Available online at www.sciencedirect.com

ScienceDirect

Physics Procedia 64 (2015) 204 – 210

Physics Procedia

Scientific Workshop on Nuclear Fission dynamics and the Emission of Prompt Neutrons and Gamma Rays, THEORY-3

Modeling of neutron spectra based on activation analysis

N. Jovancevic[a,b]*, M. Fridman[c], L. Daraban[a], F.-J. Hambsch[a], S. Oberstedt[a], M. Hult[a], G. Lutter[a], G. Marissens[a], H. Stroh[a]

[a]European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retisewerg 111, 2440 Geel, Belgium
[b]Department of Physics, Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovica 3, 21000 Novi Sad, Serbia
[c]Recah Institute of Physics, Hebrew University of Jerusalem, Jerusalem 91904, Israel

Abstract

Safe and economical use of nuclear energy and particularly the development of GEN-IV reactors impose a better understanding of prompt neutron emission in fission, as well as of the fission process as such. Therefore, accurate measurements of the prompt fission neutron spectra (PFNS) are very important. In this work, we are testing the possibility to determine the PFNS by an activation method called DONA (DOsimetry and Spectroscopy using Neuron Activation) recently developed at IRMM (Wieslander et al., 2010, Lövestam et al., 2009). This type of modeling of the neutron spectra, based on the activation analysis, can provide new information about an old problem which still exists today, i.e. the discrepancy between measured integral and differential data (Capote et al. 2012). The problem is that the calculated average cross section for a certain neutron reaction, by using the differential experimental PFNS, in many cases cannot reproduce satisfactorily the integral measured cross section values. The modeling of the neutron spectra by the DONA technique was tested with the standard neutron spectrum of the spontaneous fission of ^{252}Cf. We analyzed the sensitivity of the unfolding procedure to the initial neutron energy spectrum, the influence of the neutron scattering, the possibility of using different activation reactions and we also made an estimation of the lowest measurable neutron fluence rate.

Peer-review under responsibility of the European Commission, Joint Research Centre – Institute for Reference Materials and Measurements

* Corresponding author. Tel.: +381214852815; fax: +381214852801.
*E-mail address:*nikola.jovancevic@df.uns.ac.rs

Peer-review under responsibility of the European Commission, Joint Research Centre – Institute for Reference Materials and Measurements
doi:10.1016/j.phpro.2015.04.028

Keywords: Prompt fission neutron spectra, activation measurement, unfolding technique, ^{252}Cf neutron spectrum;

1. Introduction

Reliable experimental data on the PFNS are necessary for improving the theoretical models, as well as the design of an innovative reactor, because the PFNS has a very strong influence on the nuclear reactor parameters, such as k_{eff}. The PFNS can be measured in different types of experiments. The existing experimental data show a discrepancy between the measured integral and differential data (Capote et al. 2012). The problem is that the calculated average cross section for certain neutron reactions, by using the differential experimental PFNS, in many cases cannot reproduce satisfactorily the integral measured cross section values. For example, this problem is presently not solved for ^{235}U, which is one of the most important isotopes in the nuclear energy applications (R. Capote 2014; N. Kornilov, 2011). Moreover, a recently published overview on the measured integral crosses section with the PFNS shows missing or not satisfactory data for some reactions (Capote et al. 2012). Also, the new evaluation of the PFNS is simultaneously included in the differential an integral experimental data. Because of that, we are testing the possibility to use the DONA activation method in the PFNS measurement. In this experiment, the PFNS should be determined, but generally, the DONA method can, at the same time, provide data about the integral cross section values with the PFNS for the considered nuclear reactions (Wieslander et al., 2010, Lövestam et al., 2009).

The DONA method (DOsimetry and Spectroscopy using Neuron Activation) recently developed at IRMM is based on the measurement of the neutron induced saturated gamma activity A_k , which is proportional to the product of the cross section for a certain reaction and the used neutron flux:

$$A_k \propto \sum_i \sigma_{ki} \Phi_i \qquad (1)$$

where σ_{ki} is the corresponding activation excitation function and Φ_i is the neutron spectrum content in energy bin *i*. Proceeding from this fact that a certain numbers of reactions with a very well-known neutron activation cross section are activated in the same neutron flux, and using an unfolding procedure, it is possible to obtain the neutron spectrum. Because of that, 9 circular metal disks (Ti, Ni, Fe, Co, In, Mg, Al, Zr and Au) and two Au foils are included in the DONA detector for measurement of the neutron spectrum. For the chosen materials, the neutron activation functions for certain reactions with different energy thresholds, are well-defined. In this work, we analyzed the sensitivity of the unfolding procedure to the initial neutron energy spectrum, the influence of the neutron scattering, the possibility of using different activation reactions and we also made an estimation of the lowest measurable neutron fluence rate. This analysis will be useful for the future planning of the PFNS activation measurements.

2. Measurements

2.1. Activation of the DONA disk

Two DONA rings with 11 disks in each ring, were activated at the IRMM in the neutron field from a ^{252}Cf source. The estimated total flux of neutrons at the place of the detector, was around 200 n cm^{-2} s^{-1}.The neutron source inside the aluminum holder was placed between the two DONA rings. The distance between the ^{252}Cf source and the DONA rings was about 7.7 cm. The activation was 43 days long. A possible presence of contamination and residual activity in the disks, which have very high chemical purity, was checked using the gamma-ray spectrometry measurements before the irradiation.

2.2. The gamma ray spectroscopy measurements

The measurements of the neutron induced gamma activity were carried out in the underground laboratory HADES (Andreotti et al., 2011) with eight HPGe-detector systems. These ultra-low level gamma-ray spectrometry systems were employed for this purpose since a very low level of gamma activity was expected due to the low neutron flux used for irradiation. The first measurement started 2 hours after the irradiation. Based on these gamma ray measurements, the saturation activity A_{sk} per atom at the end of activation was calculated for each detected radionuclide. The obtained results with the general information about the activation reactions are presented in Table 1.

Table 1. General information about the activation reaction and the detected values of the specific activity (<*E*> is the mean neutron energy of the integrated response of each reaction in the ^{252}Cf field).

Activation reaction	<E> [MeV]	Half life	Main gamma-ray line [keV]	A_{sk}[10^{-24} Bq atom^{-1}]
^{197}Au(n,g)^{198}Au, foil	0.72	2.68 d	411	26.2(25)
^{197}Au(n,g)^{198}Au, foil Cd shielded	0.72	2.68 d	411	26.3(25)
^{197}Au(n,2n)^{196}Au foil	10.5	6.18 d	333	1.45(19)
^{197}Au(n,2n)^{196}Au foil Cd shielded	10.5	6.18 d	333	1.37(9)
^{197}Au(n,g)^{198}Au	0.72	2.68 d	411	26.2(23)
^{115}In(n,n')^{115}In	2.67	4.49 h	336	38.3(32)
^{47}Ti(n,p)^{47}Sc	3.82	3.35 d	159	3.84(25)
^{58}Ni(n,p)^{58}Co	4.2	70.8 d	811	20.6(14)
^{54}Fe(n,p)^{54}Mn	4.28	312 d	835	18.2(12)
^{59}Co(n,p)^{59}Fe	5.94	44.5 d	1099	0.309(14)
^{46}Ti(n,p)^{46}Sc	6.08	3.35 d	889	2.81(18)
^{24}Mg(n,p)^{24}Na	8.26	15.0 h	1369	0.430(28)
^{48}Ti(n,p)^{48}Sc	8.35	1.82 d	159	0.083(3)
^{27}Al(n,a)^{24}Na	8.67	15.0 h	1369	0.224(15)
^{96}Zr(n,2n)^{95}Zr	10.2	64.03 d	724	8.82(64)
^{197}Au(n,2n)^{196}Au	10.5	6.18 d	333	1.20(10)
^{59}Co(n,2n)^{58}Co	13.1	70.9 d	811	0.079(5)
^{90}Zr(n,2n)^{89}Zr	14.4	3.27 d	909	0.064(4)

3. Results

3.1. The Monte Carlo simulation of the DONA detector response

The Monte Carlo simulation of the DONA detector response was done by taking it to account influence of the materials surrounding the detector set up. The results (Fig 1) show high discrepancy between the initial source spectrum and the one seen by the detector in the low energy region. A reason for that is the influence from scattering neutrons. Because of that in the future calculation have to be included a scattering factor corrections. This can improve the obtained results, especially in the case of ^{197}Au(n,g)^{198}Au reaction.

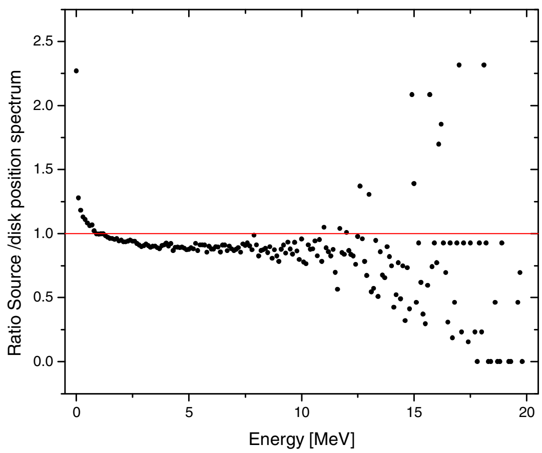


Fig. 1. Ratio of the neutron spectrum form the source and at the DONA detector position.

3.2. Comparison of the measured and calculated data

The verification of the measured gamma activity in this work was done by a comparison with the calculated reference data for the average neutron activation cross section (Reich and Mannhart, 1989). The analysis presented in this study (Reich and Mannhart, 1989) shows a very good agreement between the existing experimental and calculated values for the ^{252}Cf neutron spectrum for the used reactions, which can be considered as a standard reaction for this type of activation measurements.

A comparison of the measured and calculated data as a ratio C/E (calculated/experimental) is presented in Fig. 1. In this calculation, the detected specific saturated activities (Table.1) were normalised to the data from the ^{115}In(n,n')^{115m}In reaction. Hence, the obtained values present the average cross-sections for certain reactions normalised to ^{115}In. The reference calculated 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 a low threshold and there are no discrepancies between the existing experimental and calculated data (Reich and Mannhart, 1989).

The obtain results in Fig. 2. show a satisfactory agreement for the 11 reactions, but not for the $^{197}Au(n,\gamma)^{198}Au$, $^{96}Zr(n,2n)^{95}Zr$ and $^{90}Zr(n,2n)^{89}Zr$ reactions. The analysis of these discrepancies shows that 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. In order to correct the influence of scattered neutrons to the detected activity, it is necessary to obtain information from a Monte Carlo simulation. For the high threshold reactions $^{90}Zr(n,2n)^{89}Zr$, also an unexpected low values for the C/E ratio is observed. The analysis shows that the Zr disk was contaminated by Th and the detected gamma peak in the energy region around 910 keV comes from the standard background Th gamma lines of 911 keV, but not from the decay of ^{89}Zr. A higher value of gamma activity was also detected for the $^{96}Zr(n,2n)^{95}Zr$ reaction. The reason for that is the influence of the neutron capture on the ^{94}Zr isotope to the detected gamma activity of ^{95}Zr. Also it is necessary to note that this reaction is not a standard activation reaction. Because of that, the $^{90}Zr(n,2n)^{89}Zr$, $^{96}Zr(n,2n)^{95}Zr$ and $^{197}Au(n,\gamma)^{198}Au$ reactions were not used in the unfolding procedure for the modelling of the neutron spectrum.

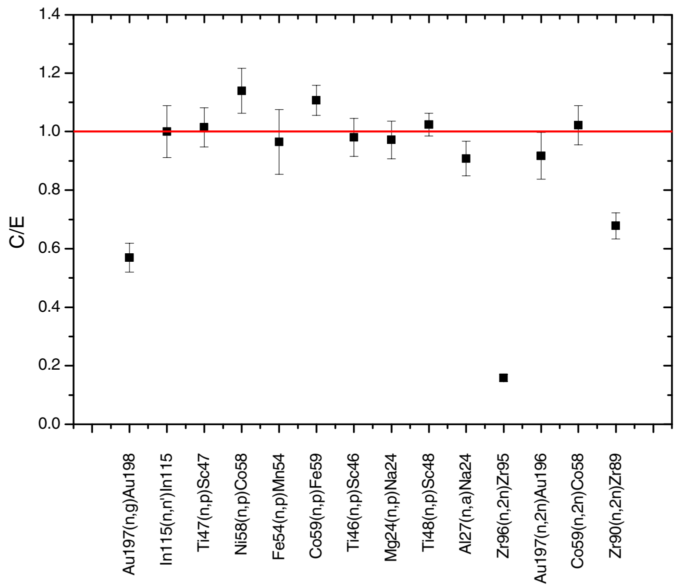
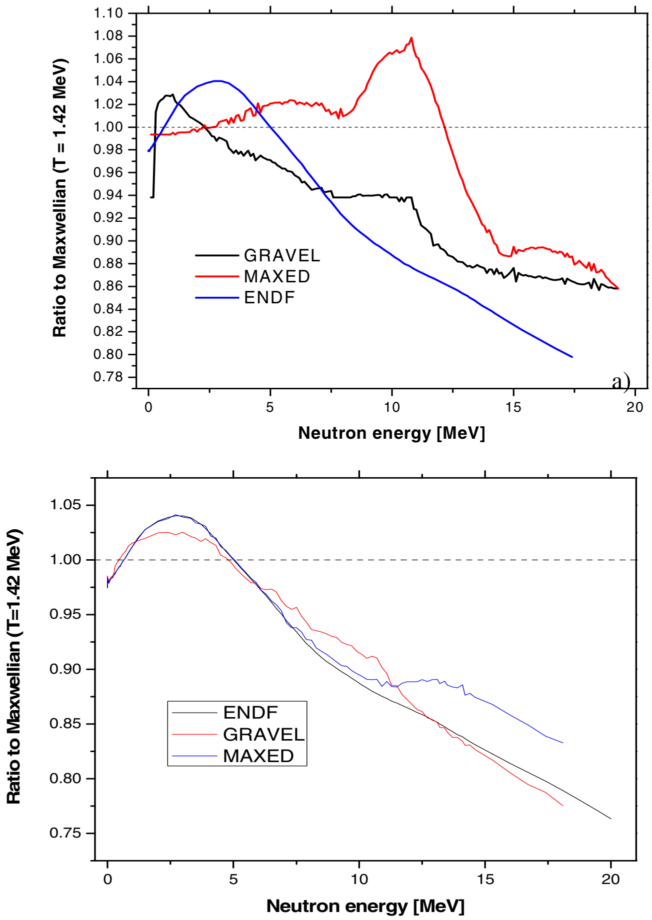


Fig. 2. Comparison of the measured data in this experiment and calculated from (Reich and Mannhart, 1989).

3.3. The spectrum unfolding procedure

The unfolded ^{252}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. The input data for the neutron spectra unfolding were the known neutron excitation function for the detected reactions, measured specific saturated activity and an initial guess neutron spectrum to start the unfolding algorithm. As an input guess spectrum we used a Maxwellian spectrum with T=1.42 MeV (which is a very good approximation of the ^{252}Cf neutron spectrum) and the well-known Mannhart calculated ^{252}Cf neutron spectrum (Reich and Mannhart, 1989). The obtained results are presented in Fig. 3. The results show that starting from a Maxwellian spectrum it is possible to obtain more or less the same trend for the unfolded spectrum and the standard ENDF spectrum by the GRAVEL algorithm. That is not the case for the

MAXED routine. However, in both cases we did not obtain a broad peak around 3 MeV, characteristic for the standard Mannhart ^{252}Cf spectrum. The determined neutron spectra with the Mannhart spectrum as default show a much better agreement between the MAXED and GRAVEL results, as well as with the standard ENDF spectrum (Reich and Mannhart, 1989; The ENDF). This demonstrated the high sensibility of the unfolding procedures on the initial guess spectrum.



b)

Fig. 3. Comparison of the unfolded spectra with the standard ENDF ^{252}Cf : (a) Maxwellian initial guess spectra; (b) the ENDF spectra (Reich and Mannhart, 1989; The ENDF).

4. Conclusion

In this work we tested the possibility of using the DONA method for obtaining realistic general information about the PFNS. The analysis stressed two main problems, the influence of the scattered neutrons and the dependence of the unfolding procedure on a initial guess spectrum. The measured activity and simulated spectra show very strong influence of scattering neutrons in the low energy region, below 1 MeV. The improvement of this technique, by taking into account the correction for the influence of the neutron scattering, should be part of a future investigation.

The testing of the unfolding technique by using two different algorithms shows the high sensitivity of the unfolding procedure on the initial guess spectrum. A diference in the final spectra provided by the MAXED and GRAVEL codes was observed. However, this disagreement becames less obvious in the case when an initial guess spectrum is closer to the real solution. This sugests the possibility of using the DONA method for testing the reliability of a different existing experimental PFNS data by using them as a first initial guess function.

The improvement of the DONA method requires a more precise measurement and the use of a source with a higher activity. This can provide more accurate data, especially in the high energy region. Also, it should be considered to use more activation reactions particularly sensitive in the energy region above 10 MeV and below 2 MeV. However, there are not a lot of standard activation reactions suitable for purpose. Some of the reactions which application should be analysed are ^{65}Cu(n,2n)^{64}Cu, ^{63}Cu(n,2n)^{62}Cu, ^{64}Zn(n,p)^{64}Cu, ^{32}S(n,p)^{32}P, as well as some of the neutron scattering reactions with the low energy treshold and a relatively high cross section in the energy region below 1 MeV (for example ^{103}Rh(n,n’)).

References

Andreotti E., Hult M., Gonzalez de Orduña R., Marissens G., Mihailescu M., Wätjen U. and Van Marcke P., Kiev, 7-12 June 2010, Publishing Department of KINR, Ukraine, 2011, p. 601-605.

Capote R., Trkov A., Sin M., Herman M., Daskalakis A., Damon, Y., Nuclear Data Sheets 118 (2014) 26-31.

Capote R., Zolotarev K. I., Pronyaev V. G., and Trkov A., 2012, PHYSOR 2012 – Advances in Reactor Physics – Linking Research, Industry, and Education, Knoxville, Tennessee, USA, April 15-20.

Goffe, W., 1996, Studies in Nonlear Dynamics & Econometrics, Vol. 1(3) (Berkeley Electronic Press), 169-176.

Kornilov N., Jornal of the Korean Physical Society, 59 (23) pp. 1717-1720.

Lövestam G., Hult M., Fessler A., Gasparro J., Kockerols P., Okkinga K., Tagziria H., Vanhavere F., Elisabeth Wieslander J. S., 2009, , Radiation measurements 44 pp 72-79.

Reginatto M., Goldhagen P., 1999, Health Phys. 77 p. 579.

Reich C.W., Mannhart W., and England T., 1989, IAEA Technical Report INDC(NDS)220/L, p.305, IAEA, Vienna.

The ENDF. Evaluated Nuclear Data File, https://www-nds.iaea.org/exfor/endf.htm

Wieslander E. J.S., Lövestam G., Hult M., Fessler A., Gasparro J., and Kockerols P., 2010, Radiation protection dosimetry, Vol. 138. No. 3, p. 205-212.