

Hydrogen isotope fractionation between melts and aqueous fluid: Implications for magmatic processes

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The δD of silicate glass inclusions trapped in minerals is an important tool in geochemistry because it provides information on the δD of deep fluids, and, therefore, on the water cycle between the atmosphere, the hydrosphere and the lithosphere. Such understanding relies on the knowledge of the D/H isotopic fractionation values between aqueous fluids, melts, and minerals at temperature and pressure conditions relevant to the interior of the Earth. Significant D/H isotopic fractionation between silicate melts and aqueous fluids were reported at hundreds of MPa and °C by using in situ measurements in hydrothermal diamond anvil cell (HDAC)[1] However, available datasets are focused on fluids and melts with D/H ratios close to unity. The relevance of such data for natural processes D/H ratios in the ‰ range may not always be clear because the a role of bulk D/H ratio of the system was not assessed. To address such concerns, the effect of the bulk D/H ratio on hydrogen isotopes partitioning between water-saturated silicate melts and coexisting silicate-saturated aqueous fluids has been determined in the $Na_2O\text{-}Al_2O_3\text{-}SiO_2\text{-}H_2O$ system. To this end, in situ Raman spectroscopy measurements were performed on fluids and melts with D/H ratios from 0.05 up to 2.67 at equilibrium in a HDAC, in the 300 - 800°C and 200 - 1500 MPa temperature (T) and pressure (P) range, respectively.

In the P-T range studied, the D/H ratios of fluids barely change with temperature (in average $\Delta H_{fluid} = 0.47 \pm 1.15$ kJ/mol). In contrast, the D/H ratios of coexisting melts display strong dependence on temperature (in average $\Delta H_{melt} = 7.18 \pm 1.27$ kJ/mol). The temperature-dependence of the D/H fractionation factor between melt and fluid ($\alpha_{melt-fluid} = D/H_{melt} / D/H_{fluid}$) is similar in all the experiments and can be written:

$$\ln(\alpha_{melt-fluid}) = -747 (\pm 75) \cdot T^{-1} + 0.65 (\pm 0.10) .$$

The $\alpha_{melt-fluid}$ is independent of the bulk D/H ratio of the melt + fluid system.

Therefore, experimental determination of $\alpha_{melt-fluid}$ using D-enriched fluids can be applied to natural systems. As a result, hydrogen isotope fractionation between silicate melts and aqueous fluids probably affect the δD values recorded in the geochemical observables such as the melt inclusions (MIs). Such knowledge alters the present understanding of the water cycle in subduction zones. Indeed, it has been assumed that the δD composition of MIs trapped in crystals in back-arc basalts represents the δD composition of the slab fluids present in the mantle wedge. This interpretation was based on the hypothesis that no fractionation occurs between the slab fluids and the deep magma. However, the present data leave this hypothesis open to question, and indicate that D and H likely fractionated between the slab fluids and the back-arc magma. Although these data do not allow a definitive quantification of the effect because of the simple chemical composition we used, they demonstrate the necessity of a more complete understanding of H and D fractionation processes between silicate melts and aqueous fluids. Such processes may have a direct impact on δD of the various isotopic reservoirs in subduction zones, and therefore influence our understanding of the water cycle in such a geological context.

[1] Mysen, B. O. American Mineralogist 98, 1754-1764 (2013)

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