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San Domenico Center

Impact of new results of the neutron capture cross section measurements for odd gadolinium isotopes on thermal-spectrum systems

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Structure of presentation

- Motivation for the use of Gd in thermal fission reactors
- Status of available data
- Performance of available data and necessity of new experiments
- New n_TOF xs
- Zed-2 benchmark and preliminary assessment of new xs
- Conclusions and future developments
- References

Importance of Gd odd isotopes in fission reactors

Use as **"burnable neutron poisons"** in nuclear reactors

- To increase the **efficiency** and economic performances of **reactor fuel, **it is necessary to **increase** the initial **enrichment of ^{235}U** in the fuel itself.
- However high enrichments pose severe safety problems due to the **high initial excess reactivity.**
- This can be **inherently compensated** by loading the fuel with **"burnable neutron poisons"**, i.e. isotopes with very high capture cross section, that are depleted together with the fissile isotopes.

It is very important to assess the **capture behavior of burnable poisons** in order to evaluate:

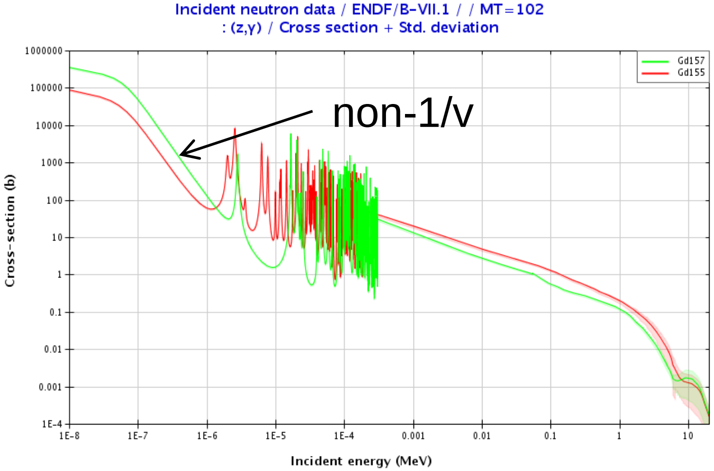
- the **economic gain due to the extension** of fuel life;
- the **residual reactivity penalty** at EOL, in terms of reactor days lost (16 pins Gd-doped FAs for PWRs = 5 full power days lost/year = 8 M€ for the electricity market in France);
- the **reactivity peak** for partially spent fuel for the criticality safety evaluations of Spent Fuel Pools.

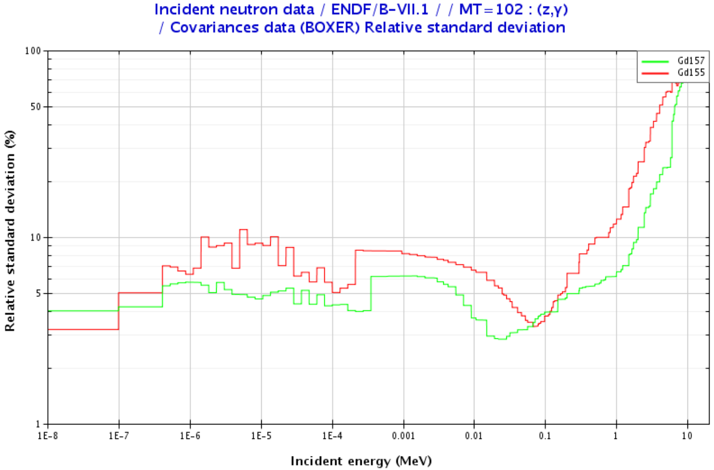
Use in Gen. II & Gen. III Reactors

Current **Gen. II and Gen. III** nuclear reactors make **extensive use of Gadolinium** as:

- **burnable neutron poison** (Gadolinia: Gd_2O_3) for **PWR, BWR, VVER** fuels
- **emergency shutdown poison** (Gadolinium nitrate, $GdNO_3$), for **CANDU.**

The reason of this choice is the **extremely high neutron capture cross sections** of the odd Gd isotopes (**^{155}Gd and ^{157}Gd**) for low energy neutrons (thermal to ≈10 eV).





157Gd(n,g) thermal

Despite their importance, the capture cross sections of the odd Gd isotopes have not been so extensively studied and are **not known with the accuracy required** by present-day nuclear industry.

Reference	Year	Thermal xs (kb)	Deviation from ENDF/B-VII
Pattenden *2nd At. En. Conf. Geneva, 16*	1958	264	+3.9%
Tattersall *Jour. Nucl. Ener. A 12, 32*	1960	213	-20%
Moller *Nucl. Sci. Eng. 8, 183*	1960	254	=
Sun *J. Radioanal. Nucl. Chem. 256, 541*	2003	232	-9%
Leinweber *Nucl. Sci. Eng. 154, 261*	2006	226	-12%
Mughabghab *Evaluation (adopted in ENDF/B-VII)*	**2006**	**254 ± 0.3%**	=
Choi *Nucl. Sci. Eng. 177, 219*	2014	239	-6%

Evaluated data vs some experimental benchmarks

ICSBEP	Config.	K Exp	K ENDF/B-VII	K JEFF-3.1	K Leinweber	Improvement?
HST-014	C2	1.0000	1.00996	1.01304	1.01903	N
	C3	1.0000	1.01827	1.01852	1.02636	N
LCT-035	C3	1.0000	0.99591	0.99556	0.99935	Y
LCT-005	C2	1.0000	1.00029	1.00006	1.00466	N
	C3	1.0000	0.99907	1.00002	1.01651	N
	C4	1.0000	0.99721	0.99846	1.01602	N
	C6	1.0000	1.00684	1.00697	1.00962	N
	C7	1.0000	1.00191	1.00258	1.00846	N
	C8	1.0000	1.00163	1.00295	1.01213	N
	C9	1.0000	1.00257	1.00379	1.01459	N
	C10	1.0000	1.00135	1.00290	1.01474	N
	C11	1.0000	1.00165	1.00342	1.01544	N
	C13	1.0000	1.01309	1.01129	1.01303	N
	C15	1.0000	1.01751	1.01750	1.02436	N

van der Marck 2012 Analysis

In 2012 S. C. van der Marck published an extensive and comprehensive analysis of **ENDF/B-VII.1**, **JENDL-4.0**, **JEFF-3.1.1** performances using MCNP6 over available benchmarks (mainly ICSBEP). The conclusion about Gd isotopes is that the evaluations above aren’t good enough to represent the experimental data, experimental uncertainties included.

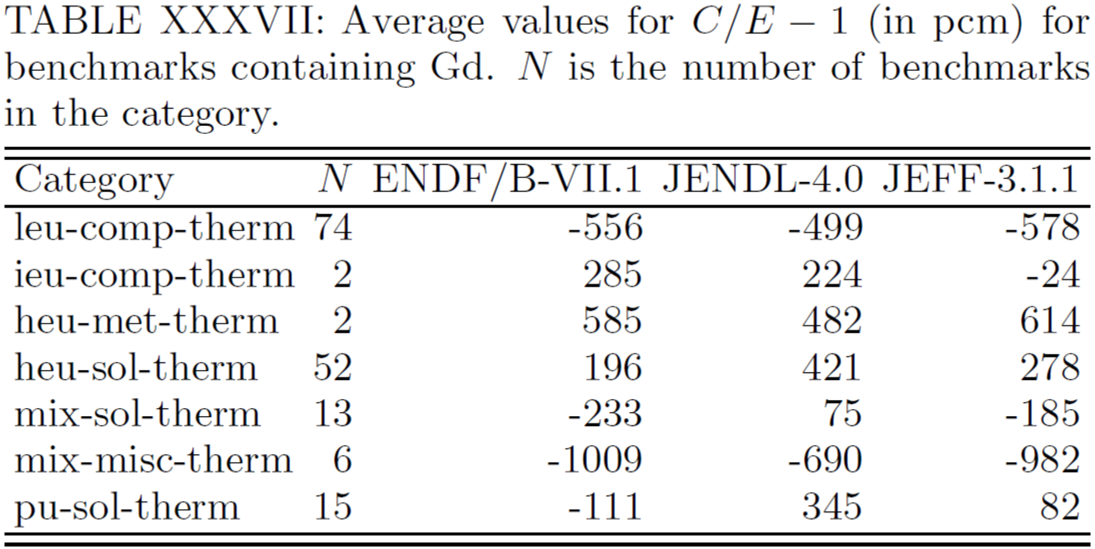
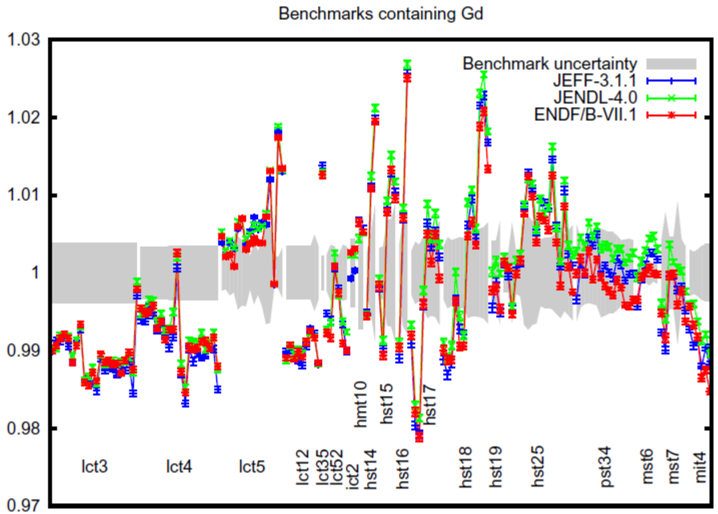


TABLE XXXVII: Average values for $C/E - 1$ (in pcm) for benchmarks containing Gd. N is the number of benchmarks in the category.

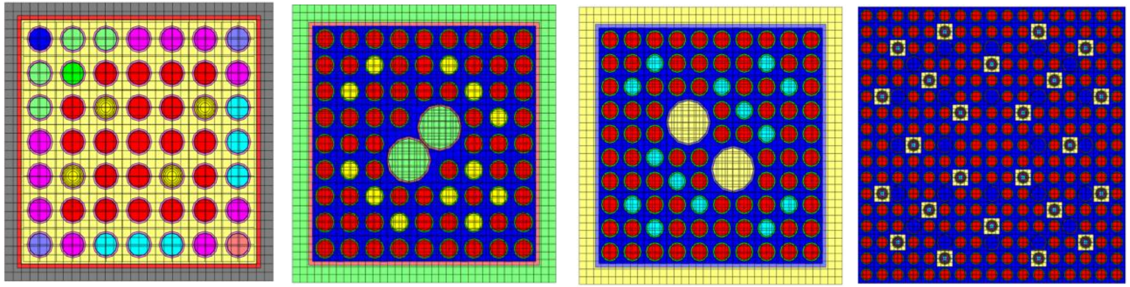
Category	N	ENDF/B-VII.1	JENDL-4.0	JEFF-3.1.1
leu-comp-therm	74	-556	-499	-578
ieu-comp-therm	2	285	224	-24
heu-met-therm	2	585	482	614
heu-sol-therm	52	196	421	278
mix-sol-therm	13	-233	75	-185
mix-misc-therm	6	-1009	-690	-982
pu-sol-therm	15	-111	345	82



ENEA S/U Analysis

- To understand and assess the importance and role of 157Gd and 155Gd in nuclear fuels, a **Sensitivity and Uncertainty (SU) analysis on k** for several different FAs has been performed at BOL, hot-full power (HFP) conditions using the US-NRC reference **SCALE 6.1** code system developed at ORNL.
- **Tsunami-2D** sequence with **ENDF/B-VII.0** evaluations.

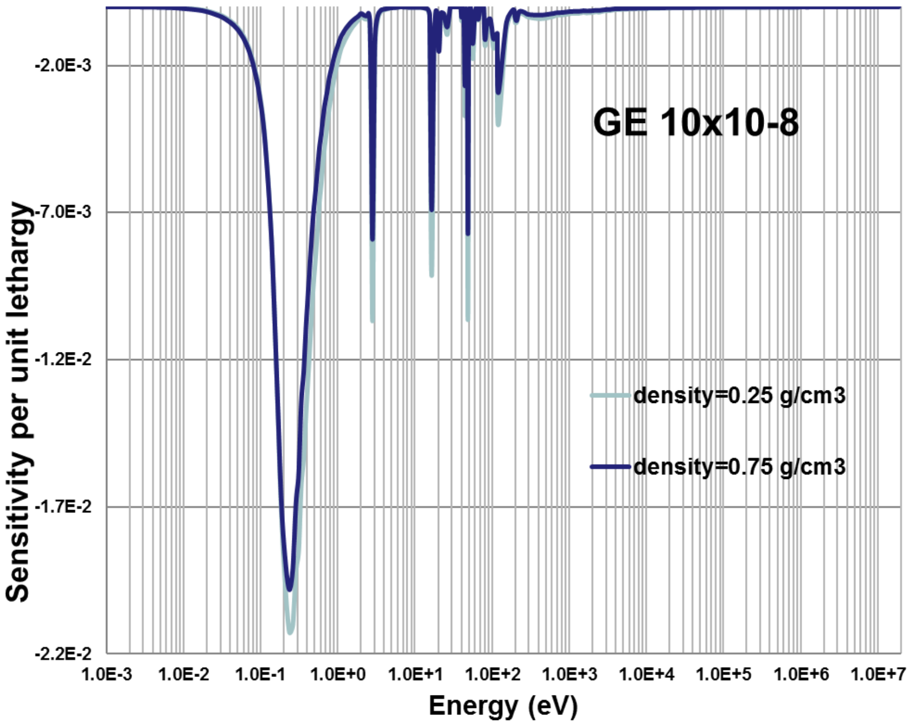
$$S_i(E) = \frac{\sigma_i}{k}\frac{\partial k}{\partial \sigma_i} = \frac{\frac{\partial k}{k}}{\frac{\partial \sigma_i}{\sigma_i}}$$


PB BWR GE 9x9-7 GE 10x10-8 EPR

Covariance Data: 44-group library (based on ENDF/B-VII.0)

ENEA S/U Analysis

- BWR GE 10x10-8 results.
- Two different moderator densities tested.
- The region of highest sensitivity **for k** is **between 0.1 and 1 eV**.



ENEA S/U Analysis

Nuclide-Reaction	**Contrib. to Uncertainty in k** ***(% Δk/k)***	**Rank**
^{235}U $\bar{\nu}$	2.70E-01	1.00
^{238}U(n,γ)	1.97E-01	0.81
^{235}U(n,γ)	1.43E-01	0.64
^{235}U(n,f)	1.43E-01	0.56
^{235}U(n,f) / ^{235}U(n,γ)	1.21E-01	0.54
^{238}U(n,n')	1.20E-01	0.51
^{235}U χ	1.13E-01	0.45
^{238}U $\bar{\nu}$	7.11E-02	0.32
^{157}Gd(n,γ)	6.03E-02	0.26
^{155}Gd(n,γ)	4.48E-02	0.20
^{92}Zr(n,γ)	4.29E-02	0.16
^{1}H(n,γ)	3.67E-02	0.14
^{91}Zr(n,γ)	3.48E-02	0.13
^{1}H(n,n)	3.13E-02	0.12
^{90}Zr(n,γ)	2.82E-02	0.10

The **uncertainty** on **Gd** cross sections gives the **largest contribution** to the uncertainty on k **after ^{235,238}U**.

Several **cross sections** in this list have already been **measured at nTOF**.

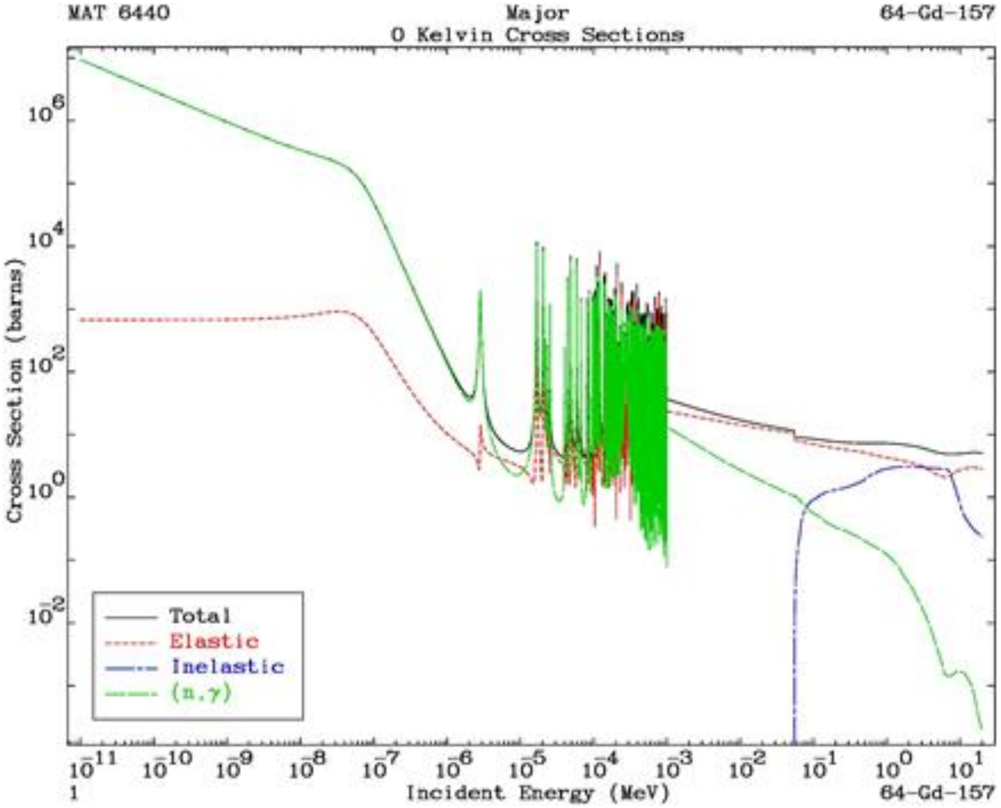
New n_TOF measurements

- The n_TOF Collaboration decided in 2015 to carry out new Gd odd isotopes (n,g) xs measurements;
- Isotopically «pure» samples were aquired from ORNL;
- Measurements were conducted in July 2016;
- Results are being published (see also M. Mastromarco presentation at this conference, Thursday 17:30).

- - - - -

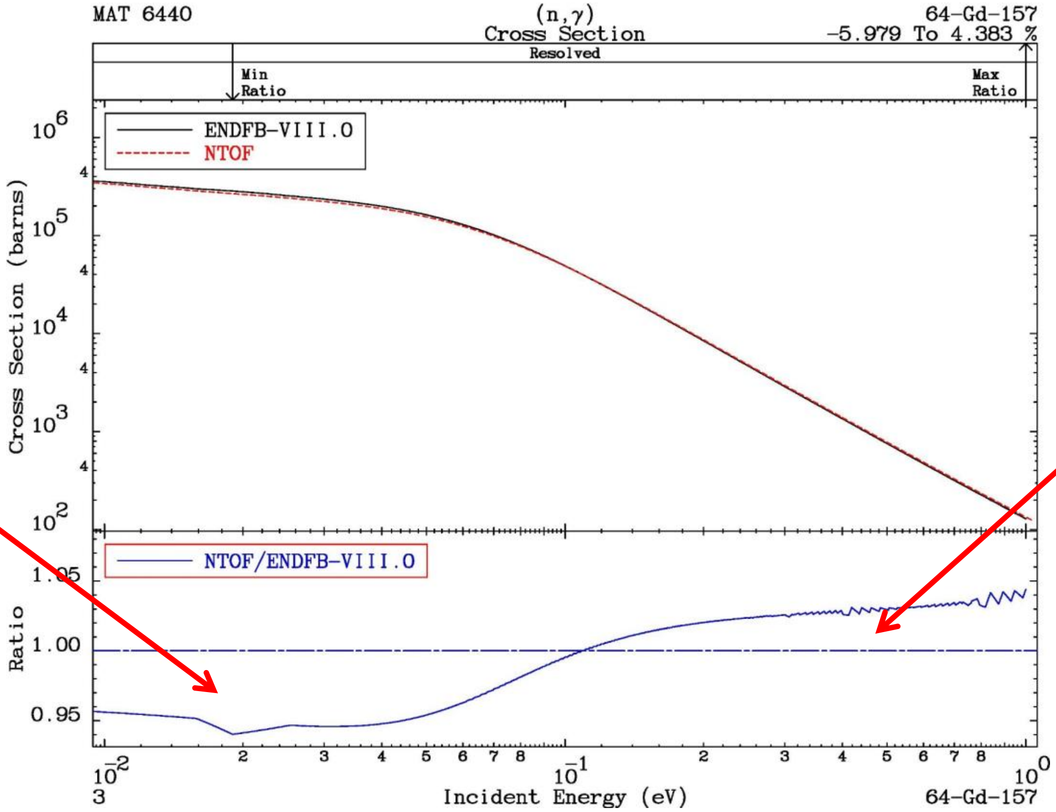
- The newly obtained xs @ 0.025 eV results 239.8 ± 9.3 kb;
- Xs uncertainty @ 0.025 eV about 3.9%;
- Uncertainty to be reduced to about 3.0% after detailed post-irradiation analysis of the samples (to be accomplished after use at GELINA).

New n_TOF 157Gd xsec



Preliminary cross sections retrieved from ArXiv 1805.04149 (2018)

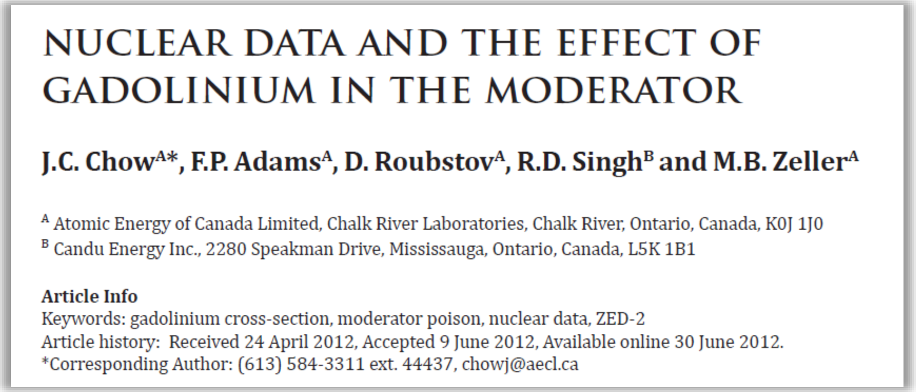
New n_TOF 157Gd(n,g) xsec vs ENDF/B-VIII



≈ -3.5%

≈ +2.5%

AECL - Chalk River results



NUCLEAR DATA AND THE EFFECT OF GADOLINIUM IN THE MODERATOR

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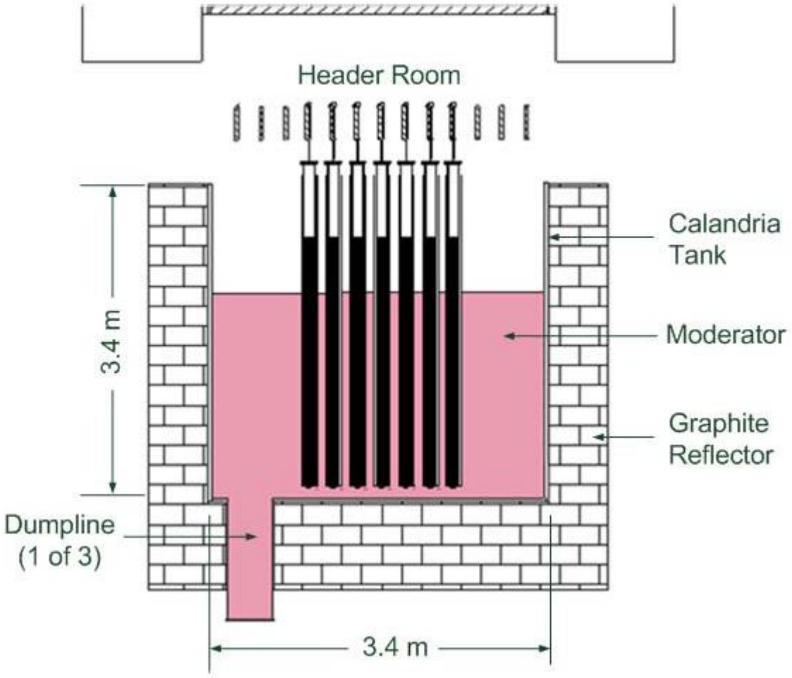
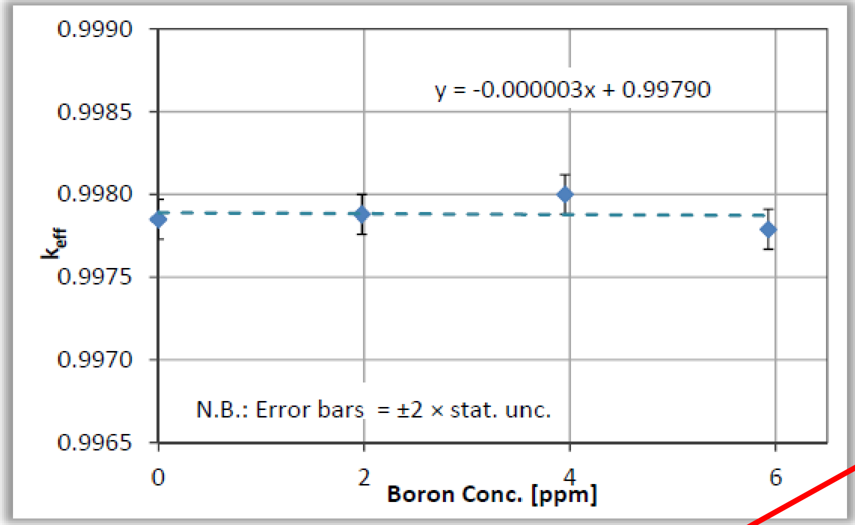


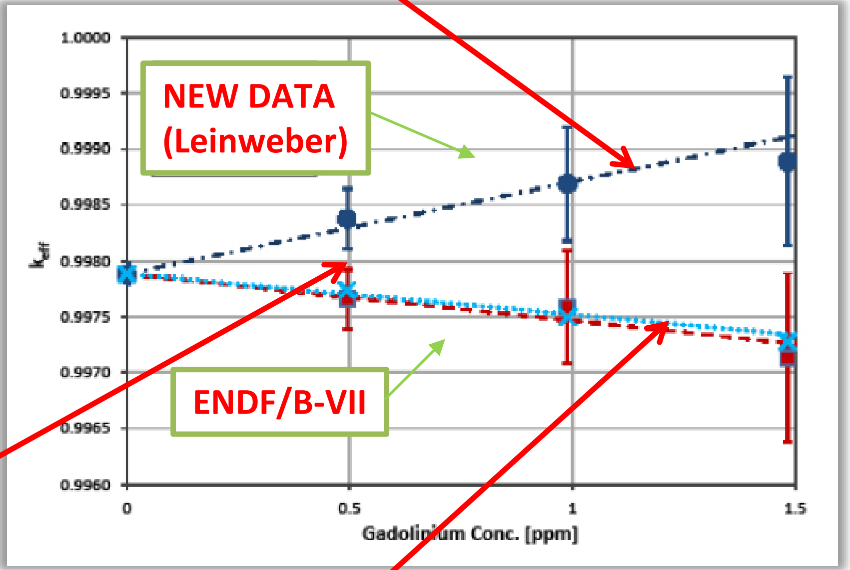
ZED-II Research Reactor

AECL - Chalk River results

(n,g) @ 0.025 eV = 226 kb



N.B.: Error bars = ±2 × stat. unc.



NEW DATA (Leinweber)

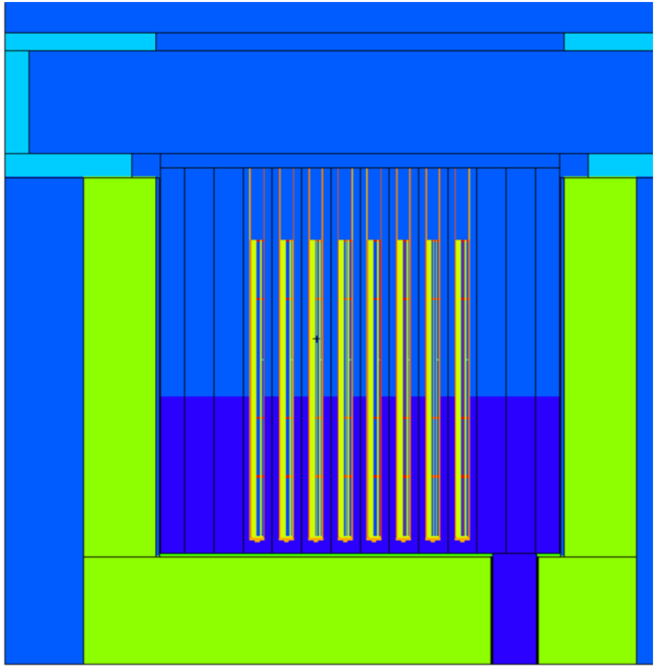
ENDF/B-VII

Real thermal xs between
226 and 254 kb!

≈ -22 pcm

(n,g) @ 0.025 eV = 254 kb

MCNP6 Calculations

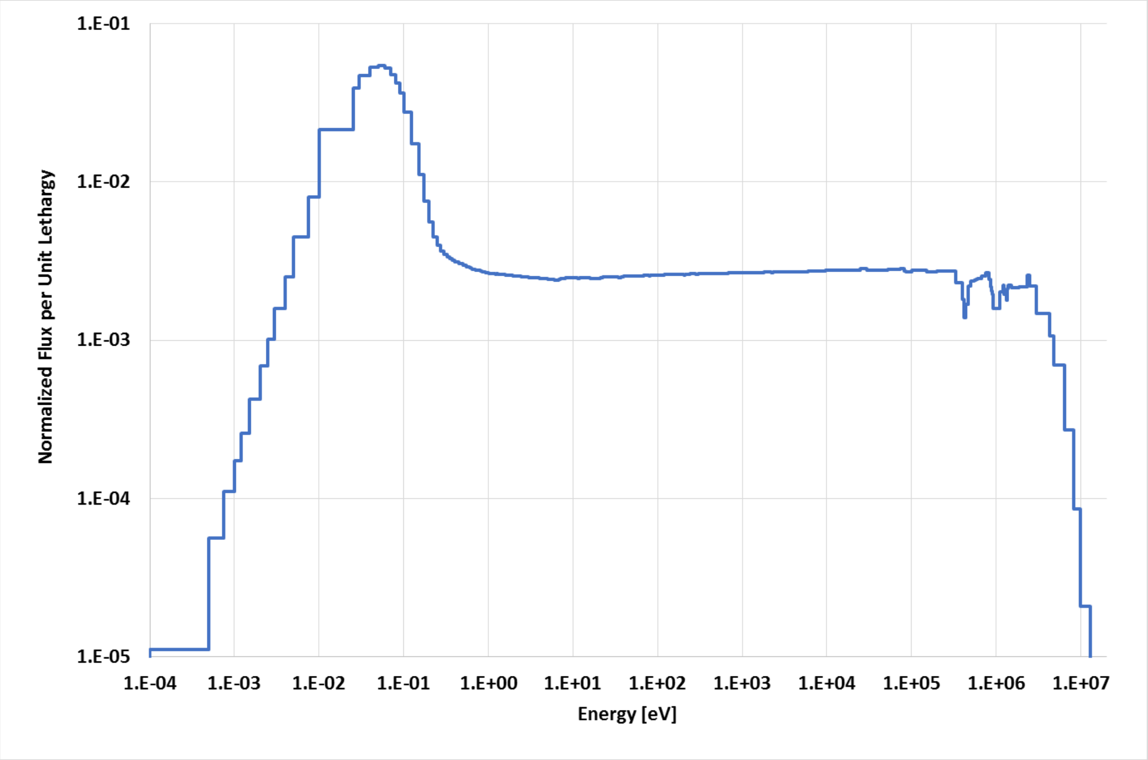


	Lower Energy bound [MeV]	Upper Energy bound [MeV]	Lower Energy bound [eV]	Upper Energy bound [eV]	Spectrum Group Score	Rel Err (1 sd)	Percentage
Case 1	1.00E-11	1.00E-10	1.00E-05	1.00E-04	1.36372E-03	0.0005	0.0005%
	1.00E-10	1.00E-07	1.00E-04	1.00E-01	1.64690E+02	0.0000	63.1422%
	1.00E-07	2.00E+01	1.00E-01	2.00E+07	9.61324E+01	0.0000	36.8572%
				total	2.60824E+02	0.0000	99.9999%
Case 2	1.00E-11	1.00E-10	1.00E-05	1.00E-04	1.33117E-03	0.0005	0.0005%
	1.00E-10	1.00E-07	1.00E-04	1.00E-01	1.61164E+02	0.0000	62.6011%
	1.00E-07	2.00E+01	1.00E-01	2.00E+07	9.62810E+01	0.0000	37.3985%
				total	2.57446E+02	0.0000	100.0001%
Case 3	1.00E-11	1.00E-10	1.00E-05	1.00E-04	1.30079E-03	0.0005	0.0005%
	1.00E-10	1.00E-07	1.00E-04	1.00E-01	1.57696E+02	0.0000	62.0626%
	1.00E-07	2.00E+01	1.00E-01	2.00E+07	9.63953E+01	0.0000	37.9372%
				total	2.54092E+02	0.0000	100.0002%
Case 4	1.00E-11	1.00E-10	1.00E-05	1.00E-04	1.26969E-03	0.0005	0.0005%
	1.00E-10	1.00E-07	1.00E-04	1.00E-01	1.54368E+02	0.0000	61.5284%
	1.00E-07	2.00E+01	1.00E-01	2.00E+07	9.65201E+01	0.0000	38.4712%
				total	2.50889E+02	0.0000	100.0001%

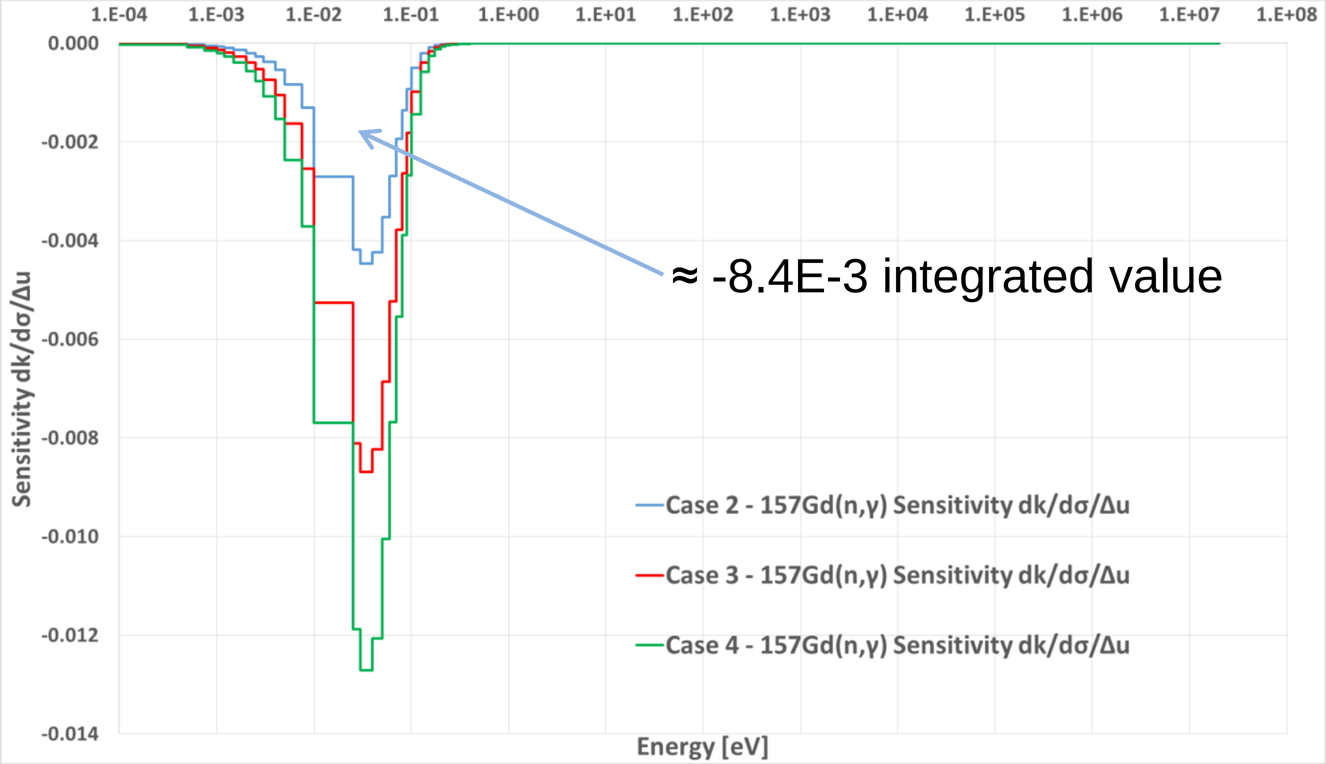
Zed-2 Flux per unit lethargy in moderator

MCNP6 calculations

Very well thermalized specturm!

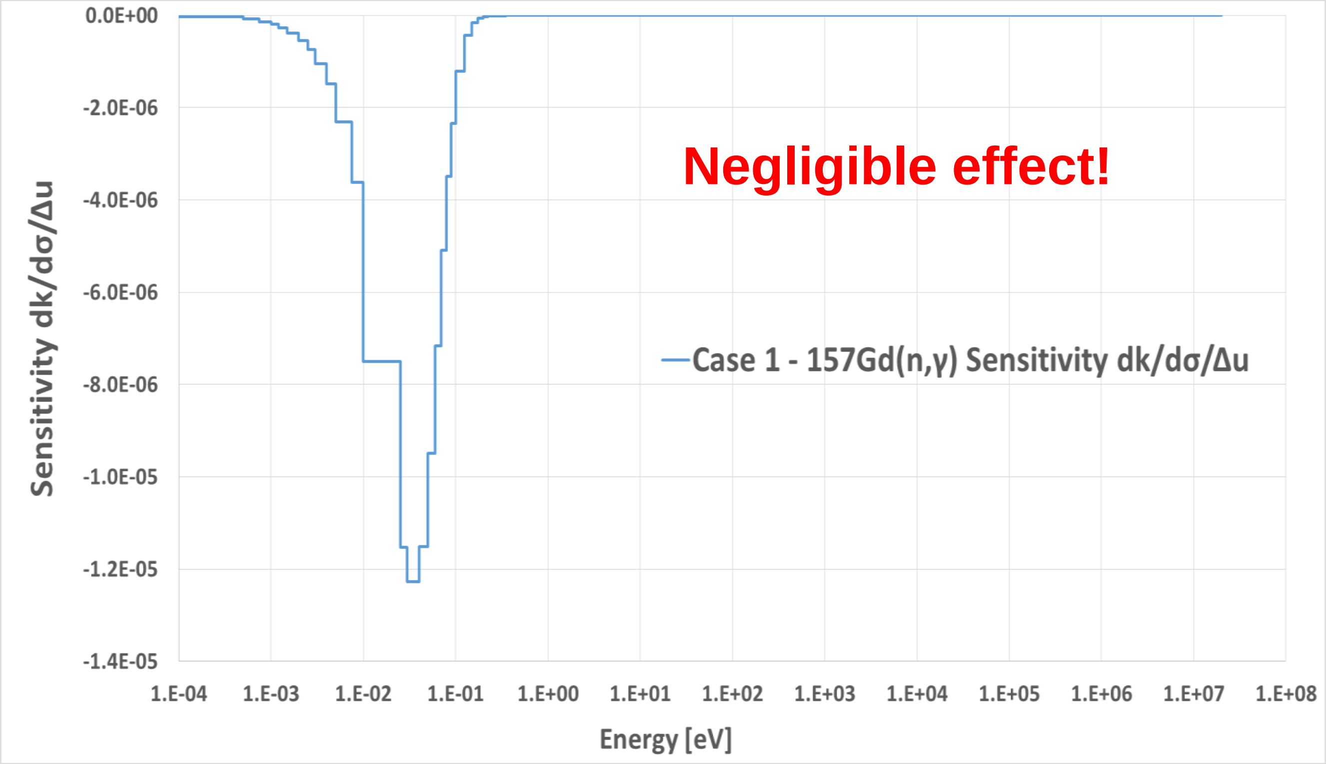


Zed-2 k sensitivity per unit lethargy to 157Gd(n,g)



≈ -8.4E-3 integrated value

Zed-2 k sensitivity per unit lethargy to 157Gd(n,g) only due to impurities in graphite reflector



Negligible effect!

1&2 groups Zed-2 sensitivities to 157Gd(n,g)

	Energy Bounds [eV]		Lethargy Interval [--]	^{157}Gd(n,γ) Sensitivity	Relative Uncertainty	^{157}Gd(n,γ) Sensitivity/ Δu	Sensitivity/ Δu Absolute Uncertainty	^{155}Gd(n,γ) Sensitivity	Relative Uncertainty	^{155}Gd(n,γ) Sensitivity/ Δu	Sensitivity/ Δu Absolute Uncertainty
Case 2	1.00E-04	1.00E-01	6.91E+00	-8.1813E-03	0.0004	-1.1844E-03	-4.7375E-07	-1.8362E-03	0.0004	-2.6582E-04	-1.0633E-07
	1.00E-01	2.00E+07	1.91E+01	-1.7510E-04	0.0003	-9.1609E-06	-2.7483E-09	-3.9074E-05	0.0003	-2.0443E-06	-6.1328E-10
		sum of 2 groups	2.60E+01	-8.3564E-03	0.0004	-3.2113E-04	-1.2778E-07	-1.8753E-03	0.0004	-7.2066E-05	-2.8676E-08
	integration	1 group calculation	2.60E+01	-8.3577E-03	0.0004	-3.2118E-04	-1.2847E-07	-1.8756E-03	0.0004	-7.2079E-05	-2.8831E-08
Case 3	1.00E-04	1.00E-01	6.91E+00	-1.5901E-02	0.0004	-2.3019E-03	-9.2076E-07	-3.5684E-03	0.0004	-5.1658E-04	-2.0663E-07
	1.00E-01	2.00E+07	1.91E+01	-3.4339E-04	0.0004	-1.7966E-05	-7.1862E-09	-7.6710E-05	0.0003	-4.0133E-06	-1.2040E-09
		sum of 2 groups	2.60E+01	-1.6244E-02	0.0004	-6.2427E-04	-2.4971E-07	-3.6451E-03	0.0004	-1.4008E-04	-5.5737E-08
	integration	1 group calculation	2.60E+01	-1.6247E-02	0.0004	-6.2437E-04	-2.4975E-07	-3.6457E-03	0.0004	-1.4010E-04	-5.6041E-08
Case 4	1.00E-04	1.00E-01	6.91E+00	-2.3277E-02	0.0004	-3.3697E-03	-1.3479E-06	-5.2231E-03	0.0004	-7.5612E-04	-3.0245E-07
	1.00E-01	2.00E+07	1.91E+01	-5.0723E-04	0.0003	-2.6537E-05	-7.9612E-09	-1.1341E-04	0.0003	-5.9334E-06	-1.7800E-09
		sum of 2 groups	2.60E+01	-2.3784E-02	0.0004	-9.1402E-04	-3.6366E-07	-5.3365E-03	0.0004	-2.0508E-04	-8.1596E-08
	integration	1 group calculation	2.60E+01	-2.3788E-02	0.0004	-9.1416E-04	-3.6567E-07	-5.3374E-03	0.0004	-2.0511E-04	-8.2046E-08

1-grp Δk

$$\Delta k \cong k \cdot S \cdot \frac{\Delta\sigma}{\sigma}$$

$$k = 0.99766 \qquad S \cong -8.36\text{E}{-3} \qquad \Delta\sigma \cong -0.035\,\sigma$$

$$\boxed{\Delta k \cong \ +29.2\ \text{pcm}}$$

The ideal, compensating gain should have been around +22 pcm.

Approximation introduced: total sensitivity attributed solely to thermal group (<0.1 eV)!

2-grps Δk

- 2 groups: 1) 0.0 – 0.1 eV,
  2) 0.1 – 2.0E7 eV

- Second group by far less important than first;

- Correction to k results now roughly **+ 28.1 pcm**;

- Full MCNP6 calculations with the new xs are needed for a more precise assessment;

- These will be accomplished in the next weeks.

Conclusions and next steps

From the preliminary and «back of the envelope» analyses conducted so far

- The new xs seems to satisfy the Zed-2 experiments **much better** than currently available evaluations;
- The new xs, again based on Zed-2 results, seems to underestimate slightly capture by about 0.8%;
- Infinite groups calculations with MCNP6 are needed for better confidence on the performance of the new xs in Zed-2;
- Many other ICSBEP benchmarks are to be used for further validation of the new product;
- Uncertainty in the thermal region needs to be further reduced;
- GELINA experiments with the same samples are underway;
- Possibility to produce new evaluations for JEFF4.

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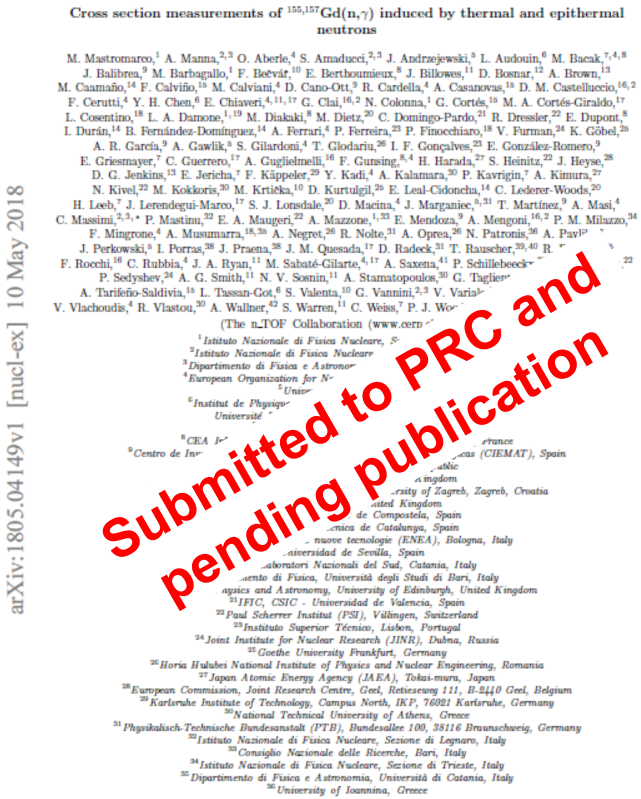
Forthcoming papers



arXiv:1805.04149v1 [nucl-ex] 10 May 2018

Cross section measurements of ^{155,157}Gd(n,γ) induced by thermal and epithermal neutrons

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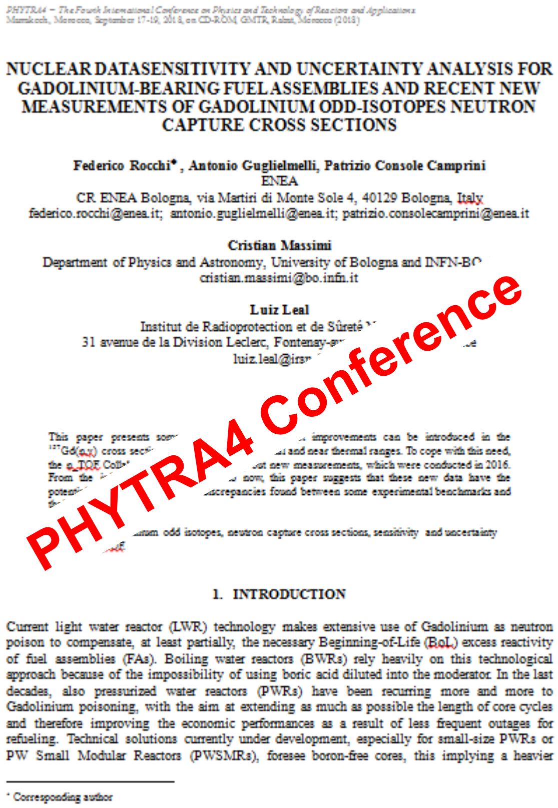

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NUCLEAR DATASENSITIVITY AND UNCERTAINTY ANALYSIS FOR GADOLINIUM-BEARING FUEL ASSEMBLIES AND RECENT NEW MEASUREMENTS OF GADOLINIUM ODD-ISOTOPES NEUTRON CAPTURE CROSS SECTIONS

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This paper presents som improvements can be introduced in the ^{157}Gd(n,γ) cross secti al and near thermal ranges. To cope with this need, the n_TOF Colla ut new measurements, which were conducted in 2016. From the now, this paper suggests that these new data have the potenti iscrepancies found between some experimental benchmarks and

inium odd isotopes, neutron capture cross sections, sensitivity and uncertainty
OF

1. INTRODUCTION

Current light water reactor (LWR) technology makes extensive use of Gadolinium as neutron poison to compensate, at least partially, the necessary Beginning-of-Life (BoL) excess reactivity of fuel assemblies (FAs). Boiling water reactors (BWRs) rely heavily on this technological approach because of the impossibility of using boric acid diluted into the moderator. In the last decades, also pressurized water reactors (PWRs) have been recurring more and more to Gadolinium poisoning, with the aim at extending as much as possible the length of core cycles and therefore improving the economic performances as a result of less frequent outages for refueling. Technical solutions currently under development, especially for small-size PWRs or PW Small Modular Reactors (PWSMRs), foresee boron-free cores, this implying a heavier

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<u>**Thank you for your attention**</u>

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