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Spin state of iron in MORB glass up to the lowermost mantle conditions

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The existence of deep magma has been suggested by the studies of physical observations or high P-T experiments subjected to the deep Earth (e.g., Lee et al., 2010; Williams and Garnero, 1996). These melts are considered to contribute to the ultra-low velocity zone on the core-mantle boundary (CMB). However, the presence of the deep magma has been controversial yet because the physical and chemical properties of silicate melts are not understood enough under high P-T conditions.

The spin state of iron in the silicate melt is one of the important factor to affect the gravitational stability of the deep magma. The iron is one of the abundant element in the Earth and its amount is critical for the density of the silicate minerals and liquids. The iron portioning into the silicate melts was reported at pressures greater than ~76 GPa (Nomura et al., 2011), which might cause the iron-rich dense melt above the CMB region. They suggested the iron partitioning was able to be changed due to the spin crossover of iron (from high-spin to low-spin) occurring around 70 GPa. On the other hand, some recent studies reported no spin transitions in the silicate glass at the pressures corresponding to the Earth's mantle (e.g., Mao et al., 2014; Prescher et al., 2014).

The previous studies about the spin state of iron discussed using the results limited up to the relative lower pressures (~80 GPa) or the simple component glass expected to the actual Earth's composition. Here, we report the results of the iron spin state measurements for the multicomponent silicate glass under high pressure up to about 130 GPa, corresponding to the lowermost mantle depth. The iron spin state was measured using synchrotron ⁵⁷Fe Mössbauer spectroscopy method, which bring the direct information about the iron spin state.

The starting material was synthesized ⁵⁷Fe-enriched silicate glass, which represented an average composition of mid-ocean ridge basalt. The glass was prepared by quenching molten mixture of oxides; SiO₂, MgO, Al₂O₃, TiO₂, ⁵⁷Fe₂O₃ and CaCO₃, K₂CO₃, Na₂CO₃. High pressure experiments were performed using a diamond anvil cell. The silicate glass powder was sand-wiched between two NaCl layers which worked as a pressure medium and a pressure scale. We used equations of state for NaCl B1 and B2 phases reported by Matsui (2009) and Fei et al. (2007), respectively. In case of experiments without XRD technique, the pressure was measured based on the pressure dependence of diamond T_{2g} mode presented by Akahama and Kawamura (2004). Energy domain Synchrotron ⁵⁷Fe Mössbauer spectroscopy was conducted at beamline BL10XU and BL11XU of SPring-8 at room temperature and pressure ranged from 1 atm to 130 GPa. Spectra were collected for 3-11 hours depending on the data qualities. The recorded spectra were fitted with Lorentzian doublets using the MossA software package (Prescher et al., 2012).

The obtained Mössbauer spectra was able to be fitted better supposing two doublet components, which might be derived from high-spin (HS) Fe²⁺ and HS Fe³⁺. Quadrupole splitting (QS) values of two doublets components were apt to increase from ambient condition to high pressure up to 130 GPa and were suited for the values of the previous studies. On the other hand, the change of the isomer shift (IS) had less pressure dependence. The ratio of ferrous iron to the total iron spiecies, Fe²⁺/ Σ Fe showed an increase tendency up to about 60 GPa. That tendency turned to decreasing at higher pressures up to 95 GPa, and then the ratio seemed to be constant over 100 GPa. The drop of Fe²⁺/ Σ Fe resembled the case of NaFe-silicate glass reported by Prescher et al. (2014) although the onset of decreasing in present study pointed at higher pressure than Prescher et al. (2014).