ARMY RESEARCH LABORATORY

Physics-based Multi-scale Modeling of Shear Initiated Reactions in Energetic and Reactive Materials

by John K. Brennan, Müge Fermen-Coker, and Linhbao Tran

ARL-MR-0744 **April 2010**

NOTICES

Disclaimers

The findings in this report are not to be construed as an official Department of the Army position unless so designated by other authorized documents.

Citation of manufacturer's or trade names does not constitute an official endorsement or approval of the use thereof.

Destroy this report when it is no longer needed. Do not return it to the originator.

Army Research Laboratory
Aberdeen Proving Ground, MD 21005

ARL-MR-0744 **April 2010**

Physics-based Multi-scale Modeling of Shear Initiated Reactions in Energetic and Reactive Materials

John K. Brennan and Müge Fermen-Coker
Weapons and Materials Research Directorate, ARL

and

Linhbao Tran
Shock Physics Insight
807 Park Road, Bellevue, WA 98004

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*	2. REPORT TYPE	3. DATES COVERED (From - To)
April 2010	DRI	

4. TITLE AND SUBTITLE
Physics-based Multi-scale Modeling of Shear Initiated Reactions in Energetic and Reactive Materials

5a. CONTRACT NUMBER

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
John K. Brennan, Müge Fermen-Coker, and Linhbao Tran

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
U.S. Army Research Laboratory
ATTN: RDRL-WML-B
Aberdeen Proving Ground, MD 21005

8. PERFORMING ORGANIZATION REPORT NUMBER
ARL-MR-0744

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

10. SPONSOR/MONITOR'S ACRONYM(S)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT
We developed a multi-scale approach for simulating shear initiated reactions that spans from the molecular scale to the mesoscale (length scale of material heterogeneities) to the full continuum scale (length scale of the weapon system) and implemented it into the Combined Hydro and Radiation Transport Diffusion (CTH) hydrocode developed by Sandia National Laboratory. We demonstrated that the new approach allows predictions not previously possible for energetic materials when subjected to loads that result in shear localizations. This computational tool provides a novel modeling capability that opens previously unavailable avenues by bridging the gaps between multiple scales to enable improved predictions towards designing armor and anti-armor devices. It also enables the development of concepts for enhanced survivability and lethality, primarily in the areas of insensitive munitions, reactive armor, and the development of novel concepts and designs using reactive materials.

15. SUBJECT TERMS
shear initiated reactions, energetic materials, reactive materials, multi-scale modeling, mesoscale

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			John K. Brennan
Unclassified	Unclassified	Unclassified	UU	20	19b. TELEPHONE NUMBER (*Include area code*) (410) 306-0678

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. Z39.18

Contents

List of Figures

1. Objective

We propose to develop a multi-scale framework to simulate and predict shear initiated reactions in energetic and reactive materials; implement this technology into the Combined Hydro and Radiation Transport Diffusion (CTH) hydrocode; verify the algorithm; and conduct preliminary studies to assess this novel framework. The final product is an improved predictive capability implemented into the CTH hydrocode developed by Sandia National Laboratories to allow for the prediction and/or simulation of high explosive (HE) and reactive material (RM) reactions when subjected to loads that result in shear localizations.

2. Approach

A numerical framework for nucleating and propagating shear localizations already exists in CTH, where a physical criterion for the initiation of shear bands has been implemented. We propose to use this framework to track shear localizations and apply the outcome of our multi-scale approach at these locations to simulate the shear initiated reactions of HEs and RMs. A schematic of the approach developed in this work is shown in figure 1. It consists of four major components: (a) mesoscale modeling, (b) converting mesoscale output to input for the reactive burn model, (c) linking the reactive burn model output to continuum level simulation of shear initiated reactions, and (d) assessing the overall approach by implementing it into the continuum mechanics code CTH.

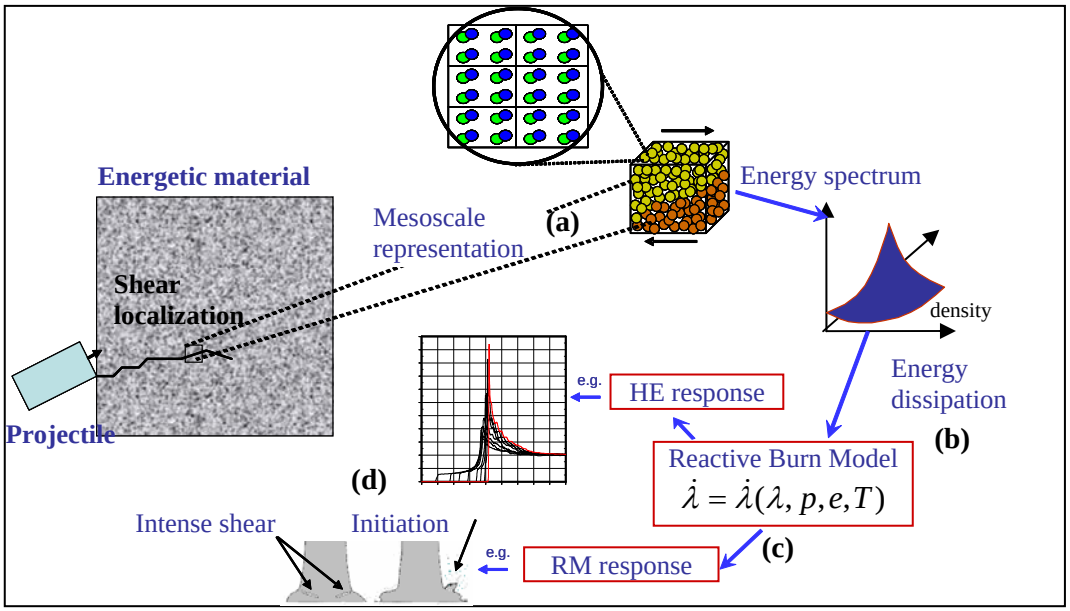

Figure 1. Schematic of the multi-scale shear initiation model.

As depicted in figure 1(a), each mesoparticle is intended to represent several atomic-level unit cells of the crystalline material. The dissipative particle dynamics method (*1, 2*) provides the mesoscale input to the reactive burn model. Thermodynamic properties (pressure, density, temperature, and energy) from the mesoscale simulations are generated at various steady-state shear rates, providing look-up data tables for use in the CTH modeling (cf. figure 1(b)), such that for each strain observed in the computational domain where a shear localization has developed, the state variables and the energy spectrum obtained by the mesoscale model are known at the continuum level. With this approach, the assumptions made in the continuum mechanics code with respect to shear localizations are circumvented. Moreover, by incorporating mesoscale level calculations into the continuum mechanics code, more accurate estimates for the thermodynamic state within the localized regions, and consequently more accurate input to the reactive burn model, are obtained.

A sub-grid model is developed to account for energy dissipation and link the mesoscale temperature estimates to the continuum level in a relatively cell-size independent manner by calculating a volumetric averaged solution of the extent-of-reaction at the sub-grid level to represent the continuum extent-of-reaction (cf. figure 1(c)). This extent-of-reaction is used in the equation-of-state (EOS) to determine the amount of energy release.

The sub-grid calculations are performed for cells that are identified to contain shear bands at the continuum level. A numerical framework for nucleating and propagating shear band localizations already exists in CTH (*3–5*). When a set of nucleation criteria is satisfied, a shear band is formed by introducing Lagrangian particles and they conform to local planes of maximum shear as they propagate in three-dimensional space, until a set of growth criteria is no longer satisfied along the points defining its boundary. This framework is used to track shear localizations, and our multi-scale approach is applied at these locations to simulate the shear initiation of energetic and reactive materials (cf. figure 1(d)). The approach is implemented into CTH and simulations are conducted for HMX to assess the algorithm. Details of the approach are further discussed in the following sections.

2.1 Mesoscale Methods

The energy-conserving version of the Dissipative Particle Dynamics (DPD) method is a particle-based mesoscale method that conserves both momentum and energy while allowing the mesoparticles to exchange both viscous and thermal energy (*1, 2*). Further details of the DPD approach can be found in the original papers (*1*, *2*) and the report for our Year-1 effort (*6*).

As in the Year-1 effort, steady planar shear flow was induced by means of the Lees-Edwards boundary conditions (*7*), in which the simulation box and its images centered at $(x, y) = (\pm L, 0)$, $(\pm 2L, 0)$, ..., are taken to be stationary, while boxes in the layer above, $(x, y) = (0, L)$, $(\pm L, L)$, $(\pm 2L, L)$, ..., are moving at a speed ϖL in the positive x direction, where ϖ is the shear rate.

Boxes in the layer below, $(x, y) = (0, -L), (\pm L, -L), (\pm 2L, -L), \ldots$, move at a speed ϖL in the negative x direction. A system under such conditions is subjected to a uniform steady shear in the xy plane.

2.2 Mesoscale Models

Mesoscale models were generated for both HMX and a nickel (Ni)/aluminum (Al) system. For the HMX case, one mesoparticle was chosen to represent a single HMX molecule (figure 2), where mesoparticles interact through a standard pairwise Morse potential (*8*). Potential parameters can be determined by analytical solution of the Morse potential expression, where the cohesive energy, density, and bulk modulus at zero temperature and pressure are inputted conditions. The inputted cohesive energy, density, and bulk modulus can be taken from either experimental or higher resolution simulation data. Details for determining the Morse parameters can be found elsewhere (*9*). For the HMX mesoscale model, good agreement was found for both the EOS properties (figure 3) and the elastic coefficients.

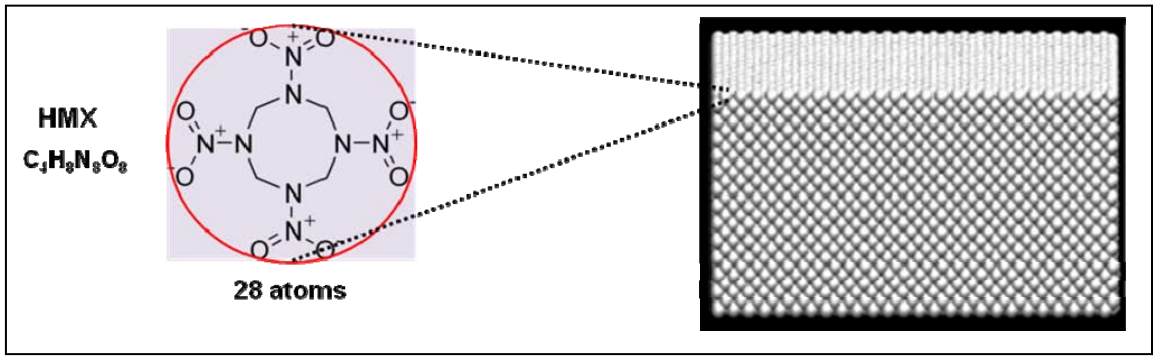

Figure 2. Mesoscale model representation of HMX.

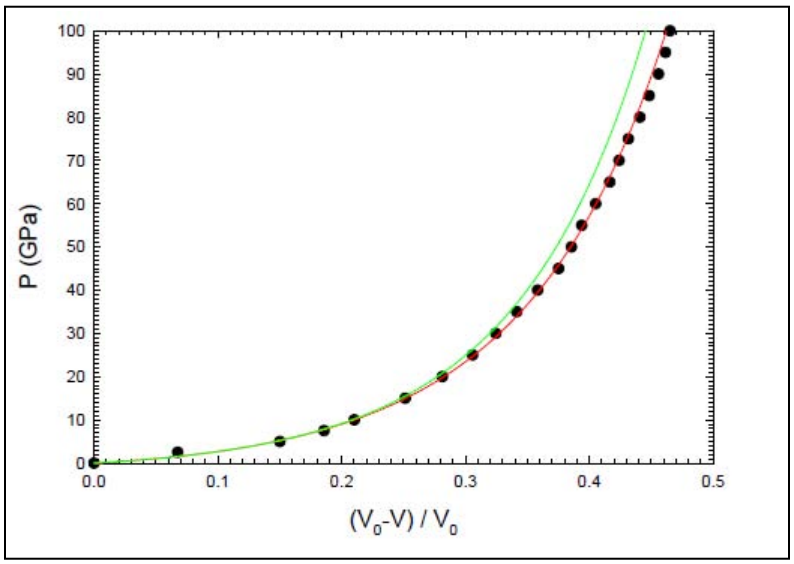

Figure 3. Pressure-volume isotherms (0 K) for an atomistic model (black), a theoretical fit (red), and our mesoscale model (green) of HMX.

For the Ni/Al system, one mesoparticle was chosen to represent four face-centered-cubic unit cells, or 16 atoms, where mesoparticles interact through a Sutton-Chen potential (*10*). The Sutton-Chen potential is the simplest of the embedded-atom type potentials, where a pairwise repulsive interaction is combined with a many-body cohesive term. The simple analytical form of the potential has contributed to its popularity for simulating metals and alloys. Analogous to the determination of the Morse potential parameters for HMX, the Sutton-Chen potential parameters can be determined through analytical solution of the potential, where the cohesive energy, density, and bulk modulus at zero temperature and pressure are inputted conditions, and again these property values can be taken from either experimental or simulation data. Details of the Sutton-Chen potential and the approach for fitting the material parameters can be found elsewhere (*10*).

The melting behavior of these models must be accurately captured since alloying of the Ni/Al system is diffusion driven. Therefore, further fitting of the model parameters to match the experimentally determined melting temperature was performed. For both the Ni and Al models, reasonable agreement was found for the EOS properties, as shown in figure 4.

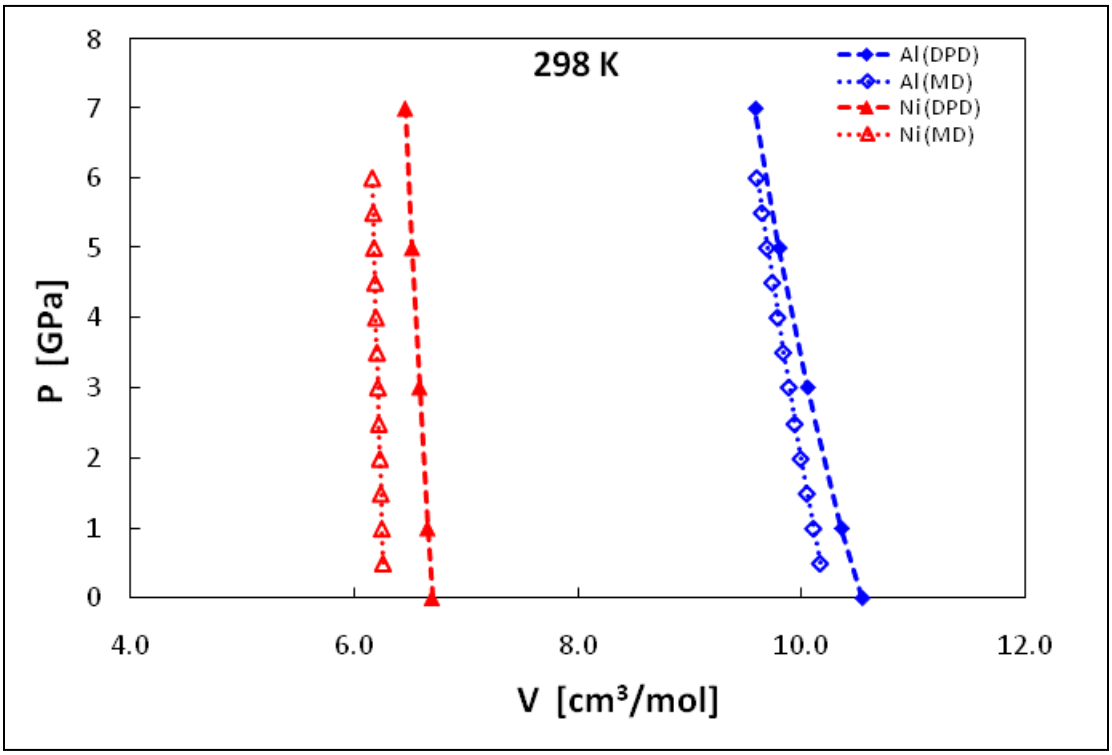

Figure 4. EOS property comparison between atomistic (MD) and mesoscale (DPD) models for Ni and Al.

2.3 Sub-Grid Models

A sub-grid model is developed to account for the dissipation of energy and allow for delayed reactions. Initiation phenomena in heterogeneous energetic materials can occur when the material is subjected to impulses such as shock waves. Under sufficiently strong shock

conditions, these shock waves can evolve into self-sustaining detonation waves. This process, shock-to-detonation transition (SDT), is fairly well understood at the phenomenological level and is based on the theory of hot spot formation (such as void collapse, visco-plastic heating, shear band, frictional heating, etc.). However, for conditions such that the SDT process does not occur, initiation of the HE leading to explosion can take place if the material is sufficiently confined. Such analysis was shown by Frey (*9*), whose study indicated that both pressure and shear rate were important parameters in controlling runaway explosion. In another study, a shear banding mechanism was shown to provide a large ignited surface area, which is believed to be necessary to explain shock initiation (*10*). The long delay between the time at impact and explosion can typically be hundreds of milliseconds, whereas for the SDT process it is on the order of microseconds. Such dominant physical features suggest energy competing processes at the micro-scale level. Therefore, at hot spot locations, e.g., shear surfaces, the possibility of initiation hinges on a balance (or lack thereof) between energy producing mechanisms (visco-plastic work, shear localization, chemical reaction, etc.) and the rate at which the energy is transported away from the zone. Energy generated from shear localization within the narrow region of the shear band width, which is on the order of micrometers, can be much higher than the bulk energy in the typical cell size of continuum simulations, which is on the order of millimeters. The large temperature gradient within a continuum cell supports the need to account for thermal diffusion. Figure 5 illustrates temperature profiles, including energy release due to reaction, as time progresses. This concept forms the basis of our sub-grid model.

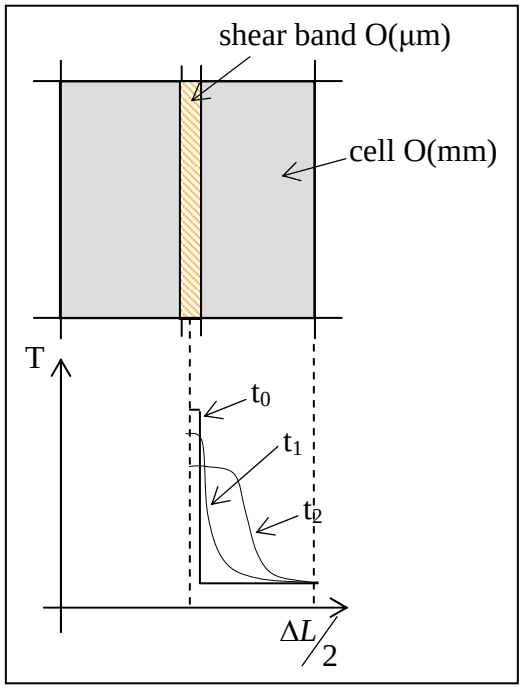

Figure 5. Sheared cell and its temperature profile.

Within each sheared continuum cell, we assume that the shear band surface is located at the mid-plane (figure 5). Such a geometric assumption leads to a simple and tractable solution at the sub-grid level. Each sheared cell is subdivided into intervals, and time-dependent equations of temperature and species are described as follows:

$$\rho C \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial^2 x} + \Delta H \dot{m}''' \tag{1}$$

$$\rho \frac{\partial \lambda_{sg}}{\partial t} = \rho D \frac{\partial^2 \lambda_{sg}}{\partial^2 x} - \dot{m}''', \tag{2}$$

where λ_{sg} is the extent of reaction at the sub-grid level, ΔH is the heat of reaction, D is the diffusion coefficient, k is the thermal conductivity, and the volumetric mass production rate, $\dot{m}'''$, is governed by an Arrhenius-type reaction:

$$\dot{m}''' = A\rho\left(1-\lambda_{sg}\right)\exp\left(-T_a \big/ T\right), \tag{3}$$

where A is the frequency and the activation temperature is T_a. The species diffusion term is assumed negligible in the current study. Solutions for the previous equations are integrated using an implicit method with the initial conditions:

$$\begin{aligned} T(x,0) &= T_{meso}, \qquad 0 \le x \le w_{sb} \\ T(x,0) &= T_{cell}, \qquad w_{sb} < x \le \Delta L \big/ 2 \end{aligned}, \tag{4}$$

where the shear band temperature, T_{meso}, is obtained from mesoscale simulations of a shear banding process, T_{cell} is the continuum cell temperature, and w_{sb} is the shear band width. Boundary conditions are given as

$$\left.\frac{\partial T}{\partial x}\right|_{x=0} = 0 \text{ and } T\left(x = \Delta L \big/ 2\right) = T_{cell}. \tag{5}$$

Within each sheared cell, a volumetric averaged solution of the extent-of-reaction at the sub-grid level is transferred to the continuum level as the continuum extent-of-reaction. This extent-of-reaction is used in the EOS to determine the amount of energy release. Such an approach ensures consistency in our internal energy calculation.

3. Results

Hydrostatic simulation was carried out with HMX mesoscale model to probe the range of validity of our model as well as to verify model parameters. Figure 6 shows curve fittings of the mesoscale data using Hugoniot relations. Values of the sound speed, the Us-Up slope, the bulk modulus, and its derivative are within ranges reported in literature.

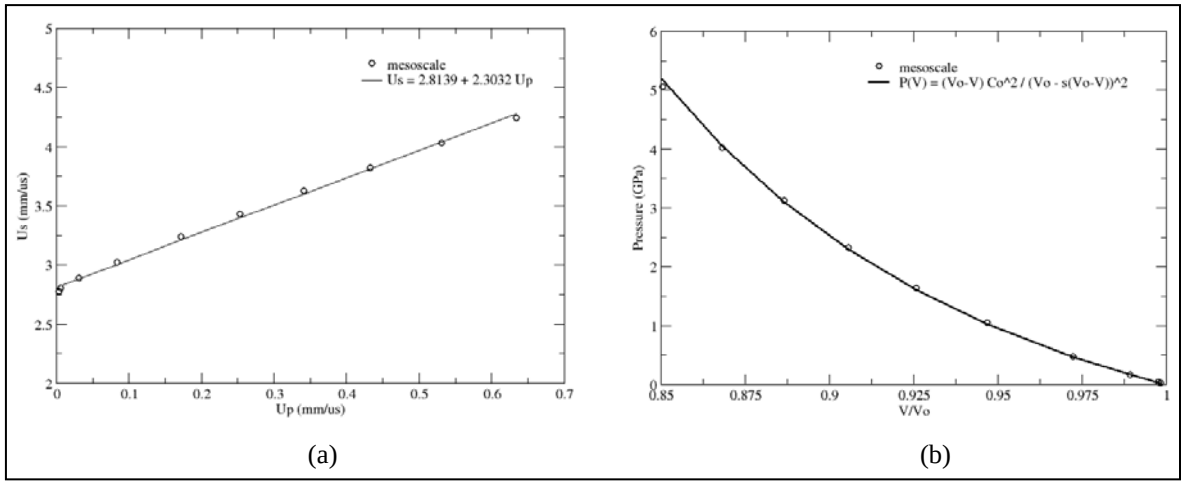

Figure 6. (a) Hugoniot fit of the Us-Up relation and (b) Hugoniot fit of the P-V relation.

Analogous to our Year-1 effort, the mesoscale look-up table was generated for a range of densities and shear rates, where the pressure and temperature were calculated. As an example of a particular data point, temporal profiles of the various energy contributions for the HMX mesoscale model under shear is shown in figure 7(a). A configuration snapshot of the HMX material just prior to the release of elastic strain energy is given in figure 7(b), where evidence of shear localization is present.

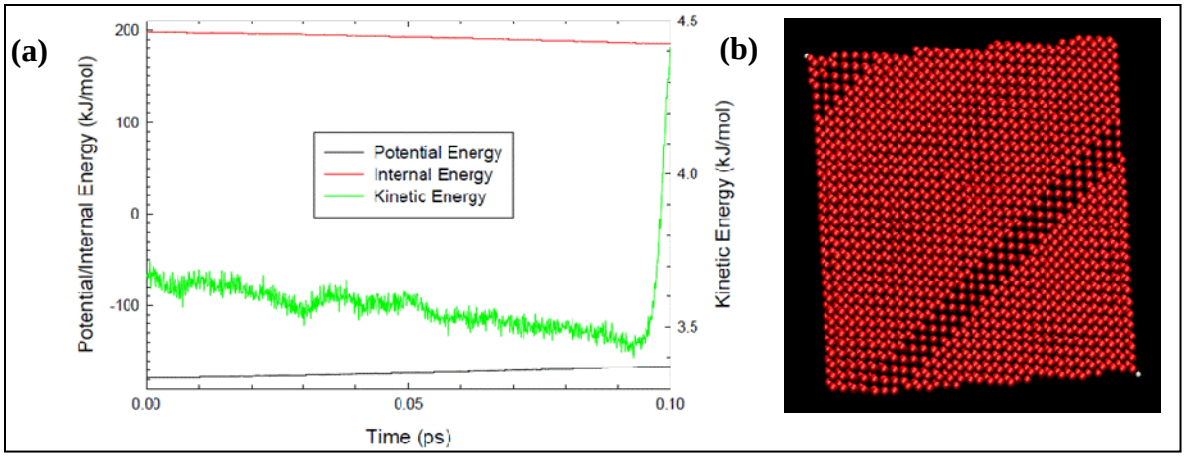

Figure 7. (a) Temporal variations of various energy contributions after the system has been sheared in the mesoscale simulation and (b) a configuration snapshot of the material sample just prior to the release of elastic strain energy.

In the continuum level simulations, a block of material is subjected to pure shear by imposing a velocity on the lower half of the block to verify the newly implemented approach in CTH, as shown in figure 8(a). Example output from a CTH simulation in which the sub-grid model has been implemented is shown in figures 8 (b) and (c).

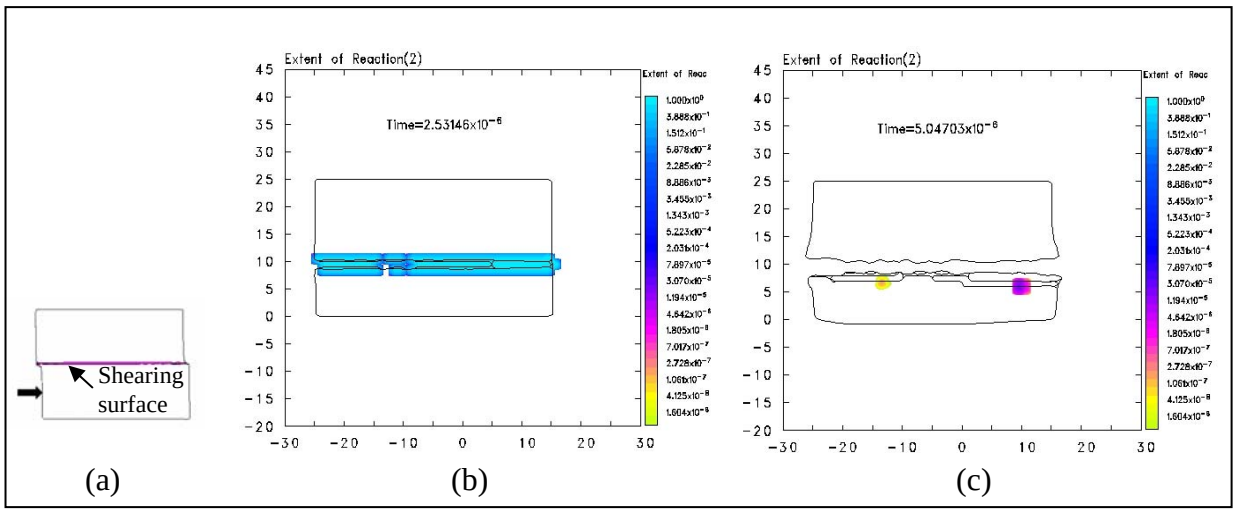

Figure 8. Example problem to verify the new multi-scale approach: (a) a sliding half of a block of material, (b) the extent-of-reaction at 2.5 μs, and (c) the extent-of-reaction at 5 μs.

4. Conclusions

We developed a multi-scale approach for simulating shear initiated reactions that spans from the molecular scale to the mesoscale (length scale of material heterogeneities) to the full continuum scale (length scale of the weapon system) and implemented it into the CTH hydrocode developed by Sandia National Laboratory. We demonstrated that the new approach allows predictions not previously possible for energetic materials when subjected to loads that result in shear localizations. This computational tool provides a novel modeling capability that opens previously unavailable avenues by bridging the gaps between multiple scales to enable improved predictions toward designing armor and anti-armor devices. It also enables the development of concepts for enhanced survivability and lethality, primarily in the areas of insensitive munitions, reactive armor, and the development of novel concepts and designs using RMs.

Accomplishments in this year's work included the development and implementation of a simple and efficient approach to generating various mesoscale models for HE and RM systems. We found the models to adequately reproduce the EOS properties for HMX and a Ni/Al system. Compared to Year 1, the approaches used to develop this year's models were more efficient while still maintaining physically relevant parameters. In this year's effort, we also refined our

sub-grid model from the previous year. Furthermore, we refined the multi-scale links between the mesoscale and the sub-grid models as well as the sub-grid and continuum descriptions. Overall, these improvements have led to more physically pertinent continuum level predictions.

5. References

1. Bonet Avalos, J.; Mackie, A. D. *J. Chem. Phys.* **1999**, *111*, 5267–5276.
2. Mackie, A. D.; Bonet Avalos, J.; Navas, V. *Phys. Chem. Chem. Phys.* **1999**, *1*, 2039.
3. Silling, S. A. *CTH Reference Manual: Three dimensional shear band model*. Unpublished.
4. Fermen-Coker, M. *Numerical Simulation of Adiabatic Shear Bands in Ti-6Al-4V Alloy Due to Fragment Impact*; ARL-RP-91; U.S. Army Research Laboratory: Aberdeen Proving Ground, MD, March 2005.
5. Fermen-Coker, M. *Implementation of Schoenfeld-Wright Failure Criterion Into a 3D Adiabatic Shear Band Model in CTH*; ARL-TR 3284; U.S. Army Research Laboratory: Aberdeen Proving Ground, MD, September 2004.
6. Brennan, J. K.; Tran, L.; Fermen-Coker, M. *Physics Based Multi-Scale Modeling of Shear Initiated Reactions in Energetic and Reactive Materials*; U.S. Army Research Laboratory: ARL-TR 4753; Aberdeen Proving Ground, MD, March 2009.
7. Lees, A.W.; Edwards, S. F. *J. Phys. C: Solid State Phys.* **1972**, *5*, 1921.
8. Allen, M. P.; Tildesley, D. J. *Computer Simulation of Liquids*; Oxford University Press: New York, 1989.
9. Girifalco, L. A.; V. G.; Weizer, V. *Phys. Rev.* **1959**, *114*, 687.
10. Sutton, A. P.; Chen, J.; V. *Phil. Mag. Lett.* **1989**, *61*, 139.
11. Frey, R. *Seventh Symposium (International) on Detonation*, Naval Surface Weapons Center, NSWC MP 82-334. White Oak, MD, 1981.
12. Howe, P.; Frey, R.; Taylor, B.; Boyle, V. *Sixth Symposium (International) on Detonation*, Office of Naval Research, ACR-211, Arlington, VA, 1970.

6. Transitions

Transitioning this modeling capability to the FY11 program goals of the Institute for Multi-Scale Reactive Modeling of Insensitive Munitions (MSRM) is planned.

List of Symbols, Abbreviations, and Acronyms

Al	aluminum
CTH	Combined Hydro and Radiation Transport Diffusion
DPD	Dissipative Particle Dynamics
EOS	equation-of-state properties
HE	high explosive
MD	molecular dynamics
MSRM	Multi-Scale Reactive Modeling of Insensitive Munitions
Ni	nickel
RM	reactive material
SDT	shock-to-detonation transition

No of. Copies	Organization
1 (PDF ONLY)	ADMNSTR DEFNS TECHL INFO CTR ATTN DTIC OCP 8725 JOHN J KINGMAN RD STE 0944 FT BELVOIR VA 22060-6218
1 HC	US ARMY RSRCH LAB ATTN RDRL CIM G T LANDFRIED BLDG 4600 ABERDEEN PROVING GROUND MD 21005-5066
9 HCs	US ARMY RSRCH LAB ATTN RDRL WMP PATRICK BAKER (1HC) ATTN RDRL WML H M FERMEN-COKER (5 HCS) CALVIN CANDLAND (1HC) ATTN RDRL WML B J BRENNAN (1 HC) JEFFREY MORRIS (1HC) ABERDEEN PROVING GROUND MD 21005-5066
3 HCS	US ARMY RSRCH LAB ATTN IMNE ALC HRR MAIL & RECORDS MGMT ATTN RDRL CIM L TECHL LIB ATTN RDRL CIM P TECHL PUB ADELPHI MD 20783-1197
2 HCS	US ARMY RDECOM-ARDEC ATTN RDAR MEE W BLDG 3028 KENNETH LEE ATTN AMSRD AAR AEE W BLDG 3022 ERNEST BAKER PICATINNY ARSENAL NJ 07806
2 HCS	SANDIA NATIONAL LABORATORIES ATTN MS 1322 STEWART SILLING ATTN MS 0836 DAVID CRAWFORD PO BOX 5800 ALBUQUERQUE NM 87185-1322

TOTAL:18 (1 ELEC, 17 HCS)

INTENTIONALLY LEFT BLANK.