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Predicting the Effects of Thermally-Induced Density Gradients on the Hydrodynamic Behavior of PBX 9502

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Abstract. High explosives are often exposed to thermal and mechanical stimuli within normal and abnormal environments. These stimuli can introduce density gradients in the explosive charge but the effect of such gradients is not well-understood. The goal of this work is to develop a comprehensive methodology to model the evolution of density gradients under various thermal conditions and then to predict shock initiation and detonation behavior of the gradated sample. PBX 9502 was chosen for these scoping studies because it has a large coefficient of thermal expansion and because the available shock initiation data indicates that initial temperature strongly affects the shock-to-detonation transition (SDT). The multiphysics code COMSOL was first used to model induced density gradients in a slab of PBX 9502 with asymmetric heating. Then, the gradated sample was constructed in the Pagosa hydrocode to simulate steel flyer impact scenarios, with SDT and detonation propagation following a calibrated Scaled Uniform Reactive Front (SURF) burn model. The gradated material exhibited very different initiation and corner turning behavior compared to homogeneous materials, and the orientation of the gradation was also found to have some effect. The methodology used here can estimate thermomechanical response of the material for non-uniform thermal environments and compute the resulting detonation properties of the PBX.

INTRODUCTION

Typical plastic-bonded explosives (PBX) are polymer matrix composites with a very high (often > 90 wt%) solids loading of explosive crystals. PBX 9502 (95 wt% triaminotrinitrobenzene (TATB), 5 wt% Kel-F 800 binder) is an insensitive high explosive, meaning that it generally cannot be detonated without a significant shock input. In part due to its insensitivity, PBX 9502 exhibits a number of non-ideal characteristics, such as poor corner turning, large reaction zone, and curved detonation front [1]. Compared to other PBX formulations, PBX 9502 shock-to-detonation transition (SDT) also exhibits high dependence on the initial temperature of the explosive [2]. Some debate exists as to whether this is inherently a temperature effect, where the temperature of the PBX contributes to a faster than normal chemical reaction rate, or whether this is actually a density effect, where the lower density material has a larger number of so-called “hot spots” which contribute to the buildup of ignition and growth of the detonation front [3,4].

Here, we hypothesize that the material density is the more critical parameter, and seek to investigate the effect of spatially-variable density across a charge of PBX 9502 under an idealized fragment or flyer impact experiment. Our other work [3,5] suggests that more subtle factors, namely the void size distribution, affect behavior. But insufficient data exist to incorporate those details at this time; however, we could employ the approach described in [3] to account for the nuanced effects of microstructural details for use in the framework described here. There are multiple approaches described in the literature to generate density gradients in PBX materials, both experimentally and in simulations. Generally, asymmetric heating is a flexible, convenient method to produce gradated samples. Recent advancements in additive manufacturing (AM) of explosives offer a more precise method of controlled density variation [6]. Shock wave and/or detonation wave shaping has already been demonstrated at the small scale using such techniques [7]. However, most AM efforts have used new explosive formulations which are optimized for the manufacturing methods but are not well-characterized. For this reason, we focus here on the legacy material PBX 9502, but suggest that this methodology may have some usefulness to the emerging AM field.

In this work, the multiphysics code COMSOL was first used to model induced density gradients in PBX 9502 with asymmetric heating and confinement. Then, the gradated sample was constructed in the Pagosa hydrocode and subjected to steel flyer impact. SDT and detonation propagation used a Scaled Uniform Reactive Front (SURF) burn model which was parameterized to intermediate (uncalibrated) PBX 9502 density. In theory, using this methodology, the effect of any thermomechanical environment on PBX material state (e.g. density) can be predicted, and then the result of that off-normal PBX state on shock initiation and detonation propagation can be assessed. This current approach still relies on shock initiation calibration experiments, such as gas gun impact or wedge tests, but SURF was built to use fundamental constituent properties to calculate mesoscale-driven ignition and growth in the manner of [3,5], mentioned above. Once such experimental data exists, it should be straightforward to implement it into SURF calculations and validate against integrated experiments.

GENERATING GRADATED DENSITY WITH COMSOL

Figure 1 shows the 2D axisymmetric geometry as it was set up in COMSOL. The PBX 9502 slab was set to a density of 1.890 g/cc and was given nominal material properties from the literature. Bulk coefficient of thermal expansion was taken from Skidmore [8], while mechanical properties (e.g. modulus, Poisson's ratio) were taken from Gibbs and Popolato [9]. The off-normal scenario chosen for this research was a moderately confined, asymmetrically heated experiment. The PBX 9502 was bordered at the top and bottom of the slab with steel and with aluminum at the lateral boundaries, with metal thermal properties assigned from the COMSOL material library. The bottom steel layer was given a constant power flux representative of a heater block, while the other boundaries served as heat sinks. The PBX was allowed to expand during heating – that is, the metal provided no confinement. This was chosen to maximize the thermal gradients across PBX 9502, giving us the best chance of observing density gradients. In principle, this problem would be simplified by imposing an arbitrary gradient, however, this problem exists within a framework where the actual mechanical confinement would play a role and it is necessary to have the capability afforded by COMSOL to account for those effects.

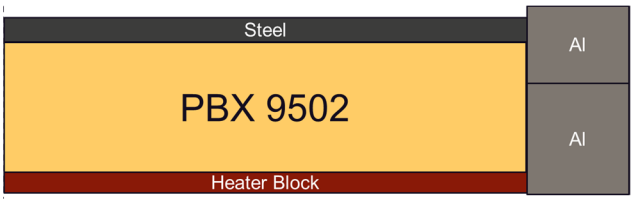

FIGURE 1. 2D axisymmetric representation of the experiment geometry. The PBX 9502 slab was 6" diameter, 1.5" thick.

PBX 9502 rapidly decomposes above 250°C, but will remain stable for a reasonable time at, or below this temperature. Consequently, this is the highest temperature for which shock initiation data exists. The heating conditions and time duration of the simulation were therefore run so that the hottest part of the PBX corresponded to approximately 250°C, while the coolest part of the slab ended up around 75°C. This lower temperature also has existing shock calibration data, but no such data exists at any intermediate temperature. Therefore, new burn model parameters would need to be generated for any intermediate temperature, or more accurately, any intermediate density.

COMSOL generates continuous density, temperature and density fields from this type of simulation. To avoid needing to generate SURF parameters for every single pixel, we performed a convergence study in Pagosa, finding that the solution converged when the slab was reconstructed with no fewer than 27 discrete density zones. Figure 2 shows the contours which bound these binned constant temperature / density regions. The results show PBX 9502 displaces mainly laterally, as would be expected with no confinement from the aluminum. The bottom center of the sample (bottom left in Fig. 2) reaches the highest temperature, being adjacent to the heater block and far from thermally conductive material. The top right of the sample is the coolest, being adjacent to effectively steel and aluminum heat sinks. The simulation used a linear CTE such that the density contours were identical to the temperature contours. The coolest material is the highest remaining density but is still lower than nominal, ambient density.

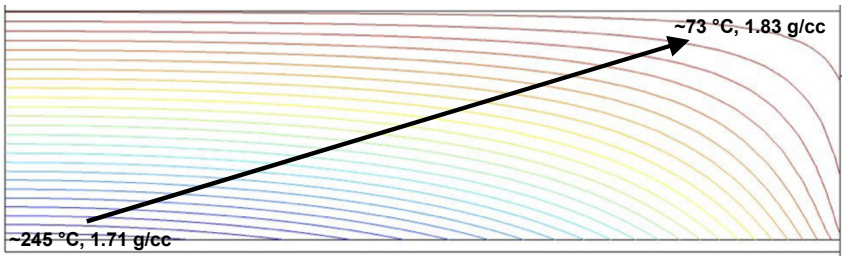

FIGURE 2. PBX 9502 temperature / density zones after heating.

SDT STUDIES IN PAGOSA USING A SURF BURN MODEL

Generation of SURF Burn Model Parameters for Intermediate Densities

The SURF burn model was developed using an ignition and growth concept, with measurable mesoscale parameters in mind [10]. After ignition, the conversion of solid material to gas products follows an exponential function which depends on a scaled length s, which is effectively the ratio of the radius r of a given burn center to the characteristic distance a_{bc} between adjacent burn centers. s is a function of the shock pressure, where on a rate basis can also be written as:

$$\dot{s}(P_s) = \dot{r}(t)\left(\frac{1}{\frac{4}{3}\pi a_{bc}^3}\right)^{\frac{1}{3}} = D\mathrm{N}^{\frac{1}{3}} \quad (1)$$

such that the scaled length rate is the product of the deflagration velocity D and the number density of burn centers N, both of which also depend on the shock pressure. Unfortunately, despite this well-intentioned mesoscale construction, D and N are quite difficult to measure experimentally, and therefore typical utilization of SURF requires calibration to shock initiation experiments. Generally, the scaled length rate is parameterized using the function $\exp(A+B*\mathrm{P_s})$, where A and B are constants for a given material state (composition, density, etc). These parameters are then implemented into a hydrocode such as Pagosa for shock initiation and propagation simulations.

For the gradient density simulations in this present work, absolute x-y coordinates of each contour within the PBX 9502 slab were exported from COMSOL and used to construct the boundaries of constant density zones. Each zone needs the A and B SURF parameters assigned in order to run the simulation. The top right zone (highest density) and bottom left zone (lowest density) had approximately the correct densities for using existing shock initiation data to calibrate the SURF burn model, but all of the intermediate density regions had no calibration data available. To generate the SURF A and B parameters over the entire density range, wedge test data from Dallman and Wackerle [11] at 76°C was used in conjunction with the existing calibrations for the SURF parameters A and B at 23°C and 250°C for PBX 9502 along with an iterative procedure to estimate A and B in the uncalibrated region. We assumed that the Pop plot time-to-detonation for a given $\mathrm{P_s}$ is proportional to $\dot{s}$. Figure 3 (left) shows an exponential fit to the three existing data points from Dallman and Wackerle at 10 GPa shock pressure (dashed line) along with the proportionally-adjusted $\dot{s}$ curve (solid line) calculated from this procedure. The unknown (uncalibrated) values of A and B at the void fraction of 0.05 (76°C) were then adjusted in an iterative process (Fig. 3 right) until the experimental rate curve and proportionally-adjusted, iterated $\dot{s}$ curve matched as closely as possible. We note that the density at elevated temperatures was never measured directly, but instead calculated from thermal expansion data. Now that A and B were adequately described as functions of density, each PBX 9502 density zone from the COMSOL simulations could be assigned complete SURF parameters.

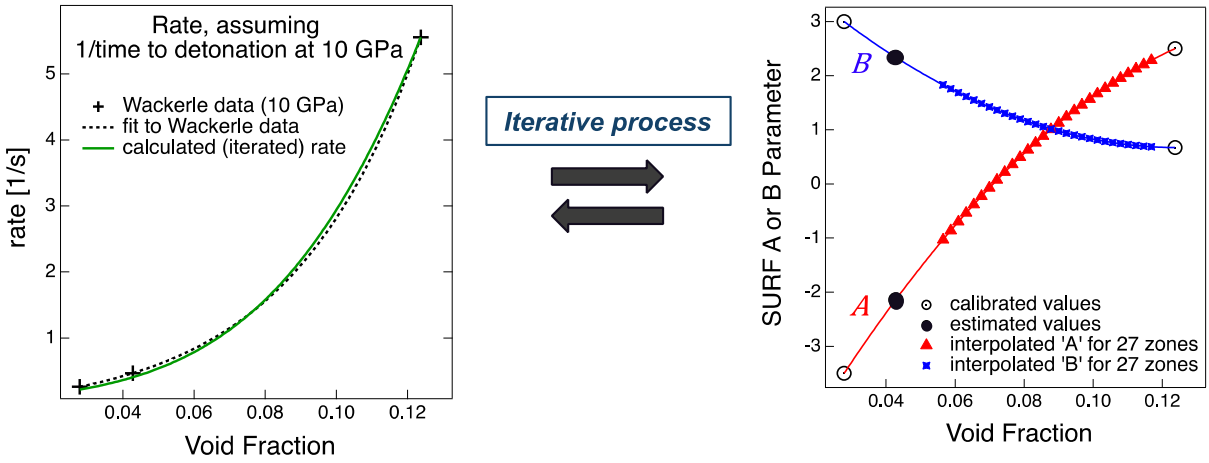

FIGURE 3. The iterative process for determining SURF parameters for PBX 9502 at non-calibrated density.

Flyer Impact Simulations in Pagosa Using Assigned SURF Parameters

The details of the Pagosa hydrocode have been discussed elsewhere and so are omitted here for brevity [12]. We used the ambient temperature PBX 9502 Us-Up relationship for all cases (Us=2.97 +1.81Up [mm/μs]), ambient JWL parameters as in [13], and resolution of 200 μm. 30 mm dia. by 10 mm thick stainless steel flyer impact simulations were run on PBX 9502 in four different conditions: (1) a uniform slab of PBX 9502 at 23°C; (2) an identical slab at 250°C; (3) the gradated density slab from Fig. 2, where the flyer impacts the higher density region first ("Gradient Up"); and (4) the gradated density slab from Fig. 2, where the flyer impacts the lower density region first ("Gradient Down"). The slab had a diameter of 20 cm, while the 1 cm thick stainless-steel flyer (SESAME ID 4272) had a diameter of 3 cm and impacted the material at 1.33 km/s. Figure 4 shows a still frame mid-simulation of (4), plotting pressure and displacement 5 μs after impact.

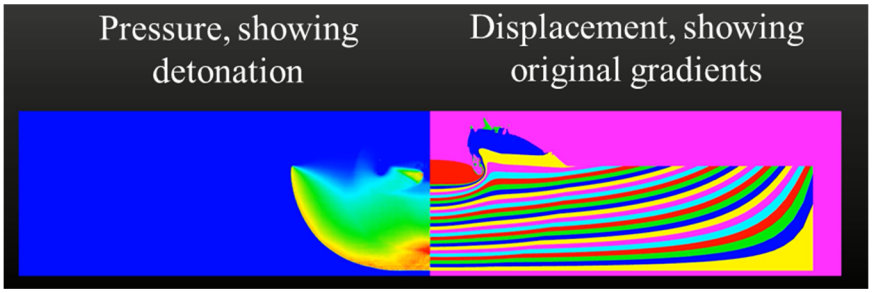

FIGURE 4. Pressure and displacement of the gradated density PBX 9502 5 μs after flyer impact.

Figure 5 shows still frames for all four cases at 4.25 μs and 15 μs. These times were chosen to generally represent the "corner turning" or initiation regime and the detonation propagation regime, respectively. Several interesting phenomena are observed. For the constant density materials, temperature has a drastic effect on corner turning, as has been proven experimentally [14]. The gradated materials do not show large differences in corner turning.

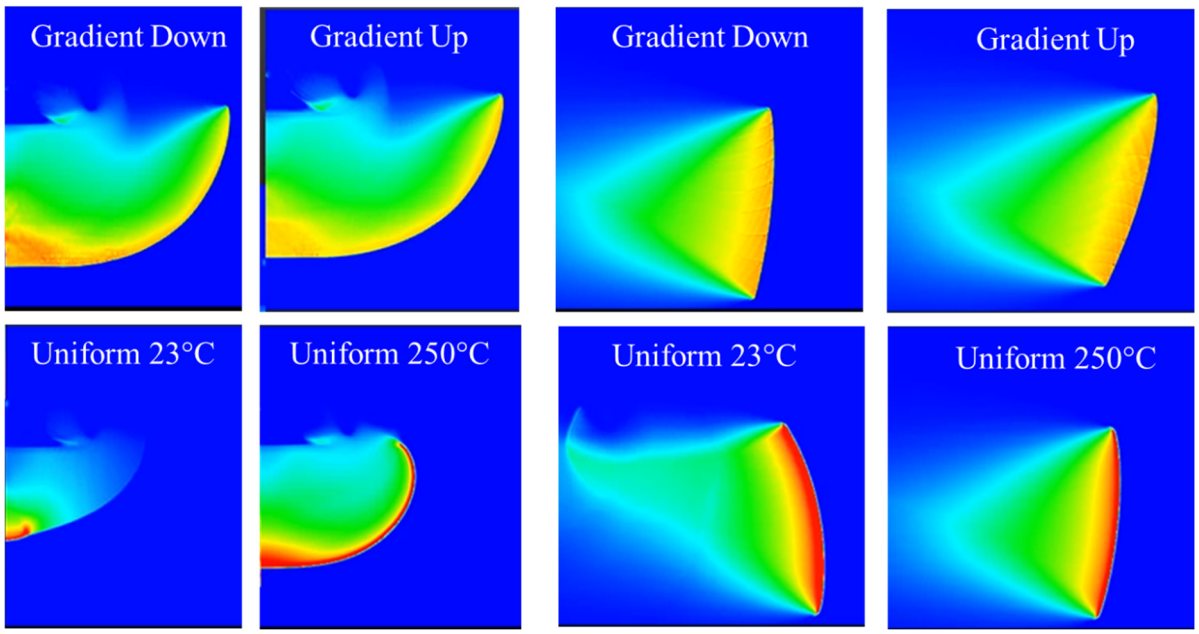

FIGURE 5. Comparison of the four materials under flyer impact initiation, showing detonation waves at 4.25 μs (a) and 15 μs (b). Detonation is proceeding from left to right. Clear differences in corner turning and wave shape are observed.

The propagation behavior does reveal some interesting differences between the cases. The wave curvature is different in all four cases. The Gradient Up case, where the highest density region is on top, has a clear tilt in the detonation front such that the detonation is proceeding faster in the higher density regions. The Gradient Down case is not the opposite, but rather the detonation is fastest somewhere in the middle density regions. This is likely a residual effect of the initiation conditions; that is, if the slab was infinitely wide, the detonation in the highest density regions would eventually overtake lower density regions. For the slab size considered here, the detonation front in this case is constantly evolving. Similarly, the 23°C slab also has a tilted wave front that appears to be a residual effect from poor corner turning at the top of the slab. Eventually, the wave shape should be identical to that in the 250°C slab, but moving faster due to the higher density. It should also be noted that there is some fine detail in the wave structure in the gradated density cases, but this is an artifact of imposing discrete boundaries of homogeneous density zones rather than a continuous density gradient.

DISCUSSION AND FUTURE WORK

This study demonstrates proof of concept for calculating thermal gradients and therefore density gradients in PBX materials under realistic heating conditions, generating burn model parameters for each material state, and running SDT simulations using that burn model in a hydrocode. This work demonstrates a viable framework to calculate and estimate the response over the entire life cycle of a PBX slab, going from a pristine to a thermomechanically damaged state, then detonated via shock initiation. The process for generating SURF parameters for material in an uncalibrated state is self-consistent, though it does rely on some assumptions about the underlying physics. However, SURF is built on mesoscale parameters that can in theory be measured. In the future, we will attempt to validate the calculated parameters by using data such as void size distributions from x-ray or neutron scattering [15] and high pressure deflagration rate [16]. Our approach could also be validated directly with flyer impact experiments, though directly measuring the exact details of the detonation front is difficult unless a diagnostic like proton radiography is used.

For the specific case studied here, we find that both initiation (and therefore corner turning) and propagation regimes in PBX 9502 were measurably affected by the density and gradients of the density with respect to the flyer. Perhaps most importantly, the detonation front curvature and tilt were strongly affected by the density gradient, even for 10+ μs after initiation began. For small samples, this introduces the possibility of intentional wave shaping through density gradients. This type of analysis should be especially interesting for AM explosive materials, where density

gradients can be introduced on-demand. While PBX 9502 is not a likely candidate for additive manufacture, the lessons learned here regarding tight control of density should have wide-ranging implications.

CONCLUSIONS

A combination of COMSOL multiphysics modeling, SURF burn model calibrations / approximations, and flyer impact simulations in Pagosa were used to examine the effects of temperature and density on detonation initiation and propagation in PBX 9502. A novel iterative process was successful in generating reasonable SURF parameters for the explosive at densities not calibrated by existing shock initiation experiments. Not only were different effects seen between uniform density and gradient density PBX 9502 samples, but the specifics of the density gradients also were found to be important for corner turning and detonation wave shape. This methodology is allows us to estimate the changes in response over the entire storage, handling and testing lifetime of an explosive part. The strong effect of density gradients has some implications for additively manufactured explosives as well. Future work could include validation experiments for the fragment impact scenario as well as shock initiation experiments at intermediate densities to test the burn model approximations. Finally, while this work shows a clear methodology to incorporate non-uniformities in an explosive charge due to the thermal and mechanical environment, the conclusions rely on several assumptions and approximations. Future work should strive to reduce the uncertainties that arise due to those issues.

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REFERENCES

1. L. G. Hill, J. B. Bdzil, T. D. Aslam, “Front curvature rate stick measurements and detonation shock dynamics calibration for PBX 9502 over a wide temperature range,” in *Proceedings of the Eleventh International Symposium on Detonation. Snowmass Village, CO, 1998,* pp. 1029-37.
2. R. L. Gustavsen, R. J. Gehr, S. M. Bucholtz, A. H. Pacheco, B. D. Bartram, “Shock initiation of the TATB-based explosive PBX-9502 heated to ~ 76°C,” *AIP Conf. Proc.* **1793**, 030017 (2017).
3. W. L. Perry, B. Clements, X. Ma, J. T. Mang, “Relating microstructure, temperature, and chemistry to explosive ignition and shock sensitivity,” Combust. Flame **190**, pp. 171-176 (2018).
4. C. M. Tarver, S. K. Chidester, A. L. Nichols, “Critical Conditions for Impact- and Shock-Induced Hot Spots in Solid Explosives,” J. Phys. Chem. **100**, pp. 5794-5799 (1996).
5. W. Lee Perry, B. Clements, X. Ma, J. D. Yeager, G. Watt, A. L. Higginbotham Duque. “Predicting the Effects of Density and Microstructure on Shock Initiation of Explosives,” in *Proceedings of the 16th International Detonation Symposium, Cambridge, MD, USA,* pp. 2-9 (2018).
6. A. M. Schmalzer, L. G. Hill, B. C. Tappan, P. R. Bowden, B. E. Clements, R. Menikoff, A.H. Mueller. “Manipulation of Corner-turning Behavior in High Explosives via Additive Manufacturing,” in *Proceedings of the 16th International Detonation Symposium, Cambridge, MD, USA,* pp. 408-414 (2018).
7. B. Branch, A. Ionita, B. M. Patterson, A. Schmalzer, B. Clements, A. Mueller, D. M. Dattelbaum, “A comparison of shockwave dynamics in stochastic and periodic porous polymer architectures,” Polymer **160**, pp. 325-337 (2019).
8. C. B. Skidmore, T. A. Butler, C. W. Sandoval, “The Elusive Coefficients of Thermal Expansion in PBX 9502,” Report No. LA-14003. Los Alamos National Lab., NM (2003).
9. T. R. Gibbs, A. Popolato, ed., *LASL Explosive Property Data*, Vol. 4, University of California Press (1980).
10. R. Menikoff, M. S. Shaw, “Reactive burn models and ignition & growth concept,” EPJ Web of Conferences, EDP Sciences, pp. 00003 (2010).
11. J. Dallman, J. Wackerle, “Temperature-dependent shock initiation of TATB-based high explosives,” in *Proceedings of the Tenth International Detonation Symposium, Boston, MA, USA,* pp. 130-138 (1993).

12. W. N. Weseloh, S. P. Clancy, J. W. Painter, “PAGOSA physics manual.” Report No. LA-14425-M. Los Alamos National Lab., Los Alamos, NM (2010).
13. X. Ma, B. Clements. “Using the Pagosa SURF Model to Simulate Fragment Impact on Energetic Materials for Safety Applications.” Report No. LA-UR-19-26765. Los Alamos National Lab., NM
14. C. M. Tarver, “Modeling Detonation Experiments on Triaminotrinitrobenzene (TATB)-Based Explosives LX-17, PBX 9502, and Ultrafine TATB,” J. Energ. Mater. **30**, pp. 220-251 (2012).
15. J. T. Mang, R. P. Hjelm, E. G. Francois, “Measurement of Porosity in a Composite High Explosive as a Function of Pressing Conditions by Ultra-Small-Angle Neutron Scattering with Contrast Variation,” Propellants Explos. Pyrotech. **35**, pp. 7-14 (2010).
16. S. F. Rice, M. F. Foltz, “Very high pressure combustion: reaction propagation rates of nitromethane within a diamond anvil cell,” Combust. Flame **87**, pp. 109-122 (1991).