \times 10^7 \text{ Pa}$
$t_4 = 8.24 \times 10^4 \text{ J/kg}$	$k_I = 1.2 \times 10^5 \text{ s}^{-1}$
$b_n(P_g < 69 \text{ MPa}) = 1.0$	$k_h = 100 \text{ s}^{-1}$
$b_n(P_g \geq 69 \text{ MPa}) = 0.8$	$n = 9.8$
$k_p(P_g < 69 \text{ MPa}) = 0.1227 \text{ m/s}$	$R_1 = 4.2$
$k_p(P_g \geq 69 \text{ MPa}) = 0.1139 \text{ m/s}$	$R_2 = 1.0$
$a_t = 1.35 \times 10^{10} \text{ Pa}$	$\omega = 0.25$
$k_{cw} = 2.5 \times 10^8 \text{ kg/m}^3 \cdot \text{s}$	$D_p = 10^8 \text{ Pa}$
$A = 2.70 \times 10^{11} \text{ Pa}$	$T_{ign} = 525 \text{ K}$
$B = 4.96 \times 10^8 \text{ Pa}$	$W_{c0} = 6 \times 10^{10} \text{ J/kg}$
$\beta_{max} = 5 \times 10^8 \text{ Pa}$	$g_v = 2100 \text{ kg/m}^3$
$k_s = 0.423 \text{ W/m} \cdot \text{K}$	$\mu_c = 100 \text{ kg/m} \cdot \text{s}$
$k_1 = 0.0943 \text{ W/m} \cdot \text{K}$	$R_{k0} = 0.2 \text{ m(Pa)}^{b_n}$
$k_2 = 9.43 \text{ W/m} \cdot \text{K}$	$m = 20$

FIGURE 2. Schematic of the interaction between the characteristic pore size (L_p) and gas-phase preheat & reaction zone (L_g). Three conditions are depicted: (1) high pressure and low ϕ_s , (2) intermediate pressure and intermediate ϕ_s , and (3) low pressure and high ϕ_s .

phase ignition that $R_k \equiv a_p \phi_g^{2.25} P_g^{b_n} > R_{k0}$. This gas-phase zone ignition constraint would be applicable to conditions where ignition leads to gas-phase driven combustion. Gas-phase driven combustion is a condition where conductive heating from the gas-phase zone plays a critical role in sustaining the pyrolyzation of the solid. A plug forms when the pore size is so small that gas-phase reactions are quenched as a result of heat loss to

the pore walls. As a consequence, ignition fails. Gas is not generated to resist the compaction process in this regime, yielding a plug.

Rate Model

This gas-phase driven mode can not be the *only* mode of initiation and burning in explosives since undamaged heterogeneous plastic-bonded explosives with very little void are shock initiated. Initiation of single crystal HMX requires an extremely strong shock. Therefore, localized heterogeneous heating is important. Here we modify the BN reaction rate, albeit crudely, to account for localized compaction heating leading to reaction with the addition of the third term in the following rate expression.

$$C = -3Sk_p (P_g/D_p)^{b_n} \phi_s \rho_s (1-f)^{2/3}/a_p \\ - k_h H(\phi_s - \phi_{s0}) ((P - P_0)/D_p)^2 \rho_s (\phi_s - \phi_{s0}) \\ - H(W_c - W_{c0}) k_{cw} \phi_s, \quad (3)$$

where $S \equiv H(T_{int} - T_{ign})H(R_k - R_{k0})$, $f \equiv (\phi_s - \phi_{s0})/(1 - \phi_{s0})$, W_c is the rate of compaction work (second term on right hand side of Eq. (2)), and W_{c0} is a threshold rate of compaction work. The parameters k_p , k_h , and k_{cw} are rate constants. The first two terms are adopted from (5) with slight modification. The extra gas-phase ignition requirement, $H(R_k - R_{k0})$, has been added to the first term. Results qualitatively similar to those obtained by (5) are obtained when $R_k > R_{k0}$ ($L_p \gg L_g$) everywhere. If $R_k < R_{k0}$ ($L_p \ll L_g$) everywhere the main reaction term (first term in Eq. 3) does not contribute. The induction time equation is also adopted (see Eq. 12 of (8)) with slight modification (the evolution of the induction time variable, I , is not allowed when $R_k < R_{k0}$). The numerical calculations, shown in the following section, utilize MacCormack's method with time splitting. The artificial viscosity described in (8) is adopted.

SIMULATIONS

In the simulations presented we assume the following conditions (see Table 1 for other parameters): $P_{g0} = P_{s0} = 10^5 \text{ Pa}$, $T_{g0} = T_{s0} = 300 \text{ K}$.

$\rho_{s0} = 1900 \text{ kg/m}^3$, $a_{p0} = 7.5 \times 10^{-5} \text{ m}$, and $u_p = 100 \text{ m/s}$ (u_p is the piston speed). We first consider the case where $W_{c0} \rightarrow \infty$; that is, compaction initiated reaction is suppressed. Under these conditions a plug will form, but detonation will not initiate on the top of the plug. At early times the rate of compaction work is seen corresponding to the initial compaction wave (see Fig. 3). After an ignition delay, vigorous reaction begins near the piston. This produces a secondary compaction wave, as indicated by the rate of compaction work profile at $t = 30 \mu\text{s}$. A combined compaction wave then forms. The maximum rate of compaction work increases throughout the simulation. The inset in Fig. 3 shows the x - t plot of the compaction waves for this simulation. Two leading nearly linear wave loci are evident (neither attributable to detonation). In experiments, x - t traces like this have been interpreted as a weak compaction driven reactive wave that may transit suddenly at some later point. Shutting off reaction completely in the high density region ($k_h = 0$ where $R_k < R_{k0}$) yields the same x - t characteristics. Therefore in these simulations the second slope is simply a stronger compaction wave driven by burning near the piston rather than weak reaction in the plug region.

Figure 1 illustrates the base case (see Table 1). The "inert" plug, supported by burning near the piston, forms an accelerating effective piston. In this accelerating system, the compaction wave at the head of the inert plug strengthens with time, as discussed above, and initiation occurs where $W_c > W_{c0}$ (in this case, near the point where the secondary compaction wave overtakes the first). These calculations are qualitatively similar to experimental observation (*cf.* (1)). If the burning rate of the explosive is increased (corresponding to small L_g) by a factor of 10 (all else the same) detonation does occur; but, an "inert" plug is *not* formed since $R_k > R_{k0}$ everywhere. This is in agreement with observations (3). Further, if burning rates are very slow (corresponding to large L_g) detonation is not readily achieved. This is the case for insensitive explosives (4).

This work has demonstrated a physically motivated scenario for plug formation. Observed characteristics and trends are reproduced, including

burning rate trends. Instead of a self-propagating reaction wave, these results indicate that the "compaction driven burning" regime of a DDT event may simply be a strengthened compaction wave augmented by burning near the piston. The reaction modeling remains deficient in numerous ways and is a current area of attention.

FIGURE 3. Evolution of the rate of compaction work when "compaction driven" reaction is not allowed. Inset is the x - t plot of compaction waves for this simulation.

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