Interpretation of In-Pile Oscillation Experiments of Fission Product in Various Spectra

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Outline

- Experimental programme
- MINERVE reactor and experimental set-up
- Interpretation scheme
- Calibration measured/calculated signal
- Interpretation
- Conclusions and prospects

Experimental programme

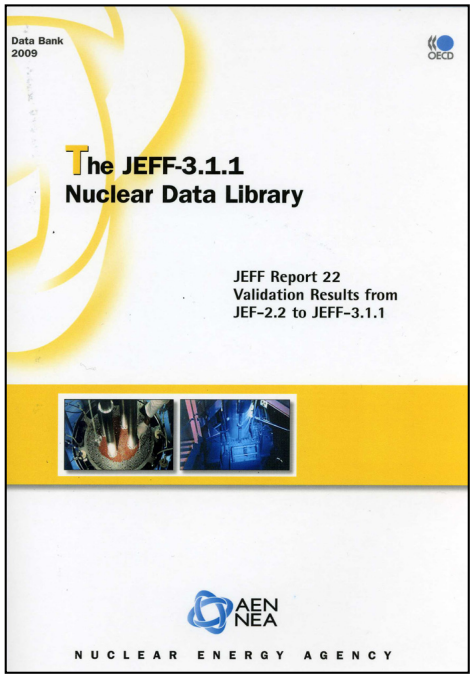
- Objective: qualification of the nuclear data of the most important fission products (FP) in PWR, responsible of 75% of the FP poisoning:

$^{147,149,152}Sm$, $^{143,145}Nd$, ^{155}Gd, ^{153}Eu, ^{103}Rh, ^{99}Tc, ^{133}Cs, ^{109}Ag, ^{NAT}Ru ,^{95}Mo

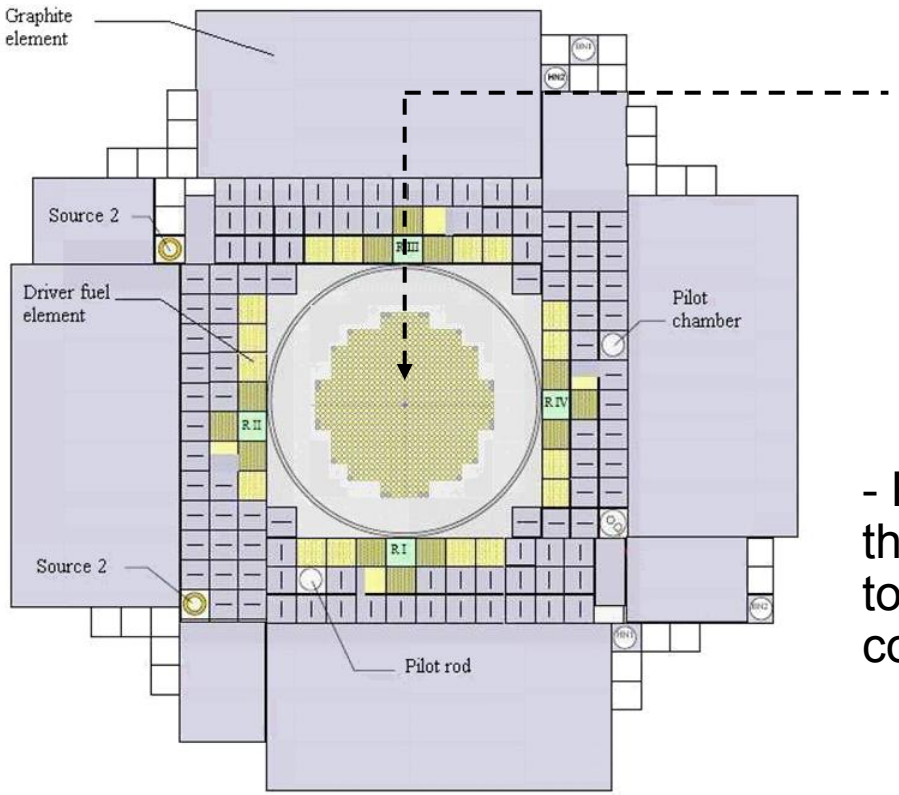
- JEFF3.1.1 library used in this study includes feedback from previous interpretations with JEF2.2 of FP oscillation in MINERVE

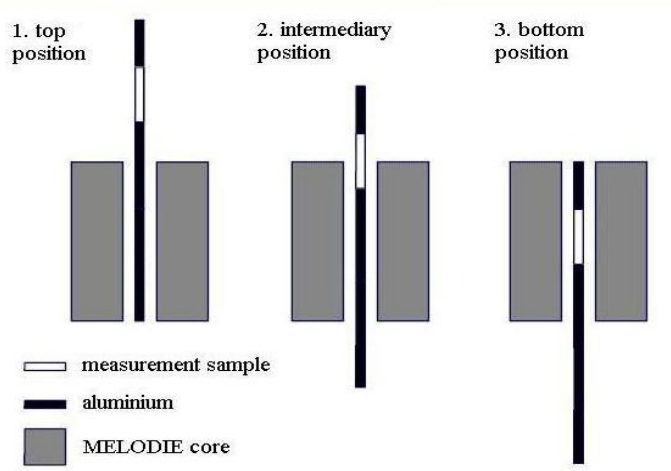
- 4 different lattices → 4 different spectrum

PWR-UO_2	(R1UO2 lattice)
PWR-MOx	(R1MOX lattice)
BWR	(REB lattice)
thermal spectrum	(R2UO2 lattice)

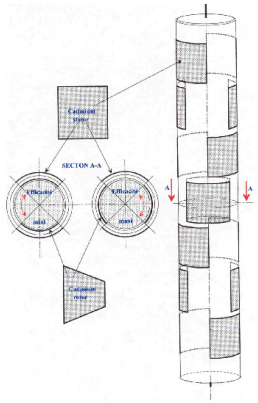


Oscillation Technique in MINERVE (1/2)





- Periodic insertion of a sample in the center of the MINERVE reactor, to measure its reactivity worth in comparison to a reference one

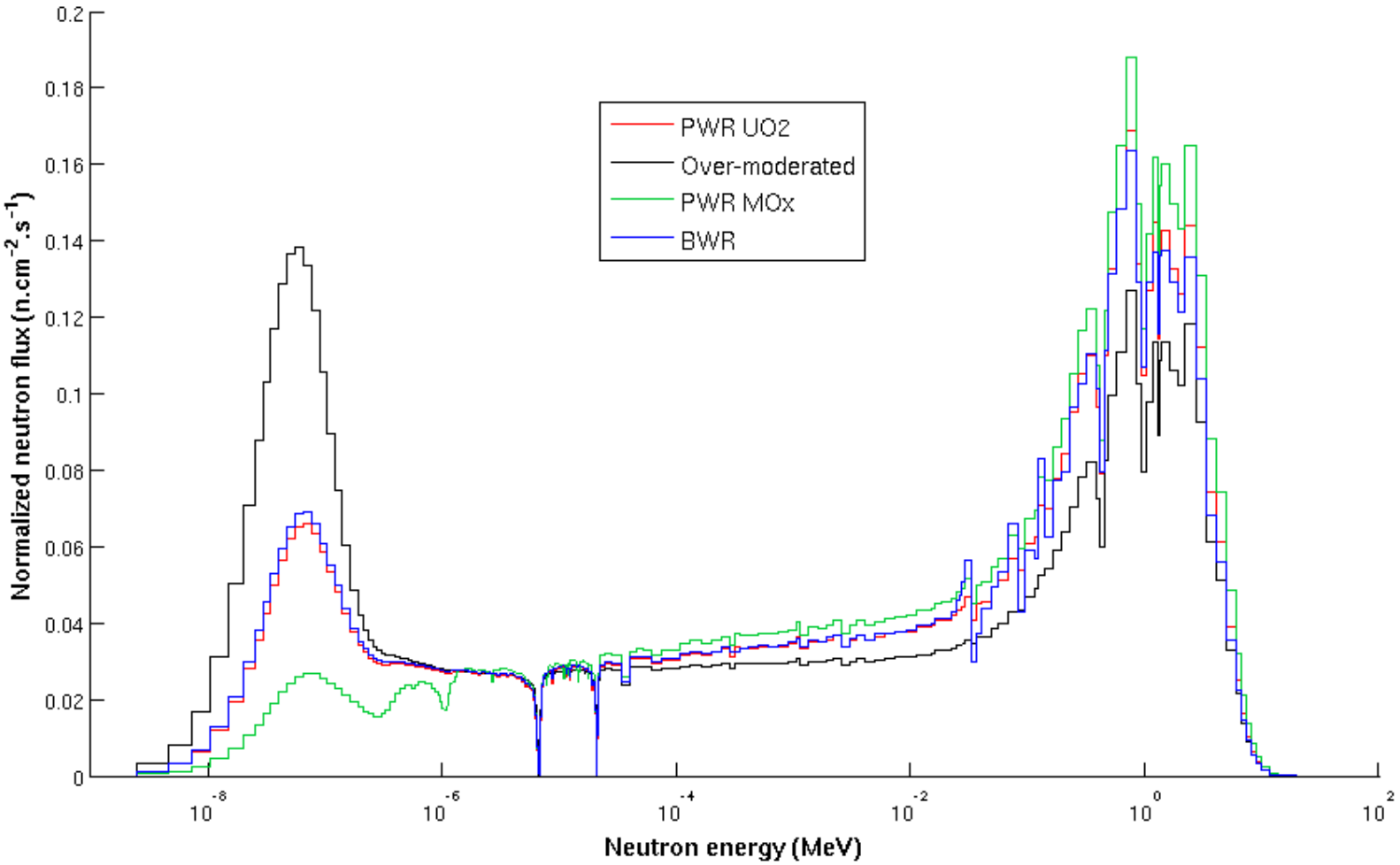


- Variation of the neutron flux detected by a boron chamber, servo-driven to an automatic pilot rod

- The automatic pilot rod compensates the reactivity variation by the overlap of cadmium sectors

Oscillation Technique in MINERVE (2/2)

Different lattices can be loaded in the center of the experimental zone, to obtain different types of neutron spectra



Calculation scheme

- **Interpretation performed using the APOLLO2.8 code package including:**

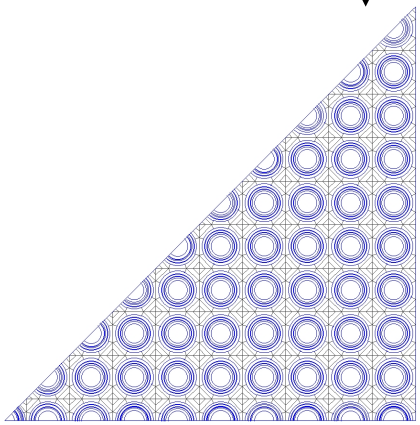
  - CEA2005V4.1.1 library based on the JEFF3.1.1 evaluations
  - 2D-MOC calculation on a 19x19 lattice
  - 281-group energy mesh SHEM
  - Recommendations from the reference *SHEM-MOC* scheme for LWR applications

- Calculation of the reactivity worth of a sample by the **Exact Perturbation Theory**

$$\Delta\rho = \frac{\left\langle \Psi_1^+, \Delta H \Psi_2 \right\rangle}{\left\langle \Psi_1^+, P_2 \Psi_2 \right\rangle}$$

1: reference case (sample with the matrix only)

2: perturbated case (sample with studied fission product)



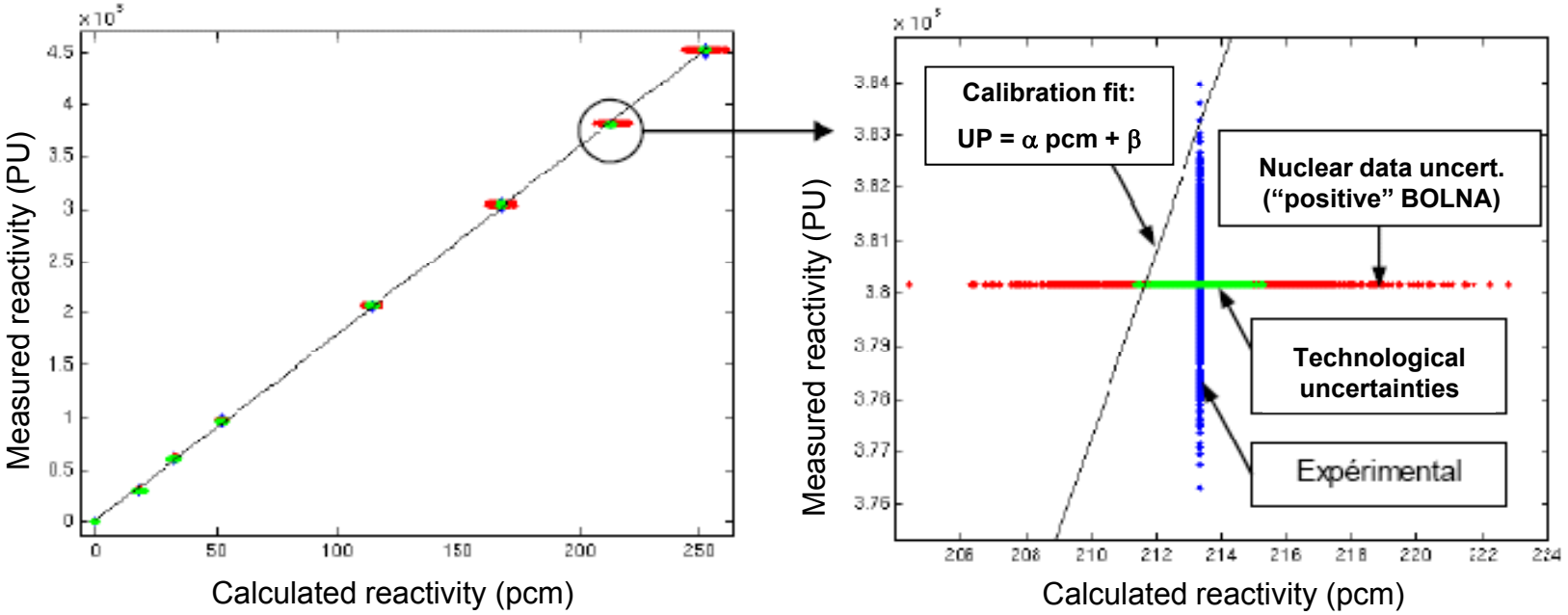
$$\Delta H = H_{matrix+FP} - H_{matrix} = H_{FP} = \Sigma_{FP(n,\gamma)}$$

Deterministic calculation validated against stochastic continuous energy code TRIPOLI4

Calibration measured/calculated signal

Determination of the calibration factor using ^{235}U and ^{10}B samples

$$\Delta S = \alpha \Delta \rho$$



Uncertainties propagation using a Monte Carlo method

- nuclear data
- technological uncertainties
- measurement uncertainties

Uncertainties

Overall uncertainties take into account:

- the material balance of the samples (chemical analysis)
- technological uncertainties (diameter of the clad and the sample)

} up to 2.5%

- the calibration factor (1.5%)
- the measurement uncertainties (about 1.5%, up to 10% on some samples)

uncertainties listed in the next slides

Interpretation, JEFF3.1.1 library

Samarium isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{149}Sm	**-3.7%**	1.2%	**-1.2%**	1.1%	**-0.2%**	1.1%	**-1.0%**	3.1%
^{147}Sm			**4.3%**	0.9%	**6.6%**	0.8%	**6.0%**	2.5%
^{152}Sm	**-3.6%**	1.2%	**-0.8%**	1.1%	**1.6%**	1.1%	**4.3%**	2.8%
^{NAT}Sm	**-3.4%**	1.2%	**-1.8%**	1.1%	**0.2%**	1.1%	**3.6%**	3.2%

→ **-1.6% ± 1.9%**

- **^{149}Sm and ^{152}Sm are well-predicted**

- Overestimation of the ^{147}Sm sample, but 70% of its reactivity effect is due to residual ^{149}Sm. Removing ^{149}Sm effect from ^{147}Sm sample ⇒ **overestimation of 38% ± 29% of the ^{147}Sm effect** in the PWR spectrum

- Increasing trends on the ^{152}Sm and ^{NAT}Sm samples *C/E*-1 with the spectrum hardness

- ^{NAT}Sm *C/E-1* consistent with ^{149}Sm (>95% of its reactivity effect)

Interpretation, JEFF3.1.1 library

Neodymium isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{143}Nd	**-4.7%**	1.7%	**-2.9%**	1.2%	**-2.2%**	1.2%	**4.4%**	3.6%
^{145}Nd	**-0.8%**	1.4%	**7.2%**	1.4%	**1.9%**	1.3%	**15.2%**	3.5%
^{NAT}Nd	**-2.5%**	1.3%	**-2.3%**	1.3%	**-1.1%**	1.2%	**1.2%**	3.5%

- Increase of the *C/E-1* ratios with spectrum hardness
- ^{NAT}Nd sample reactivity effect: 77% ^{143}Nd, 14% ^{145}Nd, *C/E* discrepancy consistent with ^{143}Nd and ^{145}Nd samples
- Trend to **overestimate the ^{145}Nd in the epithermal range**
- **^{143}Nd quite satisfactory**: *C* consistent with *E* within 2*σ* uncertainty

Interpretation, JEFF3.1.1 library

^{133}Cs isotope

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{133}Cs-1	**1.4%**	1.7%	**8.2%**	1.5%	**8.4%**	4.0%	**14.5%**	3.5%
^{133}Cs-2	**-15.0%**	5.4%	**-8.8%**	4.4%	**-6.5%**	5.8%	**-4.6%**	8.1%
^{133}Cs-3	**3.0%**	0.7%	**8.5%**	1.4%	**7.6%**	1.0%	**12.7%**	3.3%
^{133}Cs-4	**4.8%**	3.9%	**0.5%**	5.7%	**4.4%**	6.7%	**38.4%**	13.9%

- Discrepancies increase with spectrum hardness (~12%)

- High uncertainties on the material balance of the samples ^{133}Cs-2 and ^{133}Cs-4 (small amount of doping isotope), and also high experimental uncertainties (small reactivity effects)

- ^{133}Cs-1 and ^{133}Cs-3 discrepancies are consistent → **possible overestimation of ^{133}Cs in the epithermal range**

Interpretation, JEFF3.1.1 library

Silver isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{109}Ag-1	**-3.4%**	1.5%	**4.1%**	1.3%	**5%**	4%	**6.9%**	3.1%
^{109}Ag-2	**-6.9%**	7.0%	**-5.2%**	5.3%	**-0.7%**	10.1%	**-10.5%**	8.8%
^{NAT}Ag	**-0.2%**	0.6%	**7.3%**	1.2%	**4.2%**	1.3%	**13.8%**	3.0%

- Experimental uncertainties of ^{109}Ag-2 up to 10% (small reactivity effects)
- ^{NAT}Ag: 76% of reactivity effect due to ^{109}Ag.
- Consistency between ^{NAT}Ag and ^{109}Ag-1
- ^{109}Ag well predicted in thermal range and possible overestimation in the epithermal range

Interpretation, JEFF3.1.1 library

Molybdenum isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{95}Mo	**-2.1%**	1.9%			**1.1%**	2.0%		
^{NAT}Mo	**-4.7%**	0.8%	**-2.0%**	1.5%	**-3.9%**	1.5%	**-0.8%**	3.3%

- ^{NAT}Mo effect decomposed between 4 isotopes: ^{95}Mo (75%), and ^{96,97,98}Mo (23%)
- Relatively good consistency between ^{95}Mo and ^{NAT}Mo
- **^{95}Mo effect well predicted**

Interpretation, JEFF3.1.1 library

^{155}Gd, ^{153}Eu and ^{99}Tc isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{155}Gd	**-3.7%**	2.0%	**0.1%**	1.2%	**0.2%**	1.2%	**-0.2%**	3.4%
^{153}Eu	**-14.7%**	1.2%	**-11.3%**	1.1%	**-11.0%**	1.3%	**-6.6%**	2.7%
^{99}Tc	**-2.3%**	1.7%	**2.9%**	1.5%	**8.9%**	2.5%	**8.1%**	3.4%

→ **-0.4% ± 2.2%**

- **^{155}Gd well calculated**
- **Underestimation of the ^{153}Eu** (about 11 % in PWR spectrum)
- Increase of the ^{99}Tc discrepancies with spectrum hardness, trend to overestimate its reactivity effect

Interpretation, JEFF3.1.1 library

^{103}Rh and Ruthenium isotopes

δE/E are only measurement uncertainties

	R2UO2		REB		R1UO2		R1MOX	
	C/E-1	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*	*C/E-1*	*δE/E*
^{103}Rh-1	**-0.4%**	1.5%	**5.1%**	1.2%	**6.4%**	1.1%	**9.0%**	3.1%
^{103}Rh-2	**-0.4%**	1.6%	**7.2%**	1.4%	**12.8%**	1.9%	**10.8%**	3.5%
^{NAT}Ru	**8.1%**	1.5%	**13.1%**	2.2%	**19.0%**	2.3%	**24.0%**	4.3%

- ^{103}Rh samples are consistent (within exp. uncertainties)

- Thermal range well calculated, improvements still needed in the resonance range

- A part of the discrepancy of the ^{103}Rh could be linked to the double heterogeneity (unknown grain size) of the samples, trend to overestimate its absorption rate (~10 µm → -1%)

- **Overestimation of the ^{NAT}Ru sample**: 2 main isotopic components: ^{99}Ru (45% of the reactivity effect), ^{101}Ru (35%)
  → need of samples with separated isotopes

- Discrepancies increase with spectrum hardness

DIMPLE/MINERVE comparison

Results from LWR spectrum (mean of REB & R1UO2 lattices)
(overall uncertainties including material balance and calibration)

FP	*C/E-1* ratios (%)			
	MINERVE / APOLLO2.8		DIMPLE / WIMS	
	JEF2.2	JEFF3.1.1	JEF2.2	JEFF3.1
^{147}Sm	**+36** ± 28	**+38** ± 28	**+2** ± 4	**+4** ± 4
^{149}Sm	**-4** ± 3	**-1** ± 3	**-6** ± 4	**-4** ± 4
^{152}Sm	**-1** ± 3	**0** ± 3	**0** ± 4	**0** ± 4
^{143}Nd	**-6** ± 3	**-3** ± 3	**-6** ± 4	**-3** ± 4
^{145}Nd	**+4** ± 3	**+4** ± 3	**0** ± 4	**+1** ± 4
^{155}Gd	**0** ± 3	**0** ± 3	**+3** ± 4	**+3** ± 4
^{153}Eu	**-14** ± 3	**-11** ± 3	**-10** ± 4	**-6** ± 4
^{103}Rh	**+11** ± 4	**+8** ± 4	**+10** ± 4	**+6** ± 4
^{95}Mo	**+1** ± 3	**-1** ± 3	**+2** ± 4	**0** ± 4
^{133}Cs	**+8** ± 5	**+8** ± 5	**+10** ± 4	**+10** ± 4
^{109}Ag	**+4** ± 5	**+4** ± 5	**+2** ± 4	**+2** ± 4
^{99}Tc	**+4** ± 4	**+6** ± 4	**+3** ± 4	**+8** ± 4

⇒ consistent results between JEF2.2 and JEFF3.1.1

Conclusions & prospects

Conclusions

- ^{155}Gd(n,γ), ^{149,152}Sm(n,γ),^{143}Nd(n,γ) and ^{95}Mo(n,γ) well assessed in JEFF3.1.1 (≤ 3%)

- Improvements still needed for some resonant FP:
  - slight overestimation of the ^{145}Nd (4%)
  - ^{99}Tc (6%), ^{103}Rh (8%) overestimated
  - ^{153}Eu underestimated (11%)

- Oscillation of ^{147}Sm sample with Cd-filter, to cancel ^{149}Sm thermal contribution

- Need of new samples with separated isotopes of ^{99,101}Ru, and well-characterized ^{133}Cs samples (several inconsistent samples due to material balance)

- Further studies to investigate the double heterogeneity effect on the Rh-doped samples
  → measurements scheduled in 2011 with new pure metallic Rh samples

- Several samples with increasing trends on the *C/E* with spectrum-hardness
  → need to check the flux calculation in the epithermal range: new calibration samples made of gold will be available soon…

Prospects

- Projects to interpret oscillation measurements in the DIMPLE reactor next year with the calculation route (AP2.8/SHEM-MOC/JEFF3.1.1):
  - CERES Assembly II (PWR spectrum)
  - CERES Assembly III (pure thermal spectrum)

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Thank you for your attention.

Comments on the samples material balance

Until now, only most important isotopes were taken into account in the material balance of the samples

According to several chemical analysis on additional pellets, all impurities are now taken into account:

- for the sintered samples:
  - rare-earth elements (all isotopes of Sm, Nd, Gd, Eu, …)
- for the compacted ones:
  - water (up to 10% of the reactivity effect in Cs samples)
  - H, ^{NAT}Zn and ^{NAT}C