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HOT-SPOT REACTION IN UNSUSTAINED SHOCKS*

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Shock waves in reactive media create hot spots which undergo further temperature change (following creation) by means of (i) chemical reaction, (ii) thermal conduction, and (iii) adiabatic effects resulting from pressure variation. A thermodynamic description of exothermic reaction under conditions of variable pressure is presented here. The reaction rate is assumed to be a function of temperature only, and of the Arrhenius form: the effect of variable pressure enters through its influence on temperature. Decreasing pressure significantly alters adiabatic thermal explosion times, and can, under nominal conditions, completely inhibit hot-spot reaction. This effect is discussed in terms of explosive initiation by unsustained shock waves.

1. INTRODUCTION

Shock-wave initiation of solid explosives is achieved by the presence of hot spots; that is, small localized regions of high temperature which are necessary to start the reaction process. Hot spots are typically 0.1 to 10 μm (perhaps larger for very coarse-grained explosives) in radius, or half-thickness if they are planar, and 400-500 K above the mean temperature of the surroundings.[1]

If a shocked explosive containing hot spots is divided into two regions consisting of (i) hot spots (mass fraction μ) and (ii) material exclusive of hot spots (mass fraction 1-μ), the overall extent of reaction λ can be written

$$\lambda = \mu\eta + (1-\mu)\gamma \qquad (1)$$

where η is the extent of hot-spot reaction ($0 < \eta < 1$), whose rate of change is determined by the reaction kinetics at the elevated hot-spot temperature τ, and γ is the extent of reaction of ($0 < \gamma < 1$) for the remainder of the explosive. For constant μ, the overall rate of reaction is then given by

$$\dot{\lambda} = \mu\dot{\eta} + [(1-\lambda) - \mu(1-\eta)]\dot{\gamma}/(1-\gamma) \qquad (2)$$

Hot spots get the reaction started by means of the term $\mu\dot{\eta}$ in Equation (2). Growth of reaction is governed by the additive term involving $\dot{\gamma}/(1-\gamma)$.

At present, most of what is known about shock initiation of solid explosives comes from macroscopic measurement of run distance to detonation (for known initial pressure), particle velocity and pressure as functions of time at points within the sample, and free-surface motion. Exceptions to this statement consist of the work of Von Holle and Lee[2], Von Holle and Tarver[3], and Field, et al.[4] in which direct observations of hot spots have been made.

It is difficult to separate unambiguously the effects of hot-spot reaction (thermal explosion) from growth p[illegible]

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For a reactive hot spot of temperature T subject to Arrhenius kinetics with

$$\dot{\eta} = (1-\eta)\, \nu \exp(-E/RT), \qquad (3)$$

where ν is the frequency factor and E is the activation energy, the constant-pressure time to thermal explosion is given by [5,6]

$$t_p = \frac{RC_p T_o^2}{EQ\nu} \exp(E/RT_o) \qquad (4)$$

where T_o is the *initial* hot-spot temperature, Q is the heat of reaction, and C_p is the constant-pressure specific heat. If t_p is extremely short, say on the order of the risetime of the shock, then whatever time-dependent behavior that occurs behind the shock is a consequence of a reaction growth mechanism and not the thermal explosion of hot spots. However, t_p is a sensitive function of initial hot-spot temperature, which in turn depends on the shock amplitude. Thus, it is expected that for any particular explosive there is a range of shock pressures for which t_p is long compared to other characteristic times of interest, and that thermal conduction and pressure change should each have a strong influence on initiation properties.

In a sustained shock, the pressure is nondecreasing in the early part of the initiation process as shown in Figure 1B-D. At time $\bar{t}_p$ evidence of chemical reaction is seen in the region between the impact surface and the shock front *S*. The physical interpretation of $\bar{t}_p$ is the average time for hot spots to undergo thermal explosion at constant pressure. At time t^* the C^+ characteristics converge (Figure 1A) to form a detonation wave *D*. For unsustained shocks, the rarefaction wave *R*, Figure 2, may be sufficiently strong to reduce the hot-spot temperature and completely inhibit thermal explosion. A quantitative description of the conditions that make this possible is the subject of this paper.

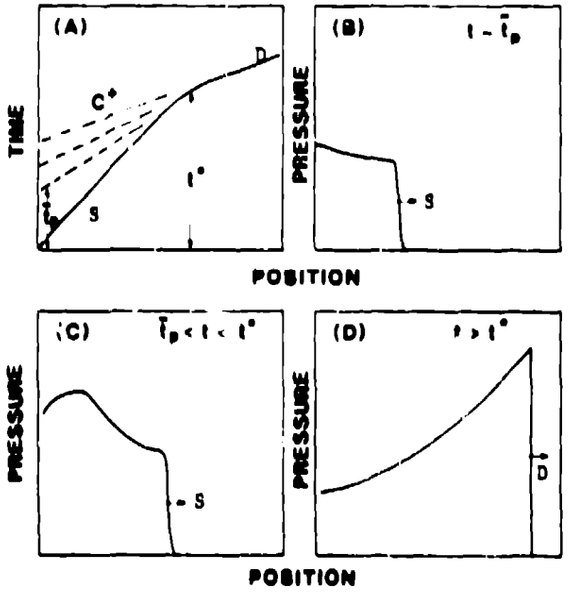

FIGURE 1
Initiation by sustained shock

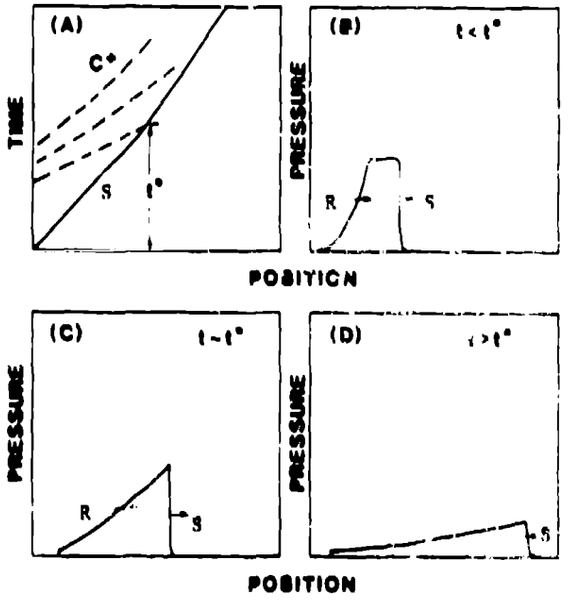

FIGURE 2
Initiation failure:
unsustained shock

2. ENERGY BALANCE

The specific model of the hot spot that is implicit here is one of a constant mass (density ρ) of reactive material at elevated, spatially uniform, temperature $\tau(t)$ embedded in inert, heat conducting surroundings at uniform initial temperature T_o. The hot spot is created within the shock front (t=0), with $\tau(0) = \tau_o$, by some highly irreversible and dissipative process such as void collapse or localized shear deformation; the precise creation mechanism is not important in the present discussion. The analysis presented here applies only to conditions *following* hot-spot creation, in which further pressure change is assumed to be continuous and mechanically nondissipative.

Mechanical equilibration within the hot spot is assumed to take place on a time scale much shorter than other (thermal) characteristic times of interest. Thus, the pressure $p(r,t)$ is taken as a function $p(t)$ involving time alone. The reaction rate is assumed to be a function of the temperature alone and of the Arrhenius form. Variable pressure influences reaction rate indirectly through its effect on temperature.

Under conditions of variable pressure, energy balance requires that the time rate of change of the hot-spot temperature τ be given by[7]

$$\dot{\tau} = (\tau\Gamma/\rho c^2)\dot{p} + (Q/C_p)\dot{\eta} + (K/\rho V C_p) \oint_S (\nabla T)\cdot\hat{n}\, dS, \qquad (5)$$

where Γ is the Grüneisen coefficient, c is the adiabatic sound speed, K is the thermal conductivity, $T(r,t)$ is the temperature in the region outside the hot spot (defined by surface S, with unit normal $\hat{n}$, enclosing volume V).

An approximate solution to Equation (5) is given in Reference 7 for hot spots with a nonzero characteristic constant for thermal conduction, $v_c = (K/\rho C_p)(S/V)$, where S/V is the surface-to-volume ratio of the hot spot; v_c has units of velocity and is a measure of how quickly heat is lost due to thermal conduction over the boundary.

For purposes of discussing hot-spot reaction in unsustained shocks, the adiabatic case, $v_c = 0$, is instructive. It is also useful to employ the exponential approximation to the Arrhenius rate law.[5] Expansion of $1/\tau$ in a Taylor Series about the initial state $1/\tau_o$ in Equation (3), and neglecting the first-order depletion term gives

$$\dot{\eta} \cong \nu \exp(\theta - E/R\tau_o), \qquad (6)$$

where $\theta = E(\tau - \tau_o)/R\tau_o^2$. In the adiabatic case, Equation (5) then becomes

$$t_p \dot{\theta} = A(1 + R\tau_o\theta/E) + \exp(\theta), \qquad (7)$$

$$A = t_p \dot{p} (\Gamma E/R\tau_o\rho c^2), \qquad (8)$$

with initial condition $\theta(0) = 0$

A typical value for $E/R\tau_o$ is 30 and hence in the early stages of reaction $R\tau_o|\theta|/E \ll 1$: under this condition and the special case of $\dot{p}$ = constant (and thus A = constant, neglecting changes in Γ, ρ, and c), a solution of Equation (7) is

$$A \exp(-\theta) = (1 + A)\exp(-At/t_p) - 1 \qquad (9)$$

The time t' at which $\exp(-\theta) \to 0$ (thermal explosion) is given by

$$t'/t_p = A^{-1} \ln(1 + A) \quad . \qquad (10)$$

When $A \to 0$, $t'/t_p \to 1$ as required. Equation (10) describes the way in which the

thermal explosion time is reduced or extended for a constant value of A given by Equation (8).

The critical condition for complete quenching of the adiabatic hot spot occurs for A = -1; that is, when $t'/t_p \to \infty$. In terms of constant $\dot{p}$ this occurs when

$$t_p \dot{p} < - \rho c^2 R t_o / E\Gamma \quad . \tag{11}$$

Nominal values for the physical quantities in Equation (11) are $\rho \sim 2 g/cm^3$, $c \sim 3000$ m/s, $\Gamma \sim 1$, and $E/Rt_o \sim 30$. For PBX-9404, an HMX-based explosive with a nitrocellulose binder, $t_p \sim 1$ μs for a 3.7 GPa shock. Thus a constant value of $\dot{p} \sim -0.6$ GPa/μs is sufficient to inhibit all hot-spot reaction, regardless of size, heat capacity, or thermal conduction.

This condition is easily achieved in unsustained shock initiation experiments. Inequality (11) provides a very useful criterion for determination of conditions under which a constant rate of pressure decay inhibits hot-spot reaction.

4. CONCLUSIONS

Reaction under transient pressure conditions is described by Equation (5) for hot spots of nonzero $v_c = (K/\rho C_p)(S/V)$. For the special case of the adiabatic hot spot, $v_c = 0$, and Arrhenius kinetics in the exponential approximation, Equation (5) can be solved to give nondimensional hot-spot temperature θ as a function time behind an unsustained shock wave in the region of constant $\dot{p}$ (for example, in the rarefaction wave R of Figure 2C). It is found that for $\dot{p}$ given by Inequality (11), time for thermal explosion becomes infinite. The critical condition corresponds to a value of $\dot{p} \sim -0.6$ GPa/μs, commonly achieved in unsustained shocks.

It is obvious that constant $\dot{p}$ cannot be maintained indefinitely, and Inequality (11) serves only as an estimate of pressure decay rates that can influence the initiation process. The formal solution of Equation (5) for the nonadiabatic hot spot and arbitrary p(t) is presented in Reference 7. However, once the pressure has dropped to zero, whether hot spots undergo thermal explosion or not probably has little to do with prompt initiation because of the strong dependence of $\dot{y}$, Equation (2), on pressure.

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