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Incorporation of Fe^{2+} and Fe^{3+} in bridgmanite during magma ocean crystallization

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Abstract

Using large volume press, samples of bridgmanites (Bg) in equilibrium with both silicate melt and liquid Fe-alloy were synthesized to replicate the early period of core-mantle segregation and magma ocean crystallization. We observe that the Fe partition coefficient between Bg and silicate melt ($D_{Fe}^{Bg/melt}$) varies strongly with the degree of partial melting (F). It is close to 1 at very low F and adopts a constant value of ~0.3 for F values above 10 wt%. In the context of a partially molten mantle, a larger F (closer to liquidus) should yield Fe-depleted Bg grains floating in the liquid mantle. In contrast, a low F (closer to solidus) should yield buoyant pockets of silicate melt in the dominantly solid mantle.

We also determined the valence state of Fe in these Bg phases using X-ray absorption near-edge spectroscopy (XANES). Combining our results with all available data sets, we show a redox state of Fe in Bg more complex than generally accepted. Under the reducing oxygen fugacities (f_{O_2}) of this study ranging from IW-1.5 and IW-2, the measured Fe^{3+} content of Bg is found moderate ($Fe^{3+}/\Sigma Fe = 21 \pm 4\%$) and weakly correlated with Al content. When f_{O_2} is comprised between IW-1 and IW, this ratio is correlated with both Al content and oxygen fugacity. When f_{O_2} remains between IW and Re/ReO_2 buffers, $Fe^{3+}/\Sigma Fe$ ratio becomes independent of f_{O_2} and exclusively correlated with Al content.

Due to the incompatibility of Fe in Bg and the variability of its partition coefficient with the degree of melting, fractional crystallization of the magma ocean can lead to important chemical heterogeneities that will be attenuated ultimately with mantle stirring. In addition, the relatively low-Fe^{3+} contents found in Bg (21%) at the reducing conditions (IW-2) prevailing during core segregation seem contradictory with the 50% previously suggested for the actual Earth's lower mantle. This suggests the presence of 1.7 wt% Fe^{3+} in the lower mantle, which reduces the difference with the value observed in the upper mantle (0.3 wt%). Reaching higher concentrations of trivalent Fe requires additional oxidation processes such as the late arrival of relatively oxidized material during the Earth accretion or interaction with oxidized subducting slabs.

Keywords: Bridgmanite, lower mantle, magma ocean, melting, partitioning, redox, ferric iron, XANES