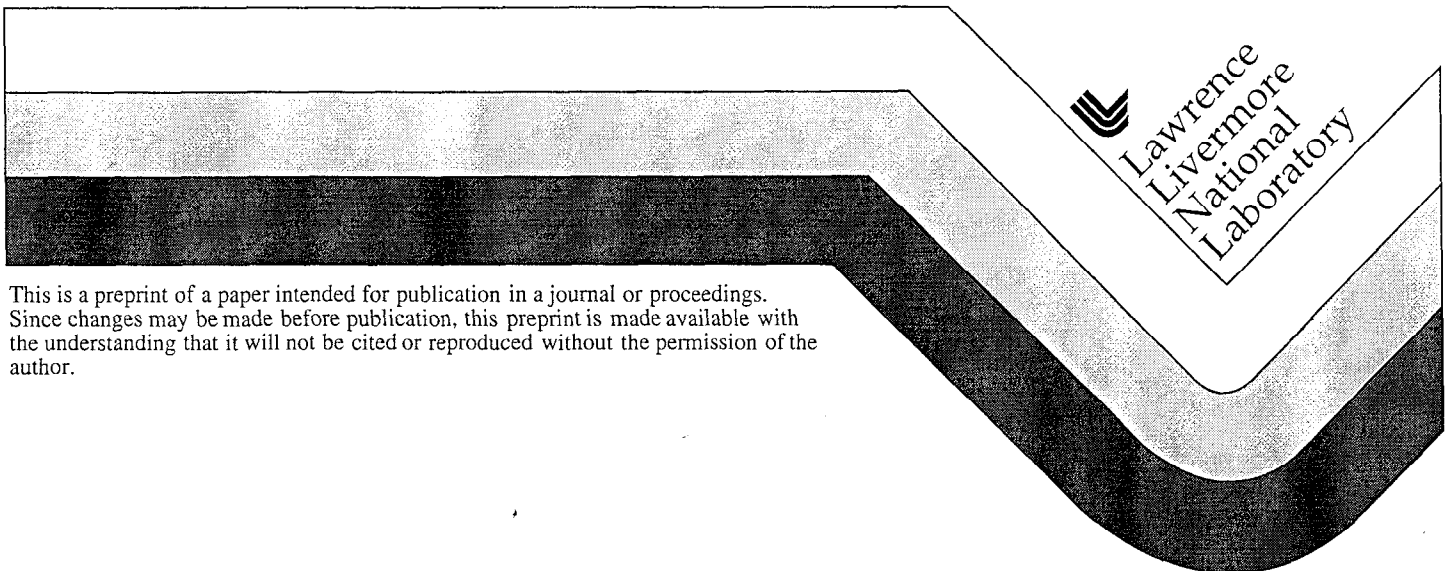


Modeling Thermally Driven Energetic Response of High Explosives in ALE3D

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MODELING THERMALLY DRIVEN ENERGETIC RESPONSE OF HIGH EXPLOSIVES

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We have improved our ability to model the response of energetic materials to thermal stimuli and the processes involved in the energetic response. Traditionally, the analyses of energetic materials have involved coupled thermal transport/chemical reaction codes. This provides only a reasonable estimate of the time and location of ensuing rapid reaction. To predict the violence of the reaction, the mechanical motion must be included in the wide range of time scales associated with the thermal hazard. The ALE3D code has been modified to assess the hazards associated with heating energetic materials in weapons by coupling to thermal transport model and chemistry models. We have developed an implicit time step option to efficiently and accurately compute the hours of heating to reaction of the energetic material. Since, on these longer time scales materials can be expected to have significant motion, it is even more important to provide high-order advection for all components, including the chemical species. We show two examples of coupled thermal/mechanical/chemical models of energetic materials in thermal environments. (U)

Keywords: thermal transport, fire, high explosives, material modeling, deflagration

Introduction

In order to understand the hazards associated with a weapon system, it is necessary to model the weapons response to a variety of conditions. The response associated with shock initiation has been well modeled with explicit hydrodynamics codes. One particular area that has not been well characterized is the response of energetic materials to an unusual thermal environment, such as a fire.

In a typical fire scenario, radiation and convection transports the heat from the fire to the exterior of the explosive device. From there, it is conducted through the outer case and then to the explosive, which begins the process of thermal decomposition. This decomposition gradually changes the material properties of the explosive. These changes range from changes in heat capacities and thermal conductivities, to changes in shear modulus, yield strength and bulk moduli, to phase changes. The change from a solid to a gas induces mass motion in the explosive confinement. Based on the strength of that confinement, the decomposition can be either slow, leading to a benign overall system response, or very fast, leading to a catastrophic event.

The characteristics of the thermal hazard environment and the type of modeling required are significantly different from those of shock initiation and propagation. First, the time scales associated with the response range from minutes to days instead of micro- to milliseconds. Second, the mechanism of energy transfer is thermal transport instead of shock propagation. Third, the change in composition is directly a function of the temperature and must be modeled as such, instead of modeling it as either a *fait-accompli* or as a pressure driven

reaction. Fourth, the process can be accompanied by relatively slow motion so that the energetic material and its confinement are subject to deformation in the elastic regime for the major portion of the response, instead of very quickly transitioning to plastic modes. Fifth, because the reactions occur slowly, the composition of the energetic material is a mixture of reactants, intermediates, and final products throughout the duration of the calculation. This is very different from detonation modeling where material is either fully unreacted or fully reacted in all but a small region of space and time. Therefore, it is more important to model the properties of the material mixture, because it is no longer the exception but rather the rule.

These characteristics required that we transform ALE3D (Sharp et al.) from a 3D ALE hydro-code into a 3D coupled thermal/chemical/mechanical code by adding several new capabilities. These include implicit thermal transport, thermally driven reactions, models for both the thermal and mechanical properties of chemical mixtures, second order species advection, and implicit hydrodynamics.

The ALE3D code

Thermal Transport. The thermal transport module in ALE3D was developed from a version of TOPAZ3D (Shapiro, 1985). The thermal transport equations are:

$$\rho C_v \frac{\partial T}{\partial t} = \nabla \cdot \underline{k} \cdot \nabla T + \dot{q}, \quad (1)$$

where T is the temperature, t is time, k is the thermal conductivity, ρ is the density, and C_v is the heat capacity at constant volume. The thermal transport equations are solved implicitly in time. In addition, the temperature derivatives of the heat capacity and heat generation terms are included in the solution, turning the solution scheme into a quasi-Newton-Raphson method.

Chemistry. The chemical reactions in ALE3D are based on the scheme that was developed in Chemical TOPAZ (Nichols, 1990). ALE3D can handle an arbitrary number of reactions and an arbitrary number of species. Each chemical reaction r is defined by the equation:

$$0 = \sum_i v_{ir} N_i \quad (2)$$

where N_i is the concentration of the i^{th} species and v_{ir} is its stoichiometry. The rate of change of a given species is given by the formula:

$$\frac{d[N_i]}{dt} = \sum_r k_r(T, P) v_{ir} \prod_j [N_j]^{\mu_{rj}} \quad (3)$$

where k is the reaction rate, and μ_{rj} is the reaction order. For most reaction forms, the reaction order is an integer like 0, 1, or 2. Several different forms are available for the reaction rate. These include a modified Arrhenius reaction formulation:

$$k_r(T, P) = A_r^0 P^{\eta_r} \exp\left(-\frac{E_r^* + PV_r^*}{k_B T}\right), \quad (4)$$

a form factor reaction formulation where non-integer reaction orders are allowed and the rate is a continuous piecewise exponential in the pressure:

$$k_r(T, P) = A_r^0 P^{\eta_r} \quad P_{i-1} \leq P \leq P_i, \quad (5)$$

and a compression ignition reaction formulation:

$$k_r(T, P) = A_r P^{n_r} \left(\frac{P}{P_0} - 1 - c \right). \quad (6)$$

These last two forms ((5),(6)) can be used to reproduce the forms of reaction developed by Lee and Tarver (Lee and Tarver, 1980) for reactive flow models.

The time integration of these chemical reaction equations is solved implicitly with a self-correcting Newton-Raphson technique.

Thermal Boundary Conditions. To model actual physical processes, it is important that the numerical boundary conditions do not interfere with the physical reality of what we are trying to model. One particular example of this is the description of a heater. It is typical to model a temperature-controlled heater in a thermal transport code as a temperature boundary condition. This works as long as the temperature of the material being heated is less than that of the heater. For explosive systems this condition is true during the initial heating phase, but is patently false from the onset of an exothermic reaction.

To account for these discrepancies, we have developed two approaches. The first one was to develop a material heat generation option based on a Proportional-Integral-Derivative (PID) thermal controller. The PID controller option requires the nodal location of an effective thermocouple. The difference between the actual temperature at the thermocouple location and the target temperature defines an error θ . The rate of energy delivery to the PID controlled elements is:

$$\dot{E}(t) = a\theta(t) + b\dot{\theta}(t) + c \int_0^t \theta(t') dt' \quad (7)$$

where a , b , c are the PID constants. The flux has both upper and lower limits. Thus, when the system becomes exothermic, the PID heat generation option simply stops adding energy to the heater elements. This form of material heat generation has been implemented and has been used to better describe the non-uniformity of the temperature field in real systems. Its one drawback is that it forces the time step into the range where the actual controller is required to operate.

The second technique we developed is a bounded boundary condition. This boundary condition will force the temperature to be equal to the target temperature only if the target temperature is greater than the current temperature. Thus, when the system goes exothermic, the boundary condition simply stops applying.

Thermal/Chemical Interaction. Chemical reactions are usually associated with changes in material properties and either absorb or release energy. In ALE3D, the energy released by the chemical reaction is based on the change of the energy between the reactants and products. This can be significantly different from specifying a predefined heat-of-reaction since the heat capacities of the two materials are not required to be identical. Simply put, in the absence of thermal diffusion, the energy in a zone must be conserved. Using a technique developed by Nichols and Westerberg (1993), we determine the amount of thermal energy that must be added to the zone to bring the total energy of the zone back to its value at the beginning of the time step.

The heat capacity for the chemical mixture is defined as the mass-weighted average of the component heat capacities. For the thermal conductivity, we include two models. The first,

associated with a uniform mixture, is a volume-weighted average of the conductivities. The second, associated with a reaction front, is a volume-weighted harmonic average of the conductivities. We use this second scheme to represent the conductivity of a mixed material that arises through advection.

Thermal/Mechanical Interactions. ALE3D accomplishes thermal/mechanical coupling by a sequence of alternating mechanical and thermal steps. The mechanical steps move the nodes while holding the entropy, S , constant. The thermal step moves heat between nodes holding the nodal locations fixed. The mechanical energy is modified by the change induced by thermal transport. Two contributions are used to influence the change in the temperature used in the thermal transport formalism during the mechanical step. The first mechanism applies to the isotropic and elastic contributions. For the isotropic contribution, we ask how does the temperature change as the volume is changed while we hold the entropy fixed. The thermodynamic derivatives that describe that process are:

$$\left(\frac{\partial T}{\partial V}\right)_s = -T\left(\frac{\partial P}{\partial E}\right)_v = -\frac{T\gamma}{V}, \quad (8)$$

where γ is the Gruniesen gamma function. The elastic stress-strain component is determined by asking the similar question: How does the temperature change while we change the material deviatoric strain $\underline{\underline{\epsilon}}$ holding the entropy fixed. The thermodynamic derivatives are:

$$\left(\frac{\partial T}{\partial \underline{\underline{\epsilon}}}\right)_s = TV\left(\frac{\partial \underline{\underline{\xi}}}{\partial E}\right)_\underline{\underline{\epsilon}} = 2TV\left(\frac{\partial \mu}{\partial E}\right)_\underline{\underline{\epsilon}}, \quad (9)$$

where $\underline{\underline{\xi}}$ is the deviatoric stress and μ is the shear modulus. These two terms are combined into one parameter ϕ which is passed from the mechanical step to the thermal step:

$$\frac{\Delta T}{T} = \phi = -\gamma\frac{\Delta V}{V} + 2V\left(\frac{\partial \mu}{\partial E}\right)_{v,\underline{\underline{\epsilon}}}\underline{\underline{\epsilon}}:\Delta\underline{\underline{\epsilon}}. \quad (10)$$

The second mechanism used to influence the temperature change is to directly add energy to the thermal equations. This mechanism is currently only used for plastic work, where we assume that all of the plastic work energy is deposited as non-reversible thermal energy.

The advantages of our ϕ method over the direct addition of energy are that it always results in a positive temperature, and that the data which is passed from the mechanical step to the thermal step is unit-less, thereby reducing the complexity that might otherwise be required. It is because of this complexity that the effect of material motion on the temperature has largely been ignored in coupled thermal/mechanical codes.

Modeling Long Time Scales. The traditional method to model the thermal response of energetic materials has been to run thermal/chemical codes, like Chemical TOPAZ. Such calculations would model the process until the chemical reaction went into thermal run-away. At that point one would transition to some form of a burn code.

ALE3D can mimic this type of coupling by the use of variable mass scaling. The principle of mass scaling is that, as long as there is no acceleration of the material, one can scale the mass without changing the results of the calculation. The adiabatic sound speed, c , is related to the density through the equation of state:

$$\rho c^2 = -V \left(\frac{\partial P}{\partial V} \right)_s, \quad (11)$$

and the Courant condition for an explicit time step is:

$$\Delta t < \Delta x / c, \quad (12)$$

where Δx is the smallest dimension in any zone. Traditionally, one does not run problems at the courant time but at some fractions of it, like 0.5. Thus, by increasing the density, we decrease the sound speed and increase the allowable time step size.

The form of variable mass scaling that we describe here changes the scaling factor as the calculation progresses. The method reduces the scaling to keep the courant time comparable to the other time scales in the problem. The other time scales currently used are the thermal stability time, the maximum allowed change in temperature, and the maximum allowed change in the composition.

As mentioned earlier, the variable mass scaling technique works as long as one can expect that there is little acceleration in the system before the energetic material reacts rapidly. For many systems this is not the case. For these systems, we have developed an implicit hydrodynamics method that replaces the standard explicit time integration scheme.

The implicit hydrodynamics method solves the mechanics problem quasi-statically in a single iteration. It is valid to use a single implicit iteration as long as there is not a significant change in the shape and compression of the material in a single step. This places a constraint on the size of the time step. The change in the location of the nodes over a time step is dependent on both the forces and their derivative at the beginning of the time step. These values create a linear set of equations that must be solved. We can solve these with either a direct or iterative matrix solver routine. The matrixes produced by the implicit hydrodynamics can be ill-conditioned and difficult to solve for certain classes of problems. Work is continuing on matrix solvers that will make this technique more robust.

Implicit-Explicit Interaction. To model the wide range of time scales involved with 'cook-off' problems, it is necessary to invoke implicit time step control for the slow processes and then transition to an explicit time step treatment for the fast time step.

From experience, we know that an implicit time step takes about a hundred times more computation time to calculate than an explicit time step. When we are running a problem that begins with only slow processes, we use the same time step controls that are in place for the variable mass scaled method. The implicit time step adds a time step constraint that no zone may change its strain by more than a user-specified value. We typically use a value of 0.001. When the time step size shrinks to less than a hundred times the courant time, the time integration method is switched from implicit to explicit. Currently, once a calculation has gone explicit we do not allow it to change back into implicit.

Species Advection. Since reaction rates depend on the exact concentration of the constituent materials, it is important that when material is advected between zones, the change in the chemical composition is accurately rendered. The chemical advection is superimposed on ALE3D's standard advection scheme: the method developed by Van Lear (1977) for pure zones and first order for mixed zones. First the volume fluxes, both pure and mixed, are calculated for each face between zones which have been allowed to advect. Based on those fluxes, all of the intensive and extensive material properties are advected. The next phase calculates the mixed material properties. When all of the materials have been processed, an

average of the mixed material zones is calculated and stored in the pure zone slot for that element. Chemical advection is then done. The chemical advection routines use the volume flux for just the chemical material. The volume flux is then used to determine the volume fraction flux for each species using second order up-wind advection. The volume flux and the volume fraction flux are then combined to create the volume flux for each species. This is used to advect the mass fractions for each species. The sum of the volume fluxes for all of the species is normalized to return the original overall volume flux. After the mass fluxes have been calculated, the new mass fractions are determined and the overall mass advection is corrected for the species effects.

Material Model for Chemical Mixtures. ALE3D currently supports only a single broad class of models for chemical mixtures. The model allows any number of species with material equations of state selected from any of the models supported in the code. The equation of state for the mixture of species is determined by equilibrating the temperature and pressure while holding the total energy and volume fixed.

Determining the strength properties of a complex mixture of materials is a more difficult. It is easy to see that the strength should depend strongly on the morphology. In ALE3D, the strength properties of the chemical mixture are determined after the temperature/pressure equilibration has been completed. Two models for the shear modulus, μ , are available. In the first, the shear modulus of the mixture is the volume fraction weighted average of the components. In the second, the reciprocal of the shear modulus is the volume fraction weighted average of the reciprocal shear moduli of the non-fluid species. In both models, the volume fractions of species with zero shear modulus are summed. When this value becomes larger than a user specified value, usually set to 40%, the mixture material is assumed to lose structural integrity, and the shear modulus is set to zero. This constraint is similar to what is done with models of sand and describes the loss of structural integrity when the amount of solid drops too small.

Burn Propagation. Once the high explosive system has ignited, one could follow the deflagration by direct numerical simulation. This would require very fine zoning and much better chemical kinetic models than we currently have available. Instead of a direct numerical simulation, we have added a front propagating capability based on level set models. To propagate the burn front with these models, one creates a field ψ that is zero at the point of ignition, and monotonically increasing away from it. One then solves:

$$\frac{\partial \psi}{\partial t} = -c(P, T, \{N_i\}) |\nabla \psi| \quad (13)$$

which is a reformulation of the wave equation. Here c is the experimentally determined burn speed of the high explosive under the appropriate conditions. The burn front is determined by finding the location where the field goes through zero. This method is similar to methods used to calculate the detonation front like the DSD (Bdzil et al., 1994) and WBL (Labourn and Swift, 1989) models. It differs in that those models may be run as a preprocessor step, since the waves they are tracking are supersonic, while this model must be run during the deflagration, since the deflagration is sub-sonic. At this stage, we are not including any of the boundary condition effects that are common in the DSD and WBL approaches.

Example Problems

We show two models of the energetic material response using ALE3D.

Sample Problem 1: The Variable Confinement Cook-off Test. We will show an example cook-off problem to illustrate these capabilities. The Variable Confinement Cookoff Test (VCCT) is a test that has been developed by Naval Surface Warfare Center as an explosive screening test. The configuration is shown in figure 1. The test fixture consists of two steel end-plates and a variable thickness steel tube. Inside the steel tube is an Aluminum tube that helps distribute the temperature uniformly within the device. A cylinder of energetic material is placed between two sets of steel washers. The purpose of the washers is to place the explosive within the uniform heating region. The washers have a hole in the middle that also provides some space for thermal expansion.

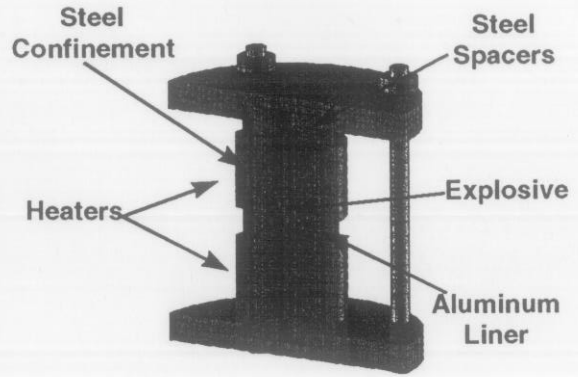


Figure 1. Initial Condition of Variable Confinement Cookoff

After an initial heat up, the heaters are used to heat up the exterior of the system at a rate of 3.3 °C/hour. The experiment continues until the confinement bursts. An experimental sequence will vary the thickness of the exterior sleeve until an explosive response is obtained.

In both of the calculations that we will show, the explosive is modeled using the most current chemical 3-step and 4-species reaction model from Tarver et al (1996). The first two species are treated as simple elastic solids with thermal expansion. The last two species are treated as a dense and light gamma-law gas respectively. All exterior surfaces are radiatively and convectively connected to the surrounding ambient temperature. The interior surfaces of the bolts are radiatively connected to the heater surfaces. The space within the washer is treated as a void material: a material whose properties are reset to their original values at the end of each step.

The VCCT results are based on a combined implicit/explicit run. These results are shown

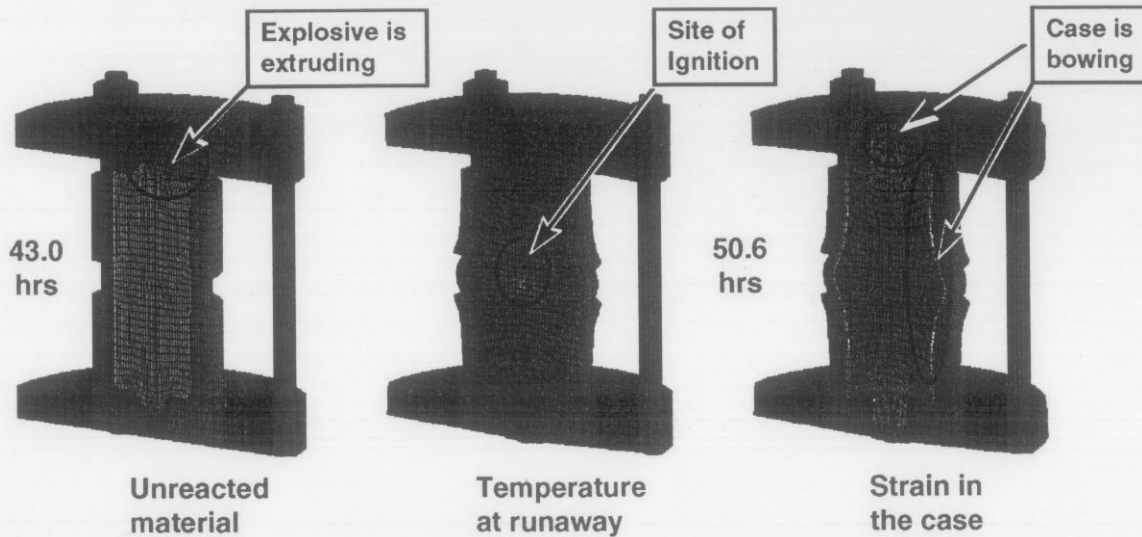


Figure 2 Typical implicit/explicit results for the VCCT test. Note that the material in the left most figure is moving within the device several hours before the violent response.

in figure 2. Note that the explosive material is moving within the fixture roughly eight hours before the violent reaction starts. The expansion into the void region is a combination of three effects. First, the explosive is decomposing and has produced a small amount of gas. Second, the explosive solid species are thermally expanding. Third, the onset of expansion is held off during the early portion of the experiment because the material strength of the solid species holds it back. However, as more gas is produced, the strength drops, letting the material flow.

An interesting point is that the time step for the problem dropped as the explosive finished filling the void space inside the washer spacers. This decrease in time step was partially due to the sudden heating of the explosive gas species by adiabatic compression and also from the implicit time step control. After the explosive has filled the void region, the time step is able to grow again.

The mass flow within the fixture long before the thermal runaway and violent response of the explosive is a result which could not be predicted by the variable mass scaling method we have described. It certainly could not be uncovered by any of the standard techniques that have been used previously. The implicit/explicit calculation predicts the temperature at which the VCCT reacted to within experimental error, and qualitatively reproduces the mechanical response seen. We currently do not have models that will predict the extent of metal fracture or pressure of HE products that would be needed to more quantitatively compare our results to experiment. We also correctly predict that without the hole in the spacer washers, the system will break before there is a violent reaction from ordinary thermal expansion.

Sample Problem 2: Deflagration in a spherical shell. Our second example shows a system where we use the deflagration model to progress the burning of the high explosive. The initial configuration is shown in figure 3. The system is a pressure vessel consisting of a 1 cm spherical shell of Aluminum surrounding a 2 cm spherical shell of HE surrounding a 1 cm spherical shell of Aluminum. The pristine HE in this system has the material properties of Aluminum, and the reaction products are treated as a gamma-law gas. A point in the middle of the explosive is ignited with a high initial temperature. This triggers the burn propagation criteria to then propagate the remainder of the user defined burn rate. For

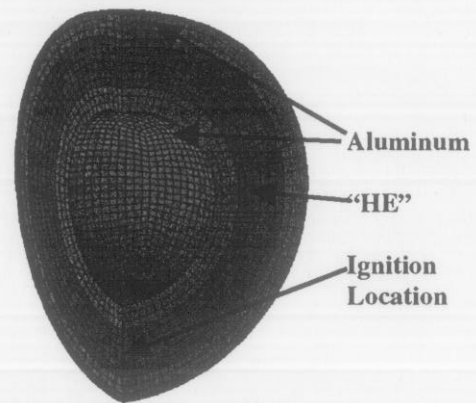


Figure 3. Initial configuration of deflagration example.

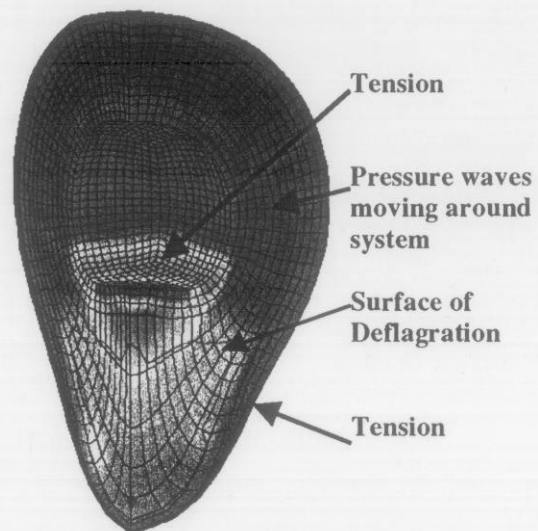


Figure 4. Late time configuration of deflagrating system. Pressure waves have time to move throughout the system.

this example, we have chosen a burn rate for the explosive of $.1 * P^{.78}$ where P is in Mbar. The system starts at an initial pressure of one atmosphere.

We show the results of our calculation in figure 4. In this system, the configuration remains unchanged for roughly 200 microseconds as the explosive burns very slowly under low pressure. As more of the explosive burns, the pressure in the vessel increase, causing the burn rate to increase according to the burn rate law. The burn rate is sufficiently slow that there is time for signals from the burning explosive to move around the entire vessel.

After roughly 200 microseconds, the pressure in the vessel is sufficiently high that the burn is too rapid for the pressure to equilibrate inside the vessel. This allows the explosive burn rate to bootstrap up. Also affecting this is the strength of the explosive material. With this particular model, the partially degraded explosive material has enough strength to resist the pressure of the decomposing explosive.

Conclusions

In this paper we have shown the variety of features added to ALE3D to calculate the response of an energetic material to thermal stimulus. The ϕ method for coupling between the mechanical and thermal steps is an important new method for coupled thermal/mechanical finite element analysis. The implicit hydrodynamics option is essential to model the very long time response of the explosive system to thermal events.

To model this response, we have chosen the route of completely coupling all of the various effects together. Although this may appear complicated, most fundamental effects are relatively simple and do not require special consideration. It is only when multiple effects are simultaneous that the effects combine to produce interesting results. Our route contrasts to other approaches that use a suite of codes to examine the behavior in different regimes. Such a methodology is appropriate when the major responses are well separated in both time and effect. The VCCT example shown here is not well separated as the early motion moves explosive material away from the heaters into a more benign region.

It is clear that there must be more work done on the material models associated with the chemically reacting mixture material. The strength of the explosive in the partially decomposed state can have a profound effect on the ensuing reaction. The higher the strength of the explosive, the more it will resist the expansion of the decomposition products. This will increase the rate at which the explosive burns, turning what could have been a benign event into a catastrophic one. Our second example problem of deflagration in a spherical vessel illustrates this effect.

The new features added to ALE3D also allow us to model several new classes of problems. The implicit hydro technique together with the thermal transport capability allows us to consider the class of problems associated with manufacturing (e.g., forging, casting, and extruding) (Couch et. al.). The time scale for these problems and the thermal effects can modify the residual stress in the material. For these systems, the residual stress can induce undesired bowing and changes in shape.

Acknowledgments

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