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A. L. Nichols, III, R. Becker, W. M. Howard, A. Wemhoff

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TOWARD IMPROVED FIDELITY OF THERMAL EXPLOSION SIMULATIONS

A. L. Nichols, III[1], R. Becker[1], W. M. Howard[1], and A. Wemhoff[2]

[1] *Lawrence Livermore National Laboratory, Livermore CA 94551*
[2] *Mechanical Engineering, Villanova University, 800 Lancaster Avenue, Villanova, PA 19085*

Abstract. We will present results of an effort to improve the thermal/chemical/mechanical modeling of HMX based explosive like LX04 and LX10 for thermal cook-off. The original HMX model and analysis scheme were developed by Yoh et.al. for use in the ALE3D modeling framework. The current results were built to remedy the deficiencies of that original model. We concentrated our efforts in four areas. The first area was addition of porosity to the chemical material model framework in ALE3D that is used to model the HMX explosive formulation. This is needed to handle the roughly 2% porosity in solid explosives. The second area was the improvement of the HMX reaction network, which included the inclusion of a reactive phase change model base on work by Henson et.al. The third area required adding early decomposition gas species to the CHEETAH material database to develop more accurate equations of state for gaseous intermediates and products. Finally, it was necessary to improve the implicit mechanics module in ALE3D to more naturally handle the long time scales associated with thermal cook-off. The application of the resulting framework to the analysis of the Scaled Thermal Explosion (STEX) experiments will be discussed.

Keywords: thermal Cookoff, HMX, LX-10, ALE3D.
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INTRODUCTION

Predicting the response of a high explosive to thermal stimuli is an important component in understanding the safety associated of those explosives in accident scenarios. Previously, Yoh et. al. [1] used the chemical kinetics model for HMX developed by Tarver and Tran [2] to model an LX-04 Scaled Thermal Explosion Experiment (STEX) [3]. Those results were moderately successful but fell short in several areas. Though the time to event was good, the overall temperature profile missed many of the characteristics of the experimental results. For the strain gauge results, the model predicted strains approximately an order of magnitude higher than the experiment, with that deviation occurring from the point where the explosive made first contact with the container walls through thermal runaway.

There are a number of possible areas where the Yoh el.al. could be deficient. These include the equation of state of the intermediate and product gases, the reactant eos under high compression, the lack of the beta to delta phase change kinetics, burn rates at elevated temperature, the possibility of gas diffusion through the explosive composite, porosity in the explosive, and the lack of a fully implicit solve of the mechanics. In general, the biggest issue is that the model creates too much pressure too early in the reaction process. The purpose of this paper is to show the changes that have been made to the model to better predict the complete thermal-mechanical-chemical response of HMX/Viton-A explosive compositions.

IMPOROVED MODEL FOR HMX

After further analysis of the Yoh results, four areas were targeted for improvement: improved gaseous equations of state, the addition of a reversible porosity model to the chemical material model, an improved kinetics model, and improvements to the solution of the mechanical problem.

We follow the methodology developed by Yoh et. al. to define the thermal and mechanical characteristics of the material models of reactant and early product species. Examination of their Viton-A chemistry showed that it was itself a significant contributor to the overall pressurization, even though it is inert to temperatures above the decomposition of HMX. Thus, we have removed the Viton-A reaction chemistry from the model.

There are two areas where the kinetics derived from thermal response only process that Tarver and Tran used have trouble. First it does not include the effect of the β-δ HMX phase change on the thermal-mechanical process.The second issue is that the original model cannot handle the pressure dependent characteristics that are seen in experiments such as the closed pan DSC, here the temperature of reaction decreases as the pressure is increased.

There are 5 species in our reaction mechanism: β-HMX, δ-HMX, the first decomposition product f, the first gaseous species set hg, and the final gaseous product lg. In our reaction network, we replaced Tarver's first reaction with 2 reactions based on Henson et.al. 's [4] work on the β-δ phase change and modified it by replacing the forward and backward Arrhenius reactions with a simpler hyperbolic sine form:

$$k = A[x_\beta]\exp\left(-\frac{E}{RT}\right)\sinh\left(\Lambda^* - \frac{E_e^*}{RT}\right) \qquad (1)$$

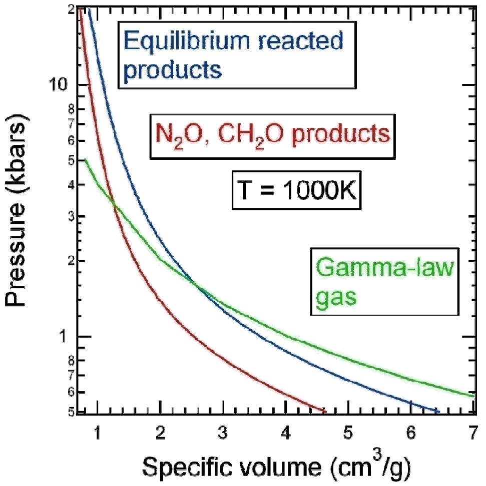

Figure 1. Pressure along the 1000K isotherm for the original gamma law gas and the new CHEETAH based products.

The sinh term captures the predominant direction of the reaction, while the Arrhenius factor describes the overall rate. In general one needs both a forward and reverse reaction of this form to be equivalent to the two Arrhenius form. However, since we are generally going in one direction we can get by with only one. The rest of the reactions continue to be based on Tarver's work but we have adjusted the Arrhenius pre-factors.

The previous gas models for the hg and lg gases were simple gamma-law gas models. We used the CHEETAH equation of state code [5] to create tabular equation of state tables for the intermediate and final product gases. The hg equation of state is based on CHEETAH's models for N_2O and CH_2O, and the final product equation of state was derived from a fully equilibrated product set. In Fig. 1 we show the comparison of 1000 K isotherms between the gamma-law gas and the new eos's. The new equations of state reduce the pressure at large volume expansions.

The scheme to optimize the reaction pre-factors used DSC and iso-thermal and ramped TGA small scale experiments to calibrate the reaction network through the first production of gaseous products. The auto-catalytic reaction between the f and hg species was calibrated using closed pan DSC results, and the hg to lg gas reactions were based on modeling the ODTX experiment. The results of the calibration are shown in Table 1. The error in either time or temperature for the TGA is now 0.02% from a previous 54%, and the DSC error now 1.1% as compared to 5.6%.

One area that has been neglected in the analysis of thermal explosions is the porosity of the composite explosive. In general, most explosives have a couple of percent porosity, without which it

TABLE 1. Kinetic parameters for HMX/viton .based HMX formulations.

Reaction(s)	Kinetic Relation	Activation Energy(cal/mol)	Prefactor lnA(sec-1)	Notes
$\beta \leftrightarrow \delta$	$k = Ax_\beta \exp\left(-\frac{E}{RT}\right)\sinh\left(\Lambda^* - \frac{E_e^*}{RT}\right)$	101847.2	107.3889	n = 1, Ee*/R = 1168 K, Λ* = 2.728
$\beta + \delta \leftrightarrow 2\delta$	$k = Ax_\beta x_\delta \exp\left(-\frac{E}{RT}\right)\sinh\left(\Lambda^* - \frac{E_e^*}{RT}\right)$	26969.9	29.4867	n = 1, Ee*/R = 1168 K, Λ* = 2.728
$(\beta, \delta) \rightarrow f$	$k = Ax_{(\beta,\delta)} \exp\left(-\frac{E}{RT}\right)$	52700	50.0535(β), 50.1073 (δ)	
$f \rightarrow hg$	$k = Ax_f \exp\left(-\frac{E}{RT}\right)$	44300	35.0655	
$f + hg \rightarrow 2hg$	$k = Ax_f x_{hg} \exp\left(-\frac{E}{RT}\right)$	44300	38.8814	Closed-pan DSC and ODTX only
$hg + hg \rightarrow 2lg$	$k = Ax_{hg}^2 \exp\left(-\frac{E}{RT}\right)$	34100	27.5655	ODTX only

becomes difficult to cause the explosive to detonate. We added a reversible porosity model to our chemical material model, based on the solution of a gas filled spherical pore in a thick walled spherical shell. We developed an implicit form for the porosity, where we define a reference porosity ν_g^R and an actual porosity of ν_g

$$\nu_g^R = \nu_g \left(\frac{1 - \nu_g \alpha}{1 - \alpha}\right)^3 \tag{2}$$

The α is an expression of the compression of the pore and is defined by:

$$\alpha = \left(P_s - P_g\right) \Big/ \left[\frac{4\mu}{3}\left(2 + \sum_{i \in s} \frac{x_i}{\rho_i} \Big/ \sum_{i \in s} \frac{x_i}{\rho_i^0}\right)\right] \tag{3}$$

Where P_s is the solid pressure, P_g is the gas pressure, x_i is the mass fraction of the ith species, μ is the shear modulus, and ρ_i and ρ_i^0 are the density and reference density, respectively, of the ith species. Eq (2) is solved implicitity for the current porosity, subject to the yield constraint as defined by Torre[6]:

$$3\left(1 - \nu_g\right)\left(P_s - P_g\right)/2Y_s \le -\ln\left(\nu_g\right) \tag{4}$$

Where Y_s is the yield strength of the solid.

Again, one of the issues that faced Yoh et.al was the need to mass scaling to achieve long time scales. By making several modifications to ALE3D we have been able to run the problem using an implicit time step. These changes include adding soft contact between disconnected objects, added an option to calculate numerical derivatives for the material properties needed in the implicit mechanics, and added a gravity load capability to keep the central HE cylinder in contact with the walls of the STEX.

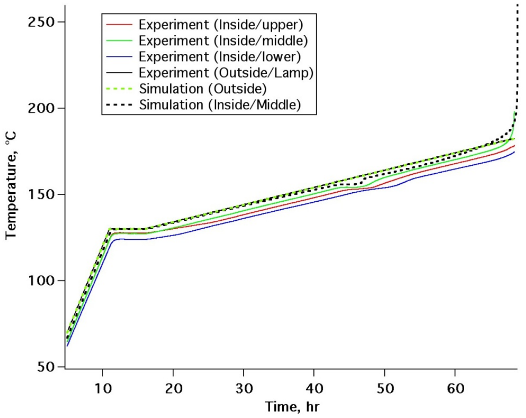

Figure 2: Comparison between experimental (solid) and modeled (dashed) temperature vs time in the TE-047 STEX experiment.

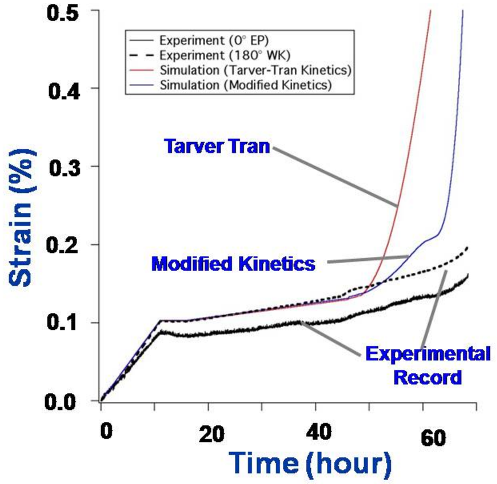

Figure 3: Comparison between model and experimental strain vs time in the TE-047 STEX experiment.

RESULTS AND DISCUSSION

Using the scheme described here, we modeled the LX-10 STEX experiment TE-047. In Fig 2. We show a comparison of the temperature vs. time between the computational model and the experimental data. The error in the time to explosion is roughly half of the previous result, and the temperature curves are much closer, with the appropriate phase change wobble that was not present in the previous model.

The strain results are shown in Fig 3. Without the porosity model, the strain goes off scale at roughly 45 hours. From this we see the importance of adding porosity to the material model. We note that the change from the Tarver-Tran kinetics to the new formulation has further improved the pressure response over the cook-off process.

One other interesting feature of the model is that the material models all failed just as the system went into thermal runaway. This is due to poor thermodynamic derivatives in that region of phase space due to CHEETAH's current inability to handle liquid-gas phase change. This fact indicates that the fluid products may not just be gasses, but that one will need to consider the phase change in the modeling process

CONCLUSIONS

We have described the kinds of improvements that have been undertaken to improve thermal explosion models of HMX-viton based systems. We believe that the porosity and implicit mechanics changes will be generally applicable to the modeling of a wide variety of composite explosives. The HMX kinetic network can be used for other HMX based explosives, but work will need to be done to incorporate the effect of the potentially reacting binder system. Finally, more work needs to be done on generating equations of state for early product gases at relatively low temperature and moderate pressure, where potential phase changes may exist.

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