FIRST-NUCLIDES: RESULTS ON RADIONUCLIDE RELEASE FROM USED FUEL

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The Collaborative Project (CP) FIRST-Nuclides ("Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel") is aimed at providing new and comprehensive knowledge of the fast release of safety relevant radionuclides from used high burn-up light water reactors (LWR) UO_2 fuel. The results cover the dissolution based instant release fraction (IRF) from PWR and BWR fuels with enrichments between 3.3 to 4.9 % ^{235}U, burn-up between 50 and 70 GWd/t_{HM}, power rates from 160 to 330 W/cm and fission gas releases (FGR) of 1 to 14 %. The experiments comprise clad and unclad pellets, segments of pellets, fragments and powders. The experiments were performed using the same bicarbonate groundwater composition.

I. INTRODUCTION

The fast / instant release of radionuclides from spent (used) nuclear fuels (SNF) was investigated in a series of European projects (such as SFS [1, 2], NF-Pro [3], MICADO [4]). Also several French research programs investigated and quantified the rapid release [5-7]. However, some important questions remained open which were elaborated within the Collaborative Project (CP) "Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (FIRST-Nuclides). This CP dealt with the understanding of the fast release of radionuclides from high burn-up UO_2 (SNF) in deep geological repositories. An outline of FIRST-Nuclides was presented previously [8]. The SNF waste type represents one of the sources for the release of radionuclides after loss of integrity of the disposed canisters. An assessment of the time-dependent radionuclide release is required for safety analyses. The fraction released first consists of radionuclides (1) in gaseous form, and (2) those showing a high solubility in groundwater. For this reason, the CP FIRST-Nuclides aimed at determining the "instant release fraction (IRF) values of the two most important long-lived fast leaching nuclides iodine and caesium, and of carbon and selenium, whose IRF contribution is still largely unknown. The elements I, C and Se tend to form anionic species. Such anions are hardly chemically retained in any repository barrier system.

The CP started January 1, 2012 and ended on December 31, 2014. During this period, a close cooperation was developed between the partners and the Associated Groups. The project was implemented by a Consortium with ten beneficiaries (Karlsruhe Institute for Technology (KIT) Germany, Amphos 21 Consulting S.L. (AMPHOS21) Spain, Joint Research Centre – Institute for Transuranium Elements (JRC-ITU) European Commission, Forschungszentrum Juelich GmbH (JÜLICH) Germany, Paul Scherrer Institut (PSI) Switzerland, Belgian Nuclear Research Centre- Centre d'Etude de l'Energie Nucleaire (SCK•CEN) Belgium, Centre National de la Recherche Scientifique (CNRS) France, Fundació Centre Tecnològic (CTM) Spain, Magyar Tudományos Akadémia Energiatudományi Kutatóközpont (MTA EK) Hungary, Studsvik Nuclear AB (STUDSVIK) Sweden). Thirteen organisations have contributed to the project without any funding as *Associated Groups (AG*), i.e. organisations from France (Commissariat à l'énergie atomique et aux énergies alternatives, CEA), United States of America (Los Alamos Natonal Laboratory, SANDIA National Laboratories), United Kingdom (Nuclear Decommissioning Authority (NDA), National Nuclear Laboratory (NNL), University Cambridge, Center for Nuclear Engineering of the Imperial College London, University Lancaster), Finland (Posiva Oy, Teollisuuden Voima (TVO)), Czech Republic (ÚJV Řež, a. s.), Spain (Ciemat) and Germany (Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH). These groups had particular interest in exchange of information. A group of six implementation and regulatory oriented organizations (SKB (Sweden), NAGRA (Switzerland), ONDRAF/NIRAS (Belgium), ANDRA (France), BfS (Germany), and ENRESA (Spain)) have participated as *"End-User Group (EUG)"*. This group ensured that end-user interests (waste management organisations and one also regulator) were reflected in the project work, and

they reviewed the project work and the scientific-technical outcome.

The CP was organized in six workpackages (WP): WP1, "Samples and tools" dealt with: the selection, characterization and preparation of the materials to be studied and the set-up of experimental and organisational tools. One of the essential requirements of the project was that typical and sufficiently well characterized SNFs were used for the experiments and modelling studies. WP2 covered the "Gas release and rim and grain boundary diffusion experiments" and WP3 addressed "Dissolution based release studies". This WP included a quantification of released radionuclides: fission gases (Kr and Xe), and fission products as ^{135}Cs, ^{129}I, ^{14}C, ^{79}Se, ^{99}Tc and some isotopes, and the characterization of selenium in the spent fuel. WP4 "Modelling" dealt with modelling of release/retention processes of fission products in the SNF structure. WP5 "Knowledge, reporting and training" was responsible for the management of the knowledge generated within the project which included the state-of-the-art report, the general reporting, up-to date documentation and the organization of training. The management of the Collaborative Project was included in WP 6.

The current contribution presents a selection of experimental results from the leaching experiments of WP3, where the fast/instant radionuclide release was determined by contacting irradiated fuel samples with a standardized bicarbonate ground water under various conditions.

II. MATERIALS AND METHODS

In the last years, the discharge burn-up of nuclear fuel has increased in all types of nuclear power plants. This increase is a consequence of the cost reduction associated with nuclear power by the utilities . As an example, the increase of the initial enrichment of the UO_2 fuel and the related peak burnups[1] are shown in Fig. 1 for the German nuclear power plants.

To achieve burnups above 50 GWd/t_{HM}, higher initial enrichments are required reaching up to almost 5 wt.% ^{235}U. Such fuels need a compensation of the excess reactivity by the burnable neutron poison gadolinium. Minimization of the potential fission gas release is anticipated by reduction of the power rates. Accordingly, the high burnup SNF investigated in FIRST-Nuclides addressed these modern fuels, which differed significantly from materials used in previous investigations. Compared to previous investigations, more data of in-pile operation parameters have been collected, which may be correlated to the IRF data (see TABLE I).

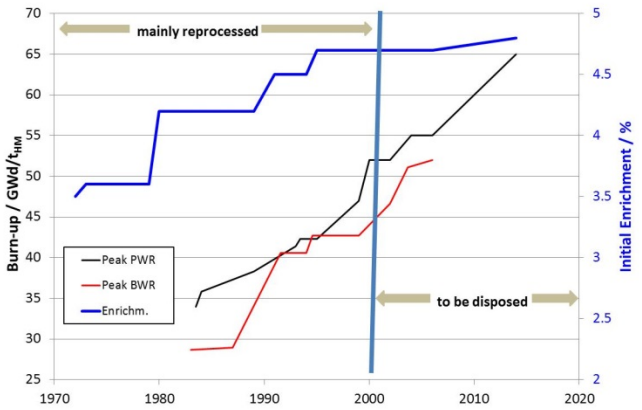

Fig. 1. Evolution of initial enrichment and the peak burn-up of German power plants. Data taken from [9, 10].

TABLE I. Overview of the fuels investigated within CP FIRST-Nuclides.

		PWR	**BWR**	**THTR/ VVER**
Discharge		1989 - 2008	2005 – 2008	
Producer		Areva	Areva/ Westing-house	
Cladding	Material	Zry-4 – M5	Zyr 2	Graphite Zr1%Nb
	∅ / mm	9.50 - 10.75	9.84 - 10.2	
	Thickness / mm	0.62 - 0.73		
Pellet	Enrich-ment / %	3.80 – 4.94	3.30 - 4.25	2.4 - 16.8%
	Grain size/ µm	5-40	6 - 25	20 -80
	Density g cm^{-3}	10.41	10.52	10.8
	Specifics	standard; NIKUSI production	standard; Al/Cr addition	
Irradiation	Burn-up / GWd/tU	50.4 – 70.2	48.3 – 57.5	10 - 107
	Cycles	2 - 14	5 – 7	
Lin. Power	average W/cm	186 -330	160	130–228
Fission Gas Rel.	%	4.9 – 23	1.2–3.1	

[1] PWR fuel assemblies have similar axial-burnup shapes relatively flat in the axial mid-section (peak burnup from 1.1 to 1.2 times the assembly average burnup) and under-burned fuel at the ends of the rods (burnup of 50 to 60% of the assembly average).

The radionuclides that are susceptible to fast release are segregated in different regions within the fuel rods, i.e. in the gap between the fuel and the cladding, in fuel fractures, in grain boundaries and on the cladding. Hence, the release to be expected in the leaching tests depends on the position in the fuel rod and the physical preparation of the fuel samples. The most complete information could be obtained by exposing different fuel regions to the leachant. Therefore, tests with following types of samples were performed:

- clad fuel segments with a length of up to 2.5 cm,
- fuel fragments without zircaloy cladding,
- fuel powder obtained by milling and sieving of the fuel and targeted for maximal grain boundary exposure studies.

The radionuclide release in the leach tests depends on the leach test conditions, i.e. the composition of the leachant and the atmosphere. To reduce the number of variables and to allow inter-comparison of the results, a common leachant was defined, consisting of 19 mM NaCl + 1 mM $NaHCO_3$. Following tests were performed:

KIT has conducted leach experiments on a spent PWR UOX fuel with an average burn-up of 50.4 GWd/t_{HM}. The tests covered both clad fuel pellets and fuel fragments. The tests were performed with the standard leachant in autoclaves under Ar/H_2 gas overpressure (i.e. under reducing conditions). After one day of pre-leaching, samples of the leachate were taken at regular time intervals, without renewal of the solution, i.e. in the static mode. The IRF of relevant radionuclides such as I, Cs, Tc, Sr and U isotopes was determined for test durations up to 245 days. The autoclave set-up also allowed the measurement of the fission gas release during the leach tests.

JRC-ITU has leached spent BWR UOX fuel with an average burn-up of 42.22 GWd/t_{HM}. In parallel leach tests were performed using a slice of a clad fuel pellet and fuel powders prepared separately from the core and the rim zone of the fuel pellet. 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.

JUELICH has investigated the instant radionuclide release fraction from spent UO_2 TRISO coated particles. These particles were designed for use in a High Temperature Reactor (HTR) and had very high burnup (~100 GWd/t)

PSI investigated spent BWR UOX fuel with an average burn-up of 57.5 GWd/t_{HM}, a spent PWR UOX fuel with an average burn-up of 56.5 GWd/t_{HM}, and a spent MOX fuel with an average burn-up of 63 GWd/t_{HM}. Leach tests were performed with clad fuel segments, fuel fragments, and separated claddings with some adhering fuel residues in the standard leachant using glass columns under air atmosphere (i.e. under oxidizing conditions). After 7 days, the leachate was removed completely and then replaced by fresh leaching solution. Afterwards, samples of the leachate were taken at regular time intervals, without renewal of the solution, i.e. in static mode. The IRF of I and Cs isotopes was determined for test durations up to 182 days.

SCK·CEN has performed leach experiments on a spent PWR UOX fuel with an average burn-up of 50.5 GWd/t_{HM}. In parallel the leaching of clad fuel segments and opened fuel segments was carried out. The opened fuel segments consisted of both the fuel fragments and the separated cladding. 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 further renewal of the solution, i.e. in static mode. The IRF of I and Cs isotopes was determined and reported for test durations up to 357 days. Apart from these, many other isotopes were analyzed, such as ^{14}C, ^{99}Tc and ^{238}U.

CTM did leach experiments on a spent BWR UOX fuel with an average burn-up of 45 GWD/t_{HM}. They have tested in parallel the leaching of clad 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.

Studsvik has conducted leach experiments on samples of six different spent fuels, i.e.

- BWR UOX fuels with an average burn-up of 50.2 GWd/t_{HM} (test on clad segment), 54.8 GWd/t_{HM} (test on clad segment), and 57.1 GWd/t_{HM} (test on fragments + separated cladding).
- An Al/Cr doped BWR UOX fuel with a burn-up of 59.1 GWd/t_{HM} (test on fragments + separated cladding).
- A Gd doped PWR UOX fuel with an average burn-up of 54.4 GWd/t_{HM} (test on fragments + separated cladding).
- A spent PWR UOX fuel with an average burn-up of 70.2 GWd/t_{HM} (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 tests on fuel powder were performed using a simultaneous grinding and leaching method.

As a complement to the leach experiments made on fuel samples under controlled laboratory conditions, **MTA EK** has collected and interpreted the isotopic dissolution data of damaged and leaking VVER fuels that had been stored in storage pools in the period 2003-2007 (damaged fuel) and 2009-2010 (leaking fuel). The damaged fuels had an average burnup in the range 10.1-26.7 GWd/t_{HM}. The leaking fuel had an average burnup of 14 GWd/tHM. The leachant was in this case the water of the storage pool. In the case of the damaged fuel, the pH of the water was ≈7 in the first 14 days after the incident (15 g H_3BO_3/ kg H_2O) and ≈4-4.5 (21 g H_3BO_3/ kg H_2O) in the remaining period, with measurements up to 1368 days. In the case of the leaking fuel, measurements up to 369 days are available. During the storage, the water purification has led to periods of increasing and decreasing radionuclide concentrations. The dissolution rate was calculated in various ways for a series of isotopes, e.g. Cs, I and U isotopes.

Although the equipment and procedures were not entirely standardized, the use of the standard leachant and similar or complementary sample preparations have made the experimental program coherent.

III. RESULTS AND DISCUSSION

Table II summarizes the most relevant isotopes analyzed by the partners in the leaching experiments

The measurements of the FGR during the leach test by KIT have shown that FGR takes place also during the leaching of the fuel (Fig. 2). In order to evaluate how much of the inventory has been released through the experiment, the fraction of inventory released for an element *i* (*FIG_i*) is calculated following eq. 1:

$$Fraction\ release = \frac{m_i}{m_{UO_2} x H_i} \qquad \text{(eq. 1)}$$

with m_i, is the mass of element (g)*i* in the gas or liquid phase, m_{UO2} is the initial oxide mass (g) in the fuel sample and H_i corresponds to the fraction of inventory for the element *i* ($g_i \cdot g_{UO2}^{-1}$). The amount of fission gases released in during the leaching experiments appears to be significantly higher than the amount of fission gases released into the plenum during the fuel operation.

Fig. 2 also shows that release rate from the same SNF material is higher in the case of fragments in comparison to a cladded pellet

TABLE II. Analyzed Isotopes in the different labs.

	$^{133/134}Cs$	^{137}Cs	^{129}I	^{79}Se	^{99}Tc	^{90}Sr	^{238}U	others
KIT		x	x		x	x	x	FG
JRC-ITU		x			x	x	x	Rb, Mo
FZJ	x							
PSI		x	x					
SCK·CEN	x		x		x	x		Zr, Mo,Ru, Cd
CTM		x			x	x	x	Rb, Mo
MTA EK		x	x				x	
Studsvik	x			x			x	Mo

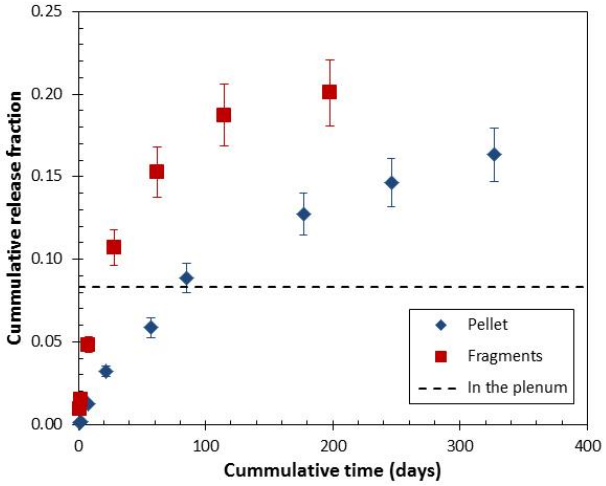

Fig. 2. Cumulative release fraction of FG (Xe+Kr) as function of time during leaching of SNF (50.4 GWd/t_{HM}) in comparison to the FGR into the plenum of the rod[11].

The IRF determined from differently prepared samples (clad segments, opened segments, fragments, powders…) are shown in Fig. 3 for caesium, and for iodine in Fig. 4. The release depends on the operation parameters, such as burnup and power rate, as well as on the nature of the exposed fuel structures (gap or grain boundaries), respectively. The release tends to increase in the order: fragments < clad pellet segments < opened clad pellets, in agreement with the exposed surface area and presence/absence of the gap inventory. The UO_2 fuel doped with Al/Cr appears to have a lower IRF, which is probably due to the fact that the larger grain size increases the distance over which Cs and I have to diffuse before

they reach the grain boundary. It was also observed that the IRF was higher for PWR fuel in comparison to BWR fuel even for similar burnup, probably because the linear power was higher in the PWR fuel.

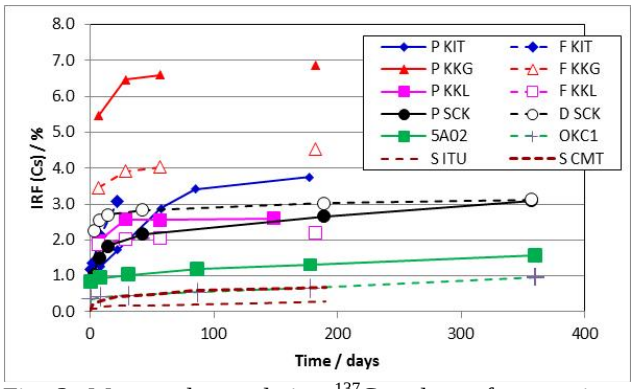

Fig. 3. Measured cumulative ^{137}Cs release from various SNF samples:

P KIT: Clad PWR pellet under reducing conditions
F KIT: PWR Fragment under reducing conditions
P SCK: Clad PWR segment, D SCK: Declad PWR segment (fragments + cladding)
P KKG: Pellet from KKG PWR (PSI)
P KKL: Pellet from BWR (PSI)
F KKG and F KKL: fragments from KKG PWR and KKL BWR (PSI)
5A02 and OKC1: BWR fragments together with cladding
S ITU and S CTM: 2.5-2.8 mm segments of BWR pellets
Uncertainty of ^{137}Cs measurements: ±10 %

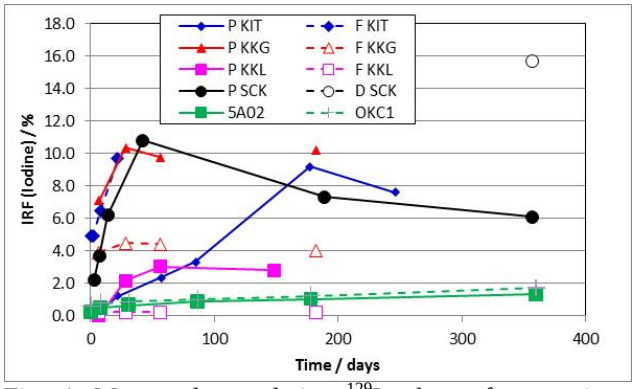

Fig. 4. Measured cumulative ^{129}I release from various SNF samples. (Nomenclature see Fig. 3). Uncertainty of ^{129}I measurements: ±20 %.

All leach tests were performed with the standard leachant. However, some tests were conducted in autoclaves under Ar/H_2 gas overpressure (i.e. under reducing conditions). The different release behavior of a series of elements is shown in Fig. 5. For this comparison the cumulative releases after a period of 150 to 180 days have been plotted.

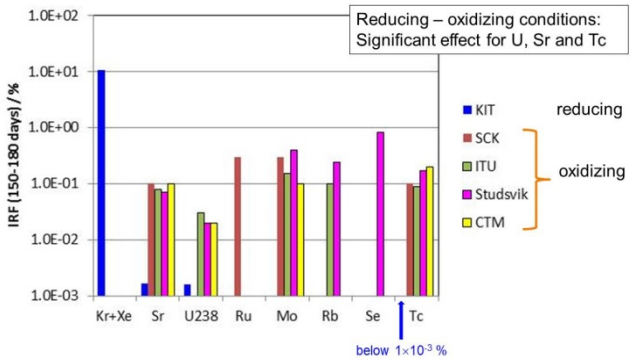

Fig. 5. Cumulative elemental releases from various SNF samples under oxidizing, anaerobic and reducing conditions. (KIT: reducing conditions).

Figs. 3 and 4 do not show differences for Cs and I release under reducing and oxidizing or anaerobic conditions. This is different for elements which are either redox-sensitive or which are incorporated in the UO_2 matrix such as strontium, uranium , technetium. For these elements, the cumulative releases after 150 to 180 days differ by almost two orders of magnitude. This observation is explained by the oxidation of the UO2 and consequent dissolution of U(VI) species at a relatively high rate [12, 13].

In almost all leach experiments, a residual dissolution rate of Cs isotopes was measured for the longest test durations. This means that the measured IRF for the longest duration (≤ 1 year) may not be the long-term maximum. The net Cs dissolution rates measured in the leach tests were all significantly higher than the expected long term UO_2 matrix dissolution rate, even for the tests in reducing conditions by KIT. Extrapolation, preferably supported by modeling, is necessary to estimate the time required for the complete release of IRF nuclides. Further interpretation via mechanistic modeling may be based on following detailed model, which describes the IRF from the main fuel structures (gap and macro fractures, accessible grain boundaries, and deeper, less accessible grain boundaries) as a function of time:

$$IRF_{total}(t) = IRF_{gap}(t) + IRF_{acc.\ GB}(t) + IRF_{deep\ GB}(t)$$

The required data can be obtained by the tests with differently prepared samples.

The long-term matrix dissolution rate, and the time it will take before the release of IRF radionuclides is controlled by this matrix dissolution, will depend on the redox conditions. In reducing conditions, the matrix dissolution rate will be much lower than in oxidizing conditions. Moreover, matrix oxidation tends to open

grain boundaries and thus causes the continuous exposure of fresh grain boundaries with high concentrations of soluble IRF nuclides.

Until recently, the IRF of spent fuel was often related to its burnup. With more data becoming available for high burnup fuel, there is growing evidence that the fission gas release (FGR) and IRF depends much more on the linear power rate of the fuel than on the burn-up. The data gathered in FIRST-Nuclides give further evidence for this. Hence, the linear power appears to be a more relevant operational parameter to predict the FGR than the burn-up is. This can be explained by the fact that the FGR depends on the temperature of the fuel pellet in the reactor during operation. This temperature depends on the linear power, rather than on the burnup.

However, the large amount of data has not yet been sufficiently evaluated. Following issues are under investigation and will be developed from the complete data sets:

- Comparison of Cs and I release for PWR and BWR fuel for high burn-up (> 55 GWd/t_{HM}).
- Determination of fission gas releases (FRG) for the fuels studied
- Correlation of IRF (Cs, I) with linear power rate.
- Correlation FGR with linear power rate. Preliminary evaluations suggest that linear power is more relevant operational parameter to predict FGR than the burn-up.
- Correlations of IRF versus Fission Gas release.
- IRF for radionuclides other than Cs and Iodine.

IV. CONCLUSIONS

Within the CP FIRST-Nuclides, dissolution based instant releases were determined for 26 different moderate to high burn-up SNF samples, investigating 5 elements or more for four up to ten time steps, each. It is obvious that not all data can be presented here. As expected from literature (e.g. [14, 15]) the release rates of Cs and I decreased after a period of a few months of leaching.

It was also demonstrated that elements which are either redox-sensitive or which are incorporated in the UO_2 matrix such as strontium, uranium, and technetium show different cumulative releases (after 150 to 180 days) by almost two orders of magnitude depending on the redox conditions of the experiments.

The End-Users stated that the subject of the CP “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (FIRST-Nuclide) is highly relevant for all waste management organisations involved in repository development for which direct disposal of spent nuclear fuel is foreseen. The Instant Release Fraction (IRF) contributes substantially to the peak release after container breaching and its potential radiological consequences. Specifically, the results include (1) data from experimental determination of rapid release fractions for moderate and high burn-up UO_2 fuels, including doped fuels, expected to be used much more by reactor operators in the future, and (2) a data base for release of Cs and I from high burnup fuel and comprehensive comparisons of IRF with fission gas release (FGR), which are necessary in order to be able to estimate IRF data for populations of fuel rods in various reactor operation conditions.

The partners of the project as well as Associated Groups and End-Users identified a series of open questions to be resolved in future investigations. The 3 years CP FIRST-Nuclides required huge investments to setup the experiments, to implement the required analytical tools and instruments and to get the clearance by the utilities to publish the spent fuel data. Some of the experiments have run only for short time, which does not justify the level of investment. For this reason, we believe that the definition of a long-term project allowing for the continuation of the experiments will maximize the outcome of the efforts invested. This will provide

- Improved statistics for the IRF of other fission products.
- In depth investigations of low concentrated but very relevant for the Safety Case isotopes such as the FP 79Se, and Pd, or activation products ^{36}Cl and ^{14}C.
- Clear correlations for predicting the IRF from nuclear power plant operational parameters (power rates, temperatures, FGR).
- A basis for delineating the instant release from long-term radionuclide release.
- Data for additional types of samples (e.g. MOX)

For this reason, it is intended to keep the experimental set-up and the materials for an interim period and to apply for a new project in the future [16].

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