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Evolution of mineralogy and radionuclide diffusion on Portland-type -cementitious materials due geochemical and thermal effects

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Experimental work within the CEBAMA project focuses on alteration and interaction studies on cementitious materials (based on Portland type CEM II, w:c = 0.45, OPC) in contact with groundwater (GW) from Underground Research Facility Josef and with bentonite suspension (Czech B75 bentonite) in pressure vessels during three sampling campaigns (9/18/27 months) under in-situ (10℃) and high temperature (95℃) load. The first two sampling campaigns after 9 and 18 months were performed, and evolved solutions and solid samples were analysed. The main goal was to understand how the interaction processes affect the transport properties of cementitious materials. The HTO through-diffusion experiments on these evolved cylindrical samples (thickness of 8 mm, diameter of 50 mm) provided three types of datasets: depletion and breakthrough curves obtained during the experiments and concentration profiles in samples, which were obtained by a newly developed abrasive technique after the experiment´s termination.
A precipitate of carbonates, mainly calcite, was found on the surface of some samples, that was the most significant on samples after interaction of samples in bentonite suspension under 95℃ load; the oversaturation of calcite can be linked to large carbonates pool in bentonite and to calcium originating from the cement paste.
Based on chemical analyzes of evolved solution of groundwater in vessels and XRD after the interaction, the high temperature speeded up the dissolution / transformation of ettringite (C₄AF), which resulted in increase of sulfate concentration. Further analyzes of evolved solution revealed an increase of potassium and calcium. The heated samples contained also hydrothermal mineral katoite (C₃ASH₄) and miss the phases with stability under lower temperatures like CO₂/Cl-hydrocalumites.
The first results of HTO through-diffusion experiments on samples from the first sampling campaign showed that the high temperature raised values of effective diffusion coefficients (*D*_e), approx. in order of magnitude, which could be related to dissolution of material under higher temperature but also to the decrease of elasticity (microcracks). The mean values of HTO effective diffusion coefficients (for evaluation using GoldSim software) from three parallel experiments are the following:
- *D*_e = 2.8•10⁻¹¹ m²•s⁻¹ in the series: OPC + B75; 95℃; 9 months;
- *D*_e = 0.3•10⁻¹¹ m²•s⁻¹ in the series: OPC + B75; 10℃; 9 months;
- *D*_e = 2.0•10⁻¹¹ m²•s⁻¹ in the series: OPC + GW; 95℃; 9 months;
- *D*_e = 0.4•10⁻¹¹ m²•s⁻¹ in the series: OPC + GW; 10℃; 9 months.

The preliminary results (the concentration profile dataset were not considered) from through-diffusion experiments on samples from the second sampling campaign which are terminated between March and April 2018 showed no significant changes between two sampling periods.

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