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Status of dissolution based fast/instant radionuclide release studies

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Introduction

This report summarizes the progress made in Work Package 3 of FIRST-Nuclides in the third year of the project. The laboratories that participate to WP3 are the Belgian Nuclear Research Centre (SCK•CEN), the Paul Scherrer Institute (PSI), the Karlsuher Institut für Technologie (KIT), Studsvik Nuclear AB (STUDSVIK), the Joint Research Centre – Institute for Transuranium elements (JRC-ITU), the Fundacio CTM Centre Tecnologic (CTM), and the Hungarian Centre for Energy Research (EK).

The major objective of WP3 is the quantification of the fast release of radionuclides by means of leach tests with spent nuclear fuel. Such leach tests are performed by SCK•CEN, PSI, KIT, STUDSVIK, ITU and CTM. The experiments are done with PWR fuels having a burnup in the range of 50 to 70 $MWd.kg_{HM}^{-1}$, with BWR fuels of 42-59 $MWd.kg_{HM}^{-1}$, and a MOX fuel of 63 $MWd.kg_{HM}^{-1}$ (average burnups). As a complement to the leach tests performed on fuel samples under controlled laboratory conditions, the leaching behaviour of damaged and leaking VVER fuels is studied by EK. An additional aim in WP3 was to determine the chemical speciation of Se in spent BWR UO_2 fuel by X-ray spectroscopic methods. This work was carried out by PSI in collaboration with STUDSVIK and KIT-INE.

Status after 33 months

The first year (2012) had been used to define the detailed experimental matrix and to prepare the leach tests and analytical methods. The sample preparation methods, leachant composition and analytical methods had been discussed between the participating institutes, to come to an optimal program in which the various contributions give complementary information, produced in conditions that are sufficiently harmonized to allow intercomparison. The only laboratory that had already started some of the planned leach tests in 2012 was STUDSVIK .

In the second year (2013), the analytical methods were further developed, the preparations of the leach tests have been continued, most planned experiments have been started, and the first results have become available.

In the fourth year (2014), the experiments were continued, with samplings for test durations up to one year in some cases, and the results were interpreted.

SCK.CEN has continued the leach experiments on a spent PWR UOX fuel with an average burnup of 50.5 GWD/tHM. They have tested in parallel the leaching of cladded fuel segments and opened fuel segments where the cladding and fuel fragments are separated from each other but leached together. The tests were performed with the standard leachant in glass columns under air atmosphere (i.e. under oxidizing conditions), identical to the columns used by PSI. After two complete leachate renewals in the first days, samples of the leachate were taken at regular time intervals, without renewal of the solution, i.e. in static mode. The IRF of iodine and caesium isotopes was determined and reported for test durations up to 357 days. Apart from these, many other isotopes were analysed, a.o. ^{14}C, ^{99}Tc and ^{238}U. The results are presented in ref [1].

PSI has continued the leach experiments on a spent BWR UOX fuel with an average burnup of 57.5 GWD/tHM, spent PWR UOX fuel with an average burnup of 56.5 GWD/tHM, and spent MOX fuel with an average burnup of 63 GWD/tHM. They have tested in parallel the leaching of cladded fuel segments, the leaching of fuel fragments, and the leaching of separated claddings with some adhering fuel residues. The tests were performed with the standard leachant in glass columns under air atmosphere (i.e. under oxidizing conditions). Samples of the leachate were taken at regular time intervals, without renewal of the solution, i.e. in static mode. The IRF of ^{129}I and ^{137}Cs was determined and reported for test durations up to 182 days. The results of X-ray spectroscopic analyses (micro-XRF and micro-XANES), performed on samples of BWR fuel in 2013 and 2014 to determine the selenium distribution on the microscale, its oxidation state and its structural environment (next- neighbour distances and coordination numbers), were further interpreted and published. The results, partly obtained in collaboration with Studsvik are presented in refs. [2, 3, 4, 5].

JRC-ITU has continued leach experiments on a spent BWR UOX fuel with an average burnup of 42.22 GWD/tHM. They have tested in parallel the leaching of cladded fuel segments and fuel powders that were taken separately from the core zone and the rim zone of the fuel. The tests were performed with the standard leachant in glass test tubes under air atmosphere (i.e. under oxidizing conditions). At each sampling, the leachate was completely renewed, so the tests were done in pseudo-dynamic mode. The IRF of Cs, Tc, Sr, Rb, Mo and U isotopes was determined and reported for test durations up to 190 days.

CTM has continued leach experiments on a spent BWR UOX fuel with an average burnup of 45 GWD/tHM. They have tested in parallel the leaching of cladded fuel segments and fuel powders that were taken separately from the core zone and the rim zone of the fuel. The tests were performed with the standard leachant in glass test tubes under air atmosphere (i.e. under oxidizing conditions). At each sampling, the leachate was completely renewed, so the tests were done in pseudo-dynamic mode. The IRF of Cs, Tc, Sr, Rb, Mo and U isotopes was determined and reported for test durations up to 190 days. The experiments of CTM were performed in the laboratories of JRC-ITU.

The results obtained by JRC-ITU and CTM are presented in refs. [6,7,8].

KIT has performed leach experiments on a spent PWR UOX fuel with average burnup of 50.4 GWDtHM. They have tested in parallel the leaching of cladded fuel segments and fuel fragments. The tests were performed with the standard leachant in autoclaves under argon/hydrogen gas atmosphere with a pH_2 of 3 bar (i.e. under reducing conditions). After one day of preleaching, samples of the leachate and the gas phase were taken at regular time intervals, without renewal of the solution, i.e. in static mode. The IRF of relevant radionuclides such as I, Cs, Tc, Sr and U isotopes and fission gases (Xe + Kr)was determined and reported for test durations up to 245 days. The results are presented in ref. [9].

Studsvik has started the last planned tests an continued leach experiments on samples of six types of a spent fuel, i.e.

- BWR UOX fuels with an average burnup of 50.2 GWD/tHM (test on cladded segment), 54.8 MWd/THM (test on cladded segment), and 57.1 MWd/tHM (test on fragments + separated cladding)
- An Al/Cr doped BWR UOX fuel with a burnup of 59.1 MWd/tHM (test on fragments + separated cladding)
- A Gd doped PWR UOX fuel with an average burnup of 54.4 MWd/tHM (test on fragments + separated cladding)
- A spent PWR UOX fuel with an average burnup of 70.2 MWd/tHM (test on fuel powder)

The tests on cladded fuel segments and fuel fragments + separated cladding were performed with a slightly modified standard leachant (10 mM NaCl and 2 mM $NaHCO_3$) in glass test tubes under air atmosphere (i.e. under oxidizing conditions). At each sampling, the leachate was completely renewed, so the tests were done in pseudo-dynamic mode. The IRF of I, Cs, Tc, Sr, Rb, Mo, Se, U and many other isotopes was determined and reported for test durations up to 364 days. The methods for measurement carbon-14 in the leaching solutions have been tested. The tests on fuel powder were performed using a simultaneous grinding and leaching method. The results are presented in refs. [10, 11, 12, 13, 14].

EK has further disseminated the results obtained in the first reporting period, regarding the dissolution rates of different isotopes from damaged and leaking VVER fuel stored in water for several years. The results are presented in refs. [15,16].

Further planning

The project stops on 31-12-2014. An overview of the work performed in Work Package 3 of FIRST-Nuclides with a global interpretation of the results will be published in a scientific journal.

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