Application of layered double hydroxides for ⁹⁹Tc immobilization.

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⁹⁹Tc is a long-lived ($T_{1/2}=2.13\cdot10^5$ y) fission product (FP) of the nuclear fuel cycle (NFC). As a component of nuclear wastes it remains a FP of concern for the final disposal option in case of uncontrolled FP release. Stabilized as Tc(VII)O₄⁻, it is well soluble, poorly sorbed and hence, highly mobile anion in the environment. A number of strategies have been proposed to reduce ⁹⁹Tc mobility by incorporating it into the structure of mineral phases ubiquitous and durable in the given environment. We have recently demonstrated the incorporation of TcO₄⁻ in layered double hydroxides (LDH) that were earlier proposed for ⁷⁹Se and ¹²⁹I remediation in water treatment technology.

This work is focused on the 99Tc(VII) incorporation into Pyroaurite (PyA) and Hydrotalcite (HTC) - Mg-Fe and Mg-Al LDHs correspondingly, on simplified CO2- and O2-free conditions. Earlier experiments with Re(VII), considered to be a chemical analogue for 99Tc(VII), have demonstrated little to no uptake of Re(VII). Similar experiments with ⁹⁹Tc(VII) have shown a significant increase in ⁹⁹Tc uptake on LDHs even in the concurrency with CO_3^2 ions. Following leaching experiments have revealed the irreversible character of 99Tc(VII)-LDH interaction. Modern spectroscopic methods are applied in order to understand how 99Tc is accommodated in between the brucite layers of LDHs and whether ⁹⁹Tc(VII) undergoes redox transformation to Tc(IV). This information will help to verify, whether LDHs can be used as a host phase for ⁹⁹Tc long-term and safe storage. Additionally, these data aquired on controlled conditions can be used for modelling of ⁹⁹Tc geochemical behavior in more complex repository relevant systems.

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