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The effect of the water-to-rock ratio on REE distribution in hydrothermal fluids: An experimental study

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High-temperature submarine MOR hydrothermalism creates high elemental fluxes into, and out of, oceanic lithosphere significantly affecting ocean chemistry. The Turtle Pits hydrothermal system discovered at 5° S on the slow-spreading Mid-Atlantic Ridge (MAR) in water depths of ∼3000 m (∼300 bar) emanates 'ultrahot' fluids > 400 °C [1] with high concentrations of dissolved gases (e.g., H_2), transition metals, and rare earth elements (REE). The normalised REE patterns of these 'ultrahot' fluids are uncommon as they exhibit depletions of LREE and no Eu-anomaly ('special' REE-signature in [2]), which is in contrast to the "typical" LREE enrichment and pronounced positive Eu-anomaly known from many MOR vent fluids observed world-wide [e.g., 3]. Although hydrothermal fluid REE-signatures may play a key role in understanding processes during water-rock interaction, only few experimental data have been published on REE distribution in seawater-like fluids reacted with rocks from the ocean crust [e.g., 4, 5]. Besides temperature, the seawater-to-rock ratio (w/r ratio) strongly affects water-rock reaction processes and, thus, has significant control on the fluid chemistry [e.g., 6, 7].

To understand how vent fluid REE-signatures are generated during water-rock interaction processes we designed a series of experiments reacting different fluid types with mineral assemblages from fresh, unaltered gabbro at 425 °C and 400 bar using cold seal pressure vessels (CSPV). Mixtures of 125-500 μm-sized hand-picked plagioclase and clinopyroxene grains separated from unaltered gabbro reacted in gold capsules with 3.2 wt.% $NaCl_{(aq)}$ fluid (similar to seawater salinity), or with natural seawater. The w/r (mass) ratio ranged from 1 to 100 and the run durations were varied from 3 to 30 d in the $NaCl_{(aq)}$ experiments, and was 3 d in the seawater experiments. The reacted fluids were extracted after quenching and analysed by ICP-OES and ICP-MS.

Only in the seawater experiments, the gabbro reacted considerably with the liquid resulting in a strong REE enrichment relative to the original seawater. Increasing w/r ratios gave rise to decreasing pH of the quench fluid and enforced the enrichment of HREE in the fluid with relative depletion of LREE and no Eu-anomaly. The 'special' REE-signatures observed in Turtle Pits vent fluids at 5°S MAR could be reproduced in our experiments at a w/r ratio of 5 (pH = ∼6), whereas at a w/r ratio of 1 (pH = ∼7) the fluid exhibited the "typical" REE pattern with a positive Eu-anomaly. Fluids from experiments with w/r ratios ≥10 (pH ∼6 to ∼2) showed higher HREE enrichment than observed in natural MOR vent fluids so far.

Concluding, elevated REE concentrations in hydrothermal fluids exhibiting the 'special' REE pattern with relative enrichment of HREE and no Eu anomaly are indicative for leaching processes under high w/r ratios (∼5-10) that might be more common in slow-spreading oceanic crust with focused hydrothermal fluid-flow along e.g., detachment faults [8].

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