Large-scale matrix diffusion in crystalline rocks revealed by natural Cl, δ^{18} O and δ^{2} H tracers in pore water

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Matrix diffusion in crystalline rocks of low permeability has long been proposed as a retardation mechanism in radionuclide transport. However, its extent and efficiency under the conditions of underground repositories (ca. 400-700 m depth) have been difficult to define.

As part of the Swedish site investigation programme at Laxemar-Oskarshamn a continuous drillcore has been sampled, extending from a transmissive fracture zone into the low-permeability rock matrix at a depth of 112 m. Special sample-handling procedures allowed the rock to be retrieved in its saturated state and allowed its in situ water content to be measured, without modifications by drilling fluid or stress release. Pore-water chloride contents were then derived from out-diffusion experiments and the isotope composition from the diffusive isotope equilibration technique. The Cl timeseries data from these experiments can be described by radial diffusion indicating that diffusion might well be important under in situ conditions. The chloride contents of the pore water are similar to that in the fracture water at the fracturematrix interface. They describe a complex curved profile over the first 6 m of the drillcore, but then remain at background values (Figure 1) over several decametres (towards the next underlying conductive fracture). The δ^{10} O and δ^2 H values show similar profiles. The variations in pore-water Cl, δ^{18} O and δ^2 H reflect changes in the fracture-water composition over geological time, which can be correlated with the local hydrological history during the Holocene and Pleistocene. Taking account of this history, the shapes of the profiles suggest diffusion-dominated transport. This indicates that in crystalline rocks, matrix diffusion occurs over decametres even under present-day conditions.



Figure 1: Changes in Cl and δ^{18} O of pore water in crystalline rock as function of distance to a transmissive fracture.

Stable isotope geochemistry of amphiboles from alkaline igneous complexes

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It is now well established that the hydrogen isotope composition of amphiboles from alkaline complexes can have an extremely wide range, but also extremely low δD values (between -227 and -70%). It has been suggested that the extreme depletion in D is controlled by internally buffered magmatic processes that may be unique to highly peralkaline magmas (Marks et al., 2004). This possibility is further explored through a complete mineralogical and geochemical characterization of selected minerals from a number of welldescribed alkaline complexes, including examples from Greenland, the Kola peninsular of Russia, and NW Namibia. Amphiboles selected have compositions typical for calcic, sodic-calcic, and sodic amphiboles. The O-isotope compositions, in contrast to those for hydrogen, are quite homogeneous and are compatible with amphibole formation from mantle-derived magmas (values close to 5.5‰). In the amphiboles, the influence of Fe^{3+}/Fe^{2+} on δD values is negligible ($R^2_{Fe3+/Fe2+} = 0.3914$; n=17) and the control of iron content is small ($R^{2}_{Fe \text{ content}} = 0.6558$; n=23). Our data do not support interaction with a highly reducing fluid as a cause of the low δD values. However, a significant correlation $(R^2_{Na+K}= 0.8196; n=23)$ between the alkali content and δD value from the Ilímaussaq complex is observed. These data suggest that H-isotope compositions are controlled by a combination of Fe-content but more notably the alkali-element contents in the amphiboles, implying complex fluid-mineral equilibration in these highly alkaline magmas.

References

Marks M., Vennemann T.W., Siebel W., and Markl G., (2004) Geochim. Cosmochim. Acta 68 3379–3395.