COMPRESSIVE PROPERTIES OF PBXN-110 AND ITS HTPB -BASED BINDER AS A FUNCTION OF TEMPERATURE AND STRAIN RATE

W.R. Blumenthal, D.G. Thompson, C.D. Cady, G.T. Gray III, and D.J. Idar
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

The compressive constitutive strength behavior of PBXN-110 and its HTPB-based binder system was measured as a function of temperature (-55°C to +20°C) and strain rate (10^{-3} and 2000 s^{-1}). PBXN-110 is a plastic bonded explosive (PBX) with relatively high binder content that contains 88wt% HMX and 12wt% HTPB-based binder. A pure analog of the PBXN-110 binder system was tested for comparison to the strain rate and temperature dependence of the composite PBXN-110. As expected, the strength of PBXN-110 was found to exhibit strong temperature and strain rate dependence, attributable to the large fraction of the very soft HTPB binder. The strength of the pure HTPB binder analog was challenging to measure using the split Hopkinson pressure bar (SHPB) because of its extreme softness, however satisfactory results were obtained at and below room temperature by optimizing the SHPB technique. These measurements provide the basis to develop and validate predictive material strength models for PBXN-110.

INTRODUCTION

Physically-based constitutive models are needed to predict the mechanical behavior, damage evolution, and performance of modern energetics for their safe application. However, the fundamental micromechanisms controlling these processes remain poorly understood. A robust constitutive model would ideally account for many important engineering effects such as: 1) thermo-mechanical loading conditions (stress-strain state, confinement, strain rate, and temperature); 2) scale (micro-to-macroscopic); 3) microstructural variables (constituent morphology and size, binder type and content); and 4) loading history (deformation and damage evolution).

A number of previous studies have probed the constitutive response of a wide variety of plastic bonded explosives (PBX).[1-10] High-rate studies[1,3,4,6,8-10] have shown that: a) the peak compressive strength is strongly influenced by strain rate and temperature, b) after the peak strength has been achieved damage accumulation reduces the flow strength, and c) specimen geometry and lubrication are important because the slow sound speed of PBX can affect stress equilibrium within the specimen and friction can affect the strength and failure process. Low-strain-rate studies on PBXs[2,5,7] have

similarly shown that the peak compressive strength and loading modulus increase with decreasing temperature and increasing strain rate.

This paper describes the results of an experimental program to measure the compressive mechanical response of PBXN-110. A pure analog of the PBXN-110 binder system was also tested for comparison to the strain rate and temperature dependence of PBXN-110. The influence of strain rate and temperature on the mechanical response of these materials provides part of the basis to develop and validate predictive material strength models for a range of energetic composites and their constituents.[11]

EXPERIMENTAL

MATERIALS

The investigation evaluated both the plastic-bonded explosive PBXN-110 and a pure analog of its multi-component binder system based on hydroxyl-terminated polybutadiene-based polymer (hereafter the polymer binder will be referred to as simply HTPB). Both the PBXN-110 and the HTPB analog were received from the U.S. Naval Surface Warfare Center, Indian Head, MD.

PBXN-110 is composed of 88wt% HMX in a binder system containing 5.32 wt% hydroxyl-terminated polybutadiene (HTPB) as a monomer, 5.32 wt% isodecyl pelargonate as a plasticizer, 0.75 wt% lecithin (processing agent) and A02246 (antioxidant), 0.51 wt% isophorone diisocyanate (IPDI) as a curative, and 0.10 wt% dibutyltin dilaurate (DBTDL) as a curing catalyst. The composition of the pure HTPB binder analog will be discussed later in this section.

The PBXN-110 was received in 3.2-mm and 19-mm thick sheets. The average density was 1.659 g/cm^3 with a standard deviation of 0.005 g/cm^3.
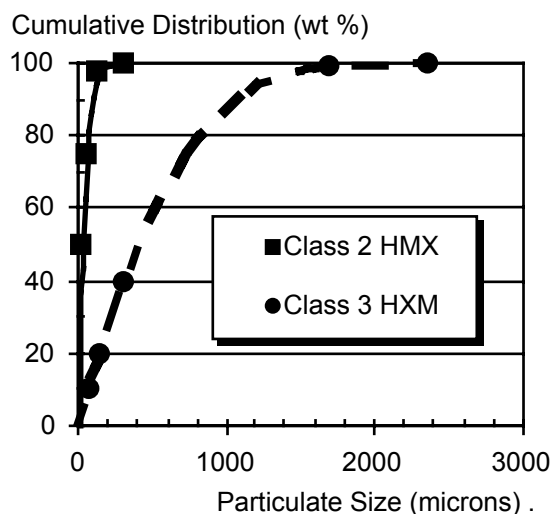

FIGURE 1. PARTICULATE SIZE DISTRIBUTIONS OF CLASS 2 AND CLASS 3 HMX USED IN PBXN-110.

The HMX in PBXN-110 is composed of a 3:1 ratio of coarse-to-fine grade HMX. The coarse grade (class 3) HMX has a very broad range of particulate sizes between 75 and 2300 microns with an average of about 400 microns. The fine grade (class 2) HMX contains particulates up to 300 microns in size, but at least 75% of the particulates are below 45 microns with an average particle size of about 10 microns. The cumulative distributions of the two HMX classes are shown in Figure 1 based on MIL-DTL-45444C. Note that there is a potential for very large HMX crystals to dominate the volume of a given Hopkinson bar specimen, strongly affect the deformation and damage evolution response, and lead to high variability in the statistical results.

The high fraction of pliable HTPB binder in PBXN-110 results in a soft composite material. This softness creates a unique machining challenge because specimens can not be machined using conventional lathe or milling techniques. Instead, PBXN-110 compression specimens were core-punched from the sheets. The first set of specimens was hand-cored (labeled *hand-cored* in figures). Approximately one year later a

second set of specimens was created by mounting the core punch in a press for better specimen precision and reproducibility (labeled *precision-cored*). Because the PBXN-110 sheets were made to a thickness equal to the required specimen length, no additional machining of the specimen faces was required.

The HTPB binder analog consisted of 46.65wt% hydroxyl-terminated poly-butadiene (HTPB) monomer, 46.65 wt% isodecyl pelargonate (IDP) plasticizer, 6.174wt% isophorone di-isocyanate (IPDI) reactant, 0.435 wt% 4,4'-methylene bis(2,6-di-tert-butylphenol) anti-oxidant, and 0.09 wt% di-butyltin dilaurate (DBTDL) catalyst. Hence, the HTPB monomer, the IDP plasticizer, and the IPDI curative were proportionally scaled; however, the weight fractions of catalyst (DBTDL) and anti-oxidant (AO4426) were nearly identical to the actual PBXN-110 binder.

The HTPB binder analog was mixed, injected into PVC tubes, then cured for 48 hours at 40°C. Specimens were made by conventional single-point lathe machining while cooling the HTPB with a liquid nitrogen spray to sufficiently stiffen the material.

LOW STRAIN RATE TESTING

Quasi-static compression tests were conducted on the PBXN-110 material using a screw-driven Instron model 5567 workstation with a Bemco environmental chamber. Strain was measured using cross-head displacement and was validated by specimen extensometer measurements. The test temperature was obtained by ramping at 1°C/min and holding the final temperature for approximately 20 minutes prior to testing. Tests were conducted in air at temperatures of -15°C, 22°C, and 50°C $\pm$ 0.5°C as determined by averaging two thermocouples mounted inside the upper and lower platens.

PBXN-110 quasi-static compression specimens were 9.5-mm in diameter by 19-mm long and were tested without lubrication. Cross-head rates of 13 mm/min and 127 mm/min were applied (initial strain rates of 0.01 and 0.1 s^{-1}, respectively). Average strength and standard deviation for each condition was determined based on three to six tests.

Quasi-static compression of HTPB specimens 6.3-mm diameter by 6.3-mm long was performed using an MTS model 880 load frame. A strain rate of 0.001 s^{-1} was applied and measured using an extensometer placed between the platens. Specimen faces were lubricated with molybdenum disulfide grease.

HIGH STRAIN RATE TESTING

High rate compression experiments were conducted on both PBXN-110 and the HTPB binder system using a split-Hopkinson pressure bar (SHPB) with 9.4-mm diameter AZ31B magnesium alloy pressure bars to maximize the resolution of stress[12].

In addition, a specimen thickness of 3.2-mm was used to help achieve rapid stress equilibrium throughout the specimen as required for valid uniaxial stress testing[12]. PBXN-110 specimens were 6.4-mm in diameter while HTPB specimens were 8-mm in diameter.

Tests were conducted at a nearly constant strain rate of 2500$\pm$500 s^{-1} at temperatures from -55°C to +22°C. Specimens were lubricated using a thin layer of molybdenum disulfide grease.

RESULTS AND DISCUSSION

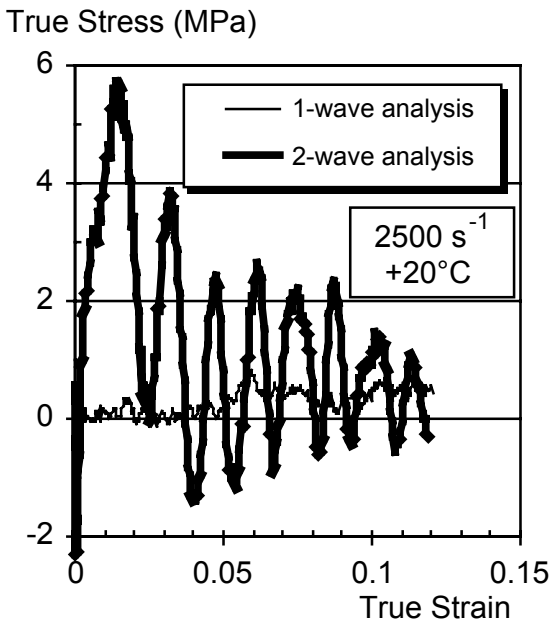VALIDITY OF THE SHPB RESULTS

True Stress (MPa)

FIGURE 2: HIGH STRAIN RATE STRENGTH OF HTPB BINDER IS ONLY VALID FOR STRAINS ABOVE 5%.

A valid, uniaxial SHPB test requires that the stress-state throughout the specimen achieve equilibrium during the test. The validity of each test was confirmed by calculating and comparing the 1-wave and 2-wave stress[12,13]. The stress-state is uniform throughout the specimen when the average 1-wave and 2-wave stresses are equal.

Figure 2 shows a comparison of the 1-wave and 2-wave stress for HTPB at 20°C. Due to the very low sound speed, strength, and dispersive/damping response of the HTPB binder, this condition represents the worst case for the validity of our SHPB results. Note that stress equilibrium is only confirmed after about 5% strain at a strain rate of 2500 s[-1]. Also note that the combination of low modulus magnesium pressure bars, low noise/high gain strain gage signal conditioners, and optimized specimen geometry allows strength measurements below the 1 MPa stress level.

PBXN-110

Figures 3 present the average compressive stress-strain behavior of PBXN-110 as a function of temperature for two quasi-static strain rates (~0.01 s[-1] and ~0.1 s[-1]). The quasi-static maximum flow stress (σ_m) is shown in the top plot; the strain-at-maximum stress value (ε_m) is shown in the middle plot; and the chord modulus (E) is shown in the bottom plot as a function of temperature. The quasi-static strength is not strongly temperature dependent compared to the modulus at these temperatures, but interestingly, the ductility parameter, ε_m, *increases* with decreasing temperature.

Fig. 3 also shows the results of three room temperature quasi-static tests of *hand-cored* specimens from an initial feasibility study along with the results obtained from testing thirty *precision-cored* specimens from the same sheet stock. Although the maximum strength of the two sets of specimens is statistically the same, the parameters ε_m and E are not. Specifically, the *precision-cored* ε_m was ~35% greater than the *hand-cored*, while the *precision-cored* modulus was ~25% softer than the *hand-cored* modulus. The reasons for these changes within a short period of a year are currently unknown.
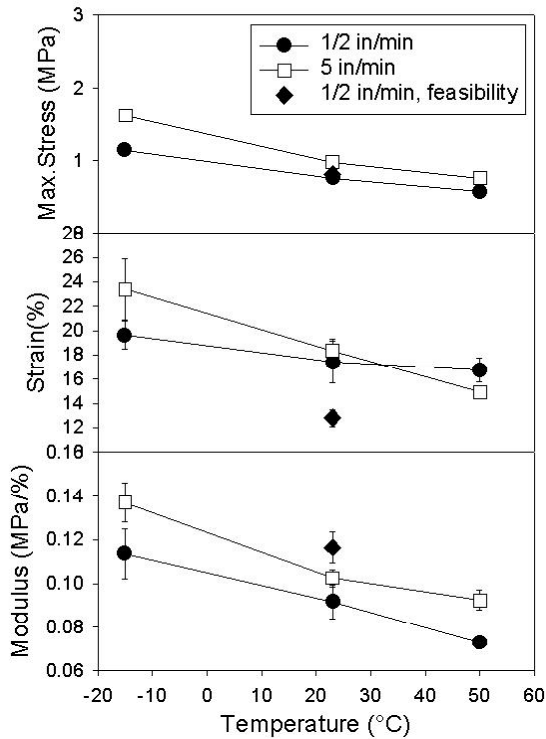
FIGURE 3. QUASI-STATIC COMPRES-
SION BEHAVIOR OF PBXN-110 AS A
FUNCTION OF TEMPERATURE.

FIGURE 4. HIGH-STRAIN-RATE
COMPRESSION OF HAND-CORED
PBXN-110 VERSUS TEMPERATURE.

The high-strain-rate stress-strain behavior of the *hand-cored* specimens of PBXN-110 is presented in Figure 4 as a function of temperature. Note that stress-state equilibrium is only achieved after about 5% strain (see Fig. 2), so that the initial loading portion of these curves *cannot* be used (e.g. for modulus or yield strength determinations). Specimens did not fail under any conditions and signs of damage were not apparent after 15% strain.

PBXN-110 shows little dependence of the strain-hardening rate on temperature, but a strong dependence of the apparent yield stress. Comparing Figures 3 and 4, PBXN-110 also exhibits strong strain-rate dependence of its flow strength (a factor of 5) at room temperature and below.
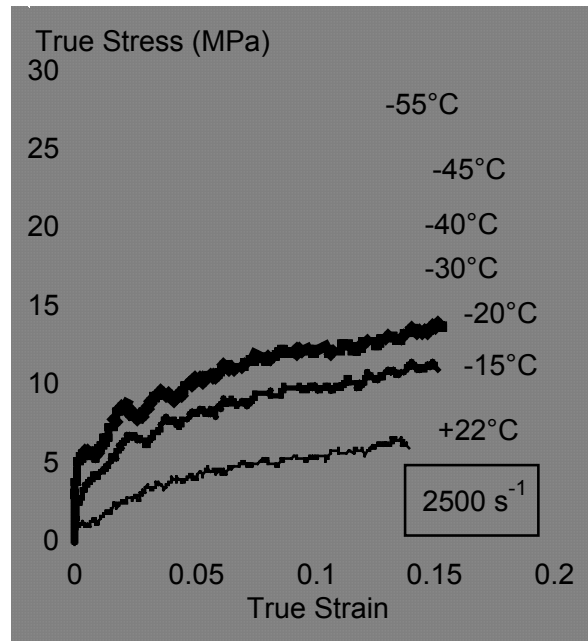
The temperature dependence of the flow stress of PBXN-110 at high strain rate (2500 s^{-1}) and an arbitrary strain of 10% is summarized in Figure 5. This plot shows a factor of 5 increase in flow stress at 10% strain from 0°C down to -55°C. Because the glass-transition temperature, T_g, of PBXN-110 is approximately -89°C, the quasi-static strength shows little temperature dependence from +40°C down to at least -15°C. High strain rate loading shifts the effective T_g to higher temperatures because of the inverse relationship of temperature and strain rate for thermally activated deformation. Hence, the strong temperature dependence of the high-strain-rate strength indicates that the dynamic T_g of PBXN-110 is being approached at -55°C.

Because of its binder phase, the behavior of PBXN-110 is distinct from other energetics: 1) its strength is very low (PBX-9501 is over ten times stronger) and 2) it is very ductile (ε_m is over 12% compared to less than 3% for PBX-9501). Note that the strain-at-maximum strength could not be

determined at high strain rates because the applied strain was restricted to about 15% for the striker bar length used.
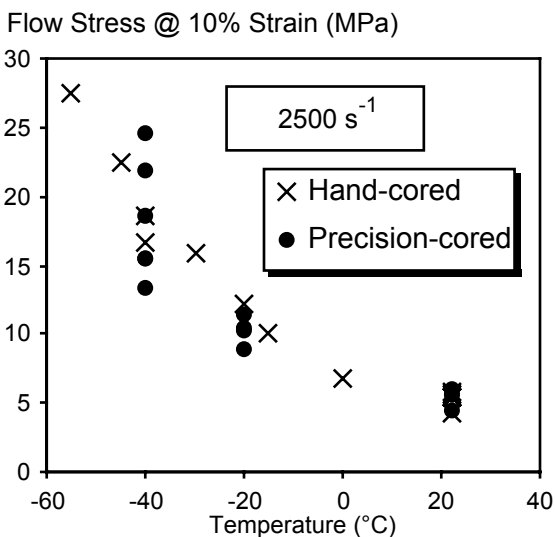
Flow Stress @ 10% Strain (MPa)

FIGURE 5. HIGH-STRAIN-RATE FLOW STRENGTH VERSUS TEMPERATURE FOR PBXN-110.

Figure 5 also shows repeated tests of *precision-cored* specimens of PBXN-110 at several temperatures in which the scatter in strength is large and increases significantly with decreasing temperature. This high level of scatter may be explained by the influence of the large HMX crystal size and distribution on the strength, however, the basis for the observed temperature dependence of the amplitude of the scatter is not understood. Perhaps fortuitously, *hand-cored* specimens fall close to the average of the *precision-cored* strengths and the shape of the stress-strain curves for the two sets of specimens are identical, in contrast to the quasi-static results.

HTPB-BASED BINDER SYSTEM

A preliminary study was conducted on an analog of the HTPB-based binder system. Figure 6 shows a plot of the high-strain-rate

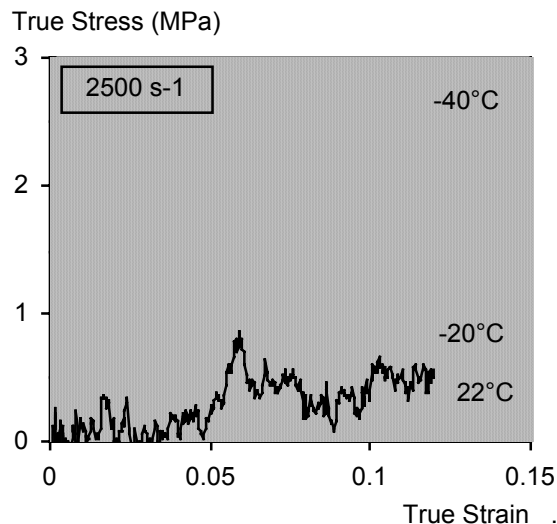stress-strain behavior from 22°C to -40°C for the HTPB binder analog.

True Stress (MPa)

FIGURE 6. HIGH-RATE STRESS-STRAIN BEHAVIOR OF PURE HTPB BINDER VERSUS TEMPERATURE.

The stress-strain response of the HTPB binder is essentially linear and completely reversible up to at least 15% strain and the strength is extremely low at both high and low strain rates. The usual noise from the SHPB diagnostic gages becomes such a significant fraction of the output signal that strengths below about 0.5 MPa cannot be accurately resolved.

A comparison of the quasi-static and high-strain-rate strength of the HTPB binder system at a nominal strain of 10% is summarized in Figure 7 versus temperature. Figure 7 shows that the HTPB binder exhibits strong strain-rate dependence at all temperatures, however the temperature dependence of the strength is very weak above -20°C. At high strain rate the strength increases dramatically by a factor of five from -20°C to -40°C.
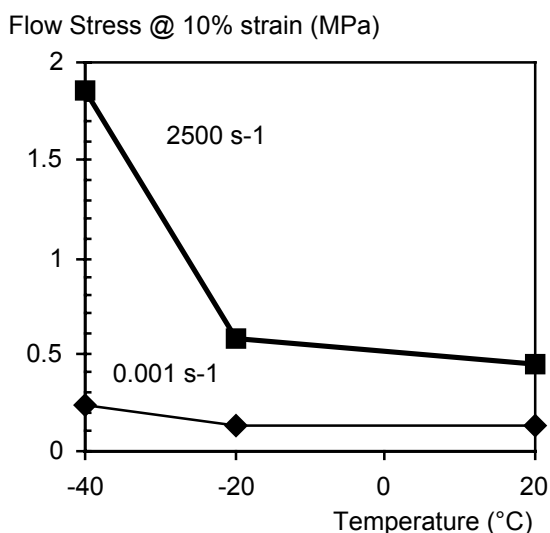
Flow Stress @ 10% strain (MPa)

FIGURE 7. FLOW STRENGTH OF THE HTPB BINDER.

CONCLUSIONS

The compressive stress versus strain response of PBXN-110 and its HTPB binder system were both found to depend strongly on the applied strain rate and to varying degrees on the test temperature.

The compressive strength of PBXN-110 at 10% strain and a strain rate of 2500 s^{-1} is over 10 times lower than PBX 9501 at 20°C[6], but PBXN-110 does not exhibit brittle failure or strain softening behavior up to at least 15% compressive strain, in contrast to PBX 9501 which peaks in strength after about 2-3% strain due to damage processes[6].

The compressive flow strength of PBXN-110 is strongly dependent on strain rate at all the test temperatures accessed in this study. Additional testing as a function of strain rate is planned at lower temperatures to evaluate the effect of the glass-transition temperature, T_g, on the strength and brittleness of PBXN-110.

The compressive flow strength of PBXN-110 is strongly dependent on temperature only below 0°C at high strain rates. At temperatures above 0°C at high strain rates and between -15°C and +40°C (well above the glass-transition temperature) at quasi-static strain rates, the compressive flow strength is only mildly dependent on temperature.

The compressive flow stress of the HTPB binder analog is very low at all temperatures tested and is not strongly temperature dependent until the temperature is reduced below -20°C at high strain rates. Again, additional testing is planned to evaluate the compressive strength behavior around the glass-transition temperature.

Comparing the compressive strength of PBXN-110 to the HTPB binder at room temperature, 15% strain, and a strain rate of 2500 s^{-1} reveals a factor of 10 difference in strength. This large strength differential is maintained down to at least -40°C at high strain rate. Quasi-static tests show similar flow stress differentials.

A significant amount of scatter was observed while testing *precision-cored* PBXN-110 at high strain rates. Since this scatter was not observed during quasi-static testing of much larger specimens the source is presumed to be due to inhomogeneities in the microstructure associated with the very coarse HMX crystals in the PBXN-110 compared to the small size of the SHPB test specimen dimensions. Further testing of PBXN-110 is planned with larger specimens to verify this hypothesis.

These measurements provide the basis to develop and validate predictive material strength models for PBXN-110 and its HTPB binder.

ACKNOWLEDGMENTS

Joint DOD/DOE Munitions Technology Development Program.

REFERENCES

1. Hoge, K. G., "The Behavior of Plastic-bonded Explosives under Dynamic Compressive Loads," Appl. Polym. Symp., vol. 5, 1967, pp. 19-40.

2. Peeters, R. L., "Characterization of Plastic Bonded Explosives," J. Reinf. Plast. Compos., vol. 1, 1982, pp. 131-140.

3. Field, J. E., Palmer, S. J. P., Pope, P. H., Sundararajan, R. and Swallowe, G. M., "Mechanical Properties of PBX's and their Behaviour during Drop-weight Impact," Proc. 8th Int. Detonation Symposium, White Oak, Maryland, USA, 1985, pp. 635-644.

4. Palmer, S. J. P., Field, J. E. and Huntley, J. M., "Deformation, Strengths and Strains to Failure of Polymer Bonded Explosives," Proc. R. Soc. Lond. A, vol. 440, 1993, pp. 399-419.

5. Funk, D. J., Laabs, G. W., Peterson, P. D. and Asay, B. W., "Measurement of the Stress-Strain Response of Energetic Materials as a Function of Strain Rate and Temperature: PBX 9501 and mock 9501," in Shock Compression of Condensed Matter-1995; AIP: Woodbury, New York, 1996, pp. 145-148.

6. Gray III, G. T., Blumenthal, W. R., Idar, D. J. and Cady, C. M., "Influence of Temperature on the High-Strain-Rate Mechanical Behavior of PBX 9501," in Shock Compression of Condensed Matter-1997; AIP: Woodbury, NY, 1998, pp. 583-586.

7. Idar, D. J., Peterson, P. D., Scott, P. D. and Funk, D. J., "Low Strain Rate Compression Measurements of PBXN-9, PBX 9501, AND Mock 9501," in Shock Compression of Condensed Matter-1997; AIP: Woodbury, NY, 1998, pp. 587-590.

8. Gray III, G.T., Idar, D. J., Blumenthal, W.R., Cady, C.M., and Peterson, P. D., "High- and Low-Strain Rate Compression Properties of Several Energetic Material Composites as a Function of Strain Rate and Temperature," Proc. 11th Int. Detonation Symposium, Snowmass Village, CO, USA, 1998, pp. 76-83.

9. Blumenthal, W.R., Gray III, G.T., Idar, D. J., Holmes, M.D., Scott, P.D., Cady, C.M., and Cannon, D.D., "Influence of Temperature and Strain Rate on the Mechanical Behavior of PBX 9502 and Kel-F 800^{TM}" in Shock Compression of Condensed Matter-1999; AIP: Woodbury, NY, 2000, pp. 671-674.

10. Idar, D. J., Thompson, D. G., Gray III, G. T., Blumenthal, W. R., Cady, C.D., Peterson, P. D., Jacquez, B. J., Roemer, E. L., Wright, W. J., "Influence of Polymer Molecular Weight, Temperature, and Strain Rate on the Mechanical Properties of PBX 9501" in Shock Compression of Condensed Matter-2001; AIP: Woodbury, NY, 2002, in press.

11. E.M. Mas, B.E. Clements, W.R. Blumenthal, C. Cady, G.T. Gray III and C. Liu "A Viscoelastic Model for PBX Binders" in Shock Compression of Condensed Matter-2001; AIP: Woodbury, NY, 2002, in press.

12. Gray III, G.T., and Blumenthal, W.R., "Split-Hopkinson Pressure Bar Testing of Soft Materials" in ASM Handbook Vol. 8, H. Kuhn and D. Medlin (Eds.); ASM International: Metals Park, OH; 2000, pp. 488-496.

13. Follansbee, P. S. and Frantz, C., "Wave propagation in the SHPB," Trans. ASME: J. Engng. Mater. Technol., vol. 105, 1983, pp. 61-66.