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Effect of iron on the structure of peridotitic silicate melts

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Abstract: Understanding the structure and physical properties of SiO_2-poor and Fe-rich peridotitic silicate melts is fundamental in discussing the nature and dynamics of magma ocean in the early Earth and planets. Electrostatic levitation furnace (ELF) at the International Space Station (ISS) enables us to investigate density and viscosity of simplified peridotitic MgO-FeO-SiO_2 melts. In addition, we investigate structure of MgO-FeO-SiO_2 melts and peridotite melts to understand physical property change in view of melt structure. This talk will review our recent study of the structure and physical properties of peridotitic silicate melts, and will discuss particularly about the effect of iron on the structure of peridotitic silicate melts.

Keywords: melt structure, iron valence, polymerization, XANES, Raman spectroscopy

1. Introduction

Knowledge of the structure and physical properties of SiO_2-poor and Fe-rich peridotitic silicate melts is fundamental in discussing the nature and dynamics of magma ocean in the early Earth and planets, and magmas in the current planetary interiors. Silicate melt is considered to be composed of polymer-like network structure, and the degree of polymerization decreases with decreasing SiO_2 content. As a result, physical properties such as density and viscosity significantly change with varying SiO_2 content. In addition, effect of iron on the structure and physical properties of peridotitic melt is important in understanding the nature of the magma ocean in the early Earth and planets. The initial magma ocean is expected to be enriched in iron, but iron content in the magma ocean would decrease during core formation process. The change in iron content in peridotitic melt may affect the melt structure and the resultant physical properties. However, effect of iron on the structure and physical properties of peridotitic silicate melts has not been well understood.

There are numbers of previous Earth science studies about the structure and physical properties of SiO_2-rich silicate melts. However, structure and physical properties of SiO_2-poor peritoditic melts have not been well investigated mainly due to two experimental difficulties. One is temperature limitation in common electric furnace up to ~1600 °C. The temperature range is enough high to melt SiO_2-rich silicate compositions, while SiO_2-poor silicate compositions have liquidus temperature higher than 1600 °C. Another experimental difficulty is high reactivity of iron in silicate melt with container material such as platinum crucible. Container

material is essential to keep molten sample in common electric furnace, while it is challenging to avoid chemical reaction between iron-rich silicate melt and container material at high temperature conditions.

In order to overcome these experimental difficulties, we utilized electrostatic levitation furnace (ELF) at the International Space Station (ISS), and we succeeded in experimentally investigating density and viscosity of simplified peridotitic melts with MgO-FeO-SiO_2 compositions at high temperatures (Kono et al., 2025). In addition, we used aerodynamic levitation furnace on the ground to conduct melting experiment of peridotitic compositions and to recover glass sample for structure analysis. In this talk, we will review our recent study of the structure and physical properties of peridotitic silicate melts, and discuss particularly about the effect of iron on the structure of peridotitic silicate melts.

2. Effect of iron and SiO_2 content on the density and viscosity of MgO-FeO-SiO_2 melts

We investigated density and viscosity of four SiO_2-poor and iron-rich peridotitic silicate melts with $Mg_{0.8}Fe_{0.2}SiO_3$, $Mg_{1.8}Fe_{0.2}SiO_4$, $Mg_{0.7}Fe_{1.2}SiO_4$, and $Mg_{0.9}Fe_{1.6}SiO_{4.5}$ compositions (Fe/(Mg+Fe) ratio = 0.10-0.63, (Mg+Fe)/Si ratio = 1.00-2.52) in the ELF at the ISS. The sample was levitated in an Ar gas environment under 2 atm pressure condition in the ELF. The volume of the melt sample at high temperature condition was determined from the sample image acquired with ultraviolet black light, and the mass of the recovered sample was weighed on the ground. Viscosity measurement was conducted by using the drop oscillation method. Sinusoidal voltages excite an oscillatory deformation on the melt sample. When the excitation voltage is stopped, the sample oscillation gradually weakens due to its viscosity. The viscosity of melt was calculate by the decay time with the density and radius of the sample.

We succeeded to obtain density of all four samples at wide range of temperature conditions between 1235 and 2465 K. Our experimentally obtained density of $Mg_{0.8}Fe_{0.2}SiO_3$ melt, which has relatively high SiO_2 content, is similar to that calculated by the previous density model based on SiO_2-rich silicate melts data (Guo et al., 2014). In contrast, there are marked differences in the densities of SiO_2-poor $Mg_{1.8}Fe_{0.2}SiO_4$, $Mg_{0.7}Fe_{1.2}SiO_4$, and $Mg_{0.9}Fe_{1.6}SiO_{4.5}$ melts between our experimental results and the previous density model. The density differences increase with decreasing SiO_2 content, which is considered to be due to uncertainty caused by extrapolation of the previous density model for SiO_2-rich basaltic melt to SiO_2-poor peridotitic melts. In addition, we found significant differences in the temperature dependences of the densities of $Mg_{0.8}Fe_{0.2}SiO_3$, $Mg_{1.8}Fe_{0.2}SiO_4$, $Mg_{0.7}Fe_{1.2}SiO_4$, and $Mg_{0.9}Fe_{1.6}SiO_{4.5}$ melts between our experimental results and those calculated by the previous density model (Guo et al., 2014), indicating that thermal expansivities of SiO_2-poor peridotitic silicate melts are much smaller than those of SiO_2-rich silicate melts. Effect of iron content on the density of $(Mg,Fe)_2SiO_4$ melts show almost linear increase of density with increasing Fe# [Fe#=Fe/(Mg+Fe)].

In addition to the density measurements, we succeeded to determine viscosities of $Mg_{1.8}Fe_{0.2}SiO_4$, $Mg_{0.7}Fe_{1.2}SiO_4$, and $Mg_{0.9}Fe_{1.6}SiO_{4.5}$ melts at the temperature conditions of 1980-2569 K. The viscosity increases with increasing iron content, while decreases with decreasing SiO_2 content. Our obtained viscosity results of the SiO_2-poor and iron-rich peridotitic silicate melts are compared with the previous viscosity model based on the data of SiO_2-rich silicate melts (Ghiordano et al., 2008). We found marked difference in the temperature dependence of the viscosities. Our results show markedly lower temperature dependences of the viscosities of the $Mg_{1.8}Fe_{0.2}SiO_4$, $Mg_{0.7}Fe_{1.2}SiO_4$, and $Mg_{0.9}Fe_{1.6}SiO_{4.5}$ melts than those calculated by the previous viscosity model (Ghiordano et al., 2008).

3. Effect of iron content on the structure of MgO-FeO-SiO_2 and peridotitic silicate melts

We investigated effect of iron content on the structure of two silicate glasses with a simplified $(Mg,Fe)SiO_3$ compositions and a peridotitic MgO-FeO-CaO-Al_2O_3-SiO_2 compositions. Melting experiments were conducted by an aerodynamic levitation furnace under air atmosphere at the Geodynamics Research Center in Ehime University. Fe^{3+} ratio was determined by Fe L_{III}-edge XANES measurement at BL27SU in SPring-8. The structure of glass was investigated in terms of Q^n and NBO/T (non-bridging oxygen per tetrahedral cation) by Raman spectroscopy.

The Fe^{3+} ratio increases with increasing iron content in $(Mg,Fe)SiO_3$ and peridotitic melts. Structure of $(Mg,Fe)SiO_3$ glass shows depolymerization with increasing Fe#. On the other hand, structure of the peridotite glass polymerizes with increasing Fe#. The data imply distinct behavior of Fe^{3+} in the glass structure between $(Mg,Fe)SiO_3$ and peridotite glasses. The Fe^{3+} may act as network modifier in $(Mg,Fe)SiO_3$ melt, while Fe^{3+}

incorporate as network former in peridotite melt. These results show that effect of iron on the melt structure depends not only on the iron content itself but also by SiO_2 content or other oxide contents.

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