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Introduction

Cement-based materials play an important role in multi-barrier concepts developed worldwide for the safe disposal of radioactive waste in deep geological repositories. In a deep geological repository very strong chemical gradients control mineral alterations at the interface between the cementitious near field (pH >12.5) and the surrounding claystone formations (pH 7-8) due to the very different chemical conditions encountered in these matrices. Reactive transport calculations showed that the alteration zone (pH plume) at the interface between the cement-based repository and the surrounding host rock spreads over the interface at very low rates (few micrometers per year). Experimental evidence for the chemical reactions and the mineral composition in the disturbed zone is almost completely lacking. The only indications have been made available from short-term laboratory experiment and natural analogue studies, such as from the Maqarin site.

Apart from short-term laboratory experiments and multi-year field experiments, a key-issue is the nature of alteration products at the cement/surrounding rock interface, which has been in contact over time periods relevant to geological disposal of radioactive and hazardous waste. For this purpose, the only known natural hyperalkaline system at Maqarin (Jordan) had been studied for more than a decade. There, high temperature and low pressure conditions led to the formation of clinker, and subsequent re-hydration to the formation of natural cement. U-Th disequilibrium series dating suggested an age of the cement mineralization of ~100000 years. Continuous leaching along fracture-bound groundwater flow-paths formed cementitious in-fills, interfaces to the adjacent bio-micritic and clay-bearing limestone, and diffusion-controlled wall-rock alteration. The fractured zones of the Maqarin samples contain zeolitic- and cement-type in-fills (e.g. ettringite, tobermorite-11 Å and 14 Å), and some calcite and siderite.

The present study aimed at developing an experimental approach based on microXRD to gain spatially resolved information on the phase assemblage at the interface between cementitious materials and the adjacent geological formation in the context of radioactive waste disposal. The approach was developed on a thin sample containing in-fill material mineralised in a fracture of clay biomicrite from the Maqarin area in northern Jordan.

Experiment and Results

Experimental technique and results are reported in the following publication:

Dähn, R., D. Popov, Ph. Schaub, P. Pattison, D. Grolimund, U. Mäder, A. Jenni, E. Wieland (2014). X-ray micro-diffraction studies of heterogeneous interfaces between cementitious materials and geological formations. Phys. Chem Earth. 70-71, 96-103.

Abstract

In the present study the challenge of analyzing complex micro X-ray diffraction (microXRD) patterns from cement-clay interfaces has been addressed. In order to extract the maximum information concerning both the spatial distribution and the crystal structure type associated with each of the many diffracting grains in heterogeneous, polycrystalline samples, an approach has been developed in which microXRD was applied to thin sections which were rotated in the X-ray beam. The data analysis, performed on microXRD patterns collected from a filled vein of a cement-clay interface from the natural analogue in Maqarin (Jordan), and a sample from a two-year-old altered interface between cement and argillaceous rock, demonstrate the potential of this method.