Investigating short-pulse shock initiation in HMX-based explosives with reactive meso-scale simulations

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Abstract. We performed reactive meso-scale simulations of short-pulse experiments to study the influence of flyer velocity and pore structure on shock initiation of LX-10 (95wt% HMX, 5wt% Viton A). Our calculations show that the reaction evolution fit a power law relationship in time and increases with increasing porosity, decreasing pore size, and increasing flyer velocity. While heterogeneous shock initiation modes, dependent on hot spot mechanisms, are predicted at lower flyer velocities, mixed heterogeneous-homogeneous shock initiation modes, less dependent on hot spots, are predicted at higher velocities. These studies are important because they enable the development of predictive shock initiation models that incorporate complex microstructure and can be used to optimize performance-safety characteristics of explosives.

1. Introduction
Shock initiation in heterogeneous explosives is primarily governed by localized hot spot formation, arising from dynamic pore collapse, and subsequent chemical kinetic mechanisms [1,2]. Reactive meso-scale simulations enable us to study the influence of heterogeneous pore structures and chemical-thermal-mechanical properties on shock initiation response of explosives. While previous computational studies have focused on inert meso-scale simulations probing the behavior of single and multiple pore collapse [3-7], only recently have studies incorporated chemistry using multi-physics codes [8-12]. Short-pulse shock initiation experiments [13] provide an excellent platform for benchmarking meso-scale modeling approaches because the entire sample domain can be simulated while resolving critical microstructural features. In the present study, we perform reactive meso-scale simulations of short-pulse shock experiments to study the influence of flyer velocity, pore size, and overall porosity on the initiation and reaction evolution in LX-10.

2. Model Description
2.1. Material Models
We performed 2D plane-strain reactive meso-scale simulations using the arbitrary-Lagrangian-Eulerian hydrocode, ALE3D [14]. HMX unreacted equation-of-state (EoS) with constant specific heat [15,16] and product EoS [17] model parameters are shown in tables 1 and 2, respectively. Due to a singularity in the Mie-Gruneisen EoS for our range of relative volumes, we employed a
seven term polynomial for the unreacted EoS, \( P = A_0 + A_1\mu + A_2\mu^2 + A_3\mu^3 + (B_0 + B_1\mu + B_2\mu^2)e \), with parameters shown in table 1 (\( B_1 = B_2 = 0 \)). The Gruneisen gamma was modified from [15] to be consistent with the specific heat and bulk modulus values. We performed a density-based correction of detonation data for pressed HMX powder [17] to obtain parameters at the target density for the product EoS and used the JWL form, \( P = A(1 - \omega/R_1\nu)\exp(-R_1\nu) + B(1 - \omega/R_2\nu)\exp(-R_2\nu) + \omega e/\nu \). An Arrhenius kinetic model based on Henson et al. [18] was used for HMX chemistry with the following parameters: \( \ln A = 29.35s^{-1} \), \( E_{act} = 35.612 \) (kcal/mol). In our simulations, the chemical kinetic model used temperatures based on the specific heat and density. For the HMX strength, we used a constant yield and shear modulus of 0.18 and 7.0 GPa, respectively, based on lower bound values in single crystal HMX experiments [19].

### Table 1. Unreacted equation-of-state parameters[15,16].

| \( \rho \) (g/cc) | \( A_0 \) (GPa) | \( A_1 \) (GPa) | \( A_2 \) (GPa) | \( A_3 \) (GPa) | \( B_0 \) | \( C_V \) (J/g-K) |
|-----------------|----------------|----------------|----------------|----------------|---------|---------------|
| 1.903           | -0.6334        | 14.29          | 10.0           | 190.0          | 1.03    | 1.10          |

### Table 2. Product JWL equation-of-state model parameters[17].

| \( A \) (GPa) | \( B \) (GPa) | \( R_1 \) | \( R_2 \) | \( \omega \) | \( q \) (GPa-cc/cc\(_0\)) |
|--------------|--------------|---------|---------|----------|--------------------------|
| 938.4941     | 21.9943      | 4.6     | 1.4     | 0.35     | 11.97                    |

For the Kapton flyer, we employed a tabular EoS with a reference density of 1.414 g/cc from the LEOS library [20]. A Mie-Gruneisen EoS was used for the Viton A with the following parameters: \( \rho = 1.865 \) g/cc, \( C_0 = 1.709 \) km/s, \( S_1 = 1.915 \), \( S_2 = -0.1578 \), \( \Gamma = 1.31 \).

### 2.2. Geometry and Microstructure

We simulated a 1.0 mm x 1.0 mm x 127 \( \mu \)m thick Kapton flyer impacting a 3.78 mm diameter x 1.89 mm long LX-10 specimen with approximately 1 \( \mu \)m mesh resolution. The experimental threshold speed for LX-10 shock initiation using this Kapton flyer geometry is 4.3 +/- 0.02 km/s [13]. For these simulations, we assumed a symmetry boundary condition at the centerline of specimen with all other outer LX-10 surfaces free of constraints. We also assumed a non-distorted flyer impacting the LX-10 specimen in a planar fashion. The specimen holder was not included in these simulations.

LLNL’s ParticlePack code [21] was used to numerically generate the LX-10 microstructure. While LX-10 has a nominal porosity of 2%, we also explored porosities of 5%, 10%, and 20%. Since there is limited data on pores in LX-10, we studied a discrete range of radii: 5, 12.5, and 25 \( \mu \)m. For numerical microstructural generations, pore radii were uniform and pore positions were spatially random but constrained not to overlap.

### 3. Results and Discussion

Figure 1 shows the progression of a reactive meso-scale simulation of short-pulse shock initiation. It shows the initial impact of the flyer and shock loading, collapse and reaction at multiple pores,
Figure 1. Progression of the meso-scale simulation of short-pulse initiation showing the flyer impact and shock, a. collapse and reaction at pores, and b. continued growth of reaction with a chemically-supported front. Pressure (0-0.3 Mbar) is shown on the upper half of the figure and mass fraction reacted (0-1) is shown on the lower half.

and the continued growth of reaction with a chemically-supported front. Simulation calculated for a flyer velocity of 4.3 km/s, 5 \( \mu \)m radius pores, and 2% porosity.

Figure 2 contrasts the predicted pressure time-histories at various horizontal positions (0, 0.57, 1.13, and 1.70 mm) near the centerline for reactive and inert simulations subjected to identical loading conditions. Following the initial short-pulse shock from the Kapton flyer, there is a sharp decay in the peak pressure with position due to the rarefactions in the inert case. While the initial shock is nearly identical in the reactive case, the decay in the peak pressure with position is relatively small in comparison. In addition for the reactive case, the post-peak decay in pressure decreases with increasing horizontal position. This suggests that chemical mechanisms are active and growing. We calculate the reaction front speed to be 5.18 km/s which is well short of the LX-10 detonation speed of 8.82 km/s but faster than the bulk sound speed. Possible reasons for this discrepancy may be that the global kinetics are not sufficiently rapid or the simulated sample domain is not large enough for the chemically-supported reaction front to develop fully into a steady-state detonation. In regards to the latter point, experimental observations [13] show only that the sample was consumed, but the reaction front speed was not measured for comparison to our predictions.

Figure 3 shows the influence of pore radii on mass fraction reacted for 2% porosity and a flyer velocity of 4.3 km/s and 5.3 km/s. The mass fraction reacted \( \left(F(t)\right) \) fit a power law relationship in time, \( F(t) = Ct^n \) with \( t \) in \( \mu \)s. At 4.3 km/s, the reaction evolution increases with decreasing pore size. Since the number density of pores swept over by the reaction front is inversely related to its size, there are fewer pores available at larger sizes. Fewer potential hot spots retards the evolution of the reaction.

At the higher flyer velocity of 5.3 km/s, the influence of pore size is relatively less apparent in the reaction evolution. In general, homogeneous shock initiation can occur at higher flyer velocities due to the higher impact-induced temperatures in the bulk material, i.e., a shock initiation mode that is not facilitated by hot spots but bulk thermal initiation observed in liquid and single-crystal explosives [9]. Mixed heterogeneous-homogeneous shock initiation modes can lead to reaction evolutions that are relatively less influenced by pore size since they do not depend solely on hot spot mechanisms. Figure 4 shows a mixed-mode shock initiation response.
Figure 2. Pressure time-histories for inert and reactive simulations at horizontal positions of 0, 0.57, 1.13, and 1.70 mm. Simulations calculated for a flyer velocity of 4.3 km/s, 5 µm radius pores, and 2% porosity.

Figure 3. Mass fraction reacted for pore radii of 5, 12.5, and 25 µm, 2% porosity, and a) 4.3 km/s flyer and b) 5.3 km/s flyer. Mass fraction reacted \((F(t))\) fit a power law relationship in time, \(F(t) = Ct^n\) with \(t\) in µs. For 4.3 km/s and 5, 12.5, 25 µm, the power law parameters are \((C, n) = (4.19, 2.08), (2.28, 2.05), \) and \((2.18, 2.25)\), respectively. For 5.3 km/s and 5, 12.5, 25 µm, the power law parameters are \((C, n) = (3.25, 1.52), (1.89, 1.38), \) and \((1.76, 1.50)\), respectively.

For a flyer velocity of 5.3 km/s, 2% porosity, and 5 µm pore radius. Homogeneous shock initiation is more prominent in the center of the domain before rarefactions have degraded it, where as the reaction front on the periphery is more dependent on hot spots to propagate outwards.

3.1. Effects of Porosity

Figure 5 shows the influence of porosity (2%, 5%, 10%, 20%) on mass fraction reacted for 5 µm pore radius and a flyer velocity of 4.3 km/s and 5.3 km/s. Once again, mass fraction reacted fit a power law relationship in time, \(F(t) = Ct^n\) with \(t\) in µs. At 4.3 km/s, the reaction evolution increases with increasing porosity. Since the number density of potential hot spots swept over by the reaction front is directly related to the porosity level, we expect the reaction to evolve more rapidly with higher porosities. In general, though, we would expect the energy expended in compaction to become increasingly larger and reduce the likelihood of initiation at higher
Figure 4. Simulation demonstrating mixed homogeneous-heterogeneous shock initiation modes. Homogeneous shock initiation is more prominent in the center of the domain, whereas the reaction front on the periphery is more dependent on hot spots to propagate outwards.

At 5.3 km/s, the influence of porosity is relatively less apparent in the reaction evolution due to the presence of heterogeneous-homogeneous shock initiation modes.

Figure 5. Mass fraction reacted for porosity of 2%, 5%, 10%, and 20%, 5 µm pore radius, and a) 4.3 km/s flyer and b) 5.3 km/s flyer. Mass fraction reacted fit a power law relationship in time, \( F(t) = Ct^n \) with \( t \) in \( \mu \text{s} \). For 4.3 km/s and 5%, 10%, 20%, the power law parameters are \((C, n) = (5.43, 2.00), (7.29, 1.95), (6.13, 1.72)\), respectively. For 5.3 km/s and 5%, 10%, 20%, the power law parameters are \((C, n) = (4.30, 1.57), (5.02, 1.59), (5.10, 1.54)\), respectively. Power law parameters for the 2% porosity case are already reported in figure 3.

4. Summary and Future Directions
Our calculations show that the reaction evolution fit a power law relationship in time and increases with increasing porosity, decreasing pore size, and increasing flyer velocity. While heterogeneous shock initiation modes, dependent on hot spot mechanisms, are predicted at lower flyer velocities, mixed heterogeneous-homogeneous shock initiation modes, less dependent on hot spots, are predicted at higher velocities. The constant specific heat assumption tends
to make our predictions more reactive, qualitatively, than would be expected for more physical specific heat values that increase with temperature.

Future directions should include validation of reactive meso-scale simulations with short-pulse experiments to improve confidence in predictions, as well as exploration of the effects of pore structure and HMX chemical-thermal-mechanical models on threshold flyer velocity for shock initiation. These types of modeling approaches may be able to answer key questions on shock sensitivity: How do pore size distributions and porosity gradients influence shock initiation thresholds? What are the lower and upper bounds of pore size that contributes to hot spot mechanisms? How do these bounds change with pore morphology and orientation? What is the upper bound on porosity before compaction energy expenditures suppress initiation? How do alternative chemical kinetic models and temperature-dependent thermal properties (e.g., specific heat) for HMX influence predictions? What is the role of numerics and mesh size on reactivity?

Acknowledgements
This work was performed under the auspices of the U.S. DOE by LLNL under Contract DE-AC52-07NA27344. This research was supported by the Joint DoD-DOE Munitions Technology Development Program. LLNL Technical Report, LLNL-CONF-644121-2013.

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