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THE HETEROGENEOUS EXPLOSIVE REACTION ZONE

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The calculated reaction zone of PBX-9404 using solid HMX Arrhenius kinetics is stable to perturbations. The calculated reaction zone Von Neumann spike pressure agrees with the experimental observations within experimental uncertainty associated with different experimental techniques. The calculated homogeneous explosive reaction zone thickness is larger than observed for the heterogeneous explosive. The effect of two volume percent air holes on the reaction zone was modeled using the three-dimensional Eulerian reactive hydrodynamic code, *3DE*. The air holes perturb the reaction zone. A complicated, time-dependent, multidimensional reaction region proceeds through the heterogeneous explosive. The experimentally observed reaction zone characteristic of heterogeneous explosives are mean values of an irregular, three-dimensional reaction region.

INTRODUCTION

The hydrodynamic stability of one-dimensional detonations in an ideal gas was studied analytically by Erpenbeck[1] and numerically by Fickett and Wood,[2] Fickett, Jacobson, and Schott,[3] and Mader.[4] The hydrodynamic stability of two-dimensional detonations in gases was studied by Mader,[4] by Taki and Fujiwara,[5] and by Markov.[6] Mader also studied the stability of detonations in the homogeneous explosives nitromethane and liquid TNT. Detonations of the condensed homogeneous explosives nitromethane and liquid TNT were found to exhibit unstable periodic behavior. The steady-state Chapman-Jouguet theory of the detonation process will not properly describe the behavior of homogeneous explosives that exhibit such unstable behavior. Most experimental studies of reaction zone characteristics of explosives have been performed using heterogeneous explosives rather than liquids or single crystals. Heterogeneous explosives are explosives containing density discontinuities such as voids or air holes. The shock interactions that occur when shocks interact with voids or air holes result in local high temperature and pressure regions called "hot spots." These "hot spots" decompose and add their energy to the flow and result in the process of heterogeneous shock initiation. The process has been modeled numerically and is described in detail by the hydrodynamic hot spot model.[7,8,9,10] The success of the three-dimensional numerical models in describing the interaction of shock waves with density discontinuities and of a detonation wave interacting with a matrix of tungsten particles in HMX,[11] encouraged us to numerically examine the interaction of a resolved reactive zone in HMX with a two volume percent matrix of air holes.[12] An objective of the study was to determine the nature of the flow being examined in experimental studies of reaction zones of heterogeneous explosives.

EXPERIMENTAL OBSERVATIONS

The heterogeneous explosive reaction zone that has been the most studied is PBX-9404 (94/3/3 HMX/Nitrocellulose/Tris-β-chloroethyl phosphate). A summary of the estimated reaction zone thickness and Von Neumann spike pressure is given in Table I along with the calculated reaction zone parameters using the solid Arrhenius HMX constants of 34.8 kcal/mole for activation energy and 3 x 10^4 μs^{-1} for frequency factor described in reference 9.

The metal free-surface measurements of B. G. Craig used the technique described in reference 13.

The infra red radiometry measurements of W. Von Holle used the technique described in reference 14.

The interferometer measurements were performed by W. Seitz. The application of this method to reaction zone measurements is described in reference 15.

The bromoform measurements were made by R. McQueen and J. Fritz using the technique described in reference 16.

The calculated reaction zone for "homogeneous" PBX-9404 is larger than the observed reaction zone for heterogeneous PBX-9404, although within the uncertainities associated with experimental interpretation and with the solid Arrhenius constants. We undertook an investigation of the effect of heterogenities on reaction zone structure to determine if they might result in a reaction zone whose effective thickness was different than the ideal steady-state reaction zone length.

The calculated steady-state reaction zone for the homogeneous explosive liquid TNT is 0.001 cm in good agreement with the Hayes conductivity thickness of 0.0013 cm and consistent with the Craig upper limit free surface reaction zone thickness of 0.01 cm reported in reference 7. The observed reaction zone thickness is probably a mean of a periodic flow since Hayes observed a 1000 megacycle (10^{-3} μs/cycle) oscillation in liquid TNT and we calculate a period of 5 x 10^{-2} μs.

TABLE I. PBX-9404 REACTION ZONE

Experimental Technique	Reaction Zone Thickness (cm)	$P_{VN\ Spike}$ (kbar)	P_{C-J} (kbar)
Bromoform	0.02	485	
Interferometer	<0.01		
Infra Red Radiometry	0.02-0.03		
Metal Free Surface	0.01	550	365
Calculated	0.07	560	365

TABLE II. PBX-9502 REACTION ZONE

Experimental Technique	Reaction Zone Thickness (cm)	$P_{VN\ Spike}$ (kbar)	P_{C-J} (kbar)
Interferometer	0.08-0.16	376	
Infra Red Radiometry	0.08		
Metal Free Surface	0.03		290
Foil/Water	0.21	375	
Calculated		377	290

TABLE III. COMPOSITION B REACTION ZONE

Experimental Technique	Reaction Zone Thickness (cm)	$P_{VN\ Spike}$ (kbar)	P_{C-J} (kbar)
Bromoform	0.04	395	
Interferometer	0.02	420	
Metal Free Surface	0.014	374	285
Conductivity	0.013		
Calculated		437	285

NUMERICAL MODELING

The steady-state reaction zone for PBX-9404 was calculated using the one-dimensional reactive hydrodynamic code, *SIN*, the HOM equation-of-state constants described in reference 7 and the Arrhenius constants for solid HMX described in reference 9.

The calculated PBX-9404 reaction zone profile is shown in Figure 1.

The time-dependent behavior of the flow in the reaction zone of detonating PBX-9404 was investigated using one-dimensional Lagrangian and three-dimensional Eulerian numerical hydrodynamics. The steady-state solution was stable and perturbations were found to decay. This is in contrast to the time-dependent, unstable, periodic reaction zones reported for liquid TNT and nitromethane in references 4 and 7.

The stable steady-state reaction zone of PBX-9404 permits us to study the effect of heterogenities on the reaction zone profile without the complication associated with a time-dependent reaction zone.

To examine the effect of heterogenities on the reaction zone, we used the three-dimensional Eulerian reactive hydrodynamic code, *3DE*.[17] It uses the techniques identical to those described in detail in reference 7 and used successfully for describing two-dimensional Eulerian flow with mixed cells and multicomponent equations of state, and for modeling reactive flow including reaction zone stability.[4]

The three-dimensional computational grid contained 30 cells in the x direction, 28 cells in the y direction, and 57 cells in the z direction, each 0.004 cm on a side. The time increment was $8 \times 10^{-4} \mu s$. At the bottom of the grid was a reaction zone piston as described in references 4 and 7, which was programmed to initialize the flow with a steady-state reaction zone. After the steady-state reaction zone had traveled one reaction zone length in solid PBX-9404, it interacted with a two percent by volume HCP (hexagonal close packed) matrix of air holes.

Thirty-four spherical air holes, each with a diameter of 0.012 cm, occupy a region in the middle of the mesh about 25 cells high. Partial air spheres occur on the boundaries as necessary. The air hole size was chosen to be representative of the actual hole size present in pressed PBX-9404.

Numerical tests with two to six cells per air sphere diameter showed the results were independent of grid size for 3 or more cells per sphere diameter.

Figure 2 shows the initial configuration of spherical air holes in PBX-9404.

The low resolution necessary for the three-dimensional calculation results in a less resolved reaction zone than described earlier using the one-dimensional *SIN* code. The reaction zone burn fraction as a function of distance is shown in Figure 3 for the *SIN* one-dimensional calculation and for the *3DE* calculation for two viscosity coefficients. The viscosity coefficient shifts the location of the start of the burn. The profile is not significantly changed by variations in the viscosity.

The *3DE* reaction zone profile for pressure, temperature, particle velocity and mass fraction as a function of distance are shown in Figure 4.

The burn fraction surfaces of a PBX-9404 reaction zone after it has interacted with the region of two volume percent air spheres is shown in Figure 5. The heterogenities perturb the reaction zone. A complicated reaction region develops and is maintained by the reactive flow.

Cross sectional plots of pressure and burn fraction through the 15th cell in the x direction ($I = 15$) are shown in Figure 6. A complicated time-dependent, multi-dimensional reaction region proceeds through the heterogeneous explosive.

CONCLUSIONS

The calculated reaction zone of PBX-9404 using solid HMX Arrhenius kinetics is stable to perturbations and a steady-state reaction zone profile is maintained. This is in contrast to the time-dependent, periodic reaction zone calculated for nitromethane and liquid TNT as described in references 4 and 7.

The effect of two volume percent spherical air holes on the reaction zone was modeled using the three-dimensional Eulerian hydrodynamic code, *3DE*. The air holes perturb the reaction zone flow. A complicated reaction region develops and is maintained by the reactive fluid dynamics.

Thus, any experimental study of a reaction region in a heterogeneous explosive is actually measuring some mean value of an irregular, complicated multi-dimensional flow. It is not surprising that different experimental techniques may give quite different reaction zone "thicknesses," Von Neumann spike pressures and profiles.

As shown in Table II for PBX-9502 and Table III for Composition B, the measured reaction zone parameters for heterogeneous explosives vary considerably with the experimental technique. The reported reaction zone thickness for PBX-9502 (95/5 TATB/Kel F, ρ = 1.894) varies by a factor of 8 between the metal free-surface measurement of Craig[15] and the foil/water measurement reported by Sheffield.[18] The reaction zone lengths for several TATB formulations reported by Campbell and Engelke[19] vary from 0.5 to 0.014 cm or by a factor of 35. Some of this variation is probably a result of the different binders and densities of the TATB formulations and the indirect estimation methods used to obtain some of the reaction zone thickness. As shown in Table III, the reaction zone thickness of Composition B (64/36 RDX/TNT, ρ = 1.713) varies by a factor of 4 between the bromoform measurement and the conductivity measurement of Hayes.[20]

The measured Von Neumann spike pressure can also vary with the experimental technique as shown in Table III where the reported Composition B Von Neumann spike pressure varies from 374 to 420 kilobars, and in Table I where it varies from 485 to 550 kilobars for PBX-9404.

The reactive region in heterogeneous explosives is complicated, time-dependent, and multi-dimensional (non laminar).

The reactive region has bounds which approach a steady-state condition, but the flow inside those bounds is multi-dimensional and time-dependent.

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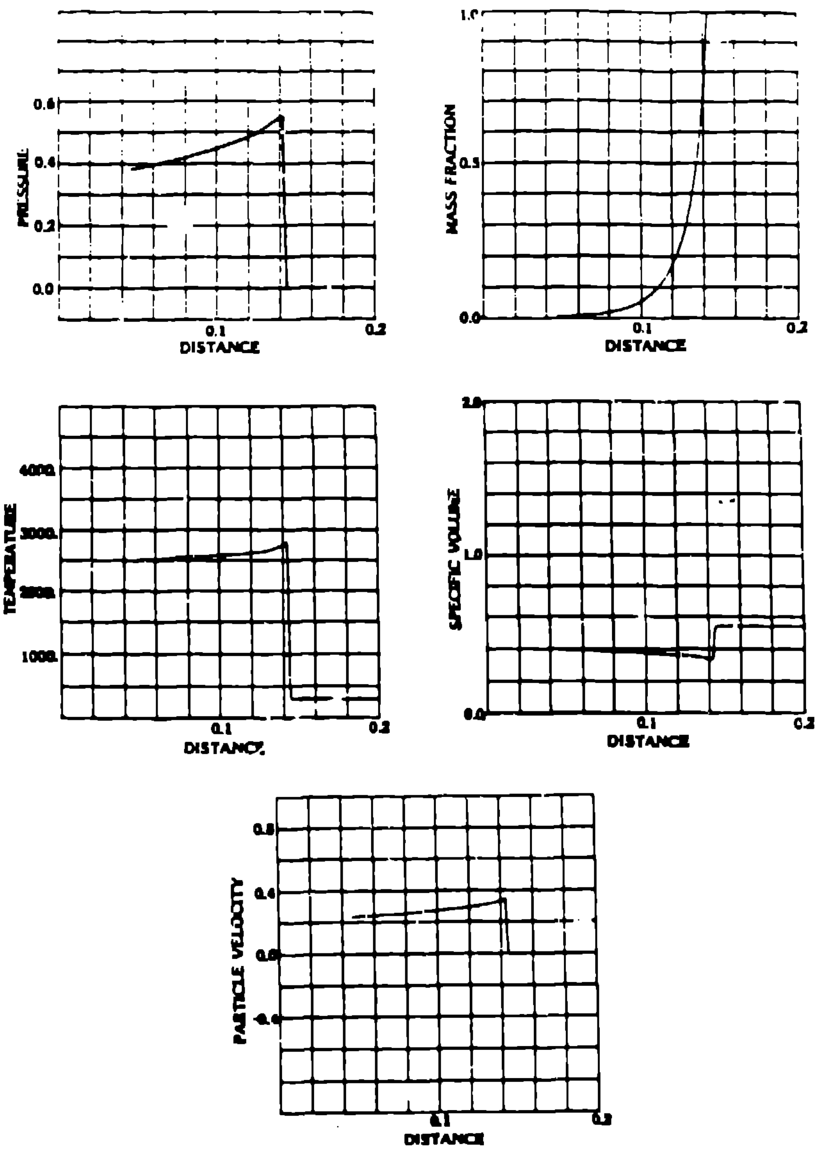

Fig. 1. The high resolution one-dimensional reaction zone profile of PBX-9404 calculated using the *SIN* code.

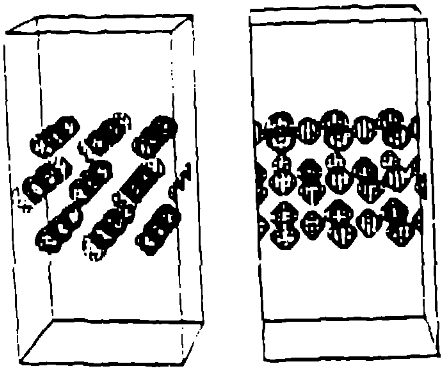

Fig. 2. The initial configuration of air spheres in a cube of solid PBX-9404.

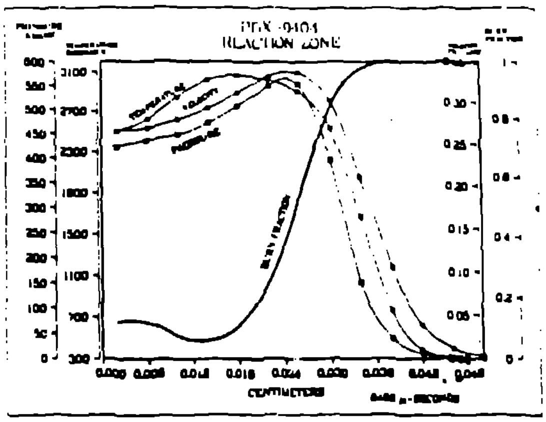

Fig. 4. The reaction zone profiles in the *3DE* calculation.

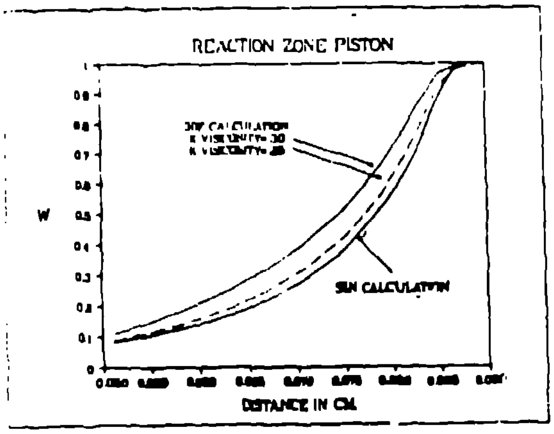

Fig. 3. The reaction zone mass fraction as a function of distance for the high resolution *SIN* calculation and low resolution *3DE* calculation.

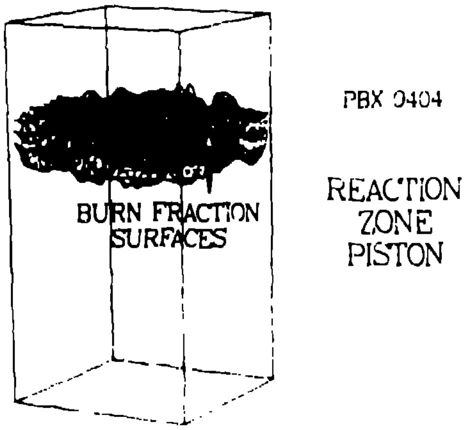

Fig. 5. Burn fraction surface profiles after reaction zone interacts with heterogenities in PBX-9404.

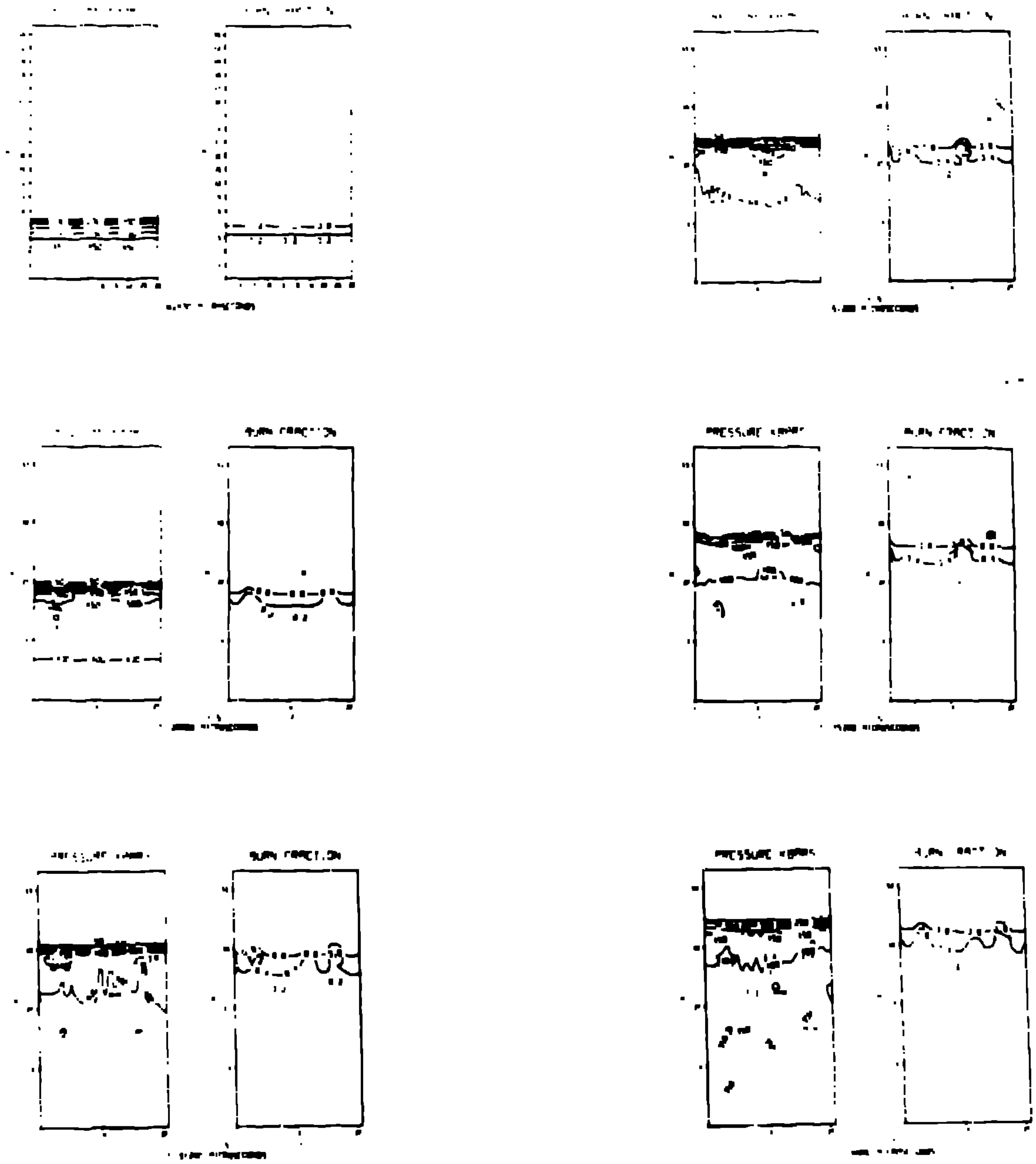

Fig. 6. Cross sectional plots through I=15 of pressure and burn fraction showing the heterogeneous PBX-9404 reaction region.