SAN096-1378C
CONF-961194--3

1996 JANNAF Combustion Subcommittee & Propulsion Systems Hazards Subcommittee Joint Meeting
Naval Post-Graduate School, Monterey CA, November 4-8, 1996

Experimental and Theoretical Investigations of Shock-Induced Flow of Reactive Porous Media*

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ABSTRACT

In this work, the microscale processes of consolidation, deformation and reaction features of shocked porous materials are studied. Time-resolve particle velocities (Sheffield, et al.[1], LANL) and stress fields (Anderson, et al.[2], SNLA) associated with dispersive compaction waves are measured in gas-gun experiments. In these tests, a thin porous layer of HMX is shock-loaded at varied levels. At high impact, significant reaction is triggered by the rapid material distortion during compaction.

In parallel modeling studies, continuum mixture theory is applied to describe the behavior of averaged wave-fields in heterogeneous media. One-dimensional simulations of gas-gun experiments demonstrate that the wave features and interactions with viscoelastic materials in the gauge package are well described by mixture theory, including reflected wave behavior and conditions where significant reaction is initiated.

Numerical simulations of impact on a collection of discrete HMX "crystals" are also presented using shock physics analysis. Three-dimensional simulations indicate that rapid distortion occurs at material contact points; the nature of the dispersive fields includes large amplitude fluctuations of stress with wavelengths of several particle diameters. Localization of energy causes "hot-spots" due to shock focusing and plastic work as material flows into interstitial regions. These numerical experiments demonstrate that "hot-spots" are strongly influenced by multiple crystal interactions. This mesoscale study provides new insights into micromechanical behavior of heterogeneous energetic materials.

INTRODUCTION

The shock response of porous energetic materials involves a complex interaction of thermal, mechanical, and chemical processes associated with dispersive waves. Detailed modeling of the compaction or the crushing processes that takes place during initiation and detonation in porous energetic materials is not currently possible. To achieve this goal, a fundamental understanding of the wave behavior in granular materials is critically needed. Although this paper covers many important topics, no attempt is made to provide a historical or comprehensive review of this area of shock physics. Rather, the intent of this work is to present new ideas in the study of low-velocity impact on porous energetic material. In particular, theoretical and experimental studies are combined to explore the nature of the microscale processes of consolidation, deformation and reaction features of porous materials.

Time-resolved measurements of shock wave structure in porous, heterogeneous materials are relatively new advances in the shock physics field. Recently, shock impact experiments have measured detailed particle velocities (Sheffield, et al.[1], LANL) and stress fields (Anderson, et al.[2], SNLA) of compaction waves in a thin porous layer of HMX. These measurements provide the means for assessing continuum models of compaction and reaction wave structure in granular materials.

Theoretical modeling of the shock response of granular materials is still an active area of research. In this work, several levels of modeling are used to investigate shocked granular materials. The response, at the macrolevel, is modeled using a continuum mixture theory developed by Baer and Nunziato[3] and implemented in the shock physics code, CTH[4]. This approach describes the wave behavior of reactive granular HMX including initiation, growth of reaction and reflected wave behavior. An important requirement for resolving the wave structure in the granular material is the modeling of the viscoelastic response of the gauge packages and confining materials used in the gas-gun experiments.

At a more fundamental level, initiation of heterogeneous materials takes place because shocks interact with material heterogeneities, causing "hot-spots" that lead to reaction. Localization of energy produces associated space-time fluctuations in the thermodynamic fields (such as pressure and temperature). When averaged over a sufficient large space, such variations produce an average heat release that is a combination of chemistry and mechanics. Although a variety of physical phenomena result in the creation of "hot-spots", it is generally agreed that plastic deformation is the key feature of the energy localization. This deformation is due to atomic rearrangement along dislocations and the prevailing theory of shock-induced "hot-spots", such as that posed by Coffey[5], is the result of photon energy transport/dis-

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sipation along moving dislocations. This atomic-level view, by itself, may be too simplistic; the chemical and physical processes associated with "hot-spots" involve fluctuating states at the mesoscale whereby multiple grains or crystals interact. The nature of the dispersion in a compaction/compression wave in porous material involve complex wave mechanical behavior.

In the sections to follow, the gas-gun experiments are described including the viscoelastic effects of the associated gauges and confining cell materials. Next, continuum mixture theory is reviewed and applied to a set of loading conditions of the experiments. Continuum mixture model is shown to replicate the wave behavior measured at the input and exit locations of a thin HMX layer at impact conditions when weak and strong reaction occur.

To complement continuum modeling, the crystal interactions in a compaction/reaction wave are modeled. This work presents a preliminary study of shock loading on discrete HMX "crystals" idealized as random cubes. A representative binder-HMX system is also studied in which a polyurethane binder fills an ordered field of spherical HMX "crystals". The fluctuating stress states and localizations of energy are resolved using three-dimensional shock physics analysis with appropriate EOS and elastic-plastic material strength descriptions. This mesoscale modeling study provides new insight into micromechanical behavior of heterogeneous energetic materials

GAS-GUN EXPERIMENTS WITH POROUS HMX

Impact experiments use gas-gun-driven projectiles to provide precisely controlled inputs to porous compacts enclosed in a plastic cell. Figure 1 shows a schematic view of a projectile impactor and target. A 3.9-mm thick HMX powder compact is confined between a polychlorotrifluoroethylene (Kel-F) plastic front disk and a poly-4-methyl-1-pentene (TPX) or polymethylmethacrylate (PMMA) plastic back disk. Magnetic particle velocity or polyvinylidene difluoride (PVDF) stress gauges are located on the front and back surface of the HMX. Projectiles faced with Kel-F impact the Kel-F front disk covering the compact. The gauge at the interface of the Kel-F front disk and HMX yields the input loading profile. The gauge at the interface of the back disk and HMX measures the transmitted wave profile.

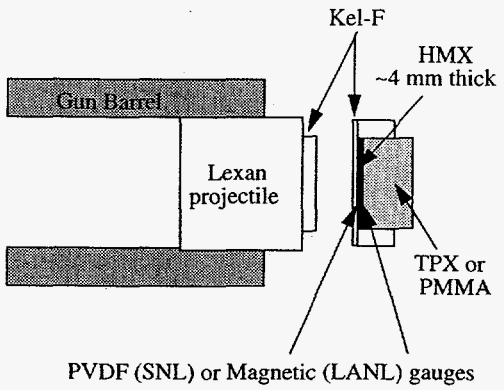

Fig. 1 Projectile and target used in shock loading experiments on porous HMX.

In these gas-gun experiments stress and particle velocity histories are measured in separate but nearly identical experiments. To achieve the same input, projectile velocities are as close to the same as possible. Experiments were performed at projectile velocities ranging from 0.27 to 0.7 km/s with corresponding stress inputs to the HMX of between 0.2 and 0.9 GPa. Additional experiments were performed at velocities as low as 0.08 km/s and as high as 0.99 km/s.

The HMX powder used in both experiments was Holston batch HOL 920-32 that has been used in other initiation studies.[6] The coarse HMX is comprised of sharp edged crystals with a mean particle size of about 120 μm. The HMX was pressed to a density of 1.24 g/cm^3 (65% TMD).

Additional tests used "fine" HMX composed of particles with an average particle size of about 10–15 μm. The powder also contains an occasional large particle of ~50 μm. Although the average particle size of the coarse and fine HMX different by about an order of magnitude, the unpressed materials have overlapping particle size distributions; no attempt was made to selectively size the coarse and fine materials. A variation in initial loading density of 1.40 g/cm^3 (74% TMD) has also been studied in these experiments. In this work, only the coarse HMX experiments are discussed and the interested reader can find details of these tests in References 1 and 7.

This technique of simultaneously measuring stress and particle velocity in similar experiments represents a significant advance in measurement technology. This technique reduces the uncertainties caused by time-dependent material response; in this case it has provided valuable information relating to the compaction process and also the reaction that takes place at higher input levels.

To appropriately model the gas-gun experiments, the coupled response of the polymer impactor, the cell front, and the backing materials must be included. Separate gas-gun tests, using symmetric impacts of polymers, were conducted for the purpose of generating visocelastic data. Shock data for polymer materials, Kel-F and PMMA are fit to a Maxwell viscoelastic description following the work of Schuler and Nunziato.[8] Briefly, one-dimensional stress-stain behavior of a Maxwell material is described by the relationship:

$$\dot{\sigma} = E(\sigma,\varepsilon)\dot{\varepsilon} + G(\sigma,\varepsilon) \tag{1}$$

where σ is the stress and ε is the material strain. Specific forms of the tangent modulus, E, and the relaxation function, G, are given in r
Reference 9 as:

$$E(\sigma,\varepsilon) = d\sigma_I/d\varepsilon \tag{2}$$

and

$$G(\sigma,\varepsilon) = [\sigma - \sigma_E(\varepsilon)]/\tau . \quad (3)$$

PVDF data[10] of symmetric impacts of Kel-F on Kel-F are used to calibrate the viscoelastic model. Figure 2 displays the impact data using PVDF stress-rate data given as stress vs. shock velocity. Steady wave analysis is used to determine the equilibrated and instantaneous stress as a function of strain. This internal state model was implemented into CTH[11] using appropriate solution methods.

As a secondary check, the Maxwell model is applied to a set of impact experiments[12] where Vistal is used as an impactor and embedded magnetic velocity gauges measure the particle velocity at various distances from initial impact. Figure 3 displays a comparison of LANL velocity gauge measurements to the CTH numerical simulation for an impact condition of 0.341 km/s. Additional impact conditions, including multiple impact and reloading, have also been mod-

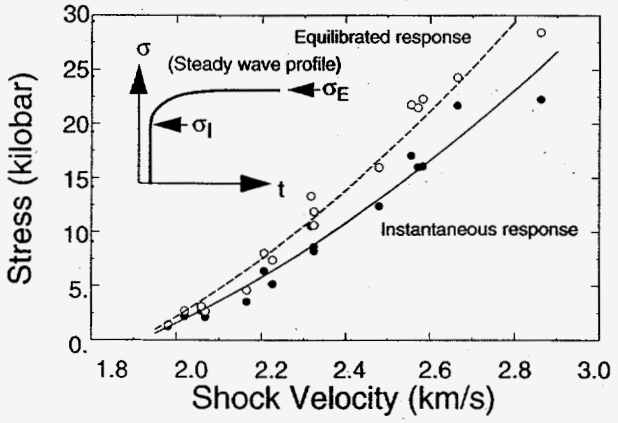

Fig. 2 PVDF data of instantaneous and equilibrated stress states observed in symmetric impacts of Kel-F.

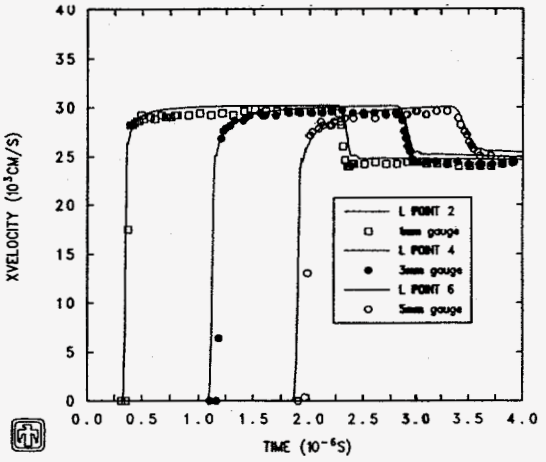

Fig. 3 A comparison of experimental data of particle velocity vs. CTH calculations using the Maxwell viscoelastic model for Kel-F.

eled.[13] In contrast to the sharp shock rise during planar impact in elastic materials, viscoelastic materials jump to a instantaneous state then relax to an equilibrated state. This behavior is shown in Figure 3. Clearly, the viscoelastic response of the gauge package and Kel-F cell front have been resolved to provide accurate loading information in the HMX tests.

OVERVIEW OF THE CONTINUUM MIXTURE MODEL AND NUMERICAL IMPLEMENTATION INTO CTH SHOCK PHYSICS ANALYSIS

In this study, the multiphase reactive flow model of Baer and Nunziato[3] is used as a theoretical framework for modeling the gas-gun tests. A complete derivation of this theory and its numerical implementation into the multidimensional shock physics code CTH is well documented in prior works (see References 4 and 15) and is not repeated here.

Mixture theory is based on a continuum approximation which averages thermal, mechanical, and chemical fields for a localized collection of materials representative of a heterogeneous mixture. This approach requires that the smallest resolved length scale is considerably larger than a typical particle or pore size in the mixture. This does not imply that processes at the microscale are negligible, but rather, that interactions associated with the discrete nature of the mixture are included as submodels. In contrast to pure materials, a mixture average for a multiphase description includes the effects of internal boundaries (or phase interfaces) across which the interchange of mass, momentum and energy occurs. Modern developments in continuum mixture theory provide a well-defined framework for a thermodynamically consistent description of compressible mixtures.[15] The theory of reactive mixtures is based on establishing balance equations and admissible constitutive relationships that are constrained by the Second Law of Thermodynamics.

In its general form, independent balance laws for each phase are derive and mixture rules are imposed and constrained to preserve overall balance of mass, momentum and energy for the total system. The entropy inequality and constitutive principles serve as guidelines for formulating micromechanical models of the interactions between phases. Most importantly, volume fraction is treated as an independent kinematic variable which brings into the description volume fraction rate dependence without any conflict with compressibility and the introduction of an effective stress which reflects mechanics effects associated with contact stresses in the granular media.

Although continuum mechanics suggests admissible relationships for the phase interactions, exchange coefficients for these relationships are undetermined. These coefficients need not be constant and can depend on all phase variables provided that symmetry and positivity are preserved. Thus, experimental guidance is required for a complete

determination of the constitutive relationships. Indeed, the interactive use of calculation and experimentation quantitatively calibrates the mixture description and assists in elucidating the important mechanisms observed in experiments. Much of this foundational work is outline in prior publications and is not repeated here, the interested reader can find such information in References 3 and 16.

The general equations of motion for a multiphase mixture are recast into an integral form consistent with the finite volume form used in modern shock physics analysis. Here, we use an implementation into the shock physics code, CTH, which is a multi-material multidimensional Eulerian finite volume code.[14]

CTH uses an Eulerian mesh which is fixed in time and space. Mixture-averaged conservation equations are solved in a Lagrangian step and distorted cells are remapped back to the fixed mesh. In addition to overall conservation equations, internal state variables are solved for various material models.[11] Overall conservation of mass, momentum and energy is preserved, and relative flow effects appear as phase diffusion effects due to velocity differences between individual phase particle velocities and the mixture mass-averaged velocity. Phase interaction effects, such as mass/energy exchange, drag and heat transfer, are cell volume-averaged quantities resolved during the Lagrangian step. As one might expect, these effects have vastly different length and time scales, hence, appropriate stiff numerical solvers are necessary. Prior to remapping the internal state variable, relative flow effects are included using a high-order convection scheme. Details of this numerical implementation are given in Reference 4.

MODEL APPLICATION TO IMPACT EXPERIMENTS

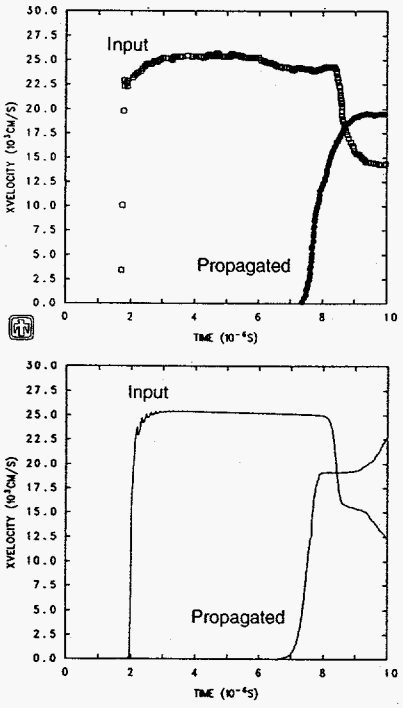

Fig. 4 Comparison of measured (top) and calculated (bottom) particle velocities at input and output positions of the HMX layer

In applying continuum mixture theory, we first consider impact conditions where reaction does not occur in the porous layer of HMX. One-dimensional simulations for an impact condition of ~300 m/s includes coupled response of the polymer impactor and backing materials. In Figures 4 and 5 the experimental data are plotted separate to the CTH calculation which superimpose Lagrangian data at the input and output side of the ~4mm layer of porous HMX (1.24 g/cm^3 density). Figure 4 compares the LANL velocity gauge measurements and with the numerical simulation using the CTH continuum mixture model.

As seen in the particle velocity measurements, the input loading reflects the viscoelastic response of the impact load transmitted through the Kel-F polymer of the cell front. The CTH calculation reproduces this behavior and numerical simulations predict a sharp rise and fall of the pressure at the input side of the HMX layer (similar to the stress measurements) due to the impedance mismatch at the Kel-F in the PVDF gauge package and HMX interface as seen in Figure 5. Thereafter, the pressure relaxes back to ~2.5 Kbar until the reflected wave from the HMX/PMMA causes additional pressure rise to ~7 Kbar. The slight relaxation observed in the velocity and stress measurements after ~5 μs is not predicted. It is suspected that this may be due to the initial nonuniformity of the pressed layer of coarse HMX particles. Repeated tests using fine particle HMX (producing a more uniform density layer) indicate that nearly constant particle velocity is sustained until the reflected wave is observed.

At the output side of the porous column, the propagated wave is dispersive, reflecting the rate processes associated with the compaction wave. The transmitted wave has a rise time in excess of 500 ns. After 10 μs, multidimensional effects are expected to take place. The CTH calculations replicate well the measurements of particle velocity and stress prior to the onset of multidimensional wave behavior.

The transient solid volume fraction profiles are given in Figure 6 indicating that rapid distortion of the HMX occurs in a thin shock-like compaction wave having a wave thickness ~ 0.5 mm. Similarly, the dispersive nature of the compaction wave is evident in the pressure wave profiles. After compaction traverses the HMX layer, wave reflection from the PMMA interface occurs rapidly as a result of a higher sound speed in the nearly fully compacted HMX material.

At higher impact conditions reaction of the granular HMX is induced. Figure 7 shows a comparison of experimental particle velocity measurements to CTH calculations at the input and output locations of the HMX layer. The figure on the left represents impact at ~ 4 kilobars. At this impact level, numerical calculations suggest that weak reaction should be evident and the particle velocity measurements indicate a slight falloff as reaction decelerates the impactor. As the compaction wave reflects off the backing surface weak reaction is evident. Although the wave speed through the porous HMX is correctly modeled in the CTH calculation, the growth of reaction is delayed in response to this reflected behavior.

The loading condition at ~7 kbars is shown on the right side of Figure 7. The CTH calculations are overlaid to measured particle velocity at the input and output ends of the HMX layer. At this impact condition, the coarse HMX

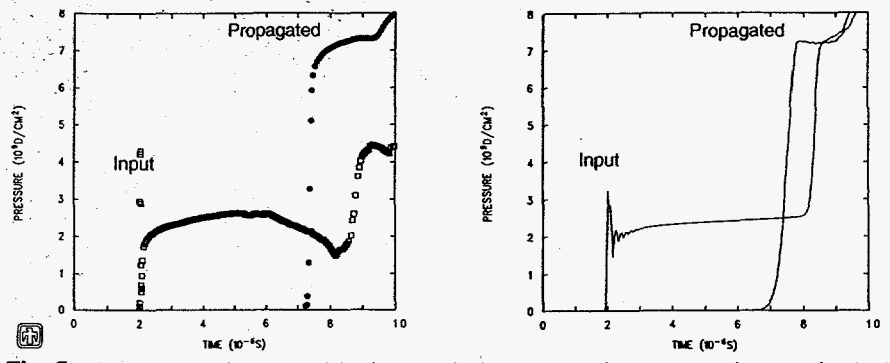

Fig. 5 Comparison of measured (top) and calculated (bottom) pressure at input and output positions of the HMX layer

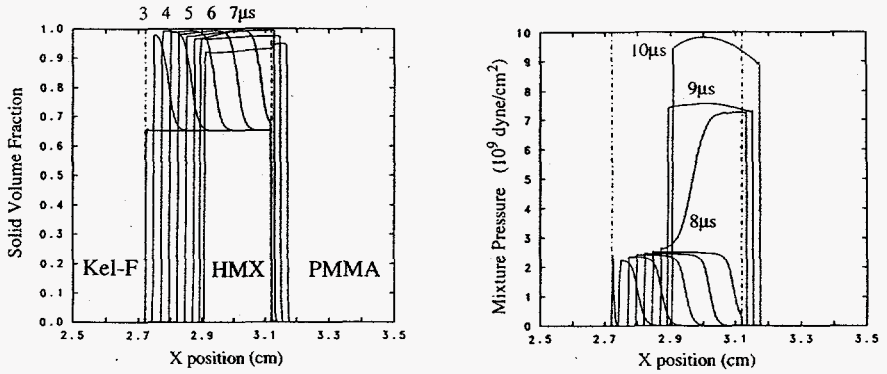

Fig. 6 Overlay of wave profiles of solid volume fraction (left) and mixture pressure (right) as a compaction wave traverses the HMX layer then reflects off of the PMMA interface.

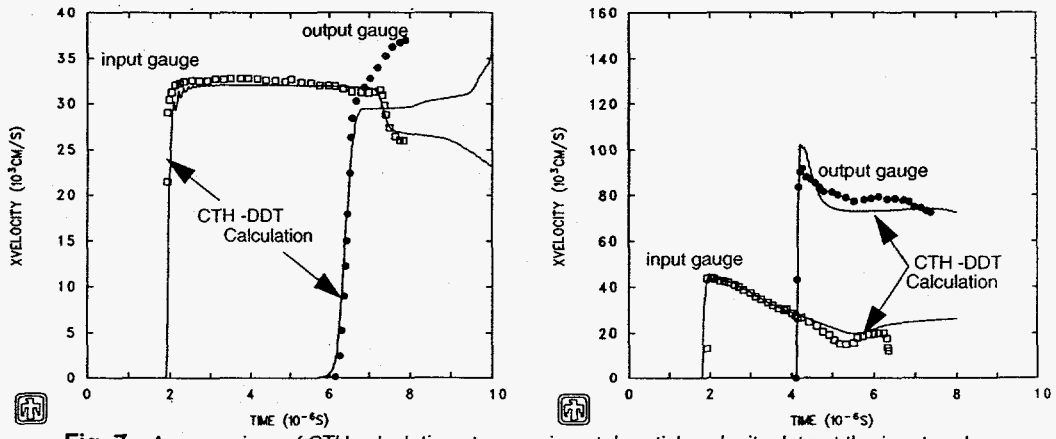

Fig. 7 A comparison of CTH calculations to experimental particle velocity data at the input and output locations of the HMX layer for impact conditions at ~4 kbar (left) and at ~7kbar (right).

begins to react as soon as the wave passes the front gauge and enters the powder. The front particle velocity decreases because the reacting HMX is decelerating the cell front where the gauge is located. Stress measurements, at this same experimental condition, indicate that the stress at this interface is increasing. The transmitted wave grows and steepens considerably. By the time the wave reaches the back of the HMX layer, the particle velocity has nearly doubled as is predicted by the continuum mixture model. Even the reflected wave behavior in the front gauge is replicated in the modeling. In all of these tests, multidimensional effects due to lateral release are expected to take place ~10 μs after initial impact, hence, the comparison of 1D modeling to experimental measurement is not meaningful.

Additional tests have provided information regarding variations of loading density and particle size. In general, higher density HMX seems to be slightly less sensitive. As one might expect, more energy is deposited in the lower density material for a given input pressure. Reactivity greatly depends on the initial particle size with reaction beginning at a much lower pressure in the coarse than in the fine HMX. However, when combustion begins in the fine HMX, the overall reaction is faster due to the effect of higher specific surface area consistent with grain-burning models[16].

MESOSCALE MODELING STUDIES

The reactivity of porous energetic materials greatly depends on the nature of "hot-spots" that are formed by a shock as it moves through the material. As observed in the gas-gun experiments, coarse HMX appears to produce "hot-spots" that are large enough to persist for a long time while fine material seems to produce "hot-spots" that are smaller in size and dissipate quickly. The threshold-to-initiation is the limit where exothermic chemical energy release is balanced by energy dissipated away from the "hot-spot" reaction. A detailed study of the nature of energy localization has been undertaken to better characterize the processes occurring at the mesoscale. Currently, there does not exist well-defined experimental measurement techniques of the nonequilibrium processes associated with "hot-spots". It is important to establish what needs to be measured and at what scale in space and time.

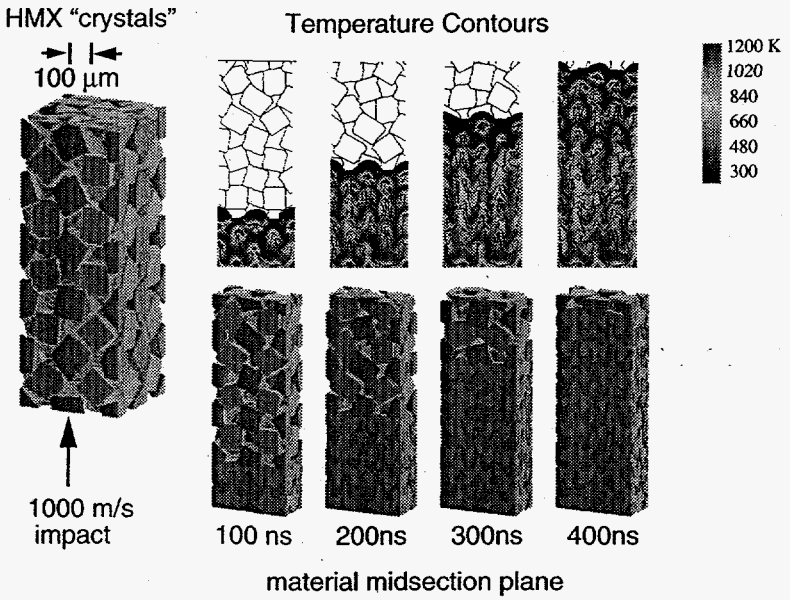

Fig. 8 A numerical simulation of impact on a collection of HMX "crystals". Material and temperature contours are displayed at various time planes.

With the advent of high performance computing, it is now possible to model three-dimensional wave behavior in complex multi-material geometries including many relevant aspects of material physics. In a preliminary study, CTH shock physics analysis is used to model the shock loading of an idealized geometry of packed HMX "crystals" represented by randomly-oriented cubes packed to a density of ~75% TMD. Similar micromechanical modeling has been previously attempted in two-dimensions[17,18]. To our best knowledge, the current work represents the first modeling of near-realistic three-dimensional geometrical effects.

A representative HMX "crystal" field is displayed on the left side of Figure 8. Although these crystals appear to be of different shades, implying different materials, all of the crystals use the same materia models. A Mie-Gruneisen equation of state is used for the HMX and an elastic-perfectly plastic model with yield at ~1 Kbar is included for material strength. The intent with the use of simple mapterial models in these simulations is to examine qualitative micromechanical behavior that is attributable to geometric interactions within crystals. Additionally, these simulations did not include any effects of thermal conduction dissipation and chemistry. Thermal conduction is a relatively new addition in CTH and the inclusion of multistep chemistry is a near future addition[19].

In these numerical simulations, a representative cluster of ~ 200 particles is shock-loaded. At the lateral boundaries of the computational domain, symmetry boundary conditions are imposed. At the bottom of the domain a

hard wall boundary condition is imposed and a reverse ballistic calculation is represented by specifying an initial velocity of the crystal array toward the hard wall boundary. At the opposite boundary, extrapolated boundary conditions are imposed. In all of these computations, the cell size in the three directions is ~ 5 μm, thus, the entire domain encompasses several million computational cells. Each crystal cube has approximately 20x20x20 internal cells. In the near future, the availability of parallel machine platforms potentially will extend computations to ~300 million cells.

Figure 8 displays a numerical calculation at an impact condition of 1000 m/s. The material boundaries are displayed at 100 ns time intervals. As the shock initially passes into the material, material deformation first occurs at contact points. Patterns of release and shock focussing takes place as the shock wave moves from crystal to crystal. At this condition, material distortion takes place near the contact points and plastic deformation causes flow into the interstitial regions. After the open regions are filled, material folding of the crystal boundaries is observed. The compaction process takes place over several particle lengths.

Above each time plane of material interfaces, contours of temperature are displayed for the center cross-section of the crystal array. Hot-spots first arise at contact points due to effects of plastic deformation. The effect of shock focusing is seen to take place within individual crystals. Since the effects of thermal dissipation are not included in these calculations the effect of thermal decay is not predicted. Numerical simulations have been performed which indicate that the size of the "hot-spots" directly scales with particle size.

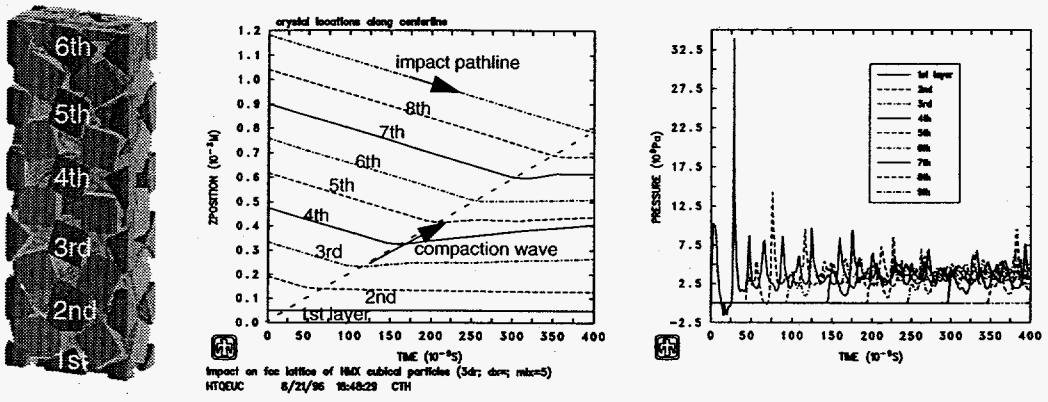

Fig. 9 Particle pathlines and stress histories along the centerline of the HMX crystal array.

Lagrangian tracer points were included at the center of each HMX crystal along the array centerline and the pathlines are plotted in the characteristic plane displayed on the left side of Figure 9. These pathlines show that compaction is normal to the impact direction and slight nonuniform motion is due to intercrystal interactions. At these locations, the stresses are also monitored and the right figure overlay the pressure histories along the centerline. These fluctuating fields are due to the effects of shocks interacting with individual material surfaces and crystal interactions. Interestingly, the amplitude of the fluctuating pressure field is of the same amplitude as the mean pressure state. For example, in this calculation, the mean pressure field is ~ 40 Kbars and the amplitudes of the shocks ringing in the crystals are of the same order. These highly fluctuating stress states persist for many particle diameters.

Clearly, nonequilibrium stress fields are induced by shock in heterogeneous material. These fluctuations may induce breakage of material. Recent experimental studies[7] has used granular sugar as a simulant to separate the effect of reaction from mechanical response. These tests indicate that the electrical "noise" in the particle velocity data is linked to the dynamic processes associated with compaction. It is speculated that this information is related to the stress fluctuations and subsequent "noise" due to crystal breakage may be a real-time diagnostic of dynamic fragmentation. Future experiments at LANL are in progress to explore this behavior.

An additional numerical simulation is displayed in Figure 9 representing HMX "crystals" in a polyurethane binder. Here the crystals are represented as spherical and the polymeric binder uses an appropriate tabular equation of state for polyurethane.[20] A elastic-perfectly plastic material strength model is applied for the binder with a yield stress of ~0.1 Kbar and a Poisson's ratio of ~0.45. The material surfaces are displayed concurrently with midplane contours of temperature and pressure at various times. The solid volume fraction of the HMX is ~0.75. Due to the higher mixture density, the shock moves nearly twice the velocity as that seen in the prior porous HMX simulation. Although the pores are filled with polyurethane, the shock passes through material interfaces and dispersion of the wave is still

seen. As one expects, much of the material distortion occur in the binder material, however, the effect of shock focusing in the HMX crystals is evident. Again, the stress field exhibit fluctuations and nonequilibrium behavior.

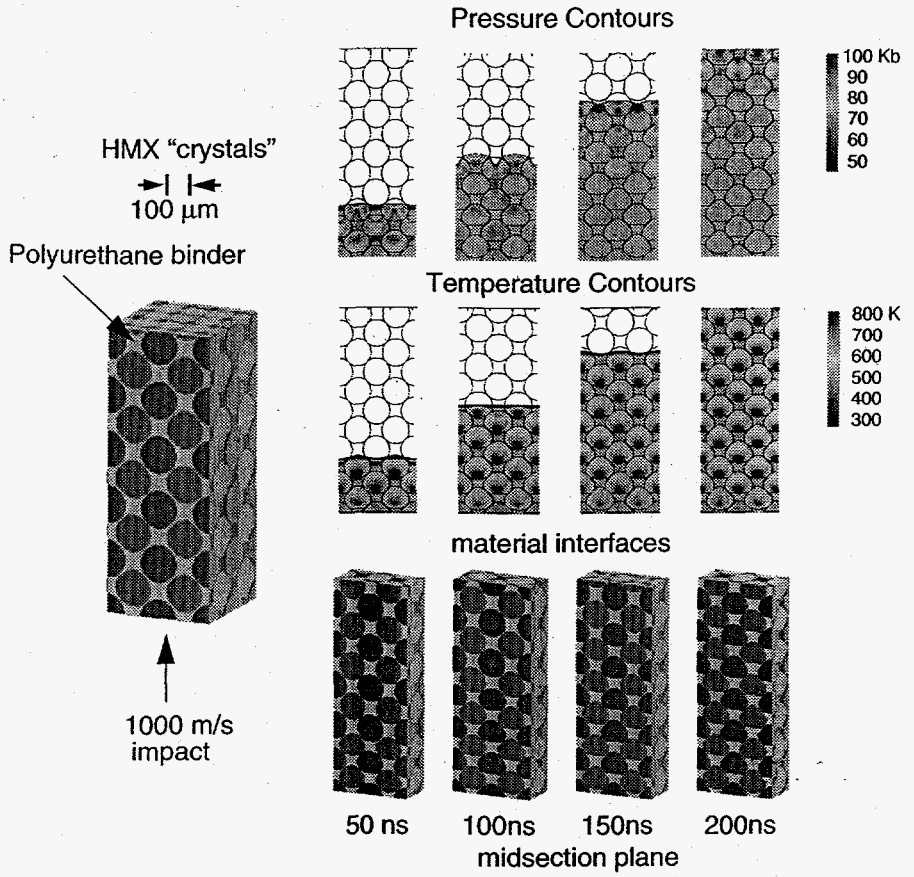

Fig. 9 Numerical simulation of impact on HMX-binder composite. Materials and contours of temperature and pressure are plotted at various time planes.

SUMMARY AND CONCLUSIONS

In this study, low-velocity impact on granular HMX has been experimentally and theoretically studied. New ideas in experiments and modeling are explored to investigate the nature of reactive compaction waves. Fundamental studies have focused on providing detailed wave information which is a critical assessment of modeling. Continuum mixture modeling has been successfully applied to replicate these measurements.

Mesoscale modeling has been applied toward a better understanding of the nature of dispersive waves which has been experimentally observed and modeled with continuum theory. Detailed shock physics analysis reveal that the shocked heterogeneous materials exhibit highly fluctuating stresses and the localization of energy due to plastic deformation include the effects of intercrystal interactions. The implication of this mesoscale modeling suggest that the classical views of shock waves (or even detonations) in heterogeneous materials are not simple jump states. Future work will extend experimental studies to explore the nature of the "noise" in the measurement data - perhaps there is important information which has overlooked in the past. Additional modeling will also investigate the effect of defects within crystals (such as bubbles or shear bands), and the blend of materials (such as metals) with improved material strength models.

ACKNOWLEDGEMENTS

The work performed at Sandia National Laboratories was supported by the U. S. Dept of Energy under contract no. DE-AC04-94AL85000. The work at LANL was supported by the United States Government Department of Energy. We thank Marlin Kipp for his assistance with the viscoelastic and crystal modeling.

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