New setup for measurement of prompt gammas from neutron interactions

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Abstract. Prompt gamma radiation from neutron interactions is an important issue as it affects the operation of nuclear facilities (radiation protection or gamma heating issues) or has various uses (non-destructive identification of elements). Despite of importance, its production is inaccurately described in present nuclear data libraries. Therefore, a set of experiments have been carried out focusing on this radiation - prompt gamma from neutron inelastic scattering on oxygen and prompt gamma from radiative capture on manganese, both present the form of an aqueous solution. The gammas were induced by the Am-Be neutron source (for oxygen) and ^{252}Cf spontaneous fission neutron source (for manganese). In the case of oxygen, the present libraries can be used, but all the libraries tested overestimated the experiment by 20–30%, considering the experimental uncertainty of 6-9%. INDEN-4.0 library is giving the best results. In the case of manganese, none of the libraries tested give acceptable results. The INDEN updated data for gamma production on manganese give usable results, but there is still a discrepancy from -36% to 55% with uncertainties from 6-15%.

1 Introduction

A new technique has been prepared in Centrum Výzkumu Řež for the measurement of prompt capture and inelastic gammas coming from neutron interactions. These gammas are essential for safety during reactor operations (worker's protection or radiation damage[1]) or have applications in many areas like element identification in soil [2] and space [3]. Despite its importance, the prompt gamma production in present nuclear data libraries is generally discrepant [4]. Fortunately, many experimenters have recently focused on this area, with [5–7] being among the most important recent publications on the subject.

The new technique is simple. A spherical tank, made of aluminium or PMMA, is filled with an aqueous solution of studied elements, e.g., manganese sulphate (for the study of manganese). Pure heavy water was used as well for the study of inelastic scattering on oxygen.

2 Methods

All measurements were performed in the laboratory with dimensions of 1000 × 800 cm and a height of 720 cm, whereas the thickness of the walls and ceiling varies from 120 cm to 150 cm of iron concrete.

The gammas were measured using well-defined HPGe with known geometry, which allows calculating efficiency curve and enables measurement in arbitrary geometry. As the source was used ^{252}Cf(s.f), which is only neutron standard, and Am-Be neutron source with higher average energy, which allows studying gammas originating in higher energy threshold reactions.

2.1 Gamma from inelastic scattering on oxygen

The first experiment was focused on the measurement of prompt gamma with an energy of 6.128 MeV coming from inelastic scattering on oxygen. The aluminium sphere, having an outer diameter of 50 cm, was filled with heavy water. Then an Am-Be neutron source was placed into the dry channel. Its neutron emission, $7.08 \cdot 10^6 \pm 2.8\%$ n/s, was determined by the manganese bath technique using a bath used in the measurement of prompt gammas from neutron capture on manganese.

The sphere was placed on the table 2 m above the floor. Then it was surrounded by borated HDPE shielding to suppress neutron leakage from the sphere (to prevent detector damage). The leakage gamma spectrum was measured with the Canberra Big Mac HPGe detector and stilbene scintillator at two distances. The distances between the centre of the neutron source and the centre of the detectors were 101 cm and 194 cm. An illustration of the geometry can be found in Fig. 1.

HPGe detector was covered with cadmium shielding to suppress neutron interaction with the detector. The response of the HPGe detector was transformed to the gamma flux using a well-defined MCNP6.2 [8] detector model. Its details and validation can be found in [9].

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The stilbene scintillator was connected to the NGA-01 spectrometer. Gamma response was separated using pulse shape discrimination and the gamma spectrum was obtained using deconvolution using the Maximum Likelihood Estimation [10].

The 6.128 MeV gammas originate not only from inelastic scattering on ^{16}O but during the disintegration of ^{16}N as well. ^{16}N is produced in a high threshold reaction $^{16}O(n,p)^{16}N$. The ratio between $^{16}O(n,n')$ and $^{16}O(n,p)$ was determined by calculation and is about 1150. Due to such a low share, the contribution of ^{16}N to total gamma flux can be neglected.

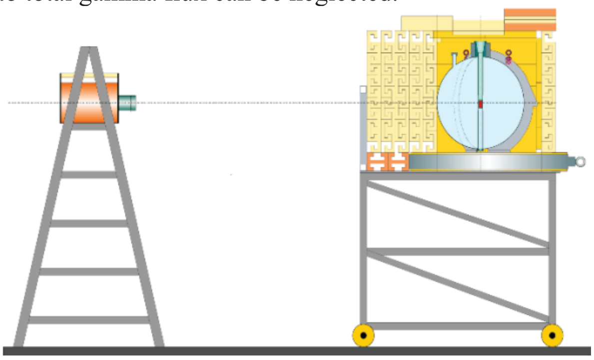

Fig. 1. Illustration of measurement setup with heavy water

2.2 Gamma from radiative capture on Mn

The second experiment was focused on the measurement of prompt gammas emitted during neutron radiative capture on manganese. ^{252}Cf spontaneous neutron source was inserted into the manganese bath, having a 34.55 cm outer radius [11]. This sphere was used in the determination of neutron emission of the used neutron sources. The bath was filled with the aqueous solution of manganese sulphate. Its composition was precisely determined in a chemical laboratory in Centrum Výzkumu Řež. The emission of the used ^{252}Cf neutron source during this experiment was determined to be $1.86 \cdot 10^6 \pm 2.8\%$ n/s.

The leakage gamma spectrum was measured with the same detectors as in heavy water measurement. The distance between the centre of the neutron source and the HPGe detector was 208.55 cm. The distance between the neutron source and stilbene was 108.55 cm.

3 Calculations

3.1 Oxygen inelastic scattering gamma

Simulation of the experiment was performed in the MCNP6.2 code. The spectrum of the Am-Be neutron source was taken from stilbene measurement [12]. As the spectrum was measured in the energy range 1-12 MeV, a correction of neutron emission below 1 MeV was necessary. The correction was taken from ISO 8529-1:2021 [13] as a fraction of the neutron spectrum above 1 MeV compared to the total neutron spectrum. As the ISO 8529-1:2021 only contains a spectrum for "small" and "large" AmBe neutron sources and the AmBe neutron source used in the experiment has dimensions approximately "medium" size, the average fraction for "small" and "large" spectrum was used. The value of this fraction is 0.858.

Several nuclear data libraries for neutron interactions were tested. The used libraries are ENDF/B-VIII.0 [14], JEFF-3.3 [15], JENDL-4.0 [16], and TENDL-2019 [17]. The data for photons were taken from MCPLIB04 [18].

3.2 Manganese radiative captures gammas

Simulation of the experiment with manganese bath was performed in MCNP6.2 as well. Standard ^{252}Cf spontaneous fission neutron spectrum [19] was used as a source spectrum.

ENDF/B-VIII.0 and JEFF-3.3 libraries were used for the simulation of neutrons, but the results contained an apparently discrepant gamma spectrum. It did not contain narrow peaks but very wide peaks. Due to that reason, a special simulation was performed. Thin (2 mm) manganese foil was irradiated with a flux of thermal neutrons (0.0253 eV), and the emitted gamma spectrum was scored in the sphere surrounding the foil. The result of this simulation is depicted in Fig. 2 for the most important energy region. It is apparent that the problem is in the gamma production data.

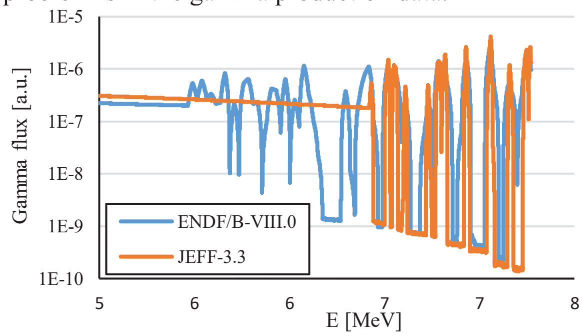

Fig. 2. Emitted gamma spectrum from thin Mn target

Further research revealed that the improved data for gamma production are available within the INDEN project [20]. These data are based on the ENDF/B-VIII.0 library but contain improved gamma production data. Details of this improvement can be found [20]. This library was tested as well; as can be seen in Fig. 3 and Fig. 4, the obtained peaks are still quite wide but significantly thinner than the original ENDF/B-VIII.0 library. Thus, the improved data were used in the simulation of the whole setup. The result of each gamma line was then subtracted from the continuum (the same approach is used to obtain Net Peak Area from HPGe measurements) and compared with the experiment.

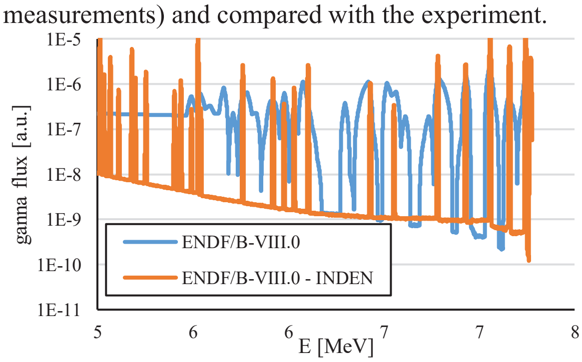

Fig. 3. Emitted gamma spectrum from thin Mn target

The same model was used for the simulation of stilbene response. The obtained spectrum was modified by the Gaussian broadening function using experimental data. Since the whole spectrum is to be compared, not just the net peak area as in the HPGe measurements, the ^{252}Cf gamma spectrum must also be included. The stilbene measured gamma spectrum of the ^{252}Cf source was used. It was normalized per one source neutron using stilbene determined ratio of photons above 1 MeV to neutrons above 1 MeV, which has a value of 0.432.

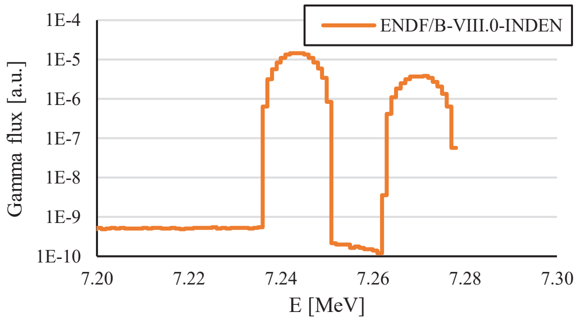

Fig. 4. Details on two most energetic peaks in gamma spectrum emitted by irradiation with thermal neutron

4 Results

4.1 Oxygen inelastic scattering gamma

The photon flux of 6.128 MeV peak from inelastic scattering of neutrons on oxygen was measured at a distance of 101 cm as 0.0907 cm^{-2}s^{-1} and a distance of 194 cm as 0.0259 cm^{-2}s^{-1}.

The results of the 6.128 MeV peak are listed in Table 1 and Table 2. The agreement between measurement and calculation is determined as the C/E-1 ratio (Calculation/Experiment - 1). The discrepancy is between 20 and 30% for all tested libraries for both distances. The best agreement is obtained with JENDL-4.0; other libraries give similar results.

Table 1. 6.128 MeV oxygen inelastic scattering gamma results for a distance of 101 cm

Neutron library	Calculated flux [cm^{-2}s^{-1}]	C/E-1	C/E uncertainty
ENDF/B-VIII.0	0.1185	30.6%	8.7%
JEFF-3.3	0.1198	32.0%	8.7%
JENDL-4.0	0.1083	19.4%	8.7%
TENDL-2019	0.1186	30.7%	8.7%

Table 2. 6.128 MeV oxygen inelastic scattering gamma results for a distance of 194 cm

Neutron library	Calculated flux [cm^{-2}s^{-1}]	C/E-1	C/E uncertainty
ENDF/B-VIII.0	0.0334	29.2%	5.9%
JEFF-3.3	0.0338	30.5%	5.9%
JENDL-4.0	0.0306	18.3%	5.9%
TENDL-2019	0.0334	29.3%	5.9%

The uncertainties in C/E-1 are 6% and 9% (including HPGe bias~ 2% [9], Net Peak Area uncertainty ~ 2–3%, geometry uncertainty ~ 2–4%, neutron spectrum uncertainty ~ 4%, neutron source emission ~ 2.8% and beam divergence uncertainty ~ 2.1–4.5%).

The comparison of stilbene measured and calculated gamma spectrum is depicted in Fig. 5. The agreement is good in the ^{12}C (4.439 MeV) peak, but peaks above 6 MeV, i.e., ^{16}O(n,n') (6.128 MeV) and neutron capture on structural materials such as aluminium (7.724 MeV), are indistinguishable from each other in the case of measurements. Measurements above 8 MeV also show a deconvolution problem at low count rates. The uncertainty in the stilbene measurement is 5–10%, in peaks up to 20%.

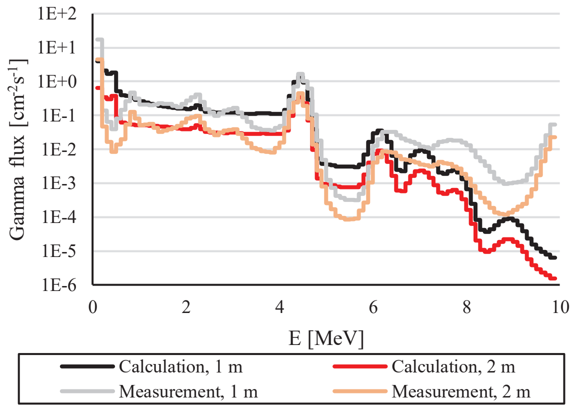

Fig. 5. Comparison of stilbene measured and calculated gamma spectrum from heavy water sphere

4.2 Manganese radiative captures gammas

The comparison of the most significant peaks calculated with INDEN update data is listed in Table 3. Other prompt gamma lines were because they were not found in the measured gamma spectrum.

Table 3. Comparison of measured and calculated peaks in gamma spectrum. Calculation obtained using ENDF/B-VIII.0 and INDEN data for manganese

E [MeV]	Calculated flux [cm^{-2}·s^{-1}]	Measured flux [cm^{-2}·s^{-1}]	C/E-1
3.409	3.40E-03	2.20E-03	54.3% ± 14.8%
4.725	4.42E-03	3.52E-03	25.6% ± 12.6%
5.015	1.21E-02	1.89E-02	-36.2% ± 6.4%
5.068	4.41E-03	4.34E-03	1.7% ± 13.1%
5.181	6.98E-03	5.40E-03	29.3% ± 11.4%
5.528	1.41E-02	1.29E-02	9.5% ± 7.0%
6.784	7.90E-03	6.35E-03	24.3% ± 11.4%
6.930	5.29E-03	4.86E-03	8.7% ± 14.0%
7.058	2.62E-02	2.28E-02	14.6% ± 6.2%
7.160	1.39E-02	1.27E-02	9.3% ± 6.5%
7.244	2.97E-02	2.47E-02	20.0% ± 5.9%
7.271	-	6.89E-03	

7.271 MeV peak was included in the calculated spectrum as well, but it is overlapped with the neighbouring peak, and their separation would be burdened with high uncertainties.

Uncertainties include Net Peak Area uncertainty (2-14%), uncertainty in the position of the detector (2%), manganese bath model uncertainty (3%), neutron emission uncertainty (2.8%), HPGe model uncertainty (2%), and beam divergence uncertainty (2%).

The measured and calculated stilbene spectra are depicted in Fig. 6. The Calculated spectrum includes stilbene measured natural gamma spectrum of ^{252}Cf. The agreement between measurement and calculation is good, but in most energy regions underestimation of calculation can be observed. The measurement uncertainties are around 10%.

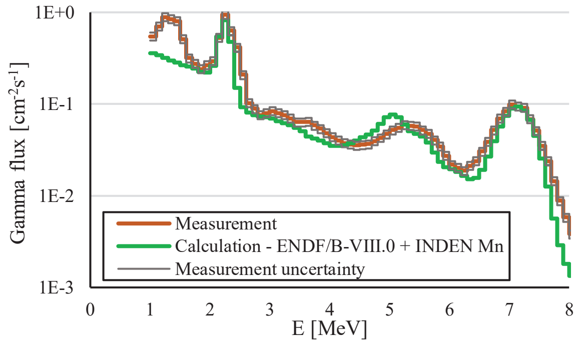

Fig. 6. Stilbene gamma leakage spectrum. The calculation was performed with ENDF/B-VIII.0 + INDEN data. Normalized to neutron source emission.

5 Conclusions

Two experiments on the measurement of prompt gamma radiation from neutron interactions were performed and reported in the paper. The experiment on inelastic scattering on oxygen has shown that the present nuclear data libraries give similar results, which overestimate the experiment by 20–30% with uncertainties of 6–9% depending on the distance of measurement. The best results were obtained with the JENDL-4.0 library.

The experiment on manganese radiative capture has revealed that the present nuclear libraries, namely ENDF/B-VIII.0 and JEFF-3.3, give significantly discrepant results. The INDEN improved data for manganese gamma production were tested as well. The results show significant improvement compared to the original data but still, the results contain wide peaks instead of narrow ones. The comparison of the most important peaks shows a C/E-1 ratio from -36% to 55%, with large differences in C/E-1 between different peaks.

It is planned to continue with a similar experiment focused on the prompt gamma measurement. The experiment on manganese is planned to be published as a benchmark.

Acknowledgments

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