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In-situ observations and reactive transport modeling of phase dissolution and precipitation processes at the granite–concrete interface in a radioactive waste repository

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Eleven core samples from the National Radioactive Waste Repository of Hungary, Bátaapáti have been studied for the aim of finding the signs of geochemical interactions among fluid, rock and building material. The cores all cross the granite–concrete interface in the repository and have been drilled from ~275 m depth from the surface, 1-15 months after concrete was placed in the chambers. Phase analytical techniques, such as XRD, TA and ATR-FTIR, as well as optical microscopy, SEM-EDS and Raman microspectroscopy were used for studying the samples. The analytical results show that calcite veins appear along the granite–concrete contact and that vaterite precipitates in the pores of concrete near the interface. This carbonation happens with the Ca^{2+}, coming from the dissolution of cement, and with the HCO_3^- content of the local granitic pore water. This process reduces the porosity and permeability. Furthermore, along the granite–concrete interface, titanite precipitation was frequently observed. The granite serves as a source of TiO_2 and the pH, Ca^{2+} and dissolved SiO_2 contents determined by the contacting cement allow the formation of this phase. Additionally, interactions between granitic pore water and hydrated cement paste have been also simulated by 1D kinetic reactive transport modeling in PHREEQC. The analytical results were used for the improvement of the model, which well matches the observations and now allows predicting long-term alterations of host rock and concrete of the repository.