

Experimental study of the Mg and Sr isotopic evolution of seawater interacting with basalt between 150 and 300 °C.

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The chemical exchange of material between seawater and the oceanic crust plays a major role in marine geochemical cycles [1]. Isotopic signatures provide an important means of tracing elemental transfer in hydrothermal environments, yet only a limited amount of experimental data on the extent of isotopic fractionation under these conditions is currently available. This study consequently investigated the extent of $\delta^{26/24}\text{Mg}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic variation during a seawater-basalt interaction experiments at 150, 250 and 290 °C. A suite of closed system experiments were run for several months at each temperature under saturated water pressure, using either crystalline or glassy basalt as the starting material and a water/rock ratio of 10 or 25.

Our results demonstrate that the dissolution of basaltic material in hydrothermal environments occurs at the same time as the precipitation of alteration minerals (mainly smectite and anhydrite), which is consistent with results from similar studies in the past (e.g. [2]). As expected, the rate of reaction using crystalline basalt was slower than with basaltic glass, and both sample types reacted faster at higher temperatures. The $^{87}\text{Sr}/^{86}\text{Sr}$ composition of the experimental fluids decreased from the initial seawater value (0.70916) towards the lower basaltic signature during the experiments (0.70317), demonstrating the progressive release of Sr during basalt dissolution. Magnesium was steadily removed from the fluid via the precipitation of clay minerals, with the residual fluids having progressively lighter $\delta^{26/24}\text{Mg}$ compositions. The mean Mg isotope fractionation factor ($\alpha_{\text{solid-solution}}$) observed at 250 °C was 1.0005 ± 0.0002 , supporting low-temperature evidence that clay minerals preferentially incorporate isotopically heavy magnesium [3].

These experiments provide quantitative information on the extent of Mg isotopic fractionation between fluids and secondary silicate minerals in hydrothermal systems, and demonstrate the potential for combined radiogenic and stable isotope analysis to track solid-fluid reactions in the oceanic crust. Further characterisation of the extent of isotopic fractionation in these systems will help establish how such processes have affected the long-term chemical evolution of the oceans.

[1] H. Elderfield and A. Schultz, "Mid-Ocean Ridge Hydrothermal Fluxes and the Chemical Composition of the Ocean," *Annu Rev Earth Planet Sci*, vol. 24, pp. 191–224, 1996.

[2] W. E. Seyfried Jr and J. L. Bischoff, "Experimental seawater-basalt interaction at 300°C, 500 bars, chemical exchange, secondary mineral formation and implications for the transport of heavy metals," *Geochim. Cosmochim. Acta*, vol. 45, no. 2, pp. 135–147, 1981.

[3] J. A. Higgins and D. P. Schrag, "The Mg isotopic composition of Cenozoic seawater – evidence for a link between Mg-clays, seawater Mg/Ca, and climate," *Earth Planet. Sci. Lett.*, vol. 416, pp. 73–81, 2015.