High Pressure Science

NE1/2009G616

High-density amorphous polymorphs of zirconium tungstate

Akhilesh K. ARORA*[1], Tomoko SATO[2] and Takehiko YAGI[2]
[1]Materials Science Division, Indira Gandhi Centre for Atomic Research, Kalpakkam, 603102, India
[2]Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba 277-8581, Japan

Introduction

Negative thermal expansion materials such as zirconium tungstate have attracted considerable attention as these are useful in obtaining zero or tailored coefficient of thermal expansion. $Zr(WO_4)_2$ turns amorphous at high pressure and the amorphization is irreversible [1]. The *a*-$Zr(WO_4)_2$ recovered from high pressure is substantially denser [2] than the cubic $Zr(WO_4)_2$ and we have analyzed its structure recently [3]. In the present work we have carried out in-situ x-ray diffraction on *a*-$Zr(WO_4)_2$ at high pressure using synchrotron radiation at Photon Factory to explore the possibility of finding amorphous polymorphs similar to those reported in ice, Si and quartz [4-6].

Results and discussion

$Zr(WO_4)_2$ powder sample was loaded in a diamond-anvil cell (DAC) along with a small gold foil. The x-ray diffraction measurements were made using synchrotron radiation (λ = 0.4131(2) Å) from the beam-line NE1 at Photon Factory, KEK. The pressure in the DAC was estimated from the equation of state of gold. An image plate was used as the detector. The 2-D image plate data was integrated to convert it to 1-D intensity versus 2θ data.

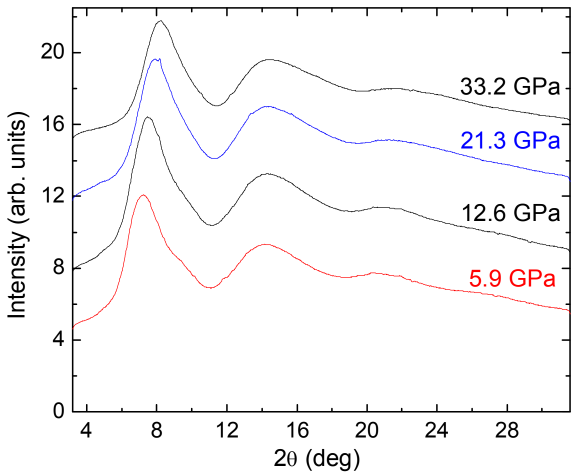

Fig. 1. Diffraction patterns of amorphous $Zr(WO_4)_2$ at different pressures.

Figure 1 shows the diffraction patterns as a function of scattering angle 2θ at different pressures. Four broad amorphous diffraction peaks (ADP) were found up to a scattering angle of 32°. One can see that the first ADP in the patterns at lower pressure has a clear shoulder between 8 and 10° 2θ. On the other hand, in the diffraction patterns at higher pressures a rapid decrease of intensity between the first peak and the first minimum is found. This suggests a qualitative difference between the patterns at low and high pressures. One can also see a systematic shift of the first ADP to higher 2θ.

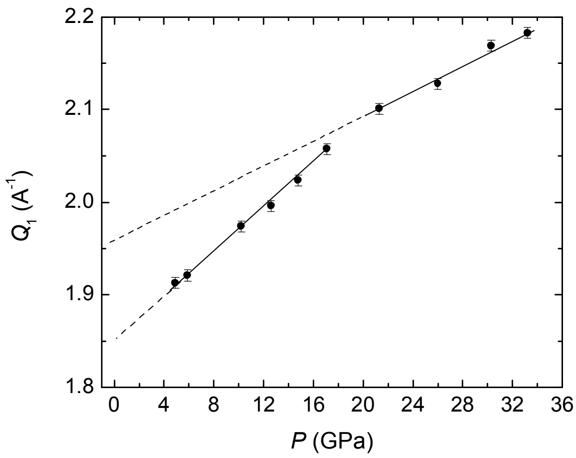

Fig. 2. Scattering vector corresponding to first ADP as a function of pressure.

The scattering vector Q_1 corresponding to position of the first ADP was calculated and is shown as a function of pressure in Fig. 2. The data shows a clear change of slope around 19 GPa. In analogy with the shift of diffraction peaks in crystalline solids at high pressure due to compression, within a given amorphous phase the position of first ADP is expected to shift monotonically to higher 2θ (larger Q_1). A change of slope can arise if a transition from one amorphous phase (say A1) to another (say A2) takes place. Smaller slope of Q_1 versus P above 19 GPa suggests that the high density amorphous phase A2 is less compressible. Thus these results suggest the possibility of existence of amorphous polymorphs in this system.

References

[1] T.R. Ravindran et al., Phys. Rev. Lett. **84**, 3879 (2000).
[2] A.K. Arora et al., J. Phys.: Condens. Matter **16**, 1025 (2004).
[3] A. K. Arora, T. Okada and T. Yagi, Phys. Rev. B **81**, 134103 (2010).
[4] O. Mishima et al., Nature **314**, 76 (1985).
[5] D. Daisenberger et al. Phys. Rev. B **75**, 224118 (2007).
[6] G.D. Mukherjee et al., Phys. Rev. Lett. **87**, 195501 (2001).

* aka@igcar.gov.in