

Boron isotope geochemistry of the hydrothermally altered oceanic crust in the Oman ophiolite

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The vertical profiles of boron contents and boron isotopic compositions were determined for the hydrothermally altered rocks through whole sequence of the oceanic crust in the Oman ophiolite. In order to determine the boron isotopic composition of rock with low boron concentration, we improved the analytical technique using P-TIMS [1]. Our new method made it possible to determine the boron isotopic composition of 100 ng of boron in natural silicate samples with satisfactory analytical precision and accuracy ($\pm 0.2\%$, 2SD). The boron contents of the basaltic and doleritic rocks altered at low temperature ($< 200^\circ\text{C}$) were higher than fresh MORB, indicating uptake of boron from seawater. In contrast to the previous observations of boron reaching from rock in water-rock interaction at high temperature [2, 3], the gabbros altered at high temperature ($> 300^\circ\text{C}$) were significantly enriched in boron than fresh gabbro, with average value of 1.3 ppm. The $\delta^{11}\text{B}$ values of rocks increased with depth, showing a negative correlation with $\delta^{18}\text{O}$ values. This indicates that the decrease in isotope fractionation between rock and fluid with increasing temperature of alteration. The calculated output flux of boron by hydrothermal alteration of oceanic crust (12×10^{10} g/y) accounts for $\sim 30\%$ of riverine input flux [4], thus the altered oceanic crust is fairly large sink of boron. Furthermore, the $\delta^{11}\text{B}$ value of the oceanic crust increases by $\sim 10\%$ through hydrothermal alteration. The boron distribution in the oceanic crust indicates that boron added to arc magma at convergent margin can be originated from not only the upper oceanic crust but also the lower oceanic crust.

[1] Nakamura *et al.* (1992) *Chem. Geol.* **94**, 193–204.
[2] Seyfried *et al.* (1984) *GCA* **48**, 557–569. [3] Ishikawa & Nakamura (1992) *GCA* **56**, 1633–1639. [4] Lemarchand *et al.* (2000) *Nature* **408**, 951–954.

Structure and properties of silicate-saturated fluids in the system $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ determined *in situ* at high temperature and pressure

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The structure of silicate-saturated aqueous fluids in equilibrium with crystals and melts in the $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ system has been determined *in situ* with the materials at temperature (up to 800°C) and pressure (up to 1350 MPa) with samples contained in an Ir-gasketed hydrothermal diamond anvil cell (HDAC). Pressure was calculated from temperature with the EOS of H_2O in constant-volume HDAC and by measuring the Raman shift of ^{13}C synthetic diamond. The pressure differences thus recorded were employed to extract silicate- H_2O mixing properties. Structure was probed with microRaman and microFTIR methods.

Coexisting molecular H_2O (H_2O^0) and OH-groups exist in silicate- and aluminosilicate-saturated fluids above $\sim 400^\circ\text{C}$ and ~ 0.5 GPa with their OH/ H_2O^0 -ratio positively correlated with temperature. Hydrogen bonding diminishes with temperature so that above $\sim 400^\circ\text{C}$, it cannot be detected, and resembles that of pure H_2O under similar temperature and pressure conditions.

Silicate speciation in fluid comprises Q^n -species similar to those reported in hydrous and anhydrous melts at high temperature and pressure. Fluids in the $\text{SiO}_2-\text{H}_2\text{O}$ system contain Q^0 and Q^1 species at $400^\circ\text{C}/760$ MPa and above, whereas at lower temperature and pressure only Q^0 species were detected. The Q^1/Q^0 abundance ratio is positively correlated with temperature (and silicate content). Negative volume of mixing, V^{excess} , derived from a comparison of PVT data of pure H_2O and those of silicate-saturated H_2O , is related to the Q^1/Q^0 abundance ratio in silicate-saturated fluid. The $V^{\text{excess}} \sim 0$ when Q^1/Q^0 approaches 0.

Aqueous fluids in equilibrium with aluminosilicate melt at similar pressures and temperatures also show OH/ H_2O^0 positively correlated with temperature and pressure with $(\text{OH}/\text{H}_2\text{O}^0)_{\text{melt}} > (\text{OH}/\text{H}_2\text{O}^0)_{\text{fluid}}$. The silicate speciation comprises Q^n -species with $n=0, 1, 2,$ and 3 . The silicate is, therefore, more polymerized than in $\text{SiO}_2-\text{H}_2\text{O}$ fluid.