INVESTIGATIONS ON THE LONG-TERM BEHAVIOUR OF HIGH LEVEL WASTE FORMS

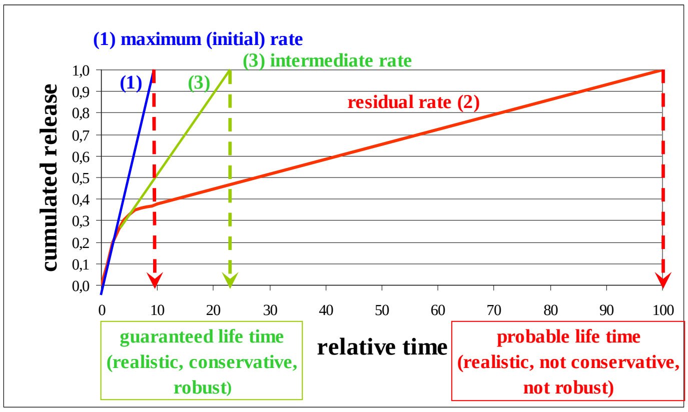
Background

The Belgian Nuclear Research Centre (SCK•CEN) has a long-standing expertise in research concerning the compatibility of waste forms with the final disposal environment, in collaboration with NIRAS/ONDRAS. For high level waste, most attention goes to two waste forms that are relevant for Belgium, namely (1) vitrified HLW (High Level Waste) from the reprocessing of spent fuel, and (2) spent fuel as such, referring to the direct disposal scenario. The expertise lies especially in the study of the chemical interactions between the waste forms and the disposal environment. This is done by laboratory experiments, supported by modeling.
Until 2004, the reference disposal design for HLW glass and spent fuel in Belgium was based on the use of a bentonite buffer. The experiments performed in that period therefore involved mostly the study of the influence of clay on the waste form behaviour. Since 2004 the Supercontainer design with Ordinary Portland Cement as buffer material (without bentonite) has been selected as the reference. The experiments related to this new design are therefore predominant now. Clay based disposal designs are still the reference in several other European countries. For this reason, the study of clay-waste interactions was not completely abandoned in the period 2004-2008, but continued in the framework of EC programmes.
The first experiments focused on the Supercontainer design were started in 2006 (HLW) and 2007 (spent fuel). The first results are available now for HLW glass. Most results generated recently are, however, still related to the bentonite concept.

Objectives

- To evaluate the minimum guaranteed durability of the waste form, which will be used as input in the safety assessment. The objective is not to obtain an absolute value for the durability or an interval of values, which will always be subject to caution, but rather to determine a lower limit for the life time of the waste form, which is conservative, realistic and robust, and to describe the related uncertainties.
- To give convincing qualitative or semi-qualitative arguments showing that the waste durability will most probably be larger than the proposed minimum life time, because a much lower 'residual rate' will be reached.

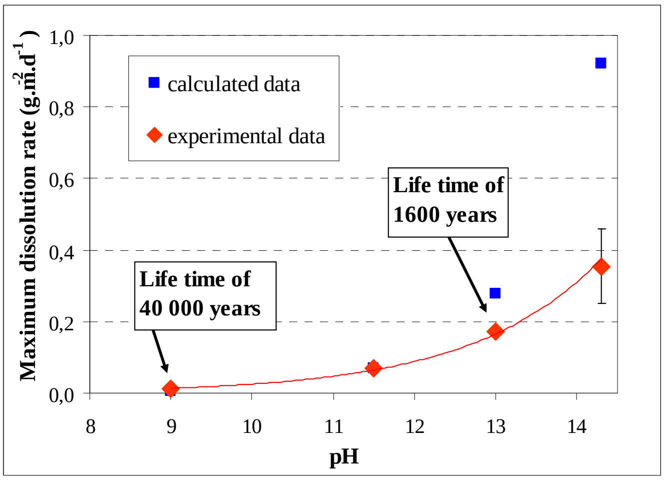
The difference between the maximum initial release rate and the much lower long term residual rate is illustrated in the graph on the right, showing the estimation of the guaranteed life time of the waste based on a maximum, conservative dissolution rate (1), and comparison with the more likely, but less conservative and less robust life time estimation, based on the residual rate (2). With sufficient argumentation, it is possible to propose an intermediate rate (3), which is still conservative and robust, but closer to the probable longer life time.



Principal results

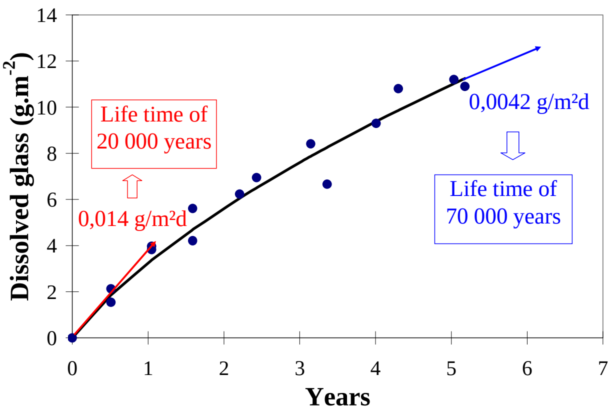
The experimental programme consists of a large number of experiments. Here, we only illustrate this with a few examples.

The maximum, initial dissolution rate in Supercontainer conditions (high pH) has been determined for several HLW glasses. An example is given for HLW glass SON68 in the graph on the right. We see that the maximum glass dissolution rate (gram.m^{-2} day^{-1}) increases at high pH, but not as much as predicted, based on calculated extrapolations of literature data. The resulting dissolution rates are nevertheless high, and correspond to a glass life time of only some thousands of years.

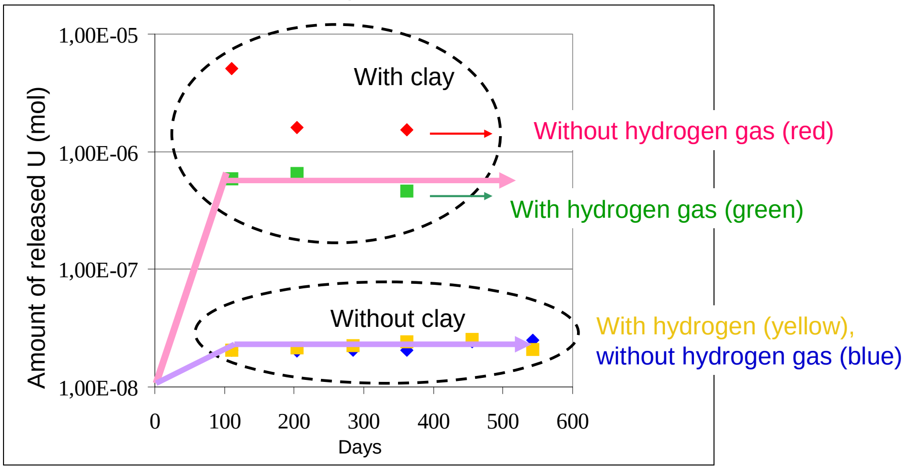


The much lower long-term rate is being determined in tests of long duration. These tests are done in realistic conditions, using percolation cells (left figure), which are filled with a layer of compact wet clay and a layer of glass, and placed in a furnace. At the end, the cells are opened and the glass and clay plug are pushed out. These tests allow monitoring of the evolution of the water composition and the amount of dissolved glass. Tests with a similar set-up have been running for 5 years at 30°C so far, for HLW glass SON68. The resulting glass dissolution is shown in the figure hereunder.

We clearly see the expected gradual decrease of the dissolution rate, from 0.014 to 0.0042 $gram.m^{-2}day^{-1}$. Such dissolution rate would give a HLW glass life time of several tens of thousands of years. The dissolution rate will probably continue to decrease in these tests, so the rate of 0.0042 $g.m^{-2}d^{-1}$ is conservative. It is still an order of magnitude higher than the long term rate of 0.00013 $gram.m^{-2}.day^{-1}$ observed in tests of more than 12 years in pure water at 90°C. In that case, the HLW glass life time will be several hundreds of thousands of years.



The presence of hydrogen gas formed by anaerobic corrosion of the metallic overpack materials in the engineered barriers may suppress the dissolution rate of spent fuel. To study this, tests were performed with UO_2, doped with alpha emitters, in the autoclaves shown in the left picture. These tests were carried out in the presence of clay and hydrogen gas at room temperature.
The results (figure hereunder) show that the addition of clay causes an increase of the initial uranium release, and that hydrogen gas has a mitigating effect. In absence of clay, hydrogen is not effective to reduce the UO_2 dissolution at room temperature.



Future work

The long term tests on HLW glass in clay conditions will be continued, to decrease further the uncertainty on the long term dissolution rate. Most attention will go, however, to the continuation of the experiments with glass and spent fuel for the Supercontainer disposal design.

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Main reference

Karel Lemmens, Christelle Cachoir, Elie Valcke, Karine Ferrand, Marc Aertsens, Thierry Mennecart, "The strategy of the Belgian Nuclear Research Centre in the area of High-level waste form compatibility research", Proceedings of ICEM2007, September 2-6, 2007, Oud Sint-Jan Hospital Conference Center, Bruges, Belgium.