^{99}Tc retention on Fe(II)Al(III)-Cl layered double hydroxides

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To assess the safety of nuclear waste repositories, possible incidents have to be considered like canister corrosion and as a consequence the release of radionuclides. Among them, the fission product ^{99}Tc is of high concern due to its long half-life ($2.13 \cdot 10^5$ years) and the high mobility of the $Tc(VII)O_4^-$ oxoanion that is barely adsorbed by common mineral phases. However, Tc migration decreases under reducing conditions due to formation of Tc(IV), whose main species is a highly insoluble solid TcO_2.Under the reducing and corrosive conditions in the near-field of the repository, Fe^{2+} will act as a reducing agent for redox sensitive radionuclides (when present in the groundwater or sorbed on mineral surfaces). Furthermore, secondary mineral phases like Fe(II)-Al(III)-Cl, a layered double hydroxide (LDH), can be formed when Fe^{2+} interacts with Al_2O_3 at circumneutral-alkaline pH [1]. LDH phases are so-called anionic clays and they are known to retain pollutants by anion exchange, incorporation, surface complexation and in the case of Fe(II)-Al(III)-Cl via reduction promoted by the structural Fe^{2+} [2].

We have analysed the ^{99}Tc uptake by Fe(II)-Al(III)-Cl LDH under varying pH (4 to 11), ionic strength (0 to 0.1 M) and Tc concentration (10^{-9} to 10^{-3} M). At pH < 6.5, the solid to liquid distribution coefficient, (log K_d in mL/g), ranges from (2 to 6) and increases with decreasing ionic strength and increasing pH. At pH > 6.5, log K_d (6.5±0.3) are independent of pH and ionic strength. Tc K-edgeX-ay absorption spectroscopy showed in all cases a reduction to Tc(IV) and enabled us to elucidate the surface bound speciation of Tc on a molecular level.

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