Uranium sorption on calcite and degraded portland cement paste

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Near-surface radioactive waste disposal facilities are widely recognized as the most practical and efficient means of safely isolating Low and Intermediate-level short-lived waste (LILW-SL). Portland cement plays a critical role as both a structural material and an encapsulant for radioactive waste storage. This material has the ability to influence the behavior of dissolved species and acts as a significant adsorbent for organics and radionuclides (RNs). The retention capacity of Portland cement can vary over time due to degradation processes such as carbonation. In addition, the presence organic compounds such as cellulose degradation products, including isosaccharinic acid (ISA), may affect cement retention capacity. However, data on the sorption of RNs on long-term degraded hardened cements and the influence of ISA on RN mobility are scarce in the literature.

Therefore, this study aims at investigating the sorption of U(VI) on degraded cement paste and calcite (the main end-product of cement carbonation).

First, the combination of leaching and carbonation of Portland cement (type CEM I according to EN 197-1) pastes was studied under $CO_2(g)$ saturated wet environments at atmospheric conditions. This investigation deeply analyzes the paste carbonation process by characterizing intermediate carbonation stages (TGA, XRD, SEM-EDS). The findings obtained from the characterization results allowed the optimization of experimental conditions for achieving a degraded paste.

Furthermore, batch sorption experiments were conducted to evaluate the capacity of both calcite and degraded cement paste in retaining U(VI). Hence, these solid phases were contacted with their corresponding porewaters and the retention of U(VI) was evaluated at increasing RN concentrations $(10^{-7.5} \text{ M} < [U] < 10^{-5.5} \text{ M})$. Adsorption experiments were carried out both in the absence and in the presence of ISA $10^{-5} \text{ M} < [ISA] < 10^{-2} \text{ M}$ and under ambient atmosphere. The results from these experiments allowed shedding light on the sorption of U(VI) on degraded paste and the role of calcite therein. Preliminary results may indicate a higher retention of U on degraded paste, probably due to the presence of other calcite polymorphs and/or remaining phases other than calcite. Further investigation will be conducted to better understand the differences between calcite and degraded