Modeling the extreme mechanics of detonation using a Taylor Galerkin Scheme

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the date of receipt and acceptance should be inserted later

1 Abstract

Design of energetic materials is an exciting area in mechanics and materials science. Energetic composite materials are used as propellants, explosives and fuel cell components. Energy release in these materials are accompanied by extreme events: shock waves travel at typical speeds of several thousand meters per second and the peak pressures can reach hundreds of gigapascals. In this paper, we develop a reactive dynamics code for modeling detonation wave features in one such material. The key contribution in this paper is an integrated algorithm to incorporate equations of state, Arrhenius kinetics and mixing rules for particle detonation in a Taylor Galerkin finite element simulation. We show that the scheme captures the distinct features of detonation waves, and the detonation velocity compares well with experiments reported in literature.

2 Introduction

Energetic composite materials are used as propellants, explosives and fuel cell components. The rapid energy release (detonation) of these materials involve tightly coupled nonlinear interactions between chemistry and mechanics. During detonation, a shock wave is sustained by the rapid chemical energy heat release from the energetic material. These waves have extreme features; they travel at typical speeds of several thousand meters per second and the peak pressures can reach about 100 GPa[1]. Laboratory experiments are seldom equipped to handle these events. On the other hand, there is significant interest in engineering the microstructures of these energetic composites for targeted shock sensitivity and energy output. Literature in this area indicate the importance of composite features, for example, smaller energetic particles have lesser run time to detonation [2] and the time to detonation increases with the strength and content of the matrix (binder) material [3]. The first step in understanding these effects is the development of a reliable computational model of the energetic particle in these composites. This work will focus on the common energetic crystal cyclotetramethylene-tetranitramine (HMX) with a molecular formula of $C_4H_8N_8O_8$[4].

Shock strength of HMX is typically an order of magnitude higher than its yield strength. The material response of HMX under shock conditions is described by an isotropic equation of state (EOS) relating pressure, volume and energy. A variety of equations of states (EOS) have been proposed, the popular ones being the Jones-Wilkins-Lee (JWL) form[5], the Murnaghan form[6] and the Gruneisen form[7]. The Gruneisen form, with a linear shock velocity versus particle velocity Hugoniot, has been employed in several studies [7; 8; 9]. For the gaseous reaction products, by far the most popular equation of state is the JWL form that was developed by measuring the expansion velocity of metal casings surrounding HMX [10].

Modeling reactive chemistry of extreme detonation events is a significant challenge. Reaction proceeds in three general steps: dissociation, formation of reactive intermediates and exothermic recombination. Intermediate reactive species only exist for a few nanoseconds and are extremely difficult to measure experimentally. Hence, reaction kinetics are often approximated in a single step [11] that transforms a solid reactant state
to gaseous product state. These reaction schemes are tuned to experiments and chemical data such as heats of formation [12; 13; 14]. In the numerical model, an equation of state is defined for each of the reaction states and a mixing rule is defined for partially reacted states. When using Arrhenius rate models, the mixing rules ensure both pressure and temperature equilibrium between the reaction states [15]. The temperature is computed using the equation of state. For example, Gruneisen equations of state typically also include analytical expressions for the temperature. While material modeling itself is challenging in these extreme conditions, modeling the pressure discontinuities and thermal spikes during detonation are a numerical challenge in itself.

These discontinuities (shocks) arise from continuous initial conditions and are governed by the reactive Euler equations, a non-linear set of hyperbolic conservation laws. Classical formulations of solid dynamics based on displacement approach are unsuitable for this problem due to locking issues at extreme mesh distortions and numerical errors in the velocity of propagation of waves. This problem has been largely addressed in the fluid dynamics community using Riemann solvers in the context of finite volume methods [16; 17]. In the context of standard finite element methods, various stabilized methods such as Petrov Galerkin (PG) methods, Galerkin/least-squares (GLS) methods and the Taylor–Galerkin (TG) methods have been developed to address the issue. In the PG and GLS methods, a stabilization term with a coefficient is added to the weak form to act as an artificial diffusion, however, the choice of the coefficient is semi-empirical [18; 19]. The basic TG algorithm was proposed by Donea [20], Baker et al. [21] and Löhner et al. [22] for first-order systems of hyperbolic equations in which Taylor expansion in time precedes the Galerkin space discretization. TG finite element schemes are especially attractive since the diffusion arises from an improved Taylor approximation to the time derivative of the fields.

While TG algorithms have been successfully applied in areas such as pollutant transport and fluid dynamics [23; 24; 25], there do not exist a prior study of the technique for detonation of energetic particles. This paper presents two key contribution to the state–of–the–art in energetic particles simulation. Firstly, we have developed an integrated algorithm to incorporate equations of state, Arrhenius kinetics and mixing rules for HMX detonation in a finite element simulation via a Newton–Raphson method. Secondly, we have tested the one-step second-order TG scheme for modeling shock waves and detonation via a series of benchmark cases.

3 Governing Equations

3.1 Euler Equations

In detonation simulations, diffusive phenomena are neglected since pressure transfer time scales are two to three orders of magnitude faster than heat or species transfer time scales [26]. The 2-D reactive Euler equations are then given by the following equations.

\[ U_1 + (F_1)_x + (F_2)_y = S \]  
\[
U = \begin{pmatrix}
\rho \\
\rho u \\
\rho v \\
\rho E \\
\rho N_A
\end{pmatrix},
S = \begin{pmatrix}
0 \\
0 \\
0 \\
\rho N_A q Z e^{-E_f/RT} \\
-\rho N_A Z e^{-E_f/RT}
\end{pmatrix}
\]

\[ F_1 = \begin{pmatrix}
\rho u \\
\rho u^2 + p \\
\rho u v \\
\rho u N_A
\end{pmatrix},
F_2 = \begin{pmatrix}
\rho v \\
\rho v^2 + p \\
(\rho E + p) v \\
\rho v N_A
\end{pmatrix}
\]  

Here, \( \rho \) is the density, \( \rho u \) and \( \rho v \) are the momentum in the \( x \) and \( y \) directions, \( p \) is the pressure and \( \rho E \) is the total energy per unit volume. The subscripts \( x, y, \) and \( t \) denote partial derivatives. The source term \( (S) \) is based upon a one-step reaction scheme for HMX described by \( A \xrightarrow{1} B \), where \( N_A \) is the mass fraction of the unreacted explosive and \( N_B \) is the mass fraction of the gaseous reaction products. The reaction rate is given by the Arrhenius form in \( S \), where \( q \) is the heat release, \( Z \) is the static frequency factor, \( E_1 \) is the activation energy and \( R \) is the molar gas constant. The Euler equations are written in the quasi-linear form with Jacobian matrices \( A_i = \partial F_i/\partial U \). The flux vectors are linearized as \( F_i = A_i U \) for the numerical implementation.

3.2 Computational model

The material behavior is given in the form of an equation of state for the unreacted solid and the explosive products. These equations are written as a function of specific volume \( \nu \) and energy \( e \). They are related to the state variables as follows:

\[ \nu = 1/\rho, \ e = E - (1/2)(u^2 + v^2) \]  

The pressure and temperature \( (p_g, T_g) \) for a solid unreacted material is given by a linear Mie-Grüneisen EOS and those for the gaseous reaction products \( (p_g, T_g) \) is
taken to be the JWL form. The EOS equations and the model parameters can be found in Ref. [15] and is available in a more condensed form in the supplementary file accompanying this letter. For modeling a mixture of solid and gaseous states, it is assumed that the unreacted explosive and reaction products are in temperature and pressure equilibrium; i.e. $T = T_s(\nu_s, e_s) = T_g(\nu_g, e_g)$ and $p = p_s(\nu_s, e_s) = p_g(\nu_g, e_g)$. Equilibrium is enforced by iterating on $\nu_s$ and $e_s$. The following system can be solved using a Newton-Raphson method.

$$
\begin{align*}
\{ p_g - p_s \} &= \left[ \begin{array}{ccc}
\frac{\partial p_g}{\partial \nu_s} & \frac{\partial p_g}{\partial \nu_g} & \frac{\partial p_g}{\partial e_s} \\
\frac{\partial p_s}{\partial \nu_s} & \frac{\partial p_s}{\partial \nu_s} & \frac{\partial p_s}{\partial e_s} \\
\frac{\partial e_g}{\partial \nu_s} & \frac{\partial e_g}{\partial \nu_g} & \frac{\partial e_g}{\partial e_s}
\end{array} \right] \{ \nu_s \} \\
&\quad - \left[ \begin{array}{ccc}
\frac{\partial p_g}{\partial \nu_s} & \frac{\partial p_g}{\partial \nu_g} & \frac{\partial p_g}{\partial e_s} \\
\frac{\partial p_s}{\partial \nu_s} & \frac{\partial p_s}{\partial \nu_s} & \frac{\partial p_s}{\partial e_s} \\
\frac{\partial e_g}{\partial \nu_s} & \frac{\partial e_g}{\partial \nu_g} & \frac{\partial e_g}{\partial e_s}
\end{array} \right] \{ e_s \},
\end{align*}
$$

(4)

To relate the unreacted solid and reaction products, a mixture rule is used, $\nu = (1 - \lambda)\nu_s + \lambda\nu_g$ and $e = (1 - \lambda)e_s + \lambda g$. Here, $\lambda$ is the burn fraction; the mass fraction of detonation products in the mixture. For the one-step reaction in this work, $\lambda = N_{fg}$. Now, the system of equations is closed and both EOS can be expressed in terms of the solid specific volume and internal energy. Convergence is achieved when $\Delta p < 10^{-4} Mbar$ and $\Delta T < 10^{-2} K$.

The 2-D reactive Euler equations given by Eq. 1 are solved using a one-step TG scheme. This widely used time-stepping algorithm is second-order accurate, explicit and analogous to the Lax-Wendroff method [20]. Taking a Taylor series expansion of $U$ (from Eq. 2) in time,

$$
U^{n+1} = U^n + \Delta t U^n_t + \frac{1}{2} \Delta t^2 U^n_{tt} + O(\Delta t^3)
$$

(5)

where $\Delta t$ is the time step, superscripts $n + 1$ denotes the current time and $n$ denotes the previous time. The second term of the RHS of Eq. 5 is found from rearranging Eq. 1 and the third term is found by differentiating Eq. 1 with respect to time. Now Eq. 5 is written as

$$
U^{n+1} = U^n + \Delta t (S - (F_1)_s - (F_2)_y)^n + \frac{1}{2} \Delta t^2 [S_t - \{ A_1 S - A_1^2 U_x - A_1 A_2 U_y \}_x - \{ A_2 S - A_1 A_2 U_x - A_2^2 U_y \}_y]^n
$$

(6)

At each time step, the equations of state and the mixing rule is used to compute $A_i$ and the source terms. The field variables are solved using the weak form given as follows:

$$
\int \int W \left[ U^{n+1} - U^n \right] dA = \int \int W S^n dA + \frac{1}{2} \Delta t \int \int W S_t^n dA
$$

(7)

To ensure stability in regions of compression where $\partial u/\partial x < 0$ and $\partial e/\partial y < 0$, flux is computed based on a group representation. After Galerkin spatial discretization the algebraic equation $MU^{n+1} = B^n$ is obtained and solved for $U^{n+1}$. A lumping scheme is then used for the purpose of adding numerical dissipation and to give non-oscillatory solutions in the presence of steep solution gradients [27]. Here, the consistent mass matrix $M$ acting on $U^{n+1}$ is replaced by the diagonal matrix $M^L$, obtained by row sum. The smoothed solution is obtained by adding a small dissipation through parameter $d$, where $0 \leq d \leq 1$; for maximum dissipation $d = 1$. The smoothed solution is obtained as

$$
U^{smooth} = dM^{L^{-1}} B^n + (1 - d)U^{n+1}
$$

(8)

4 Numerical Results

4.1 Verification using SOD Shock and ZND models

To test the stability and accuracy of the scheme described above, the classical fluid dynamics shock tube problem is solved [27]. The test consists of two fluids at differing pressures separated by a membrane. Once the membrane is removed a rarefaction wave, contact discontinuity and shock wave is formed. The solution for an ideal gas is obtained analytically using Riemann invariants and is compared with numerical results on Figures 1. A dissipation parameter of $d = 0.03$ is used in the following simulations. The numerical results on Figures 1 show good agreement with the exact solution and the distinct characteristics of the test are captured. Next, to verify that the numerical scheme described above is accurate for reactive flow, the ZND detonation model is tested [28]. The model produces a von Neumann spike in temperature that propagates at a constant speed. The result is compared with the exact solution in Figure 2 and is in good agreement.
4.2 Reactive HMX models in 1D and 2D

Next, shock loading on partially reacted HMX is tested. Initial conditions are similar to the shock tube problem and are given by \( \rho_L = \rho_R = \rho_0 = 1.89 g/cm^3 \), \( u_L = u_R = 0 \), \( \rho E_L = 0.032 Mbar \), \( \rho E_R = 0.006 Mbar \) and \( N_{AL} = 0.2 \), \( N_{AR} = 0.8 \). Initially, the HMX on the left side of the domain contains more reaction products; i.e. \( N_B > N_A \) and has a temperature well above the ignition point. This initiates a rapid chemical reaction until the side is fully reacted. A detonation wave ensues that propagates through the right-hand side. Numerical results are obtained with linear elements for a 20cm domain with \( \Delta x = 0.1 cm \) and \( \Delta t = 10^{-2} \mu s \). Dirichlet boundary conditions are used and velocity is specified to be zero at the boundary. As reaction progresses, the initial constant density shown in Figure 3, forms a discontinuity. The gas then expands behind the shock to a value of \( \rho = 1.76 g/cm^3 \) and compresses in front to a maximum value of \( \rho = 2.08 g/cm^3 \). As the solution progresses, the gaseous product on the left side of the domain reacts quickly and becomes full gaseous after approximately \( t = 2 \mu s \). The discontinuity between fully reacted products and partially reacted HMX propagates through the specimen until the partially reacted HMX becomes fully burnt. During this reaction, temperature in the expansion region rapidly increases and a von-Neumann spike is formed at the shock front as shown in Figure 4. The spike is sustained throughout the simulation. Temperature increases to a value of \( T = 1650K \) for fully reacted HMX. At the shock front temperature increases to a peak value of \( T = 1795K \). Plots for density and temperature show distinct features: rarefaction wave, contact discontinuity and a shock wave. For a time of \( t = 8 \mu s \), the rarefaction wave is located at 6.70cm, the contact discontinuity is located at 11.35cm and the shock wave is located at 14.40cm. The discontinuity in the species mass fraction is located at the contact discontinuity. The computed shock velocity of 0.5cm/\( \mu s \) for the system falls in the range of 0.33 – 0.514cm/\( \mu s \) reported in literature for HMX particle composites [29].
The same experiment was repeated in a 2D simulation. A circular detonation front was used with the initial discontinuity located at \( r = 10.6 \text{cm} \). The combustion front is represented as a quarter of a circle that expands as the detonation proceeds. A uniform mesh with a \( \Delta x = \Delta y = 0.5 \text{cm} \) and \( \Delta t = 2.5 \times 10^{-3} \mu \text{s} \) is used. No-slip boundary conditions are considered; i.e. \( u = 0 \) at \( x = [0, 20] \text{cm} \) and \( v = 0 \) at \( y = [0, 20] \text{cm} \). Figure 5 indicates that the detonation profile maintains a symmetric spherical shape with a shock front located at approximately \( 14.4 \text{cm} \) with density changes similar to the 1D case at \( t = 8 \mu \text{s} \). Figure 6 show profiles for energy showing the von–Neumann spike.

**Fig. 5** Numerical results of density for partially reacted HMX using TG scheme \( (\text{g/cm}^3) \)

**Fig. 6** 3D plot for the energy \( (\rho E) \) in the 2D simulation \( (\text{Mbar}) \).

5 Conclusion

The key contribution in this paper is an integrated algorithm to incorporate equations of state, Arrhenius kinetics and mixing rules for HMX detonation in a Taylor Galerkin finite element simulation. We have tested the one-step second-order TG scheme for modeling detonation of HMX via benchmark cases. In the detonation model, a one-step reaction scheme was used and a linear Mie-Grüneisen and JWL equations of state were implemented for unreacted and fully reacted product respectively. Temperature and pressure equilibrium between partially reacted states was enforced with a Newton-Raphson method and averaged specific volume and energy were found using rule of mixtures. An attractive feature of the TG numerical scheme is the use of linear finite elements and simplicity in implementation. The numerical scheme was tested and agreed with exact solutions for SOD shock tube problem and the ZND detonation model. The test was repeated for partially reacted HMX and we showed that the scheme captures the distinct features of the detonation process; rarefaction wave, contact discontinuity, shock wave and the von–Neumann spike, and the detonation velocity compared well with the experimental range reported in literature. Future work will include adding the equations for a polymeric binder to simulate detonation in particular composite microstructures.

Acknowledgments

This material is based upon work supported by the National Science Foundation Graduate Research Fellowship Program Under DGE 1256260 and The Defense Threat Reduction Agency HDTRA1-13-1-0009.

References


