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Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel FIRST-Nuclides

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Characterisation of spent nuclear fuel samples to be used in FIRST-Nuclides – relevance of samples for the Safety Case

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List of abbreviations

BU	burn-up
BWR	boiling water reactor
CNRS	Centre National de la Recherche Scientifique
CTM	Fundacio CTM Centre Tecnològic
EK	Magyar Tudományos Akadémia Energiatudományi Kutatóközpont
FIMA	fissions per initial metal atom
GWd $(t\ HM)^{-1}$	Giga-Watt day per tonne initial heavy metal
HM	heavy metal
IRF	fast / instant release fraction
HTR	High Temperature Reactor
JRC-ITU	Joint Research Centre - Institute for Transuranium Elements
JÜLICH	Forschungszentrum Jülich GmbH
KIT	Karlsruher Institut für Technologie
LWR	light water reactor
MOX	mixed oxide (U,Pu)O2
NAGRA	Nationale Genossenschaft für die Lagerung radioaktiver Abfälle
PSI	Paul Scherrer Institut
Pu_{fiss}	fissile Pu nuclides
PWR	pressurised water reactor
RN	radionuclide(s)
SCK·CEN	Studiecentrum voor Kernenergie / Centre d'Etude de l'Energie Nucléaire
SNF	spent nuclear fuel
STUDSVIK	Studsvik Nuclear AB
TRISO	triple Si carbide coated fuel particles
VHTR	Very High Temperature Reactor
VVER	Russian-designed pressurised water reactor
VVER / ВВЭР	Водо-водяной энергетический реактор, ВВЭР

1 Introduction

The EURATOM FP7 Collaborative Project “Fast/Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (FIRST-Nuclides)” is established with the overall objective to provide for improved understanding of the fast / instantly released radionuclides from disposed high burn-up spent nuclear fuel. Many of the published results on the fast/instant release fraction (IRF) have been determined with spent fuel irradiated either with relatively low or extremely high neutron fluxes which are not typical of present spent fuel waiting for disposal. Procedures for selection, characterization and preparation of spent nuclear fuel (SNF) samples, which are relevant to SNF disposal, are established in workpackage #1 “Samples and tools” of FIRST-Nuclides. All experimentally working project partners (KIT, JRC-ITU, JÜLICH, PSI, SCK·CEN, CNRS, CTM, EK and STUDSVIK) contribute to this workpackage. Most partners study high burn-up spent nuclear fuels irradiated in commercial nuclear power plants, while the JÜLICH group studies so-called TRISO fuel irradiated in a research reactor at the Petten EC Joint Research Centre. CNRS investigates unirradiated TRISO particles, which will be used in successive corrosion experiments under alpha irradiation. EK studies damaged and leaking VVER fuel rods with a burn-up below 27 GWd (t HM)$^{-1}$, which were stored in water for several years after an incident at the Paks-2 reactor.

One of the first activities in workpackage #1 covers the compilation of fuel characterisation data from the partners. The IRF depends on critical characteristics of the SNF such as manufacturing process, burn-up history and fuel temperature history, ramping processes and storage time. Therefore, characteristic data of various high burn-up spent nuclear fuel rods are collected, dealing with the type of nuclear reactor, its electrical power, types of fuel assemblies, fuel manufacturing information and the discharge date of the fuel to be investigated. With respect to the cladding, the characterization covers the composition, cladding diameter, thickness and the initial radial gap width between pellet and gap. The information regarding the pellet comprises the initial enrichment, geometry, grain size, density and specifics of the production process. The irradiation history covers the burn-up, the irradiation time and the number of cycles as well as the maximum and average linear power rate.

The present report (D1.1) presents *Characterisation of spent nuclear fuel samples to be used in FIRST-Nuclides* and *relevance of the samples for the Safety Case*. Data on fuel samples, which are available for the FIRST-Nuclides partners, are classified according to their data category with respect to the fuel / cladding characteristics, mentioned above. Moreover, these data-sets are classified according to three information levels: (i) essential information representing the minimum data that should be available for the fuel chosen for the study, (ii) parameters and data, which are not directly measured, but derived from calculations, and (iii) supplemental information referring to characteristics that may be needed depending on the studies to be performed. Critical parameters of the selected fuel samples are compared to those of high burn-up fuels which need to be disposed of in Europe, to assure the relevance of the samples for the Safety Case.

Spent fuel rods are owned mainly by the reactor operating utilities. KIT, JRC-ITU, JÜLICH, PSI, SCK·CEN, CTM, EK and STUDSVIK confirmed the access to the spent fuel material to be used in the project; they have the full rights to perform investigations and to publish the results.

2 Overview on selected fuel samples

Within the project FIRST-Nuclides, the experimentally working institutions study high BU fuels irradiated at different conditions. At the beginning of the project, the partners were asked to characterize the fuels, which had been proposed for the use in successive investigations. Using a questionnaire - slightly modified from a draft, which was developed by Lawrence Johnson (NAGRA) - the project partners were asked to provide those fuel characterization data, which are available to them at present. Table 1 presents the types of fuel characterization data, parameters and information categories in the questionnaire. In this D1.1 report, the listed data reflect the information on the fuels, which can be published presently. The information on the selected spent nuclear fuel samples are going to be updated throughout the project. The fuel characterization questionnaire includes essential information categories, which define the minimum data and information on the selected samples that need to be up-dated, revised and published in the course of the project FIRST-Nuclides.

Table 1: Types of fuel characterization data, parameters and information categories in the FIRST-Nuclides questionnaire.

Data category	Information level	Parameter
reactor type coolant	essential essential	BWR, PWR, VVER etc. light water
fuel material data prior to irradiation	essential essential	UO2, MOX etc. enrichment, additives etc. fuel stoichiometry nitrogen, chlorine contents fuel density (as fabricated)
fuel material data after irradiation	essential supplemental essential essential	calculated RN inventories measured RN inventories pellet dimensions grain size
fuel assembly design	essential	lattice geometry, e.g. 10X10 fuel rod diameter
fuel rod data	essential	rod type: standard or test rod internal rod pre-pressure burn-up profile (γ-scan) fission gas release
cladding composition	essential derived supplemental	alloy type and composition calculated RN inventories measured RN inventories nitrogen, chlorine contents
irradiation data	essential derived	power history cycle start and end dates fuel centre peak irradiation temperature

Table 1 (continued): Types of fuel characterization data, parameters and information categories in the FIRST-Nuclides questionnaire.

Data category	Information level	Parameter
pellet sample data	essential	calculated burn-up measured burn-up calculated RN inventories measured RN inventories position of sample in rod
	supplemental	ceramography, microscopy
cladding sample data	supplemental	oxide thickness

Fourteen spent nuclear fuel samples discharged from Belgian, Dutch, Finish, German, Spanish, Swedish, Swiss and US American light water reactors (LWR) and one non-irradiated TRISO fuel, which are in the stocks of the project partners, are chosen for consecutive investigations in FIRST-Nuclides. Table 2 presents a brief summary of characteristic information of all samples. The irradiated samples comprise six fuel rods discharged from boiling water reactors (BWR), seven discharged from pressurized water reactors (PWR) and one irradiated fuel sample discharged from a high flux reactor. In addition to these high burn-up SNF samples, 31 VVER fuel rods of the Hungarian Paks-2 nuclear power plant are studied.

The vast majority of the fuel rods had been manufactured and irradiated as standard rods; three test rods from the PWR Gösgen are provided by KIT and PSI. In addition to the spent UO_2 fuels, a single MOX fuel with an initial content of 5.5 % fissile Pu, irradiated at the PWR Gösgen (63 GWd (t HM)$^{-1}$) is studied. The initial ^{235}U enrichment of the BWR and PWR UO_2 fuels are in the range of 3.3 to 4.3 % and 2.8 to 4.3 %, respectively; some of the VVER fuel rods had been less enriched; ^{235}U enrichment of the irradiated TRISO fuel is considerably higher, i.e. 16.8 % ^{235}U (Table 2). Most of the selected BWR fuel rods are cladded with Zyr-2 type Zircaloy having diameters between 9.84 and 10.2 mm. Zyr-4 and Zr-1Nb (M5 and E110) type Zircaloy claddings with a diameter of about 9.5 mm are used for most of the selected PWR fuels.

The burn-up of BWR and PWR UO_2 fuels, selected for further experiments, range between 48.3 and 59.1 GWd (t HM)$^{-1}$ and 45 to 70.2 GWd (t HM)$^{-1}$, respectively. The burn-up of the VVER fuel rods are in the range of 10.8 to 26.7 GWd (t HM)$^{-1}$. Those average linear power values, which can be published at the present, are in the range of 130 to >330 W cm^{-1} (Table 2). The selected fuel rods were used in the BWR and PWR reactors during five to seven and two to 14 irradiation cycles, respectively.

Detailed information on the fuel and cladding materials, irradiation duration, burn-up history and discharge dates / storage time of most fuel samples are given in the following chapter.

Table 2: Summary of fuel samples selected for analytical and experimental investigations in the project FIRST-Nuclides.

Institution	LWR reactor	initial fuel enrichment	fuel type	burn-up GWd (t HM)$^{-1}$	average linear power / W cm^{-1}
KIT	PWR Gösgen (CH)	UO_2 3.8% ^{235}U	test rod	50.4	260
JRC-ITU	PWR *to be published*	UO_2 *to be published*	standard rod	45	*to be published*
JÜLICH	high flux reactor Petten (NL)	TRISO 16.8% ^{235}U	research fuel	~107 11% FIMA	*to be* determined
PSI	PWR Gösgen (CH)	UO_2 4.3% ^{235}U	test rod	62.2	*to be published*
PSI	BWR Leibstadt (CH)	UO_2 3.9% ^{235}U	standard rod	57.5 6.1 % FIMA	~160
PSI	PWR Gösgen (CH)	MOX 5.5% Pu_{fiss}	test rod	63	306
SCK·CEN	PWR Tihange-1 (B)	UO_2 4.3% ^{235}U	standard rod	52	>330
CNRS		TRISO 0.7% ^{235}U	research fuel	*non-irradiated fuel*	*non-irradiated fuel*
CTM	BWR *to be published*	UO_2 *to be published*	standard rod	54	*to be published*
EK	PWR Paks-2 (HU)	UO_2 2.4-3.8% ^{235}U	standard rods	10.8 to 26.7 *(31 VVER rods)*	130
Studsvik	BWR Olikluoto (SF)	UO_2 4.25% ^{235}U	standard rod	50.2	186
Studsvik	BWR Olikluoto (SF)	UO_2 4.25% ^{235}U	standard rod	54.8	~200
Studsvik	BWR *to be published*	UO_2 3.5% ^{235}U	standard rod	57.1	*to be published*
Studsvik	BWR *to be published*	UO_2 4.1% ^{235}U	standard rod	59.1	*to be published*
Studsvik	PWR Vandellòs (ES)	UO_2 + Gd 2.8% ^{235}U	standard rod	54.4	*to be published*
Studsvik	PWR North Anna (US)	UO_2 4.0% ^{235}U	standard rod	50.2	186

3 Fuel characterisation data-sets

A) Fuel sample selected for investigations by Karlsruher Institut für Technologie (KIT) and Joint Research Centre - Institute for Transuranium Elements (JRC-ITU)

For experimental studies at KIT an irradiated UO_2 test fuel rod segment is selected, which was irradiated at the Gösgen PWR, Switzerland, approaching a burn-up of 50.4 GWd $(t\ HM)^{-1}$. Presently, the fuel rod segment is characterized, drilled for gas sampling and cut in pellet-size slices at JRC-ITU. Gas samples are analyzed in parallel at KIT. Using pellets of this fuel rod segment, the fast / instant radionuclide release will studied by leach tests at KIT. Details on the 50.4 GWd $(t\ HM)^{-1}$ PWR fuel are given in Table 3.

Table 3: Characterisation data of 50.4 GWd $(t\ HM)^{-1}$ PWR fuel selected for investigations by KIT and JRC-ITU.

Data category	Information category	Parameter
Reactor	essential	PWR Gösgen, Switzerland
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 15x15 48 assemblies with 20 control rods/assembly
		Fuel rod diameter: 10.75 ± 0.05 mm
		Fuel rod # SBS 1108, Segment N 0203
Fuel rod data	essential	Test fuel rod
		Internal rod pre-pressure: 21.5 ± 1 bar He
Fuel material data	essential	UO2 fuel, initial enrichment 3.8% ^{235}U and U_{nat}
		O/U = 2; fuel fabrication without additives
		Pellet dimensions: ∅ = 9.2 mm, height = 11 mm
		Calculated radionuclide inventory given in Grambow et al. (2000)
		Fuel density (as fabricated): 10.41 g/cm^3
	supplemental	Measured radionuclide inventory given in Grambow et al. (2000)
Cladding data	essential	Zircaloy-4, DX ESL 0.8 Wall thickness: 0.725 mm Initial radial gap fuel / cladding: 0.17 mm
Irradiation data	essential	Calculated burn-up: 50.4 GWd $(t\ HM)^{-1}$ Number of cycles: 4 Average linear power: 260 W cm^{-1} Maximal linear power: 315 W cm^{-1} Discharge: 27. May 1989 Irradiation duration: 1226 days

B) Fuel samples selected for investigations by Joint Research Centre - Institute for Transuranium Elements (JRC-ITU) and Fundacio CTM Centre Tecnològic

Using two standard PWR and BWR fuel rods, JRC-ITU and CTM will sample pellets and powders. Static leaching experiments will be performed with these pellets and powder samples in order to differentiate radionuclide contributions from gap, grain boundary and fuel matrix. Both fuels were manufactured with UO_2 material, having standard ^{235}U enrichments. The burn-up of the PWR and BWR fuels reached 45 and 54 GWd (t HM)$^{-1}$. The ownership of both fuel rods will be transferred to JRC-ITU in the near future.

Before this transfer of ownership, characterisation data of the 45 GWd (t HM)$^{-1}$ PWR fuel and the 54 GWd (t HM)$^{-1}$ BWR fuel cannot be published. Comprehensive fuel characterization data-sets on the selected samples will be published in the course of the project FIRST-Nuclides.

C) Fuel sample selected for investigations by Forschungszentrum Jülich GmbH (JÜLICH)

JÜLICH will determine the microstructure and the elemental distributions of the TRISO fuel pebbles before and after leaching by the use of different analytical methods. The fuel pebbles were irradiated in the High Flux Reactor, Petten (Netherlands), which is owned by the Institute for Energy (IE) of the Joint Research Centre (JRC). Details on the selected TRISO fuel pebbles are given in Table 4. The fuel is based on UO_2 fuel kernels of 502.2 µm diameter, which are with several layers of pyrocarbon as well as an additional silicon carbide layer. The basic structure of fuel pebbles, TRISO spheres and fuel kernels is shown schematically in Figure 1. Pre-irradiation neutronic calculations had determined the heat from fission and photons in the pebbles. At the centre of the fuel pebbles a temperature of 1250 °C is estimated. The number of fissions at any time was determined which enabled calculation of the birth rate of the measured volatile fission isotopes of Kr and Xe by using their characteristic fission yields. A burn up of 11% FIMA (corresponding to about 107 GWd (t HM)$^{-1}$) was predicted at the end of irradiation.

Table 4: Characterisation data of TRISO high flux reactor fuel selected for investigations by JÜLICH.

Data category	Information category	Parameter
Reactor	essential	High Flux Reactor, Petten, Netherlands
		45 MW tank-in pool type reactor; Light water coolant
	derived	In-core irradiation positions: 17 Core lattice: 9 x 9 array containing 33 fuel assemblies, 6 control elements, 23 reflectors
	supplemental	Core position G3 was used
Fuel assembly	essential	HFR-EU1bis Sample Holder design; 5 pebbles and graphite half shells enclosed in AISI 321 stainless steel capsule.
Pebble material	essential	Pebble: Matrix graphite grade A3-3, matrix density: 1750 kg/m^3
		Low density graphite; Coating thickness of buffer layer: 92.3 10^{-6} m, inner PyC layer. 40.6 10^{-6} m, SiC layer: 35.9 10^{-6} m, outer PyC layer: 39.6
	essential	Prior to irradiation: enrichment 16.76 % ^{235}U
		Pebble: heavy metal 6 g per pebble
		9560 coated particles per pebble
		Coated particle: kernel diameter 502.2 10^{-6} m
		Fuel kernel: O/U ≤ 2.015; density:10.86 mg/m^{-3}
Coating data	essential	oPyC, SiC,iPyC, buffer coating thickness:174.8 µm
Irradiation data	essential	Measured and calculated burn up: 10.2 % FIMA Number of cycles: 10 Average burn-up: 11 % FIMA fast fluence (E >0.1 MeV): 3.64 x 10^{25} m^{-2} power density (fission and photons in [W/cm^3] 30
		Discharge: 18. October 2005 Duration: 249.55 days

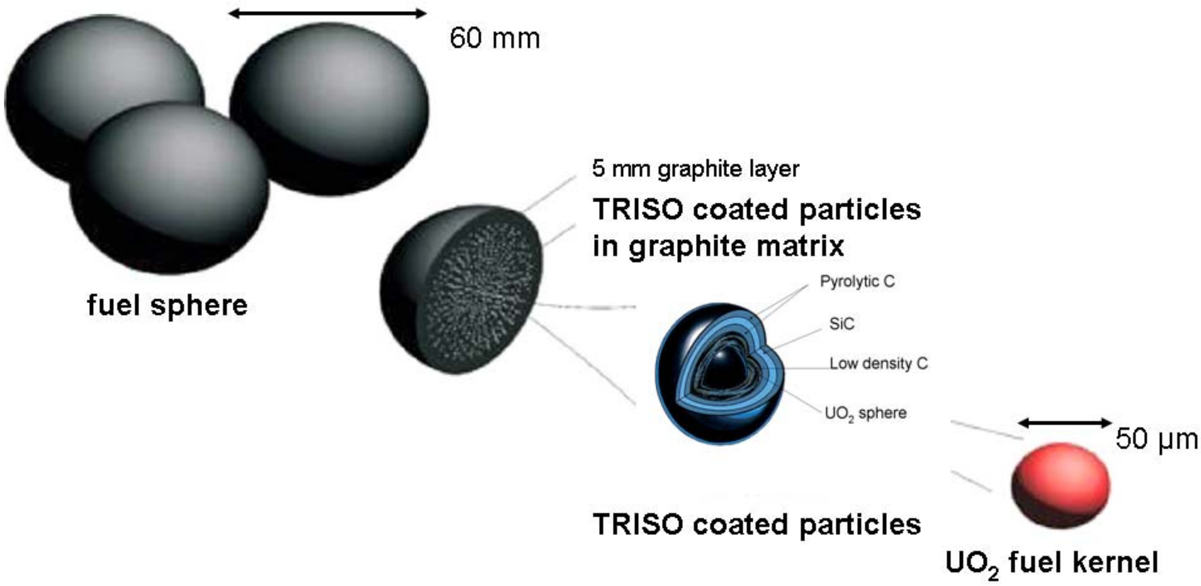

Figure 1: Schematic configuration of fuel pebbles, TRISO spheres and fuel kernels (modified after Brähler et al., 2011).

D) Fuel samples selected for investigations by Paul Scherrer Institut (PSI)

PSI will perform leach experiments on three high burn-up SNF samples, which had been irradiated in the BWR Leibstadt and PWR Gösgen in Switzerland, respectively. The preparation of the fuel samples (cutting, drilling, and breaking) is performed in the hot cell line of the PSI Hot Laboratory. In the following details on the 62.2 GWd (t HM)$^{-1}$ PWR fuel (Table 5), 57.5 GWd (t HM)$^{-1}$ BWR fuel (Table 6) and 63 GWd (t HM)$^{-1}$ PWR MOX fuel (Table 7) are given.

Table 5: Characterisation data of 62.2 GWd (t HM)$^{-1}$ PWR fuel selected for investigations by PSI

Data category	Information category	Parameter
Reactor	essential	PWR Gösgen, Switzerland
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 15x15 48 assemblies with 20 control rods/assembly
		Fuel rod diameter: 10.765 mm Fuel rod diameter after irradiation: 10.697 mm
		KKG-14B-4021-01-0129 (TM-43-06-15, 12.2006)
Fuel rod data	essential	Test rod Rod length as fabricated: 3860 mm Rod length after irradiation: 3879 mm Active length: 3550 mm
		Internal rod pre-pressure: 22 bar
		Fission gas release: 13.2%, 51.5 bar
Fuel material data	essential	UO2 fuel, initial enrichment 4.3% ^{235}U
		Fuel density (as fabricated): 10.45 g/cm^3
Cladding data	essential supplemental	Cladding outer diameter: 10.75 ± 0.05 mm Cladding inner diameter: max. 9.45 mm Max. oxide thickness: 36 µm
Irradiation data	essential	Calculated burn-up: 62.2 GWd (t HM)$^{-1}$ Number of cycles: 4 Date of loading: 28. July 1999 Discharge date: 28. June 2003 Irradiation duration: 1324.43 days

Table 6: Characterisation data of 57.5 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by PSI

Data category	Information category	Parameter
Reactor	essential	BWR Leibstadt, Switzerland
		light water coolant
Fuel assembly design information	essential	10 x 10 Lattice SVEA96 Optima, fuel assembly AIA003, rod position H6, Node 4
Fuel rod data	essential	standard rod
		Rod length as fabricated: 4146.6 mm Rod length after irradiation: 4163.3 mm
		Fission gas release: 2.26 %
Fuel material data	essential	UO_2 fuel, initial enrichment 3.9% ^{235}U
		Fuel density (spec) 10.52 ± 0.19 g/cm^3 Fuel density as fabricated 10.48 – 10.54 g/cm^3
		Pellet diameter (as fabricated): 8.77 ± 0.013 mm Pellet length: 10.7 ± 0.8 mm
		Grain size $6 \leq x \leq 25$ µm
Cladding data	essential	Zircaloy-2, designation LK3/L Cladding outer diameter: 10.30 ± 0.04 mm Cladding inner diameter: 8.94 ± 0.04 mm Liner Thickness 70 ± 40 µm
Irradiation data	essential	Rod average Burn-up: 57.5 GWd (t HM)$^{-1}$ exp. determined 6.1% FIMA Number of cycles: 7 Average linear power: ~ 160 W cm^{-1} Maximal linear power: 270 W cm^{-1} Date of loading: August 1998 Discharge date: April 2005 Irradiation duration: 2400 days

Table 7: Characterisation data of 63 GWd (t HM)$^{-1}$ PWR MOX fuel selected for investigations by PSI

Data category	Information category	Parameter
Reactor	essential	PWR Gösgen, Switzerland
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 15x15 48 assemblies with 20 control rods/assembly
		Fuel rod diameter: 10.76 mm (measured) Fuel rod diameter after irradiation: 10.736 mm
		KKG-13-5024-10-676 (TM-43-05-24, Rev.1, Oct.. 2008)
Fuel rod data	essential	Test rod
		Fission gas release: 26.7%, 76.4 bar
	supplemental	Rod length as fabricated: 3859.0 mm Rod length after irradiation: 3886.1 mm Active length: 3550 mm
Fuel material data	essential	MOX, initial enrichment: 5.5 % Pu_{fiss}
		Fuel density (as fabricated): 10.45 ± 0.15 g cm^{-3} Fuel density after irradiation: 9.903 g cm^{-3} Pellet diameter (as fabricated): 9.13 ± 0.013 mm
Cladding data	essential	Duplex ELS0.8b Cladding outer diameter: 10.75 ± 0.05 mm Cladding inner diameter: 9.30 ± 0.04 mm Max. oxide thickness: 36 µm
Irradiation data	essential	Rod average Burn-up: 63.0 GWd (t HM)$^{-1}$ Number of cycles: 4 Average linear power: ~ 306 W cm^{-}1 (average fuel rod power in 4 cycles) Maximal linear power: 430 W cm^{-1}
		Date of loading: 30. June 1997 Discharge date: 7. July 2001 Irradiation duration: 1368 days

E) Fuel sample selected for investigations by Studiecentrum voor Kernenergie / Centre d'Etude de l'Energie Nucléaire (SCK•CEN)

SCK•CEN assessed manufacturing and operational data of high burn-up (HBU)-SNF available in their stocks. For experimental studies at SCK•CEN an irradiated UO_2 standard fuel rod is selected, which was irradiated at the Tihange-1 PWR, Belgium, approaching a burn-up of 52 GWd (t HM)$^{-1}$. Fuel fragments with the suitable properties will be prepared for leaching experiments. Details on this high burn-up SNF are given in Table 8.

Table 8: Characterisation data of 52 GWd (t HM)$^{-1}$ PWR fuel selected for investigations by SCK•CEN

Data category	Information category	Parameter
Reactor	essential	PWR Tihange-1, Belgium
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 15x15 FT1x57
		Fuel rod diameter: 10.72 mm
Fuel rod data	essential	Standard fuel rod
		Rod diameter: 10.720 mm Internal rod pre-pressure: 20 bar He
Fuel material data	essential	UO2 fuel, initial enrichment 4.254 % ^{235}U
		O/U = 2; fuel fabrication without additives
		Fuel density (as fabricated): 96.3 +/- 0.1%
Cladding data	essential	M5 (Zr-Nb-Fe-O) alloy (nominal ZrNb) Cladding outer diameter: 10.72 mm Cladding inner diameter: 9.484 mm Wall thickness: 0.618 mm Initial radial gap fuel / cladding: 0.095 mm
Irradiation data	essential	Calculated burn-up: 52 GWd (t HM)$^{-1}$ Number of cycles: 2 Average linear power: possibly > 330 W cm^{-1} Date of loading: *to be published* Date of discharge: *to be published* Irradiation duration: *to be published*

F) Fuel sample selected for investigations by Centre National de la Recherche Scientifique (CNRS)

CNRS studies an unirradiated TRISO sample, which was manufactured for the use in a High Temperature Reactor (HTR). The basic structure of spherical fuel kernels within the larger TRISO spheres and HTR fuel pebbles is shown schematically in Figure 1. The analytical and experimental investigations on unirradiated TRISO particles at CNRS are complementary to those with irradiated TRISO fuel at JÜLICH (see section (C)). At CNRS, TRISO type VHTR spheres with and without grain boundaries are examined. Corrosion of the fuel particles under alpha irradiation is studied, to determine the role of grain boundary dissolution on the overall radiolytically enhanced UO_2 dissolution process.

The non-irradiated TRISO UO_2 fuel spheres were provided by JÜLICH to be examined at CNRS. Synthesis of the VHTR spheres is described in detail by Brähler et al. (2011). Results of physico-mechanical characterization and first solubility tests are published in Bros et al. (2006), Grambow et al. (2008) and Titov et al. (2004). Characteristic data of the spherical fuel kernel selected for investigations by CNRS are given in Table 9.

At CNRS, coating layers were separated from a spherical fuel kernel (Figure 2). Using SEM-EDX it was demonstrated that the chemical composition of the kernel surface is UO_2. Figure 3 shows boundaries of UO_2 grains of an untreated spherical fuel kernel and grain boundaries of a spherical fuel kernel after washing with 0.1 mol L^{-1} HCl for 15 days.

Table 9: Characterisation data of spherical fuel kernel selected for investigations by CNRS.

composition	UO_2
diameter	0.5 mm
mass	0.76 mg
density	10.96 g cm^{-3}
geometric surface area	0.00105 m^2 g^{-1}

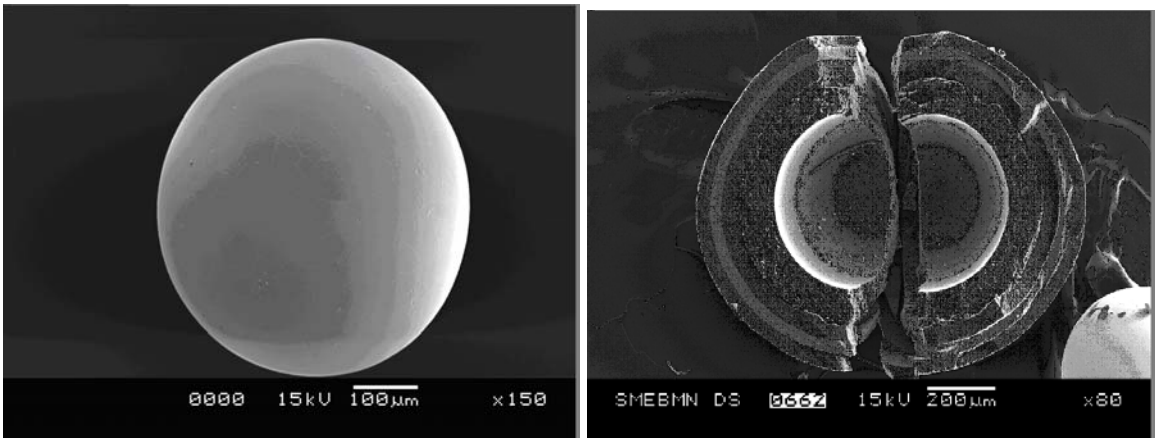

Figure 2: SEM image of a spherical UO_2 fuel kernel separated from its coatings (left) and SEM image of the cracked oPyC, SiC, iPyC and buffer coatings (right). Spherical fuel kernel separation and SEM imaging performed by J. Vandenborre, CNRS, in the framework of FIRST-Nuclides.

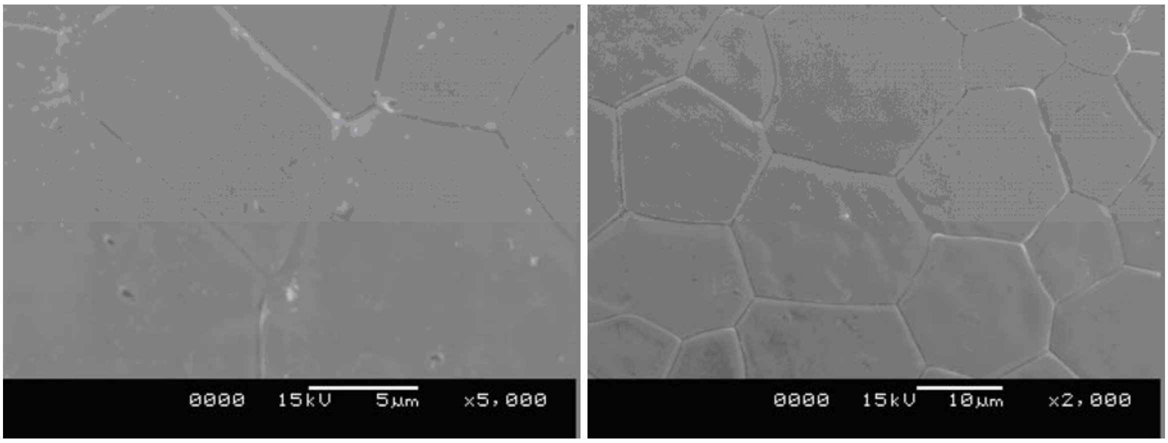

Figure 3: Boundaries of UO_2 grains of an untreated spherical fuel kernel (left) and grain boundaries of a spherical fuel kernel after HCl washing (right).

G) Fuel rods investigated by Magyar Tudományos Akadémia - Energiatudományi Kutatóközpont (EK)

EK compiles manufacturing and operational data on 31 VVER fuel rods irradiated at the Paks-2 PWR, Hungary. The damaged and leaking VVER fuel rods are stored in water for several years. Activity concentration data are going to be analyzed in order to determine activity release from into water environment. Details on the Paks-2 fuel rods are given in Table 10.

Table 10: Characterisation data of Paks-2 fuel rods to be investigated by EK.

Data category	Information category	Parameter
Reactor	essential	PWR Paks-2, Hungary, VVER-440
		light water coolant
Fuel assembly design information	essential	Lattice geometry: hexagonal 349 assemblies; 126 fuel rods per assembly
Fuel rod data	essential	31 standard fuel assemblies from Paks cleaning tank incident; leaking fuel assembly No. 70873; damaged and leaking assemblies
		Fuel rod diameter: 9.15 mm
		Internal rod pre-pressure: 5 bar He
Fuel material data	essential	UO2 fuel, initial enrichment 2.4 to 3.82% ^{235}U
		O/U = 2; fuel fabrication without additives
		Pellet dimensions: ∅ = 7.57 mm, height = 9 to 12 mm; central hole ∅ = 1.2 to 1.8 mm
		Grain size: 8–20 µm (factory data)
		Fuel density (as fabricated): 10.4 to10.8 g/cm^3
Cladding data	essential	E110 cladding (Zr1%Nb) Wall thickness: 0.65 mm Initial radial gap fuel / cladding: 0.14 mm
Irradiation data	essential	Calculated burn-up: 10.8 to 26.7 GWd (t HM)$^{-1}$ Number of cycles: one (20 assemblies), two (6 assemblies) and three (5 assemblies) cycles Average burn-up: 16 GWd (t HM)$^{-1}$ (average for 30 damaged assemblies), 14 GWd (t HM)$^{-1}$ (one leaking assembly) Average linear power: 130 W cm^{-1} Maximal linear power: 228 W cm^{-1} Date of discharge: March 2003 and April 2009 Irradiation duration: 320–795 days

H) Fuel samples selected for investigations by Studsvik Nuclear AB (STUDSVIK)

STUDSVIK will carry out leach experiments on six high burn-up SNF samples, which had been irradiated in the BWR Olikluoto (Finland), PWR Vandellòs (Spain), PWR North Anna (United States of America) and other BWR. Investigations of STUDSVIK focus on the effects of different features of the SNF samples on the fast / instant release of fission products such as Cs and I. In the following details on the 50.2 GWd (t HM)$^{-1}$ BWR fuel (Table 11), 54.8 GWd (t HM)$^{-1}$ BWR fuel (Table 12), 57.1 GWd (t HM)$^{-1}$ BWR fuel (Table 13), 59.1 GWd (t HM)$^{-1}$ BWR fuel (Table 14), 54.4 GWd (t HM)$^{-1}$ PWR fuel (Table 15) and 70.2 GWd (t HM)$^{-1}$ PWR fuel (Table 16) are given. Two subsamples of the 70.2 GWd (t HM)$^{-1}$ PWR fuel were cut from near the middle and used in previous leaching experiments (Johnson et al., 2012). Pre-irradiation data and a summary of the operating history and characterization of the fuel samples are reported elsewhere (Zwicky et al., 2011). The local burn-up of the two subsamples is significantly higher (i.e. 75.4 GWd (t HM)$^{-1}$) than the average of the fuel rod.

Table 11: Characterisation data of 50.2 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Olikluoto, Finland
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 10.05 mm
		denoted as “D07”
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 55 bar
		Fission gas release: ~1.6 %
Fuel material data	essential	UO2 fuel, initial enrichment 4.25% ^{235}U
		Pellet dimensions: ∅ = 8.7 mm
		Fuel density: 10.5 g/cm^3
Irradiation data	essential	Calculated burn-up: 50.2 GWd (t HM)$^{-1}$ Date of loading: 6. June 2003 Date of discharge: 13. June 2008

Table 12: Characterisation data of 54.8 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Olikluoto, Finland
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 10.05 mm
		denoted as “L04”
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 55 bar
		Fission gas release: ~3.1 %
Fuel material data	essential	UO2 fuel, initial enrichment 4.25% ^{235}U
		Pellet dimensions: ∅ = 8.7 mm
		Fuel density: 10.5 g/cm^3
Irradiation data	essential	Calculated burn-up: 54.8 GWd (t HM)$^{-1}$ LHGR = ~200 W/cm (to be confirmed) Date of loading: 2003 Date of discharge: 2008

Table 13: Characterisation data of 57.1 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 9.62 mm
		denoted as “O35A2”
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 81 bar
		Fission gas release: ~2.4 %
Fuel material data	essential	UO2 fuel, initial enrichment 3.5% ^{235}U
		Pellet dimensions: ∅ = 8.2 mm, length = 10.0 mm
		Fuel density: 10.53 g/cm^3
Irradiation data	essential	Calculated burn-up: 57.1 GWd (t HM)$^{-1}$ Date of loading: 2000 Date of discharge: 2008 Irradiation period: 2000-2006, 2007-2008

Table 14: Characterisation data of 59.1 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 9.62 mm
		denoted as "O3C1"
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 83 bar
		Fission gas release: ~1.4 %
Fuel material data	essential	UO2 fuel, Al/Cr doped initial enrichment 4.1% ^{235}U
		Pellet dimensions: ∅ = 8.2 mm, length = 10.0 mm
		Fuel density: 10.67 g/cm^3
Irradiation data	essential	Calculated burn-up: 59.1 GWd (t HM)$^{-1}$ Date of loading: 2000 Date of discharge: 2008 Irradiation period: 2000-2006, 2007-2008

Table 15: Characterisation data of 54.4 GWd (t HM)$^{-1}$ BWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	PWR Vandellòs, Spain
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 17x17
		denoted as "VG81"
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 2.137 MPa
		Fission gas release: ~2.2 %
Fuel material data	essential	UO2 fuel with Gd initial enrichment 2.8% ^{235}U
		Pellet dimensions: ∅ = 8.192 mm, length 9.83 mm
		Fuel density: 10.25 g/cm^3
Irradiation data	essential	Calculated burn-up: 54.4 GWd (t HM)$^{-1}$ Average linear power =1.64 kW/m (to be confirmed) Date of loading: 10. Oct. 2000 Date of discharge: 5. May 2007

Table 16: Characterisation data of 70.2 GWd (t HM)$^{-1}$ PWR fuel selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	PWR North Anna, United States of America
		light water coolant
		denoted as “AM2K12”
Fuel rod data	essential	Standard fuel rod
		Rod diameter = 9.5 mm Internal rod pre-pressure: 1.9 MPa
		Fission gas release: ~4.9 %
Fuel material data	essential	UO_2 fuel, initial enrichment 4.0% ^{235}U
		Pellet dimensions: ∅ = 8.192 mm, length 9.83 mm
		Fuel density: 10.44 g/cm^3
Irradiation data	essential	Calculated rod average burn-up: 70.2 GWd (t HM)$^{-1}$ Average linear power =18.6 kW/m Number of cycles: 14 Date of loading: 1987-07-01 Date of discharge: 2001-03-12

4 Summary and conclusions

As documented in the previous sections, the selected SNF samples were irradiated at different conditions. They cover a wide range of high burn-up fuels with respect to their critical parameters, such as fuel material composition prior to irradiation (in particular the enrichment), power history (in particular the average burn-up and linear power rate), fuel composition after irradiation and cladding properties. The vast majority of the selected fuel rods had been manufactured as standard fuel rods and were irradiated in more than six European and US American boiling water reactors and pressurized water reactors. The initial ^{235}U enrichment versus the average discharge burn-up of the selected fuels are compared to respective values for irradiated BWR and PWR fuels reported by the NEA Nuclear Science Committee (2006). As shown in Figure 4, most of the high BU fuels selected for investigations in the project FIRST-Nuclides are on or close to trend-lines for present high burn-up fuels. Non-standard fuels are included in the project, such as irradiated TRISO fuel with extremely high BU as well as damaged and leaking VVER fuel rods with relatively low BU, to enlarge the dataset over a wider range of parameters. At the present state, the characterisation of the selected high BU spent nuclear fuel samples leads to the conclusion that these samples are relevant to high burn-up fuels which need to be disposed of in Europe. As a consequence, it is expected that the knowledge and data obtained from experiments with the selected fuel samples will reduce the uncertainties and will provide for realistic data on the fast / instant radionuclide release for the Safety Cases of spent nuclear fuel disposal systems.

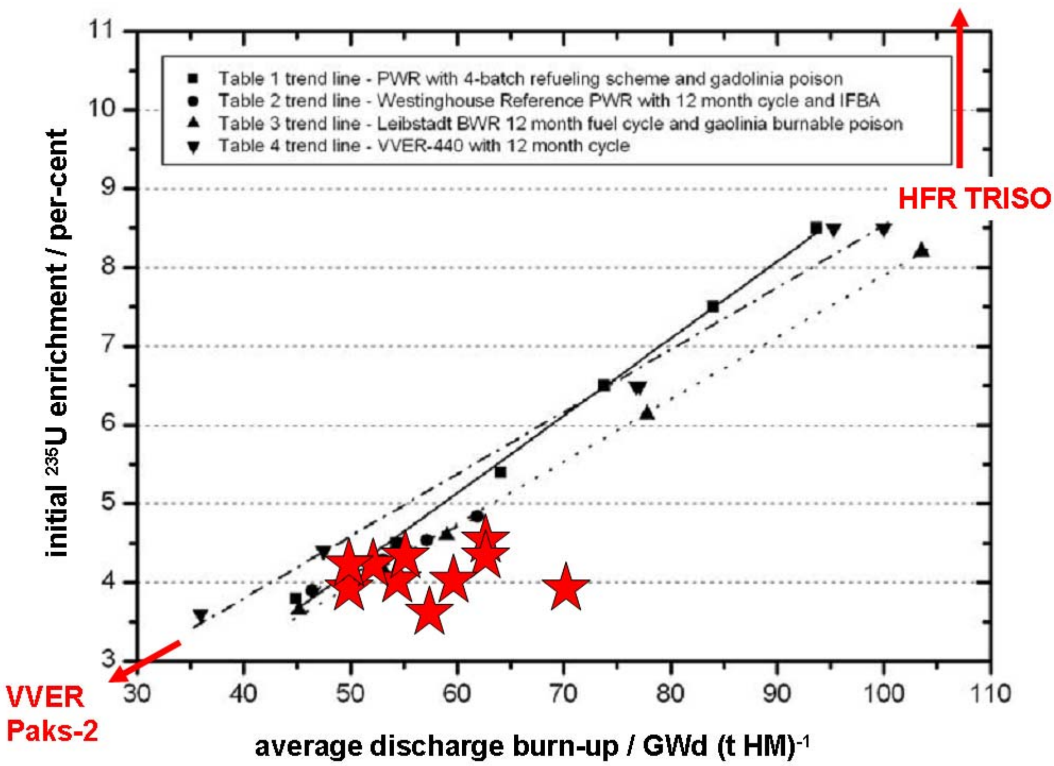

Figure 4: Initial ^{235}U enrichment versus average discharge burn-up (modified after NEA Nuclear Science Committee, 2006). Data of selected BWR and PWR fuels are shown as stars; enrichment / burn-up ratios of the irradiated TRISO fuel and of the PWR Paks-2 VVER fuel rods are out of the shown enrichment / burn-up range.

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