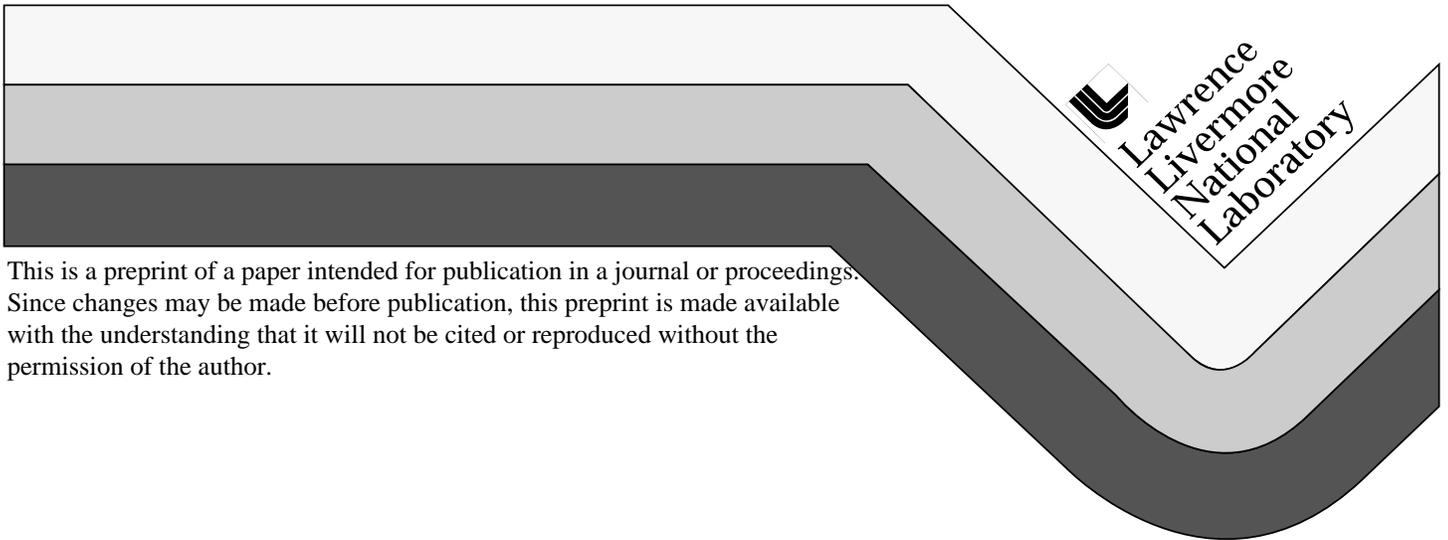


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HOT SPOT GROWTH IN A THERMAL-CHEMICAL-MECHANICAL REACTIVE FLOW MODEL FOR SHOCK INITIATION OF SOLID EXPLOSIVES

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The three dimensional Arbitrary Lagrange Eulerian hydrodynamic computer code ALE3D with fully coupled thermal-chemical-mechanical material models provides the framework for the development of a physically realistic model of shock initiation and detonation of solid explosives. The processes of hot spot formation during shock compression, subsequent ignition of reaction or failure to react, growth of reaction in individual hot spots, and coalescence of reacting hot spots during the transition to detonation can now be modeled using Arrhenius chemical kinetic rate laws and heat transfer to propagate the reactive flow. This paper discusses the growth rates of reacting hot spots in HMX and TATB and their coalescence during shock to detonation transition. Hot spot deflagration rates are found to be fast enough to consume explosive particles less than 10 μm in diameter during typical shock duration times, but larger particles must fragment and create more reactive surface area in order to be rapidly consumed.

INTRODUCTION

The Ignition and Growth reactive flow model for the shock initiation and detonation of solid high explosives has been very successful in predicting the shock initiation and detonation reaction zone properties of several explosives, especially those based on octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX) and triaminotrinitrobenzene (TATB).¹⁻⁵ Since the local temperatures in shocked, reacting solid materials can not yet be measured or calculated accurately, the reaction rate laws in Ignition and Growth and other phenomenological models⁶ are governed by the compression of the solid explosive, the geometry of the particles, and the equilibrium pressure in the reacting mixture. The Ignition and Growth reaction rate equation is:

$$\begin{aligned}
 dF/dt = & I(1-F)^b(\rho/\rho_0-1-a)^X + G_1(1-F)^cF^d p^y \\
 & (0 < F < F_{igmax}) \quad (0 < F < F_{G1max}) \\
 & + G_2(1-F)^e F^g p^z \\
 (1) \quad & (F_{G2min} < F < 1)
 \end{aligned}$$

where F is the fraction reacted, t is time, ρ is the current solid density, ρ_0 is the initial density, p is pressure in Mbars, and $a, b, x, c, d, e, g, y, z, I,$

G_1 and G_2 are constants. This rate law works well for single (sustained and short duration) shock pulses, because it models the three stages of reaction observed in shock initiation experiments (ignition of hot spots, growth of these reaction sites, and fast reaction completion). More care must be taken in modeling scenarios involving multiple shock waves in which the pressures increase much more than the temperatures in the solid particles.⁷⁻⁹ Shock initiation models are also desired which can predict the effects of the particle size and initial temperature of the explosive. Cochran and Tarver¹⁰ developed a one-dimensional statistical hot spot formation model that predicted particle size effects on shock initiation of TATB. However, that model also relied on pressure dependent reaction rates for the growth of reaction following hot spot ignition. The Ignition and Growth model is used to predict initial temperature and confinement effects when normalized to embedded pressure and/or particle velocity gauge records.¹¹⁻¹⁵

Times to thermal explosion and the subsequent violence of these explosions are calculated using standard heat transfer methods with multistep chemical kinetic mechanisms based on Arrhenius kinetics.¹⁶⁻¹⁸ With the development of large multiprocessor computers and the ALE3D

hydrodynamic computer code, these two approaches are now being combined to yield an all-Arrhenius kinetics based shock initiation and detonation reactive flow model for solid explosives. Advanced time resolved experimental techniques which can measure temperatures and species concentrations everywhere in the reactive flow process are needed. Temperature based reactive flow models can not advance very far without this experimental data.

MODEL DESCRIPTION

A detailed description of the ALE3D code and the techniques by which it calculates coupled thermal/chemical/mechanical mechanisms is given in a companion paper by Nichols et al.¹⁹ Since the local temperatures in the solid explosive are being used to propagate the chemical reactions, the equations of state for the solid and its gaseous reaction products must be more complete than those used in previous models. The details of these equations of state and other model features will be presented in detail in a longer publication.²⁰

The temperature based shock initiation model calculates the initial states produced in the solid explosive as the shock wave compresses and locally heats the porous regions by one or more of the various postulated mechanical mechanisms (rapid void collapse with material jetting, shear, friction, viscous void closure, etc.) creating high temperature sites known as "hot spots." These hot spots then either lose their thermal energy by diffusion or react exothermically. Since the exact hot spot formation mechanism which produces the most effective heating in each scenario has not yet been determined experimentally, the model is being used to test various mechanical work processes to determine which ones can produce enough heating in microseconds for shock initiation and in nanoseconds for detonation wave propagation.

The first determination that the model must make is which hot spots are hot and large enough to react exothermically before heat transfer to the surrounding cooler material lowers their temperatures. The critical conditions for impact and shock-induced hot spots in HMX and TATB have been calculated by Tarver et al.,²¹ who found that the critical temperatures for hot spot reaction

depend mainly on the chemical kinetic and heat transfer rates and the geometry of the heated region, but not on the temperature of the material surrounding the heated region.

The next step in the initiation process is the growth of reaction from these isolated ignited hot spots, and this process is the focus of this paper. It has long been known that: solid explosives with large particles are easier to ignite but ones with smaller particles grow to detonation more quickly;²² there is an average particle size that corresponds to the maximum sensitivity of a solid explosive;²³ and the surface area of the growing reaction sites must increase rapidly to account for the observed rates of buildup to detonation in shock initiation experiments.¹ This paper examines in detail the growth rates of reacting hot spots in HMX and TATB using multistep chemical kinetic models and heat conduction to determine whether this process alone can account for the decomposition rates necessary for shock initiation of solid explosives.

GROWTH OF REACTION IN HOT SPOTS

Various postulated mechanisms of hot spot formation result in different geometries and thermal conditions in and around the heated regions. Since several of these mechanisms result in spherical hot spots, which require the highest temperatures for ignition of the three simple geometries,²⁰ and since recovery experiments show that explosive particles become more spherical and somewhat smaller during shock compression, growth rates of spherical hot spots are calculated in this paper. Many of these mechanisms involve friction, shear, etc. forces that heat the outside of the explosive particle. This is particularly likely in HMX as its particles become smaller and harder with few internal voids. Lagrange analysis of embedded gauge records and Ignition and Growth modeling of relatively low pressure shock initiation experiments with HMX-based explosives have shown that the initial growth rates appear to follow the geometric factor $(1-F)^{2/3}$, where F is the fraction reacted and the ratio of 2/3 corresponds to the surface area to volume ratio of a sphere. The 1-F term corresponds in simple grain burning theory to a particle burning from the outside to the inside. Ignition of the outer

surface of an HMX particle produces hot gaseous products at temperatures up to 5000 K. Since most solid explosive charges are pressed or cast to 98-99% of their theoretical maximum densities before being compressed and heated by the shock wave, there is very little free volume into which these reaction product gases can expand and cool. In plastic bonded explosives, the particles are covered with a binder that must be ablated. After the binder is removed, the outer edge of an explosive particle can be heated by the hot gases produced by a neighboring reacting particle, and an inward moving deflagration wave will result. To estimate the shock heating of HMX and TATB particles at various shock pressures, the Ignition and Growth model uses the temperature dependent Jones-Wilkins-Lee (JWL) equation of state form:

$$p = A e^{-R_1 V} + B e^{-R_2 V} + \omega C_V T / V \quad (2)$$

where p is pressure in Megabars, V is the relative volume, T is temperature, ω is the Gruneisen coefficient, C_V is the average heat capacity, and A, B, R_1 , and R_2 are constants. More sophisticated unreacted solid explosive equations of state are being developed, with C_V defined as a function of temperature, as derived by Cowperthwaite and Shaw,²⁴ and with ω as a function of relative volume. However, this JWL form yields excellent agreement with experimental Hugoniot data and reasonable shock temperatures, because an average heat capacity between the ambient temperature and the maximum high temperature values is used. Figure 1 shows the shock temperatures calculated for HMX and TATB using the unreacted JWL equations of state for HMX (LX-10)⁴ and TATB (LX-17).⁵

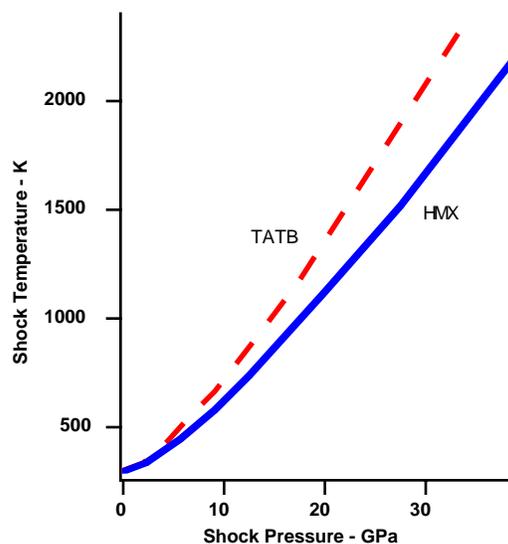


FIGURE 1. CALCULATED SHOCK TEMPERATURES VERSUS SHOCK PRESSURE FOR TATB AND HMX

The multistep chemical decomposition mechanisms, reaction rate parameters, and thermal parameters for HMX and TATB thermal modeling^{18,21} in the Chemical TOPAZ code,²⁵ which has been embedded into the hydrodynamic equations of ALE3D, are used to determine the times required for various size spherical particles to completely react. Chemical TOPAZ contains pressure dependent terms in its kinetic rate laws, but these terms were not used in this study since the effects of shock pressures on the decomposition rates of HMX and TATB are not well understood. Static high pressure has been shown to increase the times to thermal explosion of HMX.²⁶ Figure 2 shows the times required for complete reaction for spherical HMX particles at initial temperatures of 293K and 443K for several constant boundary temperatures. Figure 3 shows similar curves for three diameters of spherical TATB particles at three initial temperatures: 293K, 543K, and 673K. The calculations were ended at 200 μ s, because, even in extremely large charges, shock compression can not last that long before rarefaction waves arrive. Since HMX-based explosives initiate at much lower shock pressures than TATB-based explosives, the particle temperatures used for HMX in Fig. 2 are lower than those used for TATB in Fig. 3. Since TATB has a lower heat of reaction than HMX, the maximum

boundary temperature used for TATB is 3000K, while the maximum used for HMX is 4000K.

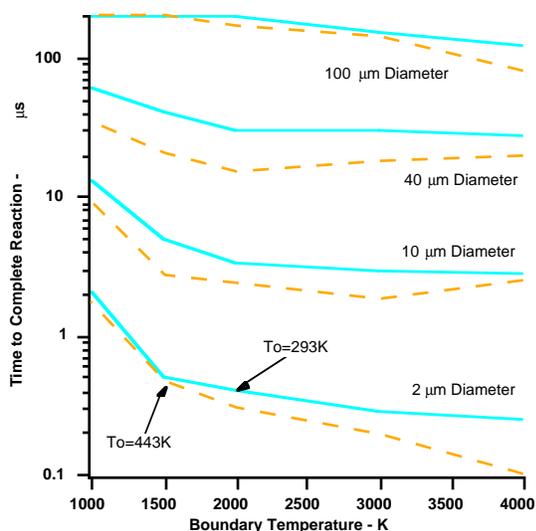


FIGURE 2. TIMES REQUIRED FOR INWARD REACTION GROWTH OF SPHERICAL HMX PARTICLES AT VARIOUS TEMPERATURES

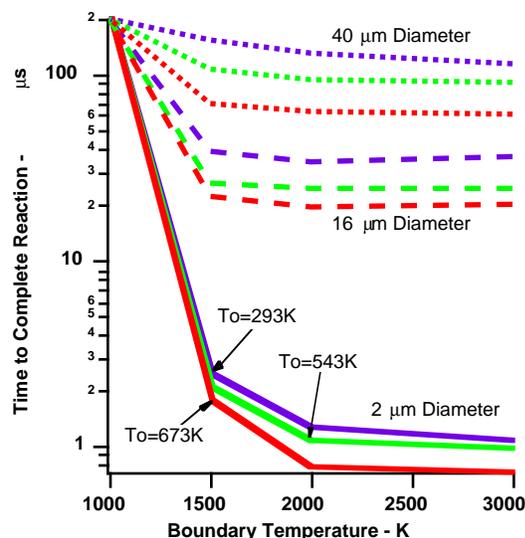


FIGURE 3. TIMES REQUIRED FOR INWARD REACTION GROWTH OF SPHERICAL TATB PARTICLES AT VARIOUS TEMPERATURES

HMX is produced in several different classes which correspond to different particle size distributions. Its particle sizes range from one μm to over 1 mm. Pressed HMX explosives usually

have a wide range of particle sizes centered around a bimodal distribution, and most of the particles are less than 100 μm in diameter. Thus HMX particles with diameters less than 100 μm are included in Fig. 2. TATB is produced in smaller particle sizes than HMX. TATB particles produced by the dry animation process are usually in the 30 - 60 μm diameter range, while the wet animation process produces some particles as large as 100 μm diameter. Ultrafine and crash precipitated TATB are produced with smaller average particle sizes of 10 μm and 0.18 μm , respectively.²⁷ TATB particles with diameters up to 40 μm are included in Fig. 3. From the calculated times shown in Figs. 2 and 3, it is clear that the smaller particles completely react within the usual time frames of shock initiation (tenths of a μs to tens of μs), while the large particles do not. For HMX, the 2 μm and the 16 μm diameter spheres reacted in less than 20 μs , especially at the elevated initial temperature of 443K, which corresponds to about 5 GPa in Fig. 1. For TATB, the 2 μm diameter particles reacted in less than 3 μs , but the 16 μm and 40 μm particles took much longer. The initial particle temperatures, 443K and 673K, correspond to shock pressures of approximately 6.5 GPa, where growth of shock induced decomposition is first observed, and 10 GPa, where growth of reaction becomes relatively fast. These calculations agree with experimental observations that smaller particles react faster.

The hot spot ignition results of Tarver et al.²¹ support the often made experimental observation that it is easier to ignite larger particles than smaller particles. There are several postulated hot spot formation mechanisms in which the interior or an edge of the explosive particle could become the ignition site. Some explosive particles, especially RDX crystals, have many occluded voids that may act as internal hot spots if compressed and ignited.²⁸ The growing reaction would then more closely resemble a spherical hot spot attempting to grow outward. In simple grain burning theory, this situation corresponds to $F^{2/3}$ for spherical geometry. To estimate the times to complete reaction for a spherical hot spot growing outward, calculations were made assuming that the spot diameter increased by a factor of 8, implying that it consumes a volume of 512 times its original size. In an idealized solid, this would correspond to 0.2% of the volume initially present as hot spots, which

is reasonable since the original 1 - 2 % void volume is compressed by a factor of 5 to 10.²³ Figures 4 and 5 shows the times to complete reaction for various hot spot diameters and particle temperatures for outwardly growing spherical hot spots in HMX and TATB, respectively. HMX particles exhibit well defined temperatures at which the time to complete reaction increases rapidly and the hot spot fails to grow at all. TATB particles grow more slowly and display less abrupt thresholds for some outward growth of the spherical hot spots. From these two sets of calculations, it is clear that the larger particles of HMX and TATB can not react completely by inward or outward deflagration in the time required for shock initiation. While not all of the explosive particles in the shock initiation region, particularly those near the initial impact surface, have to react completely to release sufficient chemical energy to cause a shock to detonation transition, most of the available chemical energy must be released or the reactive flow will fail to initiate detonation.¹

Therefore, to consume the particles fast enough, there must be other mechanisms involved in the growth process. If two or more hot spots form in very

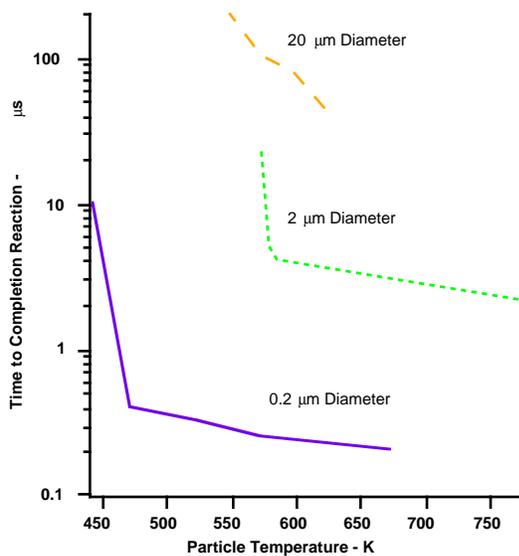


FIGURE 4. TIMES REQUIRED FOR HMX SPHERICAL HOT SPOTS TO GROW OUTWARD TO EIGHT TIMES THEIR ORIGINAL DIAMETER

close proximity, an explosive particle between them would be heated from two or more directions, thus reacting much faster. Under the high pressure, high temperature conditions produced by rapid production of hot gases, the solid particles may fragment, exposing a great deal more solid surface to hot gases. Heat transfer then causes growth of reaction in the fragmented particles and in cracks between and inside particles. Quantification of the magnitude of these increases in reactive surface area has long been one of the most difficult experimental and theoretical problems in explosive safety.²⁹

INTERACTIONS OF GROWING HOT SPOTS

To obtain some insight into the interaction of hot spot sites and the eventual coalescence of many rapidly growing hot spots at high temperatures and pressures near the region of the transition to detonation, two-dimensional calculations were run for spherical HMX and TATB particles deflagrating inward and outward using various sizes and shapes of multiple hot spots at different separation distances. An infinite number of such calculations are possible, but specific ones are being done for formation mechanisms. Heat flowing from multiple hot spots does interact and cause rapid heating and

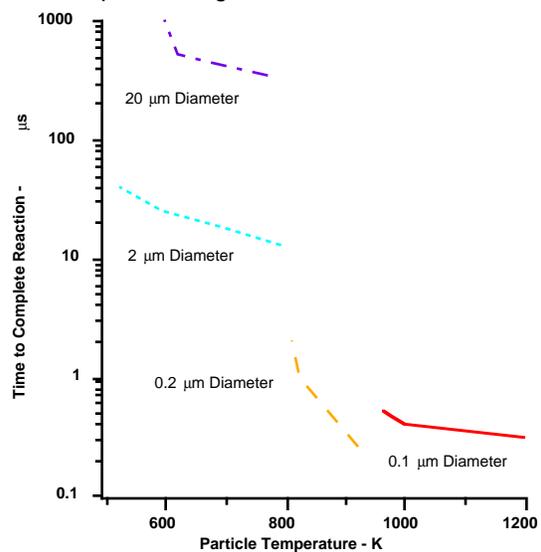


FIGURE 5. TIMES REQUIRED FOR TATB SPHERICAL HOT SPOTS TO GROW OUTWARD TO EIGHT TIMES THEIR ORIGINAL DIAMETER

decomposition in the explosive between them, provided these hot spots are large, energetic, and close enough for all of them to transfer heat to an unreacted region at similar times. The simplest example of this effect is shown in Fig. 6. Figure 6 contains the times to complete reaction for 20 μm diameter spherical 443K HMX and 673K TATB particles burning inward as functions of the number of hot spots (at temperatures of 4000K for HMX and 3000K for TATB) covering half of the outer surface area of the particle. The points for the number of hot spots equal to one assumes that the complete outer surface of the particles is heated. These times to complete reaction are 5.07 μs for HMX and 19.45 μs for TATB. When half of the surface area is covered by 8 heated regions, the times to reaction increase because these heated surfaces are quite far apart (4 μm) and coalescence of the reacted sites takes several μs . However, when 16 heated regions are used to cover approximately half of the outer surface area, they are only 2 μm apart. The reacting regions overlap rapidly, creating very high temperatures which consume the unreacted explosive in areas not covered by the initial hot spots. The times to complete reaction are less than those for particles whose complete outer surface is hot. When 32 hot spots form, they are only 1 μm apart, but they are

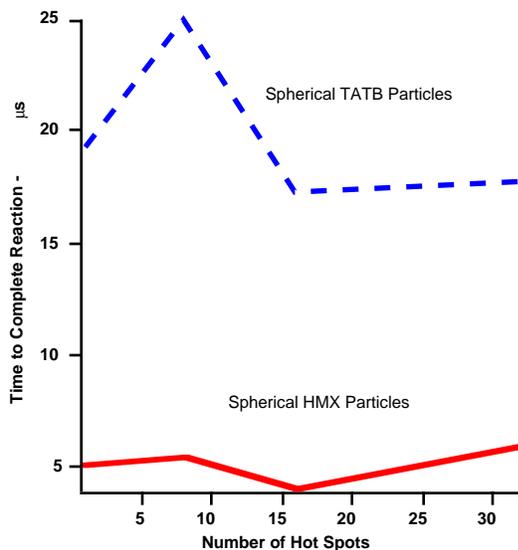


FIGURE 6. TIMES TO COMPLETE REACTION FOR 20 μm DIAMETER SPHERICAL HMX AND

TATB PARTICLES DEFLAGRATING INWARD FROM VARIOUS NUMBER OF HOT SPOT SITES

quite small and do not grow and overlap quite as rapidly as the 16 spot case. The times to complete reaction for 32 spots are greater than those for 16 spots, and for the HMX particles are longer than the reaction time when the whole surface is initially hot. This trend that faster reactions are produced by large enough and close enough multiple hot spots than for one large spot of the same volume was observed for both inward and outward burning particles. However, to date the calculated reaction times for interacting hot spots have been at most a factor of two faster than those of single spots. Therefore interactions among many rapidly growing hot spots alone do not appear to explain the rapid consumption of large explosive particles during shock initiation. Extensive fragmentation of large particles to produce more reacting surface area seems to be necessary.

SUMMARY AND CONCLUSIONS

The growth of hot spot reactions in spherical HMX and TATB particles has been studied using multistep chemical kinetic decomposition models. It was demonstrated that heat conduction alone can not account for the reaction rates in large explosive particles necessary to cause shock initiation and the transition to detonation. Small particles with diameters of 10 μm or less can be decomposed rapidly enough by either inward or outward deflagration. The interaction of several hot spots helps to transfer heat more quickly, but this mechanism alone can not account for the rapid consumption of the largest explosive particles. Therefore, in the hot, high pressure reactive flow environment of shock initiation, larger explosive particles must fragment, producing smaller, less regular particles with a great deal more surface area for the hot gaseous products to ignite. The mechanisms and extents of these fragmentation processes and the development of chemical reaction models which account for the subsequent large increases in solid surface area are the next step in this research program to build a 3D all-Arrhenius kinetics, fully coupled thermal-chemical-

mechanical shock initiation and detonation reactive flow model for solid explosives.

One of the most fascinating and important shock initiation effects that this new model can address is shock desensitization (or dead pressing) by an initial, relatively weak shock which collapses all of the voids eliminating all of the potential hot spot sites. The explosive charge then can not be initiated by a subsequent strong shock or even a detonation wave. Campbell and Travis quantitatively measured this effect in PBX 9404 and Composition B-3.³⁰ The Ignition and Growth model can prohibit detonation by a second shock wave by not allowing reaction if the explosive is ever compressed within a certain compression range using the ignition term in Eq. (1)⁷ or by using a growth rate determined by the initial shock pressure.³¹ However, as shown by Campbell and Travis, the actual process of the failure of a detonation wave in the preshocked explosive takes an amount of time which is inversely proportional to the initial shock pressure. Therefore the detonation wave continues to propagate in the preshocked material as long as the hot spots created by the first shock are forming and igniting. The measured times are similar to those required for shock initiation by short duration shock pulses.³⁰ Once the detonation wave reaches a depth in the explosive that has only fully compressed unreacted explosive and "burned out" hot spots, it no longer encounters the voids and reacting hot spot sites it needs to propagate. Correctly predicting shock desensitization is a major challenge for this new reactive flow model.

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