Measurement of the neutron spectrum using the activation method

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Abstract

A new method for the measurement of the neutron spectra has been developed (Wieslander et al., 2010, Lövestam et al., 2009). The technique is based on the measurement of γ-rays from the neutron-induced activity in a series of different metal disks followed by spectrum unfolding procedures. In this study, we measure for the first time with this technique a prompt fission neutron spectrum. Special attention is paid to the unfolding procedures and the estimation of the lowest measurable neutron fluence rate, as well as the estimation of sources of the uncertainties.

Keywords: Prompt fission neutron spectra, activation measurement, unfolding technique, 252Cf neutron spectrum

1. Introduction

The understanding of the neutron emission in fission and the mechanism of the fission process are essential for safe and economic use of the nuclear power. The energy distribution of fission neutrons has a very strong influence on nuclear reactor parameters, such as k_{eff}. Because of that, accurate measurements of the prompt fission neutron spectra (PFNS) are very important. The PFNS can be measured in different types of experiments. An overview of such experiments performed at research centers worldwide, shows that a discrepancy between the integral and different measured data still exists today (Capote et al., 2012). The integral or macroscopic experiments are proposed for the measurement of average cross sections for different nuclear reactions, in a given neutron field (in this case the PFNS). The differential or microscopic
measurements are based on the counted fission neutrons by the detector with well-known efficiency and by using the time of flight method. The differential measured PFNS can be used for the calculation of the average cross sections for certain reactions and this data can then be compared with the integral measurement. It is interesting to analyze the existed calculated and measured data for $^{235}\text{U}$, which is one of the most important isotopes for fission applications. Up to now, many PFNS measurements of $^{235}\text{U}$ were carried out at different incident neutron energies, from thermal to the fast region. In Fig. 1 the ENDF-B/VII data showing the ratio of the calculated to the measured average cross section for the $^{235}\text{U}$ and $^{252}\text{Cf}$ PFNS, is presented (Capote et al., 2012). There is a good agreement between the calculated and measured data for $^{252}\text{Cf}$, but not in the case of the $^{235}\text{U}$ in the high-energy region. The nature of disagreement between the results of these two types of measurements for $^{235}\text{U}$ (showed in Fig. 1) is not yet explained properly. These were some of the reasons to come up with new measurements of the (PFNS). For this purpose we used an activation method called the DONA (DOsimetry and Spectrometry using Neutron Activation) developed at IRMM (Wieslander et al., 2010, Lövestam et al., 2009). This detector provides the possibility of determining the neutron energy spectrum by measuring the neutron induced radionuclide specific activity in certain metal disks and using the few channel spectrum unfolding technique to extract the neutron energy spectrum. The exact materials used for the DONA detector can be varied in many different ways. For this experiment it consisted of 9 circular metal disks of Ti, Ni, Fe, Co, In, Mg, Al, Zr and Au. In addition there was one Au-foil for the thermal plus epithermal neutrons and one Au-foil covered by Cd for the epithermal neutrons. For the chosen materials, the neutron activation functions for certain reactions with different energy thresholds are well known. The specific saturated activity induced by neutrons for the activation reaction $k$ is given by equation:

$$A_{ik} = \sum \sigma_{ik} \Phi_i$$

(1)

where $\sigma_{ik}$ is the corresponding activation excitation function and $\Phi_i$ is the neutron spectrum content in energy bin $i$. From eq. (1), the neutron fluence spectrum $\Phi_i$ can be determined by the unfolding procedure. For the unfolding procedure the few channels spectrum unfolded algorithms MAXED and GRAVEL are used (Reginatto and Goldhagen, 1999). These techniques start with an initial default spectrum. A-priori information, necessary for the construction of the initial guess spectrum, can be obtained from the available experimental or calculated data. This technique is applicable for measuring of the neutron energy spectra from 0.3 to 20 MeV. Advantages of the DONA detector are that it is practically only sensitive for the neutron radiation and requires no electrical power at the measurement site. However, with the help of the DONA detector, only constant neutron fields can be measured.

Fig. 1. Ratio of calculated results to experimental average cross-section for the $^{252}\text{Cf}$ and $^{235}\text{U}$ (Capote et al., 2012).

The first step in the measurement for the prompt fission spectra measurement, via the activation technique, was the validation of the DONA detector for these types of experiments. This was done by the measurement...
of the $^{252}$Cf spontaneous fission neutron spectrum. The $^{252}$Cf PFNS is very well-known and it is one of the spectra often used for the calibration of different detectors. There is also a very good agreement between integral and differential measured data for $^{252}$Cf. Special attention is paid to unfolding procedures and the estimation of the lowest measurable neutron fluence rate, as well as the estimation of possible source of the uncertainties. This analysis should provide the information about the applicability of the DONA detector for the measurement of the $^{235}$U PFNS.

2. Measurements

2.1 Activation of the DONA disks in the neutron field from a $^{252}$Cf source

The irradiation of two DONA rings was done at the IRMM. The source was placed between the two DONA rings. The distance between the $^{252}$Cf source and the DONA centres of the DONA rings was about 7.7 cm. The Californium source was inside the aluminum holder. It was estimated that the total neutron fluence rate at the place of the detector is around 200 n cm$^{-2}$ s$^{-1}$. The irradiation was done between the 9/7/2013 and 20/8/2013. Possible presence of contamination and residual activity in the disks, which have very high chemical purity, was checked using gamma-ray spectrometry measurements before the start of the irradiation. None of the gamma lines of interest (Table 1) were detected.

2.2 The gamma-ray spectrometry measurements

Due to the low neutron flux used for irradiation in this study, the DONA disks needed to be measured using ultra-low level gamma-ray spectrometry. The measurements were performed in the underground laboratory HADES (Andreotti et al., 2011). Eight HPGe-detector systems were employed for this purpose. The first measurement started 2 hours after the irradiation. Based on these gamma-ray measurements, the saturation activity $A_{sk}$ per atom of the activated isotopes at the end of activation was calculated for each detected radionuclide. The activity results with the general information about the activation reactions are presented in Tab. 1.

3. Results

3.1 Comparison of the measured and calculated data

The measured results from this study were compared with the EXFOR data. There is a very good agreement between the existed experimental and calculated values for the $^{252}$Cf neutron spectrum (Capote et al., 2012). The detected specific saturated activities (Table 1) were normalised to the data from the $^{115}$In(n,$n'$)$^{115m}$In reaction. Hence, the values present the average cross-sections for certain reactions normalised to $^{115}$In. The ENDF data for the average cross-sections were also normalised to the $^{115}$In(n,$n'$)$^{115m}$In reaction. This reaction is chosen since it is very well known, it has low threshold and there are no discrepancies between the experimental and calculated data (Capote et al., 2012). A comparison of the measured and calculated data is presented in Fig. 2. No satisfactory agreement is obtained for the $^{197}$Au(n,$\gamma$)$^{198}$Au, $^{54}$Fe(n,p)$^{54}$Mn, $^{24}$Mg(n,p)$^{24}$Na, $^{90}$Zr(n,2n)$^{92}$Zr and $^{90}$Zr(n,2n)$^{89}$Zr reactions. In the case of the $^{197}$Au(n,$\gamma$)$^{198}$Au reaction, the high detected activity may appear due to the presence of scattered neutrons, while for the $^{90}$Zr(n,2n)$^{89}$Zr reaction, the influence of the neutron capture on the $^{94}$Zr isotope to the detected gamma activity should be analysed. For high threshold reactions such as $^{90}$Zr(n,2n)$^{89}$Zr, also it shows unexpected low values for the C/E ratio. It should be mentioned that for this energy region (above 12.7 MeV) the $^{252}$Cf spectrum is
very low in intensity and also less determined. The disagreement for the $^{54}$Fe(n,p)$^{54}$Mn and $^{24}$Mg(n,p)$^{24}$Na reactions needs future investigation.

Table 1. General information about the activation reaction and the detected values of the specific activity.

| Activation reaction               | $<E>$ [MeV] | Half life | Main gamma-ray line [keV] | $A_s [10^{-24} \text{ Bq atom}^{-1}]$ |
|----------------------------------|-------------|-----------|---------------------------|-------------------------------------|
| $^{197}$Au(n,g)$^{199}$Au, foil  | 0.72        | 2.68 d    | 411                       | 26.2(25)                            |
| $^{197}$Au(n,g)$^{199}$Au, foil Cd shielded | 0.72        | 2.68 d    | 411                       | 26.3(25)                            |
| $^{197}$Au(n,2n)$^{198}$Au foil  | 10.5        | 6.18 d    | 333                       | 1.45(19)                            |
| $^{197}$Au(n,2n)$^{198}$Au foil Cd shielded | 10.5        | 6.18 d    | 333                       | 1.37(9)                             |
| $^{197}$Au(n,g)$^{199}$Au        | 0.72        | 2.68 d    | 411                       | 26.2(23)                            |
| $^{115}$In(n,n')$^{115}$In       | 2.67        | 4.49 h    | 336                       | 36.3(32)                            |
| $^{47}$Ti(n,p)$^{47}$Sc          | 3.82        | 3.35 d    | 159                       | 3.77(25)                            |
| $^{58}$Ni(n,p)$^{58}$Co          | 4.2         | 70.8 d    | 811                       | 20.1(9)                             |
| $^{54}$Fe(n,p)$^{54}$Mn          | 4.28        | 312 d     | 835                       | 11.1(8)                             |
| $^{59}$Co(n,p)$^{59}$Fe          | 5.94        | 44.5 d    | 1099                      | 0.314(20)                           |
| $^{46}$Ti(n,p)$^{46}$Sc          | 6.08        | 3.35 d    | 889                       | 2.70(18)                            |
| $^{24}$Mg(n,p)$^{24}$Na          | 8.26        | 15.0 h    | 1369                      | 0.257(17)                           |
| $^{48}$Ti(n,p)$^{48}$Sc          | 8.35        | 1.82 d    | 159                       | 0.083(5)                            |
| $^{27}$Al(n,a)$^{24}$Na          | 8.67        | 15.0 h    | 1369                      | 0.196(13)                           |
| $^{96}$Zr(n,2n)$^{94}$Zr         | 10.2        | 64.03 d   | 724                       | 8.78(63)                            |
| $^{197}$Au(n,2n)$^{199}$Au       | 10.5        | 6.18 d    | 333                       | 1.12(10)                            |
| $^{59}$Co(n,2n)$^{59}$Co         | 13.1        | 70.9 d    | 811                       | 0.078(5)                            |
| $^{90}$Zr(n,2n)$^{89}$Zr         | 14.4        | 3.27 d    | 909                       | 0.062(4)                            |

Fig. 2. Comparison of the measured data in this experiment and calculated from Capote et al., 2012.
3.2 The spectrum unfolding procedure

Based on the measured specific saturated activity and the known neutron excitation function for the detected reactions, the unfolded $^{252}\text{Cf}$ neutron spectrum was obtained by the unfolding procedure using the WinDONA software, developed at IRMM (Wieslander et al., 2010, Reginatto and Goldhagen, 1999, Goffe, 1996), which uses the MAXED and the GRAVEL unfolding algorithms. As an input initial guess spectrum we used the Maxwellian spectrum with $T = 1.42$ MeV, which is a very good approximation of the $^{252}\text{Cf}$ neutron spectrum. The obtained unfolding results, when all detected reactions were used, are presented in the left part of Fig. 3. As it can be seen, a satisfied shape of the neutron spectra was not obtained. Due to this, the problematic $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{54}\text{Fe}(n,p)^{54}\text{Mn}$, $^{24}\text{Mg}(n,p)^{24}\text{Na}$, $^{96}\text{Zr}(n,2n)^{95}\text{Zr}$ and $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ reactions were excluded from the unfolding procedure. In this case, a realistic shape of the neutron spectrum was determined, which is presented in Fig. 3 (right part).

One of the goals of this study was to test the accuracy of the unfolding procedure. We compared the obtained unfolding spectrum with a very well known Mannhart calculated $^{252}\text{Cf}$ neutron spectrum (Reich and Mannhart, 1989). The results presented in Fig. 4 (left part) show that starting from a Maxwellian spectrum it is possible to obtain more or less the same trend for the unfolded spectrum, as it is for a standard $^{252}\text{Cf}$ spectrum in the energy region higher than 5 MeV. However, we did not obtain a broad peak around 3 MeV, characteristic for the Mannhart spectrum. Moreover, the MAXED and GRAVEL codes give different results. As a default spectrum we also used the Mannhart calculated $^{252}\text{Cf}$ neutron spectrum. The obtained results this case, presented in the right part of Fig. 4, show a much better agreement between the MAXED and GRAVEL codes, as well as with the standard ENDF spectrum.

![Fig. 3. Left part: Unfolded neutron spectrum in case when all detected reactions were used; Right part: unfolded neutron spectrum after optimisation.](image)

4. Conclusion

The necessity for new measurements of the PFNS was the main reason for the analyses of possible measurements of the PFNS via activation technique using the DONA detector. It was done by the measurement of the californium spontaneous fission neutron spectrum.

The detected specific saturated activities for 9 used reactions show a very well agreement with the calculated ENDF data. However, the disagreement obtained for the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{54}\text{Fe}(n,p)^{54}\text{Mn}$, $^{24}\text{Mg}(n,p)^{24}\text{Na}$, $^{96}\text{Zr}(n,2n)^{95}\text{Zr}$ and $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ reactions needs future investigation. For that propose, the
Monte Carlo simulation of the DONA measurement with the $^{252}$Cf source can provide information about influence of the scattering neutrons to the detected gamma activity. This can approve the quality of the obtained data since by using the DONA detector it is not possible to distinguish between the prompt and the scattering neutrons.

Our analysis of the unfolding procedure shows that both used algorithms, the GRAVEL and the MAXED, provide the unfolded spectrum, which depends on the initial default spectrum. There are also observable differences in the results of the two the GRAVEL and MAXED unfolded spectra. The results show that the GRAVEL code provides a result, which has a slightly better agreement with the ERDF data. This is particularly the case when the Maxwellian spectrum is used as the input default spectrum. The disagreement with the ENDF standard spectrum (Reich and Mannhart, 1989) was also observed when the default spectrum was the ENDF standard spectrum itself. It should be emphasized that the used activation reactions (Tab. 1) do not have a satisfactory response for the energy region below 2 MeV, as well as for the energies higher than 14 MeV. This could be the reason for the observed effect of a relatively strong influence of the input to the unfolded data. However, the results show that by using the DONA detector, it is possible to obtain realistic general information about the PFNS.

Fig. 4. Left part: Comparison of the unfolded spectra with the standard ENDF $^{252}$Cf spectrum (Reich and Mannhart, 1989) in case of the Maxwellian default spectrum; Right part: Comparison of the unfolded spectra with the standard $^{252}$Cf ENDF spectrum (Reich and Mannhart, 1989) in case of the standard Mannhart calculated spectrum as default spectrum.

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