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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 and description of methodologies and tools to be applied in FIRST-Nuclides

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[FIRST-Nuclides]

FIRST Nuclides

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BU	burn-up
BWR	boiling water reactor
CNRS	Centre National de la Recherche Scientifique
CP	coated particle
CTM	Fundacio CTM Centre Tecnològic
ELS	extra low Sn (cladding with low tin content)
EPMA	electron-probe micro-analysis
ESEM	environmental scanning electron microscope
FG	fission gas
FIMA	fissions per initial metal atom
FP	fission product
GB	grain boundary
$GWd\cdot(t\ HM)^{-1}$	Giga-Watt day per tonne initial heavy metal
HBU-SNF	high burn-up spent nuclear fuel
HM	heavy metal
HTR	high temperature reactor
ICP-MS	inductively coupled plasma mass spectrometry
IRF	fast / instant release fraction
JRC-ITU	Joint Research Centre - Institute for Transuranium Elements
JÜLICH	Forschungszentrum Jülich GmbH
KCMS	Knudsen cell mass spectrometer
KIT	Karlsruher Institut für Technologie
KKG	Kernkraftwerk Gösgen (nuclear power reactor Gösgen, Switzerland)
KKG	Kernkraftwerk Leibstadt (nuclear power reactor Leibstadt, Switzerland)
KWU	Kraftwerk Union AG
LHGR	linear heat generation rate
LSC	liquid scintillation counting
LWR	light water reactor
MC-ICP-MS	multicollector inductively coupled plasma mass spectrometry
MERARG	fission gas release study facility by annealing
MS	mass spectrometry
MTA EK	Magyar Tudományos Akadémia Energiatudományi Kutatóközpont
MOX	mixed oxide $(U,Pu)O_2$
NDT	non-destructive testing
NIKUSI	Niedrigtemperatur-Kurzzeit-Sintern / short-term fast sintering process of UO_2 pellets
NPP	nuclear power plant
OM	optical microscopy
PIE	post-irradiation examination
PSI	Paul Scherrer Institut
Pu_{fiss}	fissile Pu nuclides

PWR	pressurised water reactor
Q-GAMES	quantitative gas measurement setup
RCA	radiochemical analysis
RN	radionuclide(s)
SCK·CEN	Studiecentrum voor Kernenergie / Centre d'Etude de l'Energie Nucléaire
SEM	scanning electron microscopy
SNF	spent nuclear fuel
STUDSVIK	Studsvik Nuclear AB
TRISO	tri-isostructural (triple Si carbide coated fuel particles)
VVER / ВВЭР	Водо-водяной энергетический реактор, ВВЭР (Russian-designed pressurised water reactor)

1. Introduction

The EURATOM FP7 Collaborative Project "Fast/Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (FIRST-Nuclides)" aims to provide new and comprehensive knowledge of the fast / instantly released radionuclides from disposed high burn-up spent UO_2 fuel. The fast release of a specific fraction radionuclides will take place only after a canister failure and water penetration to the spent nuclear fuel (SNF) in a repository. The so-called instant release fraction (IRF) consists of radionuclides in gaseous form, and radionuclides showing a high solubility in groundwater.

The basic activities of the project FIRST-Nuclides – coordinated in workpackage #1 "Samples and Tools" - were to select, provide and prepare 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. 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.

Most partners dealt with high burn-up SNF, which had been irradiated in commercial nuclear power reactors, while the JÜLICH group studied so-called TRISO fuel irradiated in a research reactor at the Petten EC Joint Research Centre. CNRS worked with unirradiated TRISO particles, which are used in successive corrosion experiments under alpha irradiation. MTA EK studied damaged and leaking VVER fuel rods, which were stored in water for several years after an incident at the Paks-2 reactor. KIT, JRC-ITU, PSI, SCK•CEN, CTM and STUDSVIK investigate six SNF fuels, which were discharged from boiling water reactors (BWR), and six SNF fuels, which were discharged from pressurized water reactors (PWR). The selected BWR fuels were initially enriched with ^{235}U in the range of 3.7 to 4.3% and approached average fuel rod burn-ups between 42.2 to 59.1 GWd·(t HM)$^{-1}$ during irradiation. The selected PWR fuels cover a burn-up range of 50.0 to 70.2 GWd·(t HM)$^{-1}$ and initial ^{235}U enrichments in the range of 2.8 to 4.3%. Additionally, a PWR mixed oxide fuel with 63 GWd·(t HM)$^{-1}$ and an initial Pu_{fiss} enrichment of 5.5 % is studied by PSI.

According to available Kr and Xe analyses of eleven BWR and PWR fuel rod puncturing tests, the fission gas release (FGR) of the BWR fuels is between 1.4 and 3.9% and FGR of the PWR fuels is between 2.1 and 13.2%. Based on the data of the studied BWR fuels, there is no simple correlation of FGR with the average fuel rod burn-up or with the average linear heat generation rate (LHGR) of the SNF samples (Figure 1). Interestingly, the spent BWR fuel with the highest average fuel rod burn-up (a Al/Cr doped SNF denoted as "C1" under investigation at STUDSVIK) is characterized by the lowest fission gas release. The PWR fuel rod puncturing tests reveal a wide range of FGR values (Figure 1). The highest fission gas release is observed for a PWR Gösgen test rod with an average burn-up of 62.6 GWd·(t HM)$^{-1}$ under investigation by PSI[*]. The SNF with the highest average burn-up of 70.2 GWd·(t HM)$^{-1}$ (a standard fuel rod denoted as "AM2K12" under investigation by STUDSVIK) was irradiated at a intermediate LGHR of 18.6 kW/m and achieved an intermediate FGR of 4%. The variation of FGR values demonstrates that the fission gas release is neither correlated linearly to average fuel rod burn-ups and nor to average linear heat generation rates, but may depend on various characteristics of the SNF such as manufacturing process, burn-up history and fuel temperature history, ramping processes and storage time. Similarly, a complex dependency is expected for the IRF of the studied SNF samples. As a consequence for

[*] LHGR values for this high burn-up SNF discharged from PWR Gösgen are not available at present.

interpretation of IRF studies – and FGR studies in particular – it is essential to compile critical parameters with respect to the fuel material composition prior to irradiation, power history, fuel composition after irradiation and cladding properties, as is done in the present report.

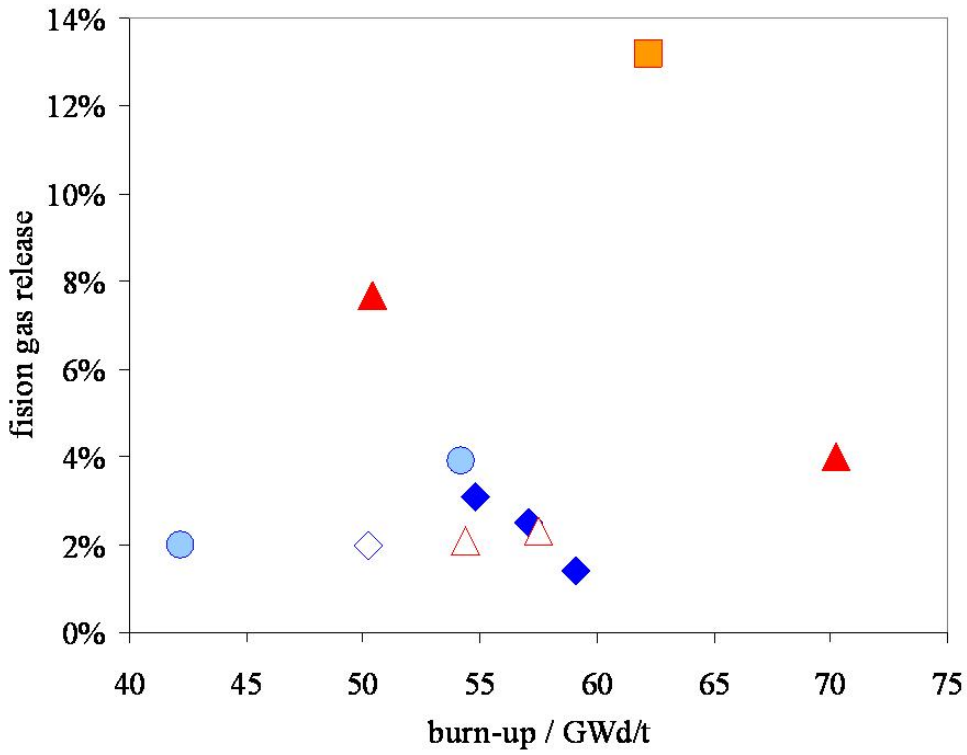


Figure 1: Measured fission gas release of studied UO_2 fuels discharged from BWR and PWR. Blue symbols denote BWR fuels and red / orange symbols PWR fuels: Open diamond denotes a BWR fuel with a LHGR of 15 kW/m, closed diamonds denote BWR fuels with LHGR ≥ 16 kW/m and the circles denote BWR fuels with un-published LHGR values; open triangles denote PWR fuels with LHGR of 13.6 and 16 kW/m, resp., closed triangles denote PWR fuels with LHGR ≥ 18.6 kW/m and the square denotes a PWR fuel with un-published LHGR value.

This report provides characterisation data of the selected SNF samples and describes the methodologies and tools, which are applied in the FIRST-Nuclides project. Tabulated fuel characterisation data-sets of the studied spent BWR and PWR fuels are provided in chapter 10. Results of non-destructive analysis of fuel rods with respect to visual examination, scanning and defect determination of the cladding, as well as activities to puncture, to cut the selected fuel rods and to prepare various kinds of fuel samples (e.g. cladded pellets, de-cladded fuel rod fragments and SNF powders) are presented in chapters 2 to 9. In addition, the report covers the preparation of extended (destructive) post irradiation examinations, quantitative determination of a variety of radionuclides by complete digestion of fuel rod fragments and by spatially resolved laser ablation mass spectrometry, deduction of the burn-up of fuel rod fragments used for SNF leaching experiments, and µ-scale synchrotron based speciation analyses.

2. KIT - SNF sample characterisation, methodologies and tools

E. González-Robles, N. Müller, V. Metz, E. Bohnert, M. Herm, M. Lagos and B. Kienzler

KIT provides a HBU-SNF rod segment in the ownership of KIT, where all data and findings can be published without restrictions. The studied fuel rod segment N0204 of the KKG-BS fuel rod SBS1108 (SBS1108-N0204) was irradiated together with the adjacent segment SBS1108-N0203 in the PWR Gösgen (*KKG*), Switzerland, and discharged in May 1989. Characteristic data of the KKG fuel were collected, dealing with the fuel manufacturing and fuel assembly design. Data on irradiation history of both segments as well as experimental data of SBS1108-N0203 (determined in previous studies) were documented. With respect to the cladding, the characterization covers the composition, cladding thickness and the initial radial gap width between pellet and cladding. The information regarding the fuel material comprises the initial enrichment, pellet dimensions, 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 fuel rod segment SBS1108-N0204 (Figure 2) was transported to JRC-ITU in March 2012 for characterisation, gas sampling, cutting and sampling of fuel pellets. Results of the non-destructive characterisation and gas sampling, conducted in the following weeks, are given in reports of JRC-ITU and KIT (Wegen *et al.*, 2012; 2013a; 2013b; 2013c; González-Robles *et al.*, 2013, respectively). After puncturing and successive cutting of the fuel rod segment, gas samples and fuel pellets were returned to KIT for further investigations.

2.1. Characterisation of a spent fuel rod segment from PWR Gösgen studied at KIT

The studied HBU-SNF consists of pure UO_2, fabricated by Kraftwerk Union AG using the *NIKUSI* sintering process (Stratton *et al.*, 1991). NIKUSI is a short-term fast sintering process under controlled oxidizing condition at a temperature (< 1300°C) below the temperature range of conventional UO_2 pellet sintering processes (Kutty et al., 2004). Stratton et al. (1991) reported an UO_2 stoichiometry of U/O = 2.002. Generally, the fuel pellets of the segment had been initially enriched with 3.8% ^{235}U. Two pellets adjacent to the upper and lower isolation pellets were made of U_{nat} (Figure 3). The cladding material is Zircaloy-4 (DX ELS 0.8) with an external and internal diameter of 10.75± 0.05 mm and about 9.3 mm, respectively; the wall thickness is specified as 0.725 mm (Kernkraftwerk Gösgen, 2010; Stratton *et al.*, 1991). The lattice geometry of the fuel assembly consisted of a 15×15 array of fuel rods. 205 of the 225 positions per assembly were occupied with fuel rods, the 20 remaining positions were available for control rods (Kernkraftwerk Gösgen, 2010). The final rod-average burn-up was estimated as 50.4 GWd·(t HM)$^{-1}$ for the fuel rod SBS1108, which had been achieved in four cycles of 1226 days total irradiation duration.

The data on the fuel rod segment SBS1108-N0204 were compiled in Deliverable 1.1 (Metz et al., 2012) and in the Proceedings of the 1st Annual Workshop of FIRST-Nuclides (Metz et al., 2013). The data-set is given in Table 13 of chapter 10 for the sake of completeness and comparison with data of all HBU-SNF studied in FIRST-Nuclides. A rim zone burn-up of 67.0 GWd·(t HM)$^{-1}$ and a maximal temperature during irradiation above 1300°C was estimated based on the data of Table 13 (Kienzler *et al.*, 2013). The average and maximal linear power was calculated as 260 and 340 W/cm, respectively.

Figure 2: Fuel rod segment SBS1108-N0204 after removal from the lead shield storage tube at the shielded box-line of KIT before transport to JRC-ITU.

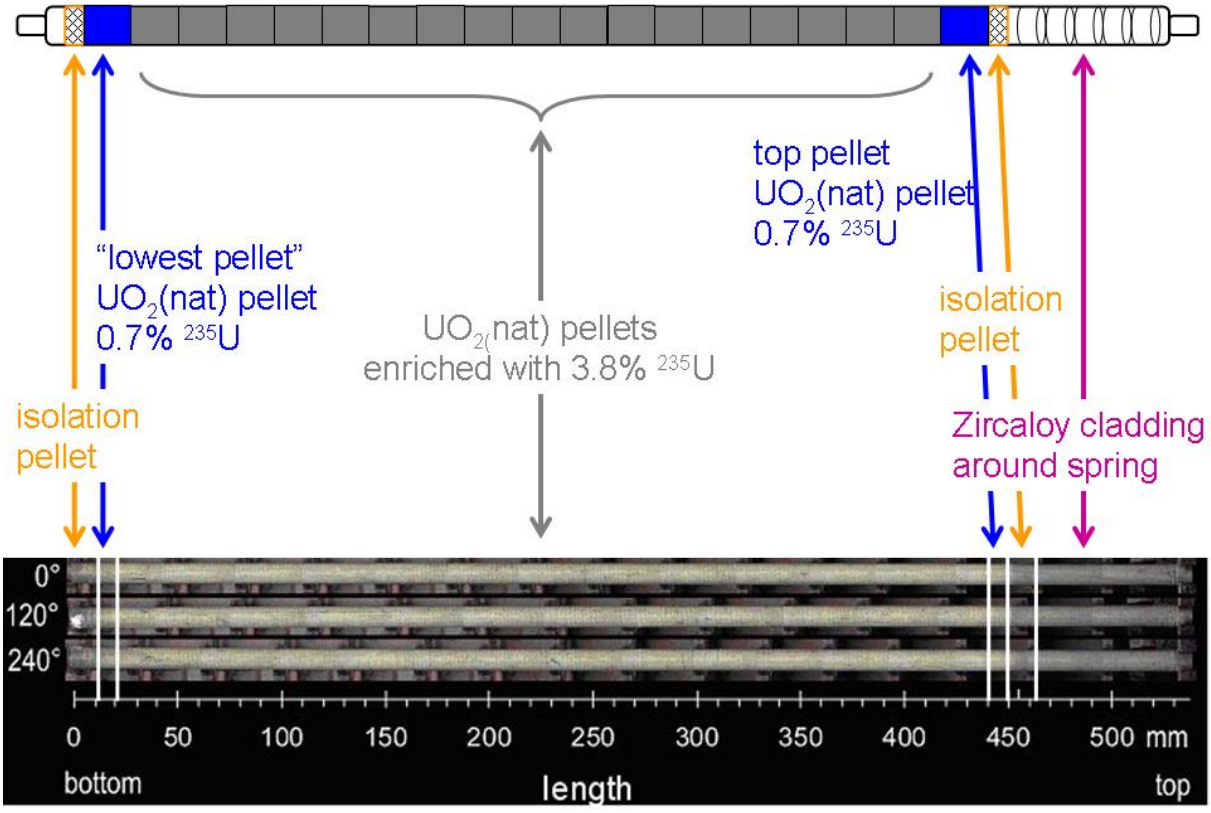


Figure 3: Schematic cross section and photos of the fuel rod segment SBS1108-N0204. The inserted photo is taken from Papaioannou et al. (2012).

2.2. Experimental and analytical methodology for SNF leaching experiments at KIT

Three leaching experiments with samples of the PWR Gösgen fuel rod segment are foreseen to be conducted under anoxic conditions. In cooperation with JRC-ITU, segmented cutting plans were compiled for the fuel rod segment SBS1108-N0204. The cutting procedure is described in section 3.5 and in Wegen et al. (2013a). In one experiment a cladded segment - pellet #3 of the rod segment - is used (Figure 4). At JRC-ITU, this segment was perfectly cut at the gaps between pellet #3 and the adjacent pellets. The zonation of the pellet dishing is visible in Figure 4. Another segment (pellet #4 in Figure 4) will be broken after cutting the cladding. Fragments of pellet #4 without cladding will be used in the second leaching experiment. In the third leaching experiment, fuel material of pellet #4 adhering to cladding will be used.

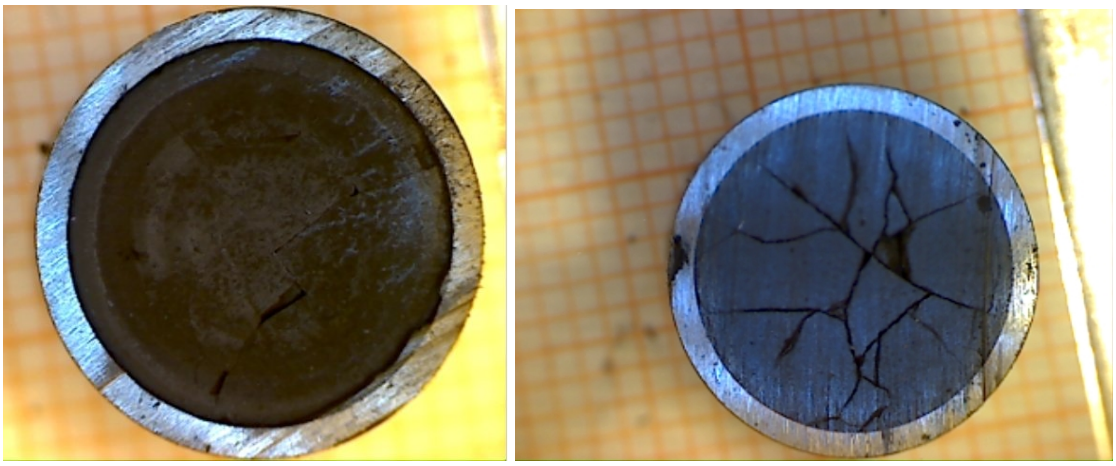


Figure 4: Pellets #3 (left image) and #4 (right image) of fuel rod segment SBS1108-N0204, used in SNF leaching experiments at KIT.

The experiments are carried out in Ti-lined VA autoclaves (total volume 250 ml; Figure 5), with two valves in the lid to allow sampling of gases and of solutions during the experiment. A 19 mM NaCl + 1 mM $NaHCO_3$ solution is chosen as leachant, and the anoxic conditions will be achieved by application of a H_2 + Ar reducing atmosphere within the autoclaves (37 bar Ar +3 bar H_2). At the beginning of each experiment a washing step is performed with the same leachant as described before. At the end of the washing step, aliquots are sampled for further analysis and the solution is completely renewed. In the following static phase of the leaching experiments, small solution aliquots are sampled at different time intervals to determine the kinetics of the IRF release. The leachant is not replenished during the static phase of each leaching experiment.

The solution aliquots are analysed to determine the specific activity of ^{134}Cs, ^{135}Cs, ^{137}Cs, ^{90}Sr, ^{129}I, ^{99}Tc, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu and – to the extend possible - ^{14}C, ^{36}Cl and ^{79}Se. Following analysis methods are applied:

- γ-spectrometry for concentration measurements of ^{134}Cs, ^{137}Cs. The separation of these radionuclides from the original solution is described in Grambow et al. (1996). Afterwards the analysis is performed by means of Ge-detectors (EGC-15-185-R and GX3018, Canberra Industries Inc, Meriden, USA).
- Liquid scintillation counting (LSC) using a Packard Tri-Carb 3110TR Low activity scintillation analyser (Perkin Elmer INC, Waltham, USA) to quantify: ^{90}Sr, ^{241}Pu, ^{14}C. Solution aliquots are homogenised with specific LSC-cocktail (Ultima Gold XR, Perkin Elmer for ^{90}Sr and ^{241}Pu; Hèonic Fluor for ^{14}C) before measurement.

- α-spectrometry is used to determine the amount of ^{238}Pu, ^{239}Pu, ^{240}Pu using an analysis chamber with a S100 field channel analysator (^{238}Pu, $^{239/240}$Pu) and passivated implanted planar silicon (PIPS) detectors (Canberra 74/01, Canberra Industries Inc, Meriden, USA) which combine high resolution and low backgrounds in a rugged alpha detector with active areas up to 1200 mm^2. It integrates into one package a stainless steel vacuum chamber for low backgrounds and ease of cleaning, a vacuum gauge, detector bias supply, preamp/amplifier, pulser, discriminator, counter, and digital display. A stainless steel shelf and sample holder are included for reproducible detector/sample spacing, which is user selectable from 1 to 49 mm, in 4 mm increments
- Inductively coupled plasma mass spectrometer (ELAN 6100, Perkin Elmer Inc, Waltham, USA) to measure the concentrations of ^{238}U, ^{99}Tc, ^{237}Np.

In parallel to the sampling of solution aliquots, the gas phases of the autoclaves are frequently sampled. The main interest of the gas analyses is in the release of the fission gases, Kr and Xe, as well as the radiolytic gases, H_2 and O_2. The autoclaves inside the shielded box-line are connected via a tubing – valve system to the stainless steel single-ended miniature sampling cylinder (Figure 6) outside the shielded box-line. After collecting the gas phase of an autoclave, the gases are analysed by means of a quadrupole gas mass spectrometer (gas MS). The gas MS (GAM400, In Process Instruments, Bremen, Germany) is equipped with a Faraday and SEV detector and a batch inlet system (Figure 6). The batch-type gas inlet system is optimised for low gas sample consumption. Within the gas dosage and inlet system, the total gas pressure is controlled at four successive positions. It applies three different expansion-volumes to charge relatively low gas contents in the desired pressure range. Calibration of the gas MS analysis is performed in the same pressure range as the respective range for analyses of the sample aliquots.

Figure 5*: Images of Ti-lined VA autoclaves used in FIRST-Nuclides leaching experiments at KIT. The left image shows an autoclave during a pressure test before transferring it to the KIT-INE shielded box line; the right image shows the autoclave during the leaching experiment with pellet #3 of SBS1108-N0204 with under a gas atmosphere of 37 bar Ar +3 bar* H_2*.*

Figure 6: **Images of the gas sampling device and gas-MS analysis system used during the SNF leaching experiments at KIT. An exemplary stainless steel single-ended miniature sampling cylinder used to collect the gases from the autoclaves is shown in the left image. A gas sampling cylinder connected to the GAM400 quadrupole gas mass spectrometer is shown in the right image.**

3. JRC-ITU and CTM - SNF sample characterisation, methodologies and tools

D. Serrano-Purroy, R. Sureda Pastor, J.Y. Colle, O. Beneš, F. Naisse, T. Wiss, R.J.M. Konings, D.H. Wegen, D. Papaioannou, R. Gretter, R. Nasyrow, V.V. Rondinella and J. P. Glatz

JRC-ITU selected two standard BWR fuel rods for investigations by JRC-ITU and CTM. Both fuels were manufactured with UO_2 material, having standard ^{235}U enrichments. During reactor operation average burn-ups of 42 and 54 GWd·(t HM)$^{-1}$ were reached, respectively. An agreement on transfer of ownership of the 54 GWd·(t HM)$^{-1}$ BWR (denoted as "BWR54") fuel to JRC-ITU was signed and irradiation data are available. With respect to the 42 GWd·(t HM)$^{-1}$ BWR fuel (denoted as "BWR42") the agreement is expected to be signed in near future. Characteristic data of both BWR fuels, includingupdated operational parameters, are given in Table 14 of chapter 10. JRC-ITU and CTM sample pellets and powders from the BWR fuels. 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. The preparation of pellet and powder samples of the BWR54 fuel is in progress. JRC-ITU performed non-destructive analyses, gas sampling, cutting and sampling of fuel pellets from KIT's PWR fuel rod segment SBS1108-N0204. Samples of this fuel rod segment were prepared for ceramography at JRC-ITU.

3.1. Preparation of samples of spent BWR fuel rods by JRC-ITU and CTM

For both selected BWR fuels, two types of fuel samples with different morphologies, fuel cladded segments and powder samples are provided for leaching experiments within workpackage 3 of FIRST-Nuclides. The preparation and physical characterization of all samples referred to in this report are performed in the hot cell laboratories at JRC-ITU.

Cladded segments

The selection of the representative disc sample from the original fuel rod was chosen after previous examination of the corresponding gamma scanning. In our case, we selected a region where the activity distribution was as constant as possible for all the samples to be cut. Moreover, the gamma-scan was used in the cutting operations to ascertain the length and the exact position of the pellets along the fuel rod. For this purpose, an initial cut of ~20 mm was performed longitudinally along the "mother" segment. Figure 7 shows the longitudinal section where it can be clearly observed the dishing at the pellet boundary for the BWR54 (a) and the BWR42 (b) samples, respectively.

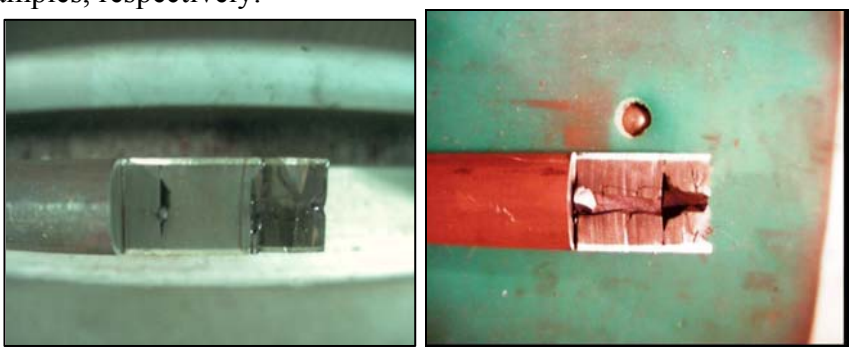


Figure 7: Longitudinal section to determine pellet boundaries in a) BWR54 and b) BWR42 fuel segments.

In both cases two cladded fuel discs samples to be used in FIRST-Nuclides were cut with a length of approximately of 2 mm at the middle of the pellet avoiding inter-pellets zones:

- One cladded segment for leaching experiments,
- One cladded disc, cut off consecutive to the previous one, for chemical determination of the experimental inventory.

These samples are characterised at the end of the corrosion leaching experiments.

Powder samples

The second type of solid samples used for leaching experiments constitute of powders from different radial positions of the fuel pellet: One obtained from the inner central part of the pellet, which is referred to as "CORE sample" and a second one obtained from the outer part of the pellet, referred to as "OUT sample". Powder samples were prepared in a dedicated procedure using hot cell technology at JRC-ITU. The different stages are numbered as follows:

i) *Cutting*: A segment of the selected fuel pin was cut into approximately 3-4 cm long pieces (Figure 8).

ii) *Drilling* out the inner part (ø<3mm), the so called CORE fraction (Figure 9). The drilling was carried out in the centre of the fuel in a number of consecutive steps to minimise the friction between the tip of the drill bit and heating-up the fuel.

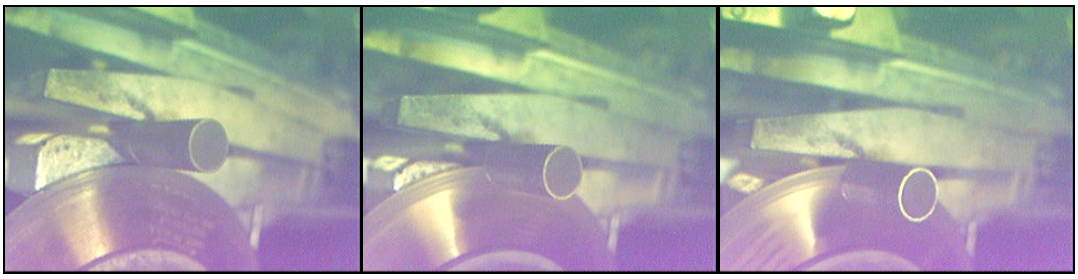


Figure 8: Cutting of cladded segments (BWR54 sample).

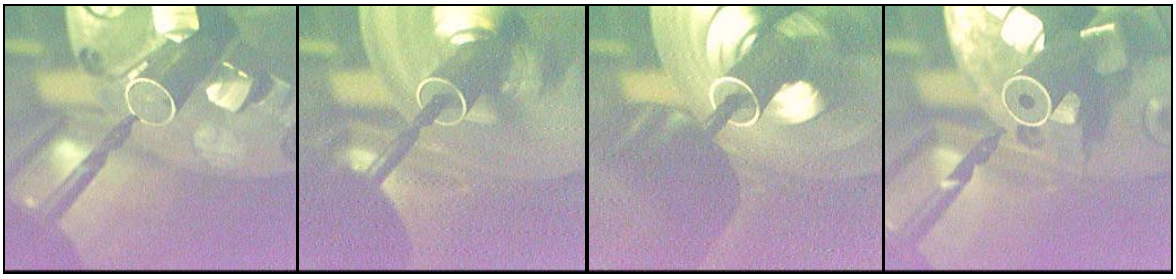


Figure 9: Drilling of cladded segments (BWR54 sample).

The OUT samples are prepared from the outer part of the fuel and are enriched in high burn-up structure. They were obtained by consecutive drilling with increasing diameter (up to 9 mm) of the drill bit. The fuel remaining inside the cladding is then detached by mechanical pressing. The OUT sample contains all fuel fractions collected from diameter 7 mm to the cladding but not the cladding itself.

iii) Sieving and Milling: Powder samples are milled (M20 KA, IKA GmbH, Germany) and sieved (Vibrax-VXR, IKA GmbH, Germany) to a fraction between 50 and 100 μm.

iv) SEM characterisation: Selected fractions are then characterised by SEM (JEOL 6400, JEOL LTd., Japan) in order to establish the correct average particle size.

The complete procedure can be found elsewhere (González-Robles, 2011; Rondinella *et al., 2*008; Serrano-Purroy *et al.,* 2007; Serrano-Purroy *et al.,* 2013).

3.2. Radionuclide composition of spent BWR fuel samples studied by JRC-ITU and CTM

The nuclide inventory of the two BWR fuels will be determined theoretically and experimentally. In the following the theoretical inventory is reported. Inventory calculations of BWR42 and BWR54 fuels were performed using the Origen-ARP code (ORIGEN-ARP, 2000) taking into account the corresponding operational parameters, see Table 14. The code is only used to determine total SNF inventories. Local inventories, corresponding to the powder SNF samples can only be determined experimentally.

3.3. Analytical methodology for inventory determination of spent BWR fuel samples at JRC-ITU

The inventory of all fuel fractions will be determined experimentally by acid dissolution. Different experimental procedures are used for SNF powder and for cladded fuel.

Powder samples

For each powder fraction, CORE and OUT, four independent samples of (30 - 60) mg are dissolved in Parr bombs by acid digestion at 180 ºC for 12 hours in (30 ± 5) mL of a mixture with HNO_3: HCl (4:1). Teflon containers are used to avoid surface interactions. The dissolution is considered to be complete when no residual solid can be observed by direct visual inspection of the vessel. After complete dissolution of the SNF powder, two duplicate samples are taken from each Teflon container to elucidate possible cross-contaminations. The first is taken directly from the container and the second is taken from a polyethylene bottle after transfer of the solution. The solution is further diluted (by weighing) in 1M nitric acid according to the expected concentrations and dose rate for posterior analysis by ICP-MS.

Cladded segment samples

In addition, the total SNF inventory corresponding to the cladded fuel samples will also be experimentally determined. In this case, the cladded samples are dissolved in a closed flask with 250 mL of an acid mixture HNO_3:HCl (4:1) during 8 hours at 120°C under reflux cooler to avoid liquid evaporation. The cladding remains in the dissolution vessel. Once the pellet dissolution is finished, the solution is filtered through a porous micro-filter-candle between 100-160 μm diameter (Robu glass, Germany) and finally recovered in a 250 mL glass bottle for interim storage (Figure 10). The cladding is recovered and stored for future examinations. Dissolution aliquots, normally second and third dilutions are analyzed following the same procedure as for powder samples.

Burn-up determination

In addition to the experimental inventories, the same samples are used for ^{148}Nd burn-up determinations (De Regge, 1977; ASTM, 1969). Another methodology which can be used

with a higher uncertainty is based on ICP-MS determinations without previous separations using Nd isotopes, free of interferences.

Table 1: Origen-ARP inventory determination for BWR54 (left) and BWR42 (right) samples

	BWR54		**BWR42**	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{85}Rb	156.9	31.41	147.9	32.61
^{87}Rb	342.6	68.59	305.7	67.39
Rb (total)	499.5		453.6	
^{86}Sr	0.9	0.08	0.6	0.07
^{88}Sr	475.6	42.67	425.6	46.82
^{90}Sr	638.2	57.26	482.7	53.11
Sr (total)	1114.7		908.9	
^{89}Y	643.5	99.97	576.5	99.98
^{90}Y	0.2	0.03	0.1	0.02
Y (total)	643.7		576.6	
^{90}Zr	156.4	3.00	229.0	4.83
^{91}Zr	844.5	16.20	757.3	15.99
^{92}Zr	917.2	17.59	818.5	17.28
^{93}Zr	1011.1	19.39	906.2	19.13
^{94}Zr	1122.6	21.53	994.4	20.99
^{96}Zr	1161.4	22.28	1032.0	21.78
Zr (total)	5213.1		4737.4	
^{95}Mo	1077.2	22.26	973.8	22.75
^{96}Mo	88.4	1.83	60.5	1.41
^{97}Mo	1135.4	23.46	1008.0	23.54
^{98}Mo	1239.4	25.61	1093.0	25.53
^{100}Mo	1299.3	26.85	1146.0	26.77
Mo (total)	4839.6		4281.3	

Table 1 (continued)

	BWR54		BWR42	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{99}Tc	1086.9	100.00	1010.0	100.00
Tc (total)	1086.9		1010.0	
^{100}Ru	218.7	7.10	143.0	5.39
^{101}Ru	1141.5	37.07	1018.0	38.38
^{102}Ru	911.5	29.60	790.9	29.82
^{104}Ru	804.4	26.12	700.4	26.41
^{106}Ru	3.2	0.10	0.0	0.0
Ru (total)	3079.3		2652.4	
^{103}Rh	579.6	100.00	569.6	100.00
Rh (total)	579.6		569.6	
^{104}Pd	441.2	19.69	319.0	17.26
^{105}Pd	614.4	27.42	533.7	28.88
^{106}Pd	547.4	24.43	461.5	24.97
^{106}Pd	337.3	15.06	286.1	15.48
^{108}Pd	228.9	10.22	189.2	10.24
^{110}Pd	71.2	3.18	58.6	3.17
Pd (total)	2240.4		1848.1	
^{109}Ag	66.8	100.00	57.3	100.00
Ag (total)	66.8		57.3	
^{110}Cd	38.5	33.67	25.9	29.09
^{111}Cd	35.8	31.32	29.9	33.51
^{112}Cd	18.0	15.79	14.8	16.63
^{113}Cd	0.1	0.08	0.1	0.14
^{113M}Cd	0.3	0.23	0.1	0.15
^{114}Cd	15.2	13.34	12.8	14.35
^{116}Cd	6.4	5.57	5.5	6.14
Cd (total)	114.3		89.1	

Table 1 (continued)

	BWR54		BWR42	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{115}Sn	0.3	0.35	0.3	0.36
^{116}Sn	5.1	5.59	3.9	4.94
^{117}Sn	6.9	7.49	6.0	7.63
^{118}Sn	6.8	7.44	5.9	7.58
^{119}Sn	7.2	7.84	6.3	8.04
^{120}Sn	5.0	5.40	4.3	5.50
^{121M}Sn	0.1	0.07	0.1	0.07
^{122}Sn	10.6	11.54	9.0	11.56
^{124}Sn	14.0	15.22	12.0	15.31
^{126}Sn	35.9	39.04	30.5	39.01
Sn (total)	91.9		78.1	
^{121}Sb	4.9	22.57	4.2	26.59
^{123}Sb	12.8	59.21	11.0	70.21
^{125}Sb	3.9	18.22	0.5	3.20
Sb (total)	21.6		15.6	
^{122}Te	0.4	0.11	0.3	0.09
^{124}Te	0.8	0.20	0.6	0.15
^{125}Te	24.0	5.87	23.3	6.51
^{125M}Te	0.1	0.01	0.0	0.00
^{126}Te	0.9	0.23	0.7	0.20
^{128}Te	56.8	13.89	48.9	13.63
^{130}Te	325.7	79.70	284.7	79.41
Te (total)	408.6			
^{127}I	67.0	22.41	59.0	22.37
^{129}I	232.1	77.59	204.8	77.63
I (total)	299.1		263.8	
^{133}Cs	1535.6	42.03	1418.0	46.93
^{134}Cs	27.2	0.74	1.5	0.05
^{135}Cs	585.3	16.02	476.8	15.78
^{137}Cs	1505.6	41.21	1125.0	37.24
Cs (total)	3653.6		3021.3	

Table 1 (continued)

	BWR54		BWR42	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{134}Ba	295.7	11.49	233.7	9.58
^{135}Ba	1.2	0.04	0.5	0.02
^{136}Ba	37.9	1.47	25.2	1.03
^{137}Ba	333.7	12.97	497.8	20.41
^{138}Ba	1905.4	74.03	1682.0	68.96
Ba (total)	2573.9		2439.2	
^{139}La	1752.9	100.00	1562.0	100.00
La (total)	1752.9		1562.0	
^{140}Ce	1838.8	52.73	1617.0	52.62
^{142}Ce	1646.6	47.22	1456.0	47.38
^{144}Ce	1.5	0.04	0.0	0.00
Ce (total)	3486.9		3073.0	
^{141}Pr	1623,7	100,00	1448.0	100.00
Pr (total)	1623,7		1448.0	
^{142}Nd	44,4	0,79	28.6	0.58
^{143}Nd	998,3	17,81	1019.0	20.53
^{144}Nd	2068,4	36,91	1702.0	34.30
^{145}Nd	944,1	16,84	868.0	17.49
^{146}Nd	784,1	13,99	664.5	13.39
^{148}Nd	503,8	8,99	449.1	9.05
^{150}Nd	261,6	4,67	231.2	4.66
Nd (total)	5604,7		4962.4	
^{147}Sm	281.4	23.58	306.5	28.33
^{148}Sm	235.8	19.76	183.7	16.98
^{149}Sm	3.2	0.26	4.2	0.39
^{150}Sm	414.8	34.75	382.5	35.35
^{151}Sm	14.3	1.20	13.9	1.28
^{152}Sm	187.3	15.69	141.7	13.10
^{154}Sm	56.9	4.76	49.5	4.57
Sm (total)	1193.7		1082.0	

Table 1 (continued)

	BWR54		BWR42	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{151}Eu	0.7	0.31	1.6	0.85
^{153}Eu	173.5	80.38	167.0	89.25
^{154}Eu	37.1	17.20	17.0	9.09
^{155}Eu	4.5	2.10	1.5	0.80
Eu (total)	215.9		187.1	
^{152}Gd	0.1	0.04	0.1	0.04
^{154}Gd	30.1	13.62	40.2	21.85
^{155}Gd	6.2	2.78	10.1	5.49
^{156}Gd	144.3	65.27	102.3	55.59
^{157}Gd	0.2	0.08	0.2	0.09
^{158}Gd	36.5	16.49	28.3	15.38
^{160}Gd	3.8	1.71	2.9	1.56
Gd (total)	221.1		184.0	
^{234}U	153.5	0.02	180.8	0.02
^{235}U	4517.7	0.55	7828.0	0.83
^{236}U	5363.9	0.65	4769.0	0.50
^{238}U	812297.3	98.78	932700.0	98.65
U (total)	822332.4		945477.8	
^{237}Np	586.2	100.00	597.5	100.00
Np (total)	586.2		597.5	
^{238}Pu	318.9	3.57	233.4	2.37
^{239}Pu	3894.4	43.61	5698.0	57.77
^{240}Pu	2659.0	29.78	2467.0	25.01
^{241}Pu	1007.1	11.28	785.3	7.96
^{242}Pu	1050.3	11.76	680.2	6.90
Pu (total)	8929.8		9863.9	
^{241}Am	376.5	59.95	801.1	84.71
^{242M}Am	0.8	0.12	0.8	0.08
^{243}Am	250.8	39.93	143.8	15.21
Am (total)	628.1		945.7	

Table 1 (continued)

	BWR54		BWR42	
	Fuel composition µg/g fuel	Elemental composition wt%	Fuel composition µg/g fuel	Elemental composition wt%
^{243}Cm	0.6	0.65	0.4	1.06
^{244}Cm	89.4	90.35	29.9	88.05
^{245}Cm	3.7	3.73	1.8	5.26
^{246}Cm	5.2	5.22	1.9	5.60
Cm (total)	98.9		34.0	

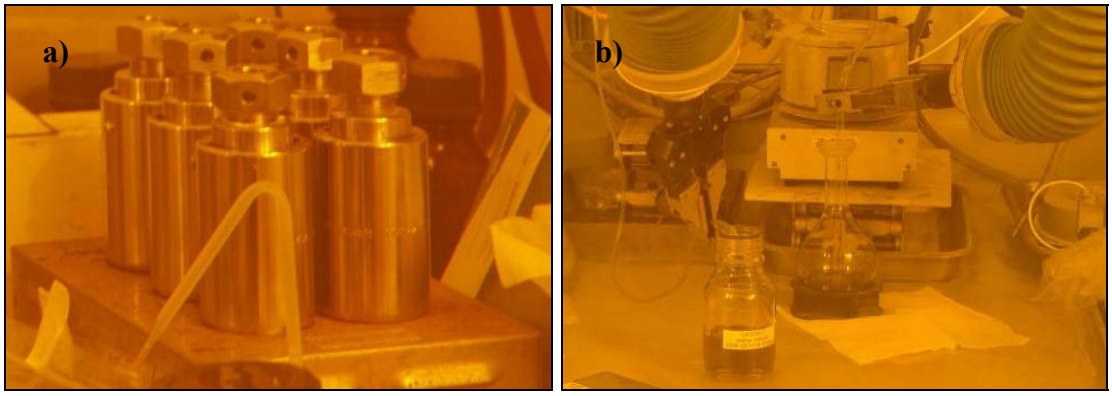


Figure 10: Experimental equipment used for inventory determinations in SNF samples. (a) Bomb dissolution for powder samples CORE and OUT and (b) dissolution of cladded SNF samples (images from González-Robles, 2011).

3.4. JRC-ITU setups for very high temperature kinetic and quantitative measurements of fission products released from nuclear materials

High temperature kinetics and quantitative measurements of fission products (FPs) released during laboratory anneals of irradiated material are performed using a combination of furnace and mass-spectrometers. The release profiles provide information on the source term or location of the FPs and quantitative measurement of the fission gases (FGs) allow to evaluate the retention capacity of the material. A description of the instrumentation is provided supported by some examples. A further facility dedicated to the study of the FGs release exists at CEA Cadarache. The facility is called MERARG (French acronym for Fission Gas Release Study Facility by Annealing). A brief comparison focused on the main principles is made between the two installations.

Knudsen Effusion Mass Spectrometry

Knudsen Effusion Mass Spectrometry is a powerful technique to study the high temperature thermodynamic and kinetic behavior of materials (Drowart *et al.* (2005), Hilpert *et al.* (1991)). It has been used at the JRC-ITU for more than 20 years to generate fundamental data and understanding for nuclear fuel safety studies with standard measurements of partial vapor pressure and vaporization behaviour (Gotcu-Freis *et al.* (2011a), Gotcu-Freis *et al.* (2011b), Gotcu-Freis *et al.* (2011c), Hiernaut *et al.* (2004)), ionization efficiency curves and ionization

and dissociation energies (Capone *et al.,* 1999)) on radioactive materials and actinide materials. It has been also intensively used for kinetic studies like fission products and actinide release behaviour from irradiated fuel (Hiernaut *et al.,* 2001; 2005; 2008a; 2008a; 2008c; 2009; Capone *et al.*, 1996; Colle *et al.* (2006), Damen *et al.,* (2002), Ronchi *et al.,* (2004), Wiss *et al.,* 2004; 2006; 2007). All those applications have some particularities in common: Specific requirements related to the radioactivity of the samples and the very high temperatures to vaporize actinide compounds, especially for oxides. The first point requires a setup built in a glove box and thus it is necessary to avoid complex design and it is also necessary to minimize the waste with a highly reliable system. The second point requires a very high temperature furnace for the Knudsen cell.

Because quantitative gas measurement can difficultly be done with the KCMS, it is complemented by an independent Quantitative GAs MEasurement Setup (Q-GAMES) able to measure within few percent the absolute quantities of released gas.

The standard procedure for the measurement of release from irradiated fuels is as follows; The 5 to 10 mg sample is introduced into the Knudsen cell, the vessel is evacuated and kept in vacuum at least for 24 hours in order to degas the system enough to have a vacuum in the order of $1x10^{-8}$ mbar, and thus a reasonable background for the mass spectrometer. The sample is then heated up with a ramp of 10 $K \cdot min^{-1}$ up to total evaporation of the uranium oxide matrix. During the all annealing, all fission products and actinides are sequentially measured with the mass spectrometer with one second per mass every 2-3 minutes. During the all annealing, the gas released is collected from the discharge of the KCMS vacuum pump to the sample chamber of the Q-Games. The collected gas (He, Kr, Xe) is continuously cleaned and measured by the Q-Games mass spectrometer. At the end of the experiment spikes are inserted in the sample chamber in order to make a quantitative measurement.

Knudsen Cell Mass Spectrometer at JRC-ITU

Figure 11 shows the actual setup of the KCMS system installed at JRC-ITU. It is equipped with quadrupole mass spectrometer (QMG422 from Pfeiffer Vacuum) which offers a mass range of 1 to 512 amu. It has a cross beam electron bombardment ion source, an axial Faraday cup and a secondary electron multiplier located at 90° to the filter axis used in ion current measurement mode. The furnace is heated by a resistance coil and shielded by seven W and Ta shields. The furnace can be heated with temperature ramps such as 10 or 30 $K \cdot min^{-1}$ or at constant temperature. The Knudsen cell can be equipped of a gas inlet allowing work with up to 100 Pa gas in the cell, thus simulating oxidizing conditions by varying the oxygen partial pressure. The system is equipped with a single blend between the ion source chamber and the furnace chamber and requires calibration for each measurement usually done with mass loss technique of a silver sample. A chopper allows making the background measurement and a liquid nitrogen (LN2) cold trap significantly improves the vacuum in the ion source chamber and thus the mass spectra background. The system is pumped with standard turbo pump backed up with dry primary pump. The vessel (out of the turbo pumps) can be baked up to 80°C by heating up the cooling water. The complete system is placed in an alpha tight glove box surrounded by movable 5 cm lead gamma shields.

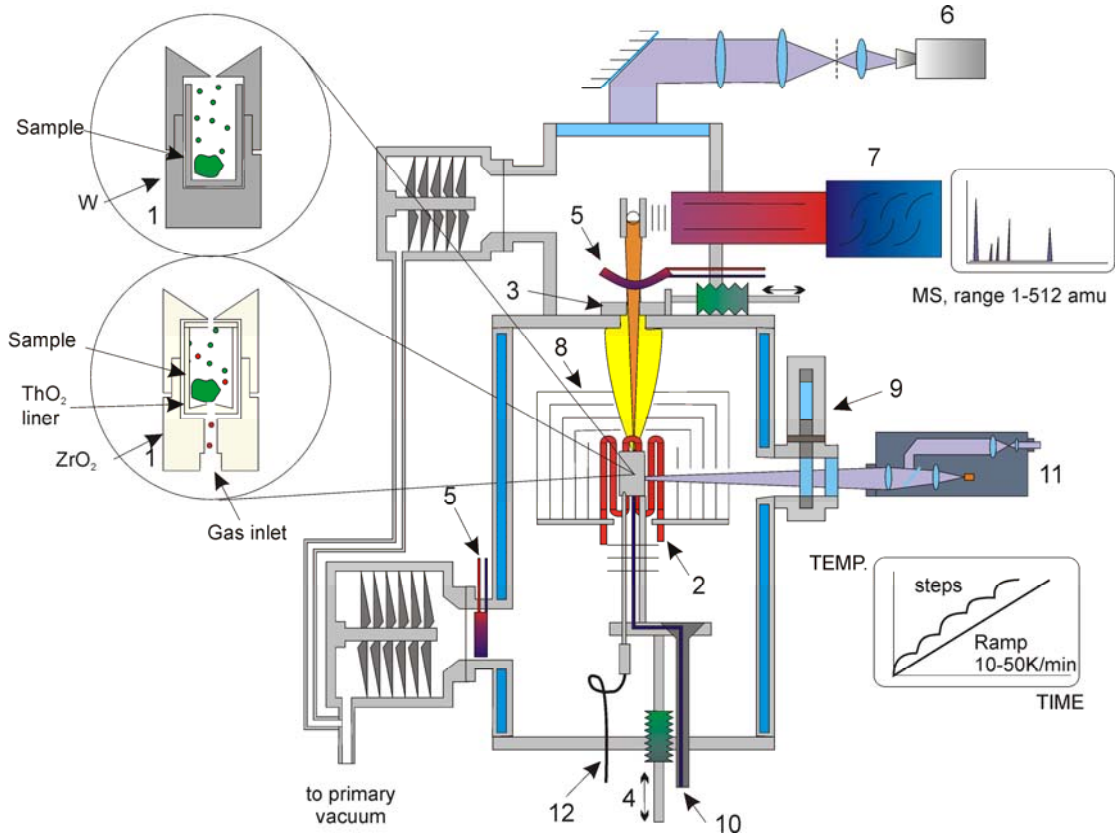


Figure 11: Schematic of the actual KCMS system at JRC-ITU. 1: Knudsen cell, 2: heating coil, 3: chopper, 4: lift, 5: LN2 cold trap, 6: camera, 7: mass spectrometer, 8: thermal shields, 9: revolving windows, 10: gas inlet, 11: pyrometer, 12: Thermocouple.

Quantitative Gas Measurement Setup

The principle of the Q-GAMES (Figure 12) consists of collecting all the gas released from a sample in a sample chamber (1). The collection can last from minutes to hours. This gas is then purified by using the proper combination of a cold trap (2), a getter pump (3), or a plasma discharge (4) to avoid or reduce the presence of parasite masses. A quantitative measurement of the gas collected into the sample chamber (1), is achieved by sampling the gas through the inlet (5) and the micro-valve (13) to the mass spectrometer chamber (7) and comparing its signals with the signals of a gas spike of the same nature using a quadrupole mass spectrometer equipped with an electron bombardment ion source (6). During the measurements process, the whole pumping system is working in close circuit, after analysis in the mass spectrometer chamber (7) the gas is pumped through the feedback (8) back to the sample chamber (1), this to avoid depletion of the gas contained in the sample chamber and to improve the sensitivity of the system.

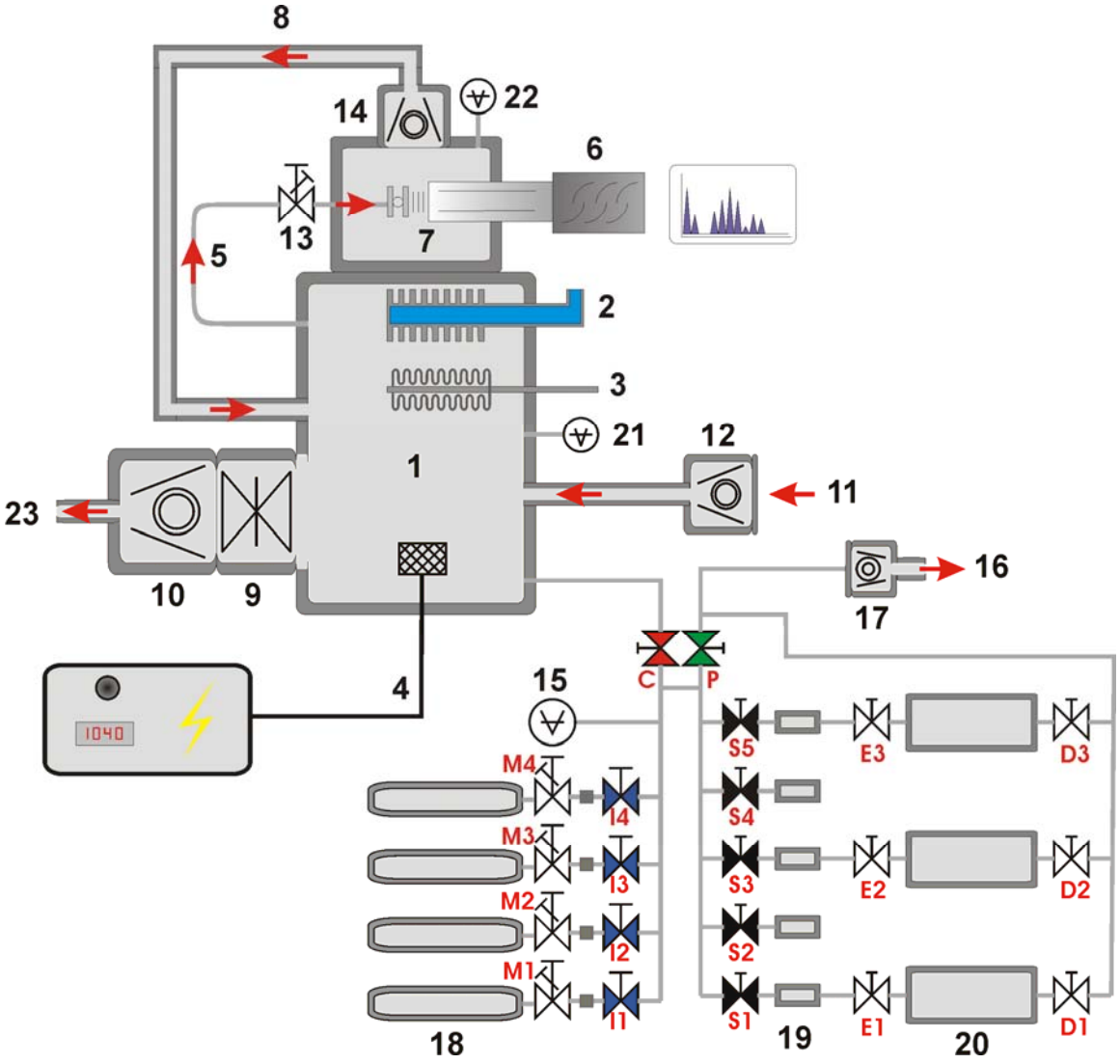


Figure 12: Simplified scheme of the Q-GAMES experimental setup (labels are explained in the text).

Sample collection and cleaning: The system consists of an ultra high vacuum (UHV) vessel - the sample chamber (1)- with a volume of about 6 liters, evacuated (<10-7 mbar) prior to the experiment via a vacuum gate (9) with a turbo pump (10). The sample gas is collected from the gas discharge set up (11), and compressed in the sample chamber (1) with a high compression ratio (Ar: >1011, H_2: ~ 10^5) hybrid turbo molecular drag pump (Pfeiffer vacuum TMU 071) (12). The gas collected in the sample chamber (1) contains the gas to measure but also undesirable gases originally adsorbed on the heated parts of the furnace or on a contaminated sample like air, water vapor and hydrocarbons. Those last gaseous species can actually represent the majority of the gas sample by orders of magnitude. They can potentially perturb the mass spectrometer measurements because gases like N_2, O_2, CO_2, CO, H_2O and large hydrocarbon molecules produce peaks in a range going from 1 to >300 amu perturbing the background and the measurement of masses of interest. In the low mass range they produce H, H_2, HD, D_2, those disturbing the measurements of ^{3}He, ^{4}He, T, in higher mass range, the hydrocarbon mass spectrum interferes directly on the measurement of the heavier gases like krypton (78-86 amu) and xenon (124-136 amu).

To neutralize or remove those unwished residual gases, three different systems are used:

-A getter pump (3) (SAES Getters GP50 with a sorption capacity of H_2: 320 Torr·L, CO 290 Torr·L and a pumping speed of ~100 $L \cdot s^{-1}$ depending of the gas and the quantity already adsorbed) is used to pump active gases N_2, O_2, CO_2, CO, H_2O, C_nH_m,… as well as H_2, DH and DD.

-A Liquid nitrogen (LN_2) cold trap (2) is used to condensate water and hydrocarbons (not appropriate for Kr and Xe measurements).

-A plasma generator (4) is used to break hydrocarbons molecules in single atoms or smaller molecules.

Depending on the needs, a combination of the different systems can be used successively or simultaneously.

Sample Measurement: Once cleaned, the gas sample is brought with an inlet (5) equipped with a micro valve (13) from the sample chamber (1) into the closed ion source of the mass spectrometer (6) located in a second UHV vessel, the mass spectrometer chamber (7), overtopping the sample chamber (1). The mass spectrometer (6), a quadrupole QMG422 from Pfeiffer Vacuum, is equipped with an electron bombardment gas tight ion source and a 90° SEM (secondary electron multiplier). The sample gas once measured is pumped from the mass spectrometer chamber (7) back to the sample chamber with two turbo pumps installed in series (14), in order to obtain a very high compression ratio (total compression ratio for N_2:10^{20}, He: 10^{12} and H_2:10^9). With this system of feedback (8) the sample gas is never depleted and the measurement can be thus continually done throughout the release process, giving information about the kinetics of the release and allowing a long time measurement.

MERARG loop at CEA Cadarache

At the Cadarache CEA centre, an annealing test facility, called MERARG (French acronym for Fission Gas Release Study Facility by Annealing) is settled in one of the hot cell laboratories (the LECA-STAR one). This experimental setup has been described in details elsewhere (Pontillon (2011)). Very generally, the corresponding key components are the induction furnace located in a shielded hot cell, the gamma spectrometry device, and the gloves box where gas coming out of the furnace is trapped.

The furnace chamber is a quartz tube; the sample is put into a crucible (Mo,Wor Pt depending on the type of experiments) which is coupled to the high frequency (50 kHz) coil placed around the quartz tube, and heated up by induction. During the whole annealing, the specimen is swept by a regulated gas flow (helium or air).

Released fission gases are carried away with the sweeping gas; it flows through aerosol filters before passing in the delay chamber situated in front of the gamma spectrometer. The gas flow ends in the gloves box where fission gases are trapped. The counting chamber and the detector are located in a shielded chamber. The aim of such measurements is to follow in a set and given point of an experimental loop, the evolution of the activity signal over time. The detector is a germanium P-type crystal. Radioactive fission gas activity is monitored by the gamma spectrometer. By taking into account fission gas dilution and flowing time between the furnace and the counting chamber, real fission gas release kinetics (i.e. at the sample position) can be reconstructed from the measured one. Furnace temperature, gas flow and pressure are continuously monitored. The sample temperature is evaluated by two ways:

(i) a thermocouple placed at the lower part of the crucible measures its temperature;

(ii) a pyrometer gives another measurement for temperatures above 1000 °C by direct sighting into the sample chamber (Menegon (2008)).

To evaluate the grain boundary inventories, the MERARG loop is combined with the ADAGIO protocol (inter-granular gas inventory determination). The ADAGIO global process has been already described in detail elsewhere (Ravel (2000), Pontillon (2007), Pontillon (2009)). According to the ADAGIO technique, the intra- versus inter-granular fission gas inventory in UO_2 fuels can be evaluated by controlled fuel oxidation. Pre-irradiated fuel samples are annealed under air flow at low temperature (between 380 °C and 450 °C) in order to induce the oxidation of UO_2 into U_4O_9c (called the 'oxidation phase' throughout the paper). As this oxidation step occurs preferentially along the grain boundaries and due to the corresponding volume expansion (caused by UO_2 to U_4O_9c phase change), grain boundary separation occurs. As a consequence, the inter-granular gas is released (Ravel (2000)) and the intra-granular gas contribution is determined by the ^{133}Xe release. After this step, the temperature is increased to extract the remaining gas inventory. During these operations, the ^{85}Kr and ^{133}Xe release are continuously monitored by online gamma spectrometry.

Summary

The KCMS coupled to the Q-GAMES and the MERARG facility together with the ADAGIO protocol are tools allowing the measurement of fission gas release from irradiated fuels during laboratory annealing. The principle of measurements differs between the two techniques. The annealing in the KCMS and measurements in the Q-GAMES are performed in vacuum although measurements under oxidative conditions are possible, and the sample can be heated up to vaporization allowing the absolute quantitative determination of the full inventory. The MERARG facility associated to the ADAGIO process uses an oxidation with assessed kinetics to measure the release of fission gases.

3.5. Preparation of samples of the fuel rod segment from PWR Gösgen at JRC-ITU

Pellets have been sampled from the PWR fuel rod segment SBS1108-N0204 at the hot cell facility of JRC-ITU. The specimens were carefully examined and some features relevant for future investigations were identified. The aim was to prepare SNF samples for leaching experiments at KIT (see section 2.1). For this purpose it was envisaged to cut whole pellets with cladding from the segment. Ideally, the segment was to be cut at the positions of the pellet/pellet gaps formed by the pellets dishings.

After performing a complete non-destructive testing (NDT) analysis of the fuel rod segment the pellet positions could be obtained from the γ-scanning along the segment (Figure 13). Five samples were consecutively dry cut from the top end of the segment and two at the bottom end. Three samples from the top end and one from the bottom end were transported back to KIT for further investigations. A detailed description of the procedure, cutting plans and the characteristics of the specimen are given in Wegen *et al. (*2012; 2013a; 2013b; 2013c). The samples left at JRC-ITU were prepared for ceramography. One of the specimen was additionally cut longitudinally. Then all three samples were embedded in epoxy resin. After curing they were first grinded and then wet polished down to 1 µm.

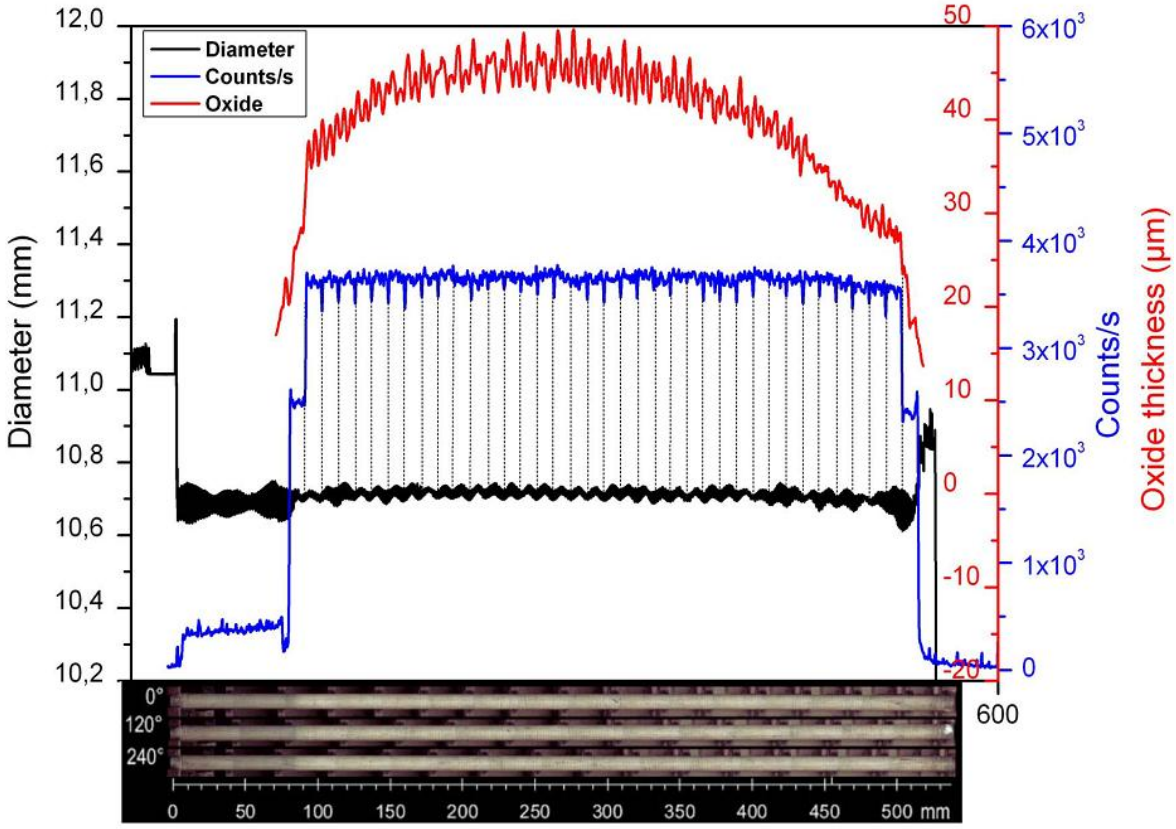


Figure 13: Measured axial outer oxide thickness, γ-scan and diameter of the fuel rod segment SBS1108-N0204 (Papaioannou et al., 2012).

4. JÜLICH – irradiated HTR fuel sample characterisation, methodologies and tools

H. Curtius, H.W. Müskes, N. Liek and D. Bosbach

4.1. Characterisation of spent fuel elements from HTR Petten studied at JÜLICH

In Germany UO_2 containing fuel pebbles were produced for High Temperature Reactors (HTR). These fuel pebbles had a diameter of 60 mm and mainly consist of graphite. Each fuel pebble contained about 9560 so called UO_2-TRISO coated particles with 502 µ diameter UO_2 kernels, having 16.76 ^{235}U wt% enrichment; coating thickness were approximately 92, 40, 35 and 40 µ for porous carbon buffer, inner dense pyrocarbon layer (IPyC), silicon carbide (SiC) and outer dense pyrocarbon layer (oPyC). A coated particle represents a miniature fuel element of about 1 mm in diameter. In Figure 14 a pebble and a UO_2-TRISO coated particle are illustrated.
Five fuel pebbles from the German production line AVR GLE-4/ were used at the High Flux Reactor in Petten for an experiment which was called HFR-EU1bis (Fütterer et al., 2006; 2008). The irradiation of the fuel pebbles at the High Flux Reactor in Petten started on 9 September 2004. After 249 full equivalent power days the experiment was terminated on 18 October 2005. One of these pebbles was named HFR-Eu1bis/2. The main data are derived from Barrachin et al. (2011) and summarized in Table 2. From this irradiated pebble spent UO_2-TRISO coated particles were isolated and used as fuel samples by JÜLICH within the project FIRST-Nuclides.

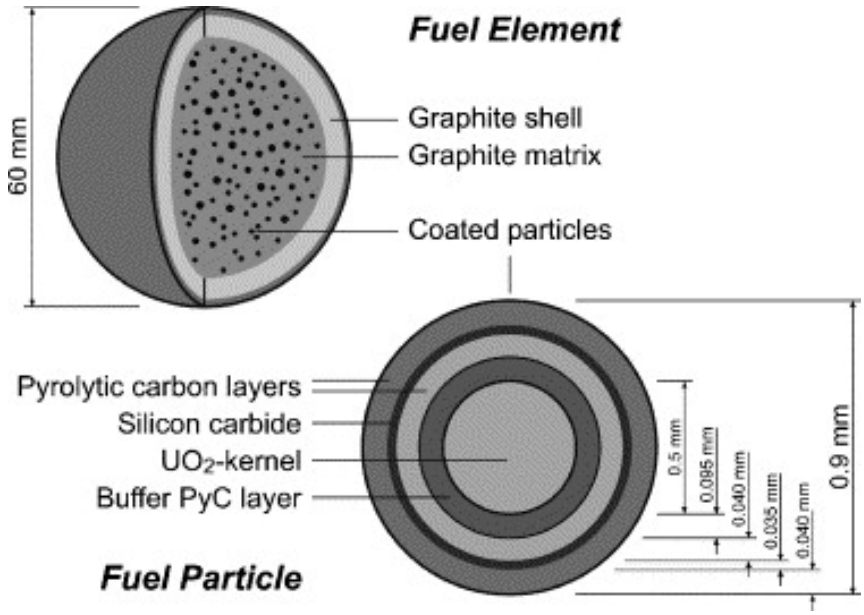


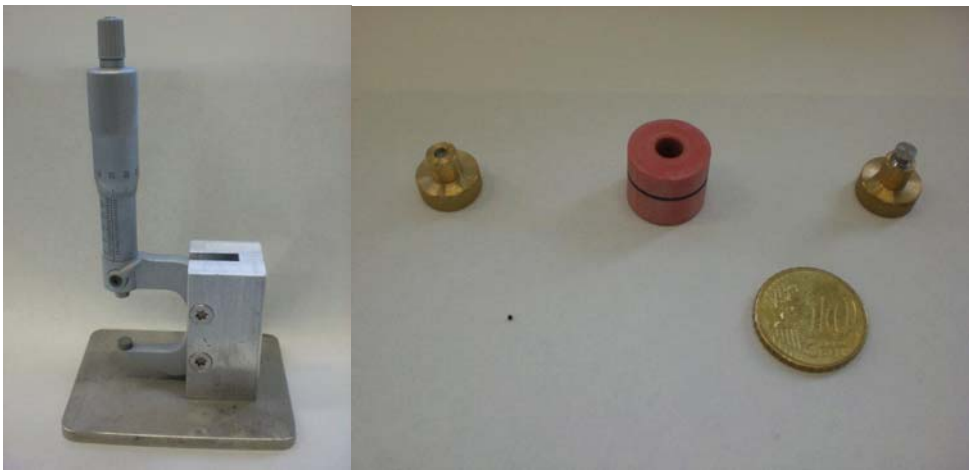
Figure 14: ***Design of a pebble and a TRISO coated fuel particle.***

Table 2: Main irradiation data for the HTR fuel pebble HFR-EU1bis/2

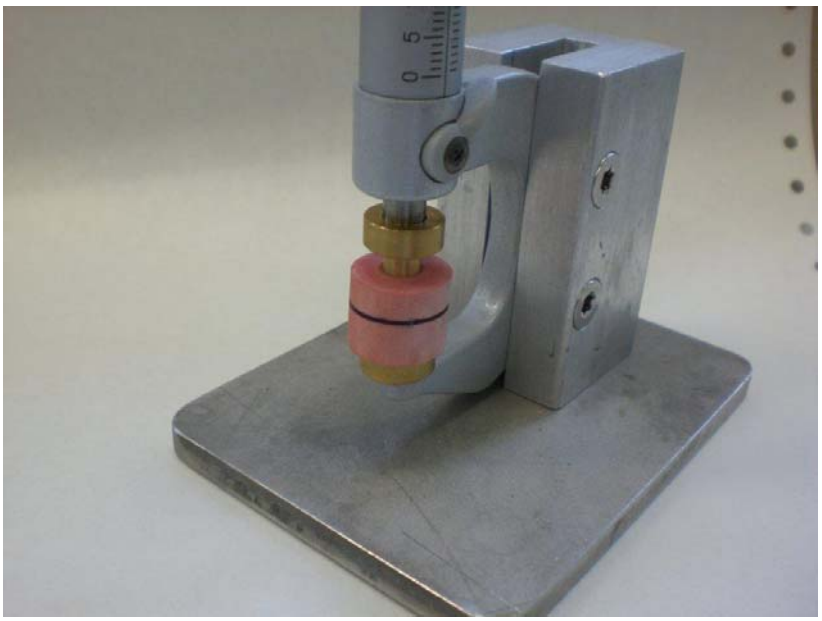
Nuclide	Bq/mgU
Enrichment	16.76 wt% ^{235}U
Irradiation at:	High Flux Reactor Petten
Reactor cycles:	10
Irradiation:	249.55 (efpds)
Thermal fluences:	2.23 x 1025 m^{-2}
Fast fluences:	3.98 x 1025 m^{-2}
Central temperature of pebbles:	1250 °C
Power density:	30 W/cm^3
FIMA:	10.2 %, (95.57 GWd/t)

4.2. Sampling and characterisation of spent HTR fuel samples studied by JÜLICH

Each TRISO coated particle represents a miniature fuel element. Due to the low dose rate no preparation of sub-samples was needed. Nevertheless in view of the planed working activities within WP2 the separation of the fuel kernel from the coatings was required. This was achieved using a modified micro meter screw with a self-manufactured special sample holder (Figure 15). Easely a separation of the fuel kernel from the coatings was achieved.
In order to gain detail knowledge of the microstructure of the fuel kernel investigations with an environmental scanning electron microscope (ESEM) were performed. The fuel kernel was sticked on a sample holder and analysed by SEM technique. A FEI Quanta 200 FEG instrument was used. The instrument is equipped with three detectors. The gaseous large field Secondary Electron-detector was used to characterize the morphology (grain boundaries, porosity). The back Scattered Electron-Detector was used to obtain chemical information due to the z-contrast (atomic number of the elements). With the Apollo X Drift detector the elemental mapping was performed (detection limit 0.1wt%).
SEM investigations were also carried out with a polished specimen. For this examination a UO_2-TRISO coated particle was embedded in a resin (Araldit DBF CH and Aradur HY 951) under vacuum. After 48 h a grinding and polishing process was performed. The wet grinding process was performed using sandpaper (SiC type, 35µm and 22µm). As last step a wet polishing of the sample was performed with sandpaper (SiC-type, 5µm).



a.) b.)



c.)

Figure 15: Instrumentation for separation of irradiated HTR fuel kernels. (a) Micro meter screw, (b) sample holder equipment and (c) complete instrumentation.

4.3. Radionuclide composition of spent HTR fuel samples studied by JÜLICH

With the OCTUPOS code the representative activities of the main radionuclides were calculated after a cool-down period of 1749 days (Table 3). In order to compare the calculated activities (Table 2) of the radionuclides present in a UO_2-TRISO coated particle to measured values, a gamma spectrometric measurement was performed. The irradiated UO_2-TRISO coated particle was placed in a polyethylene tube, closed and the measurement was started. Two different detectors were attached to the gamma spectrometer; a HPGe detector type PGC 2018, Bias: 2500 V positive and a low energy germanium (LEGe) detector. The measure time was 86400 sec. The sample has a distance to the HPGe detector of 60 cm and 50 cm to the LEGe detector. For determination of the radionuclide activities the software Gamma-W

version 2.44 was used. The uncertainties of the values obtained are in the range between 10 to 13%.

Table 3*: Main activities of radionuclides for a coated particle (uranium mass of 0.60476 mg) calculated with the OCTOPUS code (reference date: August 2010)*

Nuclide	Bq/mgU	Bq/CP
H-3	3.92E+04	2.37E+04
Se-79	2.33E+00	1.41E+00
Kr-85	9.25E+05	5.59E+05
Sr-90	6.80E+06	4.11E+06
Y-90	6.80E+06	4.11E+06
Tc-99	1.63E+03	9.86E+02
Ru-106	3.87E+06	2.34E+06
Sb-125	6.53E+05	3.95E+05
Cs-134	2.49E+06	1.51E+06
Cs-137	1.07E+07	6.47E+06
Pr-144	3.41E+06	2.06E+06
Cer-144	3.41E+06	2.05E+06
Eu-154	3.69E+05	2.23E+05
Eu-155	2.02E+05	1.22E+05
U-234	2.35E+02	1.42E+02
U-235	4.88E+00	2.95E+00
U-236	4.28E+01	2.59E+01
U-237	1.78E+02	1.08E+02
Np-237	2.44E+01	1.48E+01
Pu-238	9.50E+04	5.75E+04
Pu-239	2.26E+04	1.37E+04
Pu-240	2.79E+04	1.69E+04
Pu-241	7.26E+06	4.39E+06
Am-241	6.47E+04	3.91E+04
Cm-244	1.99E+04	1.20E+04

After the gamma spectrometric measurement of the CP (coated particle) a crack process using the modified micrometer screw was performed. The coatings were placed in a 20 mL polyethylene vial. Then 10 mL Thorex reagenz (mixture of 13 M HNO_3, 0.05 M HF and 0.1 M $Al(NO_3)_3$ x 9 H_2O) was added and leaching was performed for 7 days. The isolated fuel kernel was placed in a 20 mL polyethylene vial and dissolved completely in 10 mL Thorex reagenz.

Both sample solutions were used for further analytical steps in oder to compare the calculated values to the measured activities and to gain informations about the elemental distribution between coatings and fuel kernel. The results are summarized in Table 4.

First the activity of tritium in the chemical form as HTO was determined. From each sample solution 100 µl were diluted with 9.9 mL water and a subboil process (70°C) was performed. The condensate was collected and 1 mL was used for the β-measurement, performed with a Liquid Scintillation Counter (LSC, TRICARB 2200 A, Packard). Another β-measurement using 0.1 mL from each sample solution was performed and the activity of ^{241}Pu was determined.

Table 4: ***Representative radionuclide activities for a coated particle (CP) (uranium mass of 0.60476 mg) calculated with the OCTOPUS code (date: August 2010), measured activities ** for the coatings and for the fuel kernel from the sample solutions (date August 2012) and measured activities for the intact CP* by γ-measurement (date August 2012).***

Nuclide	Bq/CP	Bq/kernel **	Bq/coatings**	Bq/CP*
H-3	2.37E+04	6.84E+02	n.d.	n.d.
Sr-90	4.11E+06	3.18E+06	0.17E+06	
Y-90	4.11E+06	3.18E+06	0.17E+06	
Tc-99	9.86E+02	1.31E+03	0.97E+01	
Ru-106	2.34E+06	1.33E+05	7.28E+04	3.10E+05
Sb-125	3.95E+05	1.52E+04	3.54E+02	8.10E+04
Cs-134	1.51E+06	2.58E+04	0.80E+06	0.84E+06
Cs-137	6.47E+06	4.00E+05	6.36E+06	6.56E+06
Pr-144	2.06E+06	0.35E+06	1.46E+03	0.37E+06
Cer-144	2.05E+06	0.36E+06	1.18E+03	0.31E+06
Eu-154	2.23E+05	1.14E+05	2.85E+02	1.20E+05
Eu-155	1.22E+05	4.67E+04	1.52E+02	0.49E+05
U-234	1.42E+02	1.82E+02	n.d.	
U-235	2.95E+00	n.d.	n.d.	
U-236	2.59E+01	3.33E+01	n.d.	
Np-237	1.48E+01	n.d.	n.d.	
Pu-238	5.75E+04	7.03E+04	2.45E+02	
Pu-239	1.37E+04	1.09E+04	1.41E+01	
Pu-240	1.69E+04	1.35E+04	1.55E+01	
Pu-241	4.39E+06	2.47E+06	n.d.	
Am-241	3.91E+04	3.11E+04	1.05E+02	2.15E+04
Cm-244	1.20E+04	0.90E+04	1.33E+01	

n.d. : not detected

Then 1 mL from each sample solution was filled in a polyethylene vial and a γ-measurement was performed (HPGe detector type PGC 2018, Bias:2500 V positiv, counting time: 86400 s, software: Gamma-W Version: 2.44: using ^{152}Eu as standard solution with the same geometry, distance 15 cm). The uncertanties of the measured activities are in the range between 5% (high values) to 20 % (low values). The radiosotopes ^{144}Pr, ^{144} Cer, ^{154}Eu and ^{155}Eu, ^{134}Cs and ^{137}Cs were identified.

Besides $^{134/137}$Cs the radioisotope ^{90} Sr strongly contributes to the activity of the sample. The activity of ^{90}Sr was determined after the following selective separation steps; 1 mL of each sample solution was diluted with 0.67 mL of a 1 M HNO_3 solution. Then 1 mL of this solution was used to quantify the ^{90}Sr activity. A colum (6 mL in volume) was filled with a suspension of 1 g resin (Sr-Resin, Eichrom-Company) in 5 mL of a 2 M HNO_3 solution. The colum was washed two times with 5 mL of a 2 M HNO_3 solution and then 5 ml of a 8 M HNO_3 solution was added. Afterwards the sample solution was added to the column. The sample vial was rinsed with 1 mL of a 8 M HNO_3 solution and this solution was added to the colum as well. Then a washing step with 10 mL of a 8 M HNO_3solution was performed. The washing solution was collected. After the washing steps ^{90}Sr was eluated by using 10 mL of a 0.05 M HNO_3 solution. Immediatly 1 mL of the eluat was used for the β-measurement.

Then the activity of technetium was determined. First the washing solution of the Sr partition process was evaporated. The obtained residue was dissolved in a 2 M HNO_3 solution (about 2 mL) and then used as sample soltuion for the Tc stripping. A colum (6 mL in volume) was

filled with a suspension of 1 g Tc-Resin (TEVA-Resin, Eichrom-Company) in 5 mL of a 2 M HNO_3 solution. The colum was washed two times with 5 mL of a 2 M HNO_3 solution. Then the sample solution was added. Washing steps were performed with 10 mL of a 2 M HNO_3 solution. After the washing process Tc was eluated with 10 mL of a 8 M HNO_3 solution. The eluate was evaporated and the residue was dissolved in 1 mL of a 2 M HNO_3 solution and the activity of Tc was determined by LSC.
An alpha-spectrometer (Octete, company Ortec, PIPS-detector (planar implanted passivated silicon)) was used to analyze the activities of the radionuclides U, Pu, Am, Np and Cm. 0.1 mL from both samples solutions were vaporized directly on metal disks and the measurement was performed for 250000 sec..The activity for Np was below the detection limit.

5. PSI - SNF sample characterisation, methodologies and tools

I. Günther-Leopold, E. Curti, A. Froideval Zumbieh and H.P. Linder

PSI selected a standard BWR fuel rod and a standard PWR fuel rod for investigations within the project FIRST-Nuclides. Both fuels were manufactured with UO_2 material, having standard ^{235}U enrichments. During reactor operation the BWR and PWR fuel rods achieved average burn-ups of 57.5 and 62.2 GWd·(t HM)$^{-1}$, respectively. Additionally, a PWR MOX fuel rod with 63 GWd·(t HM)$^{-1}$ was selected for subsequent studies in FIRST-Nuclides. The data of the three fuel rods were compiled in Deliverable 1.1 (Metz *et al.,* 2012) and in the Proceedings of the 1st Annual Workshop of FIRST-Nuclides (Günther *et al.,* 2013). The data-set is given in Table 15 to Table 17 of chapter 10 for the sake of completeness and comparison with data of all HBU-SNF studied in FIRST-Nuclides.

5.1. Preparation of samples of spent BWR and PWR fuel rods by PSI

PSI is able to perform leach experiments on maximum nine fuel samples in parallel in the "Dissolution box" of the Hot Laboratory. The test matrix was defined by selecting three samples (cladded fuel, cladding and fuel fragments) from the UO_2 fuel rod irradiated in the Leibstadt BWR (KKL), as well as six samples (cladded fuel, fuel fragments, cladding with and without fuel residues) from the Gösgen PWR (KKG), of which four originate from an UO_2 fuel rod and two from a MOX fuel rod irradiated in the Gösgen PWR. In addition to a previously published study (Johnson *et al.,* 2012) fuel fragments and cladding of this MOX fuel rod will be leached separately in order to complete the available data set for this material.

For each of the selected fuel types one suitable fuel segment was selected and a segment cutting plan was compiled, similar to the one shown in Figure 16 for the UO_2 PWR fuel. According to these cutting plans to the Hot Cell group of the Hot Laboratory prepared segments for SNF leaching experiments, segments to be used in micrometer scale X-ray fluorescence (XRF) and X-ray absorption near edge structure (XANES) investigations as well as a segment for burn-up determination. Based on earlier experiments (Johnson *et al.,* 2012) it was decided that the length of the fuel rod specimens for the leach experiments should be 20 mm, whereas for the "burn-up sample" 10 mm will be sufficient. The cutting was performed in "Hot Cell 2" using a LECO cutting machine with a diamond plate saw. Some samples were broken into two halves by cutting the cladding on opposite sides without cutting the fuel itself. The retrieval of the fuel samples from the dry storage facility, the cutting of the segments and the further sample treatment in view of generating isolated cladding samples and fuel fragments are in process.

5.2. Experimental and analytical methodology for SNF leaching experiments at PSI

Leach equipment

Since the handling of the leaching equipment has to be performed with manipulators in the shielded dissolution box of the PSI Hot Laboratory, the design has to be as simple as possible. For the experimental setup it was decided to use the same design as the one already used in an earlier study (Johnson *et al.,* 2012). The experimental setup consists of glass columns (total volume approx. 250 mL) with a sealed outlet cock for sampling and an integrated glass filter

in order to prevent the clogging of the cock by solid particles. The equipment used in the above mentioned previous study is shown in Figure 17.

Discussions with SCK·CEN have resulted in the agreement that both laboratories will use the same design for the experimental studies in frame of the FIRST Nuclides project. The leach columns were ordered, the pistons were manufactured in-house, the entire setup was tested at PSI and the equipment dedicated to SCK·CEN was delivered in November 2012.

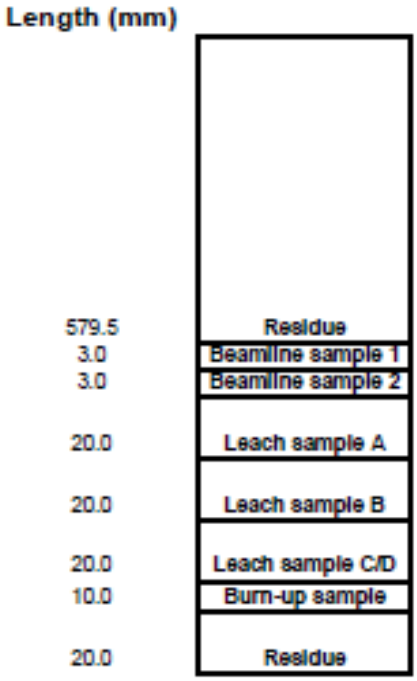


Figure 16: Cutting plan for the selected high burn-up UO_2 PWR fuel segment irradiated in the Gösgen PWR.

Figure 17: Assembly of five glass columns with pistons designed and used for earlier PSI leaching experiments (Johnson *et al.*, 2012).

Gamma spectrometry

The measurement of the ^{137}Cs activity in the leach solutions will be performed using a standard HPGe-detector with a 12% relative efficiency and 1.85 keV FWHM (full width at half maximum) at 1332.5 keV. The detector is connected to commonly used NIM-electronics with a Canberra multi-channel analyzer (MCA) AIM Model 556 and a Canberra VAX/VMS-Genie-Software. The measurement of the ^{129}I activity will be performed using a standard HPGe-planar-detector with an active area of 200 mm^2 and 0.5 keV FWHM at 122 keV. In earlier test experiments a detection limit for ^{129}I of approx. 5 Bq was determined.

In order to decrease the dose rate of the leach solutions for subsequent handling and the background for ^{129}I determination by gamma spectrometry significantly, Cs will be precipitated from the leach solutions with saturated ammonium molybdophosphate solution (Sigma Aldrich). The efficiency of the precipitation step was determined to be > 99% by gamma spectrometry. Furthermore, the recovery rate for iodine after consecutive precipitation steps was determined with a ^{131}I reference material. It could be shown that approx. 3% of ^{131}I are lost per separation step. This result was confirmed with two different samples and each time two precipitation steps. Therefore, the results of the ^{129}I measurements will be corrected for this systematic loss of iodine.

Inductively coupled plasma mass spectrometry

The inductively coupled plasma mass spectrometry (ICP-MS) system to be used in frame of the FIRST-Nuclides project is an "Element 2" from Thermo Fisher Scientific (Bremen, Germany). A schematic diagram of the system is shown in Figure 18. The combination of high sensitivity and very low background makes it very suitable for elemental trace and ultra trace analysis. The design principle is a double focusing sector field analyzer based on a reverse Nier-Johnson geometry. The torch box with the sample introduction system (nebulizer, spray chamber, plasma torch) as well as the autosampler and the interface chamber of the mass spectrometer are encapsulated in a glove-box.

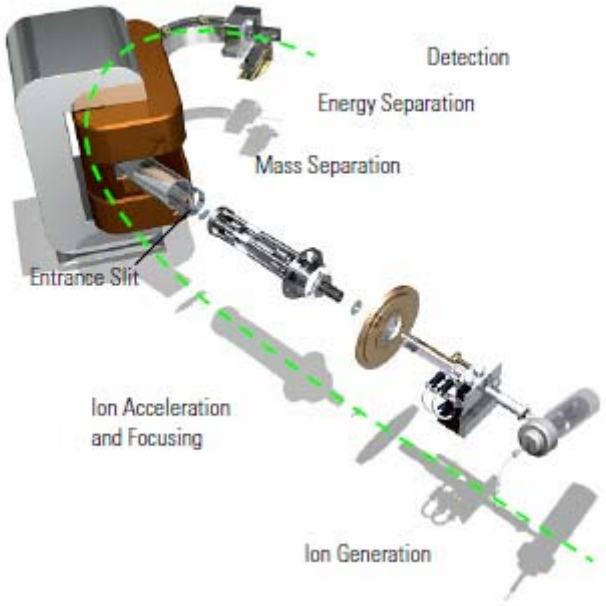


Figure 18: Schematic of the ICP-MS "Element 2" used for elemental trace and ultra trace analysis at PSI.

In order to determine the isotopic composition of Cs, aliquots of the leach solutions will be separated on a CS12a chromatographic column (Dionex, Olten, Switzerland) from the interfering element Ba. The isotopic abundances of all Cs isotopes will be determined using the online connected Multicollector ICP-MS "Neptune" (Thermo Fisher Scientific, Bremen, Germany). The MC-ICP-MS is equipped with nine Faraday cups allowing the simultaneous detection of up to nine masses. This system was especially developed for the highly precise measurement of isotope ratios. The MC-ICP-MS will also be used for the determination of ^{79}Se in the leach solutions.

In order to determine the burn-up of the UO_2 PWR fuel sample the quantitative determination of the elements uranium, plutonium and neodymium is necessary. The sample dissolution and the analytical procedure using the technique of isotope dilution, the correction of mass bias effects and the general measurement procedure will be performed as described in detail in Günther-Leopold et al. (2004, 2008).

Determination of ^{14}C by liquid scintillation counting

The experimental setup (Figure 19) to be used for the extraction and determination of ^{14}C is based on the method described by Stroes-Gascoyne *et al.* (1994). The solutions will be boiled under reflux to convert all C to CO and CO_2. Due to slight vacuum conditions the volatile species will be guided through a tube containing CuO maintained at 600°C to convert CO and H_2 to CO_2 and H_2O. Finally, the gas stream will be transferred through two wash bottles filled with 0.4 M and 0.2 M NaOH, respectively. The ^{14}C concentration in these solutions will be measured after adding of Ultima Gold LLT by the Beta-LS-spectrometer "Tricarb 2200" (Packard).

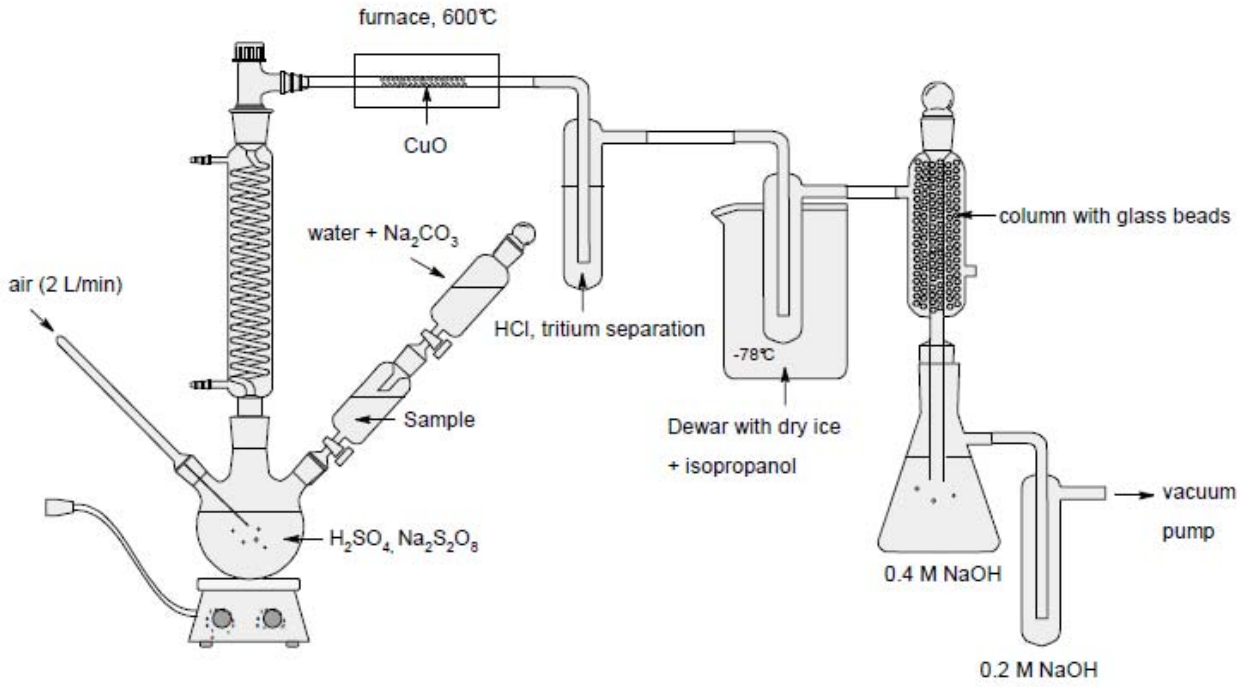


Figure 19: Experimental setup for the ^{14}C determination at PSI.

Beam line techniques

A combined micro X-ray fluorescence (micro-XRF) and micro-X-ray absorption near-edge structure (micro-XANES) feasibility study was conducted at the microXAS beamline at the Swiss Light Source in the first part of the project on high burn-up spent UO_2 fuel from the Leibstadt power plant (78.7 GWd·(t HM)$^{-1}$, 9 cycles, fuel rod KKLAGB108-G6), in order to investigate the spatial distribution of ^{79}Se and its redox speciation within a cross sectional area on the fuel pellet (Froideval *et al.*, 2012). A similar experiment is planned on fuel particles after aqueous leaching under reducing conditions, in order to detect possible changes in the oxidation state of Se. The PSI microXAS beamline is the only beamline where such investigations on radioactive materials can be conducted at a resolution close to one micron, allowing the study and even mapping of micrometer sized fuel particles (see e.g. Curti *et al.,* 2012).

The sample preparation for the microXAS beamline is performed by gently contacting a pellet cross-section (previously subject to mechanical abrasion) with Kapton tape. The obtained imprint is then covered and sealed with a second layer of Kapton, cut to a stripe representing the cross-sectional area of the pellet and mounted on a special sample holder dedicated to active measurements at SLS (Degueldre *et al.,* 2011). This method, which yields a discontinuous spent fuel sample consisting of few scattered micrometer-sized particles, is imposed by the very low activity limits allowed at the MicroXAS beamline.

In addition, test Se-K edge XANES measurements were carried out successfully at the INE-beamline (ANKA, Karlsruhe Institute of Technology) on depleted UO_2 powder samples doped with 10, 100 and 2000 ppm SeO_2. The tests were successful and showed that even EXAFS-quality measurements on spent fuel would be possible. A weak but still usable Se-XANES signal was also detected in the sample doped with 10 ppm Se. Recently, STUDSVIK has been contacted in order to ascertain whether preparation of SNF samples (studied by PSI) with their dedicated Focussed Ion Beam would be possible, with regard to future XAS measurements.

6. SCK·CEN - SNF sample characterisation, methodologies and tools

K. Govers, M. Verwerft, W. Van Renterghem, K. Lemmens, T. Mennecart, C. Cachoir, L. Adriaensen, A. Dobney and M. Gysemans

As one of the partners of FIRST-Nuclides with a hot-cell infrastructure and the required analytical laboratories, SCK•CEN will perform leach tests on spent fuel samples with a relatively high burn-up. For this purpose, a fuel rod was selected from the spent fuel stock available at SCK•CEN for which the characteristics are known and can be made public. In the past, several fuel rods irradiated under high duty conditions in Tihange-1 PWR, have been extracted from their assembly to perform post-irradiation experiments at SCK•CEN laboratories. These fuel rods and relevant results of the previous post-irradiation examination (PIE) campaign – e.g. gross-γ scanning – are available for further research. The most relevant characteristics of the fuel rod that will be used for the leach tests at SCK•CEN have been determined. The data of the fuel rod were compiled in Deliverable 1.1 (Metz *et al.,* 2012) and in the Proceedings of the 1st Annual Workshop of FIRST-Nuclides (Mennecart *et al.,* 2013). The updated data-set is given in Table 18 of chapter 10 for the sake of completeness and comparison with data of all HBU-SNF studied in FIRST-Nuclides.

6.1. Characterisation of a spent fuel rod from PWR Tihange-1 studied at SCK·CEN

The Tihange-1 NPP, located in Belgium, is a PWR reactor loaded with 15×15 fuel assemblies usually operated at an elevated linear power. Two fuel rods located at symmetrical positions (D05 and E12) in the assembly have been extracted from assembly FT1X57 for non-destructive and destructive analyses in the SCK•CEN laboratories (Sannen and Parthoens, 2003). Fuel rod D05 has been selected for use in FIRST-Nuclides.

Details about the power history at rod and sample level has been reconstructed by the operator during the first phase of the FIRST-Nuclides project, based on reactor power history and power distribution mapping of the core, but this information needs to be validated before it can be published. At present, generic data are available: the final rod-average burn-up, estimated as 50.0 GWd·(t HM)$^{-1}$ for rod D05, has been achieved in two cycles of 18 months each. The intermediate burn-up was about 28.1 GWd·(t HM)$^{-1}$ and the linear power is around 330 W/cm. Details about the characteristics of the fuel are given in the table hereunder.

The geometry of assembly FT1X57 consists of a 15×15 array of fuel rods with 21 unfuelled locations (guide tubes) for potential insertion of control rods or instrumentation. The cladding material is M5. Nominal data of the fuel rod design can be found in Table 5 and Table 18. Batch data relative to rod dimensions and fuel composition are then reported in Table 6.

Table 5: Nominal (design) data of the PWR Tihange-1 fuel rod D05, assembly FT1X57.

Assembly type		**AFA 2G**
Assembly geometry		15 × 15
# of fuel rods per assembly		204
Rod pitch	(mm)	14.3
Fuel		
Type		UO_2
Enrichment (nominal)	(% U^{235}/U_{tot})	4.25
Density (nominal)	(% TD)	96
Average grain size	(µm)	10
Cladding		
Type		M5 recrystallized
Composition		
• Nb	(wt.%)	Nominal M5
• Fe	(wt.%)	0.8 – 1.2
• O	(wt.%)	0.015 – 0.06
• Zr	(wt.%)	0.09 – 0.12
		balance
External diameter	(mm)	10.720
Thickness	(mm)	0.618
Internal diameter	(mm)	9.484

6.2. Preparation and characterisation of samples of the spent fuel rod from PWR Tihange-1 by SCK·CEN

Cutting scheme definition

Fuel rods D05 and E12 have been cut for non-destructive tests in four segments of about 1 m length each. The samples for the leach tests will be taken from the second segment (from the bottom) of rod D05, internally referred to as FT1X57-D05/R4. The burn-up profile between the end of the first span up to the end of the fifth span is indeed relatively constant, as shown by the γ-activity measurement (see Figure 20), except close to grid locations. Activity peaks are observed at regular intervals in the central zone, indicating volatile product migration to colder zones at inter-pellet locations. A large contribution to the signal indeed originates from Cs isotopes. There is, however, no indication of major redistribution of volatile fission products along the fuel rod, which would be characterized by a slightly depleted signal at the centre and higher activity in the bottom and top parts.

Table 6: Batch data of the PWR Tihange-1 fuel rod D05.

Fuel rod		
Rod total length	(mm)	3861.9
Active fuel stack length	(mm)	3634.9
Plenum length	(mm)	205.1
Diametrical pellet-clad gap	(µm)	190
He filling pressure	(bar)	20
Fuel characteristics		
Material		UO_2
Density	(% TD)	96.44
Mass metal/oxide	(g_U / g_{UO2})	88.13
U isotopic composition		
^{234}U / U_{tot}	(wt%)	0.038
^{235}U / U_{tot}	(wt%)	4.251
^{236}U / U_{tot}	(wt%)	0.001
^{238}U / U_{tot}	(wt%)	95.71
Impurities / additives		Not available yet
Pellet dimensions		
Diameter	(mm)	9.294
Length	(mm)	11.15
Dish		
• Depth	(mm)	0.34
• Spherical radius	(mm)	16.70
Chamfer		
• Height	(mm)	0.20
• Width	(mm)	0.57

The samples to be used for the leach tests will consist of two pellets. The samples are cut from mid-pellet to mid-pellet in order to keep a representative inventory of the volatile elements that relocate at pellet-pellet interfaces. Two other samples are taken for fuel characterization. The first one is used for radiochemical analysis (RCA) of the fission product and minor actinides inventory, enabling for pellet average burn-up determination. The second one is used for optical microscopy (OM) and determination of the local element composition by electron-probe micro-analysis (EPMA). An additional spare sample is foreseen as well. The proposed cutting scheme is illustrated in Figure 21. Signal peaks, regularly spaced by about 11.8 mm, were exploited to identify pellet-pellet interfaces. The samples are located in the flat γ-activity zone, far enough from the grid locations. Although the axial burn-up is expected to be homogeneous over the sampling zone, leach test samples are proposed to be flanked by the sample used for burn-up determination by RCA and the sample used for OM / EPMA. The spare sample is located the closest to the third grid location, next to the OM / EPMA sample.

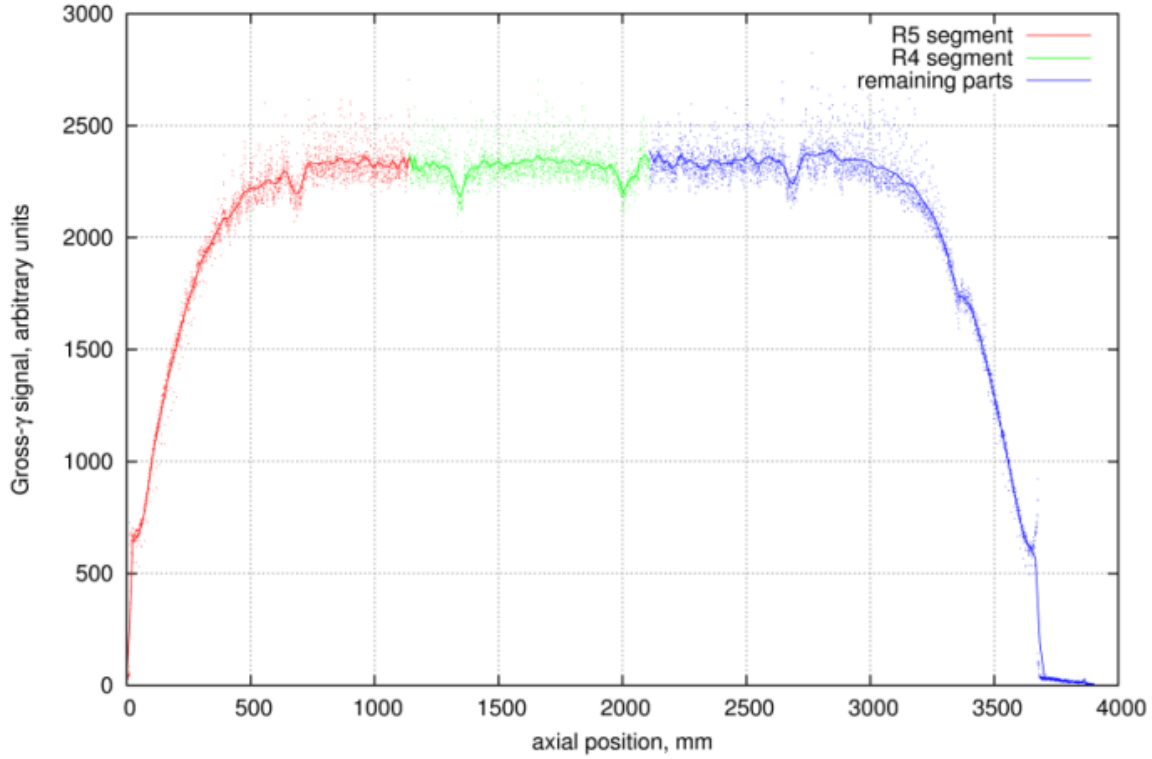


Figure 20: Gross-γ scanning of PWR Tihange-1 fuel rod D05. Measurements were acquired every 0.5 mm using a collimator. The line (guide to the eye) clearly identifies the activity drop due to support grids. The flat activity profile over most of fuel rod length is characteristic of high burn-up fuel.

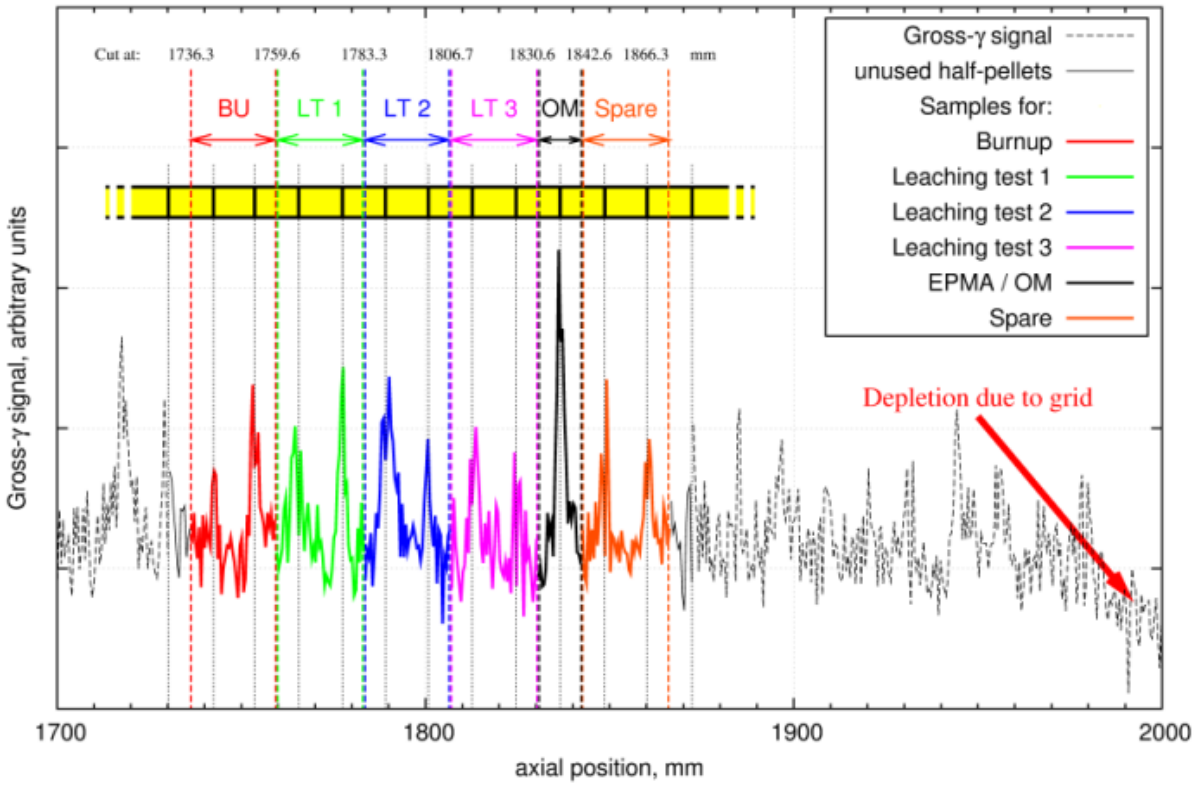


Figure 21: Proposed cutting scheme of segment FT1X57-D05/R4. Three samples are intended for leaching experiments. One sample will be used for burn-up determination and another one for optical microscopy and EPMA. An additional spare sample is presently considered.

Cutting of the spent fuel rod

The following section describes details of related to the preparation of the spent fuel samples (Figure 22). Based on the cross gamma scanning done few years ago, the exact place of the pellets inside the rod is known. The various experiments foreseen in this program need to have rod segment with two equivalent pellets. The segment will be cut from mid pellet to mid pellet in order to keep a representative inventory of the spent nuclear fuel, specially the caesium located between two pellets. In total five segments are prepared. Two of them are used for leaching tests, a third one is completely dissolved in order to determine the burn-up and the complete inventory of the spent fuel, the fourth is used for optical measurement and the fifth is a spare segment. For optical analysis with the EPMA, the segment is constituted with only two half pellets. The determination of the burn-up is done following a standard procedure of analysis.

Figure 22: Preparation of SNF samples in dry condition in the SCK·CEN hot cell facility.

A rotary disk using a cooling solution (diamond saw) is used in the case of the sample set aside for the determination of the burn-up and the optical measurements. The aim of the project FIRST-Nuclides is to study the release of the most soluble radionuclides when the spent nuclear fuel is in contact with a solution for the first time. Consequently, the classic method to cut the rod segment with the rotary disk using a cooling solution is not conceivable and the preparation of the rod segment for the leaching test have to be prepared under dry conditions. Hence, a commercial tube cutter is used (tool of plumbing). Only the cladding is cut with this tool.
Three segments are prepared:

- One for a leaching experiment with an "intact" rod segment
- The second one with a "decladded" sample. A sample is about 2.4 cm. To obtain the decladded samples, the sample is cut in several parts. Every 0.8 cm, the cladding is cut and the fuel is broken. Afterwards, the fuel is separated from the cladding by

pressure. The several pieces of the cladding and all the parts/pieces/fragments of fuel are leached together in the leaching device.

- The last one is a spare segment. It play the role of "buffer" during the preparation between the sample cut by the classic method (with cooling solution) and the sample prepared in dry conditions.

6.3. Radionuclide composition of spent PWR fuel samples studied by SCK·CEN

The fuel composition has been estimated using robust tools. The following results have been derived using the ORIGEN-ARP module available in SCALE 6.1 (ORNL, 2011). It contains pre-compiled cross-section libraries for several LWR fuel assembly designs, among which the PWR 15 × 15 lattice. Variations in fuel composition, burn-up and moderator density are handled through interpolation of the library data.
A simplified, constant, power history has been postulated over two cycles of 500 days each, reflecting the 18 months cycles of the selected high duty fuel rods. A shutdown period of 50 days has been assumed between both cycles and a storage time of 10 years after the irradiation. The power level has been varied (see Figure 23) to look at the sensitivity of the fuel composition to the burn-up after 10 years storage, which is representative of the specimen considered in the present study. Two burn-up levels are highlighted in Figure 24 and Figure 25 corresponding respectively to the rod average burn-up and sample burn-up. One should keep in mind that values calculated with neutronic codes reflect isotopic production and decay but do not model fission product redistribution (migration).

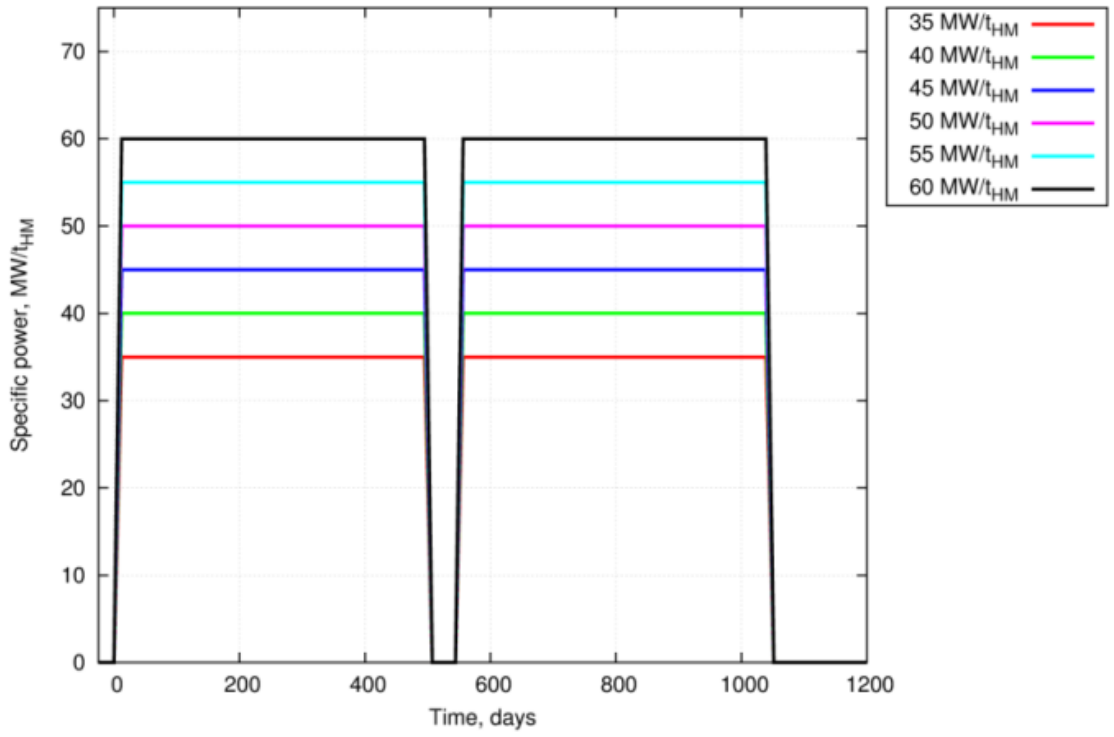


Figure 23: Simplified power history for various burn-up cases. Two cycles of 500 days at constant power are assumed.

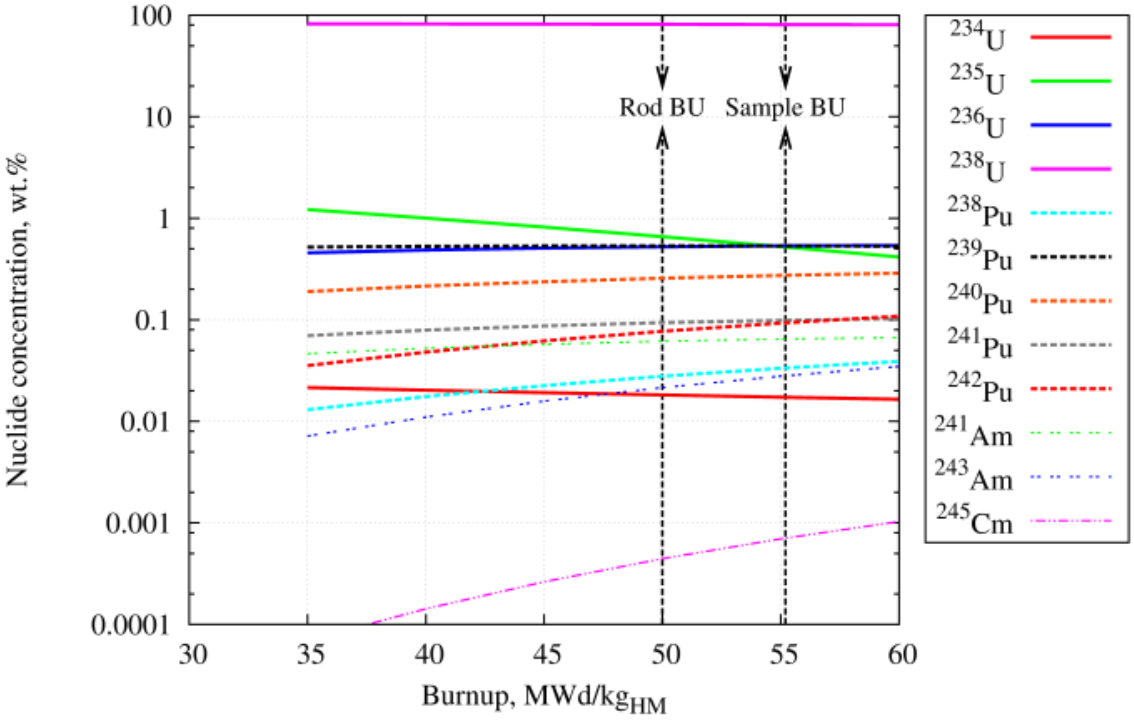


Figure 24: Actinide inventory evolution with disposal burn-up, after 10 years of decay. Vertical arrows indicate the approximated rod and sample burn-ups. Vertical arrows indicate the approximate rod and sample burn-ups.

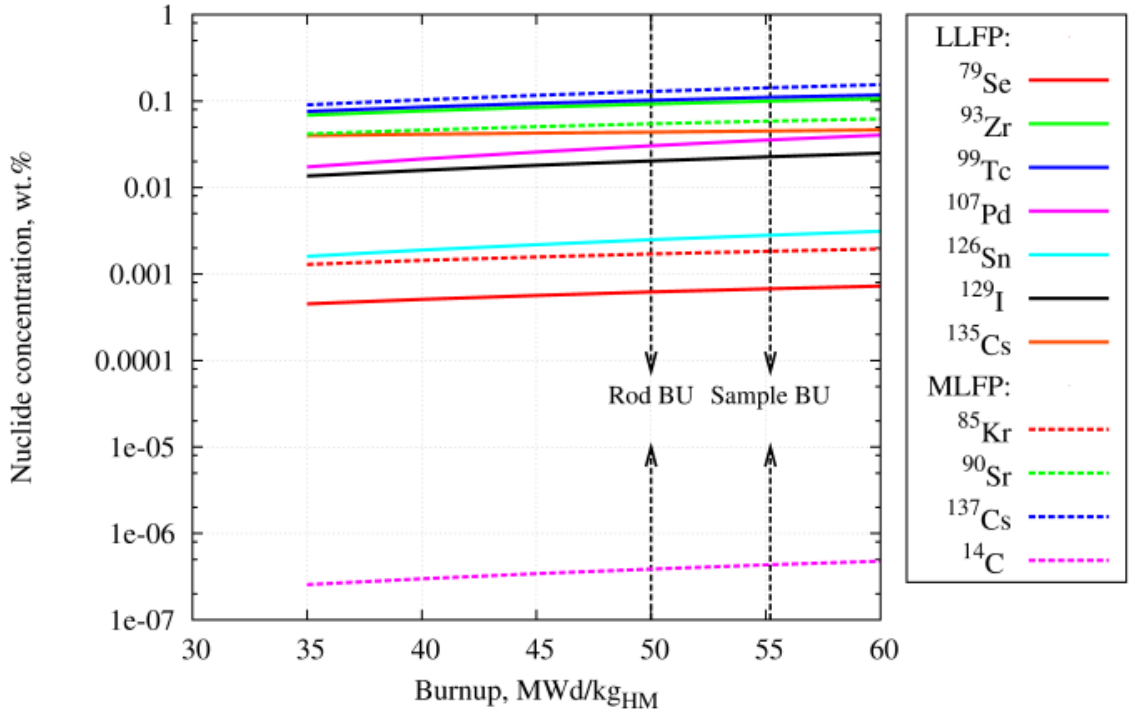


Figure 25: Relevant (in terms of the leaching test experiments) long- and medium-lived radioisotope inventory evolution with disposal burn-up, after 10 years of decay. Vertical arrows indicate the approximate rod and sample burn-ups.

6.4. Experimental and analytical methodology for SNF leaching experiments at SCK·CEN

The optical micrograph composite image of the fuel microstructure is shown in Figure 26. The crack pattern of the pellet OM1 of fuel rod D05 is typical for an intermediate to high burn-up fuel that was irradiated at moderate power. The microstructure shows radial cracks and one piece of fuel missing. This missing fuel part is a result of sample preparation. The sample was impregnated with resin to avoid further break-out and repolished. The part of the pellet with the missing fuel was of course avoided in the EPMA analysis.

Figure 26: Optical micrograph of the microstructure of sample OM1 of fuel rod D05

The SNF leaching experiments consist in two phases: (1) pre-leaching step: the spent nuclear fuel sample is put in contact with 50 ml of leaching solution during 3 - 4 days. Afterwards, the solution is completely removed. This step is repeated once again. The aim of this step is to remove the oxidised layer of the spent fuel. (2) the leaching test itself aims to study the dissolution the spent fuel matrix and the release of the radionuclides by doing 4 samplings of the solution during one year.

Three glass columns are placed in the holder foreseen for this experiment. The equipment is identified by:

1. Tests with the intact rod fragment in contact with the leaching solution
2. Test with the decladded fragment in contact with the leaching solution
3. Blanco (no spent fuel, only the leaching solution)

The experimental setup is based on previous works performed at PSI (see section 5.2 and Figure 17). The materials have a good resistance under high radiation field. Hereunder, pictures of the glass column used for the experimental work are shown in Figure 27.
Once the SNF sample is placed inside the column 1 and 2, the three columns are filled with 50 mL of solution for the preleaching phase. After 3 days, the solutions are replaced by a fresh one for the second preleaching, during 4 days. Afterwards, the solution are removed and replaced by 150 mL of fresh solution. From this moment, the "real" leaching start and a sampling of the solution (20 mL) is done after one week, one month, six months and the final samples after one year.

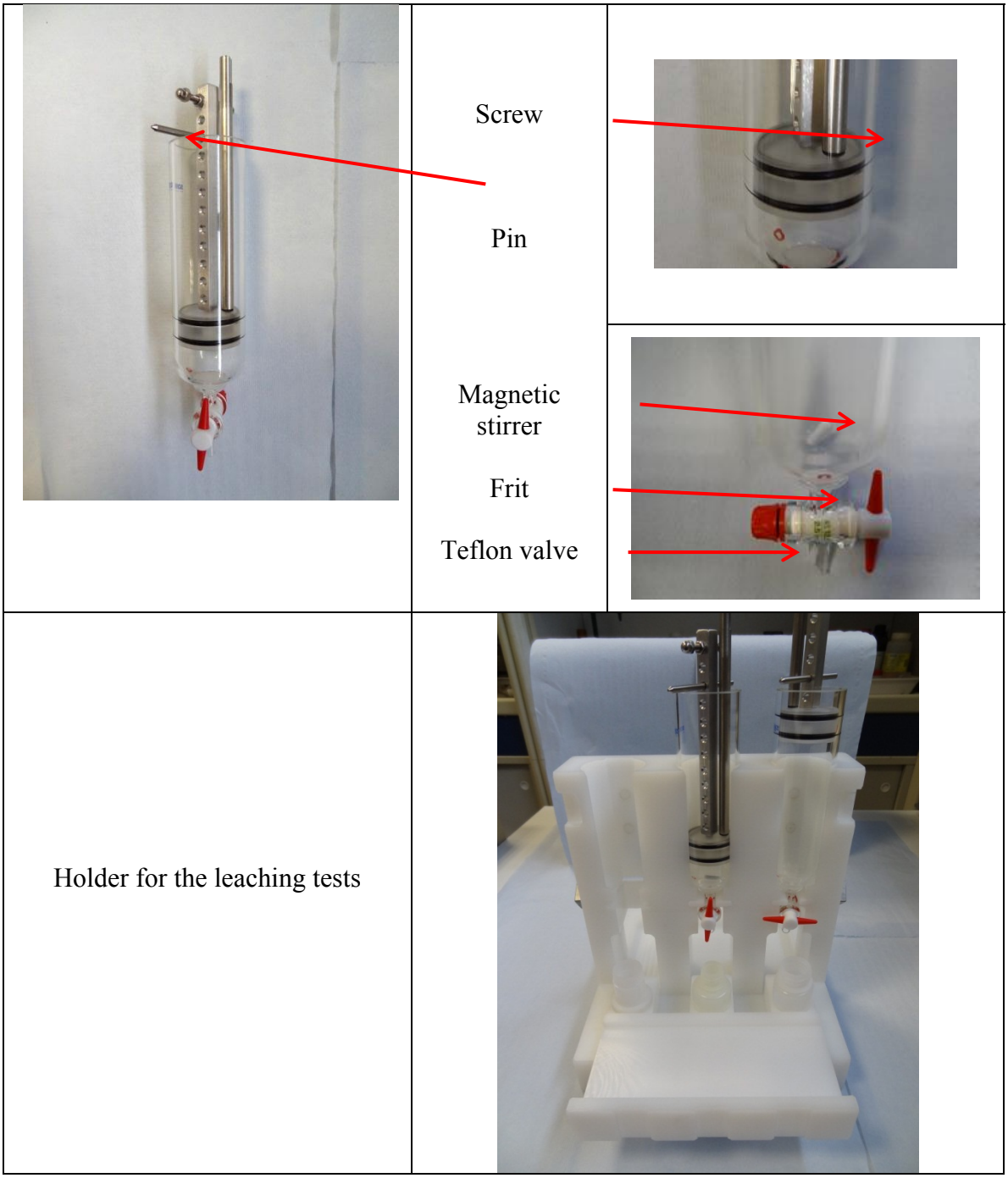


Figure 27: Experimental set-up for SNF leaching tests at SCK·CEN. The inserted images show an exemplary glass columns with specific instrumental components.

In case of the test 2 (decladded fuel), a filtration of the sampling solutions is needed before the analysis. In this test, the whole fuel is used (pieces, cores and powder of fuel). A frit was included at the bottom part of the column to prevent that the fuel pass through the tap but the porosity is too small to contain the finest particles of fuel. Consequently, a certain amount of spent fuel pass through the frit and recuperate in the bottle with the solution during the sampling. The solutions from the test 2 are previously filtered through filter paper with a porosity 4 – 12 µm. Afterwards, the filter is dried and using the weight of the filter before and

after the filtration, we estimated the mass of materials lost during the renewal / sampling of solution.
All the solutions from the tests 1 and 2 and the sampling after one week of the test 3 are analysed to determine the release of ^{93}Zr, ^{129}I, ^{135}Cs, ^{137}Cs, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{59}Ni, ^{94}Nb, ^{14}C, ^{63}Ni and ^{90}Sr.

7. CNRS – HTR fuel sample characterisation, methodologies and tools

J. Vandenborre, A. Traboulsi, G. Blain, J. Barbet, and M. Fattahi

CNRS investigates unirradiated TRISO particles, which will be used in successive corrosion experiments under $^{4}He^{2+}$-irradiation. Selected TRISO samples and sub-samples from the initial TRISO particles are prepared and characterized. An irradiation cell for microscopic studies of corrosion at UO_2 grain boundaries of (initially non irradiated) TRISO under a $^{4}He^{2+}$ cyclotron radiation is installed.

7.1. Characterisation of HTR fuel elements

UO_2 TRISO particles are purchased from JÜLICH and the synthesis (including a calcination step which at 1600°C for UO_2 crystallization) is described in detail in Brähler *et al.* (2012). The design of a UO_2 TRISO coated fuel particle with different C-layers is shown in

Figure 28 and Figure 29 Physico-mechanical characterization and first solubility tests have been performed (Bros *et al*., 2006; Grambow *et al*, 2008; Titov *et al*, 2004).

Solid analysis is performed by scanning electron microscopy (JEOL 5800 SV with a 15 kV voltage) and the SEM samples were covered by a Pt layer in order to improve electron conduction and increase the picture resolution. Mechanical separation of C-layers from the UO_2 spheres is performed in order to analyse the sphere (Figure 29). Table 7 shows the properties of the UO_2 spheres after the separation step. Moreover, a SEM-EDX analysis demonstrated that the chemical composition of the sphere surface is purely UO_2.

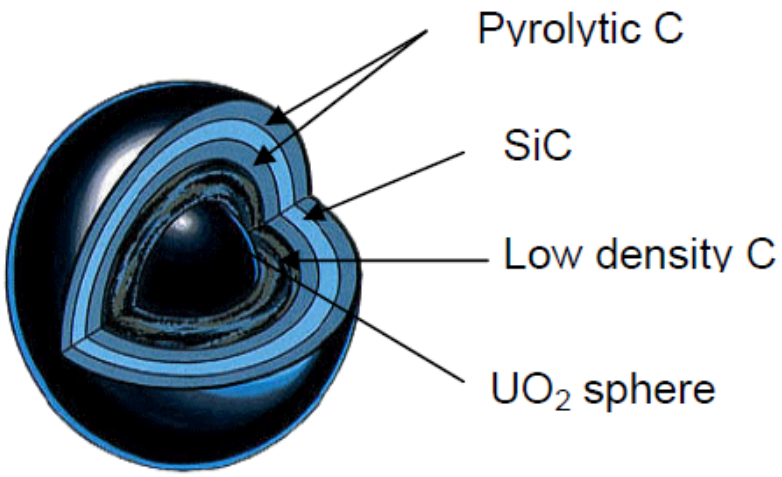


Figure 28: ***Design of a UO_2 TRISO coated fuel particle with different C-layers (from Brähler et al., 2012).***

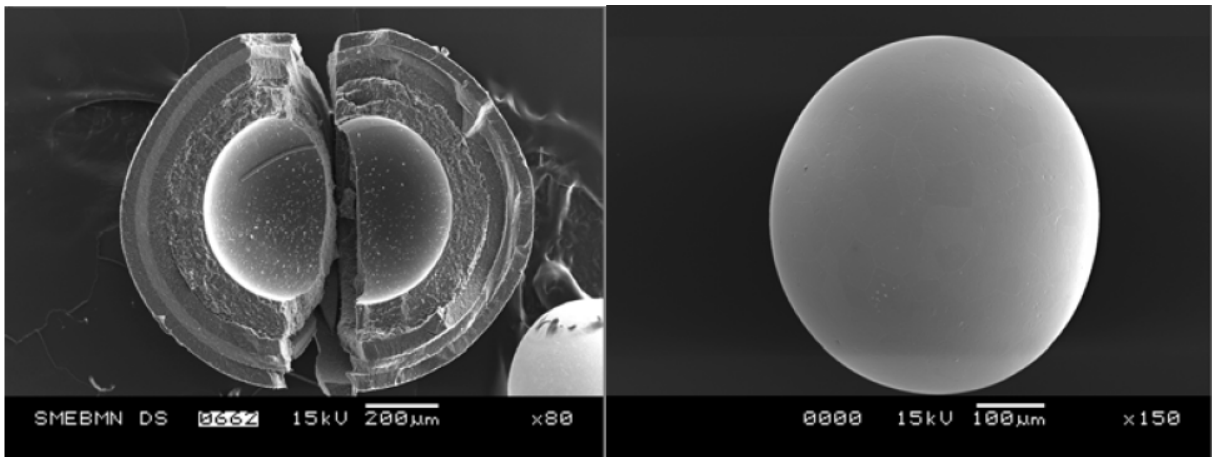


Figure 29: SEM picture of UO_2 TRISO particle after the separation step, Left: C-Layers, Right: UO_2 sphere.

Table 7: Properties determined for the UO_2 sphere after separation of the C-layers of the TRISO particle.

Sphere	UO_2(cr)
Diameter (mm)	0.50
Weight (mg)	0.76
Density ($g.cm^{-3}$)	10.96
Geometric Surface Area ($m^2.g^{-1}$)	$1.05\ 10^{-3}$

7.2. Sample preparation and characterisation of UO_2 TRISO particles studied by CNRS

Pre-washing batch experiments were performed exposing unirradiated UO_2 TRISO particles to an aqueous solution in undersaturated conditions. A HDPE (high density poly-ethylene) reaction vessel is used, containing 15 ml of a 0.1 $mol{\cdot}L^{-1}$ HCl solution under continuous stirring during 15 days in order to deplete the grain boundary (GB) phases. By this way, the impact of the GB onto the radiolytic dissolution process of the UO_2 TRISO particle will be determined. First SEM analyses were performed onto two samples. The UO_2 TRISO particle surfaces were analyzed before and after the pre-washing step, shown in Figure 30 and Figure 31, respectively). From these SEM images, the grain size average is determined to be about 15 ± 5 µm. Moreover, the pre-washing process involves a dissolution of the C-layers remained at the UO_2 surface. Moreover, new grains, with GB too, occur at the UO_2 surface with a lower grain size average.

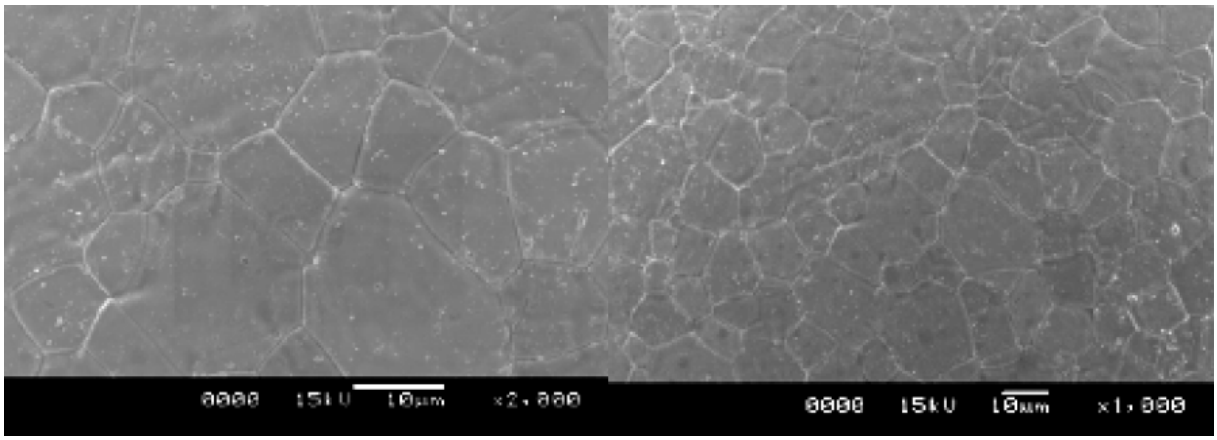


Figure 30*: SEM pictures of the UO_2 TRISO particle before the pre-washing step.*

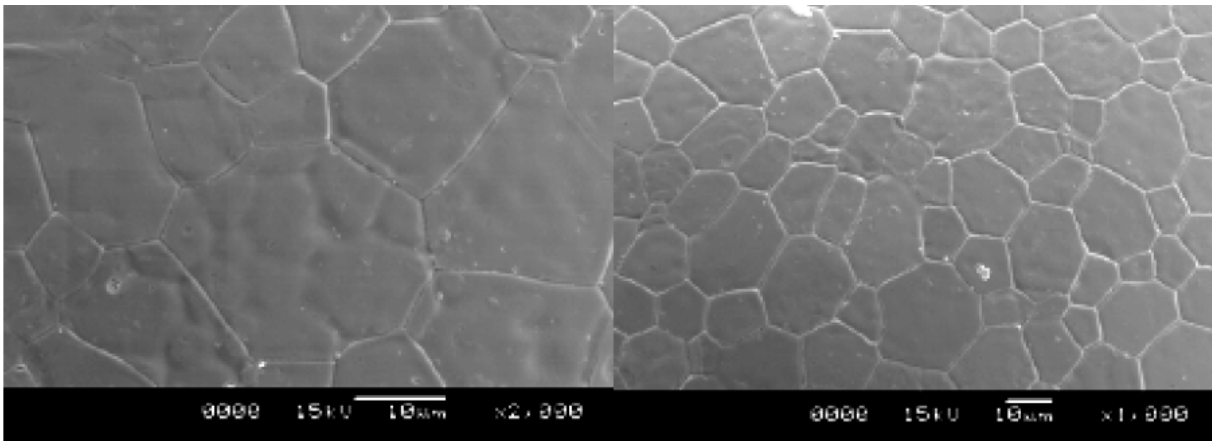


Figure 31*: SEM pictures of the UO_2 TRISO particle after the pre-washing step.*

7.3. Experimental and analytical methodology for irradiation experiments at CNRS

$^4He^{2+}$ irradiation experimental conditions

$^4He^{2+}$ ions irradiations are provided by the ARRONAX cyclotron facility (Saint-Herblain, France) onto a vertical beam-line. Experiments are carried out within the ARRONAX cyclotron at 64.7 MeV. The intensity of the particles beam, measured on an internal Faraday cup located one meter upstream, is maintained at 70 nA. The uncertainty of that current measurement is about 10 %. Fricke dosimetry is used in this study in order to determine the dose deposited into the samples. This method is based on the oxidation of Fe^{2+} to Fe^{3+} by the species produced by the water radiolysis reactions. The concentration of ferric ions is monitored in situ by UV-Vis measurements at 304 nm (ε = 2197 $L.mol^{-1}.cm^{-1}$, 298 K) with a spectrophotometer CARY4000 (VARIAN). These measurements are carried out during the irradiation by a specific probe shown in Figure 32. Super Fricke solutions are prepared by dissolving the desired quantity of Mohr's salt ($[Fe^{2+}]$ = 10 $mmol.L^{-1}$) and NaCl (1 $mmol.L^{-1}$) in aerated aqueous 0.4 $mol.L^{-1}$ H_2SO_4 solutions. All reagents are analytical grade or equivalent. NaCl is added in order to avoid any organic impurities. The irradiation time is a few minutes for ARRONAX experiments. The dose rates were measured at 4300 $Gy.min^{-1}$ during irradiation in the ARRONAX facility.

Figure 32: ***Measurement cell with UV-VIS probe for*** **$^{4}He^{2+}$ ions irradiation experiments.**

In situ Raman spectroscopy experimental conditions

μ-Raman spectroscopy is used to the analyze the surface with UO_2(cr) grain and GB(I), from the solid sintering, and GB(II), from the pre-washing process. The Raman system is purchased from the HORIBA Jobin-Yvon Company. Raman spectra are recorded with an iHR550 spectrometer equipped with two optic fibers (diameter = 100 μm, length = 20 m). The detector is a charged coupled device cooled by Peltier effect (203 K). Raman spectra are excited with a laser beam at 632.8 nm emitted by a He/Ne Laser. The laser beam is weakly focused on samples with a diameter of about 1 mm and a power of about 14 mW for a working distance of 40 mm on the sample and an acquisition time of 2 minutes. The Raman backscattering is collected through an objective system and dispersed by 1200 groves/mm gratings to obtain 5 cm^{-1} spectral resolution for Raman stokes spectra excited at 632.8 nm. The wavenumber accuracy was checked and considered better than 0.5 cm^{-1}. With the Raman spectroscopic device (laser excitation and back scattering Raman), *in situ* experiments have been performed onto the solid samples in contact with ultrapure water. Figure 33 displays the device installed onto the $^{4}He^{2+}$ beam line described above.

Irradiation cells development

First test experiments of *in situ* Raman analysis have been performed with a first version of our irradiation cell which permits analysis of the surface with Raman spectroscopy (see Figure 34). However, a modified spectroscopic cell is developed in order to analyze during the alpha irradiation, both the solid by the Raman spectroscopy and the solution by the UV-VIS spectrophotometry. Moreover, with the modified spectroscopic cell (Figure 35), radiolytic produced hydrogen will be analyzed by μ-Gas Chromatography. This complete analytic system will be useful in order to determine the uranium speciation at the surface, in the solution and to measure the hydrogen produced or consumed by the chemical system (see Figure 36).

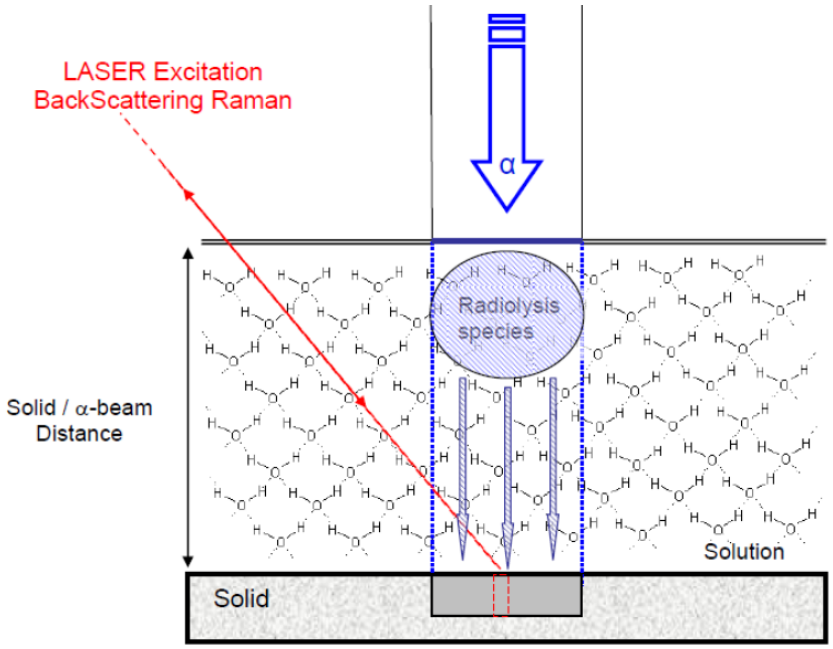


Figure 33: In situ Raman spectroscopic device experiment under $^{4}He^{2+}$ ions beam irradiation at the ARRONAX facility vertical beam line.

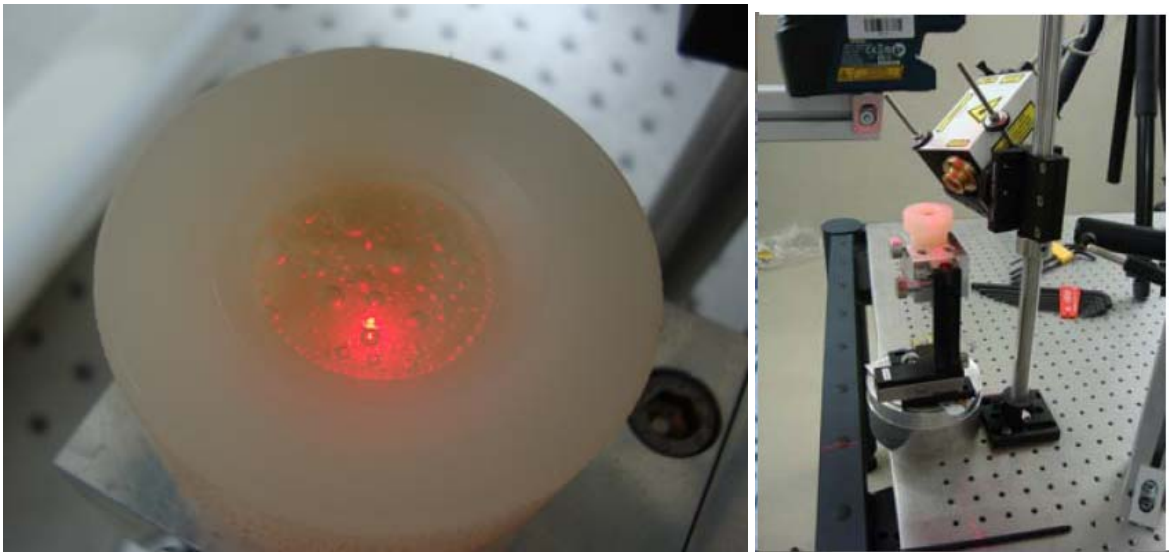


Figure 34: ***In situ Raman spectroscopic analysis under $^{4}He^{2+}$ radiation. Irradiation cell with H_2 bubbles produced by the water radiolysis (left image),in situ Raman spectroscopic device under $^{4}He^{2+}$ ions beam irradiation at the ARRONAX facility vertical beam line.***

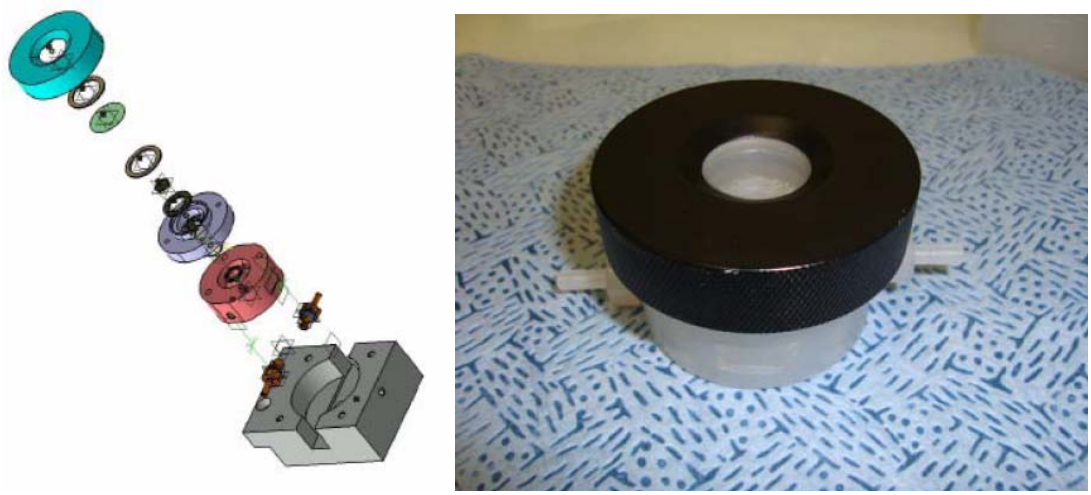


Figure 35: Schematic and photography of the modified spectroscopic cell with water circulation.

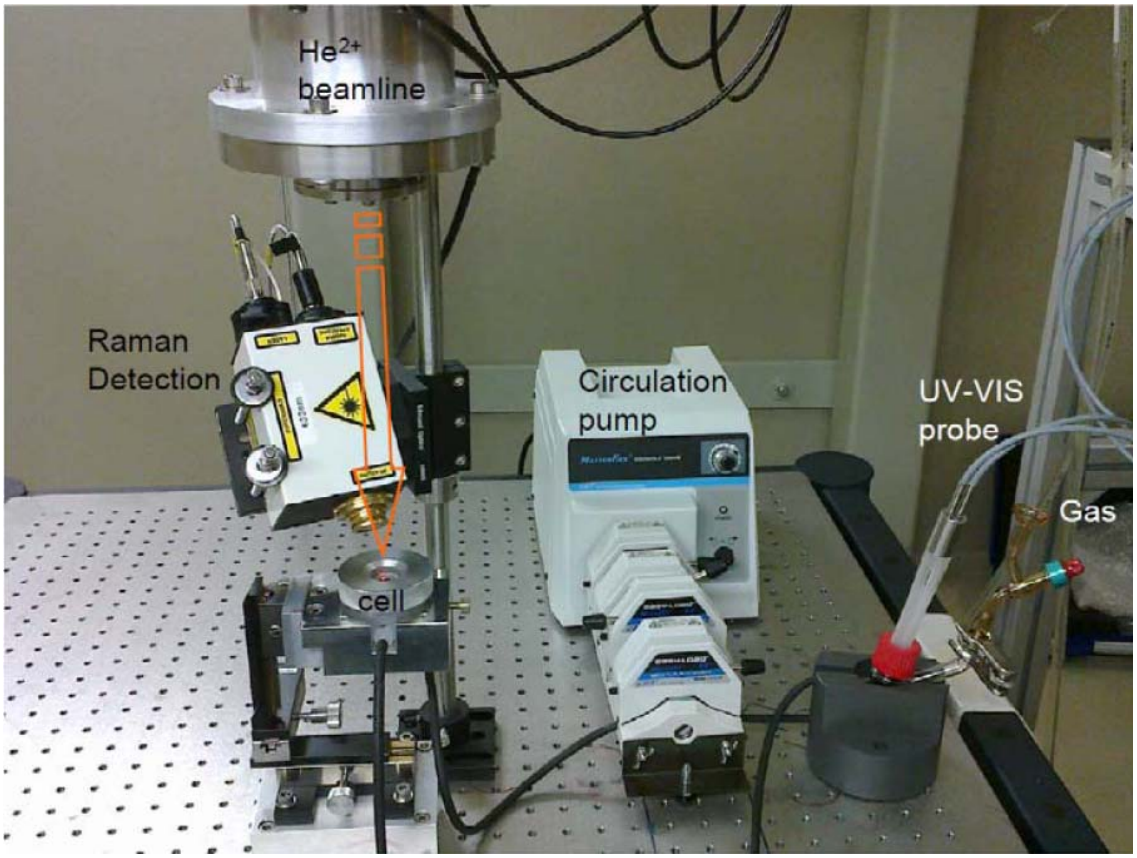


Figure 36: ***Complete experimental in situ Raman system under He^{2+} irradiation with solution circulation, in situ UV-VIS and Gas measurement.***

8. MTA EK - Determination of dissolution rates for damaged and leaking VVER fuel stored in water

E. Slonszki and Z. Hózer

MTA EK determines dissolution rates of different isotopes from damaged and leaking VVER fuel stored in water for several years. The dissolution rates were determined for two datasets:

- Fuel damage took place in a cleaning tank incident at the Paks Nuclear Power Plant in 2003. The thirty damaged assemblies stored in a special service area of the spent fuel storage pool for almost four years. Based on the measured activity concentrations, release rates for different isotopes were calculated.
- A leaking fuel assembly was identified in 2009 at the Paks NPP and a special measurement programme was carried out in the spent fuel storage pool for the investigation of activity release from the leaking fuel rod in wet storage conditions. The measured data from this programme were used for the determination of dissolution rates.

8.1. Calculation of release rates

Several isotopes were measured during and after the incident of Unit 2 of Paks Nuclear Power Plant and during the wet storage of No. 70873 leaking fuel assembly of Unit 4 of Paks NPP. Release rates of those isotopes were measured regularly and their corrected integrated release were estimated in this work. It involves 11 isotopes and uranium in the first case and 13 isotopes and uranium in the second case. MTA-EK used two methods for determination of the release rates:

- First of all MTA-EK calculated the corrected integrated releases of every isotope from the measured activity data. After that these data were divided by the total time which belongs to measurement. The calculated release rate values are given by this method.
- In the other case linear was fitted to the corrected integrated releases data which was resulted the fitted release rates. These processes are described in the following.

8.2. Release rates of isotopes from damaged VVER fuel stored in water

Calculation methods

Coolant activity data between 11th April 2003 and 8th January 2007 have been taken into account. One example of datasets (^{137}Cs) is shown in Figure 37. The data were provided by the Paks NPP. Measurements were available from the pit with the cleaning tank, from the spent fuel storage pool and from the reactor system. Several measurements were taken before and after the filters of the purification system.

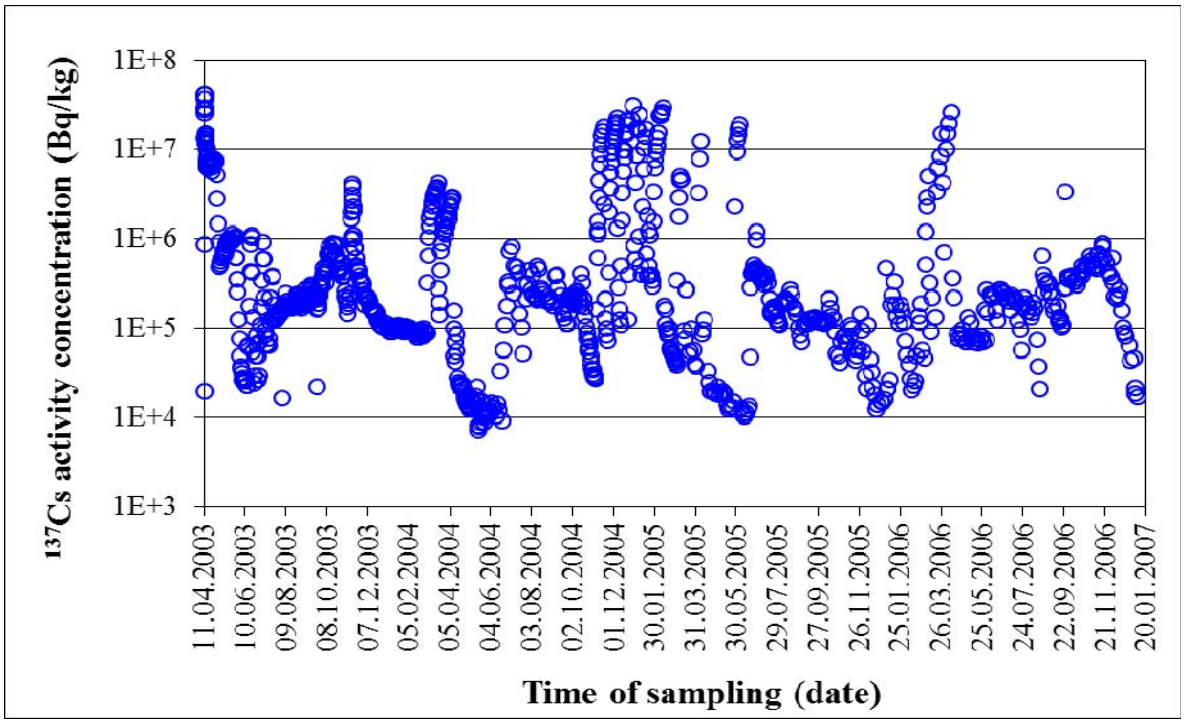


Figure 37: History of ^{137}Cs activity concentration in the pit with the cleaning tank of Unit 2 (Paks NPP).

It was supposed in the evaluation process, that the measured activity concentration was typical for the total volume of water connected to the cleaning tank with damaged fuel. The elapsed time since the incident was divided into short periods, which were characterized by stable technological conditions allowing us to consider constant release rate during a given period. The number of periods was different for the different isotopes. For example in the case of ^{137}Cs isotope 53 periods were identified.

The operation of water purification system was considered using the flowrate and efficiency values. The positions of gates between the reactor pool and between the spent fuel storage pool were taken into account.

The release rate was calculated for each period and the total release was determined as the result of integration over the total calculated time. The release rate was calculated in three different ways.

- Linear fitting was applied to those periods, where the activity continuously increased. That was typical for periods without the operation of water purification system.
- Monotone decrease or stabilization of activities was typical for periods with the operation of water purification system. In this case the following expression was applied for each isotope:

$$\alpha = g_f \frac{C - C_0 * e^{-g_f * \Delta t / M}}{1 - e^{-g_f * \Delta t / M}} \qquad (1)$$

where:

α – dissolution rate of the given isotope (Bq/h);

g_f – flowrate of water purification system (kg/h);

C – activity concentration at the beginning of the period (Bq/kg);

C_o – activity concentration at the end of the period (Bq/kg);

Δt – period (h);

M – mass of water (kg).

- In the third case the average activity value was multiplied by the flowrate of the water purification system. This approach was applied when the scatter of the measured points did not allow us to use the above methods. These periods could not be characterized by a constant dissolution rate that is why an average value was considered.

The real flowrate of the water purification system was corrected by the efficiency of filters. For this reason the efficiency was calculated from activity concentrations before and after the filters.

$$\frac{C_{before} - C_{after}}{C_{before}} \tag{2}$$

where:

C_{before} – activity concentration before the filter (Bq/kg);

C_{after} – activity concentration after the filter (Bq/kg).

The efficiency was multiplied by the flowrate and this virtual flowrate was used in the calculation of dissolution rates. The purification efficiency was calculated for each isotope and for each period.

The inventory of radioactive fission products continuously decreased after shutdown and during the storage of spent fuel in the cleaning tank. In order to get such values that could be compared with the initial inventory and the results of analytical work, corrections were applied according to the decay constants. These corrected values were higher than the measured values. The physical meaning of the corrected values for total release corresponded to the time of the incident, as the total release would have been released that time. The correction was made using this exponential formula:

$$e^{\lambda t} \tag{3}$$

where:

λ – decay constant (1/s),

t – elapsed time from the incident to the end of the given period (s).

Activity release of each period was corrected using the above formula and the total release for the considered 4 years was calculated as the sum of the individual periods. First of all this value was divided the total time - which takes from the incident to the removing of damaged fuel assemblies – to result in the release rate of the given isotope (Table 8). On the other hand linear was fitted to the corrected integrated release data and the rise of curve resulted in the release rate in case of every examined isotope. In case of uranium the activity concentration was made in µg/l. The correction was applied for the mass of metal uranium. It is mean, that the integrated activity of uranium (g) was multiplied the ratio of molar mass of UO_2 and ^{238}U (270/238), see Table 9. Finally the release rates were determined as a rate of isotope inventory of incident (Table 8 and Table 9).

Results of the calculations

Based on the calculated integrated releases the total measuring period can be divided into two parts. The activity of the first two week describes the incident while the other parts of period correspond to the wet storage. After the incident the change in the release rate was influenced first of all by the decay of short lived isotopes. However the water chemistry conditions played important role, too, which is illustrated in this figure that shows the trend of ^{238}Pu and ^{242}Cm activity and the boric acid concentration. The increase of the boric acid concentration from 15 g/kg to 21 g/kg lead to increase of ^{242}Cm and ^{238}Pu activity by two and three orders of magnitude, while the pH decreased from 7 to 4–4.5 (Table 9 and Figure 38). Similar effect was observed for other isotopes as well at the same time.

Table 8: Release rates from damaged VVER fuels - calculated and fitted release rates of the gamma radiant isotopes.

Isotope	Isotope inventory (Bq)	Calculated release rate (rate/d)	Fitted release rate (rate/d)	
			incident	wet storage
^{144}Ce	1,20E+17	2,20E-08	3,47E-08	**2,08E-08**
^{140}Ba	1,16E+17	4,71E-08	7,20E-08	**2,52E-08**
^{131}I	4,15E+16	2,39E-07	1,00E-06	**4,02E-09**
^{137}Cs	7,22E+15	3,65E-08	2,31E-07	**2,89E-08**
^{134}Cs	5,69E+15	4,79E-08	2,93E-07	**3,66E-08**
^{125}Sb	5,75E+14	1,17E-08	-	**1,36E-08**
^{154}Eu	2,32E+14	5,10E-08	-	**4,00E-08**
^{155}Eu	1,22E+14	6,56E-08	-	**4,69E-08**

Table 9: Release rates from damaged VVER fuels - calculated and fitted release rates of the alpha radiant isotopes and UO_2.

Isotope	Isotope inventory (Bq)	Calculated release rate (rate/d)	Fitted release rate (rate/d)	
			pH≈7	pH≈4-4,5
^{242}Cm	9,25E+14	7,67E-08	3,12E-10	**8,96E-08**
^{238}Pu	7,36E+13	6,29E-08	1,55E-10	**7,12E-08**
^{244}Cm	2,31E+13	3,56E-08	4,68E-11	**3,36E-08**
UO_2	-	1,06E-01 (g/d)	-	**1,17E-01 (g/d)**

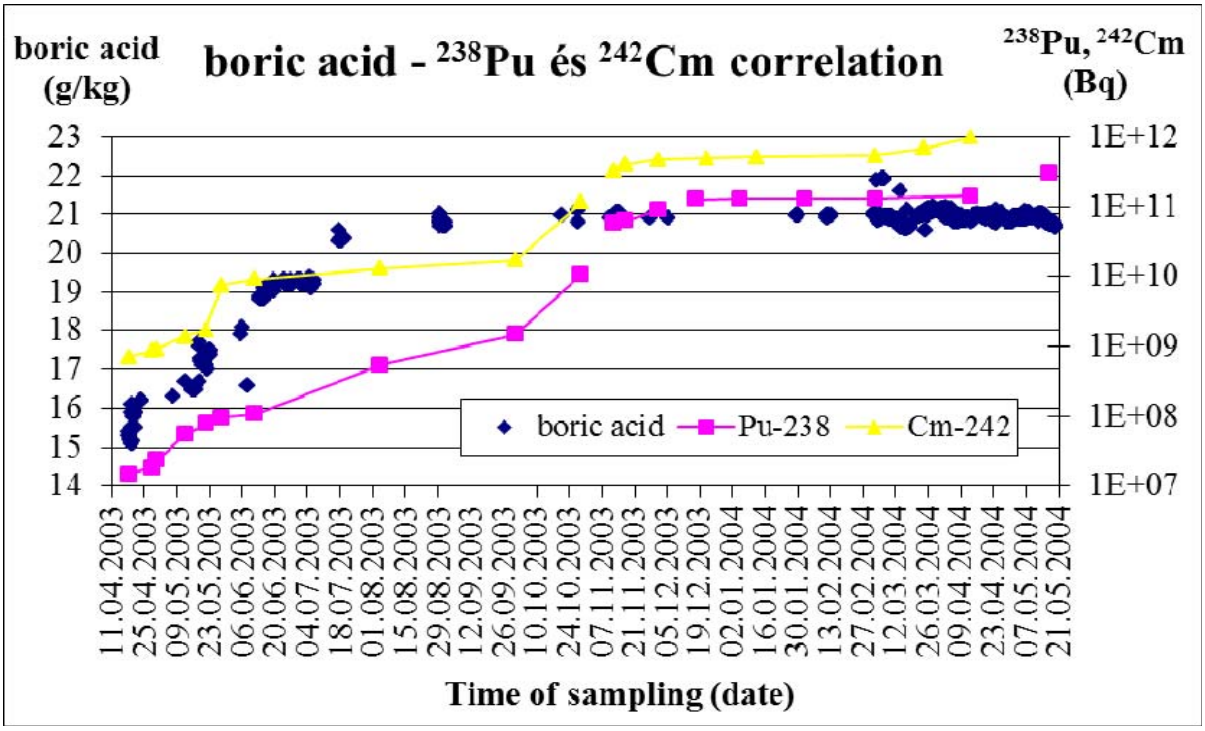


Figure 38: Change of boric acid concentration, ^{238}Pu and ^{242}Cm activity concentration.

The uranium concentration (µg/l) measurements started in five month after the incident, so there are no uranium data for the time period close to the incident. Using the available data the amount of dissolved UO_2 was calculated and compared to the original mass of the fuel. Using this approach a relative release or dissolution rate was calculated, that is similar to the release rates of the radioactive isotopes. The data shows that 1.8 % (71.5 kg) of the total mass (3969 kg) of the UO_2 was dissolved during the four years of wet storage after the incident. This means that 19 days were necessary for the dissolution of 1 kg UO_2 into the coolant and the average release rate of $UO_2 = 1.06 \cdot 10^{-1}$ g/d.

Two linears were fitted if the isotope was measured (column 4 and 5 in Table 8 and Table 9), one for the interval of the incident and one for the interval of the wet storage. In case of the long lived isotopes the release rate during the wet storage gives good agreement with the calculated release rate. For most isotopes the fitted value underestimate the ralase rate which was calculated from the integrated releases (Table 8 and Table 9). The fitted release rates of ^{144}Ce and ^{238}Pu isotopes approximate best their calculated release rates, the differences are less than 5%. In case of caesium and europium isotopes these values were between 20% and 30%. At the same time the calculated release rates of ^{125}Sb and curium isotopes exceed the fitted release rates by about 15%. The fitted release rate of UO_2 overestimates the calculated release rate by 11%.

8.3. Release rates of isotopes from leaking VVER fuel stored in water

Calculation methods

The processed activity concentration data of isotopes were originated from the measurements which were between 27th April 2009 (reactor shutdown) and 3rd May 2010. The release rate of isotopes was estimated based on the direct measurements which were available from the spent

fuel storage pool of Unit 4. The elapsed time since the reactor shutdown was divided into short periods even as in case of isotopes which were released from the damaged fuel assemblies (see section 8.1). The number of periods was different for the different isotopes. For example in the case of ^{137}Cs isotope 31 periods were identified. The method of evaluation of release rates corresponds with the above described calculation. It should be noted that only those releases were used in this evaluation which were characterized by steady-state release rates; the transients as a sipping were not taken account of (Figure 39). The isotope inventory of the reactor shutdown was used to correct the integrated releases.

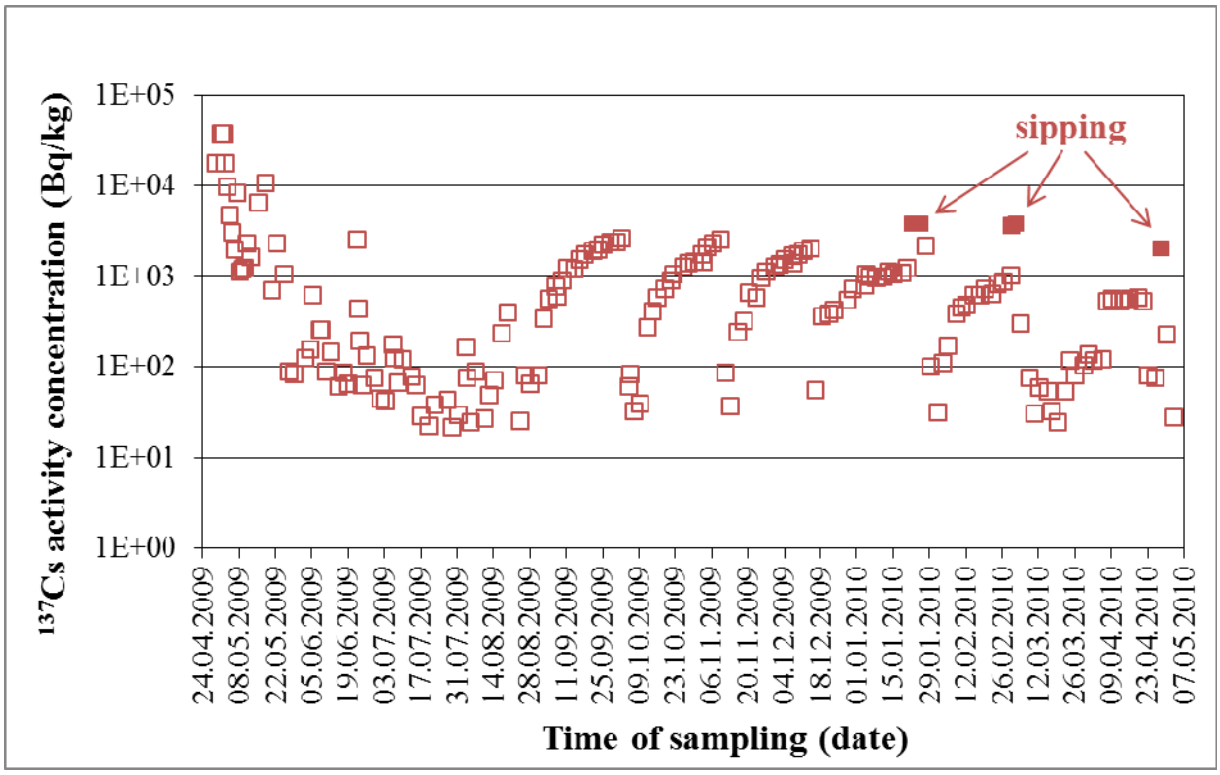


Figure 39: History of ^{137}Cs activity concentration in the spent fuel storage pool of Unit 4 (Paks NPP).

Results of the calculations

In case of the No. 70873 leaking fuel assembly, the fitted release rates of isotopes approximate well the calculated release rates. The release rates of the ^{125}Sb are given the best agreement. The fitted release rates of the other examined isotopes underestimate their calculated release rates (Table 10). The difference is under 7% for the ^{95}Zr, ^{106}Ru, ^{137}Cs and ^{134}Cs isotopes, while this value for the ^{154}Eu and ^{241}Am isotopes are under 20%. The most significant difference is in case of ^{155}Eu by 33%. The fitted release rate of UO_2 underestimates the calculated release rate too, by 16%.

During the wet storage the release rate range of long lived isotopes was $1 \cdot 10^{-8}$-$5 \cdot 10^{-8}$ rate/d (Table 8). These values are three to six time higher than during the leakage (Figure 40). The release of the ^{144}Ce isotope from the inventory two orders of magnitudes was higher during the wet storage. The fitted release rates of ^{125}Sb isotope show the best agreement.

Table 10: Release rates from leaking VVER fuel - calculated and fitted release rates of isotopes.

Isotope	Isotope inventory (Bq)	Calculated release rate (rate/d)	Fitted release rate (rate/d)
^{95}Zr	7,40E+13	6,81E-11	**6,59E-11**
^{144}Ce	3,41E+13	5,40E-11	**3,79E-11**
^{106}Ru	7,61E+12	4,22E-10	**4,11E-10**
^{137}Cs	1,59E+12	1,08E-08	**1,01E-08**
^{134}Cs	1,08E+12	1,14E-08	**1,06E-08**
^{125}Sb	1,12E+11	1,16E-08	**1,18E-08**
^{154}Eu	3,74E+10	7,87E-09	**6,63E-09**
^{155}Eu	2,35E+10	2,06E-08	**1,37E-08**
^{241}Am	6,25E+08	2,21E-06	**1,80E-06**
UO_2	-	1,49E-07	**1,25E-07**

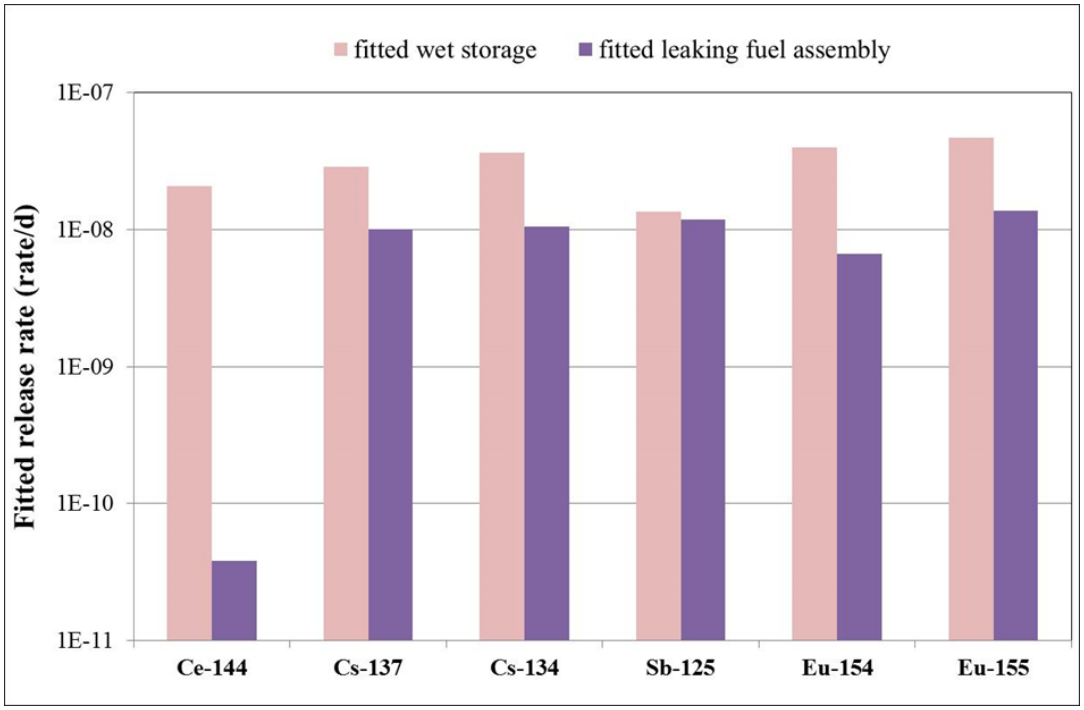


Figure 40: Comparison of the fitted release rate of long lived isotopes

8.4. Conclusions

The dissolution rates for VVER fuel stored in water for long periods were determined on the basis of data from the Paks NPP. The two datasets provided information on slightly different

conditions: in both cases the water temperature was similar, but in cases of damaged fuel the pH of coolant was significantly lower compared to the leaking fuel. This effect can explain the observed differences in dissolution rates (Figure 40).

Following a review of the reliability of measured data, the dissolution rates were determined for eleven isotopes in case of damaged fuel and for nine isotopes in case of leaking fuel. Additionally urania dissolution rates were calculated in both cases. In most of the cases the release rates of individual isotopes were comparable to that of urania.

The data produced for VVER fuel should be compared to other dissolution rates that will be determined in the framework of the FIRST-Nuclides project. Furthermore, the present VVER specific data can be used for the estimation of migration of fission product from deep geological repository with VVER fuel.

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9. STUDSVIK - SNF sample characterisation, methodologies and tools

O. Roth

Four BWR fuels with average BU of 50.2, 54.8, 57.1 and 59.1 GWd·(t HM)$^{-1}$, respectively, and two PWR fuel with 54.4 and 70.2 GWd/tHM, respectively, were chosen for spent nuclear fuel leaching experiments and laser ablation studies at STUDSVIK. The selected fuels are listed in Table Table 11. A preliminary data-set of the six fuel rods were compiled in Deliverable 1.1 (Metz *et al.,* 2012) of FIRST-Nuclides. In chapter 10, Table 19 to Table 24 give an updated data-set of the fuel rods studied by STUDSVIK.

The main focus of the investigations is to explore the effects of additives and dopants on the fast/instant release of fission products such as Cs and I. Experiments will also be performed to investigate the feasibility of measuring fast/instant release of Se and C-14. Furthermore, laser ablation experiments will be performed to study the radial distribution of I, Xe and Cs and to explore any correlation to the fission gas release (FGR) and instant release leach rates of the corresponding fuel samples.

Table 11: High burn-up fuels selected for investigation at STUDSVIK.

Sample name	Reactor type	Fuel type	Calculated BU (rod average) [MWd/kg U]
D07	BWR	standard UO_2	50.2
L04	BWR	standard UO_2	54.8
5A2	BWR	standard UO_2	57.1
C1	BWR	Al/Cr doped UO_2	59.1
VG81	PWR	Gd doped UO_2	54.4
AM2K12	PWR	standard UO_2	70.2

9.1. Characterisation of spent BWR and PWR fuels studied at STUDSVIK

Experimental and theoretical sample data

The selected fuel rods are characterized by experimental measurement of the fission gas release (FGR) and by gamma scanning. The methods for this are described below. The average linear heat generation rate (LHGR) is estimated based on core calculations and the local burn-up is determined by core calculations and/or on the basis of gamma scan results. The nuclide inventory of the samples is determined either by core calculations or by chemical dissolution and analysis (Zwicky *et al*., 2011). The data available for the selected samples are listed in Table 2. The available sample data may be extended and refined during the remaining part of the project.

Table 12: Characteristic data of studied samples from spent nuclear fuel rods selected by STUDSVIK.

	D07	**L04**	**5A2**	**C1**	**VG81**	**AM2K12**
Reactor	Olkiluoto 1	Olkiluoto 1	Oskarshamn 3	Oskarshamn 3	Vandellos	North Anna
Initial enrichment [%]	4.25	4.25	3.5	4.1	2.8	4.0
Approximate average linear heat generation rate (LHGR) [kW/m]	14.5[1]	16.2[1]	16.0[2]	17.5[2]	13.6[3]	18.6[4]
Axial position of sample for leaching [mm from rod bottom]	776-797	752-773	2648-2668	2670-2693	885-905	To be decided
Axial position of sample for laser ablation [mm from rod bottom]	N.A.	N.A.	2628-2648	2645-2670	N.A.	N.A.
Local burn-up leaching sample based on gamma scanning [MWd/kgU]	N.A.	63.2	N.A.	61.7	Verification on-going	N.A.
Local burn-up leaching sample calculated [MWd/kgU]	58.9	65.5	62.8	65.2	N.A.	N.A.

[1] At sample position
[2] At rod maximum
[3] Average segment 2
[4] At previously analyzed sample position

9.2. Experimental and analytical methodology for SNF characterisation and SNF leaching experiments at STUDSVIK

Method for analysis of fission gas release

The fission gas release is measured by puncturing the rods at the plenum, collecting the internal gas in a standard volume and determining the pressure. Samples of the gas are collected and analysed by mass spectrometry. The total internal free volume of the rod is determined by the backfill method, using argon at constant pressure. The amount of released gas is calculated from the puncturing pressure and the volume of the puncturing system and from the gas composition.
Using the UO_2 content of the rod, the average burn-up of the rod and fission yields of Xe and Kr (interpolated from standard calculations with the Origen code) the amount of generated fission gas are determined. The fission gas release (i.e. fraction of released gas relative to generated gas) is obtained from the measured amount of released Xe and Kr isotopes, and the predicted amount of generated Xe and Kr.

Method for gamma scanning

Gamma scanning is performed by moving the sample past a Ge detector with a 0.5 mm collimator. The signal from the detector is recorded in analog or digital mode. Analog recording of the signal gives the total energy detected as a function of the axial position on the rod i.e. the gross gamma spectrum. Digital recording records the energy from each nuclide separately and a spectrum for each nuclide is generated.
The gamma spectra gives information on the positions of pellet-pellet interfaces as well as the burn-up profile. By scanning a reference sample with known burn-up in sequence with the sample, the local burn-up of the sample can be calculated from the digitally recorded Cs-137 signal.

9.3. Preparation of samples of spent BWR and PWR fuel rods by STUDSVIK

Sample preparation for SNF leaching experiments

Spent fuel leaching studies will be performed using samples from the 6 different fuel rods listed in Table 11. Sample preparation has been performed for 5 of the samples; D07, L04, 5A2, C1 and VG81.
The first step of the sample preparation is gamma scanning of the rod segment. From the resulting gamma spectra the pellet-pellet interfaces are identified and the positions for cutting are determined. The cutting positions are chosen at mid-pellet positions as shown in Figure 41 in order to get a representative sample from the rod. Thereafter samples are cut from the segment using a pipe cutter. This method of cutting requires no cooling during the cutting and hence gives minimal influence on the leaching results. Each sample consists of approximately 2 fuel pellets including cladding.
The sample compartments consist of either cladded fuel segments or fuel fragments + separated cladding. The fuel segments are weighed before the start of the leaching experiments. To obtain the samples consisting of fuel fragments + separated cladding, the cladding is cut axially using a diamond wheel and bent open which causes the fuel to fragment and detach from the cladding. The fuel fragments are collected and weighed and leached together with the cladding (and any remaining fuel still attached to the inside of the cladding).

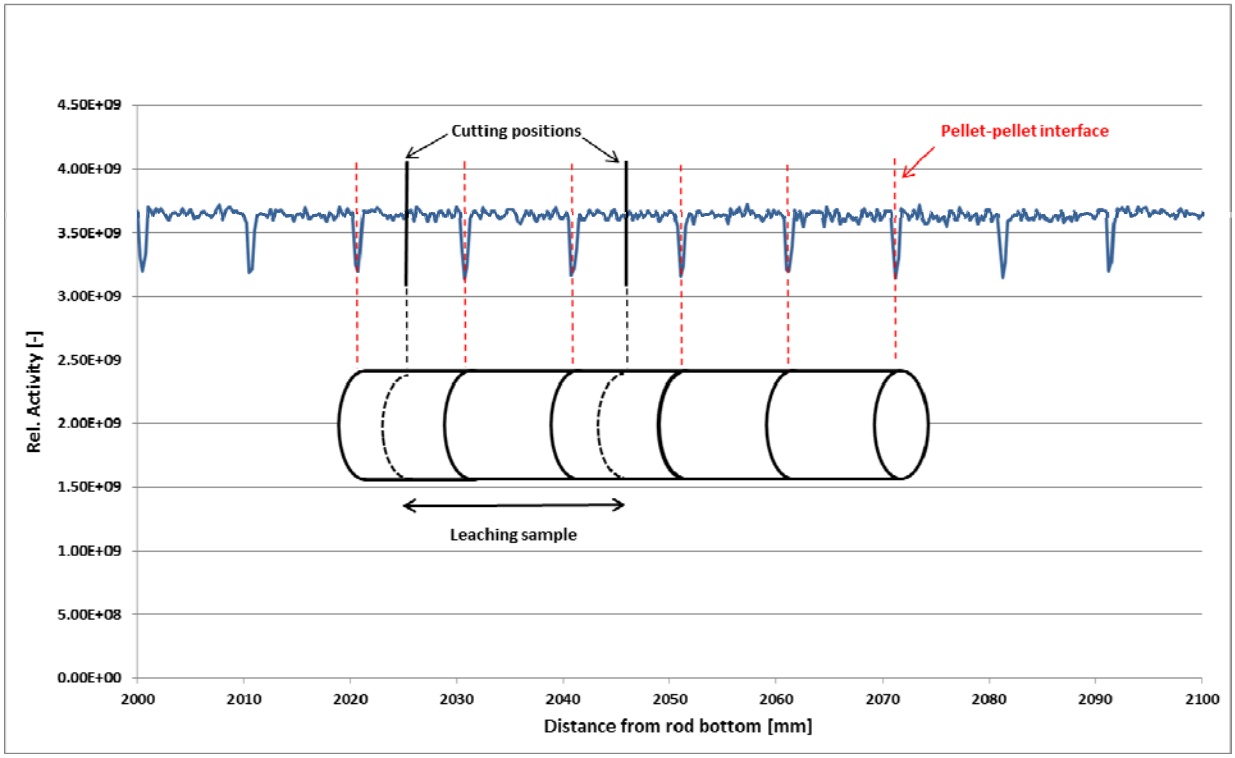

Figure 41: Schematic picture of leaching sample position relative to gamma spectrum

In a next step the leaching of powdered specimens will be performed using a simultaneous grinding and leaching technique described by Stroes-Gascoyne *et. al.* (1995). The objective of this method is to expose the grain boundaries by grinding the fuel down to the same size range as the individual fuel grains. This would ideally make the entire grain boundary inventory available for leaching. By combining the grinding and leaching into one wet grinding step surface oxidation and temperature effects (from the friction of grinding) can be minimized.

Sample preparation for laser ablation studies

Two samples were selected for laser ablation studies; 5A2 and C1 in Table 11. The two samples have a similar irradiation history but consist of different pellet types (standard UO_2 and Al/Cr doped UO_2). Laser ablation is used to study the radial distribution of I, Xe and Cs and to investigate any differences between the two pellet types.

Preparation of the two samples has been performed during 2012. The samples studied are cross sections (transversal cut, perpendicular to the axial direction of the rod) taken from a neighbour pellet to the fuel sample for the leaching investigation. The samples are prepared by cutting a rod segment approximately 2 pellets (½+ 1 + ½ pellet) long in the same manner as for the leaching sample described above (Figure 41). The sample is mounted in epoxy and then cut in half (transversal cut through the middle pellet) in order to expose a fresh mid-pellet surface to be used for the laser ablation. The surface is polished slightly before the start of the experiment.

10. Fuel characterisation data-sets of spent BWR and PWR fuels under investigation

Table 13: **Characteristic data of the UO_2 fuel rod segment SBS1108-N0204 (discharged from PWR Gösgen) selected for investigations 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 per assembly
		External fuel rod diameter: 10.75 ± 0.05 mm before irradiation 10.67 – 10.73 mm (10.71 mm mean) diameter after irradiation (Wegen et al., 2012; 2013c)
		Fuel rod # SBS 1108, Segment N 0204
Fuel rod data	essential	Test fuel rod
		Internal rod pre-pressure: 21.5 ± 1 bar He Fission gas release: 7 % Kr, 8.5 % Xe (Kienzler et al., submitted)
Fuel material data	essential	UO_2 fuel, initial enrichment 3.8% ^{235}U, U_{nat}, resp.
		O/U = 2.002; fuel fabrication without additives
		Pellet dimensions: ∅ = 9.2 mm, length = 11 mm before irradiation average length = 11.5 mm after irrad. (3.8% ^{235}U) average length = 11.3 mm after irradiation (U_{nat}) (Wegen et al., 2013b)
		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 ELS 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: 340 W cm^{-1} Discharge: 27. May 1989 Irradiation duration: 1226 days

Table 14: Characteristic data of a 54 GWd·(t HM)$^{-1}$ BWR fuel rod (denoted as "BWR54") and a 42 GWd·(t HM)$^{-1}$ BWR fuel rod (denoted as "BWR42") selected for investigations by CTM and JRC-ITU.

	BWR54	BWR42
Reactor	BWR	BWR
	light water coolant	light water coolant
Fuel assembly design information	Lattice geometry: 10 x 10	*Lattice geometry: *t.b.p.*
	Fuel rod outer diameter: 10.05 mm Fuel rod inner diameter: 8.84 mm	Fuel rod outer diameter: 11.0 mm Fuel rod inner diameter: 9.4 mm
Fuel rod data	Standard rod	Standard rod
	Fission gas release: 3.9%	Fission gas release: 2.0%
	Internal rod pre-pressure: 5.5 bar	*Internal rod pre-pressure: *t.b.p.*
Fuel material data	UO_2 fuel, initial enrichment 4.2 % ^{235}U	UO_2 fuel, initial enrichment 3.7 % ^{235}U
	Pellet diameter: 8.67 mm Pellet height: 10.5 mm	*Pellet diameter: *t.b.p.* *Pellet height: *t.b.p.*
	Fuel density: 10.45 g/cm^3	*Fuel density: *t.b.p.*
	Grain size:10.1 μm	*Grain size: *t.b.p.*
	Calculated radionuclide inventory with ORIGEN-ARP code (see Table 1)	Calculated radionuclide inventory with ORIGEN-ARP code (see Table 1)
Cladding data	Zircaloy-2 Oxide thickness: 21 μm	**t.b.p.*
Irradiation data	Rod average Burn-up: 54.24 GWd·(t HM)$^{-1}$ Number of cycles: 7 Average linear power: 160 W cm^{-1} Maximal linear power: 230 W cm^{-1}	Rod average Burn-up: 42.22 GWd·(t HM)$^{-1}$ *Number of cycles: *t.b.p.* *Average linear power: *t.b.p.* *Maximal linear power: *t.b.p.*
	Date of loading: 22. March 2002 Discharge date: 23. June 2007 Irradiation duration: 1795 days	*Date of loading: *t.b.p.* Discharge date: 15. June 1998 Irradiation duration: 1442 days
	Average fuel centre temperature at full power: 461°C	*Average fuel centre temperature at full power: *t.b.p.*

*t.b.p. – to be provided by JRC-ITU in update of this report

Table 15: Characteristic data of a UO_2 fuel rod discharged from BWR Leibstadt (average burn-up of 57.5 GWd·(t HM)$^{-1}$) selected for investigations by PSI.

Data category	Parameter
Reactor	Leibstadt NPP, Switzerland Boiling Water Reactor, BWR
Fuel assembly design information	10 x 10 Lattice SVEA96 Optima, fuel assembly AIA003, rod position H6, Node 4
Assembly / cladding material compositon	Zircaloy-2, designation LK3/L
Fuel rod data	Fission gas release: 2.26 % Rod length as fabricated: 4146.6 Rod length after irradiation: 4163.3 Internal rod pre-pressure: 7 bar
Fuel material (pellet) data	UO_2, initial enrichment: 3.9% U-235 Pellet diameter (as fabricated): 8.77 ± 0.013 mm Pellet length: 10.7 ± 0.8 mm Density (spec) 10.52 ± 0.19 g/cm^3 Density as fabricated 10.48 – 10.54 g/cm^3 Grain size $6 \leq x \leq 25$ μm
Fuel sample data	Sample position 455 mm to 520 mm from BEP
Cladding sample data	Zircaloy-2 with liner, 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	Rod average burn-up: 57.5 GWd·(t HM)$^{-1}$, exp. determined local burn-up: 6.1% FIMA Number of cycles: 7 Date of loading: August 1998 Date of unloading: April 2005 Duration of irradiation: 2400 days Average linear power: ~ 160 W/cm Max. linear power: 270 W/cm

Table 16: Characteristic data of a UO_2 fuel rod discharged from PWR Gösgen (average burn-up of 62.6 GWd·(t HM)$^{-1}$) selected for investigations by PSI.

Data category	Parameter
Reactor	Gösgen NPP, Switzerland PWR
Fuel assembly design information	KKG-14B-4021-01-0129 Lattice geometry: 15x15, 48 assemblies with 20 control rods/assembly Fuel rod diameter: 10.765 mm Fuel rod diameter after irradiation: 10.697 mm
Assembly / cladding material composition	Zircaloy 4, DX HPA4 (0.6Sn)
Fuel rod data	Test rod Fission gas release: 13.2%, 51.5 bar Internal rod pre-pressure: 22 bar Rod length as fabricated: 3860 mm Rod length after irradiation: 3879 mm Active length: 3550 mm
Fuel material (pellet) data	UO_2, initial enrichment: 4.3 % U-235 Fuel density (as fabricated): 10.45 g/cm3
Fuel sample data	Sample position 2620 mm to 2695 mm from BEP
Cladding sample data	Cladding outer diameter: 10.75 ± 0.05 mm Cladding inner diameter: max. 9.45 mm Max. Oxide: 36 µm
Irradiation data	Rod average burn-up: 62.2 GWd·(t HM)$^{-1}$, exp. determined local burn-up: to be performed Number of cycles: 4 Date of loading: 28.7.1999 Date of unloading: 28.6.2003 Duration of irradiation: 1324.43 days

Table 17: Characteristic data of a MOX fuel rod discharged from PWR Gösgen (average burn-up of 63.0 GWd·(t HM)$^{-1}$) selected for investigations by PSI.

Data category	Parameter
Reactor	Gösgen NPP LPWR
Fuel assembly design information	KKG-13-5024-10-676 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
Assembly / cladding material composition	Duplex ELS0.8b
Fuel rod data	Test rod Fission gas release: 26.7%, 76.4 bar Internal rod pre-pressure: 22 bar Rod length as fabricated: 3859.0 mm Rod length after irradiation: 3886.1 mm Active length: 3550 mm
Fuel material (pellet) data	MOX, initial enrichment: 5.5 % Pu_{fiss} Fuel density (as fabricated): 10.45 ± 0.15 g/cm3 Fuel density after irradiation: 9.903 g/cm3 Pellet diameter (as fabricated): 9.13 ± 0.013 mm
Fuel sample data	Sample position 2030 mm to 2070 mm from BEP
Cladding sample data	Cladding outer diameter: 10.75 ± 0.05 mm Cladding inner diameter: 9.30 ± 0.04 mm Max. Oxid: 36 µm
Irradiation data	Rod average burn-up: 63.0 GWd·(t HM)$^{-1}$, exp. determined local burn-up: 7.3% FIMA Number of cycles: 4 Date of loading: 30.06.1997 Date of unloading: 07.07.2001 Duration of irradiation: 1368 days Average linear power: 306 W/cm (average fuel rod power in 4 cycles) Max. linear power: approx. 430 W/cm

Table 18: Characteristic data of a UO_2 fuel rod discharged from PWR Tihange-1 (average burn-up of ~50 GWd·(t HM)$^{-1}$) 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	rod D05, fuel assembly ref.: FT1X57
		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	UO_2, initial enrichment: 4.25 % U-235
		Fuel density (as fabricated): 96% theoretical value Pellet dimensions: Diameter: 9.294 mm Length: 11.15 mm
Cladding data	essential	M5 (Zr-Nb-Fe-O) alloy (nominal ZrNb) Cladding outer diameter: 10.720 mm Cladding inner diameter: 9.480 mm Wall thickness: 0.62 mm Initial radial gap fuel / cladding: 0.095 mm, i.e. (9.480 mm – 9.294 mm)·0.5 = 0.095 mm
Irradiation data	essential	Calculated burn-up: approximate fuel rod burn-up: 50 GWd (t HM)$^{-1}$ generic BU calculation of sample: 55 GWd (t HM)$^{-1}$ Number of cycles: 2 Average linear power: around 330 W/cm Date of loading: *to be published* Date of discharge: *to be published* Irradiation duration: *to be published*

Table 19: Characteristic data of a UO_2 fuel rod D07 discharged from BWR Olikluoto-1 (average burn-up of 50.2 GWd·(t HM)$^{-1}$) selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Olikluoto-1, 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: 1.52 % Kr, 1.57 % Xe
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)$^{-1}$ Average linear power, approximate average LHGR: 14.5 kW/m (at position of studied sample) Date of loading: 6. June 2003 Date of discharge: 13. May 2008

Table 20: Characteristic data of a UO_2 fuel rod L04 discharged from BWR Olikluoto-1 (average burn-up of 54.8 GWd·(t HM)$^{-1}$) selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Olikluoto-1, 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: 3.07 % Kr, 3.06 % Xe
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)$^{-1}$ Average linear power ~20 kW/cm (to be confirmed), approximate average LHGR: 16.2 kW/m (at position of studied sample) Date of loading: 6. June 2003 Date of discharge: 13. May 2008

Table 21: Characteristic data of a UO_2 fuel rod 5A2 discharged from BWR Oskarshamn-3 (average burn-up of 57.1 GWd·(t HM)$^{-1}$) selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Oskarshamn-3
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 9.62 mm
		denoted as "5A2"
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 81 bar
		Fission gas release: 2.48 % Kr, 2.44 % Xe
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)$^{-1}$ Average linear power, approximate average LHGR: 16.0 kW/m (at rod maximum) Date of loading: 10. February 2000 Date of discharge: 2. October 2008 Irradiation period: 2000-2006, 2007-2008

Table 22: Characteristic data of a UO_2 fuel rod C1 discharged from BWR Oskarshamn-3 (average burn-up of 59.1 GWd·(t HM)$^{-1}$) selected for investigations by STUDSVIK.

Data category	Information category	Parameter
Reactor	essential	BWR Oskarshamn-3
		light water coolant
Fuel assembly design information	essential	Lattice geometry: 10x10
		Fuel rod diameter: 9.62 mm
		denoted as "C1"
Fuel rod data	essential	Standard fuel rod
		Internal rod pre-pressure: 83 bar
		Fission gas release: 1.34 % Kr, 1.46 % Xe
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)$^{-1}$ Average linear power, approximate average LHGR: 17.5 kW/m (at rod maximum) Date of loading: 10. February 2000 Date of discharge: 2. October 2008 Irradiation period: 2000-2006, 2007-2008

Table 23: Characteristic data of a UO_2 fuel rod VG81 discharged from PWR Vandellòs (average burn-up of 54.4 GWd·(t HM)$^{-1}$) 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: 1.92 % Kr, 2.30 % Xe
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); approximate average LHGR of studied fuel segment 13.6 kW/m: Date of loading: 10. Oct. 2000 Date of discharge: 5. May 2007

Table 24: Characteristic data of a UO_2 fuel rod AM2K12 discharged from PWR North Anna (average burn-up of 70.2 GWd·(t HM)$^{-1}$) 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: 5.04 % Kr, 4.85 % Xe
Fuel material data	essential	UO2 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, approximate average LHGR: 18.6 kW/m (at previously analyzed sample position) Number of cycles: 14 Date of loading: 7. July 1987 Date of discharge: 12. March 2001

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