

Retention mechanisms for safety relevant radionuclides in cementitious materials

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Cementitious materials are widely used in nuclear waste management, for example in waste solidification and stabilization, as construction materials, as backfill in (geological) disposal facilities and in certain waste containers. The migration of radionuclides in cementitious materials is controlled by solubility phenomena, diffusion, interface processes and/or incorporation into solid phases and formation of solid solutions. Within the frame of the EC-funded Horizon 2020 research project CEBAMA (www.cebama.eu), we are studying the uptake of selected long-lived fission and decay products relevant to the safety case for deep geological disposal, such as ¹²⁹I, ⁹⁹Tc and ²²⁶Ra, in a variety of cementitious systems. Within this context, a bottom-up approach is being pursued by investigating radionuclide interactions with synthetic hydrated cement phases, such as calcium silicate hydrates (CSH), various calcium aluminates (i.e. AFm and AFt), as well as hardened cement pastes with different compositions (e.g. CEM I and CEM V).

This presentation focusses on the uptake of ²²⁶Ra, ⁹⁹Tc, iodine, and molybdenum by CSH, AFm/t and C₃AH₆, respectively. Sorption/uptake kinetics were studied in static batch experiments under anoxic conditions. Structural investigations on the cementitious materials after radionuclide uptake were performed using SEM/EDX, Raman, XRD and TEM. The experiments indicate very strong retention of ²²⁶Ra by CSH phases, dependent to a minor extent on the Ca/Si-ratio. In contrast, the uptake of pertechnetate by the various model phases was negligible. Structural incorporation of iodide (I⁻) was observed for AFm-SO₄. On the other hand, uptake of iodate (IO₃⁻) by AFt as well as by AFm-SO₄, led to the formation of an iodate-substituted ettringite, formed either by anion exchange or phase transformation. SEM/EDX measurements indicated that the strong uptake of molybdate in systems with hydrogarnet was due to the formation of a molybdenum-containing AFm-like phase.