

## High Cesium Concentrations in Groundwater in a Coastal Granitoidic Fracture Network

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This study aims to increase the understanding of the sources and mechanisms by which elevated Cs concentrations build up naturally in deep and low temperature (<20°C) groundwater residing in fractured crystalline bedrock.

The hydrochemical monitoring program carried out by the Swedish Nuclear Fuel and Waste Management Co (SKB) revealed that high variability in natural Cs concentrations, with values up to several µg/L (Fig. 1), occurs in fracture groundwater at the Äspö Hard Rock Laboratory (Äspö HRL) and the nearby Laxemar area (Sweden) down to 1155m depth. Related to the various types of water residing into the bedrock [1], the highest Cs concentrations were found in both old deep-seated saline and marinogenic groundwater (Fig. 1). The SEM-EDS and XRD analyses of fracture coatings reveal that high Cs concentrations (>50 ppm) in the bulk fracture coatings correlate with a mineral enrichment in illite, together with mixed-layer clays and chlorite. Based on hydrochemical modelling, intrusion of Baltic Sea water (K and NH<sub>4</sub> rich water), together with cation exchange, constitute a consistent explanation for the high Cs concentrations observed in the marinogenic groundwater at the Äspö HRL.

The Cs sorption mechanism and results presented here are relevant for recognition of conditions of importance in term of bedrock disposal of toxic waste materials as fractured bedrock constitutes the last retention barrier separating radionuclides (including <sup>134</sup>Cs and <sup>137</sup>Cs) from the biosphere if the release of radionuclides in the repository occurs.

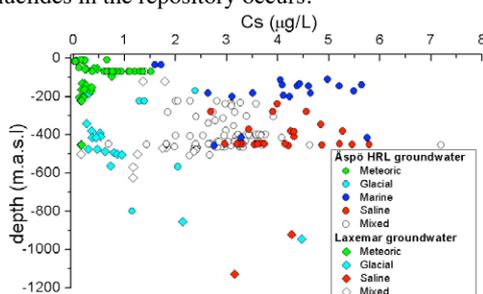


Fig. 1. Vertical distribution of the Cs concentrations in the fracture groundwater.

[1] Mathurin *et al.* (2012) *EST journal* **46**, 12779-12786.

## Control of oxygen fugacity in piston cylinder experiments

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Controlled oxygen fugacity experiments in a piston cylinder apparatus can be carried out using a double capsule technique and different solid oxygen buffers as suggested by Eugster (1957) and Jakobsson (2012). However both the capsule designs used in these studies as well as the choice of the spacer in the piston cylinder assemblage can affect the  $f_{O_2}$  in the experimental charge. The main goal of our study was concentrated on minimizing variability in  $f_{O_2}$  by testing different types of spacers, capsule materials and filling materials. In order to control and/or read the achieved  $f_{O_2}$  in the capsule we have examined the reaction of 3 different  $f_{O_2}$  sensors: NiPd, CoPd and magnetite-ilmenite. All experiments were carried out at 1GPa and 1000°C using the same piston cylinder apparatus.

The results show that use of Al<sub>2</sub>O<sub>3</sub> as spacer material in combination with noble metals such as Pd and Pt can lead to oxidation of experimental charge up to  $\Delta NNO+5$  as Pd or Pt do not prevent enough hydrogen loss during experiment. Alternatively, using wet spacer materials (e.g pyrophyllite) may reduce the experimental by diffusion of hydrogen into the capsule. Applying MgO spacer with Pt and AuPd (80/20) capsules we could achieve consistent  $f_{O_2}$  between  $\Delta NNO+2.5$  and  $+3.5$  for experiment durations up to 72 hours. In contrast, the use of pyrex (borosilicate glass) sleeve as spacer material in combination with AuPd capsules showed minimal loss of hydrogen (water) as well as no detectable boron diffusion through the capsule walls. As result, both NiPd and CoPd internal sensors indicated consistent  $f_{O_2}$  values of  $\Delta NNO+1$  ( $\pm 0.2$ ) for experiments tested up to 26 hours.

A further important factor we tested is the reaction of different solid buffers depending on the  $f_{O_2}$  we aspire to achieve. The metal sensors such as NiPd or CoPd are suitable for short and long time experiments (given the presence of an appropriate amount of water) and are able to register the changes of  $f_{O_2}$  already after first 12 hours of experiment. In contrast, magnetite-ilmenite oxide sensor needs more time for equilibration showing consistent results only after 24-72 hours runs.

[1] Eugster (1957) *J. Chem. Phys.* **26**: 1760-1761

[2] Jakobsson (2012) *Contrib. Mineral. Petrol.* **164**: 397-406