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ABSTRACT

Recent experimental and theoretical advances in understanding energy transfer and chemical kinetics have led to improved models of detonation waves in solid explosives. The Nonequilibrium Zeldovich - von Neumann - Doring (NEZND) model is supported by picosecond laser experiments and molecular dynamics simulations of the multiphonon up-pumping and internal vibrational energy redistribution (IVR) processes by which the unreacted explosive molecules are excited to the transition state(s) preceding reaction behind the leading shock front(s). High temperature, high density transition state theory calculates the induction times measured by laser interferometric techniques. Exothermic chain reactions form product gases in highly excited vibrational states, which have been demonstrated to rapidly equilibrate via supercollisions. Embedded gauge and Fabry-Perot techniques measure the rates of reaction product expansion as thermal and chemical equilibrium is approached. Detonation reaction zone lengths in carbon-rich condensed phase explosives depend on the relatively slow formation of solid graphite or diamond. The Ignition and Growth reactive flow model based on pressure dependent reaction rates and Jones-

Wilkins-Lee (JWL) equations of state has reproduced this nanosecond time resolved experimental data and thus has yielded accurate average reaction zone descriptions in one-, two- and three-dimensional hydrodynamic code calculations. The next generation reactive flow model requires improved equations of state and temperature dependent chemical kinetics. Such a model is being developed for the ALE3D hydrodynamic code, in which heat transfer and Arrhenius kinetics are intimately linked to the hydrodynamics.

1. Nonequilibrium Zeldovich-von Neumann-Doring (NEZND) Theory

The Nonequilibrium Zeldovich-von Neumann-Doring (NEZND) theory of a self-sustaining detonation wave[1-6] was developed as a framework in which to study the major chemical and physical processes that precede and follow exothermic chemical reaction. These nonequilibrium processes determine the time required for the onset of chemical reaction, control the energy release rates, and supply the mechanism by which the chemical energy sustains the leading shock wave front. The three-dimensional shock wave front structure, the nonequilibrium excitation and relaxation processes, and the chemical reaction rates in gaseous detonation waves are fairly well understood.[7] However, the high pressures (20-40 GPa), densities (2.5 g/cm^3), and temperatures (3000-5000K) generated in less than a microsecond in condensed phase detonation waves traveling at velocities approaching 10 mm/μs create environments that are difficult to study experimentally and theoretically.

Figure 1 shows the four main regions of the NEZND reaction zone for a detonation wave in a solid or liquid explosive containing carbon, hydrogen, oxygen, and nitrogen atoms ($C_wH_xO_yN_z$). The first region is the leading shock wave front, which consists of a three-dimensional arrangement of Mach stem interactions. The familiar cellular patterns of gaseous detonation wave fronts have been observed on a much smaller spatial scale in homogeneous liquid explosives.[8] The shock front in detonating heterogeneous solid explosives is more complex than those in homogeneous liquids or perfect single crystals, because the presence of voids, grain boundaries, and internal crystal flaws creates an irregular shock compression process.[5] The explosive is accelerated, and its phonon modes are excited to a high quasitemperature within a picosecond.[9]

Following shock front compression, the second region in Fig. 1 is dominated by the flow of this excess phonon energy into the low frequency vibrational modes of the molecule by "multiphonon up-pumping" and the subsequent energy flow from the low frequency modes to the high frequency modes by intramolecular vibrational energy redistribution (IVR). Recent experimental measurements and molecular dynamic simulations of these processes were discussed by Tarver.[6] Ten to a hundred picoseconds are required for complete vibrational relaxation. The establishment of vibrational equilibration behind the leading shock front is a necessary condition for chemical decomposition, because the initial bond breaking reaction proceeds through a transition state created by high vibrational excitation of one of the highest frequency modes.

The third region of Fig. 1 begins at the internally equilibrated transition state (or states), which is followed by the chemical reconstitution process in which the large organic molecules eventually produce several stable reaction product molecules. Little is known about this region which is referred to as the "von Neumann spike" or "chemical peak" in condensed phase explosives literature, because spikes or peaks in pressure or particle velocity are observed in or inferred from hydrodynamic experiments. However, recent experimental and theoretical research has yielded some information about the state which precedes the exothermic chemical reaction. Sheffield[10] has observed using laser interferometry a plateau in particle velocity preceding product expansion that lasts several nanoseconds in detonating nitromethane. Spectroscopic studies of nitromethane shocked to pressures approaching those in detonation waves have measured the rates of disappearance of some nitromethane peaks and the appearance of some reaction product peaks in the submicrosecond time frame.[11] The unreacted Hugoniot states calculated[3] using realistic high pressure, high temperature equation of state assumptions[12] agree with those measured by nanosecond time resolution probes.[13-15] Using these calculated unreacted states, Tarver[16] demonstrated that measured induction times for the onset of exothermic reaction in detonation and high pressure shock initiation experiments on homogeneous solid and liquid explosives can be calculated using high temperature, high density transition state theory with the rate constant K expressed by:

$$K = (kT/h)\, e^{-E/\kappa} \sum_{i=0}^{s-1} (E/RT)^{i}\, e^{-E/RT}/\, i! \qquad (1)$$

where E is the activation energy; T is the equilibrated temperature; k, h, and R are Boltzmann's, Planck's, and the gas constant, respectively; and κ is the average vibrational energy and s is the number of the vibrational modes interacting with the dissociation mode. When the total energy in these vibrational modes equals the activation energy, $\kappa = E/s$, and Eq. (1) becomes:

$$K = (kT/h)\, e^{-s} \sum_{i=0}^{s-1} (E/RT)^{i}\, e^{-E/RT} / \, i! \qquad (2)$$

Once the exothermic chain reaction process begins, highly vibrationally excited product molecules form and interact with the transition states and each other to greatly increase the rates of decomposition. Recently "supercollisions," in which large amounts of vibrational energy are transferred between highly excited molecules in gas phase collisions, were discovered.[17] Similar supercollisions are likely to dominate the energy transfer in the dense mixture of highly vibrationally excited detonation products.[6] The fourth region in Fig. 1 is dominated by the expansion and vibrational deexcitation of the stable reaction products plus the diffusion controlled formation of solid products, such as carbon particles in underoxidized explosives. Experimental techniques with nanosecond time resolution for measuring pressure,[18] particle velocity,[19] and temperature[20] have yielded average values for these parameters as the Chapman-Jouguet (C-J) state of thermal and chemical equilibrium is approached in several solid and liquid explosives. In addition to vibrationalrotational and vibrational-translational energy transfer, an essential process in the attainment of thermal equilibrium in the reaction products is the amplification of pressure wavelets by the energy released during transitions from higher to lower vibrational levels during compression by these wavelets. It has long been known that the complex three-dimensional structure of self-sustaining detonation waves develops because the shock wave front is unstable to pressure wavelets of certain frequencies and that a substantial fraction of the chemical energy released must be communicated to the shock front.[1] Amplification of these pressure wavelets by vibrational deexcitation is postulated to be the physical mechanism by which the internal chemical energy of the product molecules sustains the leading shock front at constant C-J detonation velocity.[6]

The NEZND model provides a microscopic description of the reaction zone in self-sustaining detonation waves. For gas phase detonation waves in which the perfect gas law can be used for the equations of state and experimental shock tube chemical kinetic measurements are available for most, if not all, of the chain reactions involved in the energy release process, reactive flow models have been developed that closely simulate the complex three-dimensional wave front structure and the overall energy release behind each individual shock front.[21] Nonequilibrium gas phase energy transfer mechanisms preceding and following chemical reaction can be added. For condensed phase explosives, the extreme pressures and temperatures reached in detonation reaction zones in nanoseconds have precluded exact experimental measurements and numerical modeling. However, average reaction zone profiles have been measured and can be modeled. The phenomenological Ignition and Growth reactive flow model of solid explosive shock initiation and detonation has been normalized to such data and thus has correctly predicted the momentum delivered by detonation waves with finite thickness reaction zones in many applications.

2. IGNITION AND GROWTH REACTIVE FLOW MODEL

The Ignition and Growth reactive flow model of shock initiation and detonation of heterogeneous solid explosives[22,23] has been used to solve many explosive and propellant safety and performance problems. This model uses two JWL equations of state, one for the unreacted explosive and another one for its reaction products, in the temperature dependent form:

$$p = A\, e^{-R_1 V} + B\, e^{-R_2 V} + \omega C_v T/V \qquad (3)$$

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. The reaction rate law for the conversion of explosive to products is:

$$dF/dt = I(1-F)^b(\rho/\rho_o-1-a)^x + G_1(1-F)^c F^d p^y + G_2(1-F)^e F^g p^z \qquad (4)$$

$$0<F<F_{igmax} \qquad 0<F<F_{G1max} \qquad F_{G2min}<F<1$$

where F is the fraction reacted, t is time, ρ is the current density, ρ_o is the initial density, p is pressure in Mbars, and I, G_1, G_2, a, b, c, d, e, g, x, y, and z are constants. As explained more fully in previous papers,[22,23] this three term rate law models the three stages of reaction observed in shock initiation and detonation of solid heterogeneous explosives. Although it has mainly been applied to hot spot controlled shock initiation,[24-31] the Ignition and Growth formulation also quantitatively models detonation in one-, two-, and three-dimensional hydrodynamic codes to within the accuracy of current experimental measurements.[32-37] Its detonation model is based on Zeldovich-von Neumann-Doring (ZND) theory, in which the explosive is shocked by the leading front of the detonation wave with velocity D to the von Neumann spike state determined by the unreacted JWL equation of state. This unreacted equation of state is fitted to the available shock Hugoniot data above and below the von Neumann spike state. Then the first term in Eq. (4) ignites a few percent of the explosive corresponding to the initial porosity of the charge. The second term in Eq. (4) then rapidly reacts most of the rest of the explosive simulating the exothermic chain reactions that form highly vibrationally excited reaction product gases which then expand and thermally equilibrate as the pressure decreases from the von Neumann spike pressure and approaches the C-J pressure. The third term in Eq. (4) completes the reaction process by simulating the slower energy release, which is attributed to the diffusion controlled processes of solid carbon coagulation or aluminum particle oxidation.

The JWL reaction product equation of state is fitted to the available data on product expansion from cylinder test, embedded gauge, and laser interferometry experiments on self-sustaining and overdriven (supracompressed) detonation waves. For many solid explosives, JWL reaction product equations are fitted only to streak camera and Fabry-Perot data on expanding copper cylinders.[38] The copper cylinders expand for tens of µs until the pressures in the reaction products are well below 1 GPa. The JWL equations of state are used in two-dimensional hydrodynamic codes to simulate the entire copper expansion process and thus are not heavily weighted toward the high pressure region. There are also experimental difficulties in observing the initial copper wall motion, even with the Fabry-Perot technique.[38] Thus JWL reaction product equations of state derived just from cylinder test data are not necessarily sufficiently accurate near the C-J state, even when the correct reaction zone momentum is included in a reactive flow model. To measure this high pressure region more precisely, smaller scale experiments using nanosecond time resolution techniques are required. The Sideways Plate Push test[39] was recently developed to precisely measure the high pressure radial expansion of the reacting explosive and its products for the first few µs. Supracompression data on reaction zone profiles and product states above the C-J state are also required for accurate axial metal acceleration modeling.[3,4]

For reaction zone profile studies, the fastest time response of an LLNL embedded gauge is approximately 4 ns for the 25 micron thick copper particle velocity probe of Hayes and Tarver.[13] Some specialized VISAR[34] and ORVIS[15] laser interferometers have subnanosecond time resolution, but they currently record the flow for very short times at inert interfaces. Figures 2 and 3 show Hayes gauge records for detonation waves in LX-14 (HMX-based) and LX-17 (TATB-based), respectively, and the corresponding Ignition and Growth calculations, along with several C-J idealized detonation calculations.[39] Since the C-J models (called program burn or beta burn in hydrodynamic codes) neglect the momentum associated with the reaction zone, they either underestimate the total momentum in the wave or incorrectly add extra momentum somewhere in the product expansion process. Either way, they do not correctly calculate the explosive energy delivery. These C-J models have many other problems, such as propagating as ramp waves rather than shocks, needing "shadow velocities" to turn corners, not modeling real detonation wave curvature effects, etc. Once the reactive flow model is normalized to a sufficient amount of experimental data, it correctly simulates all aspects of detonation energy delivery over a wide range of pressures. Figure 4 contains an example of one-dimensional metal acceleration, in which Fabry-Perot records of the free surface velocities of 0.267 mm thick tantalum discs driven by 19.871 mm thick discs of detonating LX-17 are compared to the Ignition and Growth calculation using the LX-17 model from Fig. 3. In one-dimensional calculations, the reaction zone expansion process follows the Rayleigh line from the spike to the C-J state. In multidimensional calculations, the detonation wave front is curved due to the presence of rarefaction waves, and the model's reaction rates must respond correctly to these pressure changes to accurately calculate such measurable properties as failure diameter, corner turning, divergence, front curvature, etc.[25,28,33] In all calculations, the equations of state and the reaction rate laws must respond correctly to the generation of reflected shock and rarefaction waves by interactions with surrounding inert materials.

3. The Next Generation Reactive Flow Model

Although the Ignition and Growth continues to be very successful in modeling shock initiation and detonation in solid explosives, it does have limitations and can be improved. Obviously, chemical kinetic reaction rate laws are governed by temperature rather than pressure, and thus pressure dependent reaction rate laws have difficulties calculating flows in which the pressure and temperature changes are of different magnitudes. Such flows occur in reflected shocks, in hot spots during rarefaction, in Mach stem interactions, etc.[40] Since the ignition of reaction occurs in hot spots and thus is controlled by their number, sizes, and temperatures, a compression based ignition term can not model the details of all possible scenarios, such as shock desensitization.[41]

Therefore a completely temperature based reactive flow model, which includes the physical processes that lead to hot spot formation, ignition, growth, and coalescence, is desired. The time resolved experimental techniques to measure temperature everywhere in a shocked, reacting solid explosive have not yet been developed, but a reactive flow model is being built assuming that these measurements will be forthcoming. To formulate this temperature dependent model, the heat transfer into hot spot sites, the race between heat diffusion and chemical reaction in the igniting sites, and the growth (or failure to grow) of the hot spots must be intimately coupled to the hydrodynamic equations. This linking procedure has been completed in the ALE3D hydrodynamic code,[42] in which the heat transfer code Chemical TOPAZ[43] has been embedded. Chemical TOPAZ has been used to estimate critical conditions for hot spot reaction growth in HMX and TATB based on chemical kinetic decomposition models derived from thermal decomposition experiments.[44] Advanced material models for the unreacted explosive are required to evaluate specific hot spot formation mechanisms, such as void collapse, friction, shear, viscous void closure, etc., and to correctly partition the work done between thermal energy and potential energy.

The equations of state of both the unreacted explosive and its reaction products must also be more advanced than the simple JWL form used in Eq. (3). Accurate temperature calculations for the compressed, unreacted explosive outside of the hot spots are essential for determining the growth of reaction by heat conduction from the reacting hot spots to the neighboring explosive molecules. The heat capacity of the solid is a function of temperature and probably a function of volume so these dependencies must be added. The distribution of energy between thermal and potential energy is much more critical when using temperature dependent rates than when global pressure dependent rates are used. The Gruneisen parameter ω in Eq. (3) is set equal to a constant for both the unreacted explosive and reaction products. In the case of the products, ω is generally between 0.4 and 0.6, based on chemical equilibrium code predictions at the C-J state.[45] However, ω decreases as the pressure decreases and specific volume increases toward the perfect gas value of approximately 0.25 ($C_p/C_v - 1$). Thus a reaction product equation of state in which ω is a decreasing function of specific volume is needed for accurate modeling to very low pressures. Experimental measurements are not yet available for ω as a function of shock pressure in unreacted solid explosives. The use of JWL-like exponential terms to describe the cold compression (potential) energy at high shock pressures may not be the optimum way to model the unreacted explosive, the reaction products, and the reacting mixture at high pressures and high temperatures.

Therefore a great deal of new experimental data is required to improve the equations of state and chemical reaction rate laws. The most important data is temperature measured in and around hot spots, in homogeneous explosives, in multiple shock geometries, and in detonation reaction zones. The temperature measurement techniques recently reported by Yoo et al.[46] for shock initiation and detonation of homogeneous explosives and Benson et al.[47] for shock initiation of heterogeneous solid explosives appear to hold great promise. The recent sound velocity measurements of Fritz et al.[48] in the pressure regime near the C-J state of the HMX-based solid explosive PBX 9501 provide another essential set of equation of state data. Hopefully the necessary experimental data will be forthcoming as the next generation reactive flow model is developed.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

Figure 1. Nonequilibrium ZND (NEZND) model of detonation in an organic solid or liquid explosive $C_wH_xO_yN_z$

Figure 2. Experimental and calculated particle velocity histories for a Hayes gauge in detonating LX-14

Figure 3. Experimental and calculated particle velocity histories for a Hayes gauge in detonating LX-17

Figure 4. Experimental and calculated free surface velocity histories for 0.267 mm-thick tantalum discs driven by 19.871 mm of LX-17

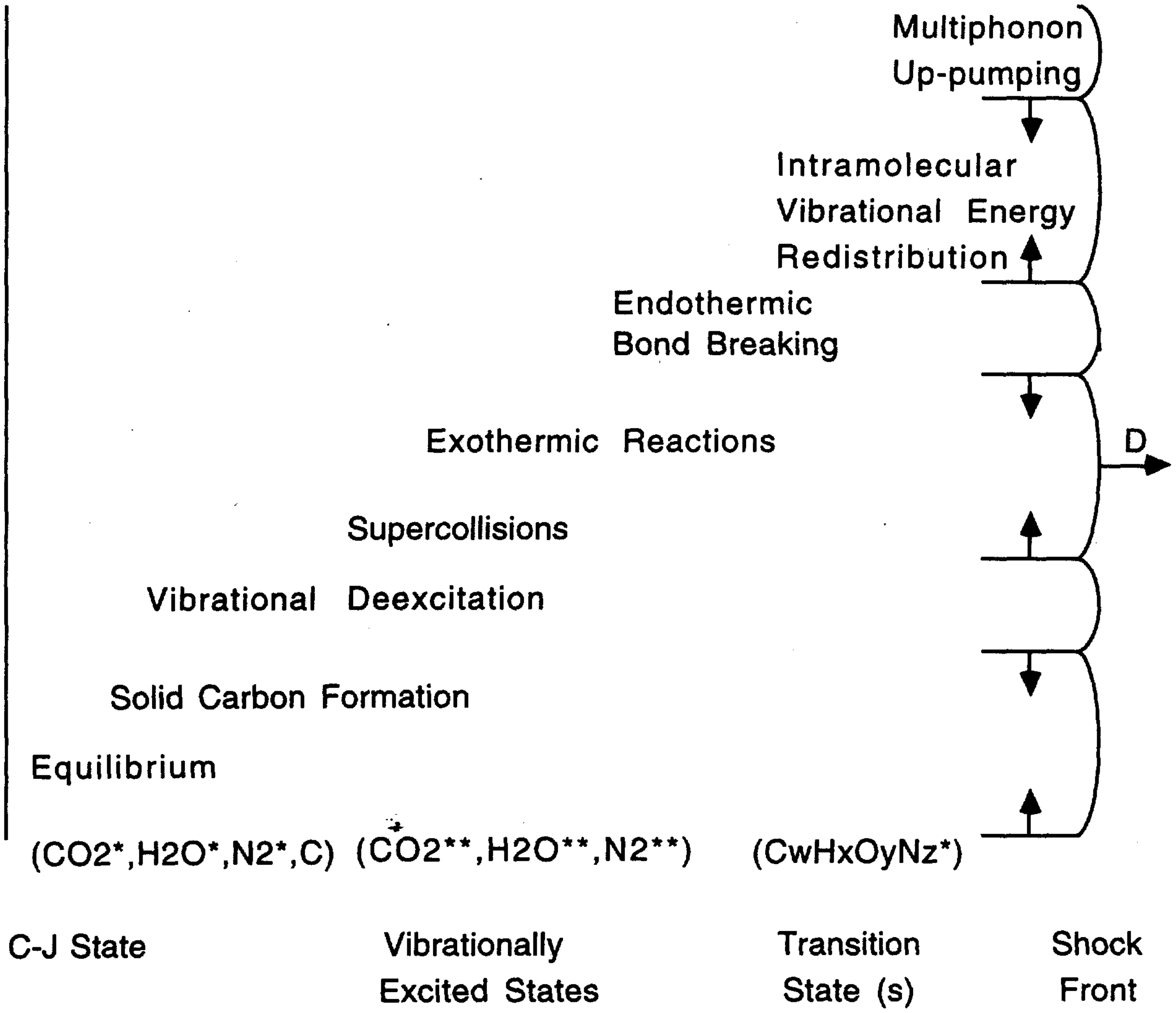

Figure 1. The Nonequilibrium ZND (NEZND) Model of Detonation in an Organic Solid or Liquid Explosive CwHxOyNz

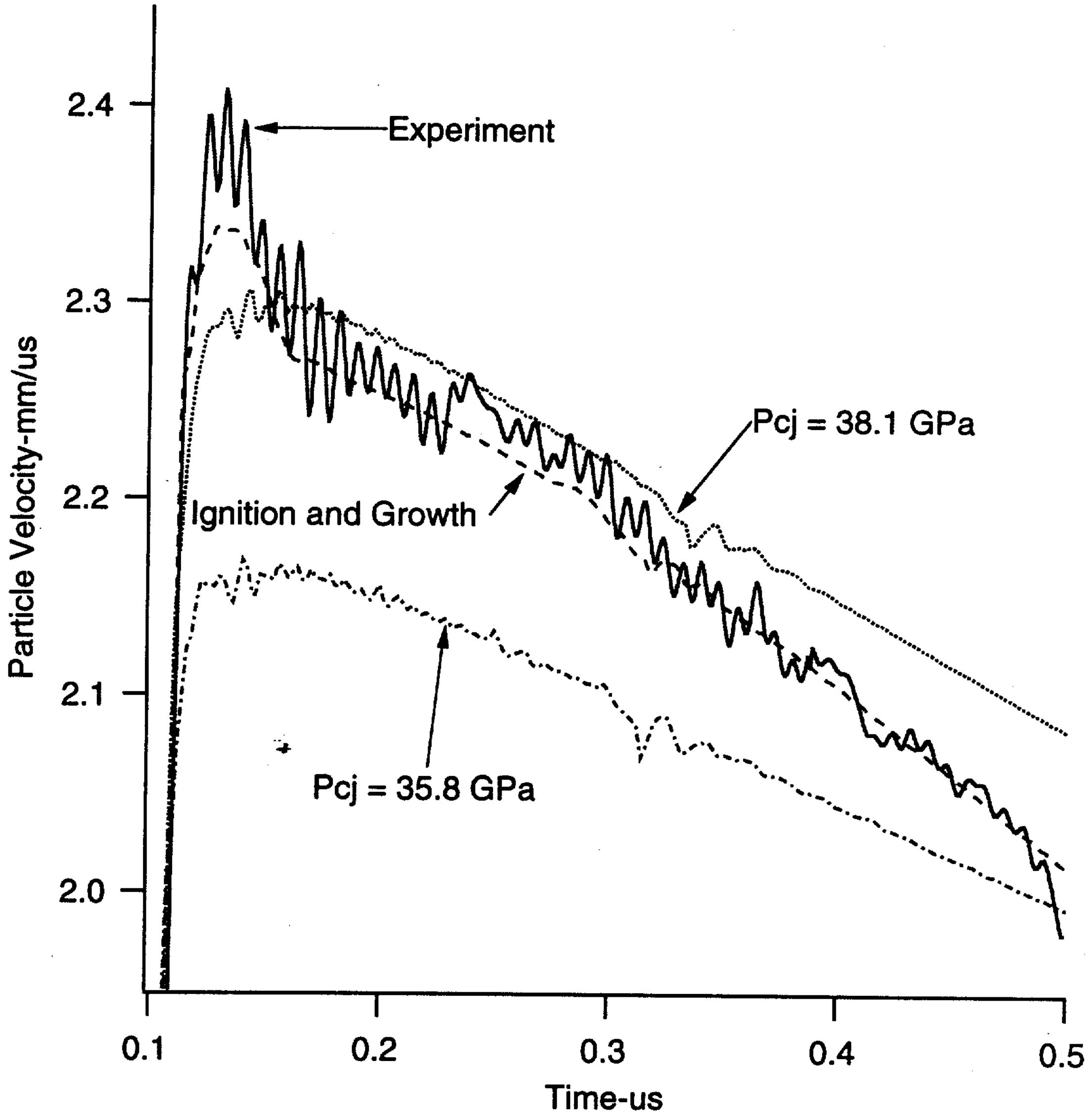

Figure 2. Experimental and Calculated Particle Velocity
Histories for a Hayes Gauge in Detonating LX-14

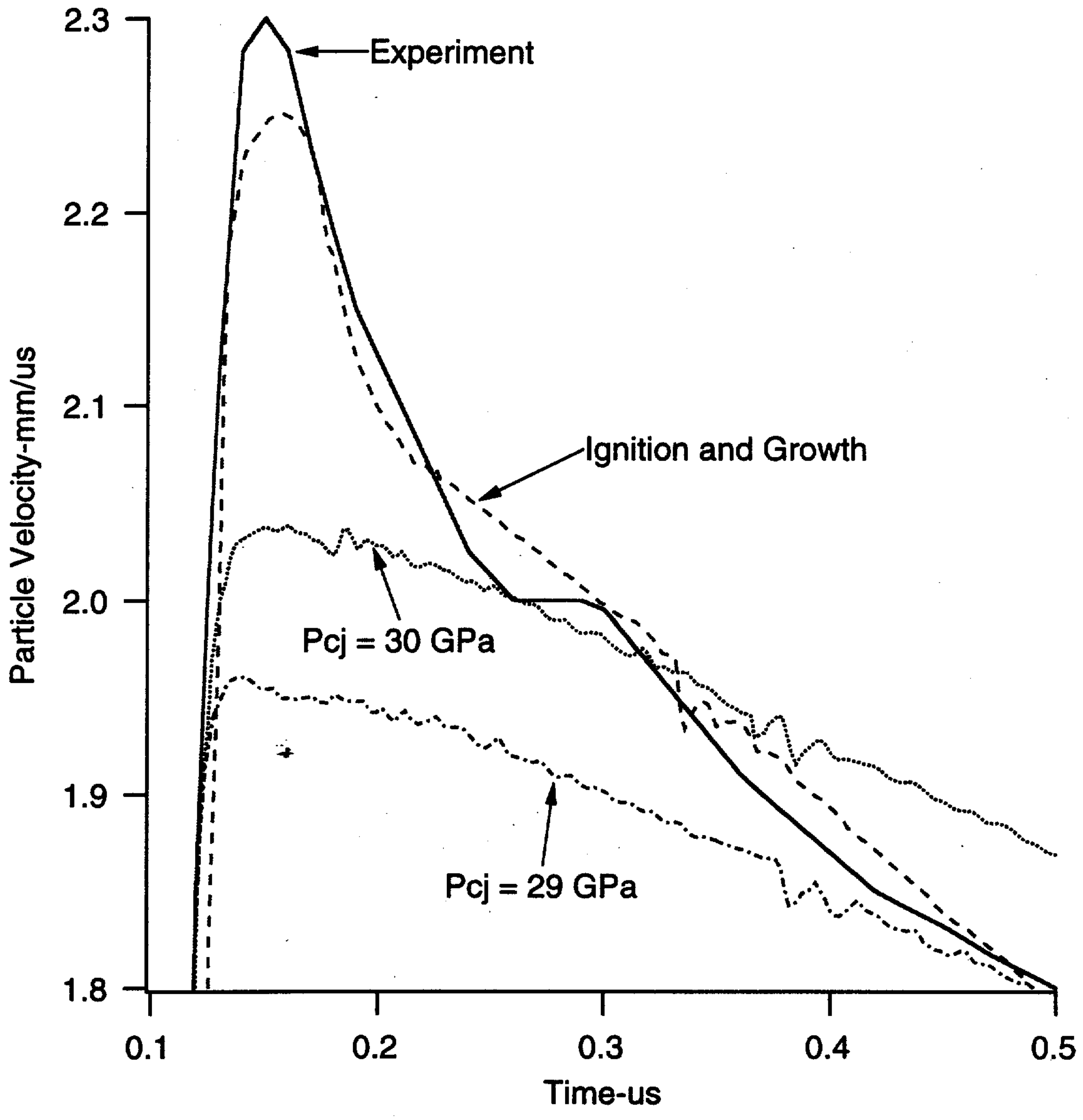

Figure 3. Experimental and Calculated Particle Velocity Histories for a Hayes Gauge in Detonating LX-17

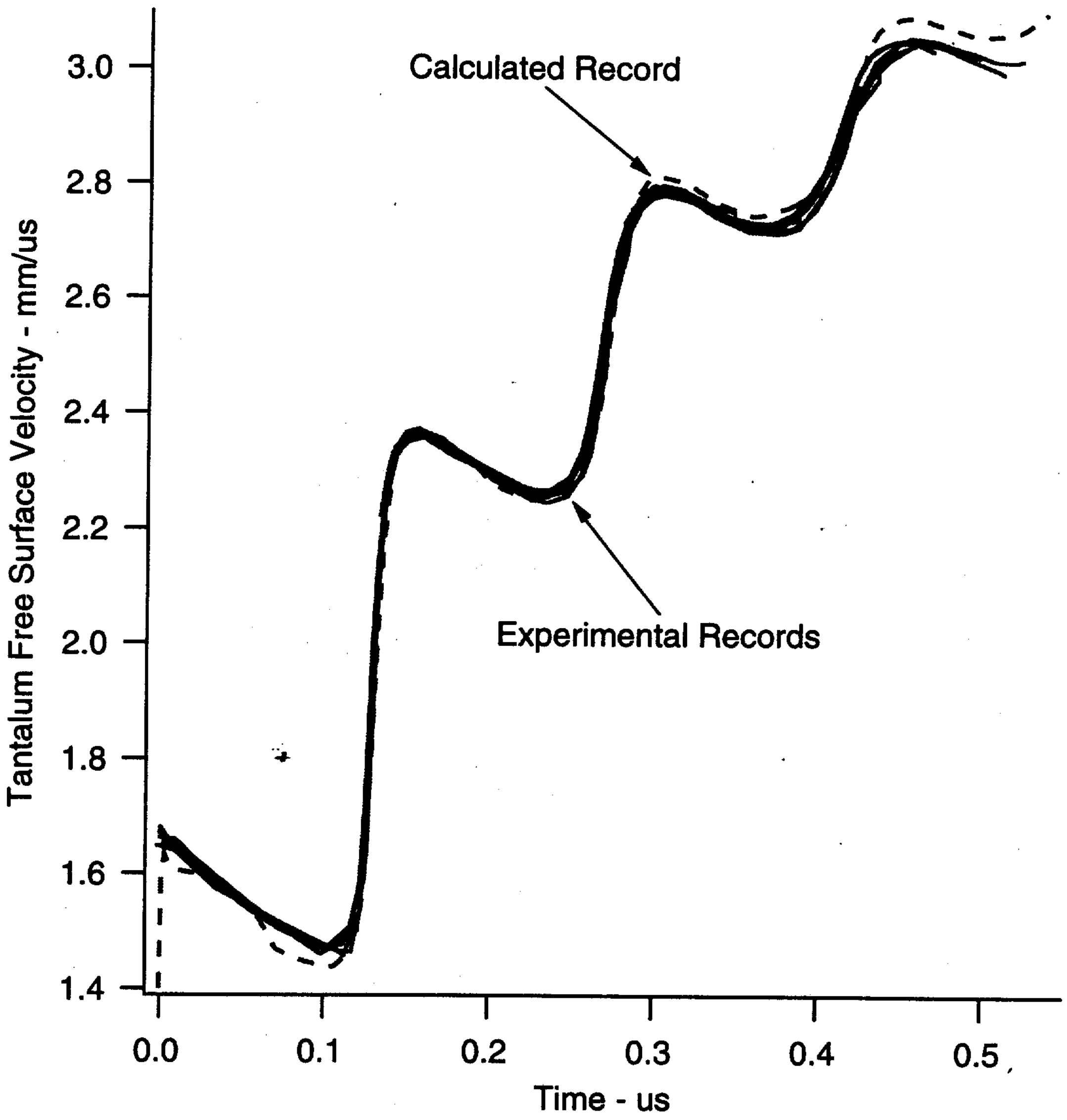

Figure 4. Experimental and Calculated Free Surface Velocity Histories for 0.267 mm thick Tantalum Discs Driven by 19.871 mm of LX-17

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