

Evolution of the δD value in water-rich basaltic melt inclusions during volcanic processes

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Melt inclusions (MIs) hosted in Mg-rich olivines are powerful tools to record magma evolution and degassing during their decompression and ascent toward the surface. However their ability to lose H₂O during magma ascent and degassing through proton diffusion is widely debated [e.g. 1].

H₂O contents and δD values were measured in two series of melt inclusions representative of Ca-rich basalts to arc basalts, rich in H₂O, in using the Cameca IMS 1270 ion microprobe (CRPG-CNRS, Nancy, France) and Raman spectroscopy (Saclay, France, [2]), respectively. In lapilli samples from Vulcano island (Aeolian arc), MIs are hosted in olivine Fo₈₉₋₉₀, they contain on average 4.4 wt% of H₂O (± 0.2 wt%; 18 measurements) and the post-entrapment olivine crystallization is negligible. In Aoba samples (Vanuatu arc), MIs are entrapped in olivine Fo₈₆₋₉₀, with 2 to 5 % of post-entrapment crystallization; their H₂O is more variable (1.7-2.3 wt%). In both cases their low CO₂ concentrations indicate shallow olivine crystallization.

As a whole the δD values (42 measurements) vary from -96‰ to +38‰ (with two values even higher). Isotopic analyses have been duplicated in single inclusions of Vulcano samples to verify the data reproducibility.

Hydrogen diffusion out of MIs is required to explain negative trends between the H₂O concentrations and δD , in particular in one series of Aoba samples showing δD values ≥ 0 ‰. Leaving apart these latter values, most of the measured δD are comprised between -19‰ and -40‰. They plot in the field of Mariana arc MIs (from -55‰ to -12‰; [3]). The question that arises is to what extent our δD values are representative of the last stage of melt equilibration with their surroundings at time of their entrapment and olivine crystallization or bring constraints on the magma source. The different processes able to reproduce the trends registered by Aoba and Vulcano MIs will be discussed.

[1] Gaetani *et al.* (2009) *Eos Trans AGU* 90(52) Fall Meeting supp. Abst. V51E-1770. [2] Mercier *et al.* (2010) *Geochim Cosmochim Acta* **74**, 5641–5656. [3] Shaw *et al.* (2008) *Earth Planet. Sci. Lett.* **275**, 138–145.

Am(III) retention by cement corrosion products under highly saline conditions

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Cementation is a common method to fix and solidify low and intermediate radioactive waste (LLW/ILW). Alteration of radionuclide bearing cement products in diluted aqueous solutions has been studied quite extensively. Yet, in chloride-rich solutions, which are relevant for final LLW/ILW disposal in rock salt, there is still a lack of thermodynamic data and understanding with respect to both cement corrosion and radionuclide behaviour. In the present work, cement interaction with MgCl₂-rich brines have been studied experimentally on laboratory-scale and with full-scale waste simulates. In addition, Am (III) retention by cement corrosion products was quantified from the laboratory experiments.

Due to dissolution of portlandite, ettringite and other solid cement phases, the initially MgCl₂-rich solutions alter to alkaline CaCl₂-rich solutions. Besides reprecipitation of gypsum, formation of hydrotalcite phases, Mg-Ca-Al hydroxochloride and brucite is observed. Compositions of the altered solution and solid phases depend strongly on the cement / brine ratio. Measured solution compositions agree well with results of thermodynamic calculations for the studied cement / brine systems. After equilibration of the systems, they are doped with small quantities of acidic ²⁴¹Am (III) and ²⁴³Am (III) solutions (10⁻⁹, 10⁻⁸ and 10⁻⁷ mol Am (kg H₂O)⁻¹). Am concentrations and solution composition have been monitored for 1000 days in the non-agitated batch experiments. Within six months a sorption equilibrium is achieved, demonstrating strong retention of Am (III) by the corroded cement products. Data of equilibrated systems are characterized by linear sorption isotherms, which allow determining apparent sorption coefficients, R_s. In MgCl₂-CaCl₂ and weakly alkaline CaCl₂ systems (7.3 < -log (m_{H+}) < 9.0), R_s is measured in the range of 3000 to 6000 ml g⁻¹, whereas in highly alkaline CaCl₂ systems (-log (m_{H+}) ~ 11.5) R_s = 10000 ml g⁻¹ is observed. The retention of radionuclides on corroded cement products has often been claimed not only for dilute solutions, but also for saline brines. This work enables to quantify the retention of Am and thus corroborates previous estimations with experimental data.