

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH
Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron capture cross section for ^{155}Gd and ^{157}Gd for Nuclear Technology

June 1, 2015

S. Lo Meo^{1,2}, C. Massimi^{2,3}, F. Rocchi¹, N. Colonna⁴, M. Barbagallo⁴, E. Berthoumieux⁵, D. M. Castelluccio^{1,2}, L. Cosentino⁶, M. Diakaki⁵, R. Dressler⁸, E. Dupont⁵, P. Finocchiaro⁶, A. Guglielmelli¹, F. Gunsing^{5,7}, N. Kivel⁸, P. F. Mastinu⁹, M. Mastromarco⁴, P. M. Milazzo¹⁰, F. Mingrone², A. Musumarra^{6,11}, D. Schumann⁸, G. Tagliente⁴, G. Vannini^{2,3}, V. Variale⁴

¹*ENEA Research Centre E. Clementel, Bologna (Italy)*

²*INFN Section of Bologna, Bologna (Italy)*

³*Physics and Astronomy Dept. University of Bologna, Bologna (Italy)*

⁴*INFN Section of Bari, Bari (Italy)*

⁵*CEA, Saclay, Irfu/SPhN, Paris (France)*

⁶*INFN, Laboratori Nazionali del Sud, Catania (Italy)*

⁷*European Organization for Nuclear Research (CERN), Geneva (Switzerland)*

⁸*PSI, 5232 Villigen PSI (Switzerland)*

⁹*INFN, National Laboratory of Legnaro, Padova (Italy)*

¹⁰*INFN Section of Trieste, Trieste (Italy)*

¹¹*Physics Dept. University of Catania, Catania (Italy)*

Spokespersons:

S. Lo Meo (sergio.lomeo@enea.it), C. Massimi (cristian.massimi@bo.infn.it)

Technical coordinator: O. Aberle (oliver.aberle@cern.ch)

Abstract: We propose to measure the neutron capture cross-section of ^{155}Gd and ^{157}Gd 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_6D_6 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 highly-enriched 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 C_6D_6 detectors

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



1 Introduction and Scientific Motivations

Fuel assemblies (FAs) of current Light Water Reactors such as Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs) of 2nd and 3rd Generation make extensive use of the so-called “burnable neutron poisons” in various forms and with different technical solutions. Burnable poisons are chosen among isotopes characterized by a neutron capture cross section at thermal energy comparable or higher than the one of neutron-induced fission of ^{235}U , so to compete with ^{235}U in the absorption of thermal neutrons. Being their absorption parasitic for the neutron chain reaction, burnable poisons can compensate an higher initial amount of fissile material that would not be otherwise allowed for safety reasons, thus resulting in an extended life and improved efficiency of the nuclear fuel.

Several types and forms of burnable poisons have been successfully tested over the past decades. The most common one is gadolinia (Gd_2O_3) mixed directly within the UO_2 fuel matrix, taking advantage of the very large neutron capture cross section below 1 eV of two isotopes, the 155 and 157. The capture cross section of these two Gd isotopes is therefore extremely important in assessing the neutronic performances and safety features of FAs and of the whole core. The proper knowledge of these cross sections is not only important at the Beginning of Life (BOL) of a FA, but also during its life cycle; in fact accurate predictions of the burning rate of the two odd Gd isotopes is fundamental for predicting the appearance of the FA reactivity peak and its intensity.

In order to verify the prediction of Gd burnup and depletion in neutronic codes and the precision of the associated multigroup cross sections, a series of experiments using the Melusine research reactor in Grenoble have been performed by CEA. The most recent work on the comparison between experiment and calculations is that of 2015 by Bernard and Santamarina [1]. While the overall predictions on Gadolinium isotopes look quite good, still some non-negligible biases are found for ^{157}Gd , suggesting a possible underprediction of the capture cross section. A reliable prediction of the depletion of Gadolinium isotopes is also necessary to estimate the so-called “residual reactivity penalty”, an unavoidable effect associated to the high-burnup, equilibrium concentrations of odd and even isotopes; this parameter is important because if it is too high it can induce a limitation on the total amount of time a given FA can be used at full power. In order to achieve a good fuel management scheme, one must rely on accurate predictions of the depletion of Gd isotopes. As a rough example, the reactivity penalty due to 16 gadoliated pins with initial 8.0 w% of gadolinia in UO_2 for a 17×17 PWR FA (average ^{235}U enrichment of 4.5 w%) corresponds roughly to the “loss” of 5 full-power days per year [2]. In the electricity energy market of France, 5 full-power days of a III-Generation EPR reactor correspond roughly to 8 M€ [3].

The large neutron capture cross section of $^{\text{nat}}\text{Gd}$ is due to the odd stable isotopes ^{155}Gd and ^{157}Gd . The importance and role of these isotopes in the nuclear fuels has been assessed by means of a Sensitivity and Uncertainty (SU) analysis of several different FAs [4]. Calculations have been performed at BOL, hot-full power (HFP) conditions using the US-NRC reference SCALE 6.1 [5] code system developed at ORNL. The calculations have been performed using a 238-group cross section library based on ENDF/B-VII.0 [6] ; cross section uncertainties are taken into account by means of the covariance library available

within the code. The FA studied, as an example, is the General Electric 10×10-8 BWR (GE10×10-8). The sensitivity analysis [4] allows to estimate, for each fuel assembly, the contributions to the uncertainty in the multiplication factor k , of any quantity involved in the reactor operation.

Table 1 presents the first 15 most important contributions, in decreasing importance, to the uncertainty in k for the GE 10×10-8 FA at moderator density of 0.45 g/cm³. For a given quantity, the “Rank” is defined as the ratio between the contribution to the uncertainty on k of that quantity, and the one of the most important contributor which, therefore, has always rank = 1. It can be seen that the (n,γ) reaction of odd isotopes ¹⁵⁷Gd and ¹⁵⁵Gd rank between 0.26 and 0.20 with respect to the most important contributor, the average neutron multiplicity of ²³⁵U. Moreover, the neutron capture cross section of the two Gd isotopes are just after the ²³⁵U and ²³⁸U main physical quantities which are either already under assessment or not easily measurable at the n_TOF facility. It should be noted that other quantities in this list, like various Zr(n,γ) cross sections, have been successfully measured at n_TOF in recent years.

Nuclide/Reaction	Contributions to Uncertainty (% $\Delta k/k$)	Rank
²³⁵ U $\bar{\nu}$	0.270	1.00
²³⁸ U(n,γ)	0.197	0.81
²³⁵ U(n,γ)	0.143	0.64
²³⁵ U(n,f)	0.143	0.56
²³⁵ U(n,f) ÷ ²³⁵ U(n,γ)	0.121	0.54
²³⁸ U(n,n')	0.120	0.51
²³⁵ U ξ	0.113	0.45
²³⁸ U $\bar{\nu}$	0.0711	0.32
¹⁵⁷ Gd(n,γ)	0.0603	0.26
¹⁵⁵ Gd(n,γ)	0.0448	0.20
⁹² Zr(n,γ)	0.0429	0.16
¹ H(n,γ)	0.0367	0.14
⁹¹ Zr(n,γ)	0.0348	0.13
¹ H(n,el)	0.0313	0.12
⁹⁰ Zr(n,γ)	0.0282	0.10

Table 1: Contribution to overall uncertainty in k for the GE 10×01-8 FA: first 15 quantities

The sensitivity analysis of [4] indicates that the most important energy range for the ¹⁵⁷Gd(n,γ) reaction is between about 0.1 and 1 eV; the region of resolved resonances gives little contribution to the energy-integrated sensitivity, just because of the rather good moderation of neutrons in LWR systems. As a consequence, any improvement in the ¹⁵⁷Gd(n,γ) cross section in the low energy range, in particular between thermal and 1 eV neutron energy, can have a significant impact on the overall assessment of the neutronic properties of FAs using this neutron poison.

Another important application of Gd is related to its use in neutron capture therapy. While this element has been considered less efficient than ¹⁰B in delivering a lethal dose

to tumor cells, recently a new approach has been proposed, based on the use of a dual boron/Gd agent to improve the efficacy of Boron Neutron Capture Therapy in lung cancer treatment [7]. In this case as well, the cross sections of the two odd Gd isotopes are important for the dose prediction. Finally, neutron capture cross sections of $^{155,157}\text{Gd}$ isotopes are important for the development of innovative neutrino detectors [8]. Tagging neutrons by capture in Gd may in fact help in significantly reducing the backgrounds and increasing the signal sensitivity of present and future megaton-size Water Cerenkov detectors, like SuperKamiokande [9] and HyperKamiokande [10]. Gd salts dissolved in water increase the neutron capture cross-section and create a specific signature of an 8 MeV gamma cascade tens of microseconds after the neutrino nucleon interaction happens. As the visible energy in a water Cerenkov detector is about 4-5 MeV that signal is much easier to detect than the 2.2 MeV gamma cascade coming from the 10 times slower thermal neutron capture in pure water. As an example, anti-neutrino induced processes like $\bar{\nu}p \rightarrow \mu^+n$ may be discriminated from their neutrino counterparts by neutron tagging, without the need to magnetize the entire detector. A precise knowledge of the capture cross-sections and decay gamma distributions would have a significant impact on the capability of such experiments to precisely simulate the detector response and quantify the effect of Gd doping on their physics potential.

Despite the importance of this cross section, only few experimental data are available on these two isotopes in the energy region below the resolved resonances. For $^{157}\text{Gd}(n,\gamma)$, a single data-point was published in 1958 [11] for the thermal cross section, which was determined to be roughly 264 kb. In the same year, BNL-325 Report indicated a value of 240 kb for the same cross section [12], while in 1960 a second set of data was extracted from total cross section measurements [13], which gave a value of 254 kb. One has then to wait 2006 with the work [14] of Leinweber et al., in 2006, is as of today, the most complete dataset on the capture cross section of the two odd Gd isotopes. This measurement, while reducing the gaps between reactor experiments and calculations [15, 16], is however not fully able to resolve other reactor physics problems related to Gadolinium [17]. Even the measurement by Choi et al. [17] cannot be considered definitive; the authors state that accurate experimental studies are further needed to understand some differences with respect to other older determinations. Moreover the work of Ref. [17], based on the use of a natural Gadolinium target which might introduce biases in the experimental results, does not cover the 0.1 – 1 eV range which is similarly important in reactor physics than the thermal range. Other discrepancies revealed by reactor physics indicate that “the gadolinium neutron capture cross-section has been over-corrected, relative to previous evaluations, in a beta version of ENDF/B VII.1 [18], which incorporates the Leinweber data” [19]. All in all, it is evident that the n_TOF experiments should clarify all the situation, by providing an accurate cross section and solving the discrepancy between the recent results in the thermal energy region, while improving and extending to higher energy the Resolved Resonance Region.

2 Proposed experimental setup

We propose to measure with high-accuracy and high-resolution the neutron capture cross section of ^{155}Gd and ^{157}Gd between thermal and 1 MeV neutron energy. The measurement should be performed in the first experimental area, using an array of four optimized C_6D_6 liquid scintillator detectors [20] specifically developed for capture measurements at n_TOF, and taking advantage of the Pulse Height Weighting Technique. Since the cross section for both isotopes drops by several orders of magnitude going above 0.1 eV, a unique sample cannot be used to perform the measurement in the whole energy range. On the contrary, to avoid saturation of the capture yield, very thin samples should be used to measure the highest cross sections, around thermal energy, while above 1 eV much thicker samples can be used in order to obtain a good signal-to-background ratio, and to collect the necessary statistics in a reasonable time.

As in many of the n_TOF experiments, the availability and the preparation of suitable samples is rather important for the success of the measurement. Highly enriched samples (around 90%) are available from Oak Ridge National Laboratory, with a cross-contamination of the two isotopes of less than 1 % (the level of accuracy required). The effect of the even isotopes will be evaluated and subtracted, taking advantage of a dedicated measurement for Nuclear Astrophysics, being submitted to this Committee as a separate proposal. In addition to the 4 gadolinium samples, a gold sample will be used for normalization purposes, while a graphite and a lead sample will be used to study the background. All samples must have the same dimension, and will be prepared as disc of 1 cm radius. They will be characterized at PSI, in particular in terms of contaminations.

3 Beam time request

The gadolinium isotopes are characterized by a very high capture cross-section, as a consequence a favourable signal to background ratio is expected from the proposed measurement. Figure 1 shows the expected counting rate for mono-isotopic gadolinium samples: ^{155}Gd (10 mg and 100 mg) and ^{157}Gd (5 mg and 200 mg). The calculation is based on the capture cross section retrieved from ENDF/B-VII.0 [6] and considering a neutron irradiation corresponding to 5×10^{17} protons for each sample. In all cases the uncertainty due to counting statistics in the unresolved resonance region is better than 3% with 100 bins per energy decade. The mass of Gd results from a compromise between the need of reducing the requested beam time and the optimization of the expected count rate in the resonance region. Samples with a mass exceeding few hundred milligrams would result in a faster measurement, but would produce saturation of the experimental observable, i.e. the capture yield, thus preventing one from extracting information on the capture cross-section. A conservative value of 20% for the detection efficiency is assumed in the calculation shown in Figure 1 and a correction factor is applied in order to take into account the area of the samples, which is smaller than the dimension of the neutron beam, intercepting approximatively 65% of the beam intensity.

As in previous C_6D_6 measurements, the estimation of the different components of the background for the proposed measurement is based on $^{\text{nat}}\text{Pb}$ and ^{12}C measurement.

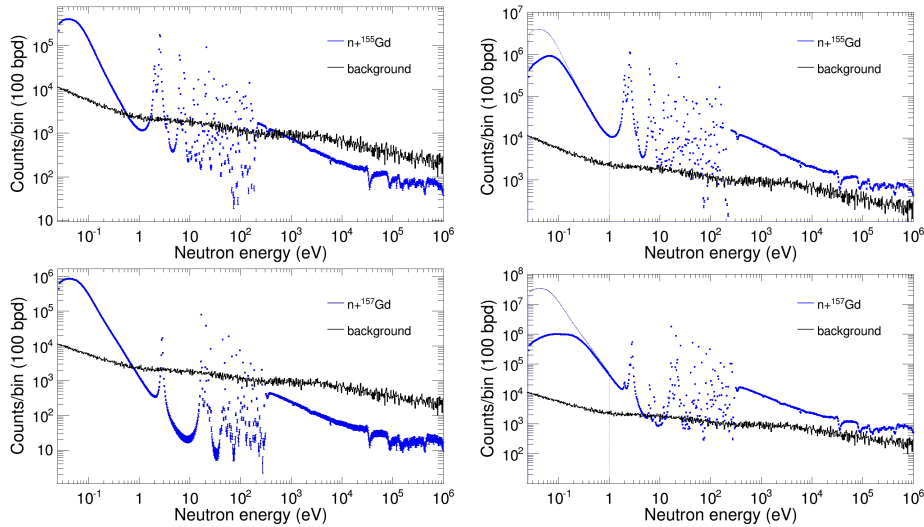


Figure 1: Expected counting rate for the (n, γ) reaction on ^{155}Gd (10 mg on the left and 100 mg on the right) and ^{157}Gd (5 mg, left, and 200 mg, right). In each calculation the neutron irradiation corresponds to an intensity of 5×10^{17} protons. The black line represents the expected background level during the measurement. For the thicker samples (right panels) the dashed curve represents the expected count rate without the self-shielding effects.

This background study requires a total number of 0.4×10^{18} protons. In addition, the normalization of capture data, the validation of the measurement at high energy and the cross-check of the flux stability is achieved by means of a periodic measurement of the Au sample. This further study requires a neutron fluence corresponding to 0.4×10^{18} protons. In summary the total proton request is 2.4×10^{18} protons on target.

4 Conclusion

We propose to measure the capture cross section of ^{155}Gd and ^{157}Gd isotopes relevant to Nuclear Technology in order to get very accurate cross sections and covariance values to improve the neutronic analyses of nuclear fuels. A series of measurements will be performed on enriched gadolinium samples, in the first experimental area, with an array of C_6D_6 detectors. In total 2.4×10^{18} protons are requested to successfully complete the measurement.

Summary of requested protons: 2.4×10^{18} protons on target.

References

- [1] D. Bernard, A. Santamarina, A. Qualification of gadolinium burnable poison: Interpretation of MELUSINE GEDEON-II spent fuel analysis. Ann. Nucl. Energy (2015), <http://dx.doi.org/10.1016/j.anucene.2015.02.034>, in print.

- [2] J. P. A. Renier, M. L. Grossbeck, Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors, ORNL-TM-2001/238, 2001.
- [3] French Law 1488, 2010: Nouvelle Organisation du Marché de l'Electricité.
- [4] F. Rocchi et al. ENEA Internal Report UTFISSM-PRONOC 2015
- [5] M. L. Williams and B. T Rearden, SCALE-6 Sensitivity/Uncertainty Methods and Covariance Data, Nuclear Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge TN 37831, USA
- [6] M.B. Chadwick, et al., Nuclear Data Sheet **107** (2006) 2931.
- [7] D. Alberti et al., A theranostic approach based on the use of a dual boron/Gd agent to improve the efficacy of Boron Neutron Capture Therapy in the lung cancer treatment. *Nanomedicine: Nanotechnology, Biology, and Medicine*, **11** (2015) 741.
- [8] E. Radicioni and G. Catanesi, private communication
- [9] S. Dazeley, A. Bernstein, N. S. Bowden and R. Svoboda, *Nucl. Instrum. Meth. A* **607** (2009) 616
- [10] K. Abe et al. [Hyper-Kamiokande Proto-Collaboration], Submitted to: *PTEP* (2015) [arXiv:1502.05199 [hep-ex]]
- [11] N. J. Pattenden, Some Neutron Cross Sections of Importance to Reactors ^{99}Tc , ^{143}Nd , ^{145}Nd , ^{149}Sm , ^{152}Sm , ^{151}Eu , ^{153}Eu , ^{155}Gd , ^{157}Gd , ^{240}Pu , Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva 1958, Neutron Cross Sections, Session A-11, P-11.
- [12] D. J. Hughes, R. B. Schwartz, Neutron Cross Sections, BNL-325, US Government Printing Office, July 1958.
- [13] H. Bjerrum Moller, F. J. Shore, V. L. Sailor, Low-Energy Neutron Resonances in Erbium and Gadolinium, *Nuclear Science and Engineering*, **8** (1960) 183-192.
- [14] G. Leinweber, D. P. Barry, M. J. Trbovich, J. A. Burke, N. J. Drindak, H. D. Knox, R. V. Ballad, R. C. Block, Y. Danon, L. I. Severnyak, Neutron Capture and Total Cross-Section Measurements and Resonance Parameters of Gadolinium, *Nuclear Science and Engineering*, **154**, (2006) 261-279.
- [15] G. Perret, M. F. Murphy, F. Jatuff, J-Ch. Sublet, O. Bouland, R. Chawla, Impact of New Gadolinium Cross Sections on Reaction Rate Distributions in 10×10 BWR Assemblies, *Nuclear Science and Engineering* **163** (2009) 17.
- [16] Jatuff F., Perret G., Murphy M., Grimm P., Seiler R., Chawla R., Impact of newly-measured gadolinium cross sections on BWR fuel rod reaction rate distributions, *PHYSOR 2008*, 14-19 September 2008, Interlaken, Switzerland, CD-ROM, 2008 (ISBN 978-3-9521409-5-6).

- [17] H. D. Choi, R. B. Firestone, M. S. Basunia, A. Hurst, B. Sleaford, N. Summers, J.E. Escher, Z. Revay, L. Szentmiklosi, T. Belgya, M. Krticka, Radiative Capture Cross Sections of $^{155,157}\text{Gd}$ for Thermal Neutrons, Nuclear Science and Engineering **177** (2014) 219.
- [18] <https://www-nds.iaea.org/exfor/endl.htm>
- [19] J.C. Chow, F.P. Adams, D. Roubstov, R.D. Singh, M.B. Zeller, Nuclear data and the effect of gadolinium in the moderator, AECL Nuclear Review, **1** (2012) 21. 10.12943/ANR.2012.00004.
- [20] P.F. Mastinu, et al., “New C_6D_6 detectors: reduced neutron sensitivity and improved safety” n_TOF-PUB-2013-002; CERN-n_TOF-PUB-2013-002 (2013)