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Influence of the alpha radiation on the UO2 dissolution at high pH cementitious water.

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Since 2004, the supercontainer design has been selected by the Belgian authority for the management of the nuclear waste (NIRAS/ONDRAF) as the preferred new reference design for disposal of High Level Waste and Spent Fuel. This design is based on a buffer and backfill materials of Ordinary Portland Cement (OPC).
In this supercontainer, the spent fuel will be surrounded by a 30 mm thick carbon steel overpack and a 540 mm thick OPC concrete buffer. The supercontainer will contribute to the containment of radionuclides within the waste matrix but will also have an effect on the retardation of radionuclide release from the waste and will retard the migration of radionuclides. Because the supercontainer design (alkaline cementitious environment) differs significantly from designs for HLW and spent fuel used in other national programs, a specific evaluation of the behaviour of spent fuel in such high-pH conditions has been started at SCK•CEN in collaboration with NIRAS/ONDRAF. An experimental programme has been initiated to study the behaviour of UO2 as an analogue of real spent fuel in the cementitious environment of the supercontainer design, more specifically to determine the UO2 stability (dissolution rate), the UO2 solubility and the influence of alpha radiation on the behaviour of the UO2.

The experiments are carried out at 25-30°C in glove box under anoxic conditions in static (SA/V ratio of 6, 17 or 257 m-1) or dynamic set-up at pH of 13.5, 12.5 or 11.7, representative of the ageing steps of cement. The influence of the alpha radiation is investigated using Pu doped UO2. The activity of the doped UO2 batches is in the range 244 to 1.4 MBq/g of UO2, representative of the ageing of the spent fuel from 150 years to 89000 years after discharge. The results are compared with those obtained with depleted UO2, which simulates very old spent fuel. In an additional series of experiments, a H2 overpressure was imposed (0.5 or 5 bar).

The experiments in hyperalkaline conditions confirm the behaviour generally observed with UO2 at neutral pH. In most tests, the uranium concentration is close to 10-8 mol.L-1 , which could correspond to thermodynamic equilibrium. The experiments with Pu doped UO2 indicate that the dissolution rate and the solubility of UO2 increases with increasing alpha activity. Due to the radiolysis of water, the produced oxidizing species, oxidise the U(IV) to U(VI), leading to an increase of the uranium concentration. In the tests at high SA/V, equilibrium with U(VI) phases may be reached. Based on the results, we estimated a dissolution rate from 0.01 µg.m-2.d-1 for the least active (depleted) UO2 to 40 µg.m-2.d-1 for to the most active doped UO2. Imposing a H2 overpressure had no significant effect on the UO2 behaviour, probably because the used UO2 did not contain epsilon particles, which are known to catalyse the H2 reduction processes on the UO2 surface.

Primary author: Dr MENNECART, Thierry (SCK-CEN)

Co-authors: Dr CACHOIR, Christelle (SCK-CEN); LEMMENS, Karel (SCK-CEN)

Presenter: Dr MENNECART, Thierry (SCK-CEN)

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