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# PROJECT FINAL REPORT

**Grant Agreement number:** FP7-295722

**Project acronym:** FIRST-NUCLIDES

**Project title:** FAST / INSTANT RELEASE OF SAFETY RELEVANT RADIONUCLIDES FROM SPENT NUCLEAR FUEL

**Funding Scheme:** COLLABORATIVE PROJECT

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**Name of the scientific representative of the project's co-ordinator<sup>1</sup>, Title and Organisation:**

Dr. Bernhard Kienzler  
Karlsruher Institut für Technologie (KIT),  
Institut für Nukleare Entsorgung (INE)  
Hermann-von-Helmholtz-Platz 1  
76344 Eggenstein-Leopoldshafen

**Tel:** +49 721 608 24467

**Fax:** +49 721 608-24308

**E-mail:** [bernhard.kienzler@kit.edu](mailto:bernhard.kienzler@kit.edu)

**Project website address:** <http://www.firstnuclides.eu/>

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<sup>1</sup> Usually the contact person of the coordinator as specified in Art. 8.1. of the Grant Agreement.

## 1. Executive summary

The EURATOM FP7 Collaborative Project “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (CP FIRST-Nuclides)” aimed on providing for “instant release fractions (IRF)” for relevant radionuclides from high burn-up spent nuclear  $\text{UO}_2$  fuel (SNF). The project supported the needs of safety analysis for different host rocks but also for the variety of SNFs to be disposed of in each repository. Shortly after failure of a canister, long-lived radionuclides can be released, mainly those with a high solubility in groundwater, also contributing significantly to dose to man. The project was implemented by a consortium of 10 European beneficiaries. 13 Associated Groups joined the project being involved in FIRST-Nuclides related fields. National Waste Management Organizations (WMO) participated in the End User Group guiding the Project with respect to the usefulness of the project achievements for the purpose of improving confidence in the Disposal Safety Case.

After completion of the CP FIRST-Nuclides, a significantly broadened knowledge is achieved by investigations covering 12 different types of high burn-up LWR SNF, IRF measurements at 45 different time steps, for up to 3 sample preparations, and up to 20 isotopes. The experiments were performed in a standard leachant guaranteeing comparable results. The data comprise the dissolution based radionuclide release for different samples, sample sizes and preparations from PWR and BWR SNF under aerobic, anaerobic and reducing conditions in the standard leachant. The data include cumulative gap and grain boundary releases for caesium and iodine up to one year experimental duration as well as the relevant rates. Release rates for other elements incorporated in the  $\text{UO}_2$  matrix, redox sensitive elements, and IRF of selenium was also determined. For  $^{14}\text{C}$  only few data could be obtained, the analyses required specific analytical techniques. The results are still outstanding. One important improvement of FIRST-Nuclides in comparison to previous studies was the clearance of the fuel owners to publish the power plant specific data, such as burn-up, power rating and calculated fission gas releases. Releases from non-standard SNFs were also investigated, such as from extremely high burn-up TRISO fuel and from leaking VVER fuel assemblies.

Results on the chemical state of Se were obtained by micro X-ray absorption near-edge structure (XANES) spectroscopy. A model is available for predicting the wetting of SNF and the time behaviour of radionuclide release. For evaluation and interpretation of the results, operational parameters have been made available. All data of the CP FIRST-Nuclides as well as the previously published data are compiled in a database available for application in safety analyses. Some relations could be derived, relating the IRF of Cs and I with the fission gas release and the power rating of the high burn-up  $\text{UO}_2$  fuel.

Up to now, 166 reports, publications and presentations at conferences document the achievements and results of FIRST-Nuclides.

## 2. Summary description of project context and objectives

The general principles of the COUNCIL DIRECTIVE 2011/70/EURATOM of 19 July 2011 establishing a Community framework for the responsible and safe management of spent fuel and radioactive waste<sup>2</sup> demand among others that “spent fuel and radioactive waste shall be safely managed, including in the long term with passive safety features”. Within this framework, the Strategic Research Agenda (SRA) of the “Implementing Geological Disposal – Technology Platform (IGD-TP) ([www.igdt.eu](http://www.igdt.eu))” defines key topics which are needed for the assessment of the safety of a deep waste repository system over extended periods of time. One of the key topics concerns the understanding and the behaviour of various wastes in geological repositories in particular for high burn-up spent uranium oxide (UO<sub>2</sub>) fuels. In many countries, this waste type represents the most important source for the release of radionuclides after loss of the canister integrity. For safety analysis, the time-dependent release of radionuclides from used high burn-up UO<sub>2</sub> fuel is required. The EURATOM FP7 Collaborative Project “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (CP FIRST-Nuclides)” elaborated these questions experimentally and theoretically.

The project tackled the challenge of realistically describing the release of the first batch of radionuclides from disposed spent nuclear fuel upon canister failure (fast/instant release fraction). This first release consists of radionuclides in gaseous form, and with a high solubility in groundwater. Some important nuclides show only marginal retention on their way to the biosphere. In present safety analyses, these radionuclides have a significant contribution to dose to man. The basis for the calculated significant dose contribution are simplified descriptions of the release functions. FIRST-Nuclides aimed to provide realistic release functions.

The CP started January 1<sup>st</sup>, 2012 and ended on December 31<sup>st</sup>, 2014. 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 National 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,

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<sup>2</sup> Council of the European Union, COUNCIL DIRECTIVE 2011/70/EURATOM of 19 July 2011 establishing a Community framework for the responsible and safe management of spent fuel and radioactive waste. Official Journal of the European Union, 2011. L 199(2.8.2011): p. 48-56.

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. During the duration of the project, a close cooperation was developed between the partners and the Associated Groups. A group of six implementation 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 regulators) were reflected in the project work, and they reviewed the project work and the scientific-technical outcome.

The fast release of radionuclides takes only place after a canister failure and water penetration to the SNF in a repository. The amount of fast released radionuclides is proportional to the mass of SNF contained in a defective canister, to the distribution of the relevant radionuclides in the plenum of the fuel rods, in the gaps between cladding and pellets, in the grain boundary, and in the  $\text{UO}_2$  grains. The CP FIRST-Nuclides aimed to determine and quantify the “instant release fraction (IRF)” values of fission gases, caesium, iodine ( $^{129}\text{I}$ ),  $^{14}\text{C}$  compounds, selenium ( $^{79}\text{Se}$ ), technetium ( $^{99}\text{Tc}$ ) and tin ( $^{126}\text{Sn}$ ). It was intended to determine the release functions of these isotopes from the gap of fuel pellets and from the  $\text{UO}_2$  grain boundaries. The determination of the dependency of the fast/instant release fraction from high burn-up  $\text{UO}_2$  fuel, the burn-up history, linear power and fuel temperature history, including possibly ramping processes, and storage time was of high importance. The manufacturing processes were also taken into account. Another objective was the transition between instant/fast release and the significantly slower release processes due to radiolytically driven  $\text{UO}_2$  matrix corrosion. For this reason, the accessibility of, and transport properties on grain boundaries and the quantification and characterization of exchange processes along the grain boundaries was investigated. Conceptual and numerical modelling was also implemented in the project in order to assess (1) boundary conditions for different systems and compare with analytical methods, (2) fission product migration along the grain boundaries, (3) effects of fractures in the pellets as well as of holes/fractures in the cladding on the fast release and (4) modelling the chemical state of the relevant elements. Direct applications of the results to safety assessments were not foreseen within the project.

Spent fuel rods are owned mainly by the reactor operating companies. Due to their competition, it is not easy to get the complete information on the actual burn-up history of the fuel samples, as well as the right to publish the data. To get approval by the utilities required a lot of work and negotiations between the related institutions. The spent nuclear fuel considered in the CP FIRST-Nuclides consisted of  $\text{UO}_2$  fuel. The burn-up covered the range of 45 to 70  $\text{GWd/t}_{\text{HM}}$  (test rod with 14 cycles in a PWR), which is representative for present fuels in pressure water reactors (PWR) and boiling water reactors (BWR). The average linear power rate was between 160 and 330  $\text{W/cm}$  and the calculated FGR between 1.2 % and 23 % (MOX fuel).

The dissolution based release studies were performed using different kinds of SNF samples: (1) Pellet: according to the production process, (2) Segment: cuts through the middle planes of pellets, (3) Slice: cut through a single pellet, (4) Fragment: piece or portion of fuel, (5) Powder: fuel material obtained after declad, sieving and milling. (6) Cladding with attached fuel. Pellets, Segments and Slices can be prepared with and without cladding. In order to improve the basis for comparison of the results from different laboratories, a standard leachant was defined, consisting of 19 mM NaCl + 1 mM NaHCO<sub>3</sub> at pH 7.4 to 8.2. The experiments were performed under aerobic, anaerobic and reducing conditions.

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 SNF was used for the experiments. WP2 covered the “Gas release and rim and grain boundary diffusion experiments” whereas WP3 addressed “Dissolution based release studies” and WP4 “Modelling”. WP5 “Knowledge, reporting and training” was responsible for the knowledge management generated within the project, including a state-of-the-art report, the general reporting, keeping the documentation and web page up-to-date and organizing training measures. The management of the Collaborative Project was included in WP6.

An important objective of CP FIRST-Nuclides was dissemination of the achieved knowledge. Detailed documentation of the scientific results can be found in three Annual Project Workshop Proceedings (2012, 2013, 2014), published along with the progress of the project as well as a large number of reviewed papers, institutional reports and other publically available sources. The outcome of the project is to be published in peer-reviewed journal articles. It will cover the dependence of IRF of Cs and I on sample properties, the gap and grain boundary release as function of the reactor operational conditions (initial enrichment, burn-up, FGR, average power rating, etc.), the IRF of other elements/isotopes on the fuel samples and sample preparations, and experimental conditions. The efforts on improving the knowledge for specific elements (<sup>79</sup>Se and <sup>14</sup>C), modelling approaches, and data for non-standard SNF are described. The state-of-the-art is summarized in the IRF-Database, where the available information on previous and present IRF studies is compiled.

The project aimed also to training and education of young scientists. This aim was achieved by a Lab exchange meeting, a training course at KIT-INE and by mobility measures for 3 PhD students.

### 3. Description of the main S&T results

FIRST-Nuclides project has been highly productive and high amount of important scientific results have been generated. All the obtained results have been presented at the annual project workshops and published in the corresponding proceedings. The proceedings of the three project workshops are available at the project website ([www.firstnuclides.eu](http://www.firstnuclides.eu)).

In the following sections, a summary of the research done during the three years of FIRST-Nuclides and the generated knowledge is presented.

#### 3.1. WP1 “Samples and tools: Selection, characterization and preparation of materials and set-up tools”

##### Objectives

The overall objectives of WP1 were to select, provide and prepare spent nuclear fuel (SNF) samples for subsequent experimental investigations. The objectives of this workpackage included the complete characterization of the selected SNF materials with respect to the individual fuel characteristics and irradiation history, achieving permission by the fuel owners for publication of key parameters as well as the installation of experimental and analytical tools.

##### Description of work performed in the workpackage

All experimentally working partners of the FIRST-Nuclides project (i.e. KIT, JRC-ITU, JÜLICH, PSI, SCK•CEN, CNRS, CTM, MTA EK and STUDSVIK) contributed to this workpackage. Samples (cladded pellets, pellets, powders, TRISO kernels etc.) of a series of high burn-up fuel materials were comprehensively characterized and prepared for various spectroscopic, metallurgic and chemical analyses as well as subsequent investigations in WP2 and WP3. For experimental studies within WP2 and WP3, experimental set-ups (incl. autoclaves, irradiation cells, reaction vessels), specific sampling devices and analytical equipment were provided and installed in the hot cells, shielded box-line and He<sup>2+</sup> irradiation facility, respectively.

**KIT** provided the PWR spent nuclear fuel rod segment SBS1108-N0204, which achieved an average burn-up, BU, of 50.4 GWd/t<sub>HM</sub> during irradiation in a PWR. Data on irradiation history of the segment were documented. Segment SBS1108-N0204, which is owned by KIT, was transported to JRC-ITU for characterisation, analyses and sampling of fuel pellets. After return of fuel pellets to KIT, they were further analysed. Fragments and a cladded fuel sample were prepared for consecutive leaching experiments.

**JRC-ITU** performed non-destructive analyses, gas sampling, cutting and sampling of fuel pellets from the segment SBS1108-N0204. Ceramographic samples were prepared from an irradiated initial enriched PWR UO<sub>2</sub> fuel as well as from irradiated initially depleted UO<sub>2</sub>



PWR fuel of this segment. With respect to HBU-SNF samples for studies by JRC-ITU and CTM, two commercial BWR fuels (denoted as BWR42 and BWR54) with average burn-up of 42 GWd/t<sub>HM</sub> and 54 GWd/t<sub>HM</sub>, were selected. Selected sub-samples of these SNF were dissolved in acidic media in order to determine the inventories and the burn-ups of these BWR SNFs were determined. Powder samples and clad fuel samples were prepared for consecutive leaching and diffusion experiments.

**JÜLICH** provided key parameters of the fuel history and irradiation characteristics on irradiated High Temperature Reactor (HTR / TRISO) fuel. From the selected HTR / TRISO material, polished samples and isolated UO<sub>2</sub> fuel kernels were prepared for subsequent investigations in WP2.

**PSI** selected a BWR UO<sub>2</sub> fuel with an average BU of 57.5 GWd/t<sub>HM</sub> and a PWR UO<sub>2</sub> fuel with 62.2 GWd/t<sub>HM</sub>. Additionally, a PWR MOX fuel with 63 GWd/t<sub>HM</sub> was selected for subsequent studies in WP3. A BWR UO<sub>2</sub> HBU-SNF sample similar to the BWR fuel sample selected for leaching experiments was prepared for two consecutive micrometer scale X-ray fluorescence (XRF) and X-ray absorption near edge structure (XANES) investigations performed at the microXAS beamline (Swiss Light Source (SLS), PSI). A complementary XAS feasibility study on non-irradiated depleted UO<sub>2</sub> powdered samples containing Se, Cs and I was also performed at the INE-Beamline (ANKA, KIT).

**SCK•CEN** collected manufacturing and operational data of a PWR fuel with an average BU of 52 GWd/t<sub>HM</sub>. The fuel studied at SCK•CEN was structurally and chemically characterized, including a determination of the local burn-up and radionuclide inventory, and comparison with the calculated burn-up and inventory.

**CNRS** installed an irradiation cell for microscopic studies of corrosion at UO<sub>2</sub> grain boundaries of (initially non-irradiated) TRISO under cyclotron radiation. CNRS prepared and characterized selected TRISO samples and sub-samples from the initial TRISO particles.

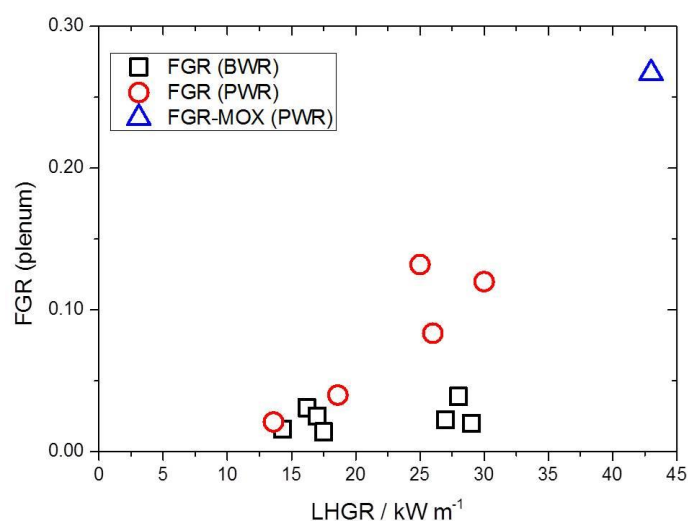
**MTA EK** compiled manufacturing and operational data on VVER fuel rods stored in water for several years. Isotope inventories of the rods were determined to support the calculation of activity release rates.

**STUDSVIK** selected four BWR fuels with average BU of 50.2, 54.8, 57.1 and 59.1 GWd/t<sub>HM</sub>, respectively, and two PWR fuel with 54.4 and 70.2 GWd/t<sub>HM</sub>, respectively. Relevant manufacturing and operational data of these HBU-SNF materials were collected and reported. Fuel samples were prepared for consecutive investigations.

Initially the activities within WP1 were planned to be concluded at the end of the first reporting period (i.e. project month 18). Since there were delays in the documentation and with the sample preparation of SNF materials used by JRC-ITU and CTM, these institutions continued their activities within WP1. Moreover, STUDSVIK and SCK•CEN extended their activities related to characterization of the selected SNF materials with respect to the individual fuel characteristics.

## Main results

The basic activities of the project FIRST-Nuclides, in particular the preparation and characterization of the SNF samples, were performed. Fission gas release (FGR), generally determined by puncturing the plenum of irradiated fuel rods, is considered as an indicator for the extent of the fast release of labile radionuclides, such as  $^{137}\text{Cs}$  and  $^{129}\text{I}$ . Data of such puncturing tests of the studied HBU-SNF samples demonstrated that FGR of the plenum is virtually correlated to the linear heat generation rate (LHGR) rather than the burn-up of the fuel (Figure 1). Since results on radionuclide inventories, structural features and other basic characteristics are used in the following work packages for correlations and interpretations of the experimental results, this information is given in the following sections.



**Figure 1.** Fission gas release measured in puncturing tests (FGR of the plenum) as function of the linear heat generation rate (LHGR) of the studied high burn-up spent nuclear fuels.

### 3.2. WP2 “Gas release + Rim and grain boundary diffusion: Experimental determination of fission gases release. Rim and grain boundary diffusion experiments”

#### Introduction

In the first project year the focus was on setting up experimental facilities, characterisation and preparation of samples. In the second year, in WP2 first results have been obtained by the partners but also new challenges arose. In the last project year most of the experiments have been finalised and evaluated.

Work package two is divided into the two components “Experimental determination of fission gas release” and “Rim and grain boundary diffusion”. In the first component, the focus is on the quantification of fission gases and fission gas release in high burn-up (HBU)  $\text{UO}_2$  spent



nuclear fuels (SNF). Fission gas sampled in the plenum of a fuel rod are analysed as well as the grain boundary inventory and the cross sectional distribution of fission gases and volatile fission products.

The second component “Rim and grain boundary diffusion” deals with investigations on oxygen diffusion in spent  $\text{UO}_2$  fuel. The examination of diffusion mechanisms will result in the quantification of water penetration into the fuel (grain and grain boundaries) structures and subsequently couple the diffusion/corrosion phenomena. Furthermore, investigations on irradiated and unirradiated fuel kernels separated from high temperature gas cooled reactor (HTR) fuel were performed on materials which are complementary to those on light water reactor (LWR) fuel.

The experimental part of WP2 started in project month 4 and ended in project month 36 (end of the project).

#### WP2, Experimental determination of fission gas release:

**JRC-ITU** was the leading organization of WP2. JRC-ITU has measured the fission gas release (FGR) from a spent fuel rod owned by KIT and determined the inventory of fission gas and fission products in grain boundaries.

**KIT** analysed fission and activation products in the fission gas sampled at JRC-ITU from the plenum of a fuel rod segment by puncturing. The development, testing and implementation of analytical methods for measuring fission and activation products have been done by KIT as well as leaching experiments.

**STUDSVIK** investigated the radial fission gas and volatile fission product distribution (Xe, I, Cs and Se) by Laser-Ablation Mass Spectroscopy (LA-MS) on HBU boiling water reactor (BWR) SNF.

#### WP2, Rim and grain boundary diffusion:

**JRC-ITU** has investigated on diffusion processes starting with the characterisation and preparation of spent fuel samples to be used for corrosion experiments in  $\text{H}_2^{18}\text{O}$  water at room temperature. In 2013 it was planned to determine the  $^{18}\text{O}/^{16}\text{O}$  depth profiles using a shielded SIMS (secondary ion mass spectrometry) to quantify the oxygen diffusion into spent nuclear fuel (SNF).

**JÜLICH** was working on spent high temperature reactor fuel. The radionuclide inventory in the fuel kernel and in the coatings were determined and compared to calculated values as well. Further on investigations of the microstructure and of the elemental distribution of the fuel kernel and of the coatings were performed before and after leaching. After cracking of the tight coatings the fission gas release fraction was measured. Then static leaching experiments with the separated fuel kernels and coatings were started to determine the fast instant radionuclide release fraction.

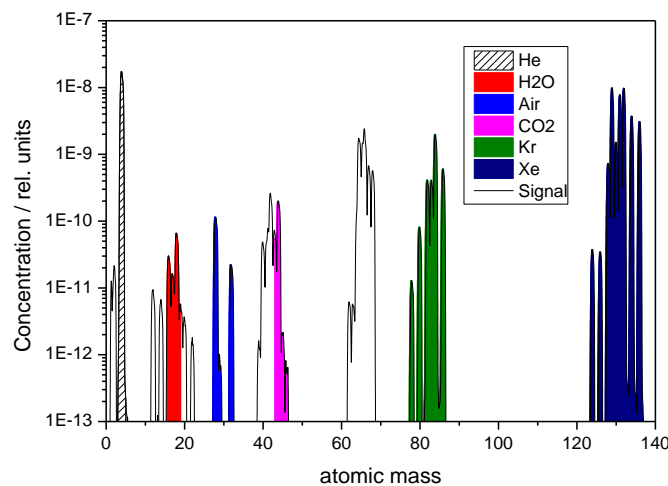
Unirradiated tri-structural-isotropic (TRISO) fuel particles were investigated by **CNRS** at the ARRONAX cyclotron. The particles were irradiated using a  $\text{He}^{2+}$ -beam in the dose rate range of 0.8 – 4.4 kGy/min. The corrosion of  $\text{UO}_2$  TRISO particles was investigated in view of grain boundary effects, secondary phase formation and under the influence of hydrogen. The experiments focused on investigating the role of grain boundaries and fuel particle corrosion under hydrogen and in varying dose rates were performed.

## Achievements

### Experimental determination of fission gas release

JRC-ITU has examined the release of the fission products  $^{88}\text{Sr}$ ,  $^{87}\text{Rb}$ ,  $^{137}\text{Cs}$ , and  $^{136}\text{Xe}$  from powder of irradiated BWR  $\text{UO}_2$  (54GWd/t<sub>HM</sub>) originating from the core region of the fuel pin. The sample was heated in a Knudsen cell coupled to a mass spectrometer under vacuum conditions at a rate of 10 K/min until complete vaporization of the fuel at 2460 K. The measured release profiles indicate two release mechanisms for  $^{137}\text{Cs}$ , one with rather low intensity starting just below 1000 K and a second significant release after 1500 K. The release of the fission products  $^{88}\text{Sr}$ ,  $^{87}\text{Rb}$ ,  $^{137}\text{Cs}$ , and  $^{136}\text{Xe}$  has been semi-quantified.

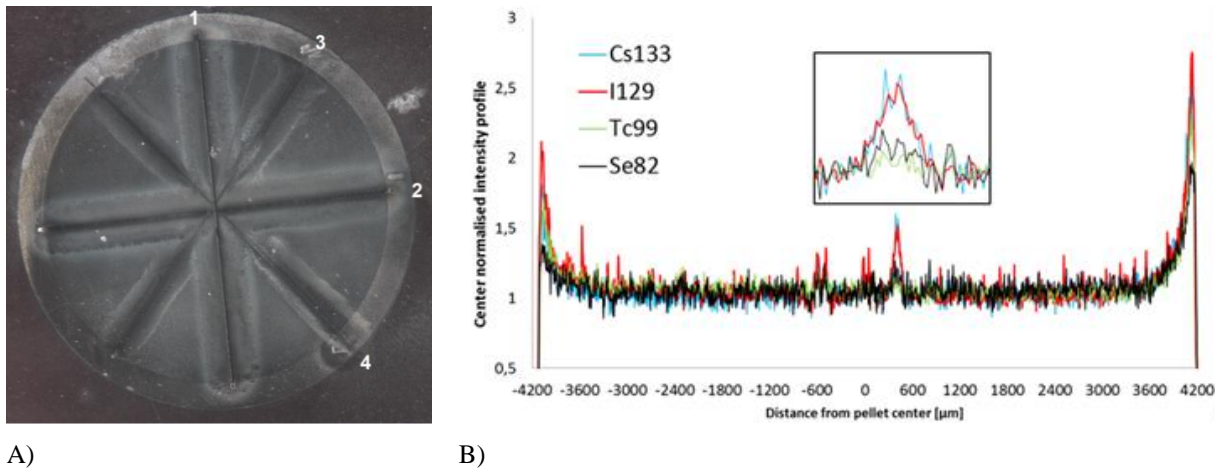
KIT has performed leaching experiments on a well characterised samples of clad PWR fuel and PWR fuel fragments under  $\text{Ar}/\text{H}_2$  (pT = 40 bar; p $\text{H}_2$  = 3 bar) in bicarbonate water (19 mM  $\text{NaHCO}_3$  + 1 mM  $\text{NaCl}$ ). After a cumulative contact time of 246 days, 15% of the fission gases (Kr+ Xe) were released into the gas phase for the clad fuel pellet while for fuel fragments 11% of the fission gases (Kr+ Xe) inventory was released after 27 days.



**Figure 2:** Example of fission gas measurements carried out at KIT/INE

STUDSVIK finished the evaluation of Laser Ablation data on cross sections from a standard  $\text{UO}_2$  fuel and an Al/Cr-additive fuel. The caesium and iodine profiles measured on both fuels are very similar and follow the radial burn-up profile. Caesium, iodine and to some extent selenium also appear to collect in some fuel cracks. Selenium was tentatively identified by the

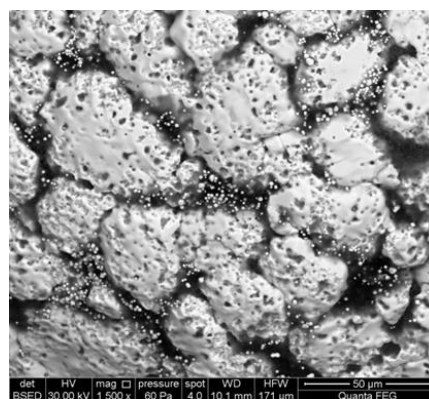
good agreement of the isotopic ratios of mass 77, 79 and 82 with the calculated inventory. Chromium and especially aluminium are heterogeneously distributed in the Al/Cr-additive fuel.



**Figure 3:** a) Overview of Al/Cr additive sample after laser ablation. B) Ablation profiles of the Al/Cr-additive fuel sample.

### Rim and grain boundary diffusion

JÜLICH has investigated  $\text{UO}_2$  TRISO coated particles from spent HTR fuel (burn-up  $\sim 100 \text{ GWd/t}_{\text{HM}}$ ).  $\text{UO}_2$  fuel kernels were exposed to synthetic groundwater (19 mM NaCl and 1 mM  $\text{NaHCO}_3$ ) for 276 days under oxidic (air) and anoxic/reducing ( $\text{Ar}/\text{H}_2$ ) conditions. The U release was below detection limit. During 276 days maximal 0.2 %  $^{90}\text{Sr}$  and 8 %  $^{134/137}\text{Cs}$  were released from grain boundaries. Depending on the environmental conditions, different release functions were observed. ESEM (environmental scanning electron microscope) investigations show on samples leached in oxidic environment, numerous intragranular open pores acting as new accessible leaching sites and white spherical spots containing Mo and Zr. Under anoxic/reducing conditions numerous metallic precipitates (Mo, Tc and Ru) filling the intragranular pores and white spherical spots containing Mo and Zr, were detected.



**Figure 4:** SEM picture of a spent  $\text{UO}_2$  kernel after the leaching experiment under anoxic conditions.

CNRS has studied the oxidation of  $\text{UO}_2$  in unirradiated HTR fuel kernels by  $\alpha$ -radiolysis products of water. Radiolysis products were produced by 66.5 MeV  $\text{He}^{2+}$  beam irradiation of water in the ARRONAX cyclotron giving a dose rate of 4.37 kGy/min. The obtained results show that gaseous  $\text{H}_2$  produced by water radiolysis completely inhibits the  $\text{UO}_2$  corrosion by interaction between  $\text{H}_2$  and the  $\text{UO}_2$  surface. The  $\text{UO}_2$  radiolytic corrosion expressed as a function of the absorbed dose show a ten-fold higher dissolution rate for the lower dose rate (0.8 kGy/min) than that found for the higher one (4.4 kGy/min). If OH radicals are produced near the  $\text{UO}_2$  surface they interact directly with the  $\text{UO}_2$  surface and not with each other to form  $\text{H}_2\text{O}_2$ . In this case also the radiolytic corrosion is higher than in presence of molecular  $\text{H}_2\text{O}_2$ . It is also shown that the  $\text{UO}_2$  radiolytic corrosion under  $\text{He}^{2+}$  radiation occurs essentially on the grain boundaries and not on the crystallized  $\text{UO}_2$  grains.

JRC-ITU analysed and modelled diffusion profiles measured on  $\text{UO}_2$  by parallel diffusion processes. No new experimental results on oxygen diffusion experiments have been obtained because during refurbishment of the autoclave in the hot cell, intended for oxygen diffusion experiments, a Ti-welded tube joint broke. For safety reasons a replacement of the complete autoclave setup is necessary. Despite all efforts undertaken it was not possible to get the set-up back in working conditions because of the delivery time of special valves needed. It was decided to prolong WP2 from 33 to 36 months and to publish the results in an open journal with reference to FIRST Nuclides.

### **3.3. WP3 “Dissolution Based Release: Dissolution based fast radionuclide release”**

#### **Objectives**

The overall objectives of WP3 were the quantification of the fast release of gaseous and non-gaseous activation and fission products into the aqueous phase during spent nuclear fuel leach tests, and – to the extent possible - the determination of their chemical speciation. The experiments covered high burn-up spent nuclear fuels (HBU-SNF) having a burn-up mostly in the range of 50 to 60  $\text{GWd/t}_{\text{HM}}$ , different irradiation histories, reactor types and sample positions in the fuel rods. Special emphasis was given to the determination of IRF values of caesium, iodine, selenium and carbon-14 (from fuel and cladding) as well as to the redox speciation of selenium. Additionally, dissolution rates for relevant isotopes were determined for damaged VVER fuel elements (to be disposed of in Germany, Hungary and Finland). The results from experiments using fuel samples with/without cladding allow an evaluation of the effect of the presence of the cladding material.

## Description of work performed in the project

Seven institutes were involved in WP3, i.e. KIT, JRC-ITU, PSI, SCK•CEN, CTM, STUDEVIK and EK. All of these institutes, except for EK, have performed leach experiments on samples of high burn-up fuels in laboratory conditions. The CTM experiments were performed in the laboratories of JRC-ITU. EK did not perform laboratory experiments, but has contributed by collecting and interpreting 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 leach experiments by the various institutes were harmonized as much as possible to allow a better comparison of the leach data. For this reason, a standard leachant (19 mM NaCl and 1 mM NaHCO<sub>3</sub>) was used by most laboratories, and tests with similarly prepared samples (cladded fuel segments) were added by several laboratories. Nevertheless, there were also differences in experimental approaches, such as the redox conditions (reducing or oxidizing atmosphere), leaching mode (static or pseudo-dynamic), and alternative sample preparations (declad fuels, fuel fragments, fuel powders...). Both PWR and BWR fuels were tested. One test series with a MOX fuel was added as well. Test durations up to 1 year were planned, but for part of the experiments, this duration will be reached (or the results will be available) after the end of the project. The leachates were analysed for a large number of radionuclides, depending on the techniques available in the participating laboratories. However, most laboratories have measured (or planned to measure) the most critical IRF radionuclides, i.e. Cs and I isotopes. Other isotopes that were measured by several, but not all laboratories are <sup>14</sup>C and <sup>79</sup>Se. For some isotopes, the analytical methods were developed in the framework of the project. Special efforts were made to characterize selenium in the spent fuel by solid state analyses. More details about the contribution of each institute are given hereunder.

**KIT** has performed leach experiments on a spent PWR UOX fuel with average burn-up of 50.4 GWd/t<sub>HM</sub>. 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 (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 static mode. The IRF of relevant radionuclides such as I, Cs, Tc, Sr and U isotopes was determined and reported for test durations up to 245 days. The autoclave set-up also allowed the measurement of the fission gas release during the leach tests. A test with cladding and adhering residues was prepared as well, but these results were not yet available at the end of the project. A method for the extraction and determination of <sup>14</sup>C has been developed.



**Figure 5:** Dissolution based release experiments conducted under hydrogen pressure.

**JRC-ITU** has performed leach experiments on a spent BWR UOX fuel with an average burn-up of 42.2 GWd/t<sub>HM</sub>. 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.

An analytical method has been developed for the determination of <sup>90</sup>Sr at ultratrace levels using the seaFAST system and ICP-MS detection. A method for the determination of <sup>79</sup>Se species at trace levels by high resolution ICP-MS coupled to an automated chromatographic system has been developed. The analytical figures of merit of the method for the determination of <sup>90</sup>Sr at ultratrace levels are a detection limit of 36 fg/g (0.2 Bq/g) representing an absolute amount of 9 fg (50 mBq). The repeatability is below 2%. The method has been tested using nuclear spent leachates analogues prepared in a buffer of NaCl 19 mol/L and NaHCO<sub>3</sub> 1mmol/L pH 8.06 with a uranium concentration from 1·10<sup>-7</sup> to 10<sup>-5</sup> mol/L. The method to determine <sup>79</sup>Se species at trace levels was applied to diluted nuclear spent fuel leachates analogues consisting in a buffer solution (NaCl 19mM, NaHCO<sub>3</sub> 1mM pH 7.4) with uranium concentration varying from 10<sup>-7</sup> to 10<sup>-5</sup> M with individual Se species in different concentrations. The detection limit is 12 pg/g, representing an absolute amount of 3 pg, with repeatability between 0.2 and 1 %. The method preserves Se species presents in the sample.

**PSI** has performed leach experiments on a spent BWR UOX fuel with an average burn-up of 57.5 GWd/t<sub>HM</sub>, spent PWR UOX fuel with an average burn-up of 56.5 GWd/t<sub>HM</sub>, and spent MOX fuel with an average burn-up of 63 GWd/t<sub>HM</sub>. They have tested in parallel the leaching of clad 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). 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 and reported for test durations up to 182 days. Tests with completely cleaned claddings (without fuel residues) were planned as well, but the results were not yet available at the end of the project. In collaboration with STUDSVIK, samples of the BWR fuel were analysed with micro-XRF and micro-XAS to determine the selenium distribution on the microscale, its oxidation state and its structural environment (next-neighbour distances and coordination numbers).

**SCK•CEN** has performed leach experiments on a spent PWR UOX fuel with an average burn-up of 50.5 GWd/t<sub>HM</sub>. They have tested in parallel the leaching of clad 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 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 analysed, a.o. <sup>14</sup>C, <sup>99</sup>Tc and <sup>238</sup>U. SCK•CEN was also work package leader of WP3.

**CTM** has performed leach experiments on a spent BWR UOX fuel with an average burn-up of 54 GWd/t<sub>HM</sub>. 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. CTM has collaborated with JRC-ITU also for the improvement of the analytical procedures for the measurement of <sup>90</sup>Sr and <sup>79</sup>Se (see higher under activities JRC-ITU).

**STUDSVIK** has performed leach experiments on samples of six types of a spent fuel, i.e.

- BWR UOX fuels with an average burn-up of 50.2 GWd/t<sub>HM</sub> (test on clad fuel segment), 54.8 GWd/t<sub>HM</sub> (test on clad fuel segment), and 57.1 GWd/t<sub>HM</sub> (test on fragments + separated cladding)
- An Al/Cr doped BWR UOX fuel with a burn-up of 59.1 GWd/t<sub>HM</sub> (test on fragments + separated cladding)
- A Gd doped PWR UOX fuel with an average burn-up of 54.4 GWd/t<sub>HM</sub> (test on fragments + separated cladding)
- A spent PWR UOX fuel with an average burn-up of 70.2 GWd/t<sub>HM</sub> (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<sub>3</sub>) 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. Laser Ablation Inductively Coupled Plasma Mass Spectroscopy analysis was applied on fuel cross-sections from the BWR fuel with burn-up 57.1 GWd/t<sub>HM</sub> and the Al/Cr doped BWR fuel, to reveal the radial profile of Cs, I and Se.

**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 a burn-up in the range 10.1-26.7 GWd/t<sub>HM</sub>. The leaking fuel had a burn-up of 14 GWd/t<sub>HM</sub>. The leachant was in this case the water of the storage pool. In the case of the damaged fuel, the pH of water was ≈7 in the first 14 days after the incident (15 g boric acid per kg of water) and ≈4-4.5 (21 g boric acid per kg of water) 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 system has led to periods of increasing and decreasing radionuclide concentrations. The dissolution rate was calculated in various ways for a series of isotopes, a.o. Cs, I and U isotopes.

An overview of the leach test matrix is given in Table 1.

**Table 1:** Overview of the leach test matrix of WP3, with for each tested fuel the average burn-up in GWd/t<sub>HM</sub> and the involved laboratory. The measurements by MTA-EK are not included in this table.

	<i>BU</i>	Cladded fuel segments	Opened cladding + fuel fragments	Fuel fragments (mm size)	Fuel powder	Cladding + adhering residues	Cleaned Cladding
<b>PWR</b>	50.4	KIT		KIT		KIT	
	50.5	SCK	SCK				
	56.6	PSI		PSI		PSI	PSI
	54.4		STUDSVIK*				
	70.2				STUDSVIK		
<b>BWR</b>	42.2	ITU			ITU <sup>†</sup>		
	50.2	STUDSVIK					
	54	CTM			CTM <sup>†</sup>		
	54.8	STUDSVIK					
	57.1		STUDSVIK				
	57.5	PSI		PSI			PSI
	59.1		STUDSVIK <sup>#</sup>				
<b>MOX</b>	63			PSI			PSI

\*Gd doped; <sup>#</sup> Al/Cr doped; <sup>†</sup> separately for rim and core.

## Main results and preliminary conclusions

- Until recently, the IRF of spent fuel was often related to its burn-up. With more data becoming available for high burn-up fuel, there is growing evidence that the fission gas release (FGR) and IRF depends much more on the linear power rating of the fuel than on the burn-up. The data gathered in FIRST-Nuclides give further convincing 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 operator, while this temperature depends on the linear power, rather than on the burn-up.
- The measurements of the FGR during the leach test by KIT have shown that FGR takes place also during the leaching of the fuel. The amount of fission gases released in this way appears to be higher than the amount of fission gases released in the plenum during the fuel operation.
- The IRF values measured for Cs tend to be proportional to the FGR with a proportionality factor of 0.6. The IRF values measured for I tend to be proportional to the FGR with a proportionality factor of 1. The difference between the Cs or I release and FGR appears to increase in absolute terms with higher FGR, where the FGR increasingly overestimates the Cs or I release. To explain the increasing differences, a detailed analysis of each test should be performed. From the perspective of performance assessment, the upwards deviations (i.e.  $\text{IRF} > 0.6 \times \text{FGR}$  for caesium, or  $> 1 \times \text{FGR}$  for iodine) are more relevant than the downwards deviations. To include all measurement points, both from FIRST-Nuclides and from older experiments, an extra term should be added.
- The IRF of Cs or I determined from differently prepared samples (cladded segments, opened segments, fragments, powders...) are coherent. They depend on the exposed surface area and the nature of the exposed fuel structures (gap or grain boundaries). These data can be used further to estimate the relative contributions of the various fuel structures.
- The doping of  $\text{UO}_2$  with Al/Cr appears to have a favorable effect on the 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.
- The solid state analyses of the spent fuel suggest that selenium may be present in the redox states 0 and +IV or in -II. In this form it replaces oxygen as Se(-II) in the  $\text{UO}_2$  lattice.
- The Cs dissolution rate decreases with time in the leach tests. The net dissolution rates measured for the damaged and leaking VVER fuel (after subtraction of the  $\text{UO}_2$

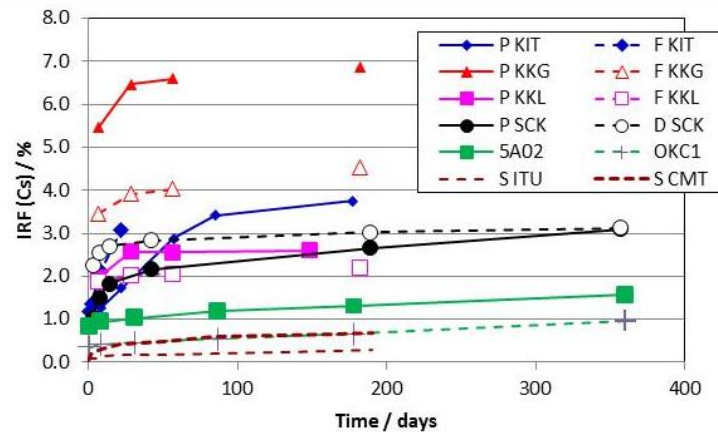
matrix dissolution) are of the same order of magnitude as the rates determined in the leaching tests, and also decrease with time. Nevertheless, in almost all leach experiments, a residual dissolution rate of Cs isotopes was measured for the longest test durations. This means that the measured IRF for the longest duration ( $\leq 1$  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  $\text{UO}_2$  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. 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.

- The IRF values measured in the leach tests can be compared with Performance Assessment oriented best estimate and pessimistic IRF estimations from a previous program (NF-PRO). It appears that Cs and I are confirmed as the most important IRF isotopes, and that the previous pessimistic estimates may still underestimate the IRF of fuels that have undergone a high linear power rating. The new IRF data available for  $^{14}\text{C}$  and Pd suggest that the previous estimations may be too pessimistic. The previous estimates for Sr and Tc are roughly confirmed by the new results. Se and Sn (left out as IRF nuclides in NF-PRO) may have small IRF contributions.

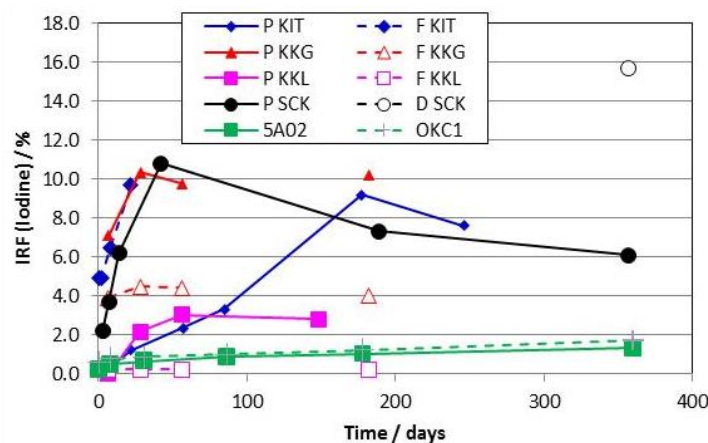
As an example of the outcome of WP3 the cumulative release fractions of I and Cs determined from differently prepared samples (clad segments, opened segments, fragments, powders, etc.) are shown in Figure 6 and Figure 7 including all the results of all beneficiaries.

The release depends on the operation parameters, such as burn-up 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. It was also observed that the IRF was higher for PWR fuel in comparison to BWR fuel even for similar burn-up, probably because the linear power was higher in the PWR fuel.

All leach tests were performed with the standard leachant. However, some tests were conducted in autoclaves under  $\text{Ar}/\text{H}_2$  gas overpressure (i.e. under reducing conditions). No differences are observed for Cs and I release under reducing and oxidizing or anaerobic conditions.



**Figure 6:** Measured cumulative  $^{137}\text{Cs}$  release from various spent nuclear fuel samples studied within the project.



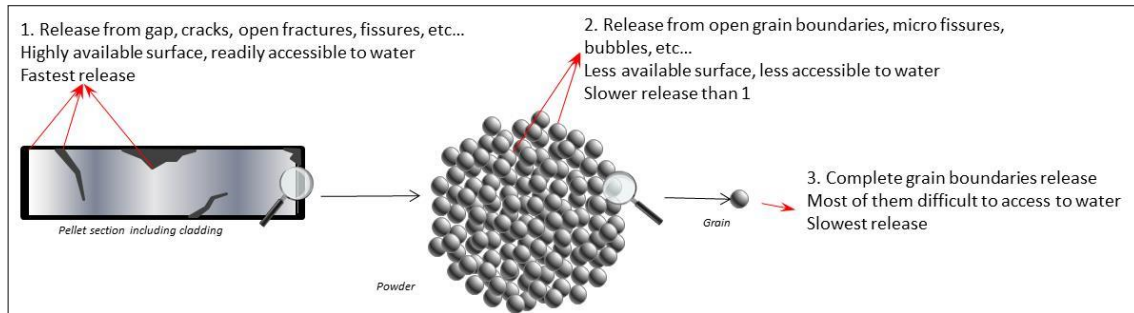
**Figure 7:** Measured cumulative  $^{129}\text{Is}$  release from various spent nuclear fuel samples studied within the project.

### 3.4. WP4 “Modelling: Modelling of migration/retention processes of fission products in the spent fuel structure”

#### Objectives

The objectives of WP 4 cover initial speciation of fission products in LWR fuel, and multi-scale modelling of the migration / retention processes of fission products in the HBU spent fuel, in the cladding, and the estimation of the fission product total release through the spent fuel rod.

On the other hand, a semi-empirical model was developed to predict fission product release to water from gap, grain boundaries and grains, taking into account Figure 8.



**Figure 8:** Conceptual description of the experimentally determined release of different radionuclides segregated from the matrix based on sample location, availability to water and sample pre-treatment.

### Description of work performed in the project

Three institutions were involved in WP4: KIT, AMPHOS 21 and CTM-UPC, ITU had a small contribution related to the use of the TRANSURANUS code.

INE-KIT focused its modelling on describing the boundary and initial conditions of the SNF, the thermodynamic data database for evaluation of the speciation and the formation of compounds of  $^{14}\text{C}$  in the fuel as well as the fission products diffusion as a function of the temperature during reactor. AMPHOS 21 modelled water penetration and saturation of segmented fuel samples with respect to the existence of micro and macro-cracks. CTM-UPC developed a semi-empirical model based on spent fuel leaching experiments to relate the observed release to the radionuclide inventories in the gap, the grain boundaries and the matrix.

More details about the contribution of each beneficiary are given below.

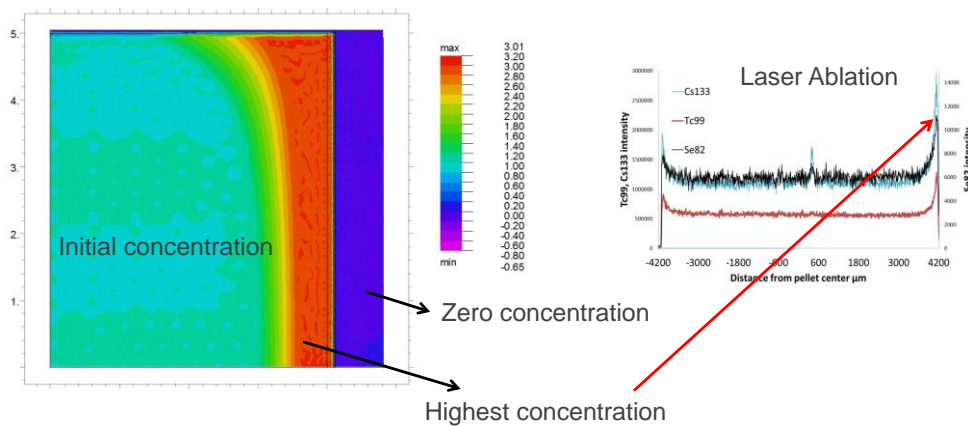
**KIT** performed calculations of burn-up, thickness and porosity of rim zone for initial and boundary conditions of a fuel sample with a burn-up of  $50.4 \text{ GWd/t}_{\text{HM}}$ , as well as Xe location in this zone.

They combined thermodynamic considerations with experimental investigation aiming on the speciation of  $^{14}\text{C}$  in the spent nuclear fuel matrix. Carbon-14 is a key radionuclide in the safety assessment and in disposal concepts it is assumed, that  $^{14}\text{C}$  bearing species are not retained.  $^{14}\text{C}$  is an activation product in spent nuclear fuel (SNF) showing low concentrations; its chemical speciation is widely unknown. After formation of  $^{14}\text{C}$  by a  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction, the highly excited and charged carbon competes with available reactants within the fuel matrix and will likely form either oxides or in reactions with metals, carbides. In contact with water, these compounds react by forming carbonates or hydrocarbons. The potential for the formation of oxides or carbides was investigated. Free energy of formation for some relevant carbides and oxides were provided. The results of thermodynamic considerations for  $^{14}\text{C}$  were compared with experimental gas measurements. According to the initial nitrogen concentration in the fuel between 4 and 11 ppm, the maximal nitrogen content of KIT's fuel rod sample was estimated to be  $2.1 \cdot 10^{-4} \text{ mol}$ . Using the KKG operation characteristics, a formation of  $4.3 \cdot 10^{-6} \text{ mol } ^{14}\text{C}$  was expected whereas the measured amount of  $^{14}\text{CO}_2$  was only



$7.2 \cdot 10^{-7}$  mol, corresponding to 17% of the total measured  $\text{CO}_2$ . However, the speciation of  $^{14}\text{C}$  is not yet resolved.

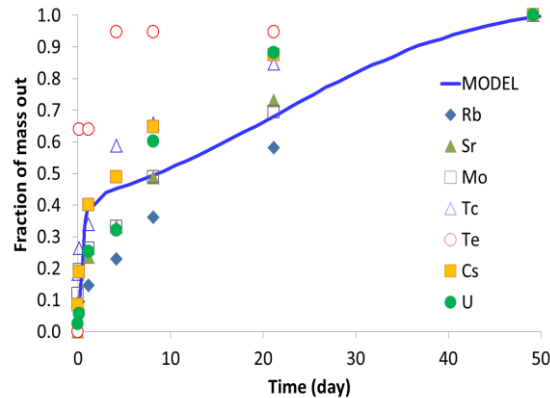
**KIT** also studied the diffusion of fission products in a fuel rod by modelling the temperature and diffusion coefficient of some radionuclides. Modelling was able to explain experimental LA-ICP-MS results (see Figure 9). Some conclusions were extracted from this modelling such as: i) Small gaps between the pellets or between pellet and cladding affect the temperature significantly; ii) Radial distribution of I, Xe, and Cs in a spent fuel pellet was modelled by assuming a temperature-dependent sorption; iii) The calculated total concentration in the colder region of the pellet corresponds to LA-ICP-MS measurements; iv) Calculated release of volatile radionuclides into the gap regions corroborate with the fission gas release (FGR) and v) After discharge from the reactor, diffusion processes of volatiles do not play any role due to the negligible diffusion coefficients at interim storage temperature.



**Figure 9:** Comparison of calculated volatile element profiles to LA-ICP-MS experimental results.

**AMPHOS 21** proposed a conceptual model that assumes that the rate of release for IRF radionuclides from a SNF pellet (or a fragment of it) upon contact with water takes place after wetting of surfaces of cracks present in the pellet. Calculations were carried out to quantify time required for water penetration into a pellet. This time is of significant relevance for interpretation of short-time leaching experiments. Subsequently, the mass (moles) of radionuclides released from the crack surfaces and accumulated in the external reservoir could be computed as a function of time. This allows direct comparison with data available from laboratory leaching experiments.

Modelling results indicate that wetting of crack surfaces present in a SNF pellet may quantitatively explain radionuclide release patterns observed in laboratory leaching experiments performed on SNF pellets or their fragments. Specifically, the time scales of initial fast radionuclide release and the later slower releases are on a scale comparable with the times calculated for complete saturation of the “macro” and “micro” crack systems. The proposed model is capable of capturing these general trends in a semi-quantitative manner as it is shown in Figure 10. In order to increase the predictive capacity of the model further data on the statistical properties of cracks and on the distribution of radionuclides on the surfaces of cracks is needed.



**Figure 10:** Experimental laboratory data (González-Robles, PhD Thesis 2011) on radionuclide release from SNF pellet (normalized to unity) and model prediction assuming that 40 % of total tracer mass is initially associated with “macro cracks”.

**CTM-UPC** developed a semi-empirical model based on the experimental fitting by using three different first-order kinetic equations corresponding to different parts of the fuel: gap, cracks, external or internal grain boundaries, rim structure, and finally grains (matrix). This model was applied to different spent fuel samples: segmented *vs.* powder, 6 different burn-ups (42, 48, 52, 53, 54 and 60 GWd/t<sub>HM</sub>) and PWR fuel *vs.* BWR fuel. The behaviour of the following fission products was studied: Cs, Sr, Tc, Mo and Rb; moreover, U was also studied as matrix dissolution indicator.

Normalized dissolution rate of caesium is always higher than for the rest of measured radionuclides, especially when looking at results obtained from cladded samples. For powder samples this trend is also observed except for Rb, which shows more similar release rates. This is also true for Sr rates obtained for PWR samples. This is assumed to be due to the degree of matrix segregation. The higher the fraction of a radionuclide is segregated, the higher is its release because it is assumed that this release is practically instantaneous as soon as the water gets in contact with them.

In general, the percentage of radionuclide released is higher for powdered than for cladded samples, except in the case of Cs in PWR samples. It can be assumed that for Cs its content in gaps and fractures is similar than its content in grain boundaries and micro fissures, which is not the case for the rest of radionuclides, that show greater content in grain boundaries due to their lower migration once segregated from the matrix.

When comparing PWR and BWR fuel samples, clear differences in terms of normalized release rates are observed, being the values obtained for PWR fuels larger than for BWR fuels. Once again, such differences become more significant as we look to Cs and Rb and to some extent to Sr, which are the ones expected to be segregated from the matrix in higher degrees. When looking at the percentages dissolved, they seem to be higher for pellets (higher availability to water) than for powders, except for Cs and PWR fuels.

## Conclusions

- Calculations of the burn-up and decay history, of the rim zone burn-up, of the rim zone thickness and of the porosity are well established.
- Speciation of  $^{14}\text{C}$  in SNF is not yet resolved.
- Modelling of temperature and radionuclide diffusion can explain element profiles determined by LA-ICP-MS measurements.
- After discharge from the reactor, diffusion processes of volatiles do not play any role due to the negligible diffusion coefficients at interim storage temperatures.
- Water Saturation Model indicates that penetration of water (wetting of crack surfaces present in a SNF pellet) and subsequent diffusion processes quantitatively explain radionuclide release patterns observed in laboratory leaching experiments performed on SNF pellets.
- To increase the predictive capacity of the model further data on the statistical properties of cracks and on the distribution of radionuclides on the surfaces of cracks is needed.
- Semi-empirical model based on the experimental fitting allows to identify the fission product location in different parts of the fuel.
- Normalized dissolution rate of caesium is always higher than rates of Tc, Rb, Mo and Sr.
- PWR release rates are higher than those of BWR.

### 3.5. WP5 “Knowledge, reporting and training: Documentation, State-of-the-Art report, up-date. Dissemination and Training”

#### Objectives

WP5 is focused in three different aspects related with the dissemination of knowledge which are summarized in the following objectives of this task:

- (1) To provide access to all scientific-technical results for all the interested parties (scientific community). This means the organization and coordination of several dissemination activities.
- (2) To elaborate a state of the art report which should be annually updated and which at the end of the project must include the improvements from the project.
- (3) To organize training activities for the next generation of spent nuclear fuel specialists.

## Description of work performed in the project

Most of the WP5 work has been performed by AMPHOS21 with the support of KIT-INE. The rest Beneficiaries have collaborate mainly with the state of the art report and disseminating their results either in conferences or peer review journals.

A brief description of the tasks done during the three years of FIRST-Nuclides in order to achieve each of the three WP5 objectives is provided below.

### Dissemination activities

A high amount of dissemination activities has been performed and different ways of disseminate have been exploited. Table 2 summarizes the resources used in order to disseminate the generated knowledge in FIRST-Nuclides project.

**Table 2:** Dissemination tools used during the FIRST-Nuclides project.

<i>Dissemination action</i>	<i>Description</i>
<i>Generic poster</i>	A generic poster of the project which is regularly updated. The last version of the poster is available at the project webpage. This final version it includes a summary of the obtained results.
<i>Newsletters</i>	Three newsletters have been edited during the project (one per year). The main objective of the newsletter is to communicate to the broader community the progresses achieved every year, as well as presenting the last news and the coming events related with the project. All of them are available at the project webpage.
<i>Deliverables</i>	Each WP has several deliverables where it is compiled all the information related with the work done. Most of them have been updated at least one time. Those reports are available at the project webpage.
<i>Web portal</i>	A web portal has been created and constantly updated to inform both the consortium members and the broader community. For this reason, the website consists of two different parts; one public and another one restricted to the consortium members. All deliverables, proceedings, newsletters, etc. are available at the public website. The intranet is a way of internal communication.
<i>Annual workshops</i>	A workshop has been organized once a year by the coordination team and the host institutions (AMPHOS21, MTA-EK, SCK·CEN and KIT/INE). 1 kick-off meeting and 3 annual workshops took place during the project life. The outcome of those workshops is the edition of the proceedings.
<i>Presentation at international conferences</i>	Both the coordinator of the project and members of the beneficiary institutions have attended several international conferences giving oral presentations either on the project as a whole or specific work carried out in the frame of the project
<i>Peer review publications</i>	The coordination team encourages all beneficiaries to publish their work in peer review journals such as Journal of Nuclear Materials. During the last workshop, the WP leaders have planned to publishing 4 papers which would cover a review of the scientific achievements within all the technical WP.

More information on dissemination activities can be found in *Section 4.3*.

### State of the Art report

The first version of the State-of-the-Art was reported at the beginning of the project and it provides a view on basic information related with the spent nuclear fuel and the IRF studies done in the past decades. A description of different type of spent fuels, processes induced during SF irradiation or the modelling tools used for fuel performance are included in the basic information section. The second part of the report includes a summary of more than 100 published experiments which have used different samples, experimental techniques, experimental conditions, type of solutions, etc. This report has been updated after the second year of the project.

The third version of this report is composed of two different parts: (1) State of the Art report and (2) IRF database. This last version of the report not only contains fuel information and previous IRF studies but it also includes a summary of the results obtained in the frame of the FIRST-Nuclides project.

The IRF database has been developed in excel format during the third year of the project and compiles both the previous published experiments and the ones done during the project. Four different types of spreadsheets can be found in the database:

- I. A single spreadsheet consisting on a *table* that contains a *summary* of the relevant parameters and results of each sample<sup>3</sup> included in the database.
- II. An *interactive* spreadsheet where the user can select the data to be *plotted* (selection of both the “x” and “y” axis; e.g. Cs IRF vs. burn-up). Data result of the experiments carried out in the frame of FISRT-Nuclides project are shown in a different color in order to see if they are in agreement with previous literature data. In addition, an interactive table allows the user to select which of the parameters from the summary table must be shown in each column. This option may be useful for understanding the presence of an outlier point.
- III. Several spreadsheets, one per publication, providing *detailed information* on the samples, materials, experimental conditions, analysis techniques, results, uncertainties, etc.
- IV. A single spreadsheet with the list of *references* used in this database

More information of this database can be found in the corresponding deliverable (D5.1, 3<sup>rd</sup> version) which is available at the project website.

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<sup>3</sup> The table is not organized by author study but by the studied samples. From each of the samples, several parameters describing its characteristics, experimental conditions and obtained results are provided.

### Training activities

The following actions have been done with the aim of providing training and education to young scientist.

- Lab-Exchange meeting: 17 participants from four partner institutions (KIT-INE, JRC-ITU, STUDSVIK and PSI) have attended at this meeting organized by PSI on March 2013. The aim of the meeting was to serve as a discussion forum for experimentalists of the project to discuss analytical and technical experimental details.
- Organization of a training course: KIT-INE, JRC-ITU and AMPHOS21 organized a training course (9<sup>th</sup> - 10<sup>th</sup> July 2013, Karlsruhe). The course focused on the radionuclide release from LWR spent nuclear fuel (SNF), experimental methods available to quantify radionuclide release and relevant characteristics of LWR SNF. Twelve participants attended the course, from six project partners and three associated groups.
- Training mobility measures: The project has allocated resources for three training mobility measures, which consider the visit of one student from a participant institution to a different partner organization.
  - Albert Martínez (PhD student from UPC/CTM) was hosted at KIT-INE to improve experimental abilities in working in an inert gas glovebox, gas sampling in a shielded box (gas-MS) or handling of 0.1 w.t.% <sup>238</sup>Pu doped UO<sub>2</sub>(s) pellets, among other technical issues.
  - Péter Szabó (PhD student from EK-MTA) was also at KIT-INE, being trained on sample preparation for <sup>14</sup>C analyses (LSC), uranium analyses, introduction to geochemical modeling, laser fluorescence measurements and other skills of interest for his scientific development within the project.
  - David García (PhD student from AMPHOS21) has benefited from a training mobility also at KIT-INE on development of sorption models.
- 7 travel grants to attend the Final workshop: 7 PhD students have taken advantage of the travel grants. The grant covered travelling, accommodation and workshop fee expenses in order young people have the opportunity of attending the final workshop of the project.



## 4. Potential impact and the main dissemination activities

### 4.1. Potential impacts

The potential project impacts are evaluated according to the degree of accomplishing the initially defined objectives:

- Determination and quantification of the “instant release fraction (IRF)” of fission gases, caesium, iodine ( $^{129}\text{I}$ ),  $^{14}\text{C}$  compounds, selenium ( $^{79}\text{Se}$ ), technetium  $^{99}\text{Tc}$  and tin ( $^{126}\text{Sn}$ ) from the gap of fuel pellets and from the  $\text{UO}_2$  grain boundaries.
- Determination of the dependency of the IRF on power plant operational parameters (burn-up history, linear power and fuel temperature history, including possibly ramping processes, and storage time).
- Determination of the transition between instant/fast release and the significantly slower release processes due to radiolytically driven  $\text{UO}_2$  matrix corrosion.
- Quantification and characterization of exchange processes along the grain boundaries by conceptual and numerical modelling.
- Dissemination of the achieved knowledge.
- Training and education of young scientists.
- Direct applications of the results to safety assessments were not foreseen within the project.

#### Potential Impacts

- IRF of fission gases, Cs, I,  $^{14}\text{C}$  compounds, selenium ( $^{79}\text{Se}$ ),  $^{99}\text{Tc}$ ,  $^{126}\text{Sn}$ , etc. from gap and  $\text{UO}_2$  grain boundaries:
  - The investigations cover 12 different types of high burn-up LWR fuel, cumulative IRF measurements at 45 different time steps, for up to 3 sample preparations, and up to 20 isotopes. For Cs and I, the available database has been improved significantly. The data could be related to dissolution based fission gas releases. Differences and similarities from gap and grain boundary release processes have been shown. Relations have been derived to be applied in safety analyses.
- Dependency of the IRF on power plant operational parameters:
  - In comparison to previous studies, the clearance of the fuel owners was obtained to publish the power plant specific data. This fact allowed relating the observed cumulative IRF and IRF rates to data such as burn-up, power rating and calculated fission gas releases. The relations are available to be applied in safety analyses.

Releases from non-standard SNFs were also investigated, such as from extremely high burn-up TRISO fuel and from leaking VVER fuel assemblies.

- Other elements:
  - The relation between the observed IRF of redox sensitive elements and elements incorporated in the  $\text{UO}_2$  matrix demonstrated clearly the effect of the leaching conditions. It is concluded that for the elements Tc, Sr, U and perhaps Se experiments should be performed under realistic environmental conditions.
- Transition between IRF and the significantly slower release processes due to  $\text{UO}_2$  matrix corrosion:
  - A way to define this transition was the application of different samples, sample sizes and sample preparations comprising pellets, segments, slices, fragments, powders, and cladding with attached fuel. The partners agreed upon a standard leachant which allowed better comparisons of results generated by the different labs. It could be shown that the fast / instant release rates of Cs and I reached after about 0.5 to one year the release rates from the matrix.
- Modelling:
  - Exchange processes along the grain boundaries were modelled. This includes the penetration of water into the fuel sample under investigation and the subsequent migration of radionuclides along the wetted fractures and grain boundaries. Modelling also provided significant input for the definition of state of the fuel samples under investigation.
- Dissemination:
  - Up to now, 166 reports, publications and presentations at conferences document the achievements and results of FIRST-Nuclides.
- Training and education of young scientists:
  - This aim was achieved by a Lab exchange meeting, a training course at KIT-INE and by mobility measures for 3 PhD students.
- Application to SC:
  - Until recently, the IRF of spent fuel was often related to its burn-up. With more data becoming available for high burn-up 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 burn-up.

- A series of relations have been derived for estimating the IRF of different fuel properties. These relations are required for safety analysis of the disposal of a multitude of different SNFs in one single disposal.
- 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 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<sub>2</sub> 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 burn-up 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.
- Further investigations:
  - 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 <sup>79</sup>Se, and Pd, or activation products <sup>36</sup>Cl and <sup>14</sup>C.
    - Clear correlations for predicting the IRF from nuclear power plant operational parameters (power rates, temperatures, FGR).
    - An improved 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 proposed to keep the experimental set-up and the materials for an interim period and to apply for a new project in the future.

## 4.2. Societal impacts

Following societal unities are affected by the outcome of CP FIRST-Nuclides. These are the implementers and regulators of a SNF disposal which will use the obtained data and knowledge for direct application in safety analyses and the improvement of the knowledge basis described in the Safety Case. This fact has also impact on general public as the trust into the argumentation by implementers and regulators is improved due to the broader knowledge of the processes in the SNF, release processes and chemical speciation of the relevant elements. Concerning liaisons with socio-technical groups, meetings with the InSOTEC project (July 2013 at Karlsruhe and at the 2<sup>nd</sup> AW at Antwerp) took place, as well as with persons involved in the German ENTRIA project.

A close trustworthy scientific cooperation has been developed between the different labs and organizations which work under specific national regulations. This cooperation includes not only the Beneficiaries, but also the Associated Groups in Europe and USA. It includes the research organizations, and consultants but also the waste management organizations, utilities, and the producer of the fuel.

The process of the EURATOM funded cooperation as demonstrated by CP FIRST-Nuclides gained high attention outside of Europe, where, for example in USA, a close and trustworthy cooperation between different laboratories could not be established as competitive considerations would dominate (statement by a representative of an Associated Group from USA).

## 4.3. Dissemination activities

One of the workpackages of the project was mainly focused on the dissemination of the generated knowledge to the interested parties. Thus, several activities and actions have been conducted in order to achieve this objective.

The main dissemination tool of FIRST-Nuclides is its webpage ([www.firstnuclides.eu](http://www.firstnuclides.eu)) where all the information related with the project as well as the produced documentation are available (e.g. deliverables, proceedings, newsletters, generic poster...). Private information for the consortium can be exchanged by means of the intranet.

The results obtained during the 3 years of the project has been regularly presented at the annual project workshops and published at the corresponding proceedings (available at the project website). Table 3 summarizes the workshops organized in the frame of FIRST-Nuclides.

**Table 3:** Workshops organized in the frame of FIRST-Nuclides project.

<i>Workshop</i>	<i>Date</i>	<i>Place</i>	<i>Hosting institution</i>
Kick-off meeting	9-10 <sup>th</sup> February 2012	Barcelona (Spain)	AMPHOS21
1 <sup>st</sup> Annual Workshop	9-11 <sup>th</sup> October 2012	Budapest (Hungary)	MTA EK
2 <sup>nd</sup> Annual Workshop	5-7 <sup>th</sup> November 2013	Antwerp (Belgium)	SCK·CEN
Final Workshop	1 <sup>st</sup> -2 <sup>nd</sup> September 2014	Karlsruhe (Germany)	KIT-INE

A list of all the scientific and technical contributions submitted and published in the project workshop proceedings is provided in *Annex A*. The active participation of the beneficiaries is highlighted as each partner provided in average two S+T contributions per year. Most of the contributions presented at the 1<sup>st</sup> Annual Workshop were related with WP1 “Samples and tools” as this WP delivered the basic data used for the remaining WP activities. In contrary, contributions to WP3 reached the highest level with respect to number and quality at the Final Workshop. In the last project year, WP3 was the biggest task in terms of beneficiaries and person-months of the project.

FIRST-Nuclides results were not only presented in the frame of the project circle and its collaborators. Most of the partners actively reported their results and/or the whole project to the international scientific community by means of oral presentations in conferences, contributions to conference proceedings, publications in peer -reviewed journals, etc (see Figure 11).

The beneficiaries actively presented the newly generated knowledge at many conferences. Table 4 lists the oral conference contributions given by the coordinator presenting the project as a whole and by the beneficiary institutions presenting details of their research carried out in the frame of FIRST-Nuclides.

**Table 4:** List of oral presentation given by the coordinator and beneficiaries at international conferences presenting FIRST-Nuclides.

Authors	Title	Conference
Curtius, H.	HTR Spent Fuel-Effects of irradiation to the microstructure.	Spent Fuel Workshop, Avignon, France, 18 <sup>th</sup> - 19 <sup>th</sup> April 2012.
Kienzler, B., Montoya, V., Metz, V.	Euratom 7 <sup>th</sup> Framework Programme Collaborative Project (2012) “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel.”	Spent Fuel Workshop, Avignon, France, 18 <sup>th</sup> - 19 <sup>th</sup> April 2012.
Kienzler, B., Valls, A., Metz, V.	Euratom 7 <sup>th</sup> Framework Programme Collaborative Project (2012) “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel.”	4 <sup>th</sup> International ATALANTE Conference on Nuclear Chemistry for Sustainable Fuel Cycles, Montpellier, France, 2 <sup>nd</sup> – 7 <sup>th</sup> September 2012.
Kienzler, B., Valls, A., Metz, V.	Euratom 7 <sup>th</sup> Framework Programme Collaborative Project (2012) “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel.”	EURADISS 2012, Montpellier, France, 25 <sup>th</sup> – 26 <sup>th</sup> October 2012.
Roth, O., Low, J., Granfors, M., Spahiu, K.	Effects Of Matrix Composition on Instant Release Fractions from High Burn-Up Nuclear Fuel.	MRS Fall Meeting - Symposium LL – Scientific Basis for Nuclear Waste Management XXXVI, Boston, USA, 25 <sup>th</sup> – 30 <sup>th</sup> November 2012.
Aldave de las Heras, L., Olszewski, G., Sandow, M., Serrano-Purroy, D., Van Winckel, S., Skwarzec, B., Klemmens, P.	Determination of <sup>90</sup> Sr at Ultratrace Levels Using an Automated Chromatographic System Coupled to High Resolution ICPMS.	European Winter Conference on Plasma Spectrochemistry, Krakow, Poland, 10 <sup>th</sup> – 15 <sup>th</sup> February 2013.
Kienzler, B., Metz, V., González-Robles, E., Bohnert, E., Loida, A., Müller, N.	Recent Activities on HLW and Spent Nuclear Fuel Related Research at KIT-INE	International Workshops ABC-Salt(III), Santa Fe, USA, 14 <sup>th</sup> – 16 <sup>th</sup> April 2013.
Kienzler, B., González-Robles, E., Metz, V., Valls, A., Duro, L.	Euratom 7 <sup>th</sup> Framework Programme Collaborative Project (2012) “Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel.”	14 <sup>th</sup> International High-Level Radioactive Waste Management Conference (14 <sup>th</sup> IHLRPMC), Albuquerque, USA, 28 <sup>th</sup> April – 2 <sup>nd</sup> May 2013.



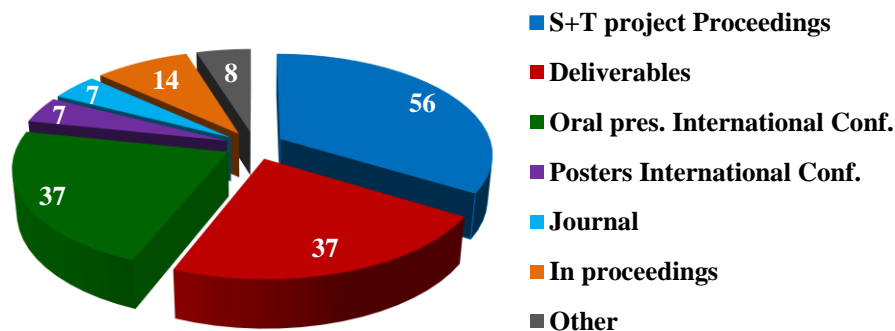
Authors	Title	Conference
Puranen, A., Granfors, M., Roth, O.	The Radial Distribution of Potential Rapid Release Radionuclides in Irradiated Nuclear Fuel.	E-MRS 2013 Spring Meeting, Strasbourg, France, 27 <sup>th</sup> – 31 <sup>st</sup> May 2013.
Aldave de las Heras, L., Sandow, M., Serrano-Purroy, D., Sureda Pastor, R., Van Winckel, S., Glatz, J.P.	Speciation of <sup>79</sup> Se at Ultratrace Levels Using an Automated Chromatographic System Coupled to High Resolution ICPMS, Application to Nuclear Spent Fuel Corrosion Studies.	14 <sup>th</sup> International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere, Brighton, United Kingdom, 8 <sup>th</sup> – 13 <sup>th</sup> September 2013.
Traboulsi, A., Vandenborre, J., Blain, G., Barbet, J., Fattahi, M.	Impact of Water Radiolysis on Uranium Dioxide Corrosion.	Migration 2013, Brighton, United Kingdom, 8 <sup>th</sup> – 13 <sup>th</sup> September 2013.
Casas, I., Espriu, A., Martínez-Esparza, A., Serrano-Purroy, D., de Pablo, J.	IRF Modelling from High Burn-Up Spent Nuclear Fuel Leaching Experiments.	MRS 2013, Barcelona, Spain, 29 <sup>th</sup> September – 3 <sup>rd</sup> October 2013.
González-Robles, E., Bohnert, E., Metz, V., Wege, D.H., Papaioannou, D., Kienzler, B.	Physical Characterisation and Calculation of the Initial and Boundary Conditions of a Commercial UO <sub>2</sub> Spent Nuclear Fuel Regarding the Radionuclide Release.	MRS 2013, Barcelona, Spain, 29 <sup>th</sup> September – 3 <sup>rd</sup> October 2013.
Kienzler, B., Loida, A., González-Robles, E., Metz, V.	Rapid Radionuclide Release: Effect of Solution Compositions.	MRS 2013, Barcelona, Spain, 29 <sup>th</sup> September – 3 <sup>rd</sup> October 2013.
Pekala, M., Idiart, A., Duro, L., Riba, O.	Modelling of Spent Nuclear Fuel Saturation with Water: Implications for the Instant/Fast Release Fraction.	MRS 2013, Barcelona, Spain, 29 <sup>th</sup> September – 3 <sup>rd</sup> October 2013.
Roth, O., Low, J., Spahiu, K.	Effects of Matrix Composition and Sample Preparation on Instant Release Fractions from High Burn-Up Nuclear Fuel	MRS 2013, Barcelona, Spain, 29 <sup>th</sup> September – 3 <sup>rd</sup> October 2013.
Kienzler, B.	FIRST-Nuclides: Investigation of Fast/ Instant Radionuclide Release from High Burn-Up Spent Nuclear Fuel and Its Impact on the Performance of Geological Repositories	EURADWASTE, Volnius, Lithuania, 14 <sup>th</sup> – 17 <sup>th</sup> October 2013.
Slonszki, E., Hózer, Z., Szabó, P.	Activity Release from the Spent Nuclear Fuels in Deep Geological Repository – The FIRST-Nuclides Project (Aktivitás-kikerülés a fűtőelemekből mélygeológiai tárolóban - A FIRST-Nuclides projekt - in Hungarian).	MNT Nucleáris Technikai Szimpózium, Budapest, Hungary, 5 <sup>th</sup> – 6 <sup>th</sup> December 2013.

Authors	Title	Conference
Cui, D., Low, J., Roth, O., Puranen, A., Low, J., Askeljung, C., Granfors, M., Spahiu, K.	On Radionuclide Release from Spent Fuel.	REDUPP FWS, Stockholm, Sweden, 18 <sup>th</sup> – 21 <sup>st</sup> February 2014.
Marchetti, I	Surface Species in UO <sub>2</sub> Oxidative Dissolution.	REDUPP FWS, Stockholm, Sweden, 18 <sup>th</sup> – 21 <sup>st</sup> February 2014.
Puranen, A., Granfors, M., Roth, O., Spahiu, K.	The Radial Distribution of Potential Rapid Release Radionuclides in Irradiated Nuclear Fuel.	REDUPP FWS, Stockholm, Sweden, 18 <sup>th</sup> – 21 <sup>st</sup> February 2014.
Roth, O., Granfors, M., Low, J., Spahiu, K., Nilsson, K., Jonsson, M.	Effects of Matrix Composition on Dissolution of High Burn-Up Nuclear Fuel.	REDUPP FWS, Stockholm, Sweden, 18 <sup>th</sup> – 21 <sup>st</sup> February 2014.
Kienzler, B., González-Robles, E., Metz, V.	FIRST-Nuclides: Selected Results.	IGD-TP Geodisposal 2014, Manchester, United Kingdom, 24 <sup>th</sup> – 26 <sup>th</sup> June 2014.
Bulgheroni, A	Surface Characterisation of Oxidised UO <sub>2</sub> by SIMS.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Curti, E., Froideval-Zumbiehl, A., Martin, M., Bullemer, A., Puranen, A., Jädernäs, D., Roth, O., Grolimund, D., Borca, C.N.	X-Ray Spectroscopic Evidence on the Chemical State of <sup>79</sup> Se in High Burn-Up UO <sub>2</sub> Spent Fuel.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Curtius, H.	Leaching of Spent UO <sub>2</sub> TRISO Coated Particles- Instant Radionuclide Release Fraction and Microstructure Evolution.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
González-Robles, E., Lagos, M., Bohnert, E., Müller, N., Herm, M., Metz, V., Kienzler, B.	Fast Radionuclide Release from a PWR Fuel Rod Segment (50.4 GWd/t <sub>HM</sub> ) Under Hydrogen Overpressure	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Kienzler, B.	Results of the 7 <sup>th</sup> FP Collaborative Project FIRST-Nuclides.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Puranen, A., Granfors, M., Roth, O.	Aqueous Leaching of <sup>79</sup> Se from Spent Nuclear Fuel.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Roth, O., Puranen, A., Granfors, M., Spahiu, K., Jonsson, M., Nilsson, K.	Matrix Dissolution and Instant Release from Spent Nuclear Fuel Containing Dopants/Additives.	27 <sup>th</sup> Spent Fuel Workshop, Karlsruhe, Germany, 3 <sup>rd</sup> – 5 <sup>th</sup> September 2014.
Kienzler, B., González-Robles, E.	State-Of-The-Art on Instant Release of Fission Products from Spent UO <sub>2</sub> Fuel	15 <sup>th</sup> ICEM Brussels, 9 <sup>th</sup> – 12 <sup>th</sup> September 2013.

Authors	Title	Conference
Curtius, H., Deismann, G., Bosbach, D.	Corrosion of Spent Fuels from Research and Prototype Reactors under Conditions Relevant to Geological Disposal.	DAEF, Cologne, Germany, 24 <sup>th</sup> – 26 <sup>th</sup> September 2014.
Kienzler, B.	FIRST-Nuclides: Outcome, Open Questions and Steps Forward.	IGD-TP Exchange Forum n°5, Kalmar, Sweden, 28 <sup>th</sup> – 30 <sup>th</sup> October 2014.
González-Robles, E., Fuß, M., Bohnert, E., Müller, N., Herm, M., Metz, V., Kienzler, B.	Study of the Release of the Fission Gases (Kr and Xe) and the Fission Products (I and Cs) under Anoxic Conditions in Bicarbonate Water	MRS 2014, Boston, USA, 30 <sup>th</sup> November - 4 <sup>th</sup> December 2014.

As explained above, the dissemination of the detailed FIRST-Nuclides results was mainly done via the project workshop proceedings and project reports, i.e. deliverable, (see dark blue and red portions in Figure 11) and via oral presentations at international conferences (see green portion in Figure 11). Other type of publications includes the following dissemination activities (see grey portion in Figure 11):

- Publication of a half a page notice at the Quarterly, Research Review Policy Focus of the Parliament Magazine, (Issue 387, 31 March 2014).
- Publication of modelling results in the *iMaGe platform* (blog, 21/05/14) which stands for interfacing Multiphysics and Geochemistry and aims to launching a platform to foster the development of software solutions for communicating geosciences modelling codes.
- Internal reports of the beneficiaries.
- Communication to the IGD-TP.



**Figure 11:** Dissemination material generated in the frame of FIRST-Nuclides classified by type of dissemination tool.

An important dissemination route needs to be highlighted. It was decided to disseminate the FIRST-Nuclides outcome by publishing four papers in a peer review journal which will cover the scientific achievements within the technical WPs. The list of the four papers and the leaders of submitting each paper are listed below.

- Summary of the project and application to the Safety Case including the reflections of the End-Users (Bernhard Kienzler and Volker Metz)
- Fission gas and dissolution based release (Karel Lemmens)
- Grain boundary effects (Dettef Wegen)
- Modelling (Lara Duro and Joan de Pablo)

These papers should be published in a special section of an issue of the Journal of Nuclear Materials in mid-2015. Due to delay due to the defects of an experimental system and long-lasting repair measures at JRC-ITU, the draft on Grain Boundary Effects will be available later.

## 5. Address of the project public website

The project website was set-up at the beginning of the project (January 2012) and has been regularly updated by AMPHOS21. The website has the objective of being a platform to disseminate information of the project as well as the obtained results.

The web is divided in two different parts, one of them opened to the public and a second one restricted to the members of the consortium (intranet).

The address of the webpage is [www.firstnuclides.eu](http://www.firstnuclides.eu) (Figure 12)



**Figure 12:** FIRST-Nuclides website (home page on the public part).

## ***6. Use and dissemination of foreground***

This section aims on showing the dissemination measures done during the project and the ones expected once the project has been finished. The complete list of published journals and dissemination activities has been uploaded in the on-line system Participant Portal

No patent applications have been made within FIRST-Nuclides, thus, Exploitable Foreground is not reported.



## 7. Report on societal implications

The following questionnaire is filled in in order to report in societal implications.

<b>A General Information</b> <i>(completed automatically when Grant Agreement number is entered).</i>	
Grant Agreement Number:	FP7-295722
Title of Project:	Fast / Instant Release of safety relevant radionuclides from spent nuclear fuel (FIRST-Nuclides)
Name and Title of Coordinator:	Dr. Bernhard Kienzler
<b>B Ethics</b>	
<b>1. Did your project undergo an Ethics Review (and/or Screening)?</b> <ul style="list-style-type: none"> <li>If Yes: have you described the progress of compliance with the relevant Ethics Review/Screening Requirements in the frame of the periodic/final project reports?</li> </ul> <p>Special Reminder: the progress of compliance with the Ethics Review/Screening Requirements should be described in the Period/Final Project Reports under the Section 3.2.2 'Work Progress and Achievements'</p>	No
<b>2. Please indicate whether your project involved any of the following issues (tick box) :</b>	
<b>RESEARCH ON HUMANS</b>	
• Did the project involve children?	
• Did the project involve patients?	
• Did the project involve persons not able to give consent?	
• Did the project involve adult healthy volunteers?	
• Did the project involve Human genetic material?	
• Did the project involve Human biological samples?	
• Did the project involve Human data collection?	
<b>RESEARCH ON HUMAN EMBRYO/FOETUS</b>	
• Did the project involve Human Embryos?	
• Did the project involve Human Foetal Tissue / Cells?	
• Did the project involve Human Embryonic Stem Cells (hESCs)?	
• Did the project on human Embryonic Stem Cells involve cells in culture?	
• Did the project on human Embryonic Stem Cells involve the derivation of cells from Embryos?	
<b>PRIVACY</b>	
• Did the project involve processing of genetic information or personal data (eg. health, sexual lifestyle, ethnicity, political opinion, religious or philosophical conviction)?	
• Did the project involve tracking the location or observation of people?	
<b>RESEARCH ON ANIMALS</b>	
• Did the project involve research on animals?	
• Were those animals transgenic small laboratory animals?	
• Were those animals transgenic farm animals?	
• Were those animals cloned farm animals?	
• Were those animals non-human primates?	
<b>RESEARCH INVOLVING DEVELOPING COUNTRIES</b>	

• Did the project involve the use of local resources (genetic, animal, plant etc)?	
• Was the project of benefit to local community (capacity building, access to healthcare, education etc)?	
<b>DUAL USE</b>	
• Research having direct military use	No
• Research having the potential for terrorist abuse	No

## C Workforce Statistics

**3. Workforce statistics for the project: Please indicate in the table below the number of people who worked on the project (on a headcount basis).**

Type of Position	Number of Women	Number of Men
Scientific Coordinator	1	1
Work package leaders	1	4
Experienced researchers (i.e. PhD holders)	14	32
PhD Students	3	4
Other	23	62

<b>4. How many additional researchers (in companies and universities) were recruited specifically for this project?</b>	<b>1</b>
Of which, indicate the number of men:	1

## D Gender Aspects

5. Did you carry out specific Gender Equality Actions under the project? ☐ Yes ☒ No

6. Which of the following actions did you carry out and how effective were they?

	Not at all effective	Very effective
<input type="checkbox"/> Design and implement an equal opportunity policy	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>
<input type="checkbox"/> Set targets to achieve a gender balance in the workforce	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>
<input type="checkbox"/> Organise conferences and workshops on gender	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>
<input type="checkbox"/> Actions to improve work-life balance	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>	<input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/> <input type="radio"/>
<input type="radio"/> Other: <input type="text"/>		

7. Was there a gender dimension associated with the research content – i.e. wherever people were the focus of the research as, for example, consumers, users, patients or in trials, was the issue of gender considered and addressed?

☐ Yes- please specify

☒ No

## E Synergies with Science Education

8. Did your project involve working with students and/or school pupils (e.g. open days, participation in science festivals and events, prizes/competitions or joint projects)?

☐ Yes- please specify

☒ No

9. Did the project generate any science education material (e.g. kits, websites, explanatory booklets, DVDs)?

☐ Yes- please specify

☒ No

## F Interdisciplinarity

10. Which disciplines (see list below) are involved in your project?

☐ Main discipline<sup>4</sup>: 1.3

☐ Associated discipline<sup>4</sup>: 1.2

☐ Associated discipline<sup>4</sup>: 1.4

## G Engaging with Civil society and policy makers

11a Did your project engage with societal actors beyond the research community? (if 'No', go to Question 14)

☒ Yes ☐ No

11b If yes, did you engage with citizens (citizens' panels / juries) or organised civil society (NGOs, patients' groups etc.)?

☒ No

☐ Yes- in determining what research should be performed

☐ Yes - in implementing the research

☐ Yes, in communicating /disseminating / using the results of the project

<sup>4</sup> Insert number from list below (Frascati Manual).

<b>11c In doing so, did your project involve actors whose role is mainly to organise the dialogue with citizens and organised civil society (e.g. professional mediator; communication company, science museums)?</b>		<input type="radio"/> <input checked="" type="radio"/>	Yes No	
<b>12. Did you engage with government / public bodies or policy makers (including international organisations)</b>				
<input type="radio"/> No <input type="radio"/> Yes- in framing the research agenda <input checked="" type="radio"/> Yes - in implementing the research agenda <input checked="" type="radio"/> Yes, in communicating /disseminating / using the results of the project				
<b>13a Will the project generate outputs (expertise or scientific advice) which could be used by policy makers?</b>				
<input type="radio"/> Yes – as a <b>primary</b> objective (please indicate areas below- multiple answers possible) <input checked="" type="radio"/> Yes – as a <b>secondary</b> objective (please indicate areas below - multiple answer possible) <input type="radio"/> No				
<b>13b If Yes, in which fields?</b>				
Agriculture Audiovisual and Media Budget Competition Consumers Culture Customs Development Economic and Monetary Affairs Education, Training, Youth Employment and Social Affairs	<input checked="" type="radio"/> <input checked="" type="radio"/>	Energy Enlargement Enterprise Environment External Relations External Trade Fisheries and Maritime Affairs Food Safety Foreign and Security Policy Fraud Humanitarian aid	<input checked="" type="radio"/>	Human rights Information Society Institutional affairs Internal Market Justice, freedom and security Public Health Regional Policy Research and Innovation Space Taxation Transport

**13c If Yes, at which level?**

- ☐ Local / regional levels  
☐ National level  
☐ European level  
☒ International level

**H Use and dissemination**
**14. How many Articles were published/accepted for publication in peer-reviewed journals?**
**7**
**To how many of these is open access<sup>5</sup> provided?**
**7**
**How many of these are published in open access journals?**
**6**
**How many of these are published in open repositories?**
**1**
**To how many of these is open access not provided?**
**Please check all applicable reasons for not providing open access:**

- ☐ publisher's licensing agreement would not permit publishing in a repository  
☐ no suitable repository available  
☐ no suitable open access journal available  
☒ no funds available to publish in an open access journal  
☐ lack of time and resources  
☐ lack of information on open access  
☐ other<sup>6</sup>: .....

**15. How many new patent applications ('priority filings') have been made?**  
*("Technologically unique": multiple applications for the same invention in different jurisdictions should be counted as just one application of grant).*
**0**
**16. Indicate how many of the following Intellectual Property Rights were applied for (give number in each box).**

Trademark

**0**

Registered design

**0**

Other

**0**
**17. How many spin-off companies were created / are planned as a direct result of the project?**
**0**
*Indicate the approximate number of additional jobs in these companies:*
**18. Please indicate whether your project has a potential impact on employment, in comparison with the situation before your project:**

- |   |   |
|---|---|
| <input type="checkbox"/> Increase in employment, or                       | <input type="checkbox"/> In small & medium-sized enterprises                        |
| <input type="checkbox"/> Safeguard employment, or                         | <input type="checkbox"/> In large companies   |
| <input type="checkbox"/> Decrease in employment,                          | <input checked="" type="checkbox"/> None of the above / not relevant to the project |
| <input type="checkbox"/> Difficult to estimate / not possible to quantify |   |

<sup>5</sup> Open Access is defined as free of charge access for anyone via Internet.

<sup>6</sup> For instance: classification for security project.

<b>19. For your project partnership please estimate the employment effect resulting directly from your participation in Full Time Equivalent (FTE = one person working fulltime for a year) jobs:</b>	<i>Indicate figure:</i>		
Difficult to estimate / not possible to quantify	<input checked="" type="checkbox"/>		
<b>I Media and Communication to the general public</b>			
<b>20. As part of the project, were any of the beneficiaries professionals in communication or media relations?</b> <div style="display: flex; justify-content: space-around;"> <span><input type="radio"/> Yes</span> <span><input checked="" type="radio"/> No</span> </div>			
<b>21. As part of the project, have any beneficiaries received professional media / communication training / advice to improve communication with the general public?</b> <div style="display: flex; justify-content: space-around;"> <span><input type="radio"/> Yes</span> <span><input checked="" type="radio"/> No</span> </div>			
<b>22 Which of the following have been used to communicate information about your project to the general public, or have resulted from your project?</b> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; vertical-align: top;"> <input checked="" type="checkbox"/> Press Release  <input type="checkbox"/> Media briefing  <input type="checkbox"/> TV coverage / report  <input type="checkbox"/> Radio coverage / report  <input checked="" type="checkbox"/> Brochures / posters / flyers  <input type="checkbox"/> DVD / Film / Multimedia         </td> <td style="width: 50%; vertical-align: top;"> <input type="checkbox"/> Coverage in specialist press  <input type="checkbox"/> Coverage in general (non-specialist) press  <input type="checkbox"/> Coverage in national press  <input type="checkbox"/> Coverage in international press  <input checked="" type="checkbox"/> Website for the general public / internet  <input type="checkbox"/> Event targeting general public (festival, conference, exhibition, science café)         </td> </tr> </table>		<input checked="" type="checkbox"/> Press Release <input type="checkbox"/> Media briefing <input type="checkbox"/> TV coverage / report <input type="checkbox"/> Radio coverage / report <input checked="" type="checkbox"/> Brochures / posters / flyers <input type="checkbox"/> DVD / Film / Multimedia	<input type="checkbox"/> Coverage in specialist press <input type="checkbox"/> Coverage in general (non-specialist) press <input type="checkbox"/> Coverage in national press <input type="checkbox"/> Coverage in international press <input checked="" type="checkbox"/> Website for the general public / internet <input type="checkbox"/> Event targeting general public (festival, conference, exhibition, science café)
<input checked="" type="checkbox"/> Press Release <input type="checkbox"/> Media briefing <input type="checkbox"/> TV coverage / report <input type="checkbox"/> Radio coverage / report <input checked="" type="checkbox"/> Brochures / posters / flyers <input type="checkbox"/> DVD / Film / Multimedia	<input type="checkbox"/> Coverage in specialist press <input type="checkbox"/> Coverage in general (non-specialist) press <input type="checkbox"/> Coverage in national press <input type="checkbox"/> Coverage in international press <input checked="" type="checkbox"/> Website for the general public / internet <input type="checkbox"/> Event targeting general public (festival, conference, exhibition, science café)		
<b>23 In which languages are the information products for the general public produced?</b> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; vertical-align: top;"> <input type="checkbox"/> Language of the coordinator  <input type="checkbox"/> Other language(s)         </td> <td style="width: 50%; vertical-align: top;"> <input checked="" type="checkbox"/> English         </td> </tr> </table>		<input type="checkbox"/> Language of the coordinator <input type="checkbox"/> Other language(s)	<input checked="" type="checkbox"/> English
<input type="checkbox"/> Language of the coordinator <input type="checkbox"/> Other language(s)	<input checked="" type="checkbox"/> English		

**Question F-10:** Classification of Scientific Disciplines according to the Frascati Manual 2002 (Proposed Standard Practice for Surveys on Research and Experimental Development, OECD 2002):

## **FIELDS OF SCIENCE AND TECHNOLOGY**

### 1. NATURAL SCIENCES

- 1.1 Mathematics and computer sciences [mathematics and other allied fields: computer sciences and other allied subjects (software development only; hardware development should be classified in the engineering fields)]
- 1.2 Physical sciences (astronomy and space sciences, physics and other allied subjects)
- 1.3 Chemical sciences (chemistry, other allied subjects)
- 1.4 Earth and related environmental sciences (geology, geophysics, mineralogy, physical geography and other geosciences, meteorology and other atmospheric sciences including climatic research, oceanography, vulcanology, palaeoecology, other allied sciences)
- 1.5 Biological sciences (biology, botany, bacteriology, microbiology, zoology, entomology, genetics, biochemistry, biophysics, other allied sciences, excluding clinical and veterinary sciences)

### 2 ENGINEERING AND TECHNOLOGY



- 2.1 Civil engineering (architecture engineering, building science and engineering, construction engineering, municipal and structural engineering and other allied subjects)
  - 2.2 Electrical engineering, electronics [electrical engineering, electronics, communication engineering and systems, computer engineering (hardware only) and other allied subjects]
  - 2.3. Other engineering sciences (such as chemical, aeronautical and space, mechanical, metallurgical and materials engineering, and their specialised subdivisions; forest products; applied sciences such as geodesy, industrial chemistry, etc.; the science and technology of food production; specialised technologies of interdisciplinary fields, e.g. systems analysis, metallurgy, mining, textile technology and other applied subjects)
3. MEDICAL SCIENCES
- 3.1 Basic medicine (anatomy, cytology, physiology, genetics, pharmacy, pharmacology, toxicology, immunology and immunohaematology, clinical chemistry, clinical microbiology, pathology)
  - 3.2 Clinical medicine (anaesthesiology, paediatrics, obstetrics and gynaecology, internal medicine, surgery, dentistry, neurology, psychiatry, radiology, therapeutics, otorhinolaryngology, ophthalmology)
  - 3.3 Health sciences (public health services, social medicine, hygiene, nursing, epidemiology)
4. AGRICULTURAL SCIENCES
- 4.1 Agriculture, forestry, fisheries and allied sciences (agronomy, animal husbandry, fisheries, forestry, horticulture, other allied subjects)
  - 4.2 Veterinary medicine
5. SOCIAL SCIENCES
- 5.1 Psychology
  - 5.2 Economics
  - 5.3 Educational sciences (education and training and other allied subjects)
  - 5.4 Other social sciences [anthropology (social and cultural) and ethnology, demography, geography (human, economic and social), town and country planning, management, law, linguistics, political sciences, sociology, organisation and methods, miscellaneous social sciences and interdisciplinary, methodological and historical S1T activities relating to subjects in this group. Physical anthropology, physical geography and psychophysiology should normally be classified with the natural sciences].
6. HUMANITIES
- 6.1 History (history, prehistory and history, together with auxiliary historical disciplines such as archaeology, numismatics, palaeography, genealogy, etc.)
  - 6.2 Languages and literature (ancient and modern)
  - 6.3 Other humanities [philosophy (including the history of science and technology) arts, history of art, art criticism, painting, sculpture, musicology, dramatic art excluding artistic "research" of any kind, religion, theology, other fields and subjects pertaining to the humanities, methodological, historical and other S1T activities relating to the subjects in this group]