

157Gd and 155Gd (n,g) xs Project

From new measurements to new evaluations

JEFF Meeting 2017, OECD-NEA, April 26th 2017

Federico Rocchi / ENEA FSN-SICNUC-PSSN Bologna - Italy



























Structure of presentation

- Introduction
- Scientific motivations
- Scientific background
- ENEA S/U analysis for LWRs
- Experimental campaign at NTOF
- Further experiments
- Future developments of the project



Introduction

- At the Italian national nTOF meeting in **March 201**: idea of dedicating scientific efforts towards a better (n,g) xs for reactor applications.
- While present evaluations of these xs perform acce for improvements.
- At the international nTOF meeting in **May 2015** in § Proposal to the ISOLDE and nTOF Committee was and positively accepted, opening the path to dedicate experiments in **Summer 2016**.
- The Proposal received endorsements by, among of
- Later on, statements of interest arrived also from **IF**

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron capture cross section for 155Gd and Gd for Nuclear Technology

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Abstract: We propose to measure the neutron capture cross-section of ¹⁵⁵Gd and ¹⁵⁷Gd from thermal to 1 MeV neutron energy. The main motivation is related to the need of accurate data for applications to nuclear reactors, but new data could also be useful for recent developments in Neutron Capture Therapy, and for new detector concepts in neutrino research. The measurement should be performed in EAR-1 with cutting edge C_nD_n detectors specifically designed for n_TOF. Since the cross section of these two isotopes changes by orders of magnitude as a function of neutron energy, two highlyenriched samples for each isotope will be measured: a very thin one for neutron energies up to 100 meV, and a thicker one for neutron energies above 100 meV.

Requested protons: 2.4×10^{18} protons on target

Experimental Area: n_TOF EAR-1 (185 m flight path) Detection system: Array of 4 C₆D₆ detectors

Samples: 10 mg, 100 mg 155Gd and 5 mg, 200 mg 157Gd enriched on a 1 cm radius disc for each isotope



Gd

ce

mal

armly

PSI.

ab.

Introduction

- Several persons and institutions are involved in the whole project.
- People involved at ENEA Bologna:
 - F. Rocchi (reactor physics aspects)
 - D. M. Castelluccio (experiments at nTOF and data analysis)
 - A. Guglielmelli (reactor physics aspects)
 - G. Clai (data analysis)
 - S. Lo Meo (spokesperson for Proposal at nTOF)



Scientific motivations

The main motivation is related to the use of "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 ²³⁵U 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 PWR = 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.

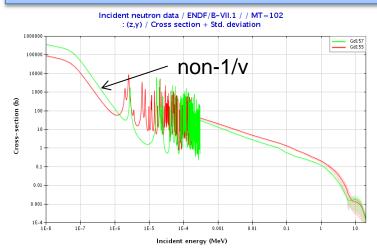


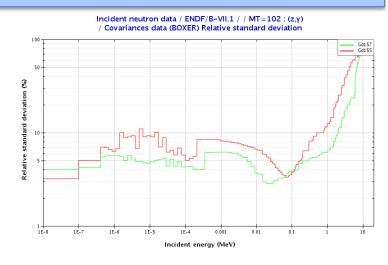
Use in Gen. II & Gen. III Reactors

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

- burnable neutron poison (Gadolinia: Gd₂O₃) for PWR, BWR, VVER fuels
- emergency shutdown poison (Gadolinium nitrate, GdNO₃), 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 $\approx 10 \text{ eV}$).



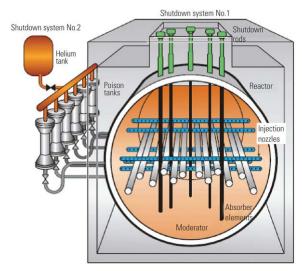




Use in CANDU Reactors

Emergency Shutdown Poison

- In CANDU reactors, in case of severe accidents due to or leading to criticality excursions, Gadolinium nitrate is injected into the moderator heavy water, to reduce (eliminate) criticality risks or excursions.
- However, uncertainties in the (n,γ) cross section of Gd odd isotopes may **impose special care** in the **safety** calculations for the licensing of CANDU reactors.





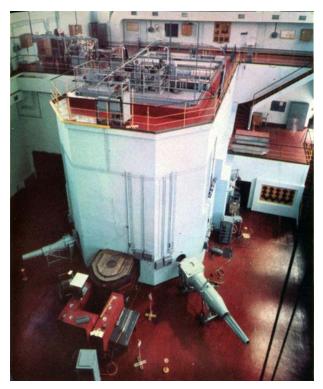
157Gd(n,g) thermal

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 (b)	Deviation from ENDF/B-VII
Pattenden 2 nd At. En. Conf. Geneva, 16	1958	264000	+3.9%
Tattersall Jour. Nucl. Ener. A 12, 32	1960	213000	-20%
Moller Nucl. Sci. Eng. 8, 183	1960	254000	=
Groshev Izv. Akad. Nauk, SSSR, 26, 1118	1962	240000	-6%
Sun J. Radioanal. Nucl. Chem. 256, 541	2003	232000	-9%
Leinweber Nucl. Sci. Eng. 154, 261	2006	226000	-12%
Mughabghab Evaluation (adopted in ENDF/B-VII)	2006	254000 ± 0.3%	=
Choi Nucl. Sci. Eng. 177, 219	2014	239000	-6%



CEA Melusine/GEDEON-II results



CEA Qualification Program for French LWR using the Melusine reactor in Grenoble in 1985 (2015 re-analysis based on JEFF 3.1.1 evaluations for EPR).







CEA Melusine/GEDEON-II results

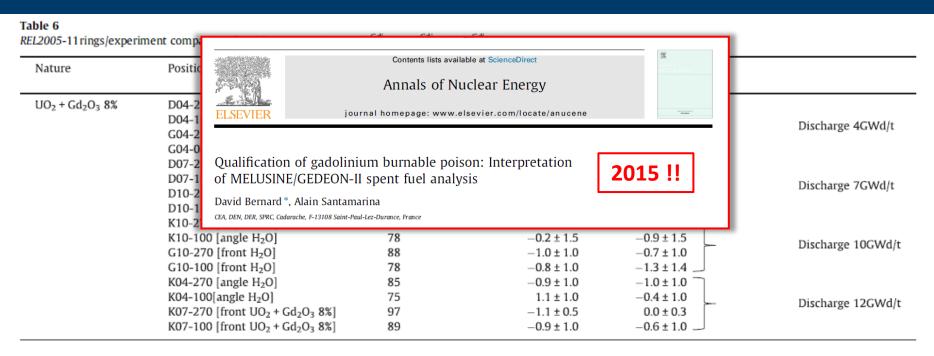
Isotope Concentrations (C/E-1)

Nature	Position	Consumption [%] [155Gd]	¹⁵² Gd/ ²³⁸ U [%]	¹⁵⁴ Gd/ ²³⁸ U [%]	¹⁵⁵ Gd/ ²³⁸ U [%]	¹⁵⁶ Gd/ ²³⁸ U [%]	¹⁵⁷ Gd/ ²³⁸ [%]
UO ₂ + Gd ₂ O ₃	D07-270 [front	26	-3±5	-2 ± 1	1 ± 3	-3 ± 1	7 ± 5
5%	$UO_2 + Gd_2O_3 5\%$						
	D07-100 [front	58	-4 ± 5	-1 ± 1	-2 ± 2	–1 ±1	2 ± 3
	$UO_2 + Gd_2O_3 5\%$						
	G04-270 [front H ₂ O]	23	-11 ± 5	-1 ± 1	0 ± 6	-2 ± 1	9 ± 10
	G04-100 [front H ₂ O]	32	-12 ± 5	-1 ± 1	0 ± 3	-2 ± 1	8 ± 5
$UO_2 + Gd_2O_3$	D04-270 [angle H ₂ O]	40	-7 ± 5	-1 ± 1	4 ± 2	−3 ± 1	10±2
8%							
	D04-100 [angle H ₂ O]	33	-8 ± 5	-1 ± 1	6 ± 1	-4 ± 1	14 ± 2
	G04-270 [front H ₂ O]	40	-8 ± 5	0 ± 1	1 ± 1	–1 ± 1	6 ± 2
	G04-080 [front H ₂ O]	34	-6 ± 5	-1 ± 1	0 ± 1	–1 ± 1	3 ± 2
	D07-270 [front	53	-11 ± 5	-1 ± 1	3 ± 2	-3 ± 1	13 ± 3
	$UO_2 + Gd_2O_3 8\%$						
	D07-100 [front	46	-8 ± 5	-1 ± 1	2 ± 2	-2 ± 1	10±2
	$UO_2 + Gd_2O_3 8\%$						
	D10-270 [angle H ₂ O]	66	-12 ± 5	-1 ± 1	3 ± 3	−2 ± 1	13 ± 4
	D10-100 [angle H ₂ O]	58	-9 ± 5	0 ± 1	1 ± 3	-2 ± 1	8 ± 3
	K10-270 [angle H ₂ O]	87	-15 ± 6	-1 ± 1	17 ± 9	-3 ± 1	43 ± 15
	K10-100 [angle H ₂ O]	78	-15 ± 5	0 ± 1	1 ± 4	-1 ± 1	10±6
	G10-270 [front H ₂ O]	88	-17 ± 6	0 ± 1	9 ± 10	−2 ± 1	24 ± 17
	G10-100 [front H ₂ O]	78	-18 ± 5	0 ± 1	4 ± 5	–1 ± 1	16 ± 7
	K04-270 [angle H ₂ O]	85	-15 ± 5	0 ± 1	7 ± 9	–1 ± 1	25 ± 16
	K04-100[angle H ₂ O]	75	-14 ± 5	-1 ± 1	-4 ± 6	–1 ± 1	3 ± 8
	K07-270 [front	97	-17 ± 5	0 ± 1	56 ± 30	-1±1	9 ± 40
	$UO_2 + Gd_2O_3 \ 8\%$] $KO7-100 \ [front \ UO_2 + Gd_2O_3 \ 8\%]$	89	-15 ± 5	0 ± 1	9 ± 11	-1 ± 1	24 ± 20

Large
differences
between
calculations
and
experiment



CEA Melusine/GEDEON-II results



Some non-negligible biases are found for 157Gd, suggesting an underprediction of the capture xs.



AECL - Chalk River results

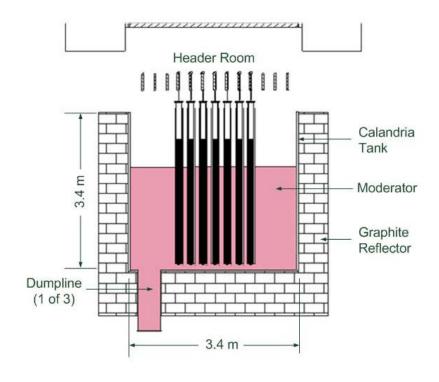
NUCLEAR DATA AND THE EFFECT OF GADOLINIUM IN THE MODERATOR

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ZED-II Research Reactor

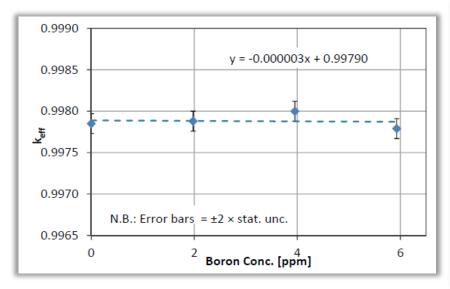


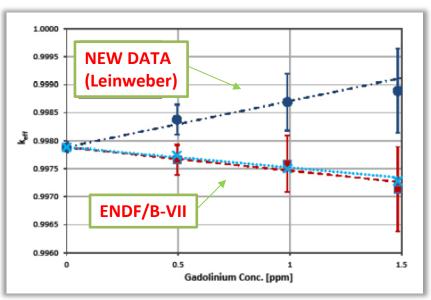


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AECL - Chalk River results







PSI 2008-2009 Validations for BWR Fuels

- PSI C-E comparisons using the <u>Leinweber data of 2006</u> within the PROTEUS reactor research programs (SVEA96+).
- Results for the total fission rate and 238U capture rate are generally much improved wrt using previous Gd evaluations.

BUT...

ICSBEP LCT-035, LCT-005, HST-014 not well reproduced (JEFFDOC-1210, 2007)



PROTEUS



PSI 2008-2009 Validations for BWR Fuels

ICSBEP	Config.	K_ref	ENDF/B-VII	JEFF-3.1	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	Υ
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 Marcl ENDF/B-VII.1, JENDL-4.0, benchmarks (mainly ICSBE

The conclusion about Gd is to represent the experiment



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Nuclear Data Sheets 113 (2012) 2935-3005

Nuclear Data Sheets

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I comprehensive analysis of sing MCNP6 over available

Benchmarking ENDF/B-VII.1, JENDL-4.0 and JEFF-3.1.1 with MCNP6

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Recent releases of three major world nuclear reaction data libraries, ENDF/B-VII.1, JENDI-4.0, and JEFF-3.1.1, have been tested extensively using benchmark calculations. The calculations were performed with the latest release of the continuous energy Monte Carlo neutronics code MCNP, i.e. MCNP6. Three types of benchmarks were used, set, criticality safety benchmarks, (fusion) shielding benchmarks, and reference systems for which the effective delawed neutron fraction is reported.

For criticality safety, more than 2000 benchmarks from the International Handbook of Criticality Safety Benchmark Experiments were used. Benchmarks from all categories were used, ranging from low-enriched uranium, compound fuel, thermal spectrum ones (LEU-COMP-THERM), to mixed uranium-putonium, metallic leuk, fast spectrum ones (MEM-MET-FAST), For fusion shielding many benchmarks were based on IAFA specifications for the Oktavian experiments (for Al, Co, Cr, Cu, LIF, Mm, Mo, Si, Ti, W, Zr), Puison Neutronics Source in Japan (for Be, C, No, Pe, Pb), and Pulsed Sphere experiments at Lawrence Livermore National Laboratory (for *Li, ¹Li, ¹Be, C, No, Q, Mg, Al, Ti, Fp, Pb, Do, Hp, Occurret, polysylvenee and tellon). The new functionality in McNP6 to calculate the effective delayed neutron fraction was tested by comparison with more than thirty measurements in widely varying systems. Among these were measurements in the Tank Critical Assembly (TCA in Japan) and IPEN/MB-DI (Brazill), both with a thermal spectrum, two cores in Measurca (Prancio and three cores in the Past Critical Assembly (FCA), Japan), all with fast spectra.

The performance of the three libraries, in combination with MCNP6, is shown to be good. The results for the LBU-COMP-TIEREM category are on awarege very close to the benchmark value. Also for most other categories the results are satisfactory. Deviations from the benchmark values do occur in certain benchmark series, or in isolated essess within benchmark series. Such instances can often be related to nuclear data for specific non-fissile elements, such as C, Fe, or Gd. Indications are that the intermediate and mixed spectrum cases are less well described.

The results for the shielding benchmarks are generally good, with very similar results for the three libraries in the majority of cases. Nevertheless there are, in certain cases, strong deviations between calculated and benchmark values, such as for Co and Mg. Also, the results show discrepancies at certain energies or angles for e.g. C, N, O, Mo, and W.

The functionality of MCNP6 to calculate the effective delayed neutron fraction yields very good results for all three libraries.

* Corresponding author, electronic address: vandermarck@nrg.eu

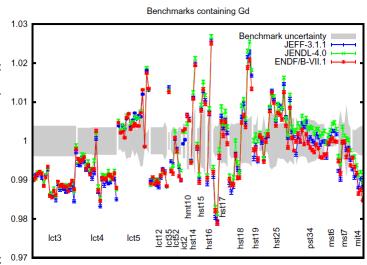
Editorial Note: All papers in the present issue, including this paper, are printed in black and white. Color version is available online at www.sciencedirect.com, see also www.elsevier.com/locate/nds. s above aren't good enough ainties included.



van der Marck 2012 Analysis

TABLE XXXVII: Average values for C/E-1 (in pcm) for benchmarks containing Gd. N is the number of benchmarks ^{1.03} 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.
- <u>Tsunami-2D</u> sequence with <u>ENDF/B-VII.0</u> evaluations.

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

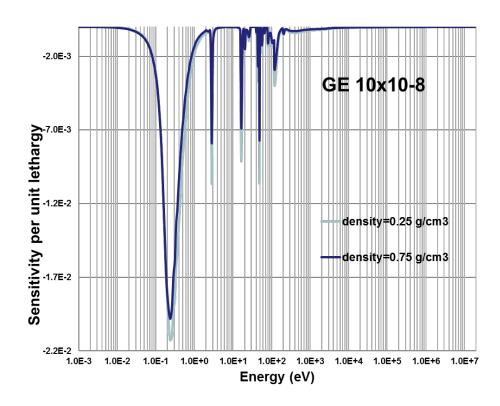
$$PB BWR \qquad GE 9x9-7 \qquad GE 10x10-8 \qquad EPR$$

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



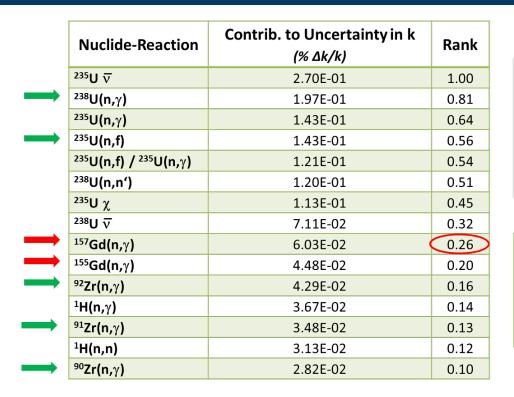
ENEA S/U Analysis

- 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



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.



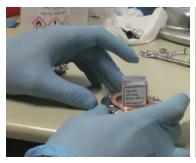
Measurement of the Gd Cross Sections

- ✓ Neutron capture measurements were performed by the time-of-flight technique using metallic Gd samples;
- ✓ The facility (CERN-nTOF) makes use of the spallation mechanism as a strong source of neutrons by using a proton beam impinging a lead target (1GeV/c -> ≈ 300 n);
- ✓ The source can be concentrated in short time pulses (≈7 ns rms) with a low duty cycle (0,5 Hz);
- ✓ Neutrons produced are canalized to an experimental area located ≈185 m downstream through a vacuum pipe to irradiate samples. Long flight path allows to gain high resolution in energy (10⁻³-10⁻⁴);
- ✓ Gamma capture measurements were performed by hydrogen-free deuteraded benzene (C6D6) detectors in combination with the Pulse Height Weighting Technique (PHWT).



155Gd – 157Gd Samples







- ✓ Samples were acquired by the nTOF collaboration from ORNL;
- √ 4 samples were isotopically enriched in either

 155Gd or 157Gd;
- ✓ The quantity of Gd in the samples results from a compromise between the need of the reducing the requested beam time and the optimization of the expected count rate in the resonance region;
- ✓ Since cross section changes by orders of magnitude as a function of the neutron energy, two highly enriched samples for each isotope were measured: a very thin one up to 100 meV, and a thicker one for cross section determination above 100 meV.



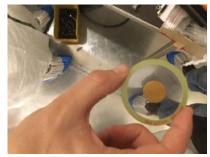
155Gd – 157Gd Samples

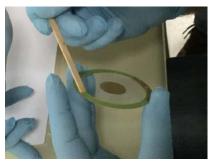
Isotope	Form	Geometry	Radius	Isotopic Purity [%]	Weight [mg]	Areal Density
¹⁵⁷ Gd	metallic	disc	1 cm	88.32	191.6±0.1	61.0 mg/cm ²
¹⁵⁷ Gd	metallic	disc	1 cm	88.32	4.7±0.1	1.5 mg/cm ²
¹⁵⁵ Gd	metallic	disc	1 cm	91.74	100.6±0.1	32 mg/cm ²
¹⁵⁵ Gd	metallic	disc	1 cm	91.74	10.0±0.1	3.2 mg/cm ²



Samples preparation









- ✓ Samples were sandwiched between two Mylar foils (to prevent oxidation) and centered avoiding any damage (they are extremely fragile and have to be handled with maximum care) in an annular frame to be correctly positioned along the line during irradiation;
- ✓ All samples had the same dimension in order to cover the same fraction of the neutron beam

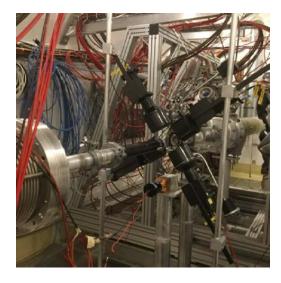


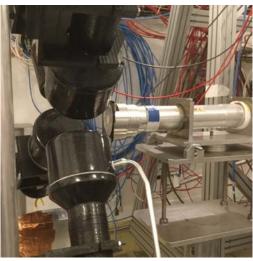
Experimental Campaign

- ✓ Measurements were performed from 17th June to 8th July 2016;
- ✓ In addition to the four Gd samples, a Gold sample, a Graphite sample and a Lead sample were used to study the background;
- ✓ Black resonance filters (Co, Ag, W, Cd) positioned along the flight path are used to determine the energy dependence of the background; they are chosen thick enough that the neutron beam is completely absorbed at the energies of the resonances;
- ✓ To validate the entire analysis procedure, 197Au(n, g) reaction cross section measurement was carried out with sample similar to Gd;
- ✓ Beam off measurements were carried out to characterize the room background;
- ✓ Energy calibration was performed using standard sources of ¹³⁷Cs, ⁸⁸Y, AmBe, CmC.



Experimental Setup





- ✓ For the detection of the prompt gamma rays resulting from the capture events, fast liquid scintillation detectors were used;
- ✓ The experimental setup for the measurements consisted of an array of four C6D6 scintillators opposite each other at 45 degree wrt the beam.

Details of time allocation for the experiments

	^	No Filters	With Filters		
Sample	#Protons	Running Time [days]	#Protons	Running Time [days]	
155 Gd thin	3.54E+17	4.00	-	-	
¹⁵⁵ Gd thick	3.29E+17	4.00	4.17E+16	0.50	
¹⁵⁷ Gd thin	3.96E+17	5.00	-	-	
157 Gd thick	4.12E+17	5.00	-	-	
Sample Out	1.71E+17	1.00	-	-	
Beam Off	ı	0.25	-	-	
Calibrations	-	0.87	-	-	
^{nat} Pb	5.75E+16	0.50	-	-	
¹⁹⁷ Au	3.63E+16	0.25	-	-	
^{nat} C	4.71E+16	0.20	-	-	
Total	1.81E+18	21.07	4.17E+16	0.50	



Further Experiments

A request for beamtime at the Gelina fa has been submitted in December 2016 JRC-Geel.

Moreover, if it proves possible, also the Research Reactor will be performed, ho



e samples sement from

Budapest

.



Future developments of the project

After the nTOF data analysis is completed, and after publication of the results on EXFOR, INFN (C. Massimi et al.) and ENEA will try to produce new evaluations for the xs and will initiate the validation process making recourse to the relevant ICSBEP benchmarks, in particular:

- ✓ LEU-COMP-THERM-005 (Pacific Northwest-BNFL, 16 cases)
- ✓ LEU-COMP-THERM-035 (JAERI TCA, 3 cases)
- ✓ HEU-SOL-THERM-014 (Institute of Physic and Power Engineering, 3 cases)

A possible completion timeframe could be envisaged at around Spring 2018.





























