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PWR AND BWR FUEL ASSAY DATA MEASUREMENTS

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Outline

- Overview on isotopic composition of spent fuel research in Spain
- PWR project
  - Description: scope, participants, fuel, samples, isotopes
  - Results & conclusions
- BWR project description
  - Description: scope, participants, fuel, samples, isotopes, experimental and analytical works
  - Schedule

Objective

- To obtain representative isotopic composition data of PWR & BWR irradiated fuel, to be used as a basis for the validation of the composition calculation methodologies (reactivity, decay heat, shielding) used by the participants.

- A cooperative spent fuel research effort was started in 2003:
  - CSN
  - ENRESA
  - ENUSA

BUC applications in Spain

- BUC is applied in Spain in criticality safety analysis of:
  - all the spent fuel pools: full BUC level is used for PWR storage, while burnable absorber BUC is applied in BWR pools.
  - spent fuel cask for dual purpose storage/transportation recently licensed (Holtec design: sealed canister containing the spent fuel, can be used for either storage or transport depending on the type of outer structure in which the canister is inserted):
    - Does not need to apply BUC techniques during the canister loading/ unloading operations, due to the presence of Boron in the pool water.
    - Does not need to apply BUC techniques for dry storage conditions
    - Assumption of cask flooding with pure water required by the transport regulations leads to the use of BUC for these conditions. Following the US regulations (ISG-8), the Actinide-only BUC level has been used.

Projects data

- PWR project
  - 17x17 fuel Vandellos II, 4,5% U^{235}, BU 70 MWd/kgU
  - 9 samples, 3 rods, 40 MWd/kgU<BU<75 MWd/kgU
  - 2 campaigns: 2003, 2006/07
  - Studsvik lab: FGR, γ-scan, chemical analysis, BU analysis
  - More than 50 isotopes of 16 different elements
  - Concluded, results will join SFCOMPO NEA Database

- BWR project
  - GE-14 10x10 fuel Forsmark, 3.95% U^{235}, BU 41 MWd/kgU
  - 8 samples, 1 rod, 39 MWd/kgU<BU<53 MWd/kgU
  - 1 campaign: 2009
  - Studsvik lab: FGR, γ-scan, chemical analysis, BU analysis
  - More than 50 isotopes of 18 different elements
  - On-going: end scheduled 2010

PWR FUEL PROJECT

PWR Project: Background

- Irradiation extension program:
  - Performed at the Vandellós II reactor
  - Fuel rods irradiated up to 70 MWd/kgU (5 cycles)
  - Transported to Studsvik's hot cell laboratory for post-irradiation examination (PIE)
- Valuable material for safety research:
  - Fully characterised
  - High enrichment and high burnup
  - Already included in CABRI, ALPS, SCIP…
- Measurements of the isotopic composition of fuel pellets from three of these rods have been performed

PWR: Scope

- The isotopic composition of nine samples from 3 rods has been analyzed, with burnups ranging from 40 to 75 MWd/kgU
- Isotopes to be measured have been selected on the basis of the nuclide importance for reactivity, decay heat, shielding and BU calculations.
- Experimental work includes γ-spectrometry of the rod, chemical analyses of dissolved fuel pellet samples, burnup determination and rod puncturing. Some of the analyses have used experimental work already performed
- Analytical work: evaluation of the experimental results and code validation
- Experimental work performed in Studsvik

PWR: samples position

- Samples selected for the first campaign

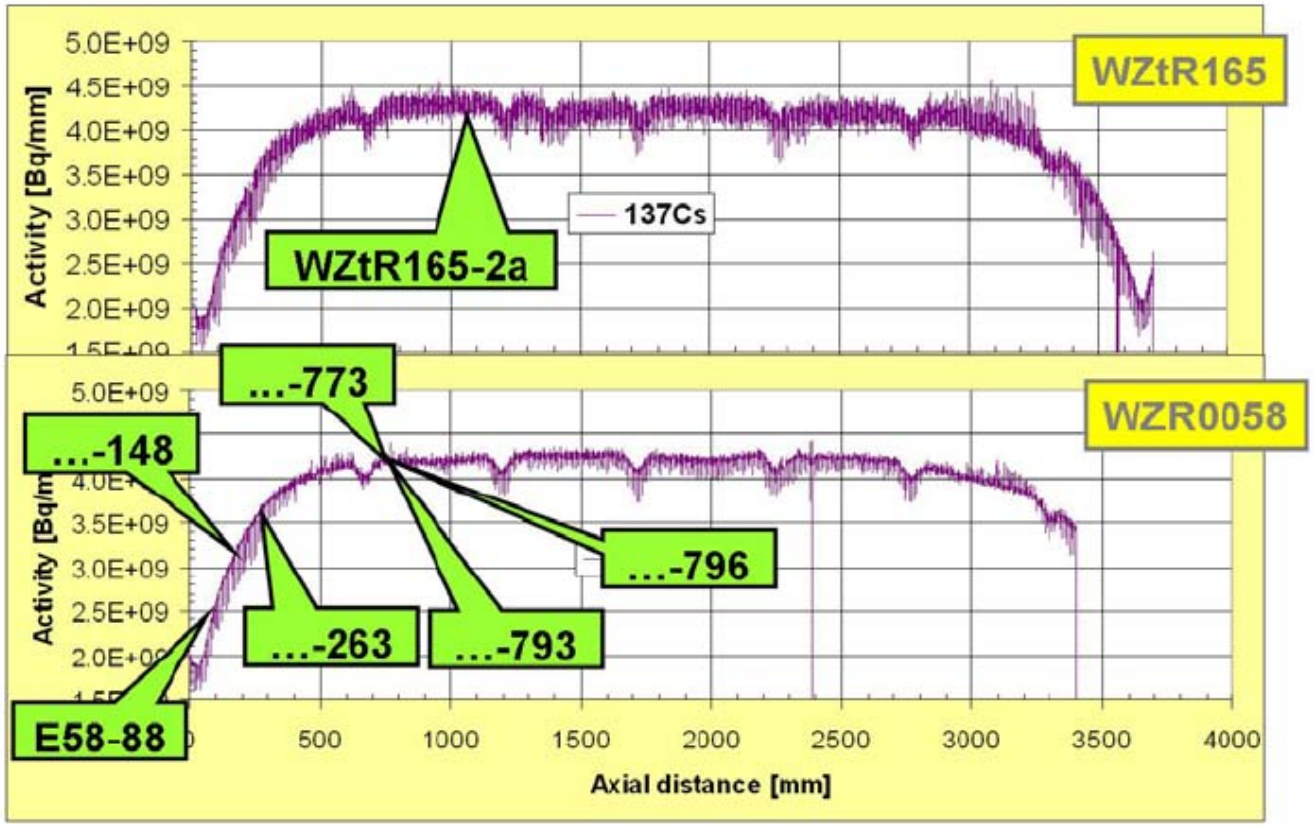


Figure 1 **Position and designation of samples selected for radio-chemical analyses**

PWR: Analyzed samples

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

PWR: Selected isotopes

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

PWR: experimental work (1)

- γ-spectrometry:
  - Axial scanning corrected for dead time losses and decay of the different nuclides since the end of the irradiation
  - Steps between 0.2 and 0.5 mm along the whole length of the rod
  - Additional corrections to account for the γ attenuation, escape probability and detector efficiency in order to calculate the radionuclide inventory in Bq/mm
- Chemical analysis:
  - Sample dissolution
  - Isotopic Dilution Analysis by ICP-MS (Inductively Coupled Plasma Mass Spectrometer)
    - Without separation
    - With separation by HPLC (High Performance Liquid Chromatography)
  - Results are reported as $^{n}X/\ ^{238}U$ for each isotope analysed.

PWR: experimental work (2)

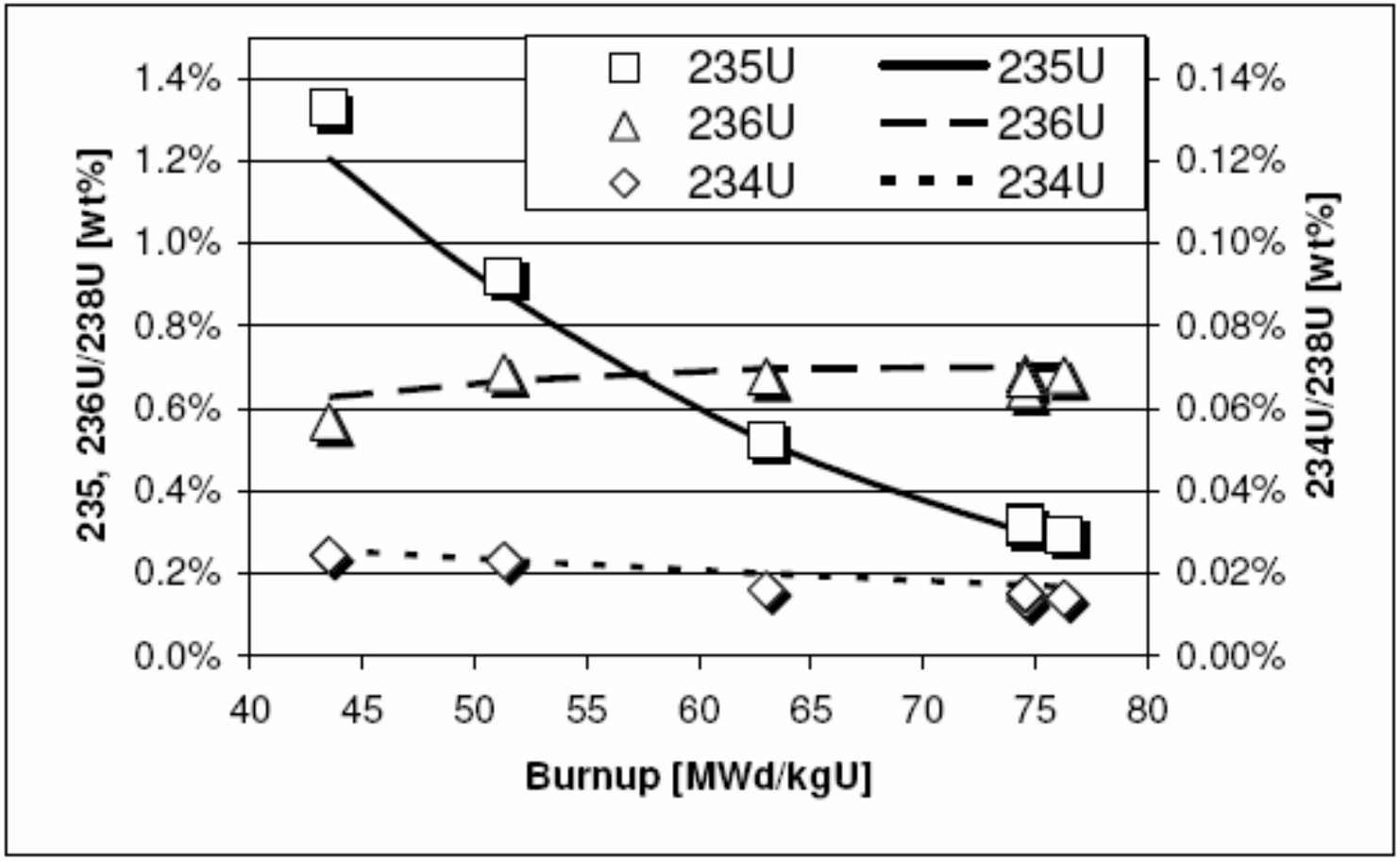
- Burnup determination:
  - Experimental isotopic abundances of uranium and plutonium composition as well as some fission products (Nd) measured by chemical analyses.
  - Comparison with corresponding CASMO based data for the same operating conditions.
  - Results consistent with values derived independently from γ-scanning and from core design data and plant operating records

- Analysis of released fission gases
  - Isotopic composition of the released fission gas is determined by mass spectrometry of samples of gas retrieved from puncturing of the rod plenum

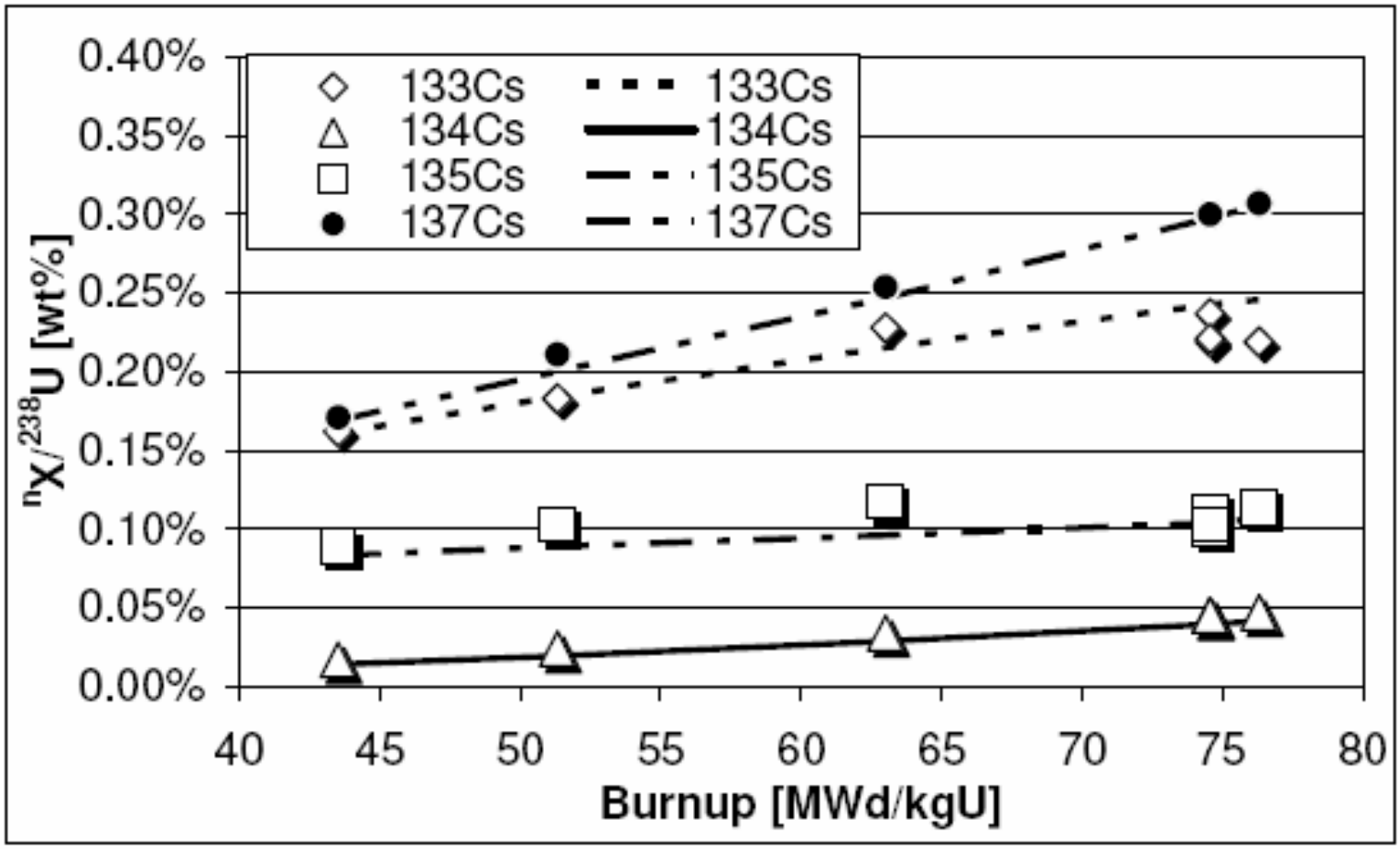
PWR: analytical work

- ENUSA has been for analytical activities:
  - Evaluation of the experimental results by comparison with analytical results of reactor irradiation obtained with the SCALE system: calculation of the expected isotopic composition of each sample and comparison with the measured concentrations, focusing in the Actinides
  - Assessment of isotopic abundances using the industry standard SAS2H of the SCALE code system, for each sample, with good agreement between measured and calculated values.

PWR: Results (I) Uranium abundance



PWR: Results (II) Caesium abundance



PWR: Results (III) burnup

Sample	Gamma scan value	Burnup [MWd/kgU] based on $^{146}Nd/^{238}U$	$^{148}Nd/^{238}U$	$^{150}Nd/^{238}U$	Weighted average Nd values	based on abundance of ^{235}U	^{239}Pu	Overall weighted average	Core design & plant data
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

PWR: Summary

- Lack of experimental data on the isotopic composition of irradiated commercial fuel
- Irradiation research programs have provided high-enrichment, high-burnup PWR fuel (well characterized, correct behaviour under normal operating conditions..)
- Nine samples from three rods and more than 50 nuclides experimentally analyzed.
- Over 400 data points giving in general a consistent picture of the isotopic content of the irradiated fuel as a function of burnup.
- SAS2H results for high-burnup samples are not subject to higher uncertainty and/or other biases than lower burnup samples.
- Isotopic experimental measuring methods provide accurate results with acceptable precision.
- Results joining soon SFCOMPO NEA database.

BWR FUEL PROJECT

BWR: Background

- A GE-14 fuel rod manufactured in ENUSA, irradiated for 5 cycles in the BWR Forsmark 3 swedish reactor was sent to Studsvik's hot cell laboratory for PIE:
  - The rod has been made available by Vattenfall
  - Well characterized, in connection with the PIE programme:
    - Fabrication data available
    - Pool-side inspected during irradiation and at discharge
    - Some PIE data already available
- Material suitable for research activities:
  - Isotopic composition measurements
  - Cladding behaviour (creep)
  - Other projects

BWR: Participants

- An agreement has been reached with Vattenfall for this project:
  - The material is available at the hot cell
  - The design data and irradiation history has been provided
  - Reference calculations made
- Support by ORNL has been obtained for this project:
  - Technical advice and support
  - Financial contribution that has allowed for increasing the project scope
  - Validation calculations

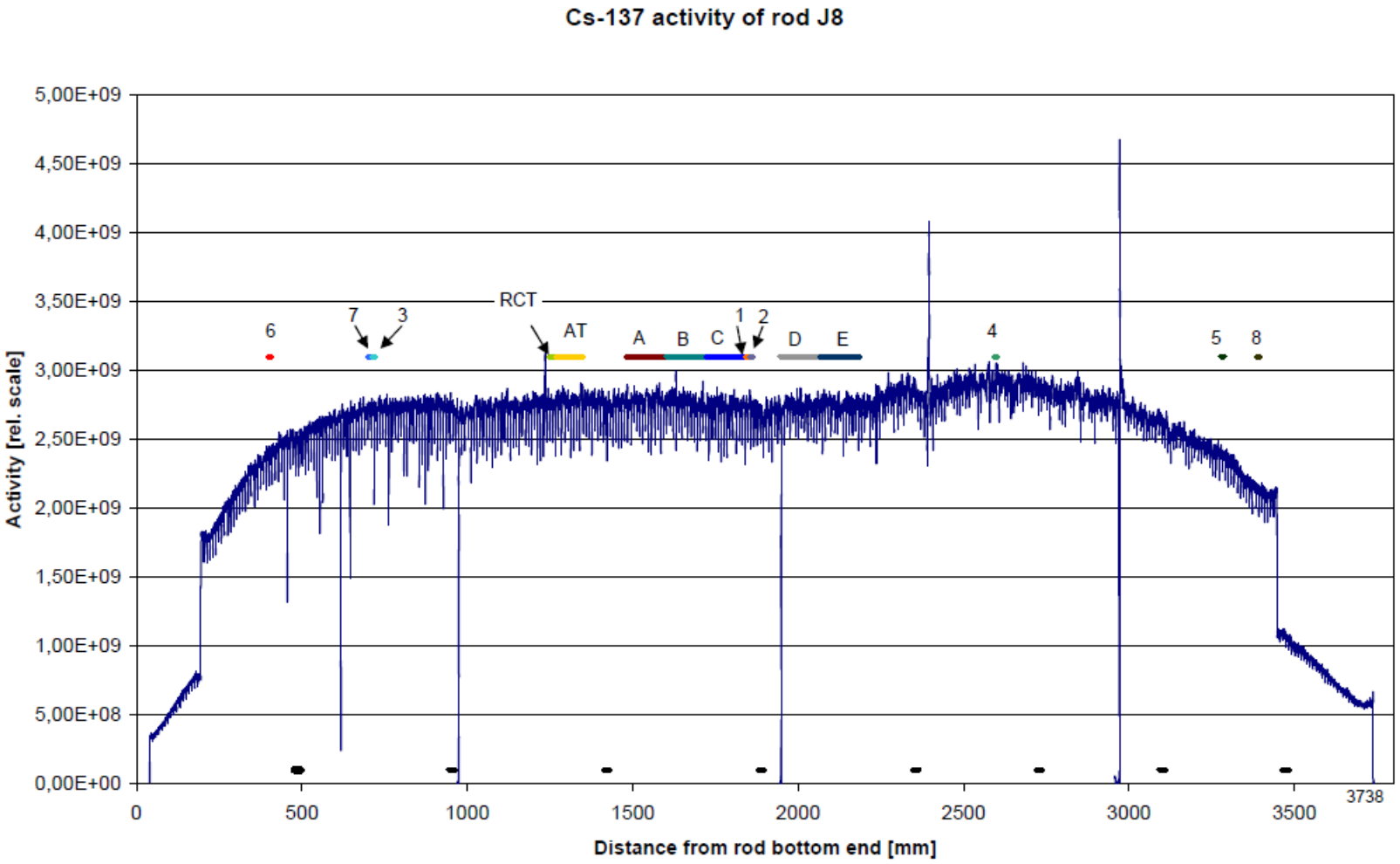
BWR: Scope

- 8 fuel samples will be analyzed
- Isotopes relevant for reactivity, decay heat and shielding
- Experimental work 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 with different codes: evaluation of the experimental results and code validation
- Measurements performed in Studsvik

BWR: Fuel rod description

- BWR GE-14 10x10 fuel design
- 3.95% enrichment
- Axially uniform (no Gd)
- Zircaloy-2 cladding
- Average burnup ~ 41 MWd/kgU
- Local burnup ~ 53 MWd/kgU
- Corrosion thickness ~ 20µm
- Hydrogen content ~ 175 ppm

BWR: Sample positions



BWR: Analyzed samples

Sample	Position (mm) from rod bottom end	Estimated BU (Mwd/kgU)	Comments
6	398-408	~42	With sample 5 effect of different void content (lower/upper)
7	702-712	~50	With sample 3 assess repeatability of measurement
3	712.6-722.6	~50	Effect of void content
1	1841.8-1851.8	~50	Assess 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/lower)
8	3384-3394	~39	Effect of void content

BWR: Isotopes

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
Mo	95 97 98 100	IDA/HPLC-ICP-MS
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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 / γ-scan / IDA/HPLC-ICP-MS
Gd	154 155 156 157 158 160	IDA/HPLC-ICP-MS
Sr	All isotopes	IDA/ICP-MS
I	129	IDA/ICP-MS

BWR: Experimental work (1)

- γ-spectrometry:
  - Measurements have already been performed in previous hot cell examinations (axial scan of the full rod length, corrections by γ attenuation, escape probability and detector efficiency).
  - Data evaluation

- Chemical analysis:
  - Sample dissolution
  - Isotopic Dilution Analysis by ICP-MS (Inductively Coupled Plasma Mass Spectrometer)
    - Without separation
    - With separation by HPLC (High Performance Liquid Chromatography)
  - Results are reported as $^{n}X/\ ^{238}U$ for each isotope analysed.

BWR: Experimental work (2)

- Burnup determination:
  - Inventory of major heavy metal nuclides (U and Pu) and some fission products (Ce and Nd)
  - Comparison with CASMO calculations based on detailed values for the same operating conditions

- Released fission gases:
  - Rod puncturing at the plenum
  - Mass spectroscopy of gas samples (Xe, Kr)

BWR: Analytical work

- Essential to verify the quality of the experimental work:
  - Consistency of the experimental and calculated results
  - Relative abundances of Nd-146,-148,-150, U-235 and Pu-239 compared
- Code validation calculations to be performed by the participants:
  - SCALE/SAS-2H
  - SCALE/TRITON
  - CASMO-SIMULATE
- Final workshop for results benchmarking

Schedule

- Preparation of the samples started in February 2009
- Cutting and dissolution of the samples was completed
- Analysis of mother solution was completed by June
- Analysis of I and Sr were completed by September
- Analysis of metallic particles and Mo are ongoing
- Experimental phase scheduled to be finished by November
- Results evaluation is ongoing and draft final report will be completed by the end of the year


CSN