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LX-04 is a widely used HMX-based plastic bonded explosive, which contains 85 weight % HMX and 15 weight % Viton binder. The sensitivity of LX-04 to a single stimulus such as heat, impact, and shock has been previously studied. However, hazard scenarios can involve multiple stimuli, such as heating to temperatures close to thermal explosion conditions followed by fragment impact, producing a shock in the hot explosive. The sensitivity of HMX at elevated temperatures is further complicated by the beta to delta solid-state phase transition, which occurs at approximately 165˚C. This paper presents the results of shock initiation experiments conducted with LX-04 preheated to 190˚C, as well as density measurements and small scale safety test results of the δ phase HMX at room temperature. This work shows that LX-04 at 190˚C is more shock sensitive than LX-04 at 150˚C or 170˚C due to the volume increase during the β to δ solid phase transition, which creates more hot spots, and the faster growth of reaction during shock compression.

INTRODUCTION

The shock sensitivity of unconfined and confined LX-04 heated to 150˚C and 170˚C has been studied using embedded manganin pressure gauges (1) and the results compared to that of ambient temperature LX-04 (2-4). Both series of experiments showed increased shock sensitivity of heated LX-04 compared to ambient. However, they did not show significant differences between the two temperatures, even though the temperature of 150˚C is below the predicted temperature for the beta to delta phase transition and 170˚C is above. Increased sensitivity of LX-04 at these temperatures was explained to be mainly due to the density changes and to faster growth of hot spot reactions following ignition. The experiments with LX-04 heated to 170˚C were fired approximately 20 minutes after the explosive reached 170˚C (2). Since solid phase transitions are sometimes quite slow, the HMX in these LX-04 charges held at 170˚C for 20 minutes may not have been fully or partially converted from the beta to delta phase.

A series of shock initiation gun experiments were conducted with LX-04 preheated to 190˚C and held at temperature for over an hour. The LX-04 chemical kinetic decomposition model of Tarver and Tran (5) predicts that 90 mm diameter cylindrical charges can be held at 190˚C for 6 hours without undergoing thermal explosion. The phase transition is predicted to completely convert producing a 6% volume expansion of LX-04. The paper also presents the density measurements and other characteristics of the delta phase HMX cooled to room temperature. There is controversy as to how long HMX remains in the delta phase after cooling. Saw (6) claims that the HMX remains in the delta phase for several months, but Smilowitz et al. (7) show an immediate reversal. Ignatov et al. (8) have presented evidence that the friction sensitivity of HMX-based high explosives in the delta phase is much greater than in the beta phase. In fact, they claim that its sensitivity is close to that of primary explosives. The purpose of this work is to determine the shock sensitivity of LX-04 containing delta HMX at 190˚C and compare it to previous LX-04 studies at 150˚C and 170˚C.

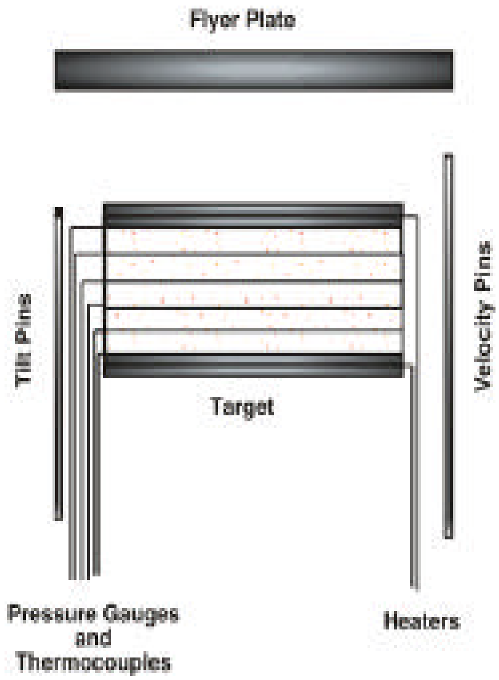

FIGURE 1. Target assembly for the 100 mm gas gun thermal experiment.

EXPERIMENTAL TECHNIQUES

Experiments were carried out with a 100 mm powder driven gas gun. The target consisted of LX-04 slowly heated to a temperature of 190˚C and held there for over an hour to assure a complete β to δ phase transformation before projectile impact. The unconfined target assembly is illustrated in Fig. 1. Nichrome foil spiral heaters were placed on both sides of LX-04 and separated from it by a thin aluminum disc for better heat distribution into LX-04. Packages containing thermocouples (TC's) and manganin pressure gauges were imbedded between the individual discs of LX-04. Details of the manganin gauges were described in our previous publications (1-4). The heating rate of the target was held constant at 3˚C/min until the temperature reached 180˚C. Then the heating rate was changed to 1˚C/min until the controlling TC reached the temperature of 190˚C. The target was held at that temperature for one hour before the gun was fired. The TC temperature profiles at various gauge locations are shown in Fig. 2. The decrease of temperature shown by internal TC's is an indication that the endothermic transition is occurring (9). The impact pressure was measured at the Al-LX-04 interface, and the build-up pressures were registered at various depths in LX-04. Pressure records from one of the experiments are shown in Fig. 3.

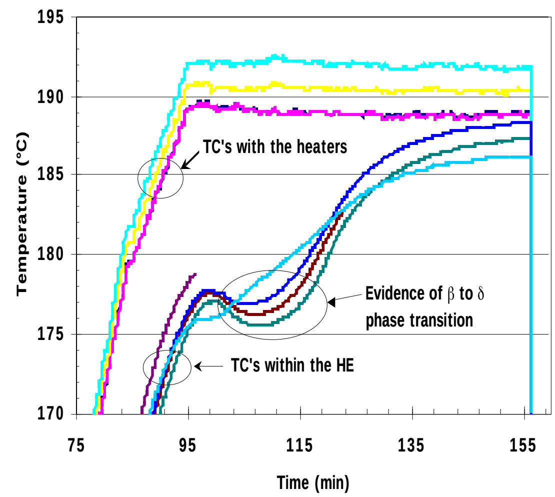

FIGURE 2. Thermal profiles of heated LX-04 as measured by thermocouples at various locations

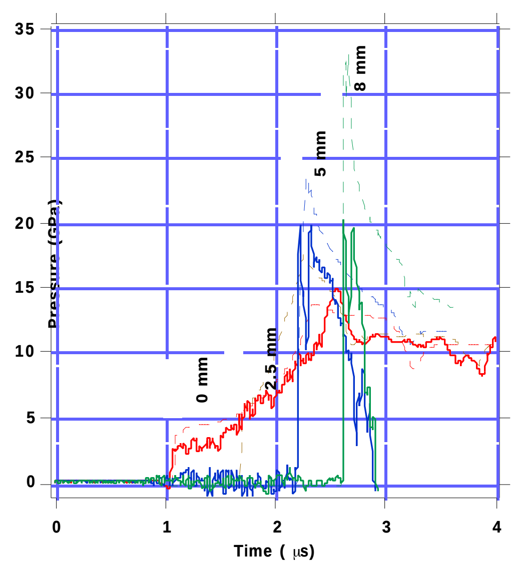

FIGURE 3. Pressure profiles in the heated LX-04 sample impacted by an aluminum flyer plate at a velocity of 0.92 mm/µs. Experiments are represented by solid lines and calculations by broken lines.

for an impact velocity of 0.92 km/s, resulting in the impact pressure of 3.0 GPa. Four experiments were conducted with impact velocities of 0.920, 0.888, 0.765, and 0.679 km/s, yielding impact pressures of 3.00, 2.83, 2.30 and 1.90 GPa, respectively.

Small-scale tests were performed on LX-04 samples heated to 140˚C and 190˚C and then cooled to room temperature. The results are shown in Table 1. Densities and sound velocities experience drastic changes after heating to 190˚C. The three small-scale qualitative sensitivity tests, drop hammer, friction, and Differential Scanning Calorimetry (DSC), show no significant sensitization when these heated samples were tested at room temperature. No significant weight losses were observed during these tests.

REACTIVE FLOW MODELING

The Ignition and Growth reactive flow model (10) was used with equation of state parameters for ambient and hot aluminum and hot Teflon. Ignition and Growth rate law parameters were determined for hot LX-04 at 190˚C. Table 2 shows the comparison of these parameters for the four temperatures. The decreases in LX-04 density when heating from room temperature to 150˚C or 170˚C were attributed to thermal expansion. The linear coefficient of thermal expansion (CTE) of LX-04 is 98.9 μm/m˚C [11].

TABLE 1. Small Scale Test Results on Heated and Then Cooled LX-04 Samples

Test	Ambient LX-04	140˚C for 22 hours	190˚C for 2 hours
Weight Loss %	0	-1.12	-0.25
Density (g/cm^3)	1.859	1.784	1.543
Sound speeds (km/s)	2.62/1.392 (Longitu-dinal/Shear)	2.18/1.255	1.32/0.988
Drop Hammer (cm)	84 (70.7-99.6)	74 (66.3-82.1)	119 (107.4-130.8)
Friction (5mg)	0/10@36kg	1/10@32kg	1/10@36kg
DSC (˚C)	284.4	283.5	283.3

The lower density at 190˚C is caused by an additional 6% volume expansion due to β to δ phase transformation in HMX. The ignition term of the reaction rate law depends on the degree of shock compression of the solid explosive and thus ignites more material as the initial density decreases. The growth of the ignited hot spots into the surrounding explosive particles depends on the shock temperature of these particles. The growth of reaction coefficient G_1 was increased from 100 at 25˚C to 210 at 150˚C and 170˚C and finally to 300 at 190˚C to match the experimental pressure histories and run distances to detonation. The value G_1 = 300 yielded good agreement with all four experiments. The calculated traces (dashed lines) for the highest impact pressure experiment are compared to experiment in Fig. 3.

SHOCK SENSITIVITY RESULTS

The 100 mm gun impact tests of LX-04 at 190˚C show a dramatic increase in shock sensitivity, which is illustrated by the Pop Plots shown in Fig 4. 190˚C LX-04 is considerably more sensitive than 150˚C LX-04 and 170˚C LX-04, which exhibited approximately the same shock sensitivity as ambient temperature PBX 9404, the most sensitive HMX-based plastic bonded high explosive. The 190˚C LX-04 run distances are similar to those of 1.72 g/cc PETN. This increased sensitivity is mainly due to the β to δ phase transformation, which takes place at temperatures exceeding 176˚C, as illustrated by the temperature decreases on the thermal records in Fig. 2. It appears that, in the previous experiments on 170˚C LX-04, there was not sufficient time allowed for the phase transition to occur before firing. Finally, the measured pressures at the aluminum-LX-04 interfaces were used to construct a new EOS for the hot unreacted LX-04 at 190˚C, which can be represented by $U_s = 1.25 + 1.6\ u_p$, where U_s is the shock velocity and u_p the particle velocity.

TABLE 2. Ignition and Growth reaction rate and density changes for LX-04 at various temperatures

Temperature (C)	25	150	170	190
Density (g/cm^3)	1.866	1.79	1.77	1.66
Growth Rate G_1	100	210	210	300
Reference	2	4	2	New

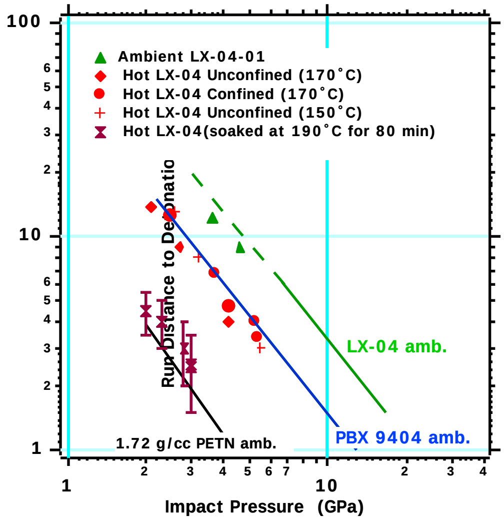

FIGURE 4. "Pop Plot" for heated LX-04 showing its sensitivity to impact as compared to other sensitive high explosives.

CONCLUSIONS

LX-04 with HMX in the δ phase at 190˚C is found to be more shock sensitive than LX-04 with HMX in the β phase at 150˚C and 170˚C. The faster growth rates of hot spot reaction sites at this elevated temperature increase the shock sensitivity. This is evident from the reaction growth parameter G_1, which had to be increased about 50% for the 190˚C case to properly model the pressure build-up to detonation. However, the β to δ phase transformation also plays an important role by increasing the void volume and thus the number of hot spot ignition sites formed during shock compression. Slow chemical decomposition of the LX-04 at 190˚C (5) may also contribute to increased sensitivity. Thus LX-04 and other HMX-based explosives that have undergone the β to δ transition should be handled carefully. Whether the HMX in LX-04 remains in the δ phase when it is cooled to room temperature and, if so, for how long still needs to be determined.

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REFERENCES

1. Urtiew, P. A., Erickson, L. M., Hayes, B., and Parker, N. L., *Combustion, Explosion and Shock Waves* **22,** 597-614 (1986).
2. Urtiew, P. A., Tarver, C. M., Forbes, J. W., and Garcia, F., *Shock Compression of Condensed Matter-1997*, Schmidt, S. C., Dandekar, D. P., and Forbes, J. W., eds.. AIP Press (429), 1998, pp. 727-730.
3. Forbes, J. W., Tarver, C. M., Urtiew, P. A., and Garcia, F., *Eleventh International Detonation Symposium*, Office of Naval Research, Aspen, CO, 1998, pp. 145-152.
4. Tarver, C. M., Forbes, J. W., Urtiew, P. A. and Garcia, F., *Shock Compression of Condensed Matter-1999*, Furnish, M.D., Chhabildas, L.C. and Hixson, R.S., eds., AIP Press (505), 2000, pp. 891-894.
5. Tarver, C. M. and Tran, T. D., "Thermal Decomposition Models for HMX-Based Plastic Bonded Explosives", *Combustion and Flame*, 2003, in press.
6. Saw, C. K., LLNL, private communication, 2003.
7. Smilowitz, L., Henson, B. F., Asay, B. W. and Dickson, P. M., "A Model of the Beta – Delta Phase Transition in PBX 9501", Twelfth International Detonation Symposium, San Diego, CA, August 2002, in press.
8. Ignatov, O. L., Lashkov, V. N., Lobanov, V. N., Strikanov, A. V. and Shestakov, A.N., "Effect of HMX Polymorphous Transformations after Thermal Treatment on Its Sensitivity to Impact Friction", International Workshop on New Models and Hydrocodes, Edinburgh, UK, May 2002, in press.
9. Maienschein, J. L., Wardell, J. F., Weese, R. K., Cunningham, B. J. and Tran, T. D., "Understanding and Predicting the Thermal Explosion Violence of HMX-Based and RDX-Based Explosives – Experimental Measurements of Material Properties and Reaction Violence", Twelfth International Detonation Symposium, San Diego, CA, August 2002, in press.
10. Tarver, C. M., Hallquist, J. O., and Erickson, L. M., *Eighth Symposium (International) on Detonation*, Naval Surface Warfare Center NSWC MP 86-194, Albuquerque, NM, 1985, pp. 951-961.
11. Weese, R. K., LLNL, private communication, 2003.