

Effect of cerium incorporation on the structure of hydrous rhyolitic melt

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Cerium (Ce) in silicate melts is considered to provide important information about the nature of silicate magmas in the Earth's and planetary interiors, and especially their oxidation state. However, the behavior of Ce in a silicate melt structure may not be simple, particularly in hydrous silicate melt where a strong depression of the Ce⁴⁺/Ce³⁺ ratio can occur (c.f. Smythe and Brenan, 2015). We investigated the effect of cerium incorporation into the structure of hydrous rhyolitic melt. Hydrous rhyolitic glasses quenched from melt at 1 GPa and 1300 °C show a significant decrease of H₂O solubility due to the incorporation of Ce. Pair distribution function measurement shows no distinct change in the local structures of the hydrous rhyolitic glasses with the incorporation of Ce, while we found a distinct change in the position of the T-OH (where T is Si or Al) Raman peak. The incorporation of Ce decreases the proportion of T-OH species and increases that of Q₄ and Q₃ species. These results imply the occurrence of a direct charge transfer reaction between hydrogen (H) in melt and incorporated Ce as: $2\text{CeO}_2 + 2\text{H}(\text{melt}) \rightarrow \text{Ce}_2\text{O}_3(\text{melt}) + \text{H}_2\text{O}$. This reaction causes a charge transfer from Ce⁴⁺ to Ce³⁺ in hydrous rhyolitic melt, and it inevitably produces Ce³⁺. The depression of Ce⁴⁺/Ce³⁺ ratio reported in hydrous silicate melts may be due to the charge transfer reaction between Ce and H, which implies that the Ce⁴⁺/Ce³⁺ ratio in hydrous silicate melts may not simply reflect the oxidation state of magmas.

Reference:

Smythe, D. J., & Brenan, J. M. (2015). Cerium oxidation state in silicate melts: Combined fO₂, temperature and compositional effects. *Geochimica et Cosmochimica Acta*, 170, 173-187.