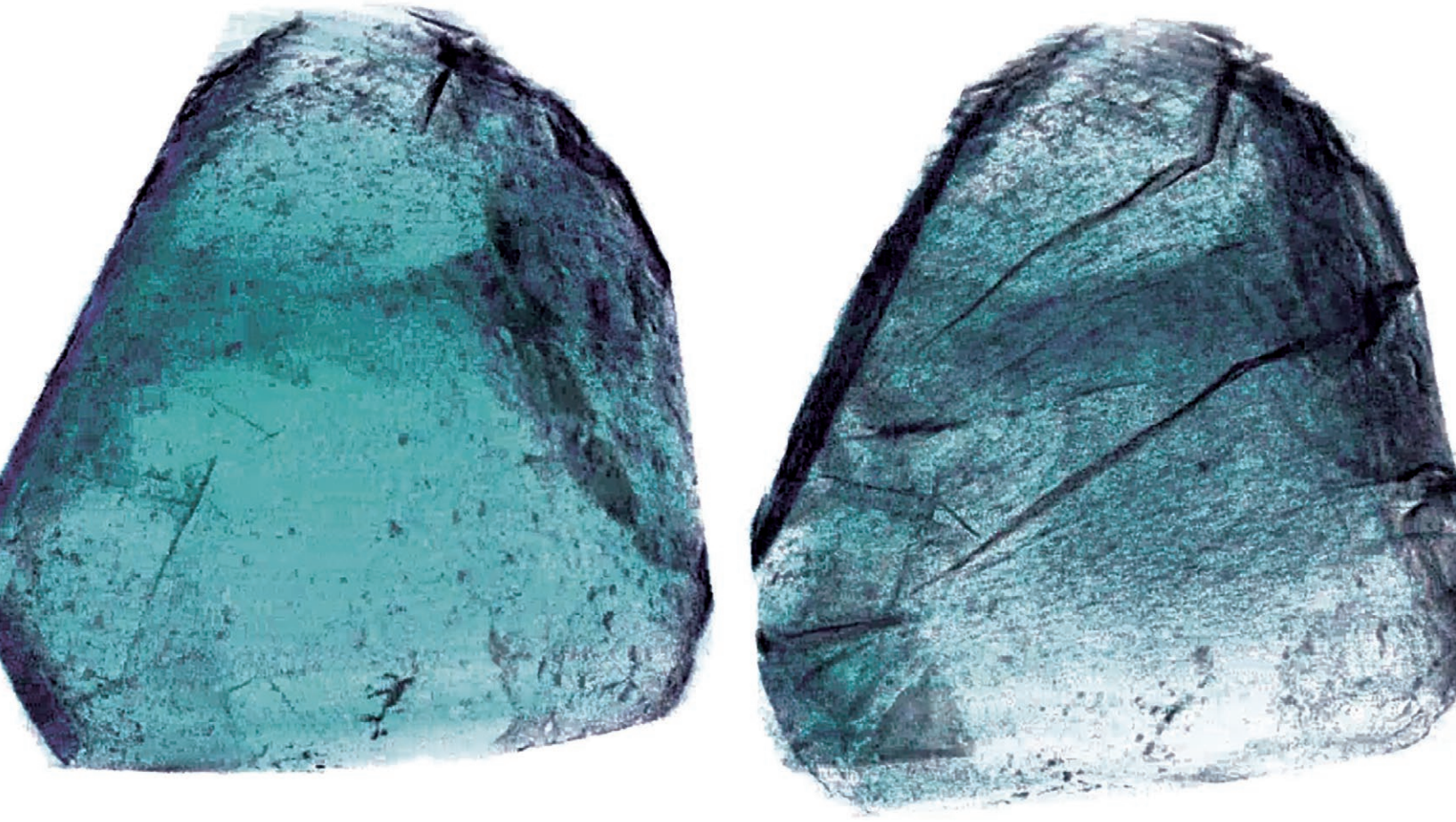

In situ Uniaxial Compression

of Single Crystals of HMX explosive during 3D XRM Imaging

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In situ Uniaxial Compression
of Single Crystals of HMX explosive during 3D XRM Imaging

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The mechanical deformation of a single crystal of high explosive HMX (octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine) using a nanoscale 3D X-ray microscope (XRM) and a purpose-built *in situ* load stage is demonstrated. This technique provides invaluable insight into how these crystals fracture and the formation of dislocations, which together affect the morphology of this material. Material damage, voids, and cracks within high explosives affect shock sensitivity and their explosive response. Therefore they may not perform as intended and are inherently less safe.

The uniaxial compression or tension of materials, typically on the millimeter or bulk scale, is critical to understanding plastic flow and fracture. This technique is often coupled to a visible camera and can measure stress and the resultant strain. Recent advances have pushed this technique to *in situ* experiments, in which other characterization techniques are used to probe the material morphology. However, to gain a complete picture of material damage, characterization on multiple length scales is needed. There is a strong requirement for further development of higher resolution *in situ* instrumentation. These techniques include electron microscopy (scanning electron microscopy, transmission electron microscopy) of quartz[1] and X-ray diffraction of aspirin and acetaminophen[2]. A review of micrometer-scale single crystals in compression is provided by Uchic et al.[3]. Here we demonstrate *in situ* compression experiments in a nanoscale X-ray microscope (XRM).

HMX is a high explosive (HE) crystal that is used in many types of plastic bonded explosives (PBX) used by the military. These crystals are typically mixed into the plastic mixture and hot molded into shape. Understanding their properties, from the bulk scale down to the single crystal nanoscale, is critical to predicting initiation and detonation, as well as assuring that the HE will function as intended. These attributes all determine performance and safety.

Experimental

The load stage was configured for uniaxial compression by screwing the sample mounting pin into the piezo actuator, which is coupled with a 9 N force sensor, and mounting the opposing diamond compression anvil (Figure 1). To facilitate holding the samples, the end of the sample mounting pin was coated with a small amount of silicone vacuum grease by using a small probe. This grease was then wiped off, leaving a very minor amount of tackiness on the surface. Single crystals of HMX, approximately 50-100 µm in diameter, were distributed into a small petri dish. One crystal was carefully chosen and mounted onto the tip of the sample mounting pin. The upper diamond anvil was then coarse-aligned to within 50 µm of the HMX crystal using a visible light camera. The position of the upper diamond anvil was further refined after the load stage was placed within the UltraXRM-L200 X-ray Microscope (now known as the ZEISS Xradia Ultra XRM). This instrument uses an 8.04 keV rotating anode copper source and a transmission X-ray microscope (TXM) architecture to achieve resolution down to 50 nm. The sample and opposing diamond were both finely aligned using X-rays.

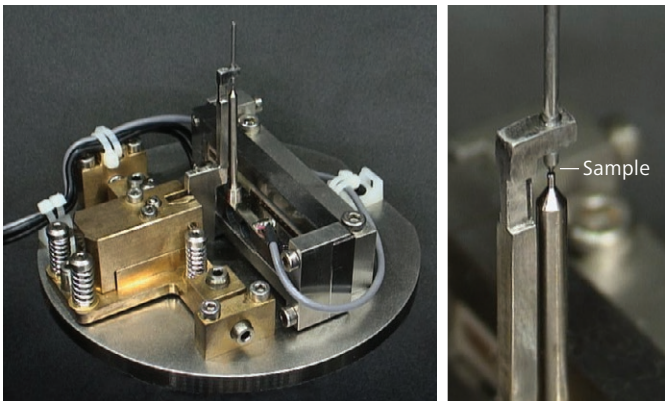

Figure 1: Photo of the load stage (left) and a close up of the top and bottom anvils in uniaxial compression configuration.

A series of 3x2 mosaic radiographs were collected in Zernike phase contrast mode (65 µm field of view per mosaic tile), as the load cell was stepped in 2 µm increments (Figure 2), pressing the crystal against the diamond anvil. The mosaic was pixel binned by 2 to produce a 1536 x 1024 pixel image with 130 nm isotropic pixels, a total field of view of 195 x 130 µm. 30 s of dwell time for each tile required 3 min for each frame.

A second crystal was mounted for tomographic imaging. The imaging conditions included a rotation of –77.25° to 75°, a 60 s exposure and 1441 radiographs leading to a 24 h total tomographic imaging time. The arm that holds the opposing diamond anvil blocked X-ray transmission and limited the tomographic rotation. The camera was not binned, therefore the dimensions of each isotropic voxel is 65 nm. The crystal was imaged uncompressed and after a compression of 5 µm.

Results

A sequence of radiographic images in Figure 2 show the uniaxial compression of a single ~140 µm diameter crystal. The images are shown every fourth µm of displacement of the load stage. Visible at the top of each frame is the diamond anvil, 100 µm in diameter. The bottom post becomes visible in the fourth image. A few gold particles, ~3 µm in diameter (useful for aligning the images when collecting tomographic data), are also visible. The simultaneously-collected load response curve is shown in Figure 3. The load climbs initially until it reaches a peak stress at ~140 mN, at which point the crystal has shattered catastrophically. The right side portion of the crystal, not under the anvil, has separated from the rest of the crystal and is rotating counter clockwise.

A second data set was collected using a fresh crystal using the Ultra XRM in tomographic, Zernike phase contrast mode. This crystal has a hexagonal cross section in one plane, and a parallelogram cross section in the other. Figure 4 shows two reconstructed slices through the crystal. In these slices, the crystal does not appear to be making contact with the pin, due to its shape. However, it is making contact in other locations. The image on the left is of the undisturbed crystal; the image on the right was recorded after compressing it a total of 5 µm. Stress cracks are visible on the left and the right of the image of the compressed crystal. Dislocations are visible as small dark spots between them. Other slices within this crystal show the formation of cracks, which also have a hexagonal shape.

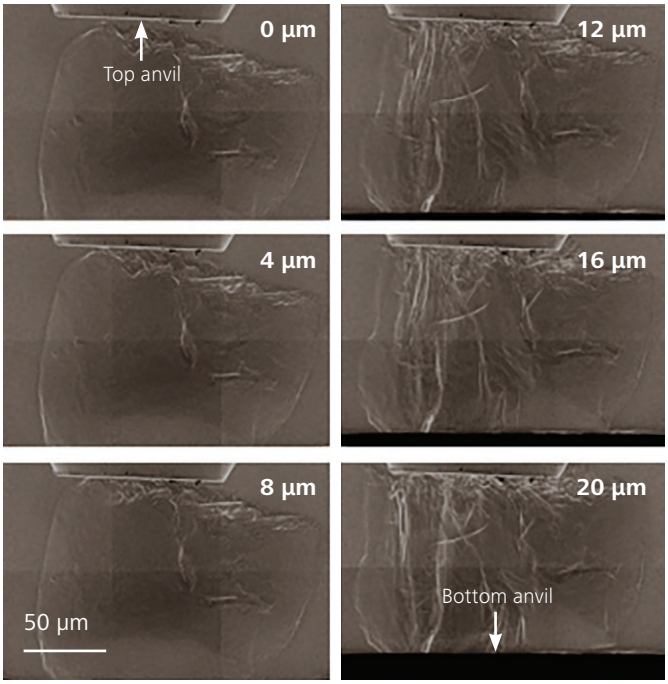

Figure 2: Sequential radiographs as a single crystal of HMX is compressed., shown in 4 µm increments. The top anvil is stationary, while the bottom anvil with the sample mounted on it is moved upwards.

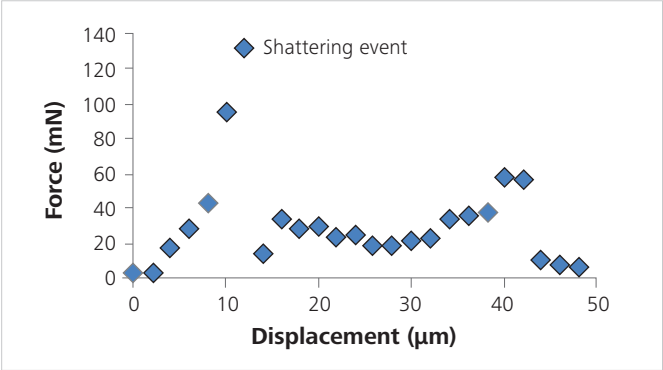

Figure 3: Force displacement plot for the uniaxial compression of a single crystal of HMX shown in Figure 2. The fourth image corresponds to the peak force at 140 mN.

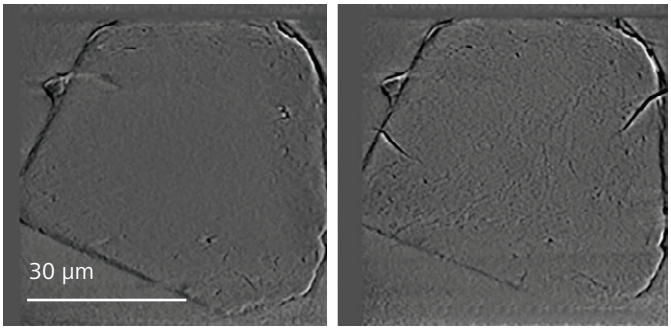

Figure 4: Reconstructed slices through a single crystal of HMX uncompressed (left) and after it has been compressed 5 µm (right). Stress cracks and dislocations are visible in the compressed crystal.

Figure 5 shows 3D volume renderings of the crystal in uncompressed and compressed state.

Conclusions

Using nano-scale X-ray microscopy, it is possible to image, *in situ*, the compression fracture of single crystal materials. Xradia Ultra X-ray microscope in combination with a mechanical load stage reveals:

- The onset of damage,
- The local strain, and
- Insight into damage progression

This was all accomplished in 3D. Plastic bonded explosives are often greater by mass greater than 90% crystal, therefore, their bulk mechanical response is strongly affected by the single crystal mechanical performance. The information collected here will help to better understand the mechanisms of damage and hotspot formation. Further experiments will relate this mechanical response to *in situ* diffraction measurements, which can better detect crystallographic deformations. Together, X-ray diffraction and tomographic imaging will help in further understanding damage and shock sensitivity in high explosives.

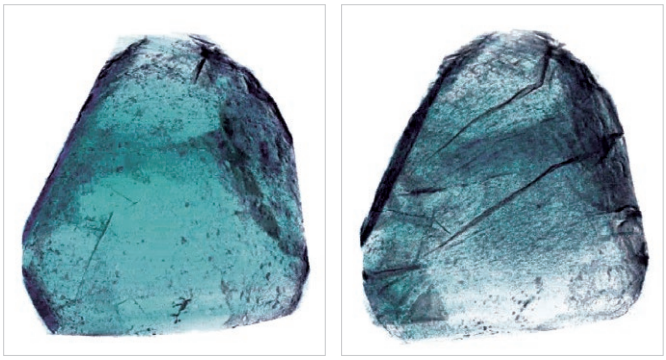

Figure 5: Volume renderings of the same crystal shown above (and on cover) in uncompressed state (left), and after compression (right) by 5 µm. Stress cracks are visible in the compressed crystal.

References:

[1] E Tochigi, et al., "*In situ* TEM observations of plastic deformation in quartz crystals," *Phys Chem Minerals* 41 (2014):757-765. DOI: 10.1007/s00269-014-0689-6

[2] RV Haware, et al., "Anisotropic crystal deformation measurements determined using powder X-ray diffraction and a new *in situ* compression stage," *Int J Pharma* 418 (2011):199-206. DOI: 10.1016/j.ijpharm.2011.06.021

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