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REACTIVE WAVE GROWTH IN SHOCK-COMPRESSED THERMALLY DEGRADED HIGH EXPLOSIVES

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We have performed experiments to study the effect of thermal degradation on shock sensitivity and growth to detonation of several high-density plastic bonded explosives, confined in stainless steel cells. Assemblies were heated *in situ* in the target chamber of a light-gas gun. Confinement was varied to allow, in some cases, for thermal expansion of the explosive, and in other cases to vent the decomposition gases. Particle velocity profiles were measured using VISAR at a LiF window interface. Results for the IHE PBX-9502 showed that its sensitivity to shock initiation could be dramatically increased or decreased depending on the confinement conditions during heating. Effects were much less pronounced for PBX-9404 and PBX-9501.

INTRODUCTION

Shock initiation sensitivity of heated explosives, particularly insensitive high explosives (IHEs) containing TATB (triaminotrinitrobenzene), has received much attention in recent years. (1,2) The susceptibility of thermally degraded high explosives (HEs) and IHEs to violent reaction is the focus of studies to guarantee safety of explosive systems. To predict DDT in HEs it is necessary to know the shock initiation behavior of the material. While it was clear that heated IHEs were sensitized to shock initiation, the controlling parameters had not been identified. We chose to focus on the effect of confinement of heated HEs subjected to shocks. In brief, we found that the sensitivity of the heated system could be either increased or decreased relative to ambient IHE, depending on the heating conditions. Much less effect was observed for heated HMX- (tetranitro tetraazacyclooctane) based HEs.

It is reasonable to expect temperature to affect shock sensitivity, but it may do so via different mechanisms, and the effects can act in opposite directions. Chemical reaction rates increase with temperature. Any decomposition products may be more or less sensitive than the pristine material. But there are also physical effects that accompany heating: phase transitions, thermal expansion (affecting porosity), trapped gases inside voids, and fractures that affect particle size. Confinement conditions may play a crucial role in the changes that accompany heating by affecting both chemical and physical processes. Confinement allows the decomposition products to remain in contact with the bulk material, perhaps enhancing autocatalytic processes. Formation of low-volume products may be favored at equilibrium. Also, dissolution of products into the lattice may alter fracture patterns, and confinement can limit thermal expansion.

EXPERIMENTAL

We conducted a series of gas-gun shots on heated PBX 9404, PBX 9501 and PBX 9502 using different physical confinement conditions. The explosive pellets (0.75-in diameter) were made at Pantex by hot-pressing the molding powders. Our experimental arrangement is shown schematically in Fig. 1. Briefly, an explosive pellet was heated inside a stainless steel cell with at least 0.5-in wall thickness to provide strong confinement. LiF was chosen as the window to allow acquisition of

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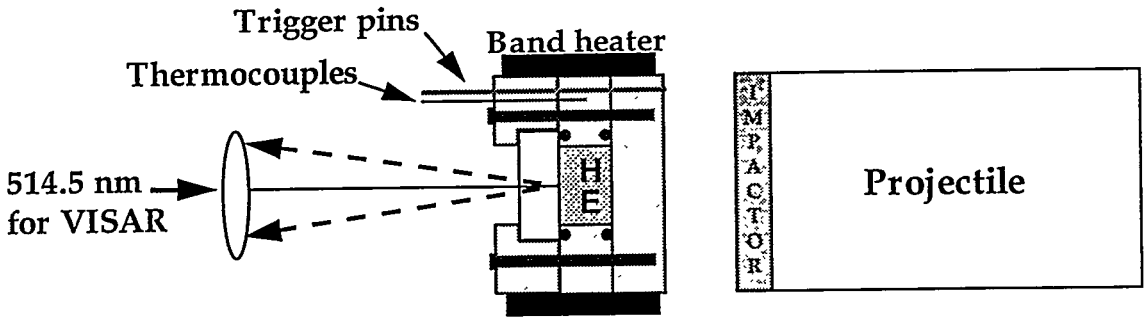

FIGURE 1. Schematic of experimental arrangement.

VISAR signal because it could be heated to the temperatures of interest. The cell was sealed with silicone or Viton o-rings, but in some cases the o-rings were cut to allow escape of the decomposition gases. Different cell thicknesses were used: some were matched to the HE pellet thickness, others were oversized to allow the pellet to expand during heating.

The target assembly was heated *in situ* inside the target chamber of a 63-mm light-gas gun. A band heater around the cell was used to heat the HE assembly at a 4°C/min ramp to a final temperature. In some experiments we allowed the HE to soak at the temperature for up to an hour. Three thermocouples were used to monitor the temperature at various positions in the cell. These temperatures agreed to within 4°C. We performed an initial series of shots on heated inert targets to check for target tilt introduced via heating, and observed no significant deviation from ambient shots. Tables 1 and 2 provide descriptions of the various shot assemblies and conditions for PBX 9502 and PBX 9501. The LiF window included a 0.020-in-thick LiF buffer to protect the diffuse VISAR reflector. We used a dual-delay leg VISAR system to record the particle velocity in the LiF.

RESULTS AND DISCUSSION

Results of 7 shots on PBX-9502 are shown in Table 1. In all cases the impact velocity was between 1.98 and 2.01 km/s, giving an initial shock to the PBX 9502 of approximately 8.6 GPa. If there was no growth in the shock wave due to reaction, the reshock from the LiF window raised the pressure in the HE to about 11.5 GPa. In shots 1, 3 and 5 there was no evidence of reactive wave growth; the HE behaved much like an inert material. Shot #2 showed wave growth after the reshock from the window, but not sufficient to lead to detonation. The target for shot #6 was heated to 240°C for one hour, then cooled overnight before firing the shot. The measured particle velocity was slightly higher than others measured

TABLE 1. Summary of shot parameters and results for PBX 9502 (ρ=1.89 g cm^{-3}), P=8.6 GPa
(PBX 9502 -- TATB/Kel-F 800 95/5 wt. %)

Shot #	Temperature (°C)	HE Thickness (in)	Cell Thickness (in)	Soak time (min)	o-rings	Result (mm/ s)
1	150	0.196	0.196	30	yes	0.65
2	150	0.197	0.221	60	yes	0.65 - 0.95
3	240	0.196	0.195	60	yes	0.65
4	240	0.197	0.227	60	yes	Detonation
5	240	0.197	0.195	30	cut	0.65
6	240 / 20	0.197	0.196	60/cool	cut	0.72 - 0.8
7	150	0.393	0.413	30	yes	Detonation

TABLE 2. Summary of Shot parameters and results for PBX 9501 (ρ=1.84 g cm^{-3}, 0.197 in thick)
(PBX 9501 -- HMX/estane/BDNPA(F) 95/2.5/2.5 wt. %)

Shot #	Temperature (°C)	Soak time (min)	Impact Velocity (mm/μs)	Shock arrival time (μs)	Result
1	20	NA	0.9	1.12	Detonation
2	200	30 min (vent)	0.9	0.96	Detonation
3	200	8 min	0.9	1.00	Detonation
4	200	20 min	0.9	1.35	Detonation
5	200	8 min	0.75	1.65	Detonation on reshock
6	200	30 min	0.75	2.0	Detonation on reshock

at elevated temperatures and did show some growth after reshock. The two samples that detonated had some additional space to allow for expansion of the pellet. These results are in rough agreement with run-distances to detonation measured in previous work of Dallman and Wackerle(1) and Urtiew, et al. (2) for heated IHEs. In those experiments the materials were not confined, allowing the material to expand and showing increased shock sensitivity at elevated temperature. These present results indicate that an important mechanism may be the change in density. Indeed, we do not expect significant thermal decomposition to occur at the temperatures and soak times of these studies. Any increase in sensitivity due to enhanced reaction rates at elevated temperatures appear to be effectively countered by the strong confinement limiting expansion. Concerns of increased sensitivity need to be seen in light of the whole system, because strong confinement can mitigate the potential sensitization.

The results of experiments on PBX 9501 are presented in Table 2. Impact velocities of 0.9 and 0.75 km/s correspond to initial shocks in the HE of 5.7 and 4.5 GPa, respectively. All targets were made with cells and pellets of matched thicknesses, and full o-rings, usually silicone, were used to seal the cells. The target on shot #2 was assembled with Viton o-rings that allowed venting of gases that had built up as a result of decomposition.

PBX 9501 showed less dramatic changes than were observed for PBX 9502. The arrival time of the shock/detonation wave at the window interface, measured referenced to an arbitrary but uniform time, t=0, was noticeably affected due to heating in the fashion described. If the arrival times corresponded to a difference in the rate of shock build-up to detonation, then it appeared as though heating for longer times acted to retard the reactive wave growth. Unlike the PBX 9502 experiments, the temperature of these shots did allow for substantial decomposition prior to the shot. Indeed, we had two experiments that cooked off before the gun shot. It is therefore reasonable to expect high gas pressures in the cell, especially in shots 4 and 6. Also note that the shot temperature was above that for the solid - solid phase transition temperature to form δ-HMX.

Five experiments were also performed on PBX 9404 (HMX/NC/CEF/DPA 94/3/3/0.1 wt %) at temperatures from 140 to 200°C and impact velocities from 0.7 to 1.2 km/s. All samples had grown to detonation within the 0.197-in thickness of the pellet. We had no uniform reference time on those shots, so relative differences between the shots are not instructive. At best we can say that no dramatic changes occurred for this HE due to heating.

Any detailed understanding of the effects of temperature on shock sensitivity ultimately requires a complete description of the thermally degraded HE material. In particular we would like to know, as functions of temperature, soak time, and confinement, parameters such as the extent of decomposition, transient and final decomposition products and their sensitivities, evolved gas pressures, and porosity (both closed and connected). To aid in understanding the results of our gas-gun shots, we performed preliminary experiments aimed at measuring the pressure of evolved gases as a function of temperature and soak time. We assembled cells with explosive samples