High-Energy Synchrotron X-Ray Diffraction Study of High-Temperature Levitated Liquids at SPring-8

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Containerless techniques allow us to study the structure of liquids at high-temperature, because they can avoid two distinct problems: (i) container interactions and contamination, and (ii) the effects of the container walls on the structural measurements. Recently a combination of containerless methods and synchrotron x-ray diffraction technique has been an essential tool to study the structure of liquids at high-temperature or supercooled liquids [1]. However most of diffraction experiments have been carried out using a reflection geometry with low energy x-rays, which makes it difficult to obtain reliable data. High-energy x-rays from synchrotron radiation source provide several advantages; high resolution in real space due to the wide range of scattering vector Q (= $(4\pi/\lambda)\sin\theta$, 2θ: scattering angle, λ: wavelength of photons), smaller correction terms (especially for absorption correction), reduction of truncation errors. Furthermore a combination of high-energy x-ray diffraction and neutron diffraction is one of the powerful tools to study short- and intermediate-range structure of disordered materials [2-4]. In this study, we tried to carry out diffraction experiment using a combination of conical nozzle levitation and high-energy synchrotron x-rays to obtain reliable diffraction data of amorphous silica at high-temperature.

The conical nozzle levitation system, integrated with a two-axis diffractometer designed for disordered materials [5] at SPring-8 high-energy synchrotron x-ray diffraction beamline BL04B2 [6] is schematically illustrated in Fig. 1. The specimen is levitated by Ar gas, and heated by CO_2 laser. The scattered x-rays are detected by a Ge solid state detector with a horizontal scattering plane. The high-energy x-ray diffraction experiments were carried out at 113.26 keV photon energy. The collected data were provided absorption, background, absorption, and Compton scattering correction and then normalized to Faber-Ziman total structure factor $S(Q)$.

Fourier transformations of the $S(Q)$ data lead to the real-space, total correlation functions, $T(r)$, of amorphous silica (T_g=1180 ˚C) at 26 ˚C and 1330 ˚C are shown in Fig. 2. The first significant change in the correlation function involves the Si-O correlation at 1.61 Å, which moves to higher r and decreases intensity with increasing temperature, indicating an increase in the Si-O thermal-vibration. The O-O correlation at 2.62 Å also decreases in intensity with increasing temperature. The position of the O-O correlation in the neutron data is almost the same with increasing temperature [7]. The position of the Si-Si correlation at 1330˚C shifts slightly to higher r and decreases intensity. However, it is remarkable that the decrease of the Si-Si correlation is more moderate than for the Si-O and O-O correlations, indicating that the 6-fold rings produced by SiO_4 tetrahedra are stable even at high-temperature.

References

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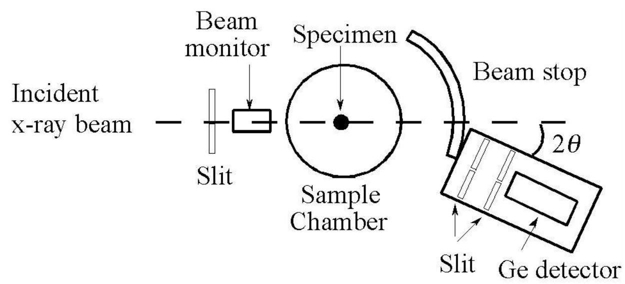
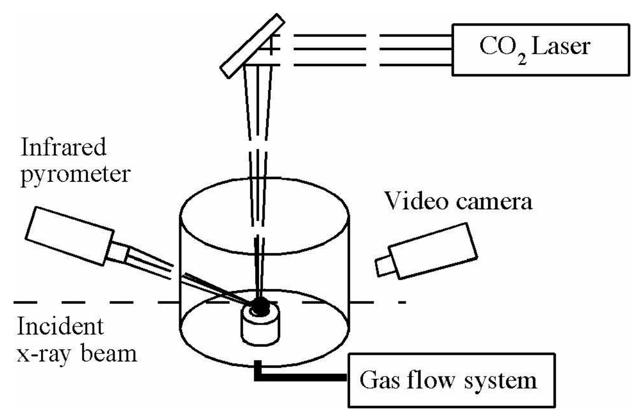


Top view



End view

Fig. 1 Schematic diagram of apparatus used for high-energy synchrotron x-ray diffraction measurements.

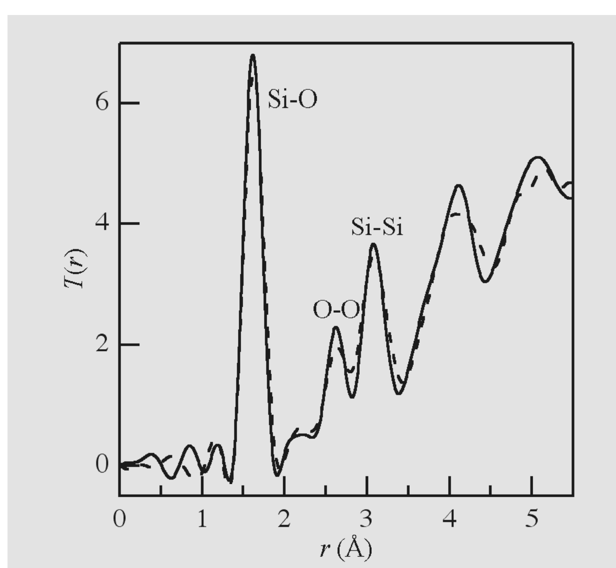


Fig. 2 The total correlation functions, $T(r)$, of amorphous SiO_2. Solid line: 26 ˚C, dashed line: 1330 ˚C ± 30 ˚C.