Analyses on the effect of hot spot density on material consumption rate

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Abstract. There is an observed effect of an explosive’s constituent grain size and density on its performance. At the mesoscale, it is the outward burning of hot spots that controls observed performance. While statistical hot spot models can integrate the mesoscale behaviour to macroscale simulations, it is unknown what the density of created hot spots is as a function of grain size and porosity. Simulating mesoscale hot spot distributions and varying hot spot density, we discuss the resultant performance as influenced by inter-pore distance and pore distribution.

1. Introduction
Shock initiation of explosives is attributed to localized high temperature regions called hot spots. Since explosives with lower pressed densities (and thus more porosity) are more sensitive to shock initiation, the mechanism of hot spot nucleation is often attributed to pore-collapse [1]. Hot spots are needed since the temperature of pure shock heating in nonporous crystal is not enough to decompose explosives on the timescales observed [2]. Every nucleated hot spot will drive chemical processes and can form outward burning flames. While there has been much analysis on the meso-scale nucleation of hot spots, their rate of outward burn is left unclear.

Simulating and understanding outward radial burn rate is difficult for a variety of reasons. Experimentally, direct observation to elucidate the dominant mechanism is complicated by high temperature and pressure, small size and time scales, and the evolving decomposition [3]. Simulation is challenging since explicit mesoscale structures are difficult to obtain and mesh. Also, the requisite mesh resolution of the flame front is high and this constrains the simulation volume and decreases the time steps. Herein, we investigate how we may approach this challenging problem.

2. Hot spot criticality
For macroscale simulation of full high explosive components (generally on the cm scale), not only must mesoscale features be neglected but their behavior generalized for much larger volumes. While there are some reactive flow models that utilize a pore size in their formulation, it is often treated as a parameter and a characteristic pore radius is used. As a shock passes, hot spots of many sizes will be created and not all will continue to burn all the surrounding explosive material. Some stagnate and die from thermal conduction on the tens of nanosecond timescales and others will continue to burn...
indefinitely being self-driven by exothermic reactions. The “critical” hot spot is that which is just hot enough to burn indefinitely.

Originally, simplified approximate solutions to empirical heat conduction equations [4] were used to eliminate subcritical hot spots from reactive flow simulation. Other estimates of hot spot criticality have come from small scale burning simulation of a hot spot as surrounded by hotter material [5]. Relating this critical hot spot size to an original pore size has been done by extracting the temperature distribution from an isolated, collapsed pore and remapping it into a simpler burn model [6]. This has given rate of burn and criticality information for the range of relevant pore sizes.

However, hot spots do not burn independently. As they all burn outward, they interact with other outward burning hot spots to consume all explosive material. Analysing them without accounting for this inter-pore distance could be an issue. A pore that burns indefinitely is required to burn much more material than a hot spot from a pore that burns in material that had 2-8% porosity. In other words, a subcritical hot spot that would be discarded by this criteria, could still burn to its nearest neighbour in a physical microstructure.

A two dimensional model of a radially burning hot spot is constructed in multiphysics hydrodynamic code ALE3D as in figure 1. Here a wedge of material approximates a spherical burn by having symmetry boundaries above and below and initialized at constant pressure. The temperature field is approximated from pore collapse simulations as in [6]. There is a symmetry boundary placed on the right end of the wedge. By moving the position of this right symmetry boundary, we simulate how close the hot spot is to its nearest neighbour. Assuming a regular distribution of pores the same size for the calculated temperature field, we can approximate porosity.

[Figure 1. Radial burn model initial conditions with remapped 2D temperature field from the collapse model of an $R = 0.05$ μm pore under a 36 GPa shock.]

Guiding the explosive decomposition is chemical-kinetics code CHEETAH which is called by ALE3D every time step to update state variables and evolve the mass fractions of products, intermediates, and reactants. CHEETAH is using a custom library built with the decomposition steps and rates extracted from quantum molecular dynamics simulations [7] and has incorporated values of thermal conductivity extrapolated [8] from experimental measurement [9]. To find the smallest size that burns to completion (the critical hot spot size), we parametrically vary pore sizes (scaling the temperature distribution linearly to the pore size) and monitor burn progress. The initial pore radius of a critical hot spot is estimated from this parametric study as the midpoint of the smallest size that consumes all simulated explosive material and the largest size that did not. In figure 2, the initial pore radius of critical hot spot size was calculated at three pressures (10, 25, and 36 GPa) for porosity $\Phi = 0.09\%$ and the error bars are the nearest neighbour simulations which bound the regime of stagnation/completion. For $\Phi = 1.6, 3.7, 6.4, 12.5\%$, we are only calculating the critical hot spot size resulting from a 10 GPa impact shock and scaling the critical hot spot trend line, maintaining the original error bars.
Figure 2 shows that there can be a significant decrease in the critical hot spot initial pore size, with an increase in porosity. This helps explain why materials with more porosity (and less chemical energy) can have shorter run-to-detonation distances than their denser counterparts. Even though more porosity means more initiation sites, it also means that as the porosity increases (and the pressure evolves to increase) the initial pore size leading to a critical hot spot can also be smaller. Since the initial pore size leading to a critical hot spot size decreases, there are many more of them since it seems the frequency distribution of pore size is an exponential [10].

3. Hot spot merging
In the above analysis, we are moving a symmetry boundary to simulate a regular grid of hot spots. In reality, there are distributions of pore sizes, as well as being distributed randomly in space. We are aware that outward burning hot spots accelerate as they approach other outward burning flames and this makes the problem highly three dimensional. We investigated the feasibility of a three dimensional burning simulation.

Constraining the simulation budget between 1,000,000-2,000,000 simulation elements and requiring the mesh refinement be about 1-2 nm for resolving flame fronts, the resulting simulation cube is about 0.1 μm on each side. This volume size is smaller than the size of what many believe to be the critical hot spot size; however it is useful for a few reasons. Firstly, it can demonstrate the effect of pore density on the ability for smaller pores to burn. Secondly, it could be used as a submodel for a zone in a much larger simulation and help dictate or parameterize a reactive flow model. Thirdly, it can demonstrate how important three-dimensional burn behaviour is on the mesoscale.

Using a measured distribution of porosity [10] described by equation (1) and table 1, we can generate volumes with different porosities as in figure 3. This process is done using a Matlab script which randomly generates a pore size and places it in a random (but not overlapping) position in the volume. This is repeated until the desired amount of porosity is reached. We note that this is also possible with the utilization of the Particle Pack module for the multiphysics code ALE3D. Once the pore sizes and locations are known, we can overlay pore locations with temperature distributions extracted from pore collapse simulations. For the simplicity of simulation, we simulated each pore center to be surrounded by shells of constant temperature. Namely, there were shells of 3100, 1800, 1450, and 1150 K at 3, 4, 5, and 6 pore radii. Surrounding this was material at post shock state with temperature of 829 K. The model was initialized at 25 GPa pressure. The resulting simulation volume looks like that in figure 4.

\[
F(x) = a_1 e^{-\frac{(\log(x) - \log(x_1))^2}{2w_1^2}} + a_2 e^{-\frac{(\log(x) - \log(x_2))^2}{2w_2^2}}
\]

(1)
Table 1. Values describing the measured pore size distribution from Willey, et al. [10] for ultrafine (UF) triaminotrinitrobenzene (TATB).

| Parameter | UF-TATB |
|-----------|---------|
| $a_1$     | 5.3e-6  |
| $10^{c_1}$| 7.3     |
| $10^{w_1}$| 1.19    |
| $a_2$     | 2.74e-5 |
| $10^{c_2}$| 36.5    |
| $10^{w_2}$| 2.51    |

Figure 3. A square 1 mm volume at representing 8% porosity constructed using the distribution in (1) containing about 30,000 pores.

Figure 4. A square 0.1 μm volume with 8% porosity from (1) where the pore locations are surrounded by shells of temperature to simulate a post-shocked volume of originally spherical pores.

With initial conditions set, the simulation can be analysed. Once again, we are utilizing a custom CHEETAH library, which incorporates previously determined decomposition kinetics from quantum molecular dynamics simulations, to guide our equation of state and composition. Every time step, ALE3D will call CHEETAH for the necessary state variables. Simulations take several weeks spread across 64 processors each. The resulting outward flame positions are plotted as surfaces in figure 5.
**Figure 5.** Merging flame fronts, plotted as isosurfaces where the composition of TATB equals 50%, from a simulation of ultrafine-TATB with 2% porosity in a square, $(0.1 \mu m)^3$, volume.

This type of simulation was constructed for different porosities and their flame speeds monitored until they decompose to 50% TATB explosive. While it is possible to continue the analysis, a divergence of simulated and real conditions would occur. The boundary conditions imposed here are symmetries on all sides. However, in a real event advection of hot gas would affect the material decomposition rate adversely. For different porosities, the rates of decomposition are plotted in figure 6.

**Figure 6.** Rates of decomposition from Monte Carlo simulations of spatial pore distribution for differing porosities.

Figure 6 is useful for a discussion of its trends. As porosity increases, the rate of decomposition increases which is to be expected; however, it is increasing less rapidly than the added amount of thermal energy each hot spot is contributing. In other words, it is mainly increasing because there is extra thermal energy from each pore instead of increasing from additional hot spot merging. This is an artefact of initial conditions. The hot spot temperature shells were chosen to be large so these small pores that fit inside our simulation volume do not stagnate, however this resulted in overlapping hot regions (which did not add thermal energy). Also, we note that since the inner shell was too large the rate of decomposition for the entire simulation volume is too fast. Initial conditions should be a subject of future investigation where calibration is more manageable in two dimensional models.
4. Conclusions
A hot spot in isolation is dependent on the amount of thermal energy it is endowed with to survive thermal conduction. The smallest (or critical) pore size to survive conduction, and continue burning, has been addressed for isolated pores in the past through both approximate solutions to the heat balance equation and one-dimensional simulation.

Our hot spot criticality calculation, presented here, has shown a pore with a neighbour, including that which is at a considerable distance with respect to the hot spot temperature field, is much more likely to burn to completion. In fact, we have demonstrated that the observed critical pore size decreases by several orders of magnitude as the hot spot temperature fields approach each other and meet.

Additionally, from our simulation of three-dimensional hot spot fields, we see that adding more hot spots clearly gives faster decomposition rates. We note that if there is excessive pore density, than there is likely to be a reduction in the amount of thermal energy generated by each pore as deformation fields interact and externally calculated, independent temperature fields overlap. As a result, if an explosive’s porosity is concentrated in areas (i.e. on grain boundaries) it could be seen as a less effective use of a part’s porosity since individually burning pores could consume more explosive if they are uniformly spaced.

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