PWR and BWR Fuel Assay Data Measurements

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Abstract - In the framework of a high burnup fuel demonstration program, rods with an enrichment of 4.5% ^{235}U were operated to a rod average burnup of about 70 MWd/kgU in the Spanish Vandellós 2 pressurised water reactor. The rods were sent to hot cells and used for different research projects. This paper describes the isotopic composition measurements performed on samples of those rods, with burnup values ranging from 40 MWd/kgU up to 75 MWd/kgU. The main results obtained and their comparison with values calculated using different methods are presented.
In 2008, a project to measure the isotopic composition of irradiated BWR fuel was started. The mother rod was irradiated in the Swedish Forsmark-3 boiling water reactor, and the fuel samples selected have a burnup ranging from 40 to 55 MWd/kgU. The paper describes the scope of the project and its current status.

I. INTRODUCTION

This paper presents an update of the research activities recently developed in Spain to obtain representative measurements of isotopic composition of spent PWR and BWR fuel irradiated under current operating conditions of Spanish reactors. The final objective of the project is the use of the resulting data as a basis for the validation of the composition calculation methodologies (reactivity, decay heat, shielding) currently applied in the country, considering in-reactor behaviour and storage and transport needs.

These activities are the result of a cooperative spent fuel research effort started in 2003 by the Consejo de Seguridad Nuclear (CSN), ENUSA Industrias Avanzadas (Enusa) and ENRESA, Spanish organization responsible for waste management in the country. In this framework, an experimental program, aimed to cover the lack of published experimental data representative of the isotopic composition of high burnup fuel currently operated in Spain, has been developed.

The program, and consequently this paper, is structured into two projects clearly distinguished in the time schedule as well as in the type of nuclear material used, PWR and BWR respectively, but throughout similar regarding to its objective, scope and experimental and analytical work performed.

The first project was aimed to measure the isotopic composition of PWR spent fuel with an initial enrichment of 4.5% ^{235}U, at several burnup values up to 75 Mwd/kgU. The measurements were performed at the Studsvik laboratories in Sweden, and the project was finished early in 2006. The results will be published in the open literature in 2010, and will be offered to the NEA for inclusion in the SFCOMPO database.

The second project is devoted to BWR spent fuel with an initial enrichment of 3.95% ^{235}U and burnup values up to 53 Mwd/kgU. The measurements have also been performed at the Studsvik laboratories, and the project will finish in 2010. It is also foreseen to offer the data to the NEA for inclusion in the SFCOMPO database.

The project raised the interest of the US-NRC and US-DOE, and as a result a cooperation agreement was established with Oak Ridge National Laboratory for these projects. This cooperation has allowed for the extension of the project with the assay of additional fuel samples and the measurement of additional isotopes.

II. PWR PROJECT

II.1 BACKGROUND AND SCOPE

In the framework of a high burnup fuel demonstration program, rods with an enrichment of 4.5% ^{235}U were operated to a rod average burnup of about 70 MWd/kgU in the Spanish Vandellós 2 pressurized water reactor. The rods were sent to hot cells and used for different research projects, the isotopic composition measurement being one of them.

Before cutting the rods into segments for further characterisation, some rods were punctured and the amount of gas in the free rod volume was determined. The composition of the gas was determined by gas mass spectrometry and the fraction of released fission gases assessed. Afterwards, local burnup and isotopic content of gamma emitting nuclides were determined by quantitatively evaluating axial gamma scans of the full rods.

Chemical analysis of dissolved pellet samples is one of the key methods to be applied in post-irradiation examination of irradiated fuel. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) after separation with High Performance Liquid Chromatography (HPLC) has been the chemical analysis method applied. Nuclide analyses in dissolved fuel samples were performed in Studsvik in two campaigns, the first one in 2003 on seven samples from two different fuel rods, the second one in 2006/07, re-analysing solutions from the first campaign and characterising two additional samples dissolved by alternative methods. Final reports of the projects were finished by April 2008.

More than 50 isotopes of 16 different elements were assessed, and the results obtained are consistent. Only in a few cases, the analysis provided unexpected results that seem to be erroneous, in most cases due to unidentified reasons. Sample burnup analysis was performed by comparing experimental isotopic abundances of uranium and plutonium composition as well as neodymium isotopic concentrations with corresponding CASMO based data. The results were consistent with values derived independently from gamma scanning and from core design data and plant operating records.

Measured isotope abundances were finally assessed using the industry standard SAS2H sequence of the SCALE code system. This exercise showed good agreement between measured and calculated values for most of the analyzed isotopes, similar to those reported previously for lower burnup ranges.

II.2 SELECTED ISOTOPES

The list of nuclides measured in this project is included in Table 1. The nuclides have been selected on the basis of their relevance for criticality safety, radiation shielding and residual heat calculations [3]. However, some relevant nuclides have been excluded due to practical reasons. This has been the case for some metallic isotopes that have an important reactivity worth, but for which a reliable concentration measurement is difficult. Not all listed isotopes have been analysed in all samples.

Table 1. Selected isotopes for PWR project

Element	Atomic mass isotope	Method
U	233 234 235 236 238	IDA/ICP-MS
Pu	238 239 240 241 242	IDA/HPLC-ICP-MS
Np	237	IDA/HPLC-ICP-MS + One-point calibration
Am	241 243	IDA/HPLC-ICP-MS
Cm	244 246	IDA/HPLC-ICP-MS + One-point calibration
Mo	95 97 98 199	IDA/HPLC-ICP-MS
Tc	99	One-point calibration
Ru	103 106	One-point calibration /γ-scan
Rh	103	γ-scan
Cs	133 134 135 137	One-point / γ-scan / One-point / γ-scan
La	139	One-point calibration
Ce	140 142 144	IDA/HPLC-ICP-MS / IDA/HPLC-ICP-MS / γ-scan
Nd	142 143 144 145 146 148 150	IDA/HPLC-ICP-MS
Sm	147 148 149 150 151 152 154	IDA/HPLC-ICP-MS
Eu	151 153 154 155	IDA/HPLC-ICP-MS / IDA/HPLC-ICP-MS/ One-point calibration /γ-scan
Gd	154 155 156 157 158 160	IDA/HPLC-ICP-MS

II.3 FUEL SAMPLES

Fuel rods with an initial ^{235}U enrichment of 4.5 wt% with three different cladding material types were fabricated by ENUSA and irradiated to a rod average burnup of about 70 MWd/kgU in the Spanish Vandellós 2 pressurised water reactor during cycles 7 to 11, between June 1994 and September 2000 in the framework of a high burnup fuel demonstration program. During cycles 7 to 10, the rods were located in four different assemblies operated in symmetric positions in the reactor core. For their last cycle, the rods were removed from their original assemblies and inserted into different positions of the same bundle. In total, nine samples were cut from three different rods and analysed in two campaigns in 2003 and in 2006/07, respectively. Sample designations, positions and approximate burnup values are compiled in Table 2.

Table 2. Analyzed samples in PWR project

Rod	Sample	Position[a] [mm]	Local burnup[b] [MWd/kgU]	Remarks
WZR0058	E58-88	88 - 90	41	Analysed in 1st campaign
	E58-148	148 - 150	52	Analysed in 1st and 2nd campaign
	E58-257	252 - 262.5	63	Analysed in 2nd campaign
	E58-263	263 - 265	63	Analysed in 1st campaign
	E58-773	773 - 775	74	Analysed in 1st campaign
	E58-793	793 - 795	74	Analysed in 1st and 2nd campaign
	E58-796	796 - 798	74	Analysed in 1st campaign
WZtR160	WZtR160-800	792 - 802.5	73	Analysed in 2nd campaign
WZtR165	WZtR165-2a	1060 - 1062	75	Analysed in 1st campaign

[a] From bottom end of rod
[b] Based on gamma scanning

II.4 EXPERIMENTAL WORK: APPLIED METHODS

Experimental work includes rod puncturing, γ-spectrometry of the rod, chemical analyses of dissolved fuel pellet samples and burnup determination. All of these methods are briefly described below.

II.4.1 Rod puncturing

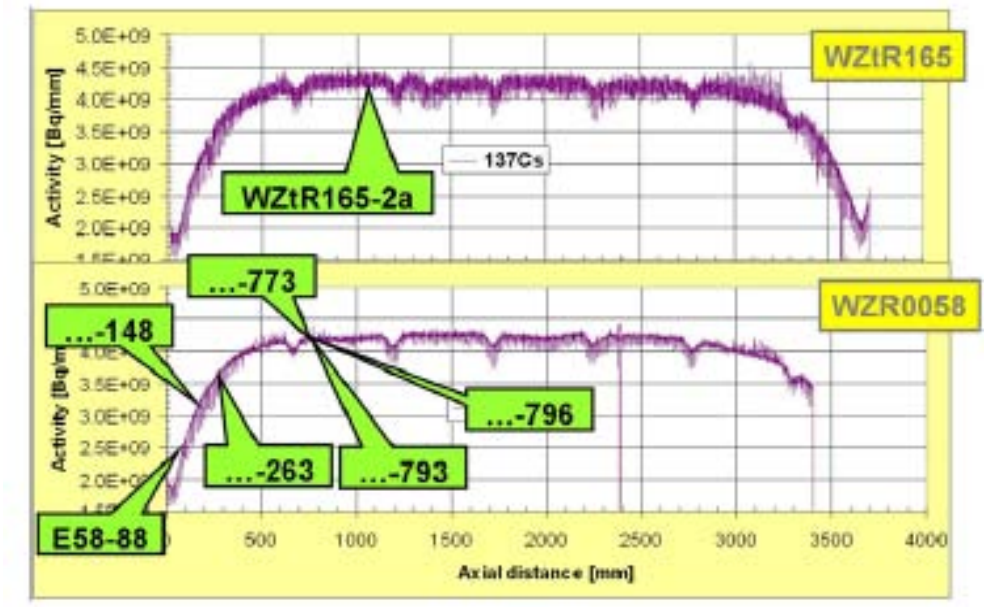
Fission gas release was determined in rods WZtR165 and WZR0058. The rods were punctured in the plenum. The internal gas was expanded into a standard volume and the resulting pressure was determined. Samples of the gas were collected for analysis by mass spectrometry. The total internal free volume of the rod was determined by the backfill method, using argon at constant pressure. The fission gas release fraction was calculated from the experimentally determined amount of Xenon and Krypton extracted from the rod and the ORIGEN calculated total inventory of fission gases generated in the fuel over its entire irradiation.

II.4.2 Gamma scanning

A high purity germanium detector behind a 0.5 mm tungsten collimator was used for the measurements. Activities were decay corrected to the end of irradiation. A well characterised reference rod was scanned together with the segments. By comparing the apparent ^{137}Cs activity measured for the reference rod with the decay corrected ^{137}Cs activity known from the characterisation of the rod, a correction factor to be applied on all apparent activities was determined. ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{154}Eu, ^{134}Cs, ^{137}Cs, ^{144}Ce and ^{144}Pr were measured by this method.

After γ-scanning, the rods were cut into segments before they were further investigated. Test samples were cut according to Figure 1 where ^{137}Cs γ-scan of rods WZtR165 and WZR0058 is plotted. Only samples selected for the first campaign are shown.

Figure 1. Position and designation of samples selected for radiochemical analysis



II.4.3 Chemical analysis

Dissolution

In the first campaign, a fuel rod slice of about 2 mm long, was placed in a glass flask together with 20 ml of concentrated HNO_3 and kept at 65°C for 6 h. Evaporation of liquid was avoided by means of an air-cooled reflux cooler. Nitrogen was bubbled through the liquid in order to stir it. The fuel matrix

together with all fission products of interest went into solution. The cladding and the metallic fission product inclusions remained undisolved.

The two 10 mm samples E58-257 and WZtR160-800, dissolved in the second campaign, were each cut in two pieces and dissolved in a Large Capacity Bomb. This high pressure bomb has a capacity of 125 ml and can be used at 250°C with a pressure of 1900 psig.

The two pieces of sample E58-257 were each dissolved in 20 ml of a 2:1 mixture of concentrated HCl and HNO3 during 6 hours at 200°C. The cladding was found to have disintegrated, with only a few larger pieces left. A grey residue was found on the bottom of the container. Therefore, the two pieces of sample WZtR160-800 were dissolved in two steps, first in 15 ml HNO3 (suprapur) during 4 hours at 180°C. After removal of the cladding ring, alloy particles were dissolved by adding 30 ml concentrated HCl, followed by a second heat-up to 200°C for 4 hours. All solutions were combined in one single flask. In the order of 0.1-0.4 g of the original fuel solution was diluted into 100 ml of HNO3 (7.5 M) in the hotcell. 20 ml of this solution were transferred to the laboratory. An appropriate aliquot was diluted with 100 ml HNO3 (0.16 M) to a target uranium concentration of about 4 µg/g. The uranium concentration was determined by an uranium analyser, measuring the characteristic fluorescence of the uranyl ion in solution after irradiation with a very short pulse of ultraviolet light from a nitrogen laser. 30 g of this solution was then mixed with all necessary spike solutions.

ICP-MS and Isotopic Dilution Analyses

The instrument and methods used for the chemical analysis have been described in [1].

II.4.4 Burnup determination

Modern physics codes like CASMO and HELIOS are able to calculate the amount of fission products and actinides formed or consumed during reactor operation in a sophisticated way, taking changes of irradiating conditions into account in a more detailed manner than old methods according to ASTM E 321 and ASTM E 244 based in isotopic composition in ^{148}Nd and uranium and plutonium respectively. The uncertainty of these methods can therefore be eliminated to a certain extent, if the experimentally determined amount of suitable fission products or actinides is compared to the result of, e.g., CASMO calculations. Cross sections applied for CASMO calculations of isotope number densities are in general well known, at least in the case of fission products that are candidates for being used for burnup determination.

The accuracy of CASMO results depends primarily on the quality of modelling operating history. In the case of the Vandellós 2 high burnup program, operation was well documented, thus allowing a quite detailed modelling. Therefore, the error of CASMO calculations is assumed to be smaller than experimental errors. The method is based on detailed information on operating conditions, number densities of all actinide and fission product isotopes of interest were calculated as a function of burnup by CASMO. The principle of the method is further described in [5]

The number densities are transformed into $^{n}X/^{238}U$ values. Experimentally determined values for ^{146}Nd, ^{148}Nd and ^{150}Nd are compared to the calculated values, thus allowing a determination of the local pellet burnup. In addition, local pellet burnup is determined by comparing ^{235}U and ^{239}Pu isotopic abundances analysed by ICP-MS to isotopic abundances calculated from CASMO number densities. Burnup was determined as well on the bases of ^{137}Cs activities measured by gamma scanning in the analysed rod and in the reference rod. The decay of ^{137}Cs during irradiation was calculated based on real power histories.

II.5 ANALYTICAL WORK

The well known (industry standard) SAS2H sequence of the SCALE4.4 code system [4] was used to assess the measured nuclide inventories for the sample rods WZR0058 and WZTR165. The aim of this analysis was twofold:

1. Serve as an independent validation check for the results obtained with a new measurement technique.
2. Identify potential limitations and/or biases of this widely used tool to accurately predict isotopic inventories for high burnup fuel.

The results obtained [2] are presented in section II.6. The input data required to perform the sample isotopic analysis with SAS2H includes the specific burnup history of the sample, the fuel temperature, the moderator temperature and density, the boron curves and the power operation factors for each sample rod. Best estimate values for all these inputs were obtained from the plant records and/or from the core design values for the operating cycles where the rods were irradiated. The burnup for each sample was obtained from the calculated fuel rod burnup profile, corrected, when necessary, with plant flux map measured data.

II.6 SELECTED RESULTS

II.6.1 Released fission gases

Fission gas release was calculated assuming two different amounts of energy released per fission. With about 8% of the amount produced during operation, the fractional release of fission gases is well comparable to published data. In rods containing fuel with an initial enrichment of 3.5 – 4.2%, operated in 15x15 fuel assembly geometry in a commercial PWR at somewhat higher average linear heat rates than the Vandellós high burnup rods, about 10 – 15% released fission gases were found at 70 – 80 MWd/kgU burnup. Isotopic ratios calculated from analysed data indicate that about one third of the released fission gases were produced in Pu fissions.

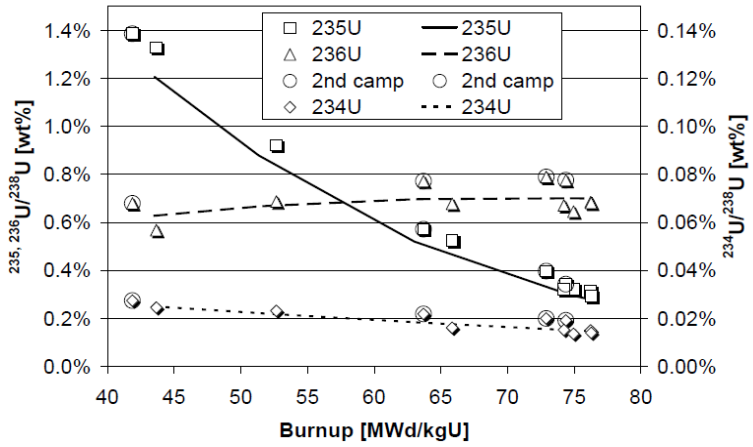
II.6.2 Gamma scanning

Figure 1 shows axial ^{137}Cs activity profile measured by gamma scanning on rods WZtR165 and WZR0058. The shapes of the profiles are typical for PWR rods. Some caesium migration to the colder pellet interfaces has occurred, in particular in the upper part of the rods. Two gaps of about 0.5 mm were found in the fuel column of rod WZtR165 at an elevation of about 3570 mm. A similar gap had formed in rod WZR0058 as well at an elevation of 2390 mm.

II.6.3 Isotope analysis

The content of Uranium isotopes in all samples is plotted in Figure 2 in terms of relative abundance to ^{238}U and as a function of burnup. The amount of ^{233}U was below the detection limit in all cases. Experimental data are represented by symbols that are depicted as a function of the burnup value based on chemical analyses and values calculated by SAS2H for the end of the irradiation are represented by lines. Uncertainties for individual data points were estimated according to the rules of error propagation, taking into account counting statistics as well as uncertainties of spike solution concentrations and isotopic compositions and of the spiking procedure.

Figure 2. Content of uranium isotopes relative to ^{238}U



The results obtained for Plutonium are presented in a similar way in Figure 3. The plutonium abundance values were corrected for the decay of ^{241}Pu and the amount of ^{240}Pu formed through decay of ^{244}Cm since the end of the irradiation and renormalized to 100%. Overall, plutonium data form a consistent set and are well comparable with calculated values.

Figure 3. Content of plutonium isotopes relative to ^{238}U

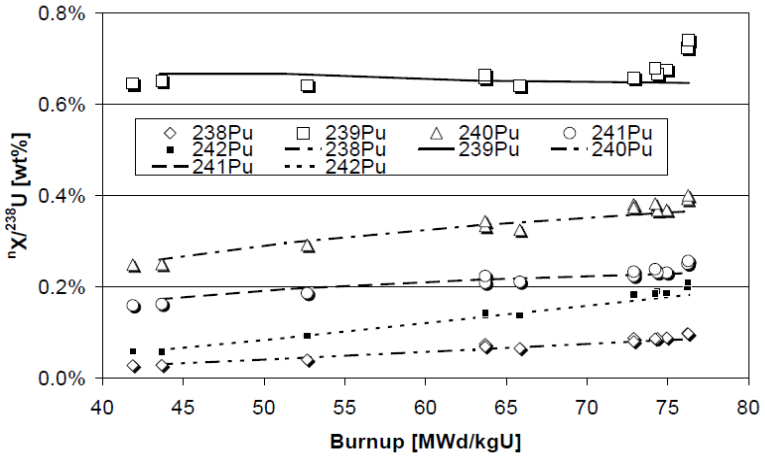
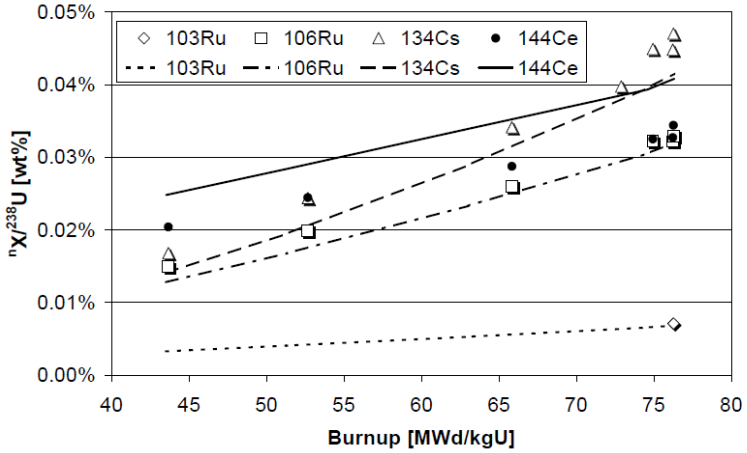


Figure 4. Content of ^{103}Ru, ^{106}Ru, ^{134}Cs and ^{144}Ce isotopes relative to ^{238}U based on γ-scanning



^{144}Ce data are shown in Figure 4. The analysed amount of ^{144}Ce at the end of irradiation is consistent with calculations, although SAS2H slightly over predicts the ^{144}Ce concentration.

^{144}Nd is subject to a decay correction. The amount formed through decay of ^{144}Ce between the end of irradiation and the date of analysis was calculated based on the analysed amount of ^{144}Ce and subtracted from the analysed ^{144}Nd amount. The neodymium data shown in Figure 5 and Figure 6 form a consistent set and are well comparable to the code calculation predictions.

Figure 5. Content of ^{142}Nd, ^{148}Nd and ^{150}Nd isotopes relative to ^{238}U

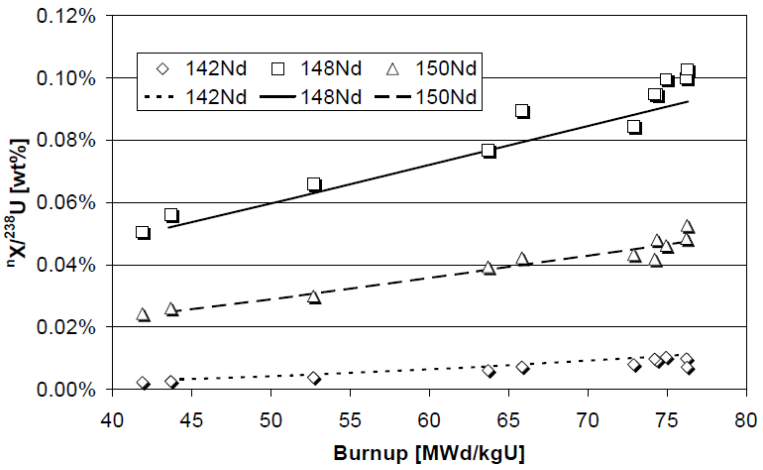
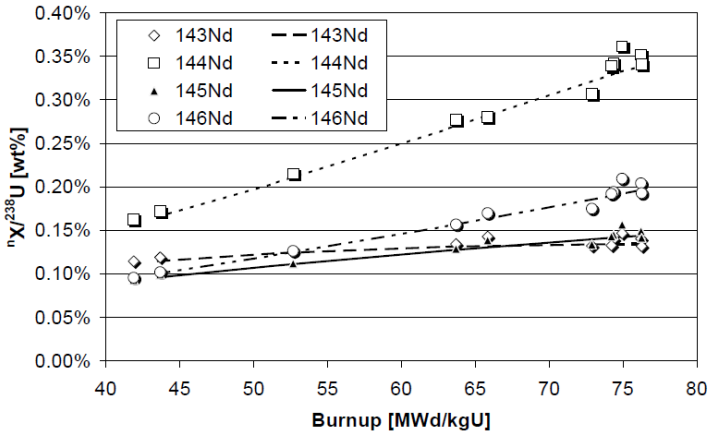


Figure 6. Content of ^{143}Nd, ^{144}Nd, ^{145}Nd and ^{146}Nd isotopes relative to ^{238}U



Samarium data are shown in Figure 7 and Figure 8. A significant amount of the analysed ^{147}Sm, about 40% in the first campaign and 50% in the second campaign, was formed through decay of ^{147}Pm between the end of irradiation and the analysis date. The corresponding correction of the experimental values was based on the amount of ^{147}Pm calculated by CASMO. Results of SAS2H at the end of irradiation agree quite well with the experimental values.

Figure 7. Content of ^{149}Sm, ^{151}Sm, and ^{154}Sm isotopes relative to ^{238}U

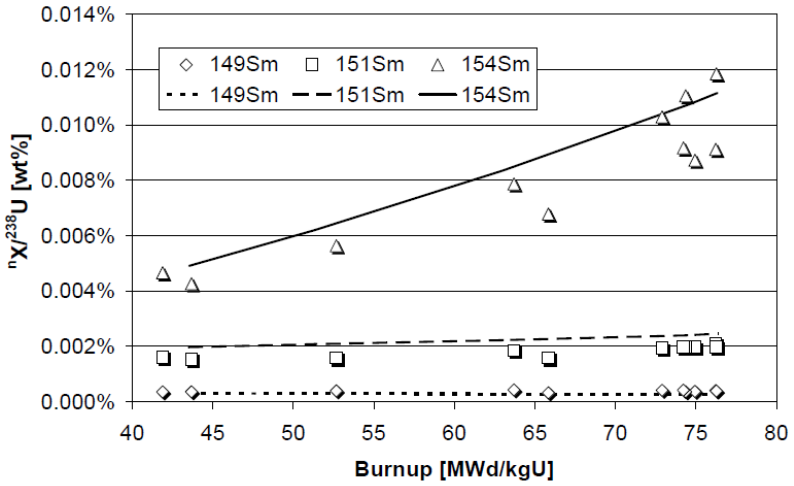
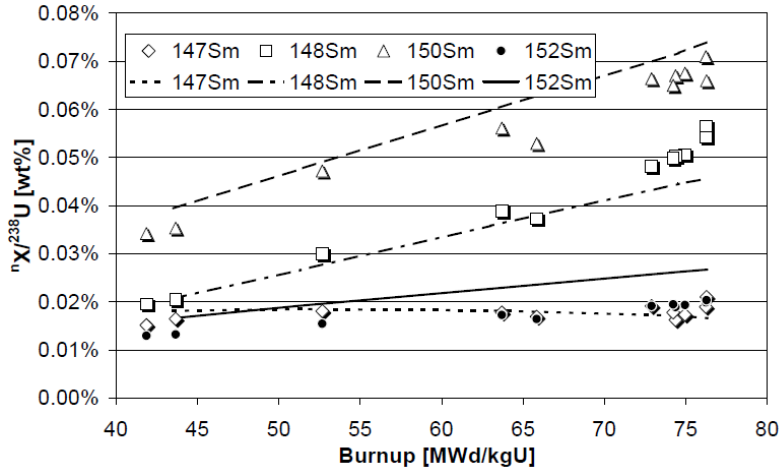


Figure 8. Content of ^{147}Sm, ^{148}Sm, ^{150}Sm and ^{152}Sm isotopes relative to ^{238}U



^{154}Eu was analysed with two independent methods, gamma scanning and IDA. As illustrated in Figure 9 the values are very similar for the two measurement methods and agree well with SA2H calculations.

Figure 9. Content of ^{153}Eu, ^{155}Eu, and ^{154}Eu isotopes relative to ^{238}U

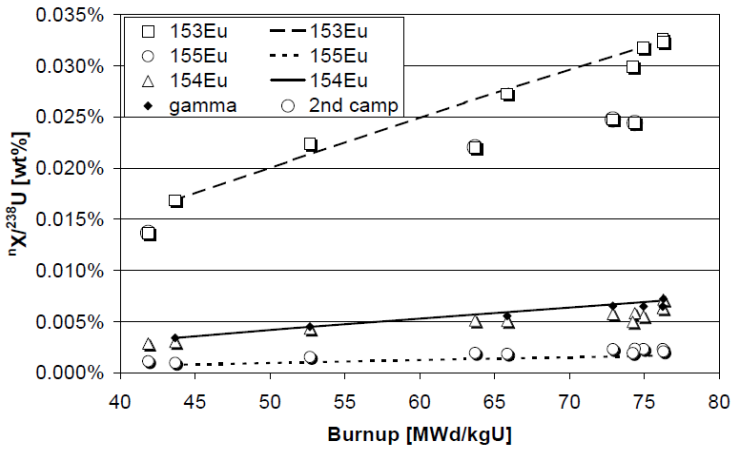
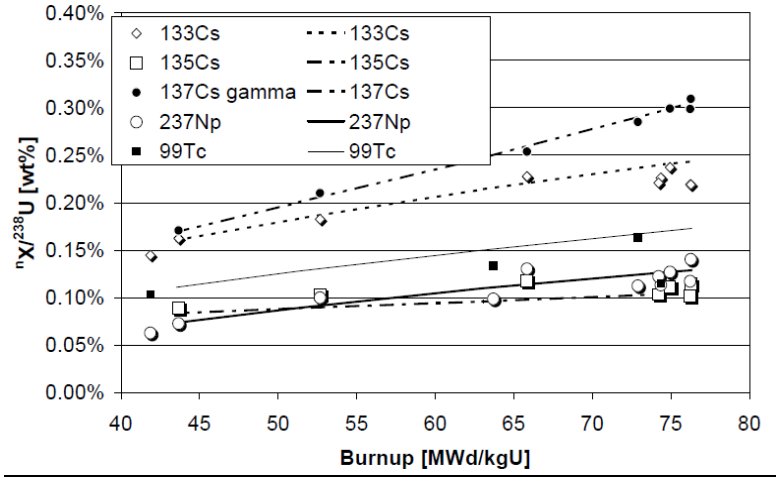


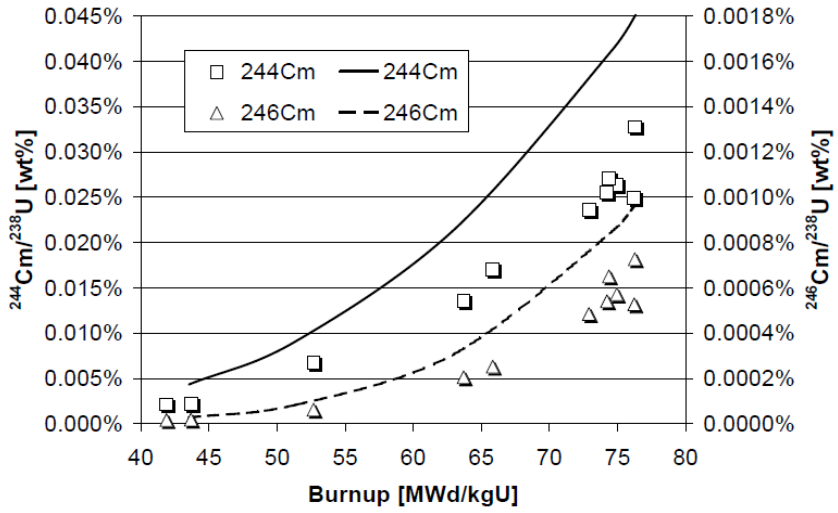
Figure 10. Content of ^{133}Cs, ^{135}Cs, ^{137}Cs, ^{237}Np, and ^{99}Tc isotopes relative to ^{238}U determined by one-point calibration and of ^{137}Cs based on gamma scanning data



Comparison of the analysed ^{99}Tc amounts with SAS2H predictions (Figure 10) shows agreement in two cases and a large difference in one case. At low burnup (sample E58-88), alloy particles seem to be still small enough to be dissolved completely, which is not the case in sample E58-793 (burnup 74.4 MWd/kgU). In sample E58-257 (burnup 64.9 MWd/kgU), alloy particles were not completely dissolved either, but the undissolved portion was probably rather small. Sample WZtR160-800 was dissolved with an alternative method, where alloy particles were completely dissolved.

Experimental curium data show a consistent pattern, compared to the code prediction curves (Figure 11). In view of the number of nuclear reactions involved in curium formation, agreement between experimental and modelling data is acceptable.

Figure 11. Content of ^{244}Cm and ^{246}Cm isotopes relative to ^{238}U determined by one-point calibration



The experimental data for the nuclides analysed by gamma scanning (Figure 4, Figure 10 for ^{137}Cs) give a consistent picture, and deviations from predicted values are not very large.

II.6.3 Burnup

All burnup data are compiled in Table 3. Weight factors proportional to the reciprocal absolute error were used for calculating the different weighted average values. Due to the low error compared to other individual values, the weighted average is somewhat dominated by ^{235}U. The indicated uncertainties do not include any uncertainty of CASMO calculations. Even if individual values are different in the strict sense, considering the indicated (1σ) uncertainties, the overall picture is rather consistent. The fact that ^{235}U analysed in the second campaign might be systematically high, as discussed above, means that the corresponding burnup values might be slightly underestimated. Comparison of results from samples supposed to have the same burnup reveals that the bias is not more than about 3%. The burnup data based on gamma scan and on comparison of experimental results with CASMO presented in Table 3 are very consistent and compare well with the burnup values derived independently from core design data and plant operating records presented in the last column of this table.

Table 3. Experimental burnup values based on γ-scan and on comparison with analysis results with CASMO, compared to values derived from core design data and plant operating records

Sample	Gamma scan value	Burnup [MWd/kgU] based on			Weighted average Nd values	based on abundance of		Overall weighted average	Core Plant & Design data
		$^{146}Nd/^{238}U$	$^{148}Nd/^{238}U$	$^{150}Nd/^{238}U$		^{235}U	^{239}Pu		
E58-88 1st c.	44.5	44.7	46.2	45.8	45.4	42.4	43.0	43.7	43.5
Uncertainty	2.2	0.9	1.1	1.6	0.7	1.3	0.4	0.5	----
E58-88 2nd c.	44.5	42.1	41.6	43.0	42.2	41.3	43.0	41.9	----
Uncertainty	2.2	0.7	0.9	0.8	0.5	0.2	0.7	0.3	----
E58-148	54.3	53.7	54.1	51.7	53.2	51.8	53.2	52.7	51.3
Uncertainty	2.3	1.0	1.4	1.3	0.7	0.5	0.5	0.3	----
E58-257	64.9	64.1	62.4	65.4	63.9	62.8	64.6	63.7	----
Uncertainty	2.8	0.8	1.4	1.6	0.7	0.3	0.3	0.3	----
E58-263	64.9	68.3	72.4	69.2	69.8	64.5	64.8	65.8	63.0
Uncertainty	2.8	1.1	1.4	1.3	0.7	0.6	0.3	0.3	----
E58-773	76.2	80.4	79.9	74.7	78.3	74.5	73.8	74.9	74.5 (*)
Uncertainty	3.3	1.8	2.4	2.0	1.2	0.6	0.5	0.5	----
E58-793 1st c.	76.2	78.8	80.1	77.7	78.9	74.9	74.1	76.2	74.5 (*)
Uncertainty	3.3	2.6	3.5	3.3	1.8	1.4	3.9	1.5	----
E58-793 2nd c.	76.2	75.9	76.1	77.3	76.3	73.3	75.5	74.4	----
Uncertainty	3.3	0.8	1.3	1.8	0.8	0.3	1.3	0.5	----
E58-796	76.2	75.3	76.3	68.7	73.3	74.5	75.1	74.2	74.5 (*)
Uncertainty	3.3	1.4	1.8	1.5	0.9	0.6	1.9	0.7	----
WZtR165-2a	78.2	75.5	82.3	83.3	79.4	76.2	74.3	76.3	76.3
Uncertainty	3.4	1.4	3.2	2.1	1.4	0.7	0.8	0.6	----
WZtR160-800	72.5	70.0	68.5	70.6	69.8	70.3	76.1	72.9	----
Uncertainty	3.1	0.8	1.9	2.1	1.0	0.3	0.3	0.3	----

(*) The burnup from these samples was very similar thus average value was used

III. BWR PROJECT

III.1 BACKGROUND AND SCOPE

As well as in the case of the PWR project, the BWR project is the result of a cooperative spent fuel research effort by Spanish organizations CSN, ENRESA and ENUSA to obtain representative isotopic composition data of irradiated BWR fuel, to be used as a basis for the validation of the composition calculation methodologies (reactivity, decay heat, shielding) used by the participants.

A GE-14 fuel rod manufactured in ENUSA and irradiated for 5 cycles in the BWR Forsmark-3 Swedish reactor was sent to Studsvik's hot cell laboratory for PIE. The material, made available by Vattenfall, was well characterized, with fabrication data, pool-side inspection during irradiation and at discharge data and also some PIE data available. Suitable for research activities, the rod has been involved in research projects related to isotopic composition measurements, cladding behaviour (creep conditions) and others.

An agreement has been reached with Vattenfall for this project: the material is available at the Studsvik hot cell, the design data and irradiation history has been provided and also some reference calculations made. Support by ORNL has been obtained for this isotopic composition project, they are providing technical advice and support as well as financial contribution that has allowed for increasing the project scope. They will also perform validation calculations when results will be available.

In the framework of the project eight fuel samples from the GE-14 rod are being analyzed in order to measure isotopes relevant for reactivity, decay heat and shielding. The experimental work, also performed in Studsvik, includes γ-spectrometry of the rod, chemical analyses of dissolved fuel pellet samples, burnup determination and rod puncturing. Some of the analyses will use experimental work already performed. Analytical work, foreseen to be performed with different codes, will allow evaluation of the experimental results and code validation works.

III.2 SELECTED ISOTOPES

The list of nuclides measured in this project is included in Table 4, as well as the experimental method used for its measurement. The nuclides have been selected on the basis of their relevance for criticality safety, radiation shielding and residual heat calculations [3]. However, some relevant nuclides have been excluded due to practical reasons. Not all listed isotopes have been analysed in all samples.

Table 4. Selected isotopes for BWR project

Element	Atomic mass isotope	Method
U	233 234 235 236 238	IDA/ICP-MS
Pu	238 239 240 241 242	IDA/HPLC-ICP-MS
Np	237	One-point calibration
Am	241 243	IDA/HPLC-ICP-MS
Cm	244 246	One-point calibration
Tc	99	One-point calibration
Ru	103 106	γ-scanning
Cs	133 134 135 137	One-point / γ-scan / One-point / γ-scan
La	139	One-point calibration
Ce	140 142 144	IDA/HPLC-ICP-MS / IDA/HPLC-ICP-MS / γ-scan
Nd	142 143 144 145 146 148 150	IDA/HPLC-ICP-MS
Sm	147 148 149 150 151 152 154	IDA/HPLC-ICP-MS
Eu	153 154 155	IDA/HPLC-ICP-MS / γ-scan / IDA/HPLC-ICP-MS
Gd	154 156 158 160	IDA/HPLC-ICP-MS
Sr	All isotopes	IDA/ICP-MS
I	139	IDA/ICP-MS

III.3 FUEL ROD DESCRIPTION AND SAMPLES

The GE-14 fuel of concern was fabricated by ENUSA and irradiated in the Swedish Forsmark 3 boiling water reactor between July 2000 and May 2005 to a calculated rod average burnup of about 41 MWd/kgU reaching local burnup values of about 53 MWd/kgU. It was part of GE-14 10x10 assembly GN592 and located in the peripheral position J8. The nominal initial enrichment was 3.95 wt% ^{235}U, the rod is axially uniform, without gadolinium, and Zircaloy-2 as cladding material. Its corrosion thickness reach about 20μm and Hydrogen content ~ 175 ppm.
A total of eight samples were cut from the rod for this project and are being analysed. According to the schedule, analytical work will be finished by the end of November. Sample designations, positions and approximate burnup values are compiled in Table 5.

Table 5. Analyzed samples in BWR project

Sample	Position (mm) from rod bottom end	Estimated BU (MWd/tU)	Comments
6	398-408	~42	With sample 5 effect of different void content (lower/uuper)
7	702-712	~50	With sample 3 asses measurements repeatability
3	712.6-722.6	~50	Effect of void content
1	1841.8-1851.8	~50	Asses repeatability of measurements
2	1852.4-1862.4	~50	
4	2503-2513	~53	Highest rod burnup
5	3277-3287	~42	With sample 6 effect of different void content (upper/low)
8	3384-3394	~39	Effect of void content

III.4 EXPERIMENTAL WORK

The rod was investigated in the Studsvik hot cells before starting this project. The examinations included puncturing and fission gas analysis, visual inspection, γ-scanning with burnup evaluation, rotational gamma scan, profilometry, eddy-current oxide thickness measurements, optical microscopy and hydrogen analysis. Afterwards, and within the scope of this project, the rod was further investigated including chemical analyses of dissolved fuel pellet samples and burnup determination. All of these methods are briefly described below.

III.4.1 Rod puncturing

Fission gas release was determined in the rod, by puncturing in the plenum. The internal gas was expanded into a standard volume and the resulting pressure was determined. Samples of the gas were collected for analysis by mass spectrometry. The total internal free volume of the rod was determined by the backfill method, using argon at constant pressure. The fission gas release fraction was calculated from the experimentally determined amount of Xenon and Krypton extracted from the rod and the ORIGEN calculated total inventory of fission gases generated in the fuel over its entire irradiation.

III.4.2 Gamma scanning

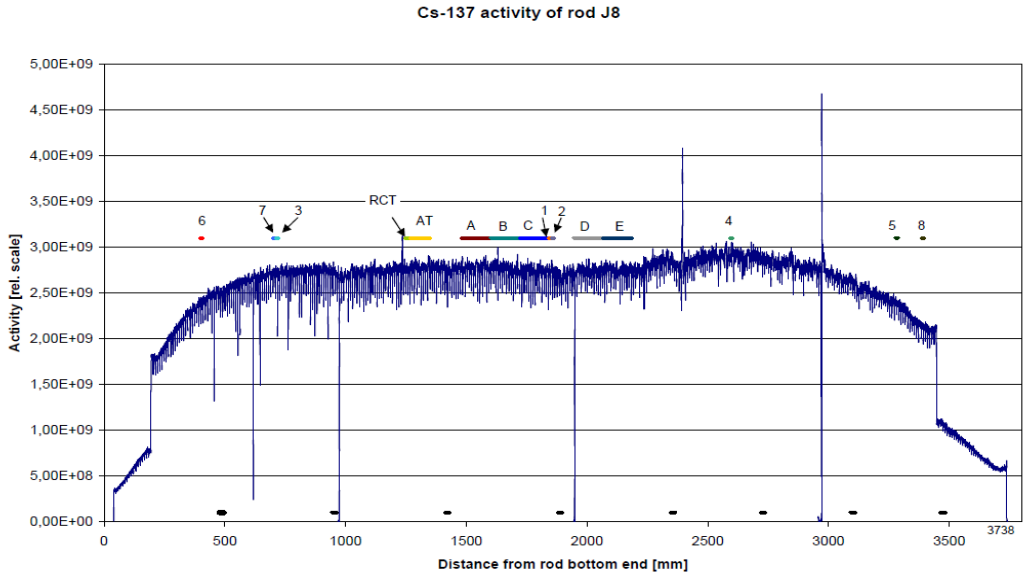
A high purity germanium detector behind a 0.5 mm tungsten collimator was used for the measurements. Axial gamma scanning was performed applying the technique of closely spaced point measurements. The efficiency file for the detector and collimator system was calibrated to give photon energy independent activity values for a fuel rod with an outer diameter of 9.5 mm with no extra absorber. Activities were decay corrected to the date May 28, 2005, corresponding to the end of irradiation. A well characterised reference rod (F3F6) was scanned together with the first segment. By comparing the apparent ^{137}Cs activity measured for the reference rod with the decay corrected ^{137}Cs activity known from the characterisation of the rod, a correction factor to be applied on all apparent activities was determined. The slightly larger diameter of the GE14 rod, compared to F3F6 rod, was

taken into account by a correction factor determined on the basis of calculated escape probabilities for the gamma energies of concern. Dead time correction of the system was checked by following the signal from a ^{60}Co source placed close to the detector. No extra correction was necessary. The absolute activity was determined.

The GE14 rod had been scanned in February/March 2006. ^{103}Ru, ^{106}Ru/Rh, ^{125}Sb, ^{134}Cs, ^{137}Cs and ^{144}Ce were measured and could be assessed. The poor quality of ^{154}Eu data did not allow an evaluation. The gamma scan results from the peak burnup region were transformed into amount of nuclide ^{n}X in weight percent relative to ^{238}U according to usual Studsvik procedure, the same applied in PWR project and described in II.4.2.

Figure 12 shows the position and designation of selected samples over the ^{137}Cs γ-scan plot of the GE14 rod. Samples labelled with a number (from 1 to 8) are the samples selected for the isotopic composition project.

Figure 12. Position and designation of selected samples from GE14 rod for chemical analyses



III.4.3 Radiochemical analysis

It is being performed also by Studsvik, and the same methods and instruments of 2nd campaign of PWR project are basically applied: dissolution and dilution of the fuel samples and subsequent Isotopic Dilution Analysis (IDA) with different techniques (ICP-MS analysis based one or two-point calibration, HPLC-MS). Results are reported as ^{n}X/ ^{238}U for each isotope analysed and certified reference standards are used.

III.4.4 Burnup determination

The same method described in II.4.4 for PWR project will be applied [5]: the experimentally determined inventory of major actinides (U and Pu) and some fission products (Ce and Nd) will be compared to the results of CASMO calculations. Therefore the uncertainty of sample specific CASMO calculations performed by Vattenfall Nuclear Fuel, based on detailed values for the same operating conditions, is assumed to be smaller than experimental uncertainties.

III.5 ANALYTICAL WORK

This work will be essential to verify the quality of the experimental work allowing to verify consistency of the experimental isotopic composition data as well as calculated burnup results based on relative abundances of ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{235}U and ^{239}Pu experimentally determined, by comparison with calculation results for expected isotopic compositions.

Assessment of measured nuclide inventories through the performance of code validation calculations by different participants and codes (SCALE/SAS-2H, SCALE/TRITON, CASMO-SIMULATE) is foreseen. A final workshop for results benchmarking will be held.

III.6 SCHEDULE

The project started in February 2009 with the preparation of the samples. At present cutting and dissolution of the samples has been finished and also analysis of mother solution, completed by June. Analysis of iodine and strontium were completed by September and metallic particles and molybdenum are ongoing. Experimental phase is scheduled to be finished by November and the draft final report, with evaluation of results will be completed by the end of 2009.

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