

Innovative microfluidic experiments for investigating the co-precipitation of metals and radionuclides with carbonate phases

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In the context of long-term safety assessments of deep geological repositories for radioactive wastes, a rigorous understanding of the immobilisation of radionuclides such as ^{226}Ra due to co-precipitation with carbonate and sulphate minerals is important for a realistic prediction of radionuclide migration behaviour in the repository near and far field [1]. The co-precipitation of ^{226}Ra in sulphate minerals, in particular barite, has been studied experimentally and numerically in detail throughout the last decade to establish thermodynamic properties and mixing behaviour of its solid solutions over a wide range of temperatures (e.g. [2-7]). However, so far few studies were dedicated to the incorporation of ^{226}Ra into carbonates, and little is known about the mixing behaviour of ^{226}Ra and calcium carbonate phases like calcite, aragonite or vaterite. The aim of this work was to develop and explore innovative microfluidic experiments in combination with 4D in-situ Raman spectroscopy/tomography. This approach enabled the investigation of co-precipitation processes of radionuclides with carbonate minerals, using, as a first step, stable Ba as chemical analogue for ^{226}Ra , due to their similar ionic radii. Different microfluidic set-ups were developed to address co-precipitation in bulk solution as well as in confinement or under diffusive flow regimes. It turned out that high amounts of Ba can be incorporated into the lattice of calcite when formed via an amorphous precursor phase, suggesting that the formation of calcium carbonates can contribute to the retention of ^{226}Ra in the repository environment. This effect is also indicated by recent experiments addressing ^{226}Ra sorption on a carbonate rich subsurface of the Opalinus Clay from the Mont Terri rock laboratory, Switzerland [8].

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