

Uranium mobility in the process waste of a conversion facility (France): static experiments

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The COMURHEX Malvési industrial site is the first step in the treatment of uranium mining concentrate. Since 1959, the process waste, resulting from the conversion of yellowcake into uranium tetrafluoride (UF₄) has been managed in settling ponds. These ponds have been constructed on mine tailings and waste resulting from the flotation process of a former sulphur mine.

The settling waste, i.e. the resulting sludge, contains a variety of chemicals (nitrates, carbonates, fluorides and sulphates) and radioactive elements (the alpha activity in the solid phase is attributable to uranium (34%) and thorium-230 (64%)).

Previous studies of the source term suggest that the mine tailings act as an efficient buffer that minimises migration to the underlying ground. However, the actual mechanisms by which this control is active are not well understood.

This contribution focuses on the mobility of uranium in the waste based on the results of static experiments (batch studies) of leaching of solid samples. We aim to examine the desorption mechanisms that may account for release of contaminants from the waste and compare the results to field conditions.

In situ GISAXS studies of carbonate mineral nucleation on mineral surfaces

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The precipitation of carbonate minerals –mineral trapping– is considered the safest CO₂ sequestration mechanisms. However, the high pressure and temperatures and the high salinity of the fluids present in many geological reservoirs play important roles in modifying the properties of the mineral faces exposed to the solution, adding more uncertainties to the factors controlling nucleation in these environments. The goal of the present study is to determine the thermodynamic factors controlling heterogeneous nucleation of carbonate minerals on mineral surfaces representative of both reservoir and cap rocks. The quartz (100) and a mica (001) faces have been selected as representative of a sandstone reservoir and as a mineral face characteristic of cap-rocks, respectively.

A technique based on Grazing-Incidence Small-Angle X-ray Scattering, previously developed in our group [1], has been used to follow mineral nucleation of the carbonate minerals on quartz (100) and K-muscovite (001). By measuring nucleation rates at different saturation indexes, effective interfacial energies (α') can be determined. Effective interfacial energies result as the combination of different interfacial energies involved in the process: α_s (liquid-substrate), α_{lc} (liquid-crystal) and α_{sc} (substrate-crystal). Literature values for α_s and α_{lc} allow determining α_{sc} , a quantity very difficult to measure using other techniques.

The first experiments have been performed nucleating CaCO₃ on quartz (100) at ambient pressure and room temperature. The results show that heterogeneous nucleation of calcium carbonate is favored on quartz (100), even though the obtained substrate-crystal interfacial energies are higher than the typical values for liquid-crystal interfaces (e.g., ~100 mJ/m² for calcite-water interfaces). This fact highlights the interplay existing between the different interfacial energies at play, and the ‘competition’ between water and the nucleated crystals for the substrate. From a thermodynamic point of view, and as a general rule of thumb, high substrate hydrophobicity and low surface mismatch are found to favor heterogeneous nucleation. In addition, a crystal growth mechanism based on aggregation of nuclei has been identified to be the dominant mechanism at the circum-neutral pH solution values used in this study.

Other mineral substrates currently under study include the edge-faces of phlogopite, a mica mineral, and olivine. These *in situ* and *ex situ* observations yield important quantitative parameters readily usable in reactive transport models of nucleation at the reservoir scale.

[1] Jun, Y.-S., Lee, B., Waychunas, G. A. (2010) Environmental Science & Technology **44**, 8182-8189