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Measurement of the ^{160}Gd(n, γ) cross section at n_TOF and its medical implications

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Abstract. Neutron-capture reactions on gadolinium isotopes play an important role in several fields of physics, in particular in nuclear Astrophysics for the understanding of the nucleosynthesis of heavy elements (beyond iron) in stars via the s- and r-processes [1] and in nuclear technology. Another important application of gadolinium is linked to the production of terbium, that offers a set of clinically interesting isotopes for theranostics, characterized by complementary physical decay characteristics. In particular, the low-energy β^- emitter terbium-161 is very similar to lutetium-177 in terms of half-life (6.89 d), β^- energy and chemical properties. Being a significant emitter of conversion/Auger electrons, greater therapeutic effect can therefore be expected in comparison to Lu-177 [2, 3]. For this reason, in the last decade, the study of the neutron capture reaction ^{160}Gd(n,γ)^{161}Gd and the subsequent β^- decay in terbium-161 is getting particular attention. As the nuclear data on the Gd-160 neutron capture reaction are quite scarce and inconsistent, a new measurement of the capture cross section of Gd-160 at the CERN neutron Time-Of-Flight facilty was performed in order to provide high resolution, high-accuracy data on this important reaction, in the energy range from thermal to hundreds of keV. In this contribution, the preliminary results of the n_TOF measurement are presented.

1 Introduction

Terbium, member of the lanthanides row, offers a *poker* of clinically interesting isotopes for radionuclide therapy with complementary physical decay characteristics; among these, the low-energy β^- emitter Tb-161 is very similar to Lu-177 regarding the half-life (6.89 d), β-energy and chemical properties. Being a significant emitter of conversion/Auger electrons, greater therapeutic effect can therefore be expected in comparison to Lu-177 [2, 3]. Moreover, it also emits low-energy photons that are useful for diagnostic applications.

Among the possible ways of producing ^{161}Tb, the neutron capture reaction on ^{160}Gd followed by the β-decay of ^{161}Gd ($T_{1/2}$ = 3.66 m) in ^{161}Tb, is poorly explored. As of today, despite of its importance only a few experimental data sets exist at thermal neutron energy (25.3 meV) and in the Unresolved Resonance Region (URR), both characterized by discrepancies larger than 50%. In the Resolved Resonance Region (RRR) experimental data are completely missing. Discrepancies of the same order are reflected in the evaluations and main nuclear data libraries (ENDF/B-VIII.0, JEFF-3.3 and JENDL-4.0). Accordingly, large discrepancies are present for the values of Maxwellian Averaged Cross Section (MACS) retrieved from Karlsruhe Astrophysical Database of Nucleosynthesis in Stars (KADoNiS) database [4].

This unsatisfactory situation for medical and astrophysical applications triggered for a new measurement of the neutron capture cross-section of Gd-160 at the CERN neutron Time of Flight facility n_TOF. Thanks to the high neutron energy resolution of EAR-1 and to the very high neutron flux in EAR-2, capture data were collected from thermal neutron energy up to a few hundreds of keV.

In the present work the sample characteristics as well as the experimental setup in both areas followed by the preliminary results are shown.

2 Samples

The gadolinium sample, supplied by the Institut Luaue-Langevin (ILL) in Grenoble (France), consisted of 317 mg GdO_2 in powdered form enchiched to 98.10% in ^{160}Gd. The main characteristic of the sample is the very low presence of the odd-isotopes Gd-155 and Gd-157 whose very high cross section [5] would affect the measurement in the low neutron energy region. The detailed sample composition is reported in table 1. The very low presence of odd-A isotopes ^{155,157}Gd is the result of 54 days of irradiation at the ILL thermal reactor. The sample was enclosed in a 2-cm-diametr PEEK capsule, 2.1 mm-thick and closed by a thin Kapton foil. A post-measurement X-ray Spectroscopy indicated negligible sample inhomogeneity.

In addition, a ^{197}Au disk, a dummy-sample (i.e. a PEEK capsule without gadolinium oxide) and an empty-sample were used in order to normalize the data and estimate the background. All samples were glued onto a 6-μm-thick Mylar foil attached to an aluminium ring of 5 cm diameter as illustrated in figure 1 for the ^{160}Gd sample.

Table 1. Gd-160 enrichment: sample isotopic composition after 54 days irradiatian at the high neutron flux reactor of ILL resulted in Gd-155 and Gd-157 at trace levels.

Isotope	%
Gd-152	3.80×10^{-5}
Gd-153	3.90×10^{-10}
Gd-154	0.02
Gd-155	3.30×10^{-5}
Gd-156	0.59
Gd-157	4.20×10^{-6}
Gd-158	1.29
Gd-160	98.10

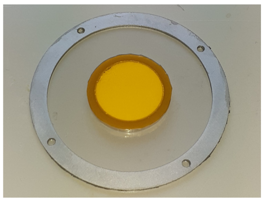

Figure 1. The Gd-160 sample in the PEEK container enclosed by the Kapton foil; the dummy-sample was made with the same procedure.

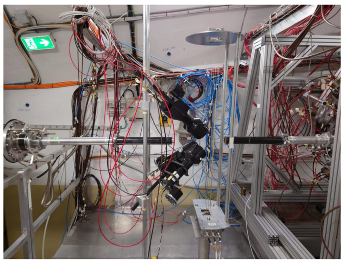

Figure 2. Experimental setup in EAR-1. Detectors placed at 125° relative the neutron direction.

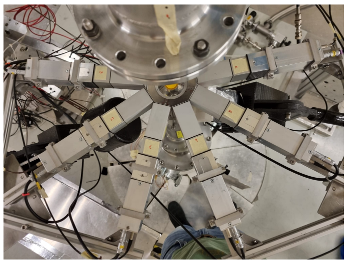

Figure 3. Experimental setup in EAR-2. Eight sTED scintillators are placed on the same plane at ≃ 5 cm from the sample, while the C_6D_6 are placed below at 125° relative to the vertical neutron direction.

3 The experimental setup

In order to extract the cross section in the neutron energy range spanning from thermal up to the Unresolved Resonances Region (URR), the neutron capture reaction measurement on ^{160}Gd was performed in both experimental areas of the neutron time-of-flight facility n_TOF at CERN.

At n_TOF neutrons are produced by spallation process on a lead target by 20 GeV/c protons coming from the CERN Proton Synchrotron. The initially fast neutrons are moderated and transported to the two experimental areas at the end of the flight paths 185 and 19 meter long, respectively. In both areas different arrays of deuterated benzene scintillators, namely C_6D_6 [6] and sTED [7] have been used. These detetection systems are characterized by low neutron-sensitivity and low γ-detection efficiency. The total-energy detection principle associated with the pulse height weighting technique (PHWT) ensures the proportionality between the detection efficiency and the corresponding energy of γ-rays coming from the cascade; see [8–10] for details.

Measurements started at the experimental area EAR-1. The incident proton-pulse coming from the Proton Syncrotron produces in EAR-1 an instantaneus flux of ≃ 10^6 neutrons/cm^2/proton-pulse spanning from thermal energies up to the GeV energy region. The main characteristics of the EAR-1 neutron flux is the high energy resolution $\Delta E_n/E_n \lesssim 3.0 \text{ x } 10^{-3}$ for $E_n \leq 100$ keV [11], which makes it particularly suitable for the study of resonant structures at high definition. As previously mentioned, the experimental setup consists of four deuterated benzene liquid scintillation detectors (C_6D_6) mounted at 125° relative to the neutron beam direction and at ≃ 10 cm upstream the samples position in order to minimize the effects of anisotropic emission (p-wave) of γ cascades (see figure 2).

To extract informations on the ^{160}Gd(n, γ)^{161}Gd reaction at energies below 1 eV, measurements were also performed at the high-flux experimental area (EAR-2) placed in the vertical direction at 19 m above the spallation target. Here, an enhanced signal/background ratio is ensured by shorter distance of EAR-2 from the lead target, the neutron beam has a 30 times higher flux ($\gtrsim 10^7$ neutrons/cm^2/proton-pulse) than in EAR-1 although with a lower resolution ($\Delta E_n/E_n = 4.0 \times 10^{-3}$ at 1 eV, $\Delta E_n/E_n = 4.0 \times 10^{-2}$ at 1 MeV) [12]. The detection setup in EAR-2 consisted of eight so-called Segmented Total Energy Detectors (sTED) and two C_6D_6. The sTED detectors, have been recently developed for (n, γ) measurements in EAR2; the smaller and segmented scintillation volume compared to conventional C_6D_6 detectors, reduce pile-up effects at high counting rate and the gain shift linked to the prompt γ-flash arising from the spallation process. The sTED scintillators, are placed at ≃ 5.0 cm from the sample in a plane perpendicular to the neutron beam direction as shown in figure 3; see [13] for details.

For each sample, data were collected in both experimental areas for a total number of protons available of 1.5×10^{18}. In the following the preliminary results are shown.

4 Results

A thorough analysis would require to use the total energy principle by combining the detection system with the Pulse Height Weighting Technique (PHWT) [8, 9]. Neverthe-

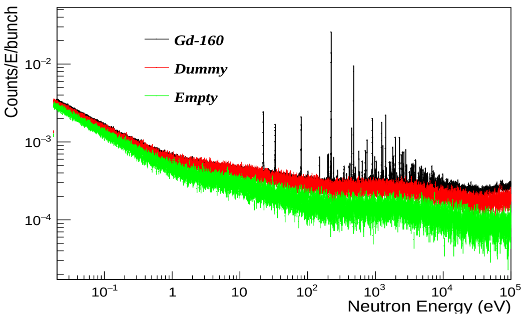

Figure 4. Counts per neutron-bunch as a function of the neutron energy for Gd-160 compared to the dummy and the empty sample in the whole neutron energy range.

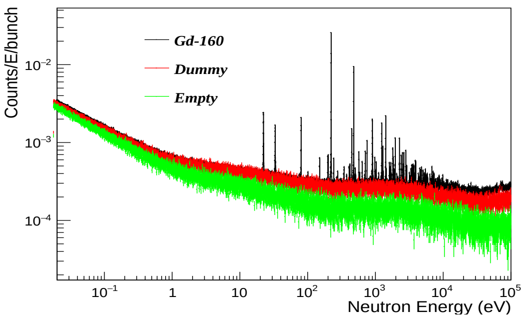

Figure 5. Count per neutron-bunch as a function of the neutron energy for Gd-160 compared to the dummy and the empty sample in the RRR; on average in the RRR Gd-160 related counts are 10% larger than dummy ones.

less, preliminary unweighted count distributions provide a good idea of the data-quality collected.

Figures 4, 5 and 6 show the counts distribution in the energy range of interest, measured in EAR-1. As expected the main background component arises from the PEEK capsule (see figures 4 and 5) whose contribution can be estimated with the dummy-sample measurement. Figure 6 shows the count rate for the Au and the empty in the range of Au saturated resonance at 4.9 eV; here, the main background source, estimated by the empty-sample is the beam-related background, although this has a quite negligible effect.

The count-rate for energies below 1 eV measured in EAR-2, is shown in figure 7. Also here the PEEK related background represents the largest contribution. Altogether, the count-rate related to neutron capture on Gd-160 is more than 15% larger compared to the main background.

5 Conclusions

The ^{160}Gd(n,γ)^{161}Gd reaction has been measured in the neutron energy range from thermal to a few hundreds of keV in both n_TOF experimental areas. Albeit an accurate analysis requires the use of PHWT, preliminary

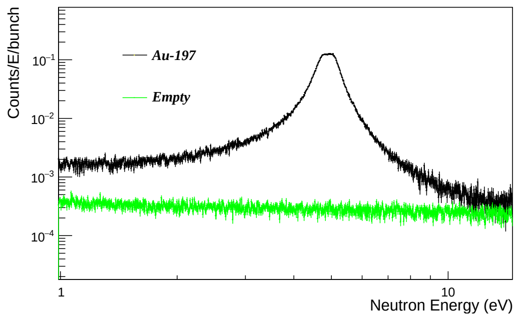

Figure 6. Au and empty count distribution in the range of the ^{197}Au satured resonance at 4.9 eV.

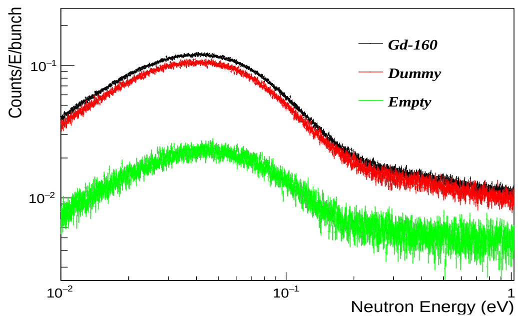

Figure 7. Count distribution of sample measured in EAR-2 for neutron energies below 1.0 eV.

results show good-data quality and an acceptable signal/background ratio.

For a precise determination of the cross section, the implementation of the setup geometry, the samples size and composition in Monte Carlo simulations will allow the precise determination of the Weighting Functions for the PHWT. We estimate that Resonance Shape Analysis (RSA) with the bayesian SAMMY code will provide resonance kernels with an accurancy better than 5%.

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