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Microstructural Effects in Shock Ignition

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INTRODUCTION
The dynamic response of explosive microstructures has been explicitly modeled, with the intention of gaining insight into initiation processes from hotspot formation to transition to detonation. In this paper, we focus attention upon the inert material response leading to hotspot ignition. Of interest are particle size and shape effects, constitutive effects of both binder and explosive, and their importance to hotspot formation. Effects of chemical reaction are considered elsewhere.

COMPUTATIONAL APPROACH
As a basis for calculation, we use the explicit Eulerian finite element code, RAVEN, developed by the second author. The calculations were performed using experimentally determined optical micrographs. The micrographs were digitally processed and image enhanced for high contrast, to the point where the HMX particles are white and the binder or void is black. If the resolution of the image is poor, some particles were connected together with small strands of white pixels that were removed by hand. A specialized program was used to read the processed image and "color" each particle so that no two particles of the same color are touching. This step ensured that the interface reconstruction method used in the calculations (developed by David Youngs and enhanced by Norman Johnson) resolves the independent particles correctly (1). A simple modification of the interface reconstruction method guaranteed that if binder or void is present in the same zone as the particle materials, the binder or void material separates the particles. A specified particle velocity was imposed on one edge of the computational domain and a transmitting boundary condition was applied to the opposite edge. The edges parallel to the shock direction had symmetry boundary conditions. Tracer particles provide the Lagrangian time histories of individual points within the calculation.

Shown in Figure 1 is a micrograph of PBX 9501, an HMX based explosive with a polyurethane binder. The micrograph is courtesy of Cavendish Laboratory, University of Cambridge. This micrograph was used for many of the calculations described in this paper. It is not actually representative of PBX 9501, in that the sample preparation technique used apparently removed a
large number of fine particles, thereby biasing the particle size distribution and reducing significantly the effective HMX volume fraction. However, for the purpose of identifying some of the principal effects, and for conducting parametric studies, it will suffice.

Figure 1: Micrograph of PBX 9501 (Courtesy Cavendish Laboratory)  
Figure 2: Digitized micrograph as used in some of the calculations.

Figure 2 shows the digitized and colored micrograph, for use in the computations. We have assigned representative properties of HMX to the particles. For the first series of calculations, we treated the remainder as void material. The effective porosity for this particular micrograph is about 47% - very high compared to the nominal 2% porosity and 5% binder content associated with PBX 9501.

EFFECTS OF PARTICLE MORPHOLOGY
Simulations to observe the inert behavior of HMX were performed on model microstructures containing over 100 particles, with shock input particle velocities of 1000 and 280 m/s. The effect of morphology upon hot-spot generation was observed by performing identical simulations on a replica of the real microstructure where the irregularly shaped particles were replaced with cylinders of the same volume and at the same centroidal location to minimize effects of changes in particle size and distribution. Both microstructures exhibited the largest heating in pockets of small particles trapped between large particles. Both microstructures showed increased connectivity of the hotspots at the higher shock particle velocity, as expected. The major effect of morphology appears to be the ability of irregularly shaped particles to rotate into orientations of predominantly tangential relative particle motion during passage of the shock. In contrast, the circular particles deform mainly by Hertzian contact with subsequent large localized deformations which tend to completely heat small particles to temperatures well above the mean aggregate temperature. The volume of hot-spots tends to be greater in the circular particles, while the magnitude tends to be greater in the irregularly shaped particles. This is significant since both volume and magnitude of the hot-spots determine the sensitivity of the explosive.

MATERIAL PROPERTIES AND CONSTITUTIVE LAWS
Good material property data is as yet unavailable for HMX and for the binder material used in PBX 9501. Thus, although we refer throughout the paper to the material behavior as that of porous HMX or of PBX 9501, the calculations should be taken as representative for these materials, rather than quantitative. For the calculations described in this paper, we used a Gruneisen equation of state, and a Steinberg Guinan constitutive law for the solid (2). The explosive melting point was assumed to depend upon pressure and, consistent with Frey (3), the melt temperature was assumed to increase by 200 K/GPa. The viscosity coefficient for the liquid phase was assumed to obey a pressure and temperature dependence according to

\[ \nu = \nu_0 \exp\left(\frac{P}{P_0}\right) \exp\left(\frac{E}{T_0} - \frac{E}{T}\right) \]
after Frey (3). We used the same material constants as did Frey. The binder material was modeled using the Steinberg Guinan model, and also using a viscoelastic model developed by Bardenhagen (4).

RESPONSE OF A POROUS EXPLOSIVE WITH NO BINDER
Figure 3 shows the compaction of the porous microstructure resulting from input shocks of various strengths. In each case, the frame was made when the shock wave had just reached the right hand side of the micrograph. The strong effect of viscosity upon the flow is very evident here. Even at the high particle velocity of 1.0 km/s, there is very little evidence of incipient vorticity. The pictures are approximately to scale; the compaction associated with the higher particle velocities is much greater than for the lower ones.

The lack of presence of incipient vorticity is in contradistinction to calculations performed by Benson for the shock compression of copper (5), and the experimental observations of Thomas et al (6). In the calculations presented here and in subsequent calculations it was found that the presence of material viscosity exerted a very strong effect upon the final particle morphology. Even small amounts of viscosity prevented vortex formation, shifting its onset to much higher input...
velocities. This is an interesting point which may be of significant importance to composite and nonideal explosive behaviors. Mixing of the ingredients will be extremely important in influencing the rate of reaction of nonideal explosives containing separate fuel and oxidizer components. It may also be of importance in ideal composite explosives, such as the one investigated here, if hotspots form in the binder and ignition results from heat transfer from the binder to the explosive component. This will have to be investigated further.

Corresponding calculated temperature distributions are shown in Figure 4. For these calculations, heating mechanisms include bulk compression, plastic work, and viscous heating. There is no gas in the void space. Viscous heating is by far the dominant heating mechanism here. For the set of input parameters used in these calculations, only a small amount of material is heated to high enough temperatures to be effective ignition sites on time scales of the order of a microsecond or so. Of interest is the serpentine nature and high degree of connectivity associated with the hotspots generated under the stronger shock loading conditions. Note, however, that the original morphology of the large particles is retained, and that the major fraction of the local plastic work occurs in the smaller particles, which undergo significant deformation and heating.

![Temperature contours for the porous microstructure subjected to various particle velocity shock inputs. There is a small temperature rise in each case at the left hand side, associated with an interface heating effect.](image)

A plot of the volume percent material at temperature corresponding to the contour plots is shown in Figure 5. Note that, at the higher particle velocities, a significant fraction of the material is heated to temperatures of 700°K or higher. For HMX, 700°K is at the lower end of temperatures important for shock initiation experiments. The calculated time to explosion for HMX, using thermal explosion theory and kinetics constants from the literature, is about 50 microseconds (7). The range of interest for shock initiation typically involves times of the order of a fraction of a microsecond to a millisecond. The corresponding temperature range, in this case, is from about 650°K to about 900°K.
Figure 5: Volume fraction of material heated to a particular temperature, as a function of input particle velocity.

EFFECTS OF BINDER
Typically, in the development of a new formulation, binder properties were chosen to aid in processibility and to improve other types of sensitivity, such as drop weight impact or skid test sensitivity. Data from the literature (8) do not indicate any strong dependence of explosives shock sensitivity upon binder properties. However, the actual range of binder properties for which there are data is very small.

We examined material property effects, such as density, heat capacity, and heat transfer coefficients, and constitutive effects, such as elastic, elastic/plastic, and viscoelastic, and Hugoniot effects. Equations of state effects were found to dominate: by proper choice of the Hugoniot parameters, we could shift the distribution of energy deposition significantly. The presence or absence of binder made a large difference upon the local thermalization of the shock energy. By comparison, variations in parameters of interest over physically meaningful ranges exhibited only small effects. Of the effects studied, the most important was that of the equation of state parameters.

As an example, the effect of variation of the density of the binder from 1.1 g/cm³ to 1.9 g/cm³ (the theoretical maximum density of HMX) is shown in the form of temperature contour plots in Figure 8.
The results show that the binder at a density of 1.9 gm/cm³ undergoes somewhat less heating than that at a density of 1.1 gm/cm³. This is intuitively appealing: one might expect the best binder material for reduced shock sensitivity would be one which matches the shock impedance of the HMX crystals. Note that the calculations show the explosive temperatures in the calculation for the high density binder to be higher than the explosive temperatures for the low density binder. It raises the interesting question as to whether, under some conditions, one might want to put the heat preferentially into an inert binder phase, to keep the reactive phase cooler, in which case, the choice of an impedance matched binder would not be ideal. This will have to be explored further.

EFFECTS OF INTRAGRANULAR VOIDS
It is well known that HMX and other explosives manufactured by standard procedures contain internal voids in the large particles. We expected that intragranular voids would serve as much more efficient hotspots than the binder material. Most modern PBX formulations have a porosity of the order of one percent. For example, hot pressed PBX 9501 has a nominal porosity of 0.6%, PBX 9404 and LX 14 have porosities of 0.9%. It is not clear how much of this is due to intragranular voids and how much is due to voids within the binder or at the binder-HMX interface.

At some point, of course, as porosity is reduced, one will reach the stage where further reduction in porosity can only occur by shattering the grains and getting binder flow about the pieces. At any rate, the issue of the importance of intragranular voids deserves special attention.

The effect of the intragranular voids on hotspot temperatures was explored with higher resolution calculations. Resolved calculations with a cylindrical intragranular void of various radii were performed, and the results compared with heating produced by intergranular porosity. Other work has shown that the heating from void collapse is sensitive to the assumption of cylindrical or spherical symmetry and several mechanisms of material heating have been identified (9). Here, we are interested in comparing the effects of intragranular and intergranular porosity, and restrict ourselves to two dimensional microstructures.

TEMPORAL BEHAVIOR OF VARIABLES OF INTEREST
In nearly all modeling studies of explosives initiation, the assumption is made of pressure equilibrium (10), temperature equilibrium (13), or both (14). It is therefore of interest to examine how closely these assumptions were met under the conditions of this study. Shown in Figure 10, 11, and 12, respectively, are plots of pressure, velocity, and temperature versus time for two
representative locations in a microstructure with 28% total porosity. The zero point in time is chosen to be the point at which the shock enters the microstructure. We note that, although the mean pressure may be approaching a steady-state value (these calculations were not carried out to sufficiently long times to demonstrate this), the fluctuations, especially at the higher shock strengths, are quite large, and the assumption of pressure equilibrium is not likely to be a good one. This is similarly the case for the particle velocity. The fluctuations are large and persistent, at least over the time scales of these calculations. Interestingly, and of considerable importance, is the fact that the temperature fluctuations are quite small, and a steady state condition is rapidly achieved. This is because the main contribution to the temperatures calculated here come from viscous heating effects, and the contribution from compressive heating is quite small. Thus, although the assumption of pressure equilibrium is a poor one, the effect of lack of pressure equilibrium is small. It will be interesting to see how the inclusion of condensed and gas phase kinetics will influence this conclusion.

Figure 10: Pressure versus time plots for two representative points in a porous microstructure.

Figure 11: Velocity versus time plots for the same two points in the microstructure.
The two dimensional calculations, using experimentally measured microstructures of PBX 9501 as a starting point, have shown that hotspot formation is much more complex than is usually modeled. Most of the heating occurs in a layer of material near the particle surface, for particles without intragranular voids. This high temperature skin is of nearly constant thickness, independent of particle size. It has the effect of making small particles appear much hotter than large ones. At least for bimodal distributions of particle sizes of the sort modeled here, the small particles are much more susceptible to heating due to plastic/viscous heating than the large ones. This effect is so severe that, at high input shock strengths, the morphology of the small particles becomes irrelevant, and the hotspot regions tend to be defined by the larger particles. How important this effect is to the initiation process will need to await evaluation of the effects of appropriate chemical kinetics.

When the porosity is replaced by binder material, hotspot formation is significantly suppressed, consistent with experimental observation. We have not found hotspot formation to be sensitive to either the binder density or yield strength. It is sensitive to equations of state effects. With binder present, intragranular voids appear to assume an extreme importance to ignition.

Viscous heating has been demonstrated by many investigators to be a very important mechanism for hotspot formation (15,16,17). The calculations performed here are consistent with this importance. While the calculations show hotspot formation and heating are strongly dependent upon viscosity, the calculations have also shown a strong sensitivity of the flow field to viscosity. Thus, low viscosity coefficients lead to near turbulent behaviors which are suppressed under conditions of high viscosity.

For the microstructural scales investigated here, the temperature at a particular point reaches a quasi-steady state on the order of 1.5 microseconds. This time is determined by the particle size distribution and porosity investigated. (Times for significant cooling by heat transfer are long with respect to the times of the calculation.) The major temperature effect in these calculations is a result of the interaction between the stress field and the strain field which causes viscoplastic heating. A secondary effect is that of compressive heating (and cooling) associated with the time varying local stress field.
Most hotspot models make either the assumption that pressure equilibrium obtains very rapidly, or
that thermal equilibrium holds. Clearly, assumption of thermal equilibrium is incorrect. However,
we found it surprising that pressure equilibrium did not obtain on time scales of interest. In these
calculations, the pressure fluctuations have only a minor influence on the temperature, as is to be
expected. However, it will be interesting to investigate this effect once chemical reaction and gas
generation are included.

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