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Acoustic wave velocity measurements of SiO₂ - Al₂O₃ glasses up to 200GPa

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Determination of the structure and physical properties of silicate melt under high pressure and high temperature is an important key to understand the stratified structure of the Earth through the global magma ocean in early Earth, and the gravitational stability of melts in Earth $^{'}$ s deep mantle. Natural silicate melts mainly consist of SiO $_2$ with various chemical components. Al $_2$ O $_3$ is one of the most abundant components in natural silicate melts after SiO $_2$ (e.g., Mysen, 2005). Thus, it is essential to understand the effect of Al $_2$ O $_3$ on the density and the structure of silicate melts under the high pressure and high temperature condition corresponding to the Earth $^{'}$ s mantle. However, the density and the structure of silicate melts under the whole mantle condition are poorly understood, and it is unclear whether an aluminous silicate melt is buoyant or not in a certain mantle condition.

In this study, in-situ high-pressure acoustic wave velocity measurements of SiO_2 - Al_2O_3 glass were performed up to around 200 GPa using Brillouin scattering spectroscopic techniques to understand the effect of Al_2O_3 on pressure-induced structural changes in silicate glasses, as the analogue of silicate melts, under the whole mantle pressure conditions. We used both SiO_2 + 3.6 mol% Al_2O_3 glass (SA1) and SiO_2 + 20.0 mol% Al_2O_3 glass (SA2) as a starting material, synthesized using container-less liquid phase processing at the SPring-8 BL04B2.

The results below 30-40 GPa showed that the acoustic wave velocity of SA1 and SA2 increases with increasing Al_2O_3 content at a certain pressure, showing that the difference of bulk modulus (K) and shear modulus (μ) occurs depends on Al_2O_3 content in SiO_2 - Al_2O_3 glasses. It is suggested that K, μ and of SiO_2 - Al_2O_3 glasses become large with progressing depolymerization in this pressure range.

At the pressures from 30-40 GPa to 100 GPa, the velocity increase with pressure is more gradual. Above 100 GPa, the sharp increase in the velocity gradient were observed in both SA1 and SA2. The pressure at which the sharp increase in the velocity gradient occurs are 130 ± 5 GPa in SA1, and 116 ± 9 GPa in SA2, and these values are smaller than those of SiO₂ glass and MgSiO₃ glass (Murakami and Bass, 2010; Murakami and Bass, 2011). The pressure condition at which the change of the velocity gradient occurs in SA1 are 10 GPa lower than that in SiO₂ glass and 3 GPa lower than that in MgSiO₃ glass. Moreover, such pressure condition in SA2 are 24 GPa lower than that in SiO₂ glass and 17 GPa lower than that in MgSiO₃ glass. This result may suggest that Si ions have a coordination number greater than 6 in both SA1 and SA2 around above mentioned pressures, and Al₂O₃ could lower the pressure condition for the formation of the Si-O coordination numbers higher than 6. The results suggest that an aluminous silicate melt is likely to become dense at much shallower depth than CMB.

Keywords: Structure of silicate glasses and melts, Brillouin scattering, Acoustic wave velocity measurement

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