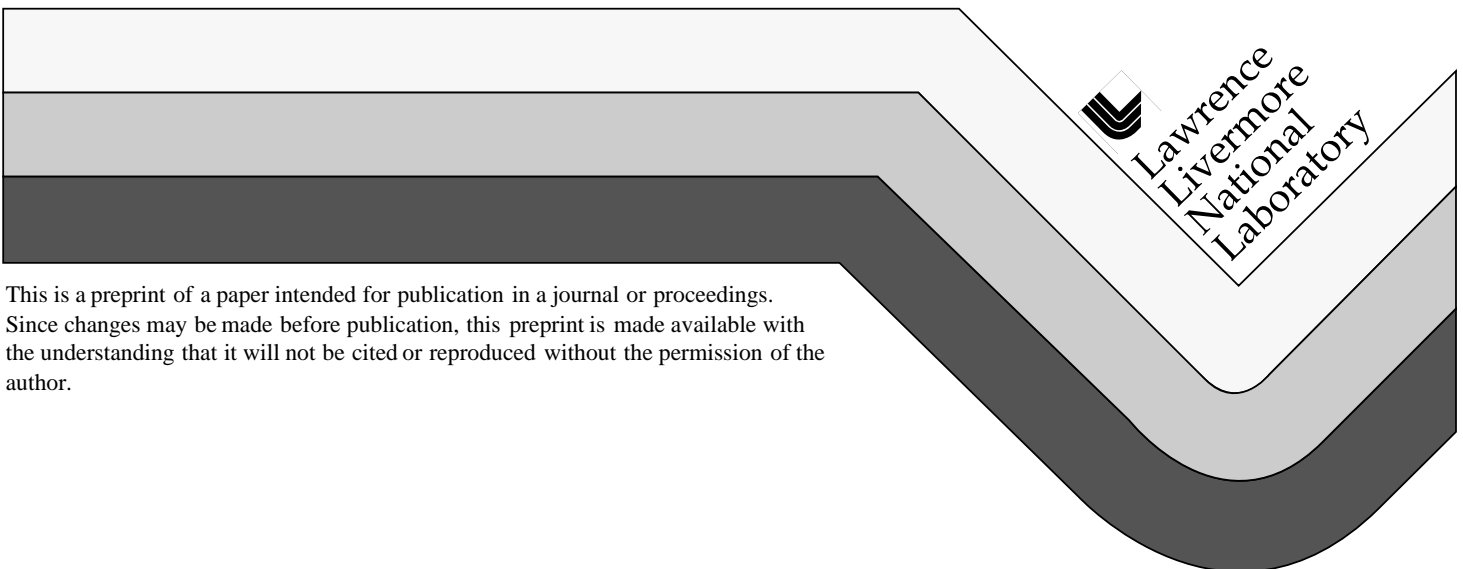


An Energy-Driven Model for HE Initiation and Burn

I. H. Zimmerman

This paper was prepared for submittal to the
1998 Nuclear Explosives Development Conference
Las Vegas, NV
October 25-30, 1998

October 1, 1998



This is a preprint of a paper intended for publication in a journal or proceedings.
Since changes may be made before publication, this preprint is made available with
the understanding that it will not be cited or reproduced without the permission of the
author.

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

An Energy-Driven Model for HE Initiation and Burn

I. Harold Zimmerman
Lawrence Livermore National Laboratory

A simple 2-state reactive flow HE burn model is described in which an approximate thermal energy is used in place of temperature to drive an Arrhenius-like rate expression. The product volume fraction and the exchange energy are determined by Newton-Raphson iteration under the twin requirements that reactant and product end up in mechanical ($P+Q$) equilibrium and that energy be rigorously conserved in the zone. The burn fraction is then adjusted by iterating the burn rate calculation. The rate expression is analytically integrable provided the rate coefficients can be taken as constant over a hydro cycle; we assume this to be true. Ignition is represented in two ways: by a void-collapse hot-spot model in porous zones and, in zones that are sufficiently energetic, by a direct-conversion reactant burn model. Neither the reactant nor the product EOS is part of the model prescription. This separates the rate law from the EOS parametrization and frees the user to choose any available EOSs to represent the reactant and product states. In particular, it is possible to model the reactant material with strength, which can be an important capability in threshold situations.

Keywords: detonation, HE burn, hot spot, ignition, reactive flow

Introduction

Reactive flow modeling of high-explosive (HE) burn is not frequently employed in large-scale numerical simulations. One reason is, that reactive flow is perceived to be mainly a research tool, a method for elucidating HE reaction rates, performance, and behavior at a high level of resolution. Another is, that reactive flow models are significantly more complex to use than the standard empirical burn models. Too often, heightened complexity reduces user confidence, and that, coupled with the increased time needed to run it, tends to leave reactive flow relegated to a dusty shelf in the user's cupboard of techniques.

Nevertheless, reactive flow HE burn models have over the years shown themselves capable of handling large-scale problems. At LLNL the ignition and growth (I&G) model (Tarver *et al.*, 1985; Lee and Tarver, 1980) has been the standard reactive flow model for nearly twenty years. At LANL the hot-spot model (Johnson *et al.*, 1985; Tang *et al.*, 1985) has been undergoing development for a comparable period. These models have reached a reasonable state of maturity. This is timely, as structures being simulated are becoming increasingly complex. Reactive-flow modeling will be needed much more often in the future than has previously been the case.

We can't completely do away with the complexity of reactive flow utilization, but I believe we can reduce it to a level where the approach will look much less formidable to users. At the same time I think it can be made more robust than is generally the case today. We can address some conceptual issues, too, so as to arrive at a method that is manifestly energy-based. We want a model that runs successfully on mm-scale zones, that is self-starting, that completely separates the rate law from the EOS, and that is consistent with the detailed conservation of energy within the zone.

The model introduced here is intended to address these issues. It divides into two parts: an (instantaneous) ignition step, and the full energy-driven burn-rate calculation. Neither of these makes any reference to temperature, in accordance with the typical (ρ, E) dependence of our hydro equations of state. During burn, reactant and product have separate artificial viscosities, so that

mechanical equilibration between reactant and product means (P+Q) equalization. Reactant and product also have separate energies. Careful consideration of the energy balance between the two components leads to a further relation that must be satisfied in order to preserve energy conservation in detail, which in turn leads to identification of the *exchange energy* E_x , a quantity that accounts for energy conveyed between reactant and product during a burn cycle.

Various considerations militate against using temperature-based kinetics in large-scale simulations. One is lack of spatial resolution. Some 4 to 10 zones are required across the HE reaction width to resolve its dynamics, and that is the level of resolution needed if the temperature calculated in a zone is to be believable. That density of zoning often is not possible, however. Even more fundamental is the difficulty of calculating a temperature in the first place. It is hard enough to obtain a thermal energy, where all one needs is the total energy and the cold compression energy. Calculating the temperature requires in addition that we have the specific heat as a function of density and energy (or density and temperature). At high energies, above 100 eV say, this may not be a problem, but where we are, in a highly kinetic situation at energies on the order of 1 eV and below, a decent specific heat is hard to define. Of course, one can introduce an approximate specific heat and arrive at a temperature by that means, assuming one has a thermal energy in hand, but I believe we do better to use the thermal energy directly. That is the approach adopted here.

The artificial viscosity Q complicates things some more by smearing the detonation over ~2-4 zones of the hydro mesh, which means that the width of the computational shock can be many times greater than the physical reaction region if the HE is zoned up to mm scale, say. This further invalidates any temperature we might assign in zones undergoing shock. For that matter it invalidates a literal interpretation of the energies we calculate. There is one thing, though, that energy has going for it: It obeys a conservation law. This proves useful in the development of the model.

There are three input parameters for the ignition stage. The subsequent burn stage features a two-term Arrhenius-like rate expression with four user input parameters. Other user input includes numerical controls, such as the equilibration convergence criterion for the product volume fraction iteration, a burn-rate convergence criterion for the burn fraction iteration, an optional time-step control, and a bound on the relative amount by which the volume fraction is allowed to change on a single iteration.

Material parameters come in through the equations of state chosen to represent reactant and product. The burn model presupposes nothing about component EOS forms.

The burn model

HE burn centers on the rate law. The form chosen here satisfies four criteria: (1) It is simple, and hence readily analyzed and modified; (2) it is analytically integrable if the rate coefficients don't change, which we assume to be true during a hydro cycle; (3) its parameters can be interpreted in reasonably physical terms; and of course (4) it is driven by thermal energy.

There are two terms in the rate expression. One, the "A" term, is unimolecular in form. It represents the direct conversion of unburned reactant that has undergone a sufficient rise in energy. This might be the result of heating, for example. The "B" term has bimolecular form and represents the rate contribution due to contact excitation of unburned reactant by burned product.

$$\frac{df_B}{dt} = A(1 - f_B) + Bf_B(1 - f_B). \quad (1)$$

The rate coefficients have an Arrhenius look to them:

$$A = \frac{1}{\tau_A} \exp\left[1 - \mathcal{E}_A / (\mathcal{E}_{\text{Reac}} - \mathcal{E}_0)\right], \quad (2a)$$

$$B = \frac{1}{\tau_B} \exp\left[1 - \mathcal{E}_B / \mathcal{E}_{\text{Prod}}\right]. \quad (2b)$$

They are not really Arrhenius expressions, even allowing for the use of thermal energy in place of temperature. For one thing, the energies that are used right now are not actually thermal. That's because we don't have a good cold-compression energy curve for the reactant and we aren't much better off in the case of the product. Once good cold-energy curves become available they should go into these formulae. For now, though, I represent the reactant cold-energy curve by a constant (\mathcal{E}_0 , the initial energy of the unburned HE) and I set the product cold energy to zero. [All energies are assumed given on a single unified scale whose reference level is defined by the convention that the product material goes to zero energy as it expands toward zero density at zero temperature.]

The other thing missing from the coefficient definitions is a thermal prefactor $\sqrt{\mathcal{E}_{\text{th}}}$. This isn't an issue if the model is always run in the same context. However, in order to address more general situations later on, we'll want to include it.

The A coefficient is zero unless the reactant "thermal" energy $\mathcal{E}_{\text{Reac}} - \mathcal{E}_0$ exceeds a user-specified reaction threshold \mathcal{E}_{RT} and Le Chatelier's principle—if reaction is to proceed, it should relieve stress in the zone—is satisfied. This threshold requirement addresses the possibility that atoms in unburned reactant might have to undergo considerable rearrangement before achieving a reactive configuration and that some level of energy is required to give them needed mobility. More prosaically, it can also be regarded as a noise limiter.

There are four other parameters to be specified, the "activation energies" \mathcal{E}_A and \mathcal{E}_B and the time constants τ_A and τ_B . These are the only parameters of the rate expression proper. A sixth datum that might be used in some variants of the full rate expression is w , the width of the reaction zone. All other input to the model applies either to the ignition stage or to numerical controls.

Assuming the rate coefficients can be considered constant during a hydro cycle, eq. (1) can be integrated exactly. If the burn fraction at time t_n is $f_{B,n}$, then the burn fraction at time $t_n + \Delta t$ is given by

$$f_{B,n+1} = \frac{A + Bf_{B,n} - A(1 - f_{B,n})\exp[-(A + B)\Delta t]}{A + Bf_{B,n} + B(1 - f_{B,n})\exp[-(A + B)\Delta t]}. \quad (3)$$

Integrability of the rate expression is especially beneficial when the burn fraction is small and is growing nonlinearly.

Ignition

Ignition can be represented by either of two mechanisms. Both are taken to be instantaneous; *ie*, they are used basically to initialize heretofore pure reactant zones for processing in the burn algorithm on subsequent cycles.

In setting up the *catastrophic void collapse* (CVC) scheme, the unburned reactant zones are given a user-specified theoretical maximum density (TMD) and a user-specified crush stress σ_{cr} . The TMD can be used with the material's initial density ρ_i to ascertain the initial void volume fraction. At the end of each hydro cycle, pure reactant zones are checked to see whether the pressure exceeds σ_{cr} . Zones meeting the criterion are changed to mixed zones by converting some reactant to burned product, enough to fill the voids and to leave the zone in internal mechanical equilibrium without changing the state of the remaining unburned reactant.

Zones that still have not initiated are then checked in the *direct reactant conversion* (DRC) algorithm to see if their energy exceeds the reaction threshold (\mathcal{E}_{RT} ; see above). Those that meet this criterion are then tested against Le Chatelier's principle to determine whether reaction is favored. If so, a converted mass fraction is calculated using the integrated rate expression with $f_{Bn}=0$ for the initial condition [see eq. (3) above]; this yields

$$f_{B,init} = A \{ 1 - \exp[-(A+B)\Delta t] \} / \{ A + B \exp[-(A+B)\Delta t] \}. \quad (4)$$

Internal equilibration is again imposed on the zone to fix the volume fraction f_V .

The newly ignited zone is ready to enter the burn algorithm on the next hydro cycle.

Conservation of energy

In this section I want to show how detailed energy conservation within the zone necessitates introduction of the exchange energy \mathcal{E}_x .

The specific energy in the zone, which I shall refer to simply as the energy, is the mass-weighted average of the component energies:

$$\mathcal{E} = (1 - f_B) \mathcal{E}_R + f_B \mathcal{E}_P. \quad (5)$$

The zonal density and its inverse can also be written in weighted form:

$$\rho = (1 - f_V) \rho_R + f_V \rho_P, \quad (6a)$$

$$\rho^{-1} = (1 - f_B) \rho_R^{-1} + f_B \rho_P^{-1}, \quad (6b)$$

where the component densities are given by

$$\rho_R = \left(\frac{1 - f_B}{1 - f_V} \right) \rho, \quad \rho_P = \frac{f_B}{f_V} \rho. \quad (7a,b)$$

Equation (5) is true at all times, and so can be differentiated:

$$\dot{\mathcal{E}} = (1 - f_B) \dot{\mathcal{E}}_R + f_B \dot{\mathcal{E}}_P + f_B (\dot{\mathcal{E}}_P - \dot{\mathcal{E}}_R). \quad (8)$$

This expression has to be equal to the derivative of the zonal energy as usually written. Neglecting contributions such as plastic work, strain energy, and the like, this is

$$\dot{\mathcal{E}} = -\Sigma \frac{d}{dt} \left(\frac{1}{\rho} \right), \quad (9)$$

where

$$\Sigma \equiv P + Q = (1 - f_V) \Sigma_R + f_V \Sigma_P. \quad (10a,b)$$

The separate component energies evolve similarly, except that there are added terms that account for energy conveyed between reactant and product wholly within the zone in consequence of the material transformation. Since this cannot be allowed to impact on the energy of the zone overall, the weighted average of these added terms must be zero. We write

$$\dot{\mathcal{E}}_R = -\Sigma_R \frac{d}{dt} \left(\frac{1}{\rho_R} \right) - \mathcal{E}_x \frac{f_B}{1 - f_B}, \quad (11a)$$

$$\dot{\mathcal{E}}_P = -\Sigma_P \frac{d}{dt} \left(\frac{1}{\rho_P} \right) + \mathcal{E}_x \frac{f_B}{f_B}. \quad (11b)$$

Then the final result is derived as follows. First, use eqs. (11) to expand the RHS of eq. (8) and then substitute the result for the LHS of eq. (9). Second, use eqs. (6b) and (10b) to expand the RHS of eq. (9). Simplifying then leaves us with the following interesting relation:

$$\frac{(\Sigma_R - \Sigma_P)}{\rho} \frac{df_V}{dt} - \left(\mathcal{E}_R + \frac{\Sigma_R}{\rho_R} - \mathcal{E}_P - \frac{\Sigma_P}{\rho_P} \right) \frac{df_B}{dt} = 0. \quad (12)$$

If we require mechanical equilibration within the zone; *ie*, if we impose the usual condition

$$\Sigma_{\mathbf{R}} = \Sigma_{\mathbf{P}}, \quad (13a)$$

then eq. (12) says that the second term also has to be zero, which means there is an additional condition that must be satisfied if energy is to be conserved in detail:

$$\mathcal{E}_{\mathbf{R}} + \frac{\Sigma_{\mathbf{R}}}{\rho_{\mathbf{R}}} = \mathcal{E}_{\mathbf{P}} + \frac{\Sigma_{\mathbf{P}}}{\rho_{\mathbf{P}}}. \quad (13b)$$

This is a key realization. It is not significantly changed by inclusion of the neglected effects mentioned above. It continues to hold in zones with more than two components. For example, if the reaction proceeds through sequential stages, as in $A \rightarrow B \rightarrow C \rightarrow \text{etc}$, then the analysis for each step follows exactly as above. More complex reaction paths lead to more complex constraints, but in any case there is a need for explicit energy exchange among participating components to account for the net energy conveyed among species by virtue of the reactions connecting them. In the present instance we have to relax \mathcal{E}_x as well as f_V in order to satisfy both of eqs. (13).

Implementation

The model is implemented in the mixed-zone package as outlined in Fig. 1 on the following page. The mixed-zone package is called on each hydro cycle (for pure zones and mixed zones alike) to calculate the zone pressure. If the material is unburned or burning HE and it is of type 1, execution passes to the control module BNTYP1. If the entire region has finished burning, however, then the type-1 burn flag is removed and control passes through the standard processing path.

A region that has not finished burning still can contain burned zones. These are updated first. Next, zones that have not yet ignited are processed. Those pure reactant zones that satisfy one or more of the ignition criteria are tagged and processed for burn starting on the next hydro cycle.

In burning zones, new burn fractions and volume fractions are first predicted by a quadratic extrapolation. The exchange energies start off with the values they had on the previous cycle. The volume fractions and exchange energies are then corrected in a 2D Newton-Raphson iteration where the two functions to be zeroed out are taken directly from eqs. (13). This leads to new component energies which changes the rate coefficients, so the burn fraction has to be corrected. This cycle repeats until the burn fraction, volume fraction, and exchange energy all have converged within their respective (user specified) tolerances, or until the burn fraction or volume fraction reaches unity.

Zonal energies, pressures, and artificial viscosities in converged zones are obtained using eqs. (5) and (10). The usual practice of iterating the EOS calculation in an explicit calculation is eliminated here in light of the fact that full convergence has already been attained. It is also proves convenient to calculate the effective zonal sound speed here at this level. If the user wishes to limit the time step to keep the change in burn fraction within a specified limit, the ability to do so is also enabled here.

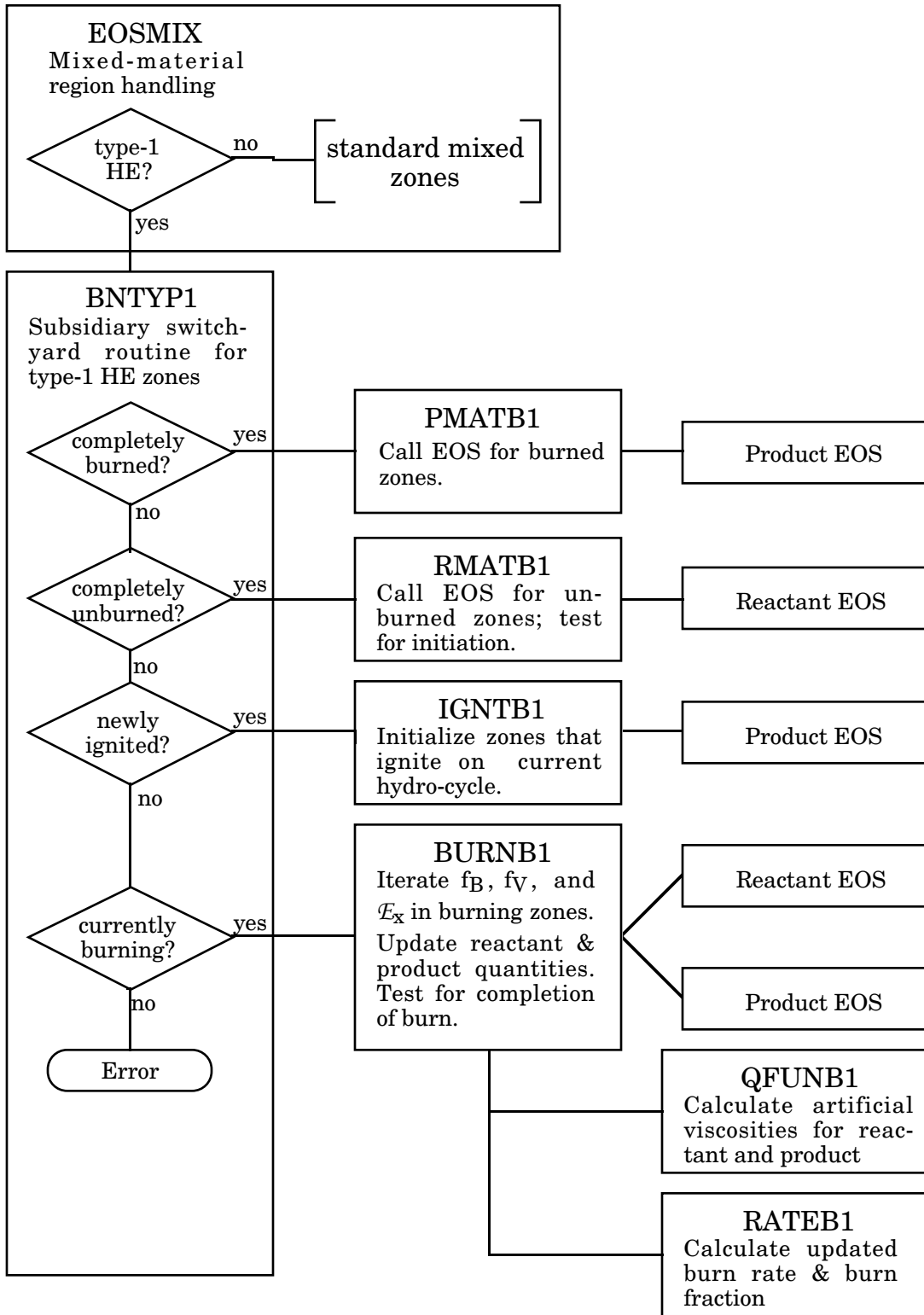


Figure 1: Logical organization of the burn scheme within the mixed-material EOS package

Variant rate expressions

The fundamental rate expression, eq. (1), takes infinitely long to finish off all the reactant. However, exigencies related to how we use our equations of state make it desirable to convert completely to product fairly quickly, before significant expansion occurs in the zone. The time expense of the iteration can be another reason to expedite the burn. One measure we might take is to declare a zone burned once f_B exceeds some fairly high criterion value. An alternative to this is to modify the rate law. I prefer the latter as providing a smoother transition to completion. Three candidate rate modifications are described here.

The first thing we consider doing is putting a floor on the reactant mass fraction that appears in the B-term of the rate expression so as to keep the rate from going to zero as the reactant is depleted:

$$f_R = \max(1 - f_B, f_{\text{floor}}), \quad (14)$$

$$\frac{df_B}{dt} \rightarrow A(1 - f_B) + Bf_B f_R. \quad (15)$$

The floor value might be adjusted through some process of successive trials so that the zone is left near a reference state, such as the CJ state, at the point where burn is complete.

Zones whose dimensions greatly exceed the reaction zone length need to burn on a time scale consistent with the shock transit time. This might be approached as follows: If the reaction zone is completely contained within the hydro zone, then some of the reactant and some of the burned product are isolated from one another. A material contact fraction f_w can be defined for the zone that records how much of the zone's material is in the presumed reaction zone. This fraction is taken to be well mixed and half-burned. None of the material in the rest of the zone contributes to the bimolecular rate term. If w is the width of the reaction zone and L_0 is the original length of the hydro zone in the direction of incidence, we then write

$$f_w = w / L_0, \quad (16)$$

$$f_{P,C} = \min(f_B, f_w / 2), \quad (17a)$$

$$f_{R,C} = f_w - f_{P,C}, \quad (17b)$$

$$\frac{df_B}{dt} \rightarrow A(1 - f_B) + Bf_{P,C} f_{R,C}. \quad (18)$$

Finally, we might simply ratchet the burn rate. A zone would be a candidate for rate ratcheting once the burn fraction had reached some criterion value well above noise, say 5%. Thereafter the rate from the previous cycle would be remembered and used on the current cycle if it exceeded the value calculated from the rate expression:

$$\frac{df_B}{dt} = \max \left[\frac{df_B(\text{old})}{dt}, \left(A + Bf_B \right) \left(1 - f_B \right) \right]. \quad (19)$$

Future directions

The model has been implemented in a 1-D code and is currently undergoing debugging. Once it is running, it will be compared with the standard Livermore I&G model (Tarver *et al.*, 1985; Lee and Tarver, 1980) to see whether it offers significant advantages with respect to run time, speed of convergence, and the like. I also want to determine how it responds to changes in the hydro. We want the new model to adapt to changes in mesh size with appreciably less fiddling than the standard model requires. I expect to have to do a considerable amount of tuning before definitive statements can be made.

Recent work (Yoo *et al.*, 1998) being done on reactant EOSs is very encouraging. The adaptability of the model to different burn scenarios will be greatly enhanced when accurate cold-compression energy curves become available. Early on, however, I will use an approximate anharmonic oscillator form such as a modified JWL energy isentrope to represent the reactant's dependence on density when $T=0$. A few such curves already exist in our data bases.

Some other modifications to the rate expression have been mentioned already. There should be a dependence in the coefficient prefactors on the appropriate thermal energies. The threshold energy for direct reactant conversion ignition and for burn via the A-term should depend on the density. Burn in large zones should be modified, perhaps by introducing a representation for the reaction zone, so as to make the result consistent with shock transit times.

Later on I want to add thermal conductivity as a means to introduce energy into the zone and thus bring about reaction. This doesn't impact the burn scheme *per se*, but it does mean that temperatures will be wanted in the reactant prior to burn. Some of our EOS data bases provide us with $(\partial T/\partial \mathcal{E})_\rho$ and $(\partial T/\partial \rho)_\mathcal{E}$, so if good reactant EOSs can be forthcoming it should be possible to get decent temperatures in the absence of shocks by integrating dT along the zone's state trajectory.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng.-48.

References

1. Johnson, J.N., Tang, P.K., and Forest, C.A., "Shock-wave initiation of heterogeneous reactive solids," *J. Appl. Phys.* **57**, 4323-4334 (1985).
2. Lee, E.L. and Tarver, C.M., "Phenomenological model of shock initiation in heterogeneous explosives," *Phys. Fluids* **23**, 2362-2372 (1980).
3. Sinz, K.H.P.H., and Morgan, D.L., *Hydro Schemes and Reactive Flow in 1D and 2D*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-ID-131619 (30 June 1998)
4. Tang, P.K., Johnson, J.N., and Forest, C.A., "Modeling Heterogeneous High Explosive Burn with an Explicit Hot-Spot Process," in *Proceedings of the Eighth Symposium (International) on Detonation*, Naval Surface Weapons Center NSWC 86-194, Albuquerque, NM (1985).
5. Tarver, C.M., Hallquist, J.O., and Erickson, L.M., "Modeling Short-Pulse Shock Initiation of Solid Explosives," in *Proceedings of the Eighth Symposium (International) on Detonation*, Naval Surface Weapons Center NSWC 86-194, Albuquerque, NM (1985).
6. Yoo, C-S., Cynn, H., Howard, W.M., and Holmes, N., "Equations of State of Unreacted High Explosives at High Pressures," preprint of an article from the *Proceedings of the Eleventh International Detonation Symposium*, Snowmass, CO, 1998; Lawrence Livermore National Laboratory, Livermore, CA, UCRL-JC-131566 (14 August 1998).