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COOKOFF OF ENERGETIC MATERIALS

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An overview of cookoff modeling at Sandia National laboratories is presented aimed at assessing the violence of reaction following cookoff of confined energetic materials. During cookoff, the response of energetic materials is known to involve coupled thermal/chemical/mechanical processes which induce thermal damage to the energetic material prior to the onset of ignition. These damaged states enhance shock sensitivity and lead to conditions favoring self-supported accelerated combustion. Thus, the level of violence depends on the competition between pressure buildup and stress release due to the loss of confinement. To model these complex processes, finite element-based analysis capabilities are being developed which can resolve coupled heat transfer with chemistry, quasi-static structural mechanics and dynamic response. Numerical simulations that assess the level of violence demonstrate the importance of determining material damage in pre- and post-ignition cookoff events.

INTRODUCTION*

An energetic material, subjected to an abnormal thermal environment, can undergo cookoff involving a variety of thermal, chemical and mechanical processes. Despite the years of study in cookoff phenomena, relatively little is known about the properties of thermally-degraded energetic materials. Recent work has suggested that thermal decomposition causes significant changes in the microstructure of the energetic material forming regions of porosity with high specific surface area.[1] These damaged states favor conditions for self-accelerated combustion and enhance shock sensitivity.

Prior work has centered on the onset of runaway reaction based on thermal-chemical effects with little regard for the mechanical behavior of the energetic material induced by decomposition. As such, cookoff modeling has traditionally decoupled thermal and mechanical response, *i.e.* the stress field is resolved as a separate calculation given an estimate of the thermal state. This approach provides leading order effects for the assessment of fast cookoff; however, uncoupled analysis is likely to not be valid for slow cookoff conditions.

The onset of self-sustained reaction is reasonably predicted provided accurate thermochemical, thermophysical properties and boundary conditions are defined. In the following section, an overview of cookoff modeling at Sandia National Laboratories is presented. Quasi-static response of confined energetic material is modeled using coupled thermal/chemical/mechanics analysis.

An assessment of the level of violence, after the onset of runaway reaction, is a more difficult modeling task requiring a determination of thermal damage states and combustion of thermally-degraded material. Detailed modeling of the changes in microstructure is not currently possible. Furthermore, a well defined test does not exist which can provide a definitive means for determining the level of violence. The variable confinement cookoff test[2,3] (VCCT) attempts to categorize the level reaction violence based on an average fragment size of the confinement case material after it has failed. Unfortunately, this only yields qualitative information on the strain rate behavior of the confinement material and does not provide a mechanistic understanding of cookoff of the energetic material.

In this work, cookoff in the VCCT geometry is modeled whereby various states of thermal damage (*i.e.* porosity and specific surface area) are specified and accelerated combustion is initiated at a localized source. Various states of damage lead to different levels of violence as the reaction competes with the release of confinement. These simulations strongly suggest that the determination of the thermal damage states, prior to the onset of ignition, is the key issue toward determining the degree of reaction violence associated with cookoff.

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THERMAL/CHEMICAL/MECHANICAL ANALYSIS

Predictive finite element-based analysis tools are being developed at Sandia National Laboratories to assess the level of violence including the coupled effects of heat transfer with chemistry, quasi-static structural mechanics and dynamic response.[4] Current research is focussed on determining appropriate combustion[5] and stress-strain material relationships[6] for thermally-damaged energetic materials.

Cookoff modeling requires multidimensional analysis capable of resolving phenomena spanning twelve orders of magnitude in time, from ten's of hours to sub-microseconds. An ambitious effort at Lawrence Livermore National Laboratory is underway in developing ALE3D as a single platform for cookoff modeling.[7] At Sandia National Laboratories, a different modeling strategy is taken. Existing analysis capabilities are coupled and applied to material characterization experiments to determine appropriate combustion physics models for cookoff. Finite element thermal and stress analysis are merged with shock physics analysis to determine the overall response of the degraded energetic material. The effects of dynamic combustion link quasi-static processes by coupling thermally-induced damage with dynamic combustion behavior.

During the early phases of a cookoff event, the thermal/mechanical behavior is dominated by the effects of heat transfer and quasi-static mechanics. These processes are coupled since materials expand or contract during heating and material thermal/mechanical properties change due to temperature, decomposition and phase transformation. The heat transfer code COYOTE[8] is a two-dimensional and three-dimensional massively parallelized finite element computer program designed for analysis of nonlinear heat conduction problems. In addition to solving standard thermal diffusion problems, COYOTE includes the effect of phase change, condensed-phase chemistry, and surface-to-surface radiation. Material deletion is included through the use of the finite element "death" capability. Elements can be removed to create a "dynamic" radiation enclosure. Gaps at material interfaces are modeled as contact surfaces. Material properties can be temperature- and/or species-dependent and either isotropic or orthotropic. A wide variety of boundary conditions are supported in COYOTE and pre- and post-processing file formats are used which permit integration with existing meshing and graphics visualization programs.

At conditions where the mechanical response has a characteristic time scale much greater than dictated by acoustic wave speeds and inertial effects are negligible, the mechanical response of the material is said to behave in a quasi-static fashion. The mechanics code JAS[9] is a three-dimensional massively parallelized finite element computer program designed for quasi-static, large deformation, nonlinear mechanical response of inert and reactive materials. The continuum equations are iteratively solved using either nonlinear conjugate gradient or dynamic relaxation algorithms. It has material models[10] describing elasticity, viscoelasticity, hardening plasticity, strain rate dependent behavior, deviatoric and volumetric creep, and incompressibility. Additionally, JAS has been coupled to COYOTE to determine combined thermal, chemical, and mechanical behavior of cookoff systems.[11] To describe the mechanical behavior of decomposing energetic materials, an algebraic stress-strain constitutive law has been developed using resolved temperature and species fields. Current studies are modifying these constitutive descriptions for degraded materials including the effects of phase change and rate-dependent deviatoric stress.

At the onset of self-sustained reaction, the rate of pressurization is said to behave dynamically and the mechanical response of the energetic material and confinement materials strongly interact. When the rate of pressurization produced by reaction is faster than the stress release due to expansion of confinement, pressure can build in the energetic material and the combustion may undergo the onset to deflagration to detonation transition. CTH[12] is a multimaterial, large deformation, shock physics computer program which has numerous models for various equations of state, multi-phases, elastic-viscoplasticity, porous and reactive/explosive material models. In this study, accelerated combustion, following thermal ignition, is modeled using a continuum multiphase reactive mixture model[13] implemented into CTH finite volume analysis[14].

VARIABLE CONFINEMENT COOKOFF TEST

Since thermal stresses and material decomposition cause a buildup of stress in the confinement, the microstructure of energetic materials can greatly change during cookoff. Given sufficient heating, exothermic energy release exceeds that dissipated by the effects of heat transfer and self-sustained reaction is possible. Once ignition occurs, the response of the confined energetic material becomes dynamic and the level of violence is determined by the evolution of pressure as a combustion wave sweeps through the thermally-damaged material. The competition between pressure buildup and stress release due to the loss of confinement determines the level of violence of the event. The outcome of the event greatly depends on the thermally-damaged state evolving prior to ignition.

As a representative cookoff experiment, the variable confinement cookoff test[2] (VCCT) is modeled to illustrate the level of violence predicted for slow cookoff accident scenarios. A schematic diagram of the VCCT geometry is shown in Figure 1. The test consists of a cylindrical geometry which confines the energetic material with an aluminum sleeve and a variable thickness steel sleeve. Steel witness plates at both ends provide axial confinement, and washers are used to center the energetic material between the plates. The heating for this test is provided by heater bands located on the outside of the steel confinement sleeve.

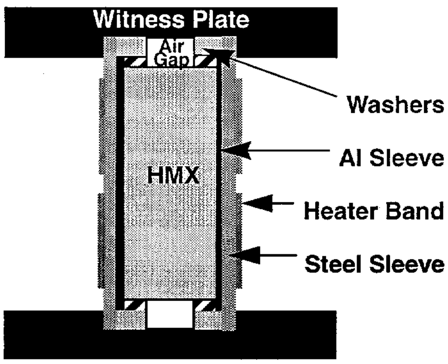

Figure 1. Variable confinement cookoff test geometry.

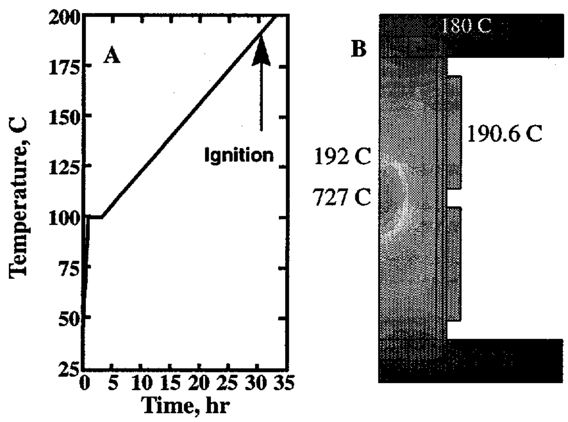

Figure 2. A) Temperature boundary condition and B) predicted thermal profile at the onset of ignition occurring at the center of the HMX.

In a typical VCCT slow cookoff test, the temperature is quickly ramped to 100 C in one hour and then the confinement is thermally-soaked for two hours at temperature. At slow cookoff conditions, heating then occurs at a rate of 3.3 C/hr until ignition. Figure 2 displays the heat transfer analysis for the VCCT geometry confining HMX. At these heating conditions, thermal ignition is predicted to occur when the temperature at the aluminum sleeve reaches ~190 C and the location of thermal runaway takes place at the center of the HMX at a time of 30 hours 27 minutes in agreement with experimental data.[3] Figure 2.B displays the predicted temperature field near the onset to self-sustained reaction. High temperatures develop at the center of the HMX with the surrounding material at much lower temperatures in the range of 180-192 C due to heat loss to the confinement.

Experimental studies[1] have suggested that as much as 10% of the energetic material decomposes prior to the onset of reaction, and the distribution of thermal damage, manifested as porosity and associated specific surface area, depends on the rate of heat transfer to the energetic material. When the heating is slow, the thermal damage of the energetic material is distributed throughout a large portion of heated material whereas during fast heating conditions localized thermal damage takes place.

At the onset of self-supported reaction, a "hot-spot" of high pressure gases spreads into adjacent material. The weak rate of combustion due to conductive burning is insufficient to support the development of shock waves; however, the effect of convective burning may lead to the onset of deflagration to detonation transition (DDT) behavior.[13]

At conditions where thermal damage is accumulated in unconnected porosity, combustion remains localized and enhanced burning takes place due to microscale flame penetration into the pores. A flame sheet model, such as that described by Schmitt, *et al.*,[15] attempts to model this phenomenon. In this approximation, the energetic material instantaneously changes from a solid to a gas across a moving interface and the local distortion of the interface due to pore penetration is treated empirically as a specific surface area correction to the normal burn rate.

When the thermal damage produces connected porosity, a different mode of accelerating combustion takes place in which rapid flame spread occurs due to convective burning. This mode of combustion is known to be the early stages of DDT[13]; the level of response of the energetic material greatly depends on the rate at which the combustion spreads in damaged energetic material and self-accelerates toward detonation.

To predict the violence of reaction during cookoff in the VCCT experiment, it is necessary to define appropriate thermal damage states that evolve prior to thermal runaway. There does not currently exist any experimental guidance of the thermal damage states; hence, the approach used here is to specify the porosity and specific surface area of thermally damaged material. In this study, HMX is

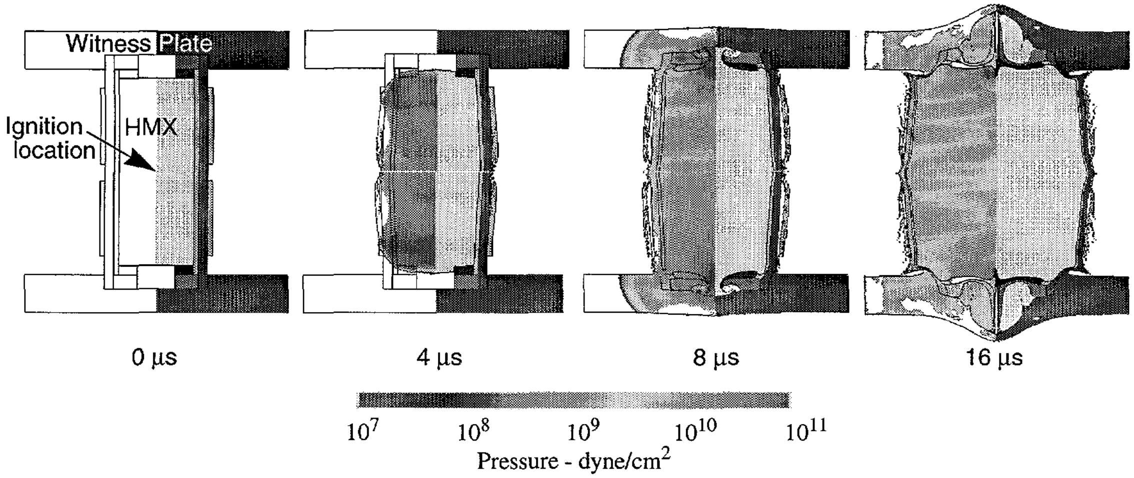

Figure 3. Several time planes of material and pressure contours during a detonation event in the VCCT geometry containing undamaged HMX.

the energetic material and burn rate and shock sensitivity are assumed to be similar to those for undamaged material.[16,17] Additionally, confinement materials are treated using elastic-perfectly plastic strength models.

As a base calculation, detonation is first considered whereby a point-initiated spherical wave spreads through undamaged HMX (100% TMD) contained in the VCCT geometry. Figure 3 presents a CTH simulation displayed at selected time planes in which materials (right) are mirrored to pressure contour fields (left). The detonation wave consumes all of the HMX in ~3.9 μs. By 8 μs, the apparatus has experienced large deformation, and breakup of the confinement begins. By 16 μs, the confinement has failed and the witness plates are breached. This case represents the most violent cookoff response since it corresponds to the highest energy release in the shortest amount of time.

In a VCCT cookoff test, the level of violence is assessed by postmortem analysis of the test apparatus. A level of violence is based upon the size and number of fragments of the confinement. Accordingly, increasing violence events are classified as burn, pressure rupture, deflagration, explosion, partial detonation, and detonation. The distinctions between categories is entirely subjective.

Although these numerical simulations indicate breakup of confinement, material fragmentation forms as a result of insertion of void in computational cells when tension states exceed a critical level. At best, this represents dynamic loading conditions near the onset of fracture of containment and a first principle resolution of breakup requires resolving to grain length scales. As traditionally done in continuum modeling, damage models are used to describe fragmentation in terms of continuum variables. In this work, the fragmentation theory developed by Grady and Kipp[18] is used in estimating the approximate number and average size of fragments based upon a calculated strain rate. The average fragment size, s, is estimated as, $s = (\sqrt{24}K/\rho c\dot{\varepsilon})^{2/3}$, where K is the fracture toughness, ρ is the density, c is the speed of sound, and $\dot{\varepsilon}$ is the local strain rate.

Table 1: Example Fragmentation Calculation

Energy Release Rate	$\dot{\varepsilon}_{hoop}$ [1/s]	s_{hoop} [mm]	$\dot{\varepsilon}_{axial}$ [1/s]	s_{axial} [mm]
D_{cj}	5.5×10^4	3.4	2×10^4	6.7
1/2 D_{cj}	4.5×10^4	3.9	5×10^3	17
1/8 D_{cj}	1.4×10^4	10.6	3×10^3	24

Lagrangian tracers are incorporated in the numerical simulations to monitor the hoop and axial stress states of the confinement. These strain rates determine the average fragment sizes in the axial and circumferential direction. To illustrate this methodology, strain-rate data is numerically obtained in cookoff simulations of detonation events. The energy release rate is varied by modifying the wave speed to one half, and one eighth of the C-J detonation velocity. In all of these simulations, 100% of the energetic

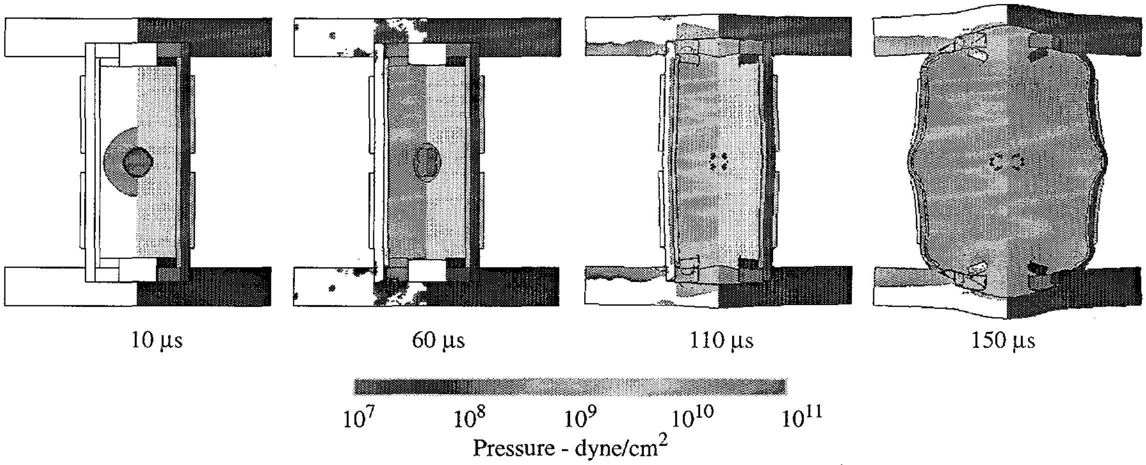

Figure 4. Selected event times of material and pressure contours during a fast deflagration event in the VCCT geometry containing 90%TMD HMX with specific surface area of 50 cm^2/cm^3.

material is consumed during the event - only the rate of energy release is modified. The calculated strain rates and average fragment size, are given in Table 1.

Many experimental studies[19,20,21] suggest that the energy release following ignition is much lower than that produced by detonation. Cookoff response is likely to induce very fast burning that can rapidly accelerate, and given enough distance, create pressure waves which coalesce into a combustion-supported shock wave. To investigate distributed combustion behavior, the CTH-DDT model of reactive multiphase mixture is used. This approach predicts self-acceleration combustion effects and the confinement responds to the combustion depressurization and/or extinguishment of reaction.

As predicted by the thermal-chemical analysis at slow cookoff conditions, the onset of reaction occurs in the interior of the energetic material. The inertia of the material acts to restrict the gas and the pressure grows until it interacts with the confinement. The effect of localized energy release is approximated as a point source of hot, high pressure gas pulse imposed at the center ignition location to trigger the dynamic event. The pressure is assumed to be representative of a constant volume explosion state.

Figure 4 displays a high speed combustion event in 90% TMD HMX containing connected porosity with specific surface area of 50 cm^2/cm^3. Following ignition, the pressure pulse initiates a deflagration which causes compaction of the porous degraded HMX to nearly 100% TMD. This compaction wave interacts with the confinement wall and after 90 μs the HMX has been pressed to fill the upper gap in the witness plate. Rapid pressurization occurs as the energetic material burns in nearly a homogeneous fashion. After 150μs the confinement has breached, thereafter, the pressure falls until reaction is quenched. In contrast to the detonation event, only 10% of the degraded HMX is consumed at the time of case failure. The associated maximum circumferential strain rate is $1.5x10^4$/s. (This implies an average fragment size of ~10mm as given in Table 1.)

Recent work has suggested that, following thermal initiation, a key process of cookoff involves the formation of large cracks in the energetic material[22]. This view is based on an experiment in which the energetic material is rapidly heated to a uniform temperature precluding the accumulation of thermal damage that would otherwise occur during slow heating. To trigger reaction, the material is preferentially-damaged by incorporating an artificial hot-wire "ignition" source. Contrary to the large cracking hypothesis, an estimate of specific surface area strongly suggest that the large cracks, by themselves, are not the key mechanism for cookoff response. There exists a wealth of prior experimental observations which provides insight on the combustion of damaged heterogeneous materials.[23,24,25]

Since the rate of pressurization is directly a measure of the rate of gas generation, an estimate of specific surface area for large scale cracks vs. distributed damage provides a basis for assessing the rate of pressure rise due to burning. Consider an energetic material with an overall length scale dimension of, L, consisting of a mixture of energetic crystals and binder with a characteristic size of, d_p, (*i.e.* a crystal diameter). Recovery observations have shown that near the onset of thermal runaway, the energetic material

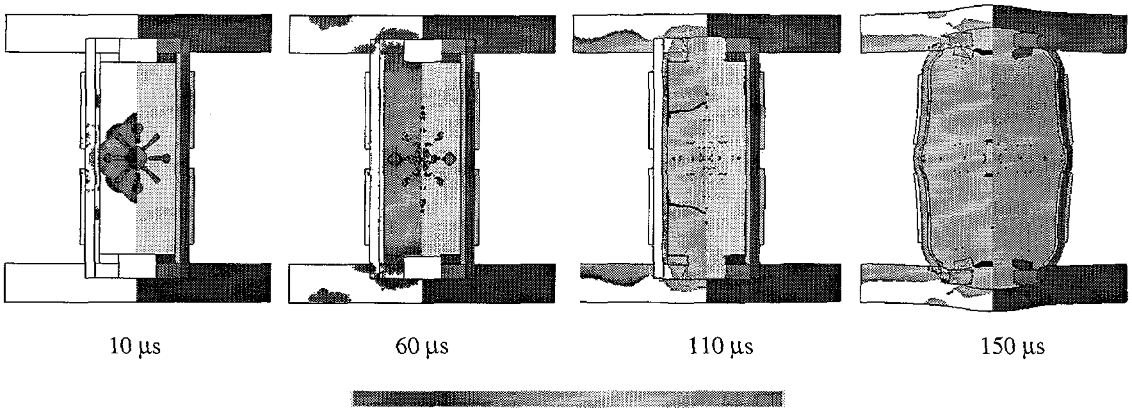

Figure 5. Selected time planes of material and pressure contours during a fast deflagration event in the VCCT geometry containing 90%TMD HMX with specific surface area of 50 cm^2/cm^3.

debonds at the grain level and defects/cracks form in the crystals.[1] To a lower bounding limit, the distributed surface area of the thermally-damaged material is estimated as: $\langle S \rangle_d = O(6\phi_s L^3/d_p)$ where ϕ_s is the solid volume fraction of the energetic material. In comparison, the surface area from N_c number of cracks having an average crack width of δ spanning the length of the total material volume is estimated by $\langle S \rangle_\delta = O(N_c L \delta)$. The ratio of surface area of microscale distributed damage to that of the large scale cracks is then estimated as:

$$\langle S \rangle_d / \langle S \rangle_\delta = (6\phi_s L^2)/(N_c d_p \delta) \cdot$$

Consider a thermally damaged material with a decomposed state of ϕ_s ~0.9 of size $L=1\ cm$ composed of energetic crystals-binder with an average diameter of d_p= 100 μm. Additionally, a damage state with $N_c=10$ cracks each having a width of $\delta = 1\ mm$ is considered. Thus, it is estimated that the ratio of distributed damage to crack surface area is of the order 10^3. Note, if the length scale of the energetic material is increased to $L \sim 10\ cm$ (typical of real systems) the surface area ratio is 10^5!

To illustrate the enhanced combustion effect due to damage specific surface area, a slow cookoff condition, similar to the prior simulation, is considered including distributed micro-damage and large-scale cracks in the energetic material. The large cracks are modeled by incorporating flow channels allowing gas flow from the ignition source. The numerical resolution is refined to sufficiently capture the flow processes in the "cracks", however, the numerical resolution is not sufficient to capture the effect of enhanced growth of surface area at the crack ends. Figure 5 displays the reacting field consistent with the time planes of the prior simulation. In this simulation, the "cracks" quickly fill with the hot high pressure gases and higher reaction rates occurs at the ends of each channel where the gas stagnates. However, as the compaction wave interacts with the confinement walls, a reflected wave closes the cracks (see time plane 60 μs). The material expands into the gap in the witness plate and pressurization to breach confinement is similar to that of the previous calculation. Roughly *10%* of the HMX has been consumed during the cookoff event. The role of the large cracks causes flame penetration in-depth to enhance convective burn with higher burning at the crack tips similar to the high speed camera observations by Kuo and colleagues.[26]

As the specific surface area is reduced, approaching that of only the crack surface area, burning of the material is significantly reduced and is incapable of sustaining accelerated combustion prior to the loss of confinement. This effect is demonstrated by reducing the specific surface area of the damage states to 5 cm^2/cm^3. Figure 6 displays several time planes after the ignition event occurs. Similar to the prior simulation, gas pressurization of the cracks first takes place, with rapid burning at the crack tips. However, the reaction is weakened as the energetic material expands into the plate gap. After 700 μs, the pressure within the VCCT geometry has dropped to less than *1 Kbar* and roughly *0.1%* of the HMX has participated in the event. To

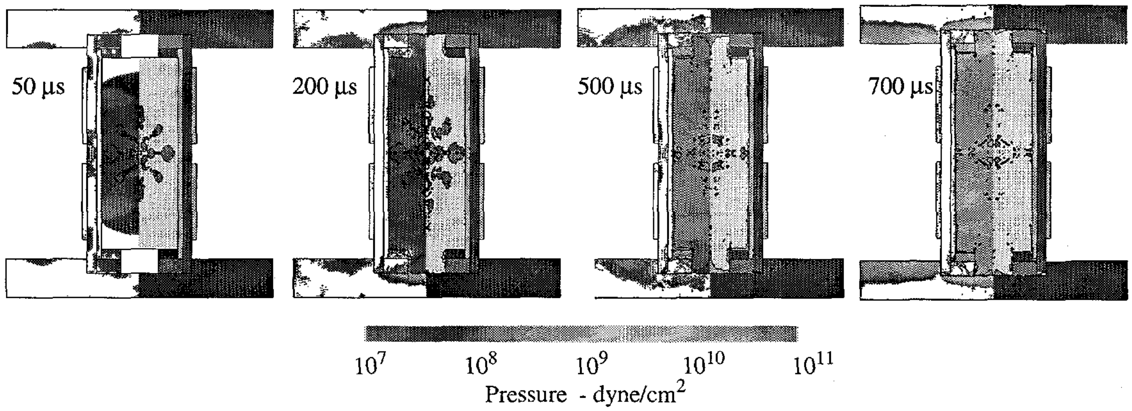

Figure 6. Selected time planes of material and pressure contours during a fast deflagration event in the VCCT geometry containing 90%TMD HMX with specific surface area of 5 cm^2/cm^3.

approach conditions such that combustion of the large cracks only takes place, the specific surface area must be further reduced by an order of magnitude lower than considered here. Clearly, it is the microscale damage state that has a more significant role in accelerating combustion. Contrary to the notion that large-scale fracture is a key to cookoff, it is the effect of the burning of distributed thermally damaged material, at the microscale, which dictates the violence of a cookoff event.

SUMMARY AND CONCLUSIONS

In this work, an overview of cookoff modeling at Sandia National Laboratories has been discussed. The cookoff modeling approach separates analysis based on quasi-static response from that of dynamic response. Post-ignition calculations of the variable confinement cookoff test emphasized the importance of the thermal damage state of the energetic material. This work has addressed the effect of specific surface area; the effect of porosity is discussed in an earlier work.[27]

This study suggests that additional material characterization work needs to be done particularly as applied to the thermally-degraded energetic material. Modeling the level of violence requires better insights into the microstructure and morphological states of the damaged material. Furthermore, improved decomposition/combustion models are needed to include phase-change effects, pressure-dependent combustion rates and the shock sensitivity of partially decomposed energetic materials.

As cookoff modeling matures, more quantitative diagnostics to probe cookoff events are required. Outcome assessments, such as those based on fragmentation characteristics, are inadequate. Instrumentation to probe the response of the confined energetic material, such as pressure or impulse[20] and stress-strain states of confinement[19], may yield better insights of cookoff behavior.

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