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CHEMISTRY RESOLVED KINETIC FLOW MODELING OF TATB BASED EXPLOSIVES

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Abstract. Detonation waves in insensitive, TATB-based explosives are believed to have multiple time scale regimes. The initial burn rate of such explosives has a sub-microsecond time scale. However, significant late-time slow release in energy is believed to occur due to diffusion limited growth of carbon. In the intermediate time scale concentrations of product species likely change from being in equilibrium to being kinetic rate controlled. We use the thermo-chemical code CHEETAH linked to an ALE hydrodynamics code to model detonations. We term our model chemistry resolved kinetic flow, since CHEETAH tracks the time dependent concentrations of individual species in the detonation wave and calculates EOS values based on the concentrations. We present here two variants of our new rate model and comparison with hot, ambient, and cold experimental data for PBX 9502.

INTRODUCTION

While detailed chemical kinetics of detonations in homogeneous gases and liquids has been extensively studied, processes governing condensed solid insensitive energetic materials are much less understood. This is due to the higher densities, shorter reactive lengths and time scales, and more energetic nature of condensed detonations. For non-ideal explosives, such as those based on TATB, chemical reaction time scales can be comparable to the characteristic flow time scales leading to non-linear coupling between hydrodynamics and chemistry. Additionally, multi-phase products form for oxygen-deficient explosives composed primarily of C, H, N, and O (TATB has the chemical formula $C_6H_6N_6O_6$), where carbon has the possibility of forming stable solid phase detonation products.

Reactive flow modeling of a non-ideal insensitive explosive, such as those based on TATB, should consider multiple time scale rates. The initial burn rate of TATB has a sub-microsecond time scale ($<\mu s$) with the gaseous product species nearly in chemical equilibrium with each other. At intermediate time scales ($\sim\mu s$) after the onset of detonation, concentrations of gaseous product species change from being in equilibrium to being approximately frozen in value. Additionally, significant late-time slow release in energy is believed to occur due to diffusion limited growth of carbon nano-clusters ($>10\mu s$) [1].

To self-consistently model the chemical kinetics of energetic material detonations the CHEETAH [2] thermochemical code has been coupled to a multi-dimensional Arbitrary Lagrangian Eulerian (ALE) hydrodynamic code. CHEETAH is used to determine the chemical properties of the reacting energetic material and to solve the chemical kinetic rate equations. We term our model chemistry resolved kinetic flow, since we track the time dependent concentrations of individual species in the detonation wave and calculate EOS values based on their concentrations.

We model here the transformation of the high explosive into a reacting fluid of small product molecules based on a simplified chemical kinetic rate scheme, whose coefficients were determined

from fitting model results to measured detonation data. Non-rate controlled product species were assumed to be in thermodynamic equilibrium or frozen depending upon local conditions. In this paper we expand upon previous CHEETAH reactive flow modeling which used pressure power-law and Arrhenius kinetic rate laws [3-4].

REACTIVE FLOW RATES

A wide range of behavior is observed in non-ideal explosives experiments dealing with differing size, geometries, initial densities and initial temperatures. These experiments sample different regions of the phase space in the detonation wave. A good predictive detonation model should be able to reproduce observed phenomena including the value of the detonation velocity and wall velocities for metal-push experiments.

With the goal of creating a single initial temperature and initial density dependent, physically motivated model treatment for TATB based explosives, we have incorporated new rate forms into our CHEETAH reactive flow model. The model uses terms for Arrhenius ignition, pressure-dependent growth, Arrhenius bulk burn with an effective activation volume, pressure-dependent completion, and Arrhenius carbon kinetics. All rates except for the carbon kinetic term are used to treat the fast and slow chemistry of burning the initial explosive to product species. Carbon species are simplified to representative nano-carbon (C_s) and bulk detonation soot (C_b) species. Both are similar to graphite, but with higher energies. The carbon reaction is $C_s \rightarrow C_b$. C_s has higher energy than C_b due to surface bonding effects.

Our initiation rate, Eq. 1, uses Arrhenius kinetics because hot spot chemistry is driven primarily by elevated local temperatures.

$$\frac{dF}{dt} = \frac{A}{F_1}(F_1 - F)(1-F)e^{-T_O^*/T}$$
$$\frac{dF}{dt} = 0 \; F \geq F_1$$
. (1)

We consider surface reactions as being responsible for reaction growth after hot spot initiation and for completion. These effects are represented by the rate form

$$\frac{dF}{dt} = A(F - F_1)^a (1 - F_2 - F)^b (P+Q)^c$$
$$\frac{dF}{dt} = 0 \; F \leq F_1$$
$$= 0 \; F \geq 1 - F_2$$
. (2)

The reaction mechanism changes for full detonation to a form that depends on temperature, T, local pressure, P, and shock pressure, P_S

$$\frac{dF}{dt} = A(1-F)(1 - S(F_1, F_2, F))S(P_{s1}, P_{s2}, P_s)e^{-T^*(P)/T}$$
$$T^*(P) = T_0\left(1 - \tanh\left((P/P_B)\right)^m\right)$$
. (3)
$$S(x_1, x_2, x) = 6y^5 - 15y^4 + 10y^3 \; y = \frac{x - x_1}{x_2 - x_1}$$

This form represents a transition to chemistry associated with bulk reaction and nano-scale porosity. S is a switching function that smoothly varies from 0 to 1 as x varies from x_1 to x_2. The need to modify the rate from a simple power law form is supported by experiments (e.g. extrapolating the pop plot to detonation). The physical basis for form of $T^*(P)$ is the observation that very high pressure significantly reduces activation barriers. F is the HE burn mass fraction. Q is the artificial viscosity. $P + Q$ (rather than P) is used in the local pressure power law terms to optimize model performance for coarse zoning. Arrhenius kinetics are used for carbon cluster growth

$$\frac{dC_b}{dt} = A C_s e^{-T_C^*/T}$$
. (4)

Species concentrations that are not kinetic rate controlled are calculated as being in chemical equilibrium for pressures greater than a specified "freezing pressure" P_F, and are held approximately constant below this pressure.

Simplifying assumptions were made to treat initial temperature effects in our kinetics model. In this model, only the early (small F) reaction rates depend explicitly on initial temperature. The pre-factor of the reaction rate A is the only portion of the rate which depends explicitly on initial

temperature. The late part the reaction rates (large *F*) and the carbon kinetics rate do not have explicit dependence on initial temperature. The same CHEETAH EOS library is used for TATB based explosives at all temperatures. In some cases, such as to account for lot-to-lot variations, the depth of the EXP-6 potential well used in the EOS treatment of gas species is scaled by a factor close to 1. This allows the apparent C-J detonation velocity to be brought into closer agreement with experiment.

To test our CHEETAH reactive flow model we have developed a HE-validation suite. The validation suite allows for automated, systematic comparison of model simulations to historic and recent high fidelity metal push experiments at cold, ambient and hot temperatures. Current TATB based materials supported are LX-17, PBX 9502, EDC-35, and Ultrafine. The current experiments considered include hot, ambient, cold rate sticks; ambient and cold Cu cylinder tests; ambient plate push; and ambient run to detonation. The total number of experiments in suite is about 50.

We are currently considering two rate model variants, which we term our basic and advanced model. Details of the basic and enhanced modes as applied to PBX 9502 are given below.

The HE-validation suite and the models account for the following physical properties of the explosive. Changes in initial temperature result in density and part size changes and in variations in initial thermal energy. Kinetic rates change with temperature. TATB-based explosives have a fast and slow component to their HE burn reaction rates.

RESULTS AND DISCUSSION

Two rate model variants were applied to PBX 9502. The basic model uses 2 pressure power-law surface reaction rates, Eq. 2, and a carbon Arrhenius rate. This model was calibrated using 80 zones/cm. The second rate model is our advanced model which has an Arrhenius ignition rate, 2 pressure power-law rates, a full detonation rate and a carbon Arrhenius rate. This rate was calibrated using 160 zones/cm.
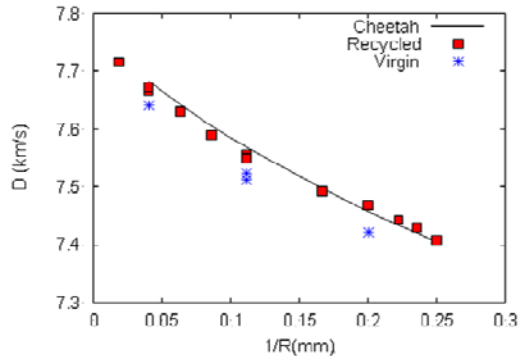
PBX 9502 modeling is complicated by there being two variants of the formulation: virgin and recycled. Recycled uses a 50/50 blend of reground HE parts and virgin. We present results here for only recycled PBX 9502. Calibrations with slight variations in parameters agree equally well with virgin results. Figure 1 shows the agreement between the calibrated basic model and size effect data [5-6] for base rate sticks. The knee in the CHEETAH results is achieved by switching from a fast to a slow surface reaction rate as the burn fraction increases. The basic model does not show failure for small rate sticks.
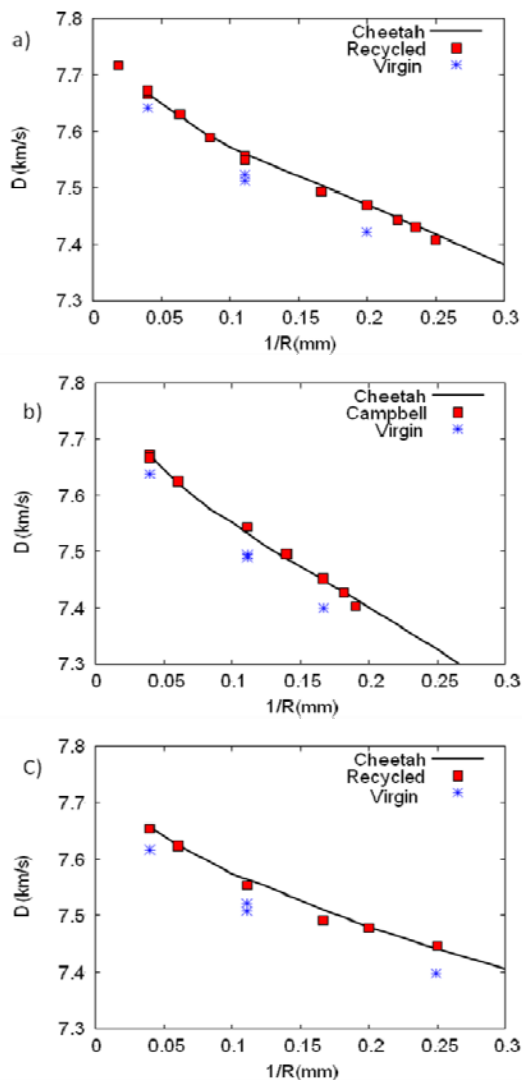
Figure 1. PBX 9502 size effect detonation velocity for (a) ambient 25C, (b) cold -55C, and (c) hot 75C [5-6]. The basic CHEETAH rate model used has been calibrated to the recycled data.
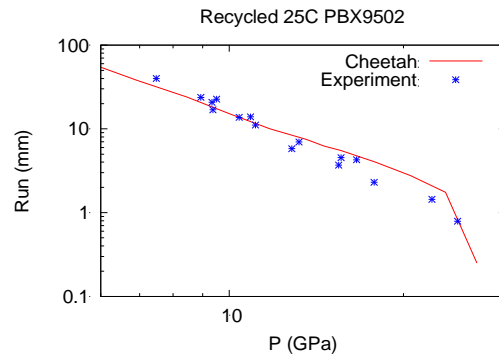
Our advanced rate model is able to treat detonation across a broader range of behavior than the basic model. Shown in Figure 2 is the ambient recycled PBX 9502 size effect curve for this model, which correctly fails below 4mm radii. The advanced model also agrees with run to detonation experimental data [7] (see Figure 3).

Figure 3. Run to detonation for ambient PBX 9502. Our model as a whole agrees with data [7].

Figure 2. Size effect detonation velocity for ambient PBX 9502 [5-6]. The CHEETAH rate model used was our advanced model calibrated to recycled PBX 9502 data.

CONCLUSIONS

Our Chemically resolved kinetic flow model has proven capable of modeling TATB based explosives at different temperatures. We have shown here that we can treat many aspects of HE behavior, including run to detonation and failure. Further calibration work needs to be done to fit all experiment types. Mesh resolved calibration also needs to be done. The CHEETAH EOS treatment for TATB explosives continues to be improved and we will be making use of new carbon and partial ionization EOS and rate models in the future.

REFERENCES

1. Viecelli, J. A. and Ree, F. H., "Carbon particle phase transformation kinetics in detonation waves," J. Appl. Phys., Vol. 88, pp. 683-690, 2000.
2. Fried, L. and Howard, M., "Cheetah 3.0 Users Manual", LLNL UCRL-MA-117541, 2001.
3. Vitello, Peter and Fried, Laurence E., "Sparse Partial Equilibrium Tables in Chemically Reactive Flow", Proceedings 13th APS Topical Conference on Sock Compression of Condensed Matter}, Portland, OR, AIP Conf. Proc. 706, 1061 (2004).
4. Vitello, P., Fried, L., Glaesemann, K. and Souers, C., "Kinetic Modeling of Slow Energy Release in Non-Ideal Carbon Rich Explosives", 13th International Detonation Symposium, Norfolk, VA, July 23-28, 2006, pp. 465-475.
5. Campbell, A. W., "Diameter Effect and Failure Diameter of a TATB-Based Explosive", PEP, Vol. 9, pp. 183-187, 1984.
6. Hill, L. G., Bdzil, J. B., Davis, W. C., and Critchfield, R. R., "PBX 9502 Front Curvature Rate Stick Data: Repeatability and the Effects of Temperature and Material Variation", 13th International Detonation Symposium, pp. 311-341, 2006.
7. Gustavsen, R. L., Sheffield, S. A., and Alcon, R.R., "Measurements of shock initiation in the tri-amino-tri-nitro-benzene based explosive PBX 9502: Wave forms from embedded gauges and comparison of four different material lots", JAP, vol. 99, pp 114907 - 114907-17, 2006.