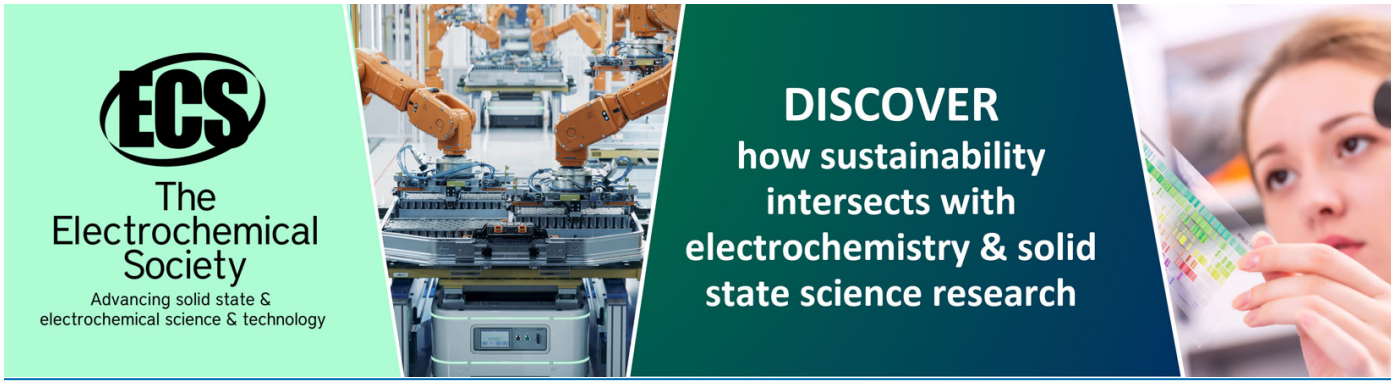
In *situ* investigation of the dynamic response of energetic materials using IMPULSE at the Advanced Photon Source



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In situ investigation of the dynamic response of energetic materials using IMPULSE at the Advanced Photon Source

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Abstract. The mechanical and chemical response of energetic materials is controlled by a convolution of deformation mechanisms that span length scales and evolve during impact. Traditional methods use continuum measurements to infer the microstructural response whereas advances in synchrotron capabilities and diagnostics are providing new, unique opportunities to interrogate materials in real time and *in situ*. Experiments have been performed on a new gas-gun system (IMPact system for Ultrafast Synchrotron Experiments) using single X-ray bunch phase contrast imaging (PCI) and Laue diffraction at the Advanced Photon Source (APS). The low absorption of molecular materials maximizes x-ray beam penetration, allowing measurements in transmission using the brilliance currently available at APS Sector 32. The transmission geometry makes it possible to observe both average lattice response and spatially heterogeneous, continuum response (1-4 um spatial resolution over ~2 x 2 mm area, 80 ps exposure, 153 ns frame-rate) in energetic materials ranging from single crystals to plastic-bonded composites. The current work describes our progress developing and using these diagnostics to observe deformation mechanisms relevant to explosives and the first experiments performed with explosives on IMPULSE at APS.

1. Introduction

Energetic materials in application are plastic-bonded composites of molecular crystals. Their mechanical and chemical response to impact is controlled by a complex convolution of bulk defects and interfaces, granular mechanics, and crystal mechanics. Chemical reactions are initiated by energy localization at hot spots but fundamental experimental insight of the mechanistic details are currently lacking. Therefore sensitivity to detonation is empirically calibrated, enabling correlation with microstructure but not fundamental understanding of the origins of initiation. Since energetic materials cannot be faithfully recovered with their microstructures intact because of mechanical and chemical fragility, the state of the material must be probed during impact with more advanced diagnostics.

Recently, the IMPact System for Ultrafast Synchrotron Experiments (IMPULSE) was developed for use at the Advanced Photon Source (APS) [1]. IMPULSE consists of a mobile, single-stage gas gun designed for fielding phase contrast imaging (PCI) and Laue X-ray Diffraction (XRD) during impact [2, 3]. The low absorption of molecular materials maximizes x-ray beam penetration, allowing measurements in transmission using the brilliance currently available at APS Sector 32. The transmission geometry enables exciting possibilities for observing both average lattice response and

spatially heterogeneous, continuum response (1-4 um spatial resolution over ~2 x 2 mm area, 80 ps exposure, 153 ns frame-rate) in energetic materials ranging from single crystals to plastic-bonded composites. This capability provides a means for linking mechanics to detonation initiation by resolving deformation mechanisms such as dislocation-mediated plasticity, phase transformation, void collapse and jetting, cracking, and compaction. Here we report our progress developing and using these diagnostics to investigate the dynamic response of energetic materials *in situ* during impact. The first set of experiments demonstrates capabilities to observe relevant deformation mechanisms in inert materials and the second are the first experiments recently performed with explosives on IMPULSE at APS.

2. X-ray diffraction and imaging at APS

In preparation for investigations of energetic materials, static measurements and dynamic impact experiments were performed to demonstrate that relevant mechanisms could be observed. Target designs were tested for Laue XRD in transmission. PCI was fielded in a series of experiments to observe void collapse and jetting, cracking, density gradients across release waves, and compaction. The setup for XRD and PCI are illustrated in figure 1 and have been described in detail elsewhere [2-5].

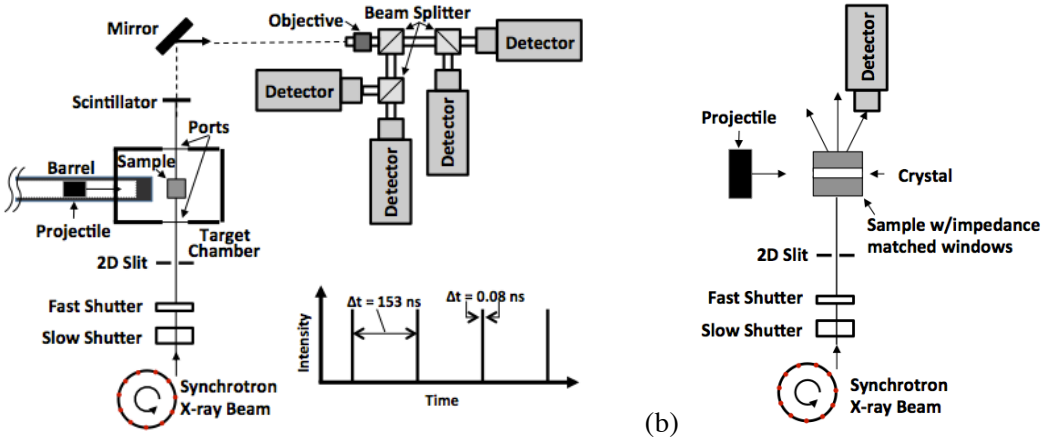


Figure 1. Experimental configuration for gas-gun driven impact experiments using (a) four-frame PCI and (b) transmission XRD with impedance matched windows at APS Sector 32. In standard mode of operation, the synchrotron provides 80 ps X-ray pulses every 153 ns.

2.1. X-ray diffraction

XRD studies in literature have set the precedent for *in situ* studies of plasticity [6-9] and phase transformations [10] but the methods are not sufficient for low symmetry molecular crystals. Monochromatic XRD limits the number of spots observed in a single experiment. Low symmetry crystals require measurement of more components of strain and phase information. Therefore, Laue XRD, with the white beam, in transmission geometry is being pursued [5, 11]. This complicates target design but allows for simultaneous measurement of XRD and PCI because of the low x-ray absorption through mm-to-cm sized samples. Initial studies have been performed to develop acceptable target designs. Figure 1(b) illustrates the target and detector configuration that has been used with oriented acetaminophen single crystals as an inert surrogate for explosive crystals. In this configuration, sample thickness was minimized to limit the broadening of X-ray peaks, and vitreous carbon was used as X-ray transparent windows for minimizing release wave effects during impact experiments. With 1-2 mm thick acetaminophen and windows, 30:1 to 15:1 signal-to-noise ratios were achieved in the static measurements shown in figure 2, and impact experiments are underway.

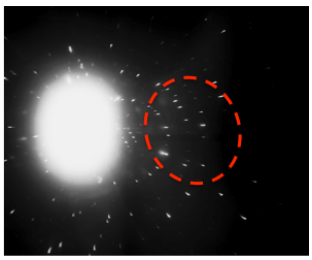
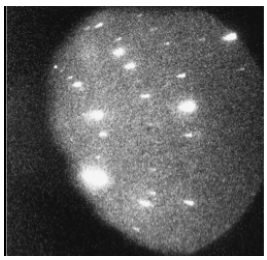
(a)  (b) 

Figure 2. XRD from (010)-oriented acetaminophen recorded on (a) 8 x 10 inch film and (b) on a Princeton PI-MAX I ICCD camera with a 75:40 mm diameter fiber taper and LSO:Ce scintillator [3]. The sample was approximately 3 inches from the film/detector, and the undulator gap was set at 20 mm. The dashed line in (a) indicates the area recorded on the ICCD.

2.2. Phase contrast imaging

PCI is much more effective for imaging low density, low atomic number materials than traditional radiography. It relies on the spatial variation in the phase of coherent or semi-coherent X-rays and their interference, rather than on their absorption [12]. It is more sensitive to interfaces through interference effects and can be used with intense polychromatic synchrotron beams that make it possible to image high-rate deformation on picosecond time scales [13, 14]. PCI had been previously applied in shock wave studies, but involved imaging of strong laser-driven shocks [15]. Our intent in this work was to image individual deformation mechanisms at lower pressures in homogeneous materials as well as heterogeneous microstructures and to demonstrate sufficient temporal, spatial, and density resolution.

2.2.1. Void collapse and jetting

Figure 3 illustrates void collapse and jetting from an oriented single crystal of acetaminophen. This experiment consisted of (010)-oriented crystal slab with five 150 μm diameter half cylinder grooves machined in the rear surface at 450 μm spacings. The crystal was impacted with copper at 468 m/s and 2 GPa, collapsing the void and producing jetting. Void collapse and jetting is a mechanism for hot spot initiation [16] but had not previously been observed in brittle molecular crystals [17, 18]. Therefore, acetaminophen was used as an inert surrogate to experimentally investigate the mechanics of void collapse because it has similar mechanical and phase behavior as HMX [19]. This experiment was representative of the front half of a hot spot void or an interface in explosives. Future PCI experiments will enable similar observations for energetic crystals with arrays of internal voids.

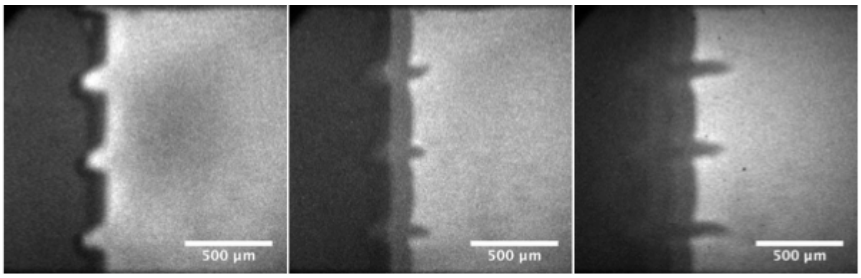


Figure 3. PCI images of void collapse and jetting from an (010)-oriented acetaminophen crystal slab after a 2 GPa planar impact. The inter-frame time was 153 ns.

2.2.2. Cylinder impact and cracking

Figure 4 illustrates a 300 μm diameter by 1 mm long copper cylinder striking a vitreous carbon plate at 809 m/s and 4.53 GPa. Significant deformation of both the cylinder and plate was evident. Cracks nucleated after impact and propagated to the rear surface where they coalesced in the spall area that developed following release of the shock wave. The ability to resolve physical interfaces of cracks,

with 3-4 μm resolution [4], will be useful for the investigation of both single crystal and composite explosives. For example, it has been speculated that cracking occurs for crystal orientations of both PETN [20] and RDX [21] in which dislocation-mediated plasticity is inaccessible by known mechanisms. In these cases, PCI experiments can be used to resolve both interpretation of velocimetry data [21] and which mechanisms should be included in constitutive models [20].

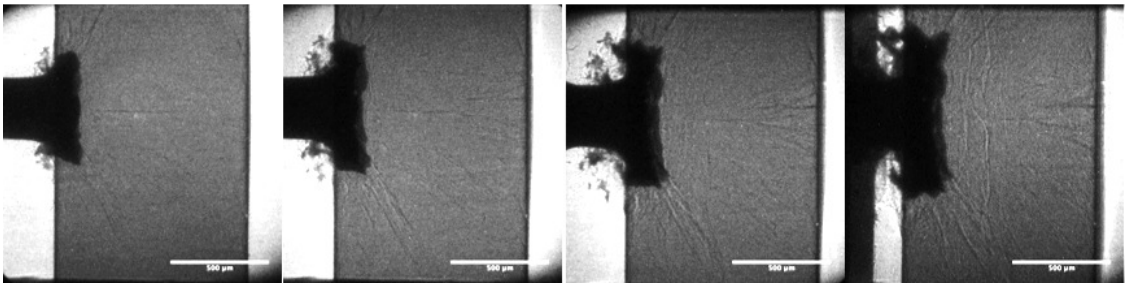
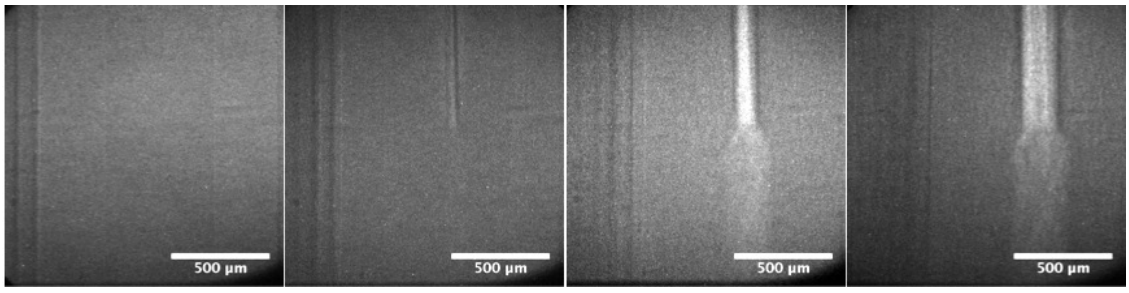
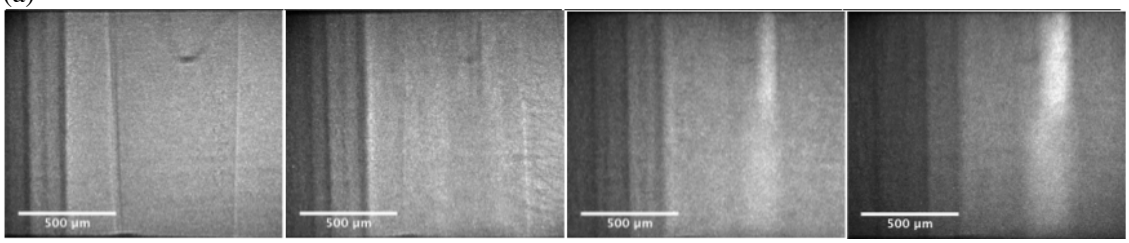


Figure 4. PCI images of a 300 μm diameter by 1 mm long copper cylinder striking a vitreous carbon plate at 809 m/s and 4.53 GPa [4]. The inter-frame time was 153 ns.



(a)



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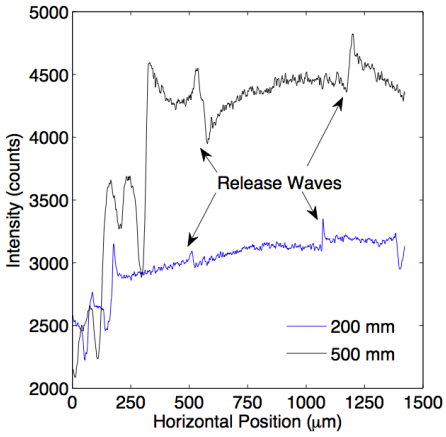


(c)

Figure 5. Cracks in vitreous carbon were put in plane strain by converging release waves following symmetric impact at 630 m/s and 2.2 GPa. The inter-frame times were 153 ns. The sample-to-scintillator distances were set at (a) 200 and (b) 500 mm to enhance spatial resolution and contrast respectively. The horizontal lineouts (c) of the first PCI image from each experiment illustrate the phase contrast effects originating from density gradients across release waves.

2.2.3. Plane strain cracking

Figure 5 illustrates the PCI results from two different symmetric impact experiments for vitreous carbon samples containing centered precracks. A symmetric impact near 630 m/s and 2.2 GPa were released off of the target's free surface and the foam backed impactor. Converging release waves put the crack in tension, causing plane strain crack propagation that appeared to be blunted by diffuse material failure on the spall plane ahead of the crack. The 7% difference in density across the release wave interfaces was directly observable with PCI. The sample-to-scintillator distances were 200 and 500 mm for (a) and (b) respectively. Increased distance enhanced the contrast, making the density gradient more visible at the release waves, but allowed the refracted X-rays to diverge, broadening the appearance of features, and degraded spatial resolution. Vertically binned, horizontal lineouts are compared in (c) and exhibit the expected phase contrast effects at the density interfaces. Additional experiments have demonstrated that release waves with as little as 2% difference in density, from the ambient 1.5 g/cc for vitreous carbon, can be observed. This will be significant for future explosive investigations. For example, the α-γ phase transition in RDX under shock loading is associated with a larger change in density [22] and the transformation front should be directly observable with PCI. If the phase transition is associated with spatial heterogeneity, such as heterogeneous nucleation of the γ phase or cracking between phases, on the 1-4 μm scale it will also be observable. This offers a new capability to study the microstructure of shock-driven phase transitions.

2.2.4. Heterogeneous microstructures and compaction of granular materials

Figure 6 illustrates an idealized representation of a granular material and the heterogeneous microstructure of plastic-bonded sugar (PBS). The two images demonstrated the difficulty of imaging the microstructure and compaction of granular composites. Discernable microstructure features in the contrast were obscured as the number and density of interfaces transited by the collimated X-rays changed, as a function of either deformation and compaction or thickness. Multiple refraction of X-rays throughout the samples caused discernable features to be replaced with texture. Although individual crack and deformation events were lost, the texture can be used as an average measure of the microstructure response. In future efforts, spatial variation in texture will be quantified and used to track compaction. Determining the optimal experimental configurations and methods of analysis (both ray tracing in PCI simulations and digital image analysis to quantify texture) for this type of data will be important for studies of shock initiation in energetic composites.

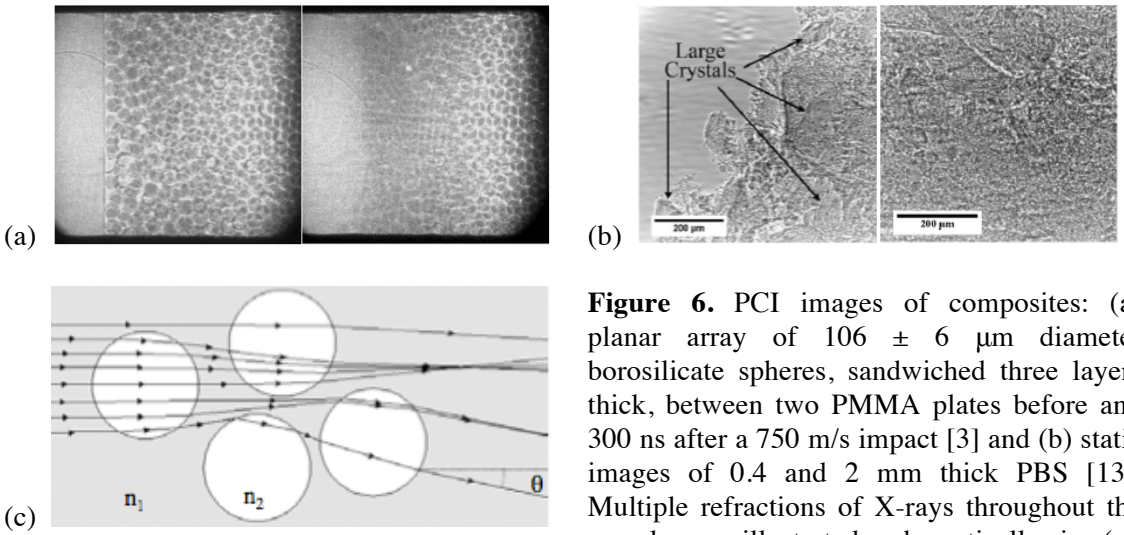
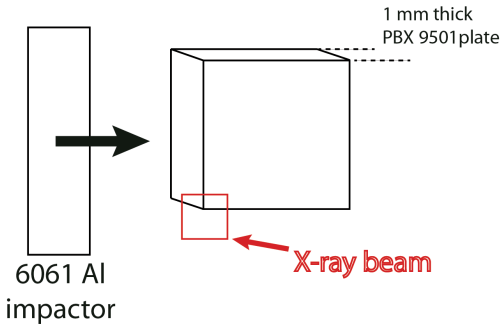


Figure 6. PCI images of composites: (a) planar array of 106 ± 6 μm diameter borosilicate spheres, sandwiched three layers thick, between two PMMA plates before and 300 ns after a 750 m/s impact [3] and (b) static images of 0.4 and 2 mm thick PBS [13]. Multiple refractions of X-rays throughout the sample, as illustrated schematically in (c), obscured microstructure features and caused texture in the images' contrast.

3. First IMPULSE experiments with explosives at APS

Prior to conducting experiments with explosives at APS, a structural analysis was performed for the entire gun system. Gas flow, expansion, and pressure time histories from a CTH hydrodynamic model were used as inputs to a structural dynamics model in the ABAQUS FEA code to predict the system's response to explosive detonation upon impact. A conservative detonation limit was set based on the yield stress of the components' materials and IMPULSE was approved for experiments containing 750 mg of TNT equivalent explosive.

The first experiments involving explosives were conducted with IMPULSE at APS in March 2013. They were performed as part of a PBX 9501 aging study intended to explore connections between environmental factors and mechanical response on the microstructure scale directly with *in situ* imaging. Prior studies have shown that storage of PBX 9501 in humid environments leads to hydrolysis of ester linkages in the Estane and reduces its molecular weight [23]. Presumably, there are commensurate effects on binder adhesion and elongation in the microstructure during impact. To observe possible effects of aging, PBX 9501 was exposed to 70 °C and 74% relative humidity for 18 days, reducing the molecular weight from 120 kDaltons to 50 kDaltons, in a manner similar to reference [24]. Thin rectangular prisms were prepared and impacted with aluminum projectiles as shown in figure 7 (a).



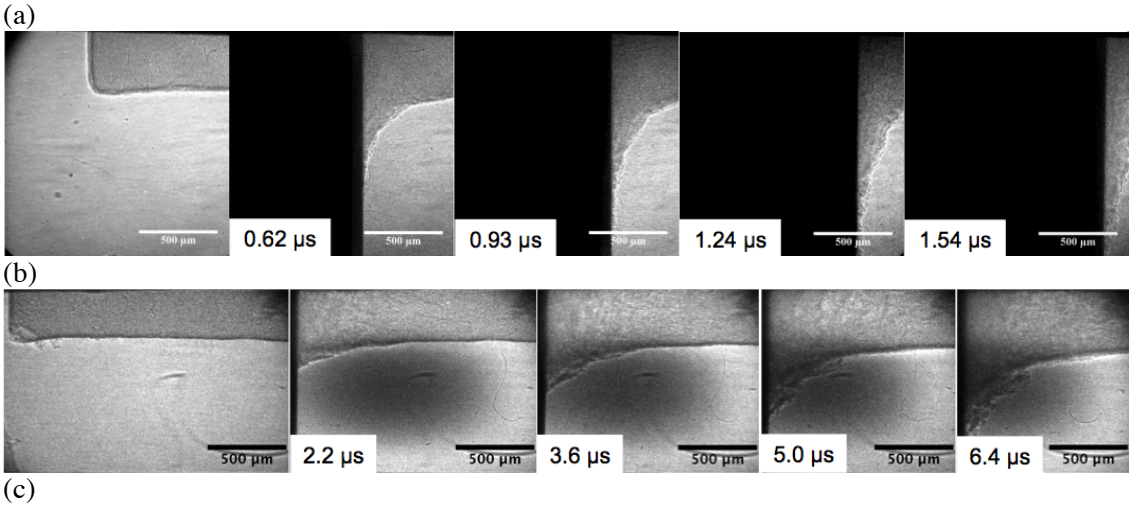


Figure 7. PCI images of planar impact of aged PBX 9501. The experimental configuration is illustrated in (a). Impact velocities and sample-to-scintillator distances were (b) 477 m/s and 200 mm and (c) 110 m/s and 500 mm.

The experimental configuration was chosen to observe axial compaction and radial flow at the edge of the PBX 9501 while also limiting the number of granular interfaces (1 mm / 200 µm grains ~ 6 interfaces) through the 1 mm thickness. Experiments were performed for 477 and 110 m/s impacts. Deformation of the prisms' edges exhibit deviations from smooth contours, in figures 7 (b-c), suggesting microstructure effects. Although individual HMX grains could not be discerned, there was corroborating spatial heterogeneity in the contrast. For the higher velocity experiment, PCI was optimized for resolution with the sample-to-scintillator distance set at 200 mm; whereas, the lower

velocity experiment was set at 500 mm for optimized contrast. At 200 mm, physical interfaces were observable but difficult to interpret because they overlapped and caused multiple refractions of the X-rays as they transited the composite. Alternatively at 500 mm, spatial variations in contrast texture, as a function of distance from the impact surface, were observed and indicated compaction as previously observed in the borosilicate sphere experiment in figure 6a. Quantitative texture analysis will be needed to track the compaction fronts. Additional diagnostics (e.g. spatially resolved velocimetry [25] or small angle X-ray scattering) will also be necessary to quantify the microstructure and mechanical state of the material behind the compaction fronts. These issues will be investigated further to determine the optimal experimental conditions for imaging and quantifying microstructure damage directly in complex composite explosives. Currently, deformation contours are being used for model validation.

4. Summary and future work

We have developed and begun utilizing PCI and Laue XRD with IMPULSE to investigate the dynamic response of energetic materials *in situ* during impact. Our initial studies of inert materials were performed to develop acceptable target designs for XRD in transmission and to demonstrate that deformation mechanisms relevant to explosives could be observed directly with PCI. The transmission XRD used impedance matched windows to limit diffraction peak broadening from sample thickness and minimize release wave effects. Sufficient signal-to-noise ratios were achieved, and impact experiments are underway to evaluate and further develop target designs. PCI experiments demonstrated that the temporal, spatial, and density resolution was sufficient for observing void collapse and jetting, cracking, density gradients with 2% difference across release waves, and compaction. The IMPULSE gun system was analyzed and approved for detonation of up to 750 mg of explosives, and the first experiments were performed in March 2013 to investigate the effects of aging on the impact response of PBX 9501. Imaging the heterogeneous microstructures was more challenging because multiple refractions of X-rays through the sample obscured individual deformation mechanisms. However, contrast texture was enhanced with sample-to-scintillator distance adjustments and used to reveal compaction fronts. Further improvements to enhance contrast are being made and digital image analysis methods and PCI ray tracing simulations are being developed. With continued development, *in situ* X-ray imaging and diffraction will provide new insights to deformation mechanisms that control the mechanical and chemical response of explosives.

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