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TRENDS IN SHOCK INITIATION OF HETEROGENEOUS EXPLOSIVES

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Various data from the literature on shock initiation were examined to ascertain the relative importance of effects of porosity, particle size, and binder composition upon explosives initiation behavior. Both pure and composite explosives were examined. It was found that the main influence of porosity is manifested through changes in Hugoniot relations. The threshold for initiation was found to be insensitive to porosity, except at very low porosities. The buildup process was found to be weakly dependent upon porosity. Particle size effects were found to depend sensitively upon the nature of the particulates. For inert particles embedded in a reactive continuum, initiation is strongly specific surface area dependent. For HMX particles embedded in inert or reactive continua, particle effects are subtle. Sparse data indicate that binder composition has a small but significant effect upon threshold velocities.

INTRODUCTION

Part of the difficulty in developing physically based models of shock initiation which have genuine predictive capability is that insufficient constraints are often imposed: models are most often applied to very limited data sets which encompass very narrow parameter ranges. Therefore, it seems to be of considerable value to examine the existing shock initiation database to identify trends, similarities, and differences which predictive models must describe, if they are to be of genuinely utility.

In this paper, open-literature data for shock initiation of detonation of heterogeneous explosives in one-dimensional geometries have been examined. The data examined were almost exclusively obtained from wedge test geometries. The intent was to identify and - where possible - isolate physically measurable and controllable parameter effects. Plastic bonded explosives with a variety of different binders and binder concentrations were examined. Data for different pressed explosive particulate materials and particle size distributions were reviewed. Effects of porosity were examined in both binderless and particle-matrix compositions. Effects of inert and reactive binders, and inert and reactive particle fills were examined. Particle size effects were examined. In several instances, the calculated data used by the original authors in their analysis was recalculated to correct for discrepancies and errors in the original analysis. In order to set the stage for the range of parameter effects, porosity influences are examined first.

POROSITY EFFECTS: PURE EXPLOSIVES

Porosity can influence the initiation process through changes in pore size and pore size distribution, which should affect hotspot formation, through changes in the specific energy density, and through changes in the shock Hugoniot. It is useful to discern which effects are strongest.

Some of the most extensive data on effects of porosity upon shock initiation were presented by Lindstrom (1). Lindstrom's initiation data in the shock velocity vs. time to detonation plane for porous tetryl are shown in Figure 1. As can be seen, there are large differences between the data for different porosities. These differences are principally due to the large effect porosity has upon the shock Hugoniot, and can largely be eliminated by re-plotting the data in the particle velocity - time plane. This can be seen by comparing Figure 1 and Figure 2. In Figure 2, the curves for the all but the lowest porosity nearly overlay. However, even in the higher porosity data, there is a small systematic trend.

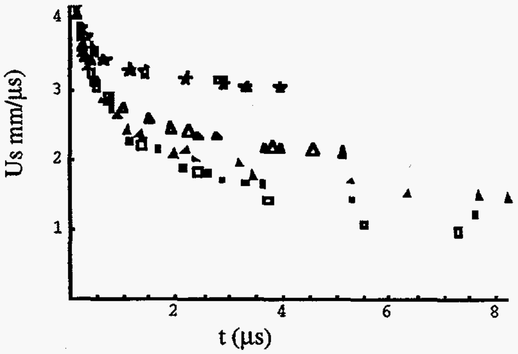

FIGURE 1. LINDSTROM'S DATA FOR TETRYL IN THE Us vs. t PLANE. THE UPPER CURVE IS FOR r = 1.70 g/cm3, FOLLOWED BY 1.60, 1.50, 1.40, 1.30, RESPECTIVELY.

The fact that buildup curves in the Up vs t plane for explosives at different porosities nearly overlay was apparently first noticed by Seely (2),

and has been reported also by Roth (3), Stresau (4), Howe (5), and others.

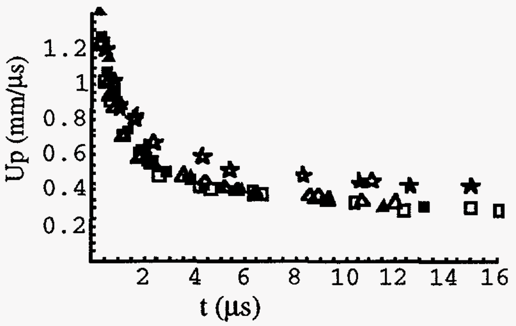

FIGURE 2. LINDSTROM'S TETRYL DATA IN THE Up vs. t PLANE. A SYSTEMATIC VARIATION IN BUILDUP EXISTS.

The nearly hyperbolic behavior exhibited in Figure 2 suggests a useful frame in which to examine the data is the $1/(Up - Up^{o})$ versus time to detonation plane, where Up is the particle velocity, and Up^{o} can be interpreted as a threshold velocity. In this frame, the data should be linear (see Figure 3). True linearity cannot persist throughout the buildup process, as the velocity versus time curve is sigmoidal. However, over the range of data published, which typically does not include the rollover region near the detonation velocity, the data are indeed linear in the $1/(Up - Up^{o})$ vs. time frame, for every explosive examined. This frame is particularly useful for determining Up^{o}. This region of linearity provides the major contribution to the time and distance to detonation and the $1/(Up - Up^{o})$ vs. scaled time representation provides a very sensitive frame for comparison of various data.. Each of the tetryl data sets was examined in the $1/(Up - Up^{o})$ vs. time coordinate system, and values of Up^{o} sought which provided best linearity. A linear least squares fitting routine was used. The dependence of Up^{o} upon porosity is shown in Figure 4 for all five porosities. The threshold velocity was found to be essentially independent of porosity, except at very low porosities, where higher thresholds obtain. This behavior was found to occur in PBX 9404 and 9501, as well.

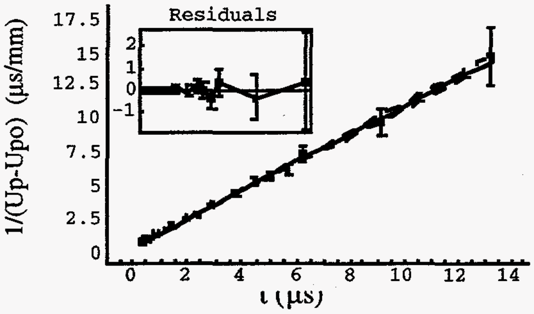

FIGURE 3. ρ = 1.40 g/cm^3 TETRYL DATA PLOTTED IN THE 1/(Up-Upo) vs. TIME TO DETONATION PLANE

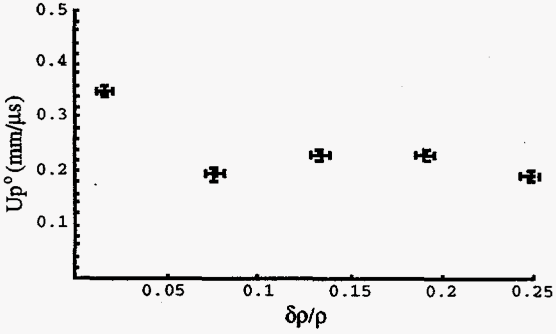

FIGURE 4. DEPENDENCE OF THRESHOLD VELOCITY UPON POROSITY.

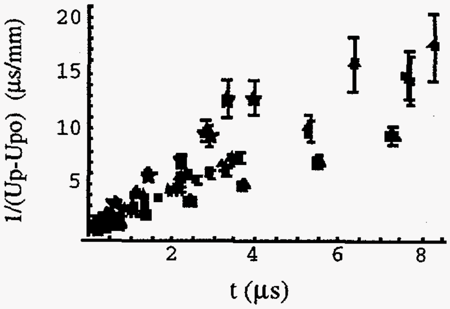

FIGURE 5. TETRYL DATA IN THE $1/(Up-Up^{o})$ vs. TIME TO DETONATION PLANE. TRENDS IN BUILDUP ARE CLEARLY EVIDENT.

Figure 5 clearly shows the existence of a signficant influence of porosity upon buildup to detonation, in that the slopes of the curves vary systematically with porosity. (Slopes are in the order ρ = 1.30, 1.40, 1.50, 1.60, and 1.70 g/cm^3.)

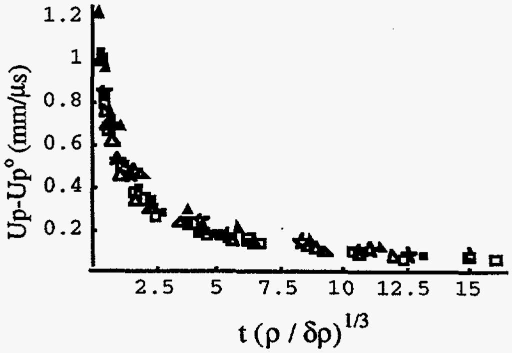

FIGURE 6. LINDSTROM'S TETRYL DATA SCALED.

Scaling the time axis by the cube root of the porosity removes this trend, within the scatter of the data (Figure 6 and Figure 7). (This scaling is likely to be processing dependent, and therefore not general. For example, Reference 6 contains large scale gap test data on TNT pressed to a fixed final density, but under different pressing temperatures. Although the initial particle size and pressing density were held constant, the sensitivities for materials pressed at different temperatures showed markedly different sensitivities.)* For PETN, RDX, and HMX, the data were too sparse to make definitive conclusions. For pure TATB, there was an exceptionally large amount of scatter between the various data sets, and no clear trend was perceived.

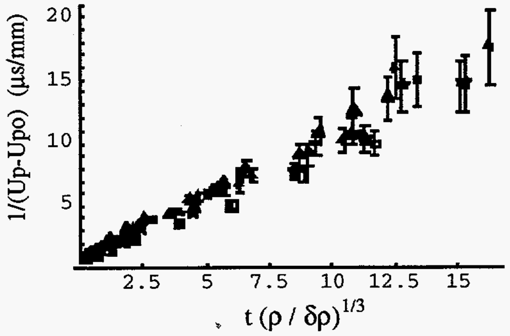

FIGURE 7. TETRYL DATA. TIME AXIS SCALED AS IN FIGURE 5.

For the porous explosives examined, it appears the greatest effect of porosity is to change the shock Hugoniot. The threshold particle velocity is influenced to a lesser extent, and the buildup curves least of all.

POROSITY EFFECTS: COMPOSITE EXPLOSIVES

As noted above, the data for porous compacts of explosives without binders showed a small, but significant, dependence of the threshold particle velocity upon porosity. The effect of this dependence becomes magnified, of course, when transformed to other frames (P vs. t, Us vs. x, etc.).

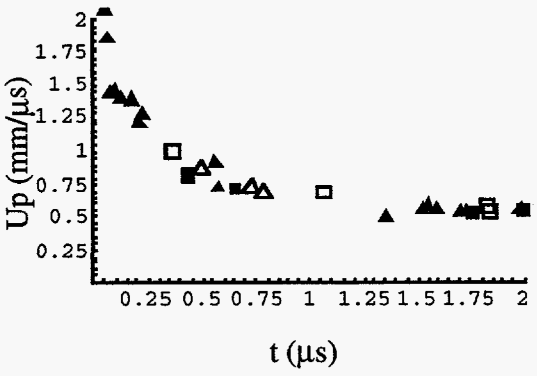

FIGURE 8. DATA FOR PBX 9404 IN THE PARTICLE VELOCITY VS TIME TO DETONATION PLANE.

For the plastic bonded explosives examined, the threshold velocity (Up^o) was similarly found to be essentially independent of porosity, except at very low porosities. The largest database examined was that for PBX 9404 (7). The buildup curve for PBX 9404 for several different porosities is shown in Figure 8. This plot contains data sets for initial densities of 1.72, 1.73, 1.82, 1.83, 1.84, and 1.85 g/cm3. The theoretical maximum density for PBX 9404 is 1.865 g/cm3 (7).

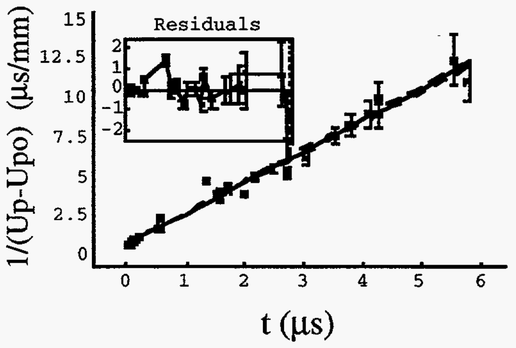

FIGURE 9. DATA FOR PBX 9404 ARE QUITE LINEAR IN THE 1/(Up- Up^o) vs. t PLANE. RESULTS ARE VERY SENSITIVE TO CHOICE OF Up^o .

For PBX 9404 all of the data, over the range of porosities for which data are available, collapse onto a single curve in the 1/(Up- Up^o) vs. t plane. Thus, various data sets for different porosities differ only in the requisite value of Up^o in this particular representation. Figure 9 shows the linearity of the data for the largest PBX 9404 data set ($\rho = 1.84$) in the 1/(Up- Up^o) vs. t plane, and Figure 10 shows the entire data set in the same plane.

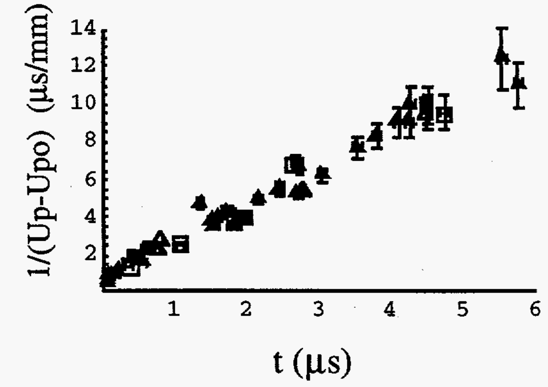

FIGURE 10. WITHIN EXPERIMENTAL ERROR, ALL PBX 9404 DATA COLLAPSE TO A SINGLE CURVE.

Measurement errors are magnified in this plane, and the data are re-plotted in Figure 11, showing indeed that the data collapse to a single

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* The author is indebted to John Ramsay of Los Alamos for bringing this to his attention.

curve. In contrast to the tetryl data, the porosity range is so small here that any porosity effect upon buildup is lost in the noise of the data, independent of the frame of reference. Figure 12 shows the dependence of Up° for PBX 9404 and PBX 9501. The four open symbols are for PBX 9501. Note the insensitivity of the threshold velocity to porosity for all but the lowest porosities. Of considerable interest is the fact that the threshold velocity for PBX 9501 lies significantly below that for PBX 9404, at a given porosity. This difference suggests that the binder itself exerts a measurable influence upon the shock sensitivity of PBXs.

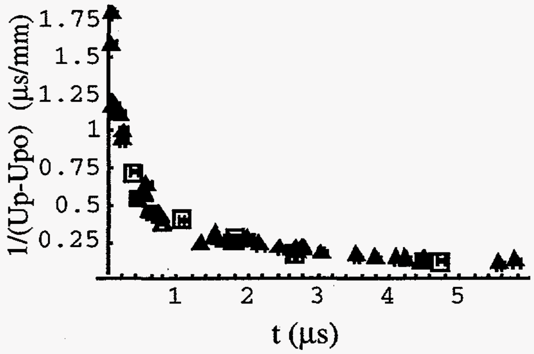

FIGURE 11. AFTER CORRECTION FOR THRESHOLD VELOCITIES, EFFECT OF POROSITY IS MINIMAL IN PBX 9404.

Both PBX 9404 and PBX 9501 have energetic binders. It is unclear whether the difference in sensitivity is due to the difference in reactivity of the binder, or in differences in mechanical behavior. Note that the PBX composites exhibit the same threshold dependence upon porosity that the pure pressed materials do.

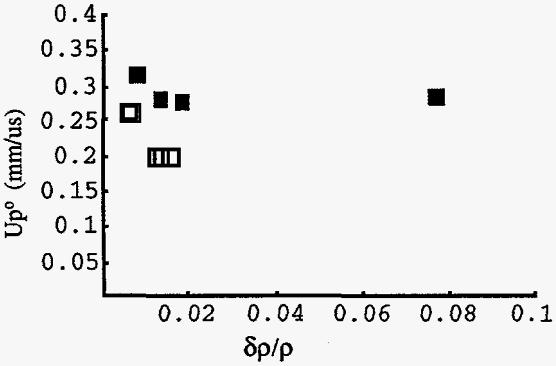

FIGURE 12. DEPENDENCE OF THRESHOLD VELOCITY FOR PBX 9404 and 9501 UPON POROSITY.

Of especial interest is the fact that PBX 9501 is more shock sensitive than PBX 9404, in the sense that it has a lower shock threshold velocity. This difference is not readily apparent from comparison of Pop Plots.

EFFECTS OF PARTICLE SIZE

Numerous investigators have explored the effects of particle size upon shock sensitivity. Thus, Taylor and Ervin (8) found for pressed TNT that large particle size samples ignited at slightly lower pressures than did the fine particle size samples, while buildup occurred more quickly in the fine particle size samples. Their samples were prepared from solvent precipitated TNT, sieve separated. No measurements were made to characterize the various particle size distributions, and no measurements were made to characterize the microstructure after pressing. Thus, it is unclear whether the differences in response were due to particle size effects, particle size distribution effects, or effects of processing.

Moulard reported studies of three different particle sizes of RDX (9). He found a rather complex behavior, which he explained by assuming that post ignition growth of reaction controlled at high pressures, and hotspot formation controlled at low pressures.

Boyle et al (10) reported upon systems of nitromethane with inert particulate additives. Their work showed a very strong dependence of buildup time upon surface area. Their results for nitromethane with Al2O3 particles and for glass particles are shown in Figure 13. Particle sizes for these systems varied from 0.5 μm to 200 μm. Note the strong dependence upon surface area. The authors note this is consistent with assuming the inert particles to be heated by passage of the shock, with subsequent heat transfer to the reactive liquid, and provides strong evidence for a grain burning mechanism.

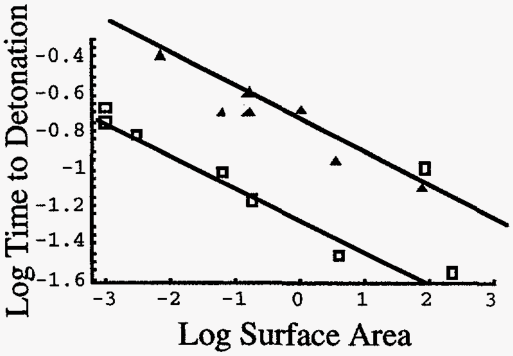

FIGURE 13. EFFECT OF PARTICLE SURFACE AREA UPON TIME TO DETONATION. UPPER CURVE IS FOR Al2O3/NM.

The data of Simpson et al (11) are for systems with relatively low particulate volume fraction (56 to 72%), and thus are likely to provide insight into particle size effects unperturbed by particle fracture associated with pressing higher volume fractions to low porosity. These data span a mean particle size range of 5 to 1700 micron diameters. This is a huge range in particle sizes and an even larger range in surface areas, and the data should be very useful for gaining insights into the dependence of ignition and buildup upon particle size effects. Since the original author was kind enough to provide the original t-x data, these data were examined more closely than those of Taylor, Boyle, or Moulard.[*]

The data for each particle size were first superposed to form a single t-x plot. A nonlinear least squares program was used to fit the t-x data for each combined data set. The data sets were then compared in both the t-x plane and the Us-t plane, where Us is the shock velocity. (Note that, for systems with the same or similar Hugoniots, a constant particle velocity threshold implies a constant or nearly constant shock velocity threshold. Direct comparison of the x-t data and the shock velocities avoids introduction of errors associated with estimates of the Hugoniots.) Shock velocity data were obtained by differentiating a curve fit to the t-x data. The threshold shock velocity and the detonation velocity were also derived from the fit.

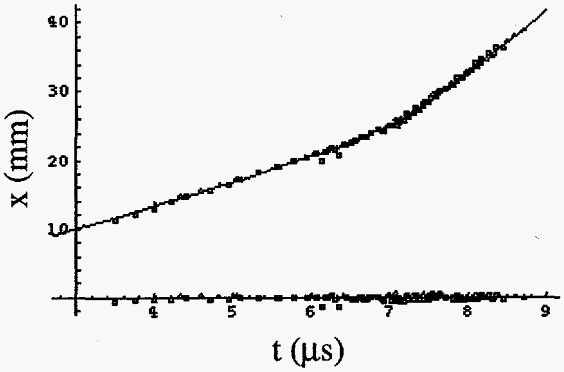

FIGURE 14. t-x DATA FOR 5 & 60 MICRON 56%v HMX/FEFO.

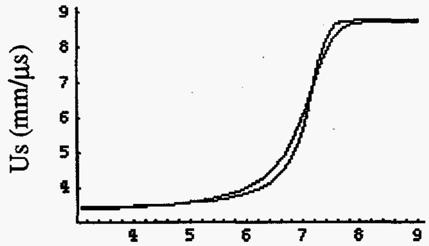

FIGURE 15. SHOCK VELOCITY-TIME FOR 5 & 60 MICRON 56%v HMX/FEFO SYSTEMS.

Figure 14 shows the combined x-t plots for the 5 and 60 micron particle size 56 volume percent HMX/FEFO system. Also shown are residuals of the two data sets compared to a fit to the combined data. There is a very small, but perhaps statistically significant, difference between the two data sets. Although perhaps statistically significant, this is an extremely modest effect. This difference persists in the shock velocity - time plane as well (Figure 15, which shows the shock velocity time results obtained from differentiating the curve fits to the two separate data sets).

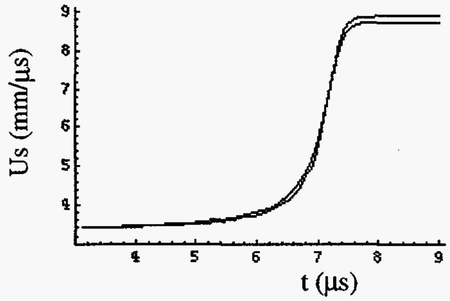

FIGURE 16. SHOCK VELOCITY-TIME FOR 60 MICRON 56%v & 72%v HMX/FEFO SYSTEMS.

Note that the particulate surface area ratio per unit volume for these two systems varies by a nominal factor of 144. For comparison, Figure 16 shows the data for 60 micron 56%v HMX/FEFO and 72%v HMX/FEFO. The effect of this variation in *composition* upon the buildup process is commensurate with that of the particle size effect - i.e, it is negligible- although the composition effect shows up clearly in the difference in detonation velocities.

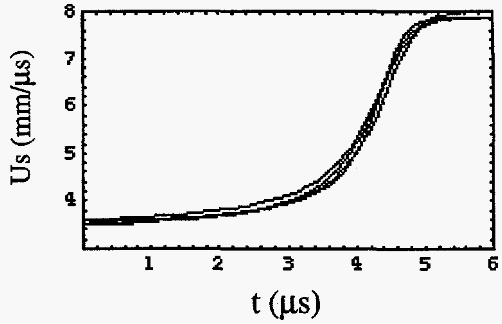

FIGURE 17. SHOCK VELOCITY-TIME FOR 5, 60, 110, & 1700 MICRON HMX/H20 SYSTEMS.

Similar results were obtained from analysis of the HMX/H20 data. Figure 17 shows shock velocity vs. time plots for HMX/H2O experiments with 5, 60, 110, and 1700 micron HMX particle sizes. Figure 18 shows the dependence of the threshold velocity, Us, upon particle size. It is clear (and quite remarkable) that, in contrast to the results for nitromethane / glass and nitromethane/Al2O3, there is a negligible effect of particle size upon the shock initiation process for both the HMX/FEFO and HMX/H2O systems. Indeed, neither the threshold velocity nor the buildup process is found to be appreciably affected by particle size for these systems.

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[*] The author is especially grateful to R. Simpson for providing this data.

Simpson et al report manganin gauge results for the both the HMX/H2O and HMX/FEFO systems. The gauge data clearly show significant differences in the buildup response for the different particle size systems. Results for HMX/H2O 5 micron and 1700 micron systems are shown in Figures 19 and 20. The data points are shock velocities values calculated using shock pressures inferred from the gauge records shown in Simpson's original paper. The curves represent velocity histories calculated from the wedge test t-x data.

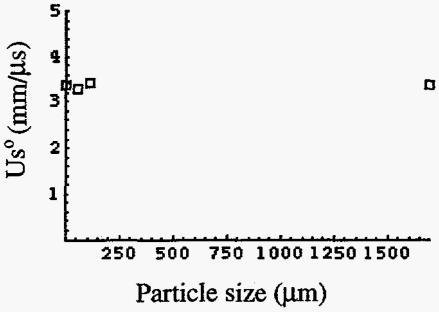

FIGURE 18. DEPENDENCE OF THRESHOLD SHOCK VELOCITY UPON PARTICLE SIZE FOR HMX/H20 SYSTEMS.

The five micron data inferred from gauge records is consistent with the calculated velocity histories using wedge test data. The 1700 micron data are totally inconsistent, however. Note that the deviation between these two sets of velocity histories for the 1700 micron data is far greater than the variations in velocity histories for the entire wedge set experiments, using just t-x data. Part of this deviation may be due to errors in the estimated Hugoniots. However, variation of Hugoniot parameters over an unreasonably large range did not lead to significant improvement of fit. Possibly, the gauges interact sufficiently with the large particles to perturb the flow, but do not appreciably affect the small particle systems.* Results for 60 micron HMX/H2O were intermediate between the 5 and 1700 micron behaviors. Gauge data for the 110 mciron HMX/H2O system were not reported.

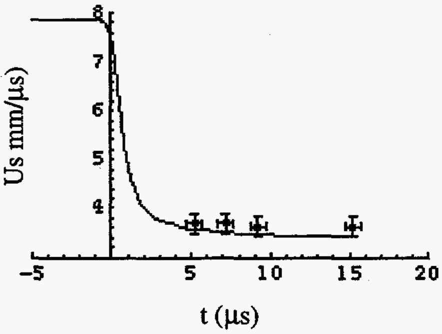

FIGURE 19. VELOCITY vs. DISTANCE FOR 5 mm. HMX/H20, AS DETERMINED FROM WEDGE TEST AND GAUGE DATA.

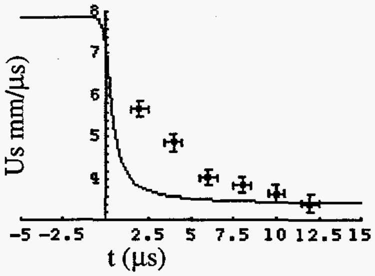

FIGURE 20. VELOCITY vs. DISTANCE FOR 1700 mm. HMX/H20, AS DETERMINED FROM WEDGE TEST AND GAUGE DATA.

DISCUSSION AND SUMMARY

Shock initiation data were reviewed with the intent of identifying trends that might be helpful to the development of initiation models. Effects of porosity, particle sizes and, to a lesser extent, binder compositions were examined. It was found that, for all systems examined, the greatest effect of porosity was upon the Hugoniot, rather than upon the initiation process. Porosity-induced changes in the Hugoniot led to large changes in sensitivity, as measured in terms of time to detonation dependence upon input pressures and shock velocities.

In contrast, porosity was found to have a relatively small effect upon either the threshold particle velocity or the buildup process. For composite plastic bonded explosives (where the range of porosity variation was small) the threshold velocity was found to be essentially independent of porosity, except for very low porosity, under which condition higher thresholds were found. Evidently, there is a small, but significant difference in sensitivity between systems having essentially no porosity and those having some porosity. Further increase in porosity affected sensitivity almost exclusively through changes in the Hugoniot.

The two HMX based plastic bonded explosives that were examined exhibited a significant difference in particle velocity threshold due to the differences in binder. This difference persisted, even when corrections for differences in porosity were made. Its not known whether this is a material property effect or is due to the differences in reactivity of the energetic binders.

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* The original author suggests that the effect is a processing artifact: The large particle size systems were difficult to load, and the gauge package may have been deformed out of the intended plane.

Data for three systems were reviewed for effects of particle size upon shock initiation behavior. The systems included non-energetic particulates in an energetic continuum, energetic particles in an non-energetic continuum, and energetic particles in an energetic continuum. The system containing non-energetic particles in a reactive matrix exhibited strong particle size effects upon the initiation behavior. In this case the time to initiation was shown by the original authors to be inversely proportional to the particle specific surface area. This is consistent with the idea of shock heated particles transferring heat to the reactive medium and supports a classical grain burning mechanism.

Neither wedge test measurements made upon the system with energetic particles in an inert matrix nor measurements made upon the system with energetic particles in an energetic matrix exhibited a significant dependence upon particle size or specific surface area. This is an important contradistinction with respect to the nitromethane data described above and suggests that current initiation models do not adequately describe hotspot behavior or subsequent growth processes.

There are numerous possible explanations. For example, the hotspot heating mechanism might be controlled by intragranular void size and number density, which may or may not track with particle size, but can be expected to be a processing artifact. It has also been suggested that the low volume fraction of the particulates in these systems may lead to very weak interparticle interactions, thereby suppressing the effects of particle size (12). However, a significant particle size effect was observed in the inert particle/nitromethane systems, which also have relatively low solids loading.

Another possible explanation is that the mass fraction of material heated to a temperature necessary for significant reaction - where "significant" in this case means that the reaction releases energy which contributes to the acceleration of the shock front - is a function of impact velocity but is independent of particle size. This hypothesis is substantiated by calculations described elsewhere (13). In these calculations, it is shown that, for monomodal particle size distributions, and where a viscoplastic heating mechanism dominates, the mass fraction of material heated to a given temperature is essentially independent of particle size over velocity ranges examined. Indeed, the calculations show a mass fraction of each particle, independent of particle size, is heated to a given temperature for a given particle velocity. Calculations and experimental data suggest a model of initiation for the HMX systems where the heated reactive material within a particle reacts and contributes energy to the flow, causing the shock wave to strengthen. As the shock strengthens and processes new material, a larger fraction of the material at the shock front is heated to temperatures leading to rapid reaction. This is in contradistinction to a model of particle size dependent hotspots with subsequent erosive burning of the particles.

The fact that the HMX/H2O and HMX/FEFO experiments did not exhibit a significant particle size effect does not eliminate its possibility for other systems, as shown by the studies of the nitromethane/inert particle systems. These showed a significant particle size dependence, manifested through the particle specific surface area. If viscoplastic work performed upon the particles is the dominant heating mechanism, this is to be expected. For the viscoplastic work upon the particles to play an important role in these systems, initiation must occur through heat transfer from the hot particles to the reactive medium. This would introduce the particle size dependence.

The absence of a particle size effect in the HMX/H2O and HMX/FEFO experiments examined also does not eliminate its possibility for other experimental configurations. Times to completion of plastic work by schock loading are particle size dependent. Large particles will take longer to deform by plastic deformation than will small particles. Short duration shocks whose time width is small with respect to particle transit times will thus be less efficient at viscoplastic work heating than longer duration shocks. Setchell (14) has shown ramp wave inputs to be sensitive to particle size effects, as well.

There is a couple of other interesting insights that result from this study. One is that single curve buildup, although it cannot be exactly true, serves as an excellent approximation for the data examined here. This is evident from examination of how well the data sets for a given particle size, porosity, etc., but for differing input velocities overlap in each of the examined frames. Within the error spread of the data, the sets were indistinguishable. To this author, this implies that the reaction rate is controlled by variables at or very close to the shock front, and is rather insensitive to the following flow. This is quite consistent with modeling the principal heating mechanism as viscoplastic work performed upon the particles, as described in Reference 13. It also implies that a significant fraction of the energy release for these systems occurs very close to the front, contributing to (and greatly influencing) the shock acceleration history (15).

It is most interesting that each of the systems examined as part of this study showed a clear and unambiguous threshold velocity, below which initiation was not observed. While the values reported here are almost certainly somewhat configuration dependent, prediction of these thresholds in terms of input material and kinetic parameters, provides a strong challenge to the modeling community.

Although not discussed in this paper, there were numerous instances encountered in this study where it was evident that the processing history associated with the explosive significantly influences its shock sensitivity, well beyond effects captured by porosity, particle size, and composition. This suggests that, at the very least, careful characterization of the microstructure of the explosive should be part of any experiment on shock initiation. In addition, it may indicate that the processing history should be made part of the explosive's pedigree, at least until that time where we have a much better understanding of the relationship between processing variables and microstructure, and microstructure and sensitivity.

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