Structure of basaltic glass at pressures up to 18 GPa

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ABSTRACT

The structures of cold-compressed basaltic glass were investigated at pressures up to 18 GPa using in situ X-ray and neutron diffraction techniques to understand the physicochemical properties of deep magmas. On compression, basaltic glass changes its compression behavior: the mean O-O coordination number (CN_{oo}) starts to rise while maintaining the mean O-O distance (r_{oo}) above about 2–4 GPa, and then CN_{oo} stops increasing, and r_{oo} begins to shrink along with the increase in the mean coordination number of Al (CN_{al}) above ~9 GPa. The change around 9 GPa is interpreted by the change in contraction mechanism from bending tetrahedral networks of glass to increasing oxygen packing ratio via the increase in CN_{ad}. The analysis of the oxygen packing fraction (\eta_o) under high pressure reveals that \eta_o exceeds the value for dense random packing, suggesting that the oxygen-packing hypothesis recently proposed cannot account for pressure-induced structural transformations of silica and silicate glasses. The rise of the CN_{oo} at 2–4 GPa reflects the elastic softening of fourfold-coordinated silicate glass, which may be the origin of anomalies of elastic moduli in basaltic glass at ~2 GPa previously reported by Liu and Lin (2014).

The widths of both the first sharp diffraction peak and the principal peak show contrastive compression behaviors between modified silicate and silica glasses. This result suggests that modified silicate represent different pressure evolutions in the intermediate- and extended-range order structures from those of silica glass, likely due to the presence of modifier cations and the resultant formations of smaller rings and cavity volume.

Keywords: Glass structure, permanent densification, high pressure, X-ray diffraction, neutron diffraction; Physics and Chemistry of Earth’s Deep Mantle and Core

INTRODUCTION

It is well known that the physicochemical properties of magmas, such as density, viscosity, and elastic moduli, are sensitive to their atomic structures (e.g., Sakamaki 2018). Thus, the structural change under high pressure has attracted considerable interest. Although it is important to explore the macroscopic properties and structures of silicate melts at high pressures and temperatures, many technical challenges are involved in performing in situ experiments of melts under extreme conditions. Glasses, which can be a quenched form of the liquid, are believed to show a similar behavior; thus, numerous studies on the glasses have been undertaken to better understand the physical properties of silica and silicate melts under pressure (e.g., Meade and Jeanloz 1988; Sato and Funamori 2008; Liu and Lin 2014). A considerable number of experiments aimed at determining high-pressure structures of silica and silicate glasses have also been conducted by X-ray diffraction (XRD) (e.g., Sato and Funamori 2010; Prescher et al. 2017; Petitgirard et al. 2018; Ohashi et al. 2018) and neutron diffraction (ND) (e.g., Wilding et al. 2012; Zeidler et al. 2014; Salmon et al. 2019). From these studies, silica and silicate glasses are believed to exhibit two types of states under pressure. The first one is characterized by the fourfold-coordinated ordinary glass, which contracts the intermediate-range order (IRO) structure at pressures below ~10–20 GPa (e.g., Wang et al. 2014; Sanloup 2016; Petitgirard et al. 2019). The second one is characterized by the highly coordinated state above ~40–60 GPa (e.g., Benmore et al. 2010, 2011; Sato and Funamori 2010; Prescher et al. 2017; Petitgirard et al. 2019). Between these states, the Si(Al)-O coordination number gradually increases from 4 to 6. Recent ab initio molecular dynamics (MD) simulation studies (Ghosh et al. 2014; Ghosh and Karki 2018) have also predicted these states. Irreversible structural changes in SiO2 glass at room temperature have led to the proposition that permanently densified SiO2 glass exhibits different decomposition behavior from that of ordinary SiO2 glass and the decomposition behavior varies with the highest pressure applied (Grimsditch 1984; Vandembroucq et al. 2008). This densified state was interpreted by Wakabayashi et al. (2011) as a mixture of low- and high-pressure IRO structures. The high-pressure IRO component is composed of small Si-O-Si rings (Zeidler et al. 2014; Ryuo et al. 2017) and inter-tetrahedral voids (Wilding et