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Relating microstructure, temperature, and chemistry to explosive ignition and shock sensitivity

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ABSTRACT

An analysis is put forth that relates explosive properties including microstructure, temperature, and chemistry to explosive ignition and sensitivity. Unlike approaches that focus only on elucidating mesoscopic mechanisms important to ignition, the present analysis seeks a methodology directly applicable to continuum explosive burn models. Because the Scaled Uniform Reactive Front (SURF) burn model has a microstructural basis, it is chosen as the starting point of the present analysis. To build upon the SURF framework, a literal translation is performed of the high-level conceptual notions for which SURF is based, to concrete ignition–combustion parameters, a statistical description of the microstructure, and other thermo-chemical data acquired by non-shock experiments. The analysis requires a void volume fraction distribution acquired from ultra-small angle neutron scattering (USANS). The shock response of PBX 9502 is used to illustrate the theory. While the analysis requires calibration to a shock experiment (the pop plot, for example), the results are a self-consistent set of realistic physical quantities that contribute to the shock initiation process including, initial temperature, chemical kinetics, a statistical description of the microstructure, and the hot spot size, spacing, and temperature. The utility of having a mesoscale based theory is that once the critical hot spot temperature, as a function of shock pressure, is known for a given explosive and initial porosity distribution, the entire set of meso- and macro-scale results, including the pop plot can be calculated for any other porosity. Thus, one can understand changes in sensitivity at different densities (void size distributions), relying only on the assumption that the hot spot temperature curve would not differ significantly if the morphology of the hots spots were similar. Another important utility of the present analysis is to address the question of the role of initial temperature on the observed shock sensitivity of PBX 9502 and other explosives.

1. Introduction

It is known that micron-sized features play a central role in the shock initiation of explosives [1–3], but exactly what the relevant physical features are that contribute to the process, and how to quantify their effects on shock sensitivity at the continuum level is still a matter of active research. For example, many explosives become significantly more sensitive to detonation at elevated temperature and some, including 2,4,6-triamino-1,3,5-trinitrobenzene (TATB)-based explosives, retain a permanent increase in sensitivity due to irreversible thermal expansion [4]. Experiments show that the porosity formed during thermal expansion is important to shock sensitivity. However, it is not understood what the exact role of temperature is and its effect on chemical reaction. As a second example, inconsistencies in manufacturing parameters can also have a significant effect on sensitivity and performance through, presumably, subtle changes in microstructure. In the present investigation, a framework is introduced that connects normally disparate, but established theories that describe fundamental combustion physics at the microstructural level to the continuum shock initiation behavior in a manner that can ultimately be integrated with contemporary hydrodynamic simulation techniques. Our approach complements other researchers that aim to investigate the microstructural response using high-fidelity modeling, with the primary focus being on elucidating relevant processes purely at the microstructural scale [5–8]. Here, we are developing the methodology to employ the appropriate statistical characterization of those features and processes in a manner that we can potentially apply to bulk materials for determining the continuum explosive response.

The reactive Euler equations form the basis for reactive flow hydrodynamic simulations of shock initiation and detonation behavior in heterogeneous high explosives [9]. So-called, 'burn models'

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account for the reactive component and several are used for a variety of problems: History Variable Reactive Burn (HVRB) [10,11], Ignition and Growth (IG) [12,13], Forest Fire (FF) [14], Program Burn and Detonation Shock Dynamics (DSD) [15–17], Johnson, Tang, Forest (JTF) [18], AWE CREST [19] and Scaled Uniform Reactive Flow (SURF) [20] models. These models have varying degrees of accuracy and utility that depend on the specific system and problem type. IG and SURF were derived using concepts that consider ignition and burning within the microstructure. Ultimately however, these models still require calibration to macroscopic shock experiments to translate the microstructural processes to the continuum response, i.e., they are empirical macroscopic models. In addition to the burden associated with generating the calibration data from full-scale shock initiation experiments, insight that is possible from these physically-based models is lost. This is especially relevant as attempts are made to understand the behavior of insensitive explosives over a wide range of environmental conditions and manufacturing variability.

Because SURF has a microstructural basis [20], it will be chosen as the starting point of the present analysis. To build upon the SURF framework, a literal translation will be performed of the high-level conceptual notions to more fundamental ignition–combustion parameters, a statistical description of the microstructure, and other thermo-chemical data acquired by non-shock experiments. The resulting analysis is then used to compute the response of a one-dimensional system of the TATB-based explosive, PBX 9502, to a well-supported (long duration, constant pressure) shock wave. This process reveals the important meso– and macroscale factors at work, and the quantitative relationship amongst them. It will also reveal, at least for this one-dimensional case, whether or not the proposed mesoscale-based theory can yield physically meaningful results. Upon validation, the analysis connects these basic factors and microstructural information at a subscale level to a continuum-level model, and thus has the potential promise of predicting shock initiation behavior with the need of only a limited number of shock initiation experiments. In addition to gaining fundamental insight, the analysis could enhance our understanding of manufacturing variability of insensitive high explosives. That is, it would benefit the development of new explosives.

The analysis requires a void volume fraction size distribution (referred to succinctly as the void size distribution), physical properties, and chemical kinetics. Here, the insensitive high explosive PBX 9502 is used as a test case. The void size distribution was acquired from ultra-small angle neutron scattering (USANS) [21,22]. The scattered intensity, $I(Q)$, observed in a USANS experiment is directly related to the structure of the sample through the squared Fourier transform of the scattering length density, $\rho(\mathbf{r})$. For the current analysis, it is assumed that the voids present in the PBX 9502 are spherical in shape. USANS measurements were performed at the National Institute of Standards and Technology Center for Neutron Research (NCNR) using the BT-5 Perfect Crystal SANS instrument and a monochromatic neutron wavelength of 0.24 nm. Our analysis then incorporates the computation of a critical hot spot size using analytic ignition theory, a characteristic hot spot size from the void size distribution, and finding the rate at which hot spots coalesce to support the shock wave (i.e., transition to a detonation) with published high pressure burn rate data [23,24].

2. Analysis

The SURF burn model is now briefly reviewed [20]. The model depends on the burning topology, laminar deflagration rate, and the number of critical hot spots that develop into ignition sites (called burn centers in the SURF literature [20]) generated by the passage of a shock wave. The topology function used in SURF is similar to that derived in the statistical hotspot model of Nichols and Tarver [25] and more recently discussed by Hill et al. [26]. The extent of reaction λ (mass fraction of the gas) depends on this burning topology, which in SURF is assigned a simple exponential function

$$\lambda = 1 - e^{s^2}. \tag{1}$$

The dimensionless variable s in Eq. (1) is a scaled length representing the ratio of the radius of the evolving burn front for a representative burn center, at some time τ after the shock wave passes, to the characteristic distance between burn centers, denoted by a_{bc}. The radius of the burn center depends on the pressure-dependent deflagration speed $\bar{D}$, which is assigned the functional dependence

$$\bar{D} \equiv D_o(P_s)\left(\frac{P}{P_o}\right)^n \tag{2}$$

where P_o and n are material parameters. For the given shock, P_s is the value of the shock pressure, while P is local pressure which may increase or decrease after the passage of the shock. a_{bc} can be expressed in terms of the number of burn centers per volume, $\mathcal{N}$. Clearly a_{bc} (and thus $\mathcal{N}$) is expected to be a strong function of P_s. That is, the greater the shock pressure, the greater the number of hot spots that will become burn centers. The specific number of burn centers (number density) is related to the characteristic separation between burn centers by associating a spherical volume with each burn center

$$\mathcal{N} = \frac{1}{\frac{4}{3}\pi a_{bc}^3}. \tag{3}$$

Combining these ideas, SURF expresses s as [20]

$$\begin{aligned} s(\tau) &\equiv \frac{1}{a_{bc}}\int_{\tau_s}^{\tau} \bar{D}(P(t))dt = \frac{D_o(P_s)}{a_{bc}}\int_{\tau_s}^{\tau}\left(\frac{P(t)}{P_o}\right)^n dt \\ &\equiv D(P_s)\ \mathcal{N}^{1/3}\int_{\tau_s}^{\tau}\left(\frac{P(t)}{P_o}\right)^n dt, \end{aligned} \tag{4}$$

where D subsumed the $\sqrt[3]{4\pi/3}$ factor. It has been found that for one-dimensional supported shocks, as is the case for pop plot data, $n = 0$ works well. That value is used in the present analysis for simplicity. Then, in terms of the rate, Eq. (4) simplifies to

$$\dot{s} = D\ \mathcal{N}^{1/3}. \tag{5}$$

At this point, the typical implementation of SURF proceeds by acknowledging the strong dependence of D and $\mathcal{N}$ on P_s by substituting a fitting form for $\dot{s}$,

$$\dot{s} = \exp\left(A + B\ P_s\right), \tag{6}$$

where A and B are fitting parameters calibrated to shock initiation data. By returning to the fundamental components expressed in Eq. (5) one can elucidate the effects of shock pressure, microstructure, ignition chemistry, initial temperature, and other physical properties. That is, Eq. (5) is the jump-off point for the present investigation. The key assumptions are that the SURF representation described is valid, spherical void morphology is appropriate, and that voids are the source of the hot spots. A classic review of other hotspot mechanisms is provided by Field [27].

USANS is sensitive to voids with diameters between about 1 nm and 10 μm. This range includes void sizes that are believed to be important as hotspots (0.1–10 μm) in shock to detonation transitions. Because the USANS measured void distribution contain the entire range of relevant voids (i.e., the critical hot spots that can be either inter- or intra-granular), we can speak in terms of those voids that become critical hot spots upon the passage of a shock (i.e., they become burn centers) as having radii exceeding a (to be