

## 5 - INTERACTION OF CEMENTITIOUS MATERIALS WITH HIGH-LEVEL WASTE

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### Abstract

Since a few years, the Belgian agency for radioactive waste (ONDRAF/NIRAS) has selected the *Supercontainer design* with an Ordinary Portland Cement (OPC) buffer as the reference design for geological disposal of High-Level Waste (HLW) and Spent Fuel (SF) in the Boom Clay formation [Bel 2006, Van G. 2009]. The Boom Clay beneath the Mol-Dessel nuclear zone is a reference methodological site for supporting R&D. Compared to the previous bentonite based reference design, described in detail in the final SAFIR 2 report [NEA 2003], the supercontainer will provide a highly alkaline chemical environment allowing the passivation of the surface of the overpack and the inhibition of its corrosion. The *Supercontainer* will contribute to the containment of radionuclides, but it will also have an effect on the retardation of radionuclide release from the waste and it will retard the migration of the released radionuclides.

In the *Supercontainer design*, the canisters of HLW or SF will be enclosed by a 30 mm thick carbon steel overpack and a concrete buffer about 700 mm thick. The overpack will prevent contact with the (cementitious) pore water during the thermal phase. On the other hand, once the overpack will be locally perforated, the high pH of the incoming water may have an impact on the lifetime of the vitrified waste or spent fuel. The behaviour of these waste forms in disposal conditions has been studied for several decades, but the vast majority of published data is related to the interaction with backfill or host rock materials at near-neutral pH. Very few studies have been reported for alkaline media, at pH >11. Hence, a research programme including new experiments, was started at the Belgian Nuclear Research Centre (SCK•CEN) and at INE (FZK) to assess the rate at which the radionuclides are released by the vitrified waste and spent fuel in such an environment.

The presence of concrete will have an impact on the behaviour of the vitrified HLW and spent fuel. For *vitrified HLW*, the high pH is expected to cause a faster dissolution of the glass matrix, driven by secondary phase formation. Low long-term dissolution rates, such as expected in neutral pH conditions, will probably be reached only after the formation of a sufficiently thick alteration layer separating the pristine glass from the bulk of the concrete, thus causing a pH gradient. So far, this process cannot yet be quantified well enough. For *spent fuel*, the preliminary results are not entirely consistent. The tests with spent fuel suggest that the high pH is not detrimental for the fuel stability. The tests with (doped) UO<sub>2</sub> suggest, however, that it may take more time to reach the threshold fuel activity where radiolytical oxidation becomes unimportant. Moreover, there are indications that the non-oxidative fuel dissolution could be accelerated at high pH by colloid formation in certain conditions. The factors affecting this colloid formation are not understood so far. Therefore it is not yet possible to assess whether this process will be relevant under in situ conditions.

The uncertainties about the present dissolution rates are still large due to the small number of data. A continuation of the programme is necessary to improve the rate estimations and to better understand the underlying mechanisms. For vitrified waste, the focus will be on the evolution at the glass/concrete interface (secondary phase formation, pH evolution...). For spent fuel, it is important to understand better the nature of the dissolution process (oxidative or non-oxidative) as a function of the fuel activity, the role of colloids, and the differences between the behaviour of alpha-doped  $UO_2$  and real spent fuel. Tests with spent fuel pellets and hydrogen gas will be started at INE to see if hydrogen reinforces the reducing capacity enough to suppress the oxidative fuel dissolution when the alpha activity of the fuel is still high. Another important remaining uncertainty is the potential effect of concrete phases close to the fuel. This may cause sorption or precipitation of U(IV) and result in accelerated fuel dissolution. Although we expect that this process will have only a temporary effect, it needs to be quantified better. It is likely that this process will drive the slow long term fuel dissolution.

This work is performed as part of the programme of ONDRAF/NIRAS (the Belgian Agency for Radioactive Waste and Fissile Materials) on the geological disposal of high-level/long-lived radioactive waste.

### References

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