In situ observation of phase transition behavior of shocked baddeleyite

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1. Introduction
Baddeleyite (ZrO_2), commonly found in mafic igneous rock, can be applied as a shock pressure barometer and a geochronometer in meteorites. It can offer essential information on the timing and intensity of a meteorites impact, which help us to understand the planetary formation and evolution. Previous researches on natural shocked-baddeleyite in the earth's rock and meteorites have observed grain refinement and the orthogonally related crystallographic orientations of the baddeleyite grains, which may infer the occurrence of phase transition. Although the phase transition behavior of baddeleyite at high pressures has been intensively investigated under hydrostatic conditions, there is little information on dynamic response of the phase transition under shock loading conditions. Therefore, it is required to clarify the phase transition behavior of baddeleyite under shock compression in order to utilized as a shock barometer.
Here, we performed *in situ* X-ray diffraction measurements on shock-loaded baddeleyite on nanosecond time scale using shock-driving laser and synchrotron X-ray.

2. Methods
We performed the experiments at NW14A beamline at PF-AR (Photon Factory Advanced Ring), High Energy Accelerator Research Organization (KEK), Japan. Baddeleyite samples were in form of square-shaped plates with dimensions 5 ×5 and a thickness of 0.06 mm. A 25 μm PET film ablator with Al coating was attached on the sample surface. The crystal structure of the sample was determined by XRD to be monoclinic (space group $P2_1/c$) with lattice parameter of a = 5.156(2) Å, b = 5.199(2) Å, c = 5.310(2) Å, β = 99.23(3)°, and V = 140.54(6) Å^3. Nd:glass laser with the wavelength of 1064 nm, pulse energy of 16 J, pulse width of 12 ns at the beamline was used for shock-driving source. The laser beam was focused by optical lens to a diameter of 500 μm on the sample surface. The velocity data was obtained by VISAR (velocity interferometer system for any reflector) measurement. For TR-XRD experiments, the synchrotron X-ray pulse of PF-AR was used as a probe source. The energy, the pulse width, and the size on the sample surface were 15.57 keV, about 100 ps, and 450 μm (H) ×250 μm (W), respectively. The XRD data at the timing within 40 ns after shock compression and that beyond 1000 ns were collected. For each XRD data, lattice parameters were refined by Rietveld method.

3. Results
The free surface velocity reached 0.914 km/s. From the velocity, the peak shock pressure determined by the U_s–u_p Hugoniots (U_s = 4.38 + 1.37 u_p km/s) (Mashimo et al., 1983) was 13.4 GPa.
The phase transition from monoclinic phase to orthorhombic-I ZrO_2 phase (the first high-pressure phase) was observed at 6.5 ns (compression state). The phase transition boundary under shock compression was determined to be 3.3 GPa, which is slightly lower than that observed in the previous static compression experiments. The compression of orthorhombic-I ZrO_2 reached to V/V_0 = 0.956, corresponding to the pressure of 14.0 GPa. Although the peak pressure in the study was beyond the phase transition boundary between orthorhombic-I and orthorhombic-II ZrO_2 (the second high-pressure phase) in phase diagram, orthorhombic-II ZrO_2 did not appear. During release state, orthorhombic-I ZrO_2 returned to monoclinic

phase, and finally orthorhombic-I ZrO_2 disappeared completely. Our direct observation revealed that the displacive-type phase transition between monoclinic and orthorhombic-I ZrO_2 can occur regardless of strain rate, but the reconstructive-type phase transition between orthorhombic-I and orthorhombic-II ZrO_2 depends on the time scale of compression.

Keywords: baddeleyite, laser shock, synchrotron time-resolved XRD

