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THE GROWING IMPACT OF GRAIN-SCALE MODELING

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ABSTRACT

Grain-scale modeling, a simulation technique that employs discrete microstructural features in order to understand “sub-grid’ phenomena, has been used in shock-physics primarily to characterize the mechanisms for hot-spot formation at voids and/or inclusions. More recently, these methods have been used on length scales of engineering interest. In this work, the unreacted equation of state (EOS) for porous hexanitrostilbene (HNS) and hexanitrohexaazaisowurtzitane (CL-20) are determined using simulated and measured microstructure grain-scale models. Calibrated Arrhenius reactive burn models are shown to be capable of predicting observed shock-to-detonation transition (SDT) behavior. Not only are measured threshold impact velocities obtained, but correct trends in pressure history for heterogeneous materials, and trends in sensitivity with pore size distribution are also reproduced. The capabilities of grain-scale methods are discussed, and a workflow is proposed for physics based performance predictions of energetic materials.

INTRODUCTION

The idea of a modeling regime that exists between scales has existed for quite some time, an example being the proposal of capturing larger turbulence length scales in fluids, i.e., the well-known large eddy simulation technique [1]. Mesoscale modeling, or the modeling of intermediate length or time scales, is employed whenever the phenomena of interest require more accurate representation, as is the case for large turbulence eddies in atmospheric models, or material grains and pores in energetic materials. In the field of energetic materials, the history of mesoscale techniques and analysis was outlined in 2007 by Baer [2]., and will be summarized briefly here.

Mesoscale modeling techniques can be categorized by three different approaches, 1) particle-based methods, 2) quasiparticle methods, and 3) direct numerical simulation. Particle based methods trace their roots to molecular dynamics (MD) scaling, and the need to perform larger and larger simulations. Computational resources limit the ability of researchers to model material feature interactions at larger scales. Generally, particle methods access larger time and length scales than MD either by using coarse-grained Lagrangian “particles”, or by using different methods to calculate particle dynamics. Quasiparticle methods use continuum conservation equations for shock physics, but employ particle dynamics to model mesoscale, or sub-grid effects. Direct numerical simulation is a method that owes its adoption to advances in computing power. In direct numerical simulation, traditional continuum methods are used with spatial resolution sufficient to discretely capture grain-scale structures. This method, aptly named here as grain-scale modeling, is the focus of this work.

To the authors' knowledge, Baer [3] was the first to report the study of energetic materials with continuum grain-scale models, in analyzing the shock response of HMX. Baer showed the rapidly fluctuating stress states due to the heterogeneous nature of the material, and the creation of hot spots from crystal interactions and deformation. Similar grain-scale simulations [4] were carried out on single pores, highlighting the importance of viscous heating and plastic work in localizing energy during pore collapse. Many others [5-10] have also used grain-scale modeling in various capacities, adding to the growing understanding of hot spot formation at the particle or pore scale, and the effects of microstructure on bulk mechanical and reactive properties.

The majority of grain-scale modeling work has focused on either the understanding of pore collapse heating mechanisms in creating hot-spots, or the simulation of many particles or pores in qualitative studies. These previous studies have led to many advances in continuum representations and understanding of heterogeneous reactive materials. This paper will discuss how grain-scale models have begun to be employed in a quantitative manner in their own right, on large domains capable of predicting the full history of energetic materials in dynamic environments, from impulse to steady state or failure.

Material Models

In continuum modeling and simulation, bulk or average material properties are used, because microstructural features are not discretely represented, and the first order or direct effects of microstructure are represented by average properties. For grain-scale models, where particles or porosity are included as part of the mesh, thus taking into account first order effects directly, every material is treated as fully dense. This poses a two-fold problem due to the lack of measured material properties for fully dense materials, and the feasibility of predicting these properties from theory. Not only are there less data for fully dense materials but also certain fully dense or crystalline materials are extremely difficult to obtain in quantities needed for traditional properties measurement techniques such as gas gun impacts for Hugoniot measurements. When data are not available for critical material models like the equation of state (EOS) or strength, other suitable means must be used to fill the gap.

For the materials under consideration in this work (HNS and CL-20), fully dense data were not available, and other methods were employed to obtain the required information. Mattson et al. pioneered an ab-initio method for determining the Hugoniot of molecular crystals and showed its applicability by first calculating the Hugoniot of pentaerythritol tetranitrate (PETN) [11]. This density functional theory molecular dynamics (DFT-MD) method was employed to obtain Hugoniot points in shock and particle velocity space (U_s-u_p) for both HNS and CL-20. These Hugoniot predictions were subsequently used to create a modified Gruneisen EOS.

Previously, a complete EOS was used for grain-scale calculations that used a temperature dependent specific heat [12]. While this method improves on a similar technique using constant specific heat, consistency was not achieved, i.e., the predicted states were path dependent. In this work, a method

equivalent to Menikoff [13] was used in calculating thermal states that are not path dependent, and is therefore consistent.

At this point, the effects of strength on grain-scale model predictions are not clear, but are being analyzed. The difficulty of parameterizing strength models for fully dense energetic materials is more difficult than the EOS, since strength effects are often secondary in a strong shock regime. However, strength properties [14] do affect the localization of energy in hot spot collapse, but the effects of this formation on macroscopic performance properties are still unknown. In this work, the strength properties of the material have been neglected, and only pure hydrodynamic response is considered.

Microstructure Representation

At the foundation of these grain-scale models is the accurate representation of the material microstructure. Two materials, HNS and CL-20, are chosen to describe two different methods of producing adequate microstructures for use in grain-scale simulations. The two methods used in this work are, 1) the simulation of measured microstructure, 2) direct use of the measured microstructure. The use of simulated microstructure has been discussed in detail in previous work [15]. It involves the statistical characterization of a measured microstructure, and reproduction of those statistics with a simpler porous structure, e.g., circular pores, that can be modified to represent other microstructures. This method can be used to determine dynamic response due to microstructural features such as pore size distribution, clustering, and porosity.

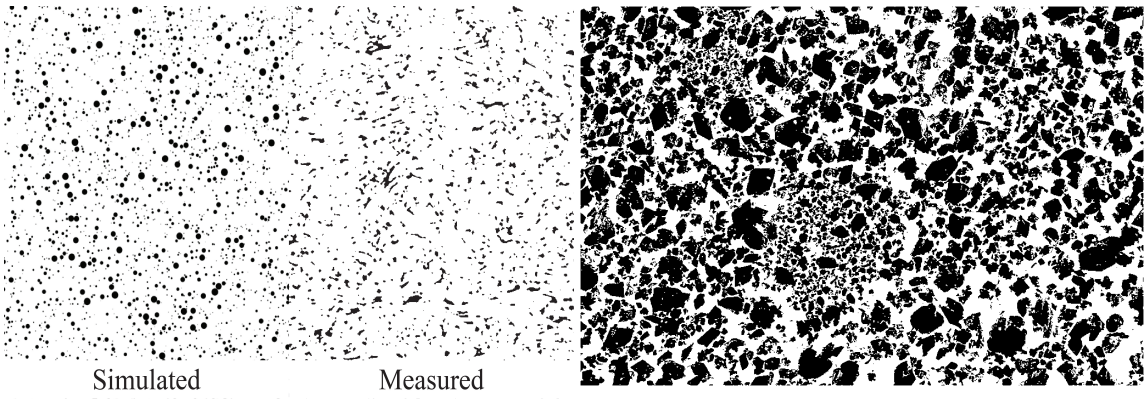

Figure 1: left) Simulated and measured HNS microstructures. right) Post-processed CL-20 microstructure.

Microstructure images can also be used directly in a simulation to model the response of the measured sample. In this method, a micrograph image is subjected to several post processing steps in order to isolate individual particles on the milled surface of a sample. The processed image is thresholded, resulting in a binary image, which is then used to discretize the outline of each particle into coordinates that are used to define the shapes in the computational mesh. This method shares many steps with those used for the statistical characterization, but departs by defining particle surfaces instead of characterizing statistics. Examples of microstructures from both methods are shown in Figure 1.

Verification of Inert Response

In order to accurately represent the transient reactive response and eventual steady state behavior of these energetic materials, the unreacted response must first be verified. Using the EOS described previously, along with several statistical reproductions of the measured HNS two-dimensional microstructure, plate impact simulations were performed at various flyer velocities using CTH [16], a multi-material, large deformation, strong shock wave, solid mechanics code developed at Sandia National Laboratories. Although three-dimensional representations are technologically achievable [8], the computational cost of performing them on large length scales is prohibitive. Performing simulations on a two-dimensional mesh results in a loss of degrees of freedom, but recent MD results on pore collapse indicate that this difference can be small [17]. The responses for the different representations at each velocity were averaged, compiled, and compared with the fully dense data, historical published data, continuum compaction methods, and theoretical methods (see Figure 2). The historical and predicted values align very favorably, and elicit sufficient confidence that this method captures the essential mechanical effects of the microstructure on the average shock response.

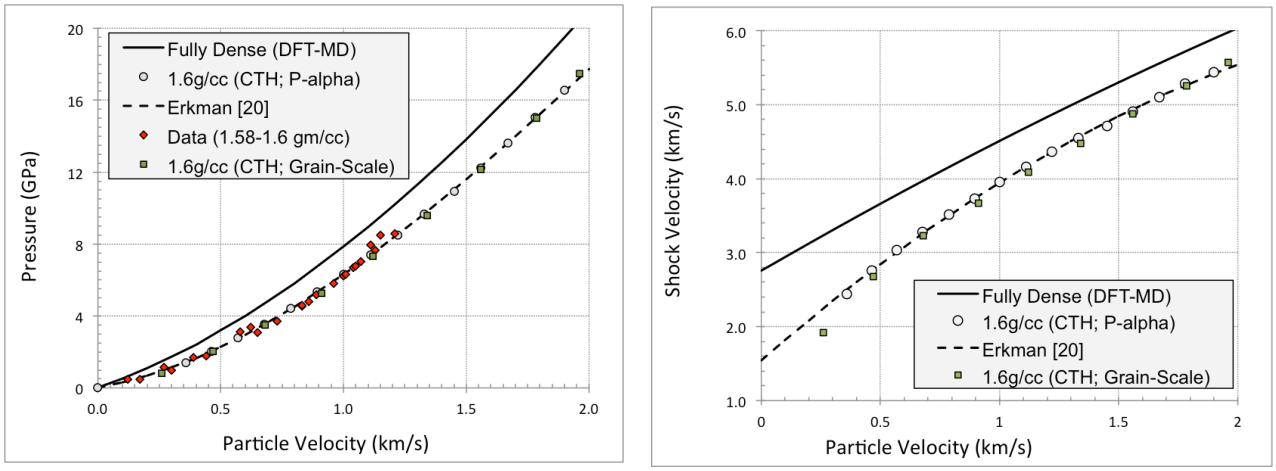

Figure 2: Fully dense and porous left) P-u_p and right) U_s-u_p HNS Hugoniot curves from historical data and other methods.

For materials that have undergone limited experimental performance characterization, there are little to no data available for comparison with and verification of predicted computational results. This is where grain-scale models will likely see greater acceptance and use. As the results continue to be verified against common materials and historical performance data, the application of the method to less common materials can be viewed as indicative of the expected behavior. As an example of such a process, a relatively recently formulated explosive[18], CL-20, was investigated to determine the unreacted Hugoniot.

Samples of CL-20 were pressed to 50% TMD, into a cylindrical cup machined into polymethyl methacrylate (PMMA), and milled with a focused argon ion beam to provide an imaging surface for scanning electron microscopy (SEM). The resulting images underwent the post-processing procedure

described previously, and were imported into CTH for impact analysis. For this study, the CL-20 micrograph was divided into 5 sections of equal width, and impact analysis was performed on each of these sections at several flyer velocities. The resultant Hugoniot curves in both U_s-u_p and pressure – particle velocity (P-u_p) space are shown in Figure 3 along with those predicted by the P-alpha continuum compaction model [19], and the method of Erkman [20]. There is a clear difference in the results of the three methods, with the grain-scale predictions falling in-between. Due to the lack of experimental data on CL20, the discrepancies between these methods cannot be explained fully, but several known assumptions can be pointed out as possible sources of error. In the method of Erkman, the use of constant Gruneisen coefficient is valid only at lower pressures. For the grain-scale results, neglecting degrees of freedom from the actual three-dimensional microstructure could affect the results at this higher porosity level. Temperature dependency is not a part of the p-alpha model, which could effect the results at higher porosities where temperature is expected to be higher upon compaction. It is expected that at lower porosities these discrepancies will be less, and the differences at high porosities highlights the need for experimental verification. This is a problem that is still being looked at, and its conclusion will be published in future work.

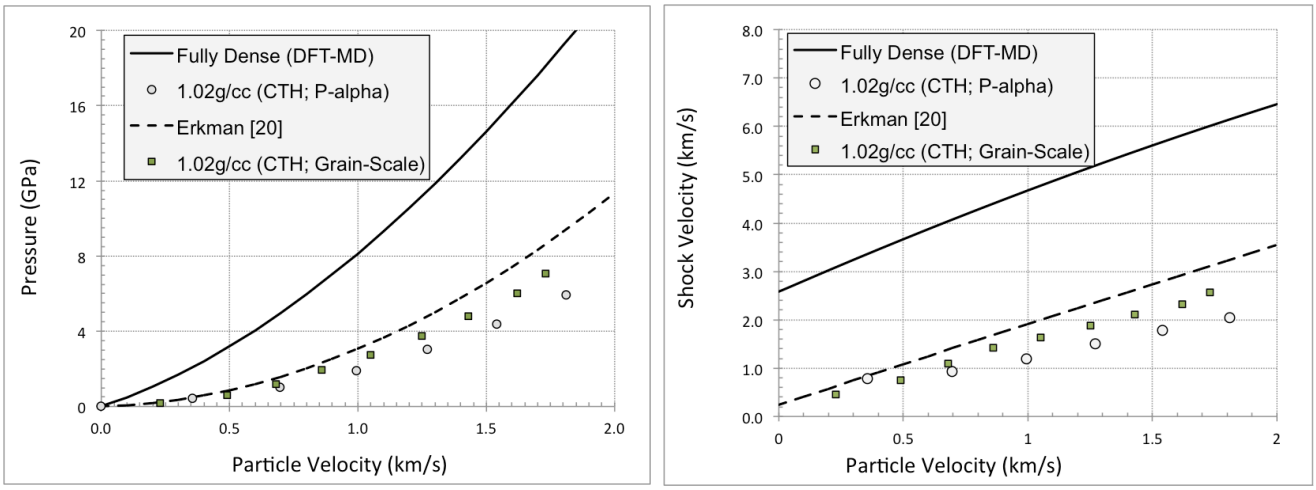

Figure 3: Fully dense and porous left) P-u_p and right) U_s-u_p CL-20 Hugoniots from different methods.

Calibration of Reactive Burn

With the inert shock response, attention is turned to the reactive burn model, which is the final piece of the grain-scale model, and will allow the prediction of transient phenomena such as SDT. Most reactive burn models in continuum codes are developed to reproduce the effects of microstructure, and are calibrated using continuum measurements. Because microstructure is explicitly represented in the grain-scale model, a basic reactive burn model is required, such as one based on Arrhenius kinetics. The ultimate goal is a predictive method that is generally applicable across materials and dynamic environments, and although currently the basic reactive model is calibrated against similar continuum measurements, the model parameters could in theory be determined by measurements of basic reactive properties of the material.

For calibrating the Arrhenius kinetics model parameters, impact threshold experiments were carried out to determine the threshold flyer velocity of HNS for several pressure pulse widths. The model parameters were then calibrated against a single point in the experimentally determined threshold space. Initial values for the parameters were chosen based on published values [21, 22], and then modified systematically resulting in successful detonation just at the experimentally measured threshold velocity. In Figure 4, snapshots of the simulation domain for velocities just under and above threshold show the model predicting failure, and successful detonation, respectively.

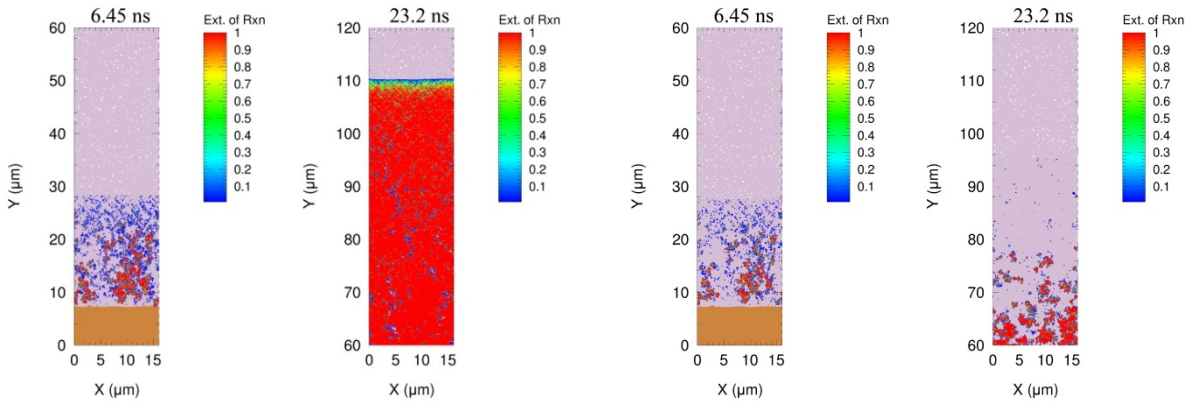

Figure 4: HNS grain scale simulation images showing left) successful SDT and right) unsuccessful SDT.

One benefit of grain-scale models, is the ability to check several aspects of a model for physical feasibility. For example, in a grain-scale model, all average state variables should be consistent with continuum measurements, as well as properties that are difficult to measure, like reaction zone thickness. The steady state inert response was already discussed previously, and the transient reactive response is of equal interest. In Figure 5, pressure traces for failed and successful SDT are shown for the HNS grain-scale model that are typical of heterogeneous reactive materials [23], where reactions progressively build up behind the initial shock wave until a release wave either quenches reactions or steady state detonation is achieved. Similar pressure histories were obtained for scaled microstructures, and correct sensitivity trends were observed, with larger and smaller pore size distributions having longer and shorter run distances, respectively. Pressure histories represent a promising source of data that can be used to further verify, or calibrate the inert and reactive aspects of the grain-scale model. A similarly difficult property to measure in energetic materials is the reaction zone thickness. In recent studies of deposited explosives for critical thickness measurements, Knepper et al. [24] measured reaction zone thicknesses for HNAB on the order of ~1 micron, and the reaction zone thickness of 1 micron from the grain-scale model support their conclusions.

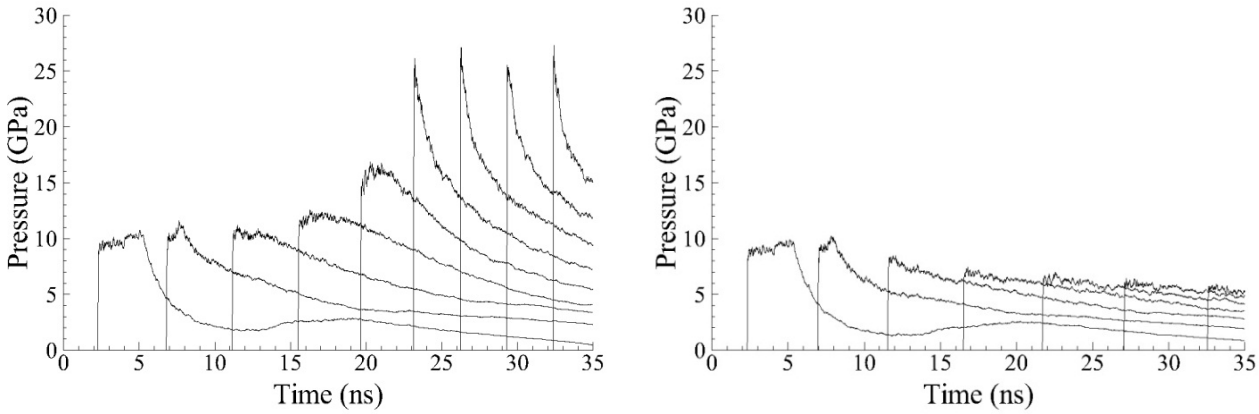

Figure 5: HNS grain-scale pressure histories from left) unsuccessful SDT and right) successful SDT.

Growing Impact

The previous sections have described a grain-scale method that has been used to ultimately match and predict the threshold behavior of HNS. In the field of energetic materials, grain-scale models on this scale are a new concept made possible by the advances in computing power and parallel processing. The results presented above represent a small portion of what is currently feasible. With just the methods discussed, the unreacted Hugoniot of any material whose microstructure can be obtained, and crystal structure is known can be obtained. With adequate experimental data, a burn model can be calibrated for the fully dense material, and applied to any microstructure of interest. It is also feasible, but not practical at this point, to do this on a relevant scale that is currently only accessible to continuum models. The interpretation of these models makes physical sense as well, since the observed effects can be tied to variables that are measured and intuitive, and not model parameters that don't always have a clear meaning.

With advances in other areas like process modeling and continuum reactive burn modeling, a workflow could be developed that, with proper validation, results in a fully predictive methodology. In such a workflow, process modeling tools produce as output a predicted microstructure based on defined process parameters. A grain-scale method uses the predicted microstructure to assess the performance of the material. A suitable continuum model, informed by the grain-scale results, then predicts the performance of devices, and is used to optimize design. A methodology such as this would help to reduce costs in performing experiments, and is motivated by emphasizing the understanding of the causes and correlations instead of the observed performance.

Conclusion

The shock response of HNS and CL-20 was analyzed using the grain-scale modeling technique. With suitable fully dense data or theoretical values for the Hugoniot, this was shown to be an accurate way of obtaining the unreacted porous Hugoniot. At high porosities, the grain-scale technique, as well as other methods, did not align as closely as for higher density samples, and this discrepancy is a current research endeavor. The reactive response of HNS was also investigated, and with available validation data, was seen to accurately match observed threshold data, as well as expected trends with changes in pore size distribution. A workflow was proposed that would allow for the prediction of the reactive

behavior of most materials, with inputs coming from basic measurements and calculations, and the output being generally applicable continuum models.

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