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TITLE **INITIATION OF PRESHOCKED HIGH EXPLOSIVES PBX-9404, PBX-9502, and PBX-9501, MONITORED WITH IN MATERIAL MAGNETIC GAUGING**

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INITIATION OF PRESHOCKED HIGH EXPLOSIVES PBX-9404, PBX-9502, PBX-95.1, MONITORED WITH IN-MATERIAL MAGNETIC GAUGING

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Desensitization of explosives by preshocking is being studied using the well-supported plane shock waves generated by a gas gun. Evolution of the waves in the explosive is monitored using in-material multiple magnetic gauges to measure particle velocity in the Lagrangian frame, over ~ 3μs of run. PBX-9404, PBX-9501, and PBX-9502 have been studied, at pressures up to 10.5 GPa. A substantial extension of the run to detonation is observed in PBX-9404, with the run beginning approximately at the end of the preshocked region. A reactive wave is observed while the preshock persists in both PBX-9404 and PBX-9501, but evidently does not contribute to the detonation wave or shorten the run to detonation. PBX-9502 is inert at pressures accessible with the gas gun, but serves to clarify the progress of multiple shocks over the off-Hugoniot EOS surface and the shock dynamics of wave coalescence.

INTRODUCTION

Detonation characteristics of multiply shocked high explosives (HE) frequently differ noticeably from those of virgin explosive, beyond variations expected due to differing initial state. A precursor shock of sufficient pressure will hinder initiation by a subsequent shock of high pressure. This phenomenon is important to considerations of safety and relevant to many applications of energetic materials.

These studies complement previous investigations. Campbell and Travis[1] studied the overlap of two shocks in detonating material, and developed the idea that the run to detonation in preshocked explosive could best be estimated by the run anticipated for the second shock, measured from the point of coalescence of the preshock and the subsequent shock. Interaction of an established detonation with a preshocked region[1] yielded a quantitative minimum criterion for desensitization. $P^{2.2}\tau = 1140$ (kbar, μs), dependent on preshock pressure P and duration τ. Their work includes data from a series of flyer plate preshock experiments done by E. Gittings[2], in which desensitization of impacted explosive was observed.

R. E. Setchell[3] used VISAR diagnostics to track behavior of materials subjected to preshocking by ramp waves which subsequently develop into shocks. The ramp waves evidently gradually compress granular materials, without provoking reaction at a pressure of 5.1 GPa. Even after shock formation, a considerable delay in transition to detonation was observed, relative to initiation by a simple shock. Subsequent experiments[4] using a separate ramp precursor wave showed similar desensitization behavior.

A similar study of the effect of short shocks and preshock pulses on detonation of PBX-9501 was done by Vorthman and Wackerle[5], using in-material magnetic gauges to record particle velocities. A Lagrangian analysis code was used to estimate the effect of the preshock on the reaction rate.

Recent work by J. P. Plotard at Vaujours[6] shows a marked increase of run to detonation with increased preshock duration, in a material composed of TATB and HMX in comparable quantities.

EXPERIMENTAL

Experiments were done on a single stage light gas gun. An inert projectile is accelerated to impact an HE target. Use of a gas gun to generate the shock waves provides the distinct advantages of a well characterized wave shape and well supported shocks. The flat ...aped pressure pulse simplifies consideration of the time-dependent behavior of the growth of the reactive wave.

Our gas gun can reach projectile velocities of up to 1.4 mm/µs, corresponding to pressures of up to about 10.5 GPa in PBX materials when single crystal sapphire impactors are used. Velocities are repeatable to 0.02 mm/µs. This maximum pressure is insufficient to cause initiation of PBX-9502 within an observable time, but will cause rapid growth to detonation in PBX-9404 or PBX-9501. Experiments have been performed on PBX-9502, to provide exactly quantifiable data on an effectively inert sample of PBX material, and on PBX-9404 and PBX-9501 to study initiation behavior under preshock conditions.

Use of embedded magnetic gauges provides unique measurements in the Lagrangian frame of the time evolution of the shocks, for a duration up to 3µs. Use of multiple gauges give independent measurements of particle velocity u_p, shock velocity U_s and, from impulse records, an estimate of the final stress. The precision of the gauges is about 1 to 2%.

The gauge package may consist of 5 nested particle velocity gauges and five impulse gauges (MIV gauge), arranged to record at well determined time intervals, or it may consist of ten particle velocity gauges (multiple magnetic gauge), to more closely monitor the time evolution of waves progressing through the material. Stirrup gauges are also used, to record input waves. The experimental setup is shown in Figure 1.

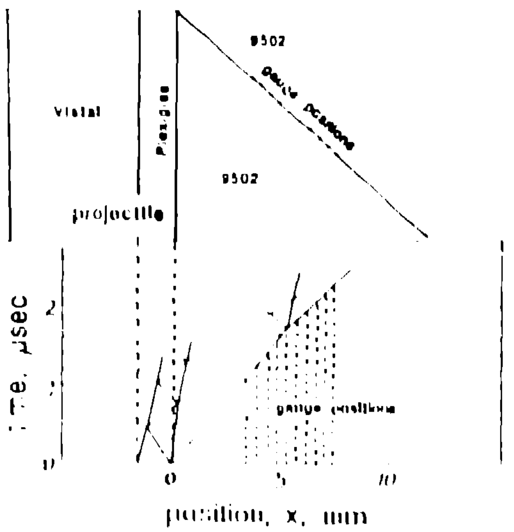

FIGURE 1. SETUP OF PRECURSOR SHOCK EXPERIMENTS

Current preshock experiments are done with a composite impactor. The stepped waves are generated using a thin (0.6 to 1.2 mm) piece of Kel-F or plexiglass on the front of a high impedence impactor. This experimental design allows accurate manipulation of time and pressure parameters, and generates reproducible flat-topped waves. Impedances of Kel-F and Vistal[7] are such that the first shock is about half of the stress of the second shock.

This projectile impacts onto a precisely machined flat explosive cylinder, into which the gauge package has been embedded at a 30° angle. Waves generated are one-dimensional for about 4 µs. Signal recording is timed with redundant shorting pins, and velocity measured with an independent set of pins set perpendicular to the path of the projectile.

Experimental wave velocities are obtained by plotting gauge responses against the known gauge positions for the particle velocity gauges. Data are compared with pressures and particle velocities estimated using the MACRAME[8] code, employing a Gamma-law equation of state (EOS) for each material.

RESULTS

Two experiments have been done with PBX 9404, and one with PBX-9501 to observe delayed detonation. In each of these experiments reaction after the second shock was observed. Four experiments have been completed on PBX 9502, which is basically an inert material under conditions achievable with our gun. Calculated pressures and measured particle velocities and wave velocities are tabulated in Table 1.

PBX-9502

Since PBX 9502 behaves as an inert material at pressures below 10.5 GPa (when observed for only a few µs), it provides a convenient test of the technique and of material response to multiple shocks. For precursor shocks, the data supplements the measured low pressure points on the principle Hugoniot, while in the subsequent shock, points on the second shock Hugoniot of PBX 9502 relative to the initial shock are measured, giving new data on the PBX 9502 EOS. Although coalescence time falls consistently later than calculated from the principal Hugoniot, measured points on the second Hugoniot are very close to the principal Hugoniot, within the experimental error of previous[9] (plane wave lens) measurements in the region studied.

Table of Data

	Experiment - 863			Experiment - 876			Experiment - 886			Experiment - 887	
	Observed	MACRAME		Observed	MACRAME		Observed	MACRAME		Observed	MACRAME
u_1	0.37	0.3882	u_1	0.56	0.5543	u_1	.57	.60	u_1	.625	.653
u_2	0.73	0.7066	u_2	0.96	0.9148	u_2	.97	.98	u_2	1.08	1.079
u_3	0.78	0.7651	u_c	0.96	0.9307	u_c	.97	1.00	u_c	1.08	1.093
u_c		0.7720	U_1	3.523	3.538	U_1	3.76	3.65	U_1	3.39	3.813
U_1	3.022	3.045	U_2	5.724	6.083	U_2	6.27	6.293	U_2	5.56	6.542
U_2	4.416	5.233	U_c	4.44	4.520	U_c	4.65	4.68	U_c	4.839	4.884
U_3	5.99	6.142	P_1		3.71	P_1		4.13	P_1		4.71
U_c		3.990	P_2		8.19	P_2		9.12	P_2		10.38
P_1		2.24	P_c		7.96	P_c		8.87	P_c		10.11
P_2		5.58									
P_3	6.102	6.63									
P_c		5.43									

Experiments have focused on the region of the PBX 9502 Hugoniot which appears to show a discontinuity at approximately 7.5 GPa[9]. According to extant data,[9] this cusp should be too small to be resolved in a single shock experiment. While single-wave experiments in PBX-9502 do not yield a two-wave structure in crossing this discontinuity, the steeper Rayleigh line of the second shock in a two shock experiment is more likely to be interrupted by this small cusp, generating two waves in the second shock, defining the location of this discontinuity. A front impactor made of Kel F, a near impedance match to PBX-9502, eliminates the third reverberation in the gauge records, allowing a search for a two wave structure in the second shock. Data from one experiment are shown in Figure 2.

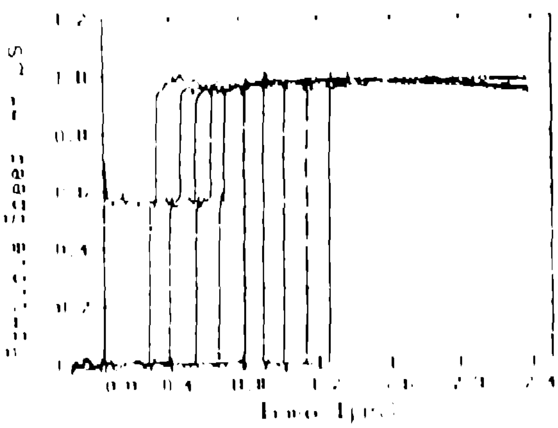

FIGURE 2. PARTICLE VELOCITY RECORDS FROM PBX 9502 PRECURSOR SHOCK EXPT

In four experiments done at different pressures in PBX-9502, no two-wave structure has been observed. Possible reasons for the lack of an observable wave are that the cusp may be too small, the pressure pairs may bracket the exact region too widely, or the process responsible for the cusp may be slow on the timescale of the shock propagation. A continuing series of experiments is investigating this region in small pressure increments, to locate exactly and characterize this apparent discontinuity and try to determine if it is due to a phase transition or other phenomenon.

Adequate data on the single wave arising after coalescence of the preshock and second shock allows the velocity of this wave to be measured. As expected, the velocity is decreased from that of the second shock. Examination of the coalescence in the x-t plane indicates that a small rarefaction should be propagated back into the doubly shocked region to accommodate this state change. Estimation of the location of the second Hugoniot relative to the principal Hugoniot in the P-u_p plane will predict the magnitude of the rarefaction wave. The MACRAME code gives a value of 0.15 GPa or 2.7% for the pressure change. This small wave is not resolved in the current data.

Hugoniot points measured in these experiments are plotted in Figure 3, showing that the second Hugoniot is very close to the principal Hugoniot for PBX 9502 at these pressures, and that the small rarefaction is unlikely to be larger than the resolution of the data.

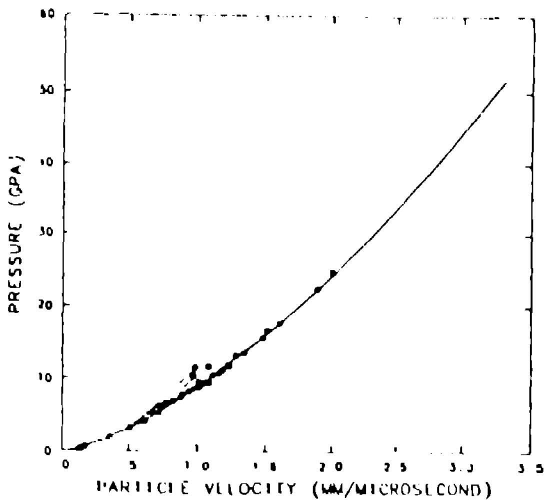

FIGURE 3. HUGONIOT DATA FOR PBX 9502.

PBX-9404

Initiation of PBX 9404 under preshocked conditions has been observed in detail, with the development of the reactive wave monitored at ten or more positions as the reaction progressed. Gauge records of the input shocks (precursor shock 2.3 GPa and second shock 5.6 GPa), and emerging reactive waves in PBX 9404 are shown in Figure 4.

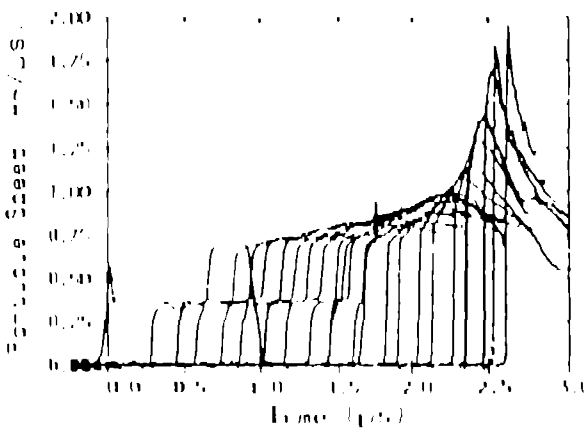

FIGURE 4. PARTICLE VELOCITY RECORDS FROM PBX 9404 PRECURSOR SHOCK EXPT

Desensitization is clearly evident in the 9404, with the run to detonation showing an increase of 270% over that expected from the pop plot, measuring from the arrival of the second shock (in the preshocked material) to where detonation was observed to occur. Onset of detonation is approximated by the overtake of the shock by the reactive wave, giving a slightly early value. At this point the wave velocity is estimated to be 7.3mm/μs (from the last two gauges) approaching the velocity of 8.1 mm/μs for PBX 9404. Pressure in this wave is estimated from the particle velocity to be about 15 GPa, well below the C-J pressure of 36 GPa. Nonetheless, detonation is delayed relative to the run anticipated from the pop plot for PBX-9404,[10] as shown in Figure 5.

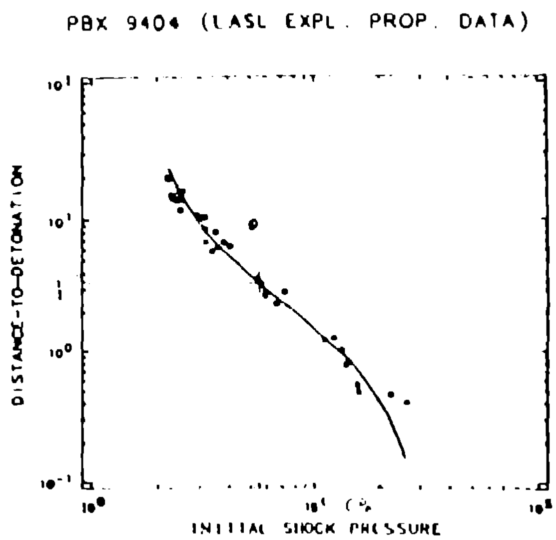

FIGURE 5. POP PLOT FOR PBX 9404.

Measuring from the coalescence of the second shock with the precursor also yields a slightly longer run than anticipated from the pop plot. The observed run is about 3% longer than the anticipated run, measuring only the region after coalescence of the two waves, giving nearly the behavior anticipated from the Campbell and Travis rule for the modified pop plot.

This slight extension may be explained by the shock interaction that takes place at the overtake of the preshock by the second shock. Shock dynamics requires a pressure reduction which is achieved by a small rarefaction wave traveling back into the doubly shocked region. Although, as in the PBX 9502, the small rarefaction is not observed directly, its passage is indicated by the reactive waves, appearing as a reduction in their velocity and a drop in their maximum particle velocity.[11] A contact discontinuity separates the doubly shocked, low temperature region from the singly shocked material at higher temperature.

The reactive waves which are clearly emerging behind the second shock as it travels through the preshocked region evidently do not contribute at all to the development of the reactive wave after wave coalescence.[11]

Overtake of the precursor by the second shock is considerably earlier than calculated by the MACRAME code. Acceleration of the second wave by reaction is a possible reason for early overtake. Another possibility is that the EOS used in MACRAME is not doing a very good job of calculating the off-Hugoniot states.

PBX-9501

Limited data obtained for PBX-9501 are shown in Figure 6. PBX-9501 differs from PBX-9404 primarily in the choice of energetic binders. Both materials have HMX as the principal constituent (PBX 9404 has 94% and PBX 9501 has 95%). PBX-9404 uses a mixture of nitrocellulose(3%) and chloroethyl phosphate (CEF) (3.9%) as the binder, with a blue indicator, diphenyl amine (DPA)(0.1%).[10] PBX-9501 includes a mixture of estane (2.0%) and a eutectic consisting of bis(2,2-dinitropropyl-acetal (BDNPA) and -formal (BDNPF) (2.5%) as the binder.[10] The process of manufacture is very similar for both materials.[10] The relative sensitivities of the two binders differ somewhat, as do the shock impedances of the different binders relative to the HMX. The rheologies[12] of the two plastics are believed to be a major factor in the very different skid responses of PBX-9404 and PBX-9501,[10,12] and provide a convenient variable related to hotspot behavior in preshock conditions.

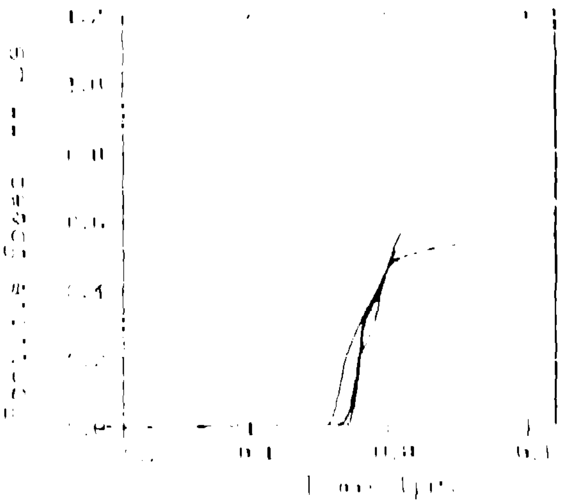

FIGURE 6. PARTICLE VELOCITY SHOCK FRONTS IN PBX-9501

An interesting feature of the several experiments done on PBX-9501, both single shock and double shock, is a set of small steps visible on the shock rise, and the long risetime consistently observed in this material. Risetimes of 35 ns in single shock experiments exceed risetimes usually seen, for example 15 ns in 9502 and 15-20 ns in 9404. Slight perturbations are seen in the shock rises, at particle velocities of 0.21 and 0.31 mm/μs. In the two shock experiment, the first shock exhibits comparable cusps at particle velocities of 0.28 and 0.33 mm/μs, giving total risetimes of between 52 and 110 ns. These small perturbations are shown in Figure 6.

Elastic response may be responsible for these waves, although these perturbations are at much higher pressures than the expected yield of the binder in PBX-9501, about 0.65 GPa.[12] Elastic waves generally have a higher wave velocity than the plastic wave, and consequently appear as a larger perturbation with increasing run distance, as do these perturbations in PBX-9501.

A reactive wave is clearly present in material which has been preshocked, as is evident in Figure 5. As was observed in PBX-9404,[11] the reactive wave is not accelerating, and neither is its particle velocity increasing. The small rarefaction has already traversed the region of the material in which this reactive wave is present.

DISCUSSION

The small rarefaction generated at wave coalescence, while not observed in the PBX-9502 experiments, apparently has a real effect[11] on reactive waves in PBX-9404 and PBX-9501. The x-t diagrams showing reactive waves are shown in Figures 7 and 8. After the second shock arrives, reactive waves emerge in the doubly shocked material, which is at a low temperature for the given pressure. When they encounter the small rarefaction, their growth is hindered, or even, in the PBX-9501 case, reversed. After wave coalescence with its accompanying contact discontinuity, the material is at a high temperature, with the shock running into material at normal porosity. In this region, a reactive wave develops which ultimately grows to detonation. This wave is probably unrelated to the wave in the preshocked region, since its growth to detonation is not accelerated by the preexisting reactive wave.

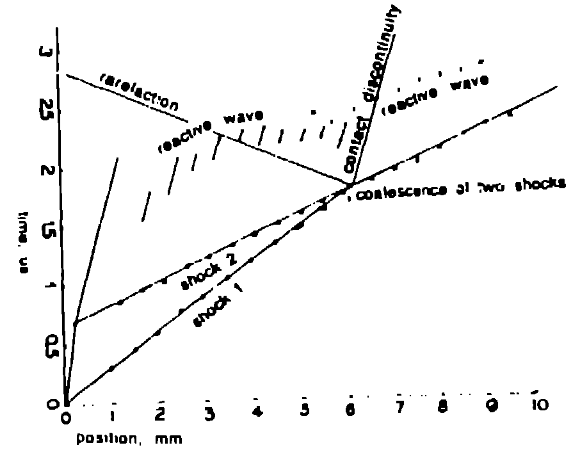

FIGURE 7. x-t DIAGRAM FOR PBX 9404 EXPT.

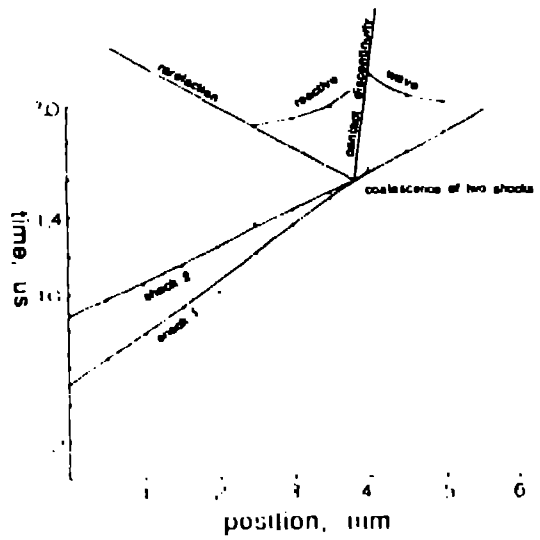

FIGURE 8. x-t DIAGRAM FOR PBX 9501 EXPT.

The small rarefaction and the contact discontinuity affect the bulk thermodynamic state of the material, rather than governing the reaction at hot spots. Only homogeneous initiation of detonation responds to bulk thermodynamic state, while heterogeneous initiation of detonation is largely sensitive to hot spots generated in the flow.

A homogeneous reaction mechanism has been suggested as active in doubly shocked material, because the density of the preshocked material is so high ([illegible] g/cm^3 in this case), with porosity largely removed. Heterogeneous character is never obliterated, because of the shock impedance mismatch between the binder and the HMX. Consequently hotspots will exist at any pressure in the plastic bonded material. The reactive waves throughout the experiment exhibit the profile of heterogeneous initiation, i.e., smooth growth behind the front. However, the growth is much slower than would be the case for a single-wave experiment.

While the reactive waves observed here resemble normal heterogeneous waves seen in single shock case, several factors suggest a homogeneous component active in the reactive waves observed in the preshocked region, in the doubly shocked material. The density of the preshocked material is high, so a very long run distance[13] is anticipated when compared to a heterogeneous detonation in a material at this porosity.

The velocity of the reactive wave is almost the same as that of the second shock in PBX-9404. In PBX-9501, the reactive wave actually decelerates relative to the second shock. The x-t planes shown in Figures 7 and 8 show the evolution of the reactive wave, with little acceleration until a wave is well established in the singly shocked region. The particle velocity at the shock arrival front never increases, strongly atypical of a heterogeneous mechanism, although the wave lacks the very localized profile and otherwise constant particle velocity seen in the classic homogeneous case.[14]

Homogeneous detonations may best be characterized by their extreme state-sensitivity.[14] The pressure drop in this rarefaction, from 5.60 to 5.54 GPa, corresponds to a drop of 20 K or less in the bulk temperature. A homogeneous wave is more likely to exhibit this extreme state sensitivity than one with substantial heterogeneous character.

A homogeneous reactive wave is unlikely to contribute to the heterogeneous detonation after wave coalescence, because the two processes proceed in different regions in the material. The x-t diagram and the long run to detonation from coalescence both suggest the non-interaction between the reactive wave in the preshocked region and that in the singly shocked material.

The usual model for preshock desensitization in PBX materials relies on hotspot removal. The $P^{2.2}\tau$ idea of Campbell and Travis suggests a time constant for hotspot removal. Sources of such a time constant may include compression time, time required for cooling of the hotspots, or some kind of reaction or pre-reaction time.

Shock traversal of hotspots with accompanying local sh s expected to occur on a timescale orders of magnitude faster[15] than the μs timescale applicable in this case. Lee and Tarver[16] give a rate of 44 us^{-1}, or a time constant of 20 ns for ignition at hotspots. R. B. Frey[17] gives a time of 35 nsec for full temperature rise and onset of ignition in shear bands at a pressure of 1.03 GPa.

Material rheology, particularly binder rheology, may entail response times on the order of 1 μs. The consistent value of τ between explosives argues against the importance of microscopic material mechanical properties. Our data indicates differences in the behaviors of PBX-9404 and PBX-9501, with the PBX-9501 showing substantially less reaction in the preshocked region. This may argue for the effect of the binder or other microscopic properties. Viscous collapse times on the order of 0.5 to 1 us have been calculated theoretically[18,19] for hotspots. These times are for complete compression, rather than the shear discussed above.

Thermal conduction is important in time-dependant behavior of hotspots. Hot spots result in local heating as opposed to bulk heating. Hot spot size, material thermal conductivity and the thermal differential between the hotspots and the bulk should be the governing factors in regulating the cooling of these hot spots.

Hotspot cooling should be about the same for both PBX-9404 and PBX-9502, since the major component, HMX, governs the thermal conductivity. Differences seen between PBX-9404 and PBX-9501 argue that bulk thermal conductivity leading to hot spot cooling is not the only factor governing the preshock desensitization, although allowances must be made for the different densities and grain sizes, hence different hotspot sizes and hotspot densities in the samples used.

Strain rate is related to both the compression time for the hotspots and thus thermal conduction argument, since adiabatic compression keeps heating local, while nonadiabatic compression allows dissipation of thermal energy during compression. In a shock, energy will remain localized as the bulk material reaches its final pressure and temperature, allowing reliable local ignition and rapid reactions in hotspots. Nonadiabatic compression permits compression with lower local temperature, (as well as bulk temperature) modifying hotspot reaction rates without manipulating other variables P and V. Thus loading rate allows some separation of hotspot behavior from bulk behavior. Varying strain rate can open up a channel for manipulation of hotspot behavior and temperature at tre same bulk pressure and density.

Comparison of PBX-9404 and PBX-9501 data may provide a test of local cooling, because of the different rheology of the binders in the two materials. A long risetime for the first shock in PBX-9501 may be related to the malleability of the estane binder,[11] and modify the input wave enough to provide information on reactive behavior as a function of strain rate.

Experiments using ramp wave inputs will extend previous work and further illuminate the importance of maintaining local temperature during compression.

Prereaction inside hotspots during preshocking may be a factor in deactivating the hotspots without removing or compressing them completely. Prereaction is supposed to deactivate hotspots either by altering the chemistry of the local material, or by increasing the internal pressure sufficiently to make the hotspot void incompressible, preventing local shear or "crushup" and transfer of energy from subsequent shocks to the material. Reaction in the preshock is not visible in our experiments. Since increasing the pressure inside hotspots is a constant volume process, our particle velocity gauging will not indicate any activity if this process is active. Extending the run of the preshock (increasing "τ") may reveal reaction and material acceleration during preshocking.

A reactive model developed by Pier Tang[20] reproduces the data and indicates nearly no reaction in the first wave, suggesting desensitization via mechanical changes during compression or temperature effects, rather than desensitization by partial chemical reaction.

SUMMARY

Desensitization may be due to bulk thermal effects or to hot spot phenomena, specifically, pore collapse, temperature differences, and changes due to chemical reaction behind the first wave.

A pressure drop is required by the shock dynamics of wave coalescence in the two shock experiment. This pressure drop is not visible in PBX 9502 records, but may nonetheless be resulting in the extended run observed in PBX 9404 after coalescence of two waves. A reactive wave is emerging in the preshocked material, but apparently does not contribute to the detonation, as indicated by an extended rather than truncated run after coalescence. This fact and the sensitivity of this reactive wave to local conditions argue for some homogeneous character.

Campbell and Travis[1] indicate that desensitization increases with increasing time between the first and second shocks, allowing more time for pore collapse, an idea supported in work done by R.E. Setchell.[3,4] However, Andreev et al.[21] indicate that material is more sensitive when the first wave creates more heterogeneities. We intend to further investigate separation of these mechanical and thermodynamic parameters by extending the work to include further comparison of PBX-9404 and PBX-9501, examination of a homogeneous detonating material with and without incompressible hot spots, and obtaining good estimates of relative temperatures in the single and double shock cases.

ACKNOWLEDGEMENTS

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