The Change in the Intermediate-Range Network Structure Plays a Key Role in the Migration of Magma in the Earth's Upper Mantle

We have found the occurrence of shear flow in usually brittle SiO_2 glass when compressed uniaxially in a diamond-anvil cell at room temperature. We have also measured the microscopic differential strain with a radial X-ray diffraction technique and clarified that SiO_2 glass transforms to a high-density low-viscosity phase by reconstructing the intermediate-range network consisting of SiO_4 tetrahedra. These results suggest that anomalous pressure dependence of the two important physical properties, density and viscosity, of silicate melts at the pressure range of the Earth's upper mantle can be attributed to the change in the intermediate-range order.

Numerous studies have long been conducted on the high-pressure behavior of glasses at room temperature because of their remarkable structural similarity to melts at high temperatures. In particular, it has been emphasized that the increase in coordination number of silicon takes place in silicate melts in the Earth's deep mantle, based on the observations in SiO_2 and other silicate glasses [1]. At the same time, SiO_2 glass has long been known for the phenomenon referred to as permanent densification [2]. This phenomenon is not related to the increase in coordination number but to the reconstruction of the intermediate-range network consisting of SiO_4 tetrahedra [3]. Our analyses have revealed that the anomalous pressure dependence of density of silicate melts at the pressure range of the upper mantle can be well explained by assuming the phase-transition-like structural change in the intermediate-range order [4, 5]. Structural changes are also expected to have strong effects on the viscosity of melts. In this study, uniaxial-compression experiments on SiO_2 glass were conducted to discuss the effects of the structural change in the intermediate-range order on the plasticity of glasses and hence the viscosity of melts.

Two types of high-pressure uniaxial-compression experiment, macroscopic-strain and microscopic-strain measurements, were conducted with a diamond-anvil cell at room temperature. In the former, the change in size of disk-shaped SiO_2 glass was measured with an optical microscope [Fig. 1(a)] [6]. In the latter, the azimuth-angle dependence of the first sharp diffraction peak (FSDP) of SiO_2 glass (at Q ~2 $Å^{-1}$) was measured with a radial X-ray diffraction technique, in which X-ray irradiated the sample from a direction perpendicular to the compression axis [Fig. 2(a)] [7]. The FSDP has been considered to be associated with the periodicity of the intermediate-range network. Radial X-ray diffraction measurements were also conducted on the recovered sample to obtain more complete structural information (up to Q ~15 $Å^{-1}$) [6].

The optical-microscope observations revealed that a large shear flow began at around 8–10 GPa, which is the onset-pressure condition of permanent densification, and continued at least up to 20 GPa [Fig. 1(b)]. Note that SiO_2 glass is usually considered to be a brittle material. The radial X-ray diffraction measurements showed significant anisotropy in the intermediate-range network at high pressures [Fig. 2(b)]. Measurements on the recovered sample suggested that the plastically deformed sample was in the fully densified state (~20% increase in density) and, very surprisingly, a large differential strain of ~3% (equivalent to ~2 GPa in differential stress) remained in the intermediate-range order [Fig. 2(c)]. However, this large residual strain (or anisotropic permanent densification) cannot directly explain the macroscopic differential strain (or shear flow) that is about an order of magnitude larger. The weakening of the Si-O-Si bond with the densification is supposed to facilitate the rearrangement of the network and make a large shear flow possible.

The density and viscosity of silicate melt are very important for understanding the dynamics of the Earth and planetary interiors. The density contrast between silicate melts and crystals and the viscosity of silicate melts are directly related to the migration of magma as the driving and resisting forces, respectively. Here, we discuss the pressure dependence of viscosity of silicate melt on the basis of our experimental results on the plasticity of SiO_2 glass. The viscosity of SiO_2-rich melts has been reported to decrease with increasing pressure [8]. This anomalous decrease in viscosity has often been explained by a model in which the increase in the coordination number of aluminum plays a key role. Several other models have also been proposed. However, none of these models can explain our experimental results on SiO_2 glass. It seems to be more appropriate to introduce a model in which the change in intermediate-range order plays a key role. The pressure dependence of viscosity is supposed to be determined by a balance between two competing effects: the negative effect due to the weakening of the Si-O-Si bond (caused by the change in intermediate-range order) and the positive effect due to the decrease in free volume. The anomalous pressure dependence of the two important physical properties, density and viscosity, of silicate melts at the pressure range of the Earth's upper mantle can be attributed to the change in the intermediate-range network structure.

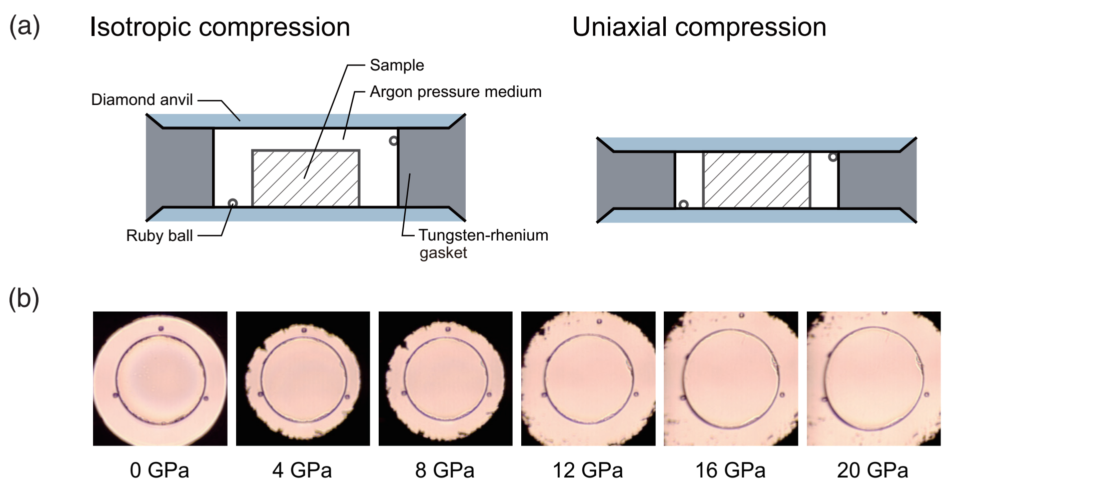

Figure 1: (a) Schematic illustrations of the cross section of the sample chamber and (b) optical-microscope images in high-pressure deformation experiments. The disk-shaped sample having an initial diameter of ~150 μm was compressed isotropically up to 6–8 GPa and its size decreased with increasing pressure. The sample was pinched directly by the two anvils and compressed uniaxially at higher pressures and its size in the radial direction increased with increasing pressure.

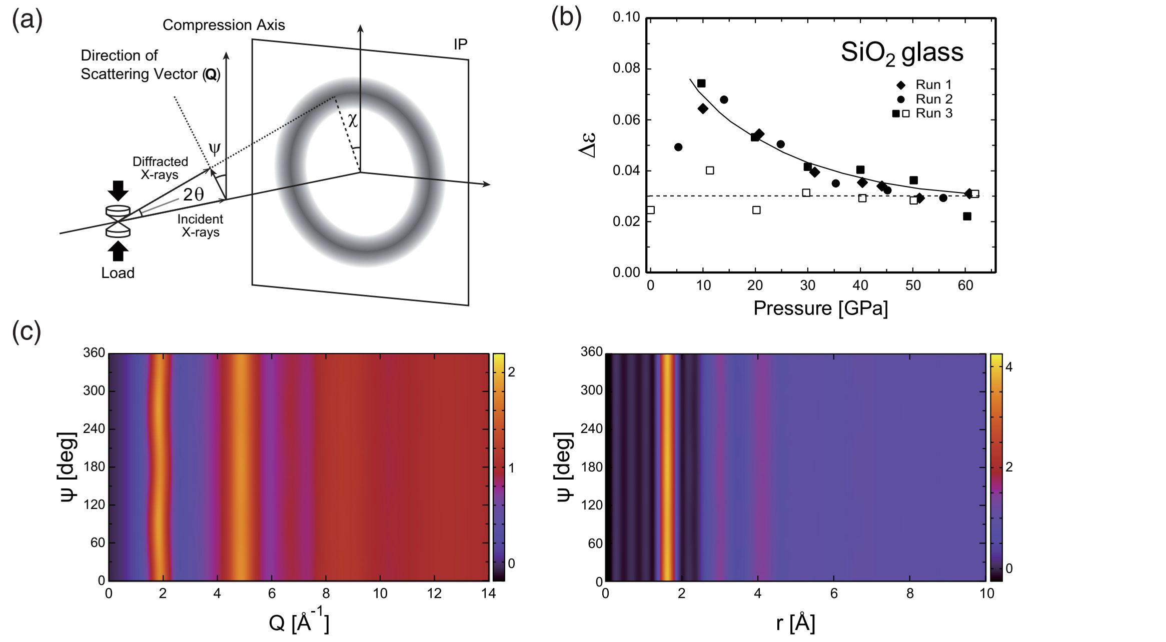

Figure 2: (a) Schematic illustration of the radial X-ray diffraction geometry. (b) Pressure dependence of the microscopic differential strain calculated from the azimuth-angle dependence of the FSDP of SiO_2 glass. Black and white symbols represent the data on compression and decompression, respectively. The solid and broken lines are to guide the eye. A large differential strain of ~3% remained after decompression. (c) Azimuth-angle dependence of the structure factor and pair distribution function of the recovered sample. Again, the FSDP showed significant anisotropy. The structure factor at a higher-Q range and the first peak of the pair distribution function (corresponding to the Si-O bond) did not show any significant anisotropy, indicating that differential strain did not remain in the basic unit of the SiO_4 tetrahedron.

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BEAMLINES

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