The Henryk Niewodniczański
INSTITUTE OF NUCLEAR PHYSICS
Polish Academy of Sciences
ul. Radzikowskiego 152, 31-342 Kraków, Poland

www.ifj.edu.pl/publ/reports/2013/

Kraków, February 2013

Report No. 2061/PN

Detection of delayed neutrons from neutron activation of fissionable substance samples. Monte Carlo modelling of response of the DET-12 device

Grzegorz Tracz[1]*, Barbara Bieńkowska*[2]*, Krzysztof Drozdowicz*[1]

[1]Institute of Nuclear Physics PAN, ul. Radzikowskiego 152, 31-342 Kraków, Poland
[2]Institute of Plasma Physics and Laser Microfusion, ul. Hery 23, 01-497 Warsaw, Poland

The work has been performed within the framework of the strategic research project "Technologies supporting development of safe nuclear power engineering" financed by the National Centre for Research and Development (NCBiR). Research task „Research and development of techniques for the controlled thermonuclear fusion", Contract No. SP/J/2/143234/11.

Abstract

Activation of fissionable elements by neutrons has been considered as a possible diagnostics of D-D and D-T fusion plasma. Fission caused by fusion neutrons leads up to emission of secondary neutrons: prompt and delayed. Proper interpretation of the time decay of delayed neutrons enables an assessment of the parameters of the primary neutron flux inducing fission. Monte Carlo calculations have been carried out by means of the MCNP code in order to elaborate the method considered. Three nuclides: pure ^{235}U, ^{238}U and ^{232}Th, and additionally sintered UO_2 have been selected as possible materials for the sample to be irradiated. Four energies of neutrons irradiating samples have been chosen: thermal, fast and two high ones. Computations have been accomplished for two variants: pure physical effect and real experimental conditions of neutron registration in the DET-12 device designed and built in IFJ PAN. Decay curves have been obtained for each case. Detection efficiency of DET-12 has been also estimated.

1. Introduction

Neutron activation method is principal for neutron-based techniques in the D-D or D-T fusion plasma diagnostics used at tokamaks and stellarators. It is applied to determine the neutron fluence or energy spectrum and based on recording of the products of induced reactions. Samples of selected materials of well known nuclear properties are placed at specific locations at or inside a fusion device and are irradiated with neutrons. Neutrons reaching the sample induce reactions which lead to creation of radioactive nuclei. Some isotopes of the samples have relatively high cross-sections for reactions with neutrons in chosen energy ranges. Usually selected reactions give the products decaying with gamma-ray emission, which can be easily detected using the gamma spectroscopy. Reactions leading to beta particle radiation are also used in the neutron activation method.

Another way is to use fissionable elements (U or Th, for instance) as the sample material. In such a case irradiation by neutrons causes fission, which leads up to emission of secondary neutrons. Most of them, called prompt neutrons, are emitted immediately (up to 10 fs) by a complex nuclei, while about 1% of particles, named delayed neutrons, is emitted by fission fractions later – from milliseconds up to even a few minutes after fission. Time resolved measurements of the delayed neutrons from the fissionable materials give the decay curve. Appropriate deconvolution of that curve enables to determine the parameters of the primary neutron flux inducing fission.

Recently a new device (named DET-12) for measurements of delayed neutrons decay was designed and constructed at the Institute of Nuclear Physics PAN in Kraków (Poland), Figs. 1 and 2.

Fig. 1. Built measuring chamber

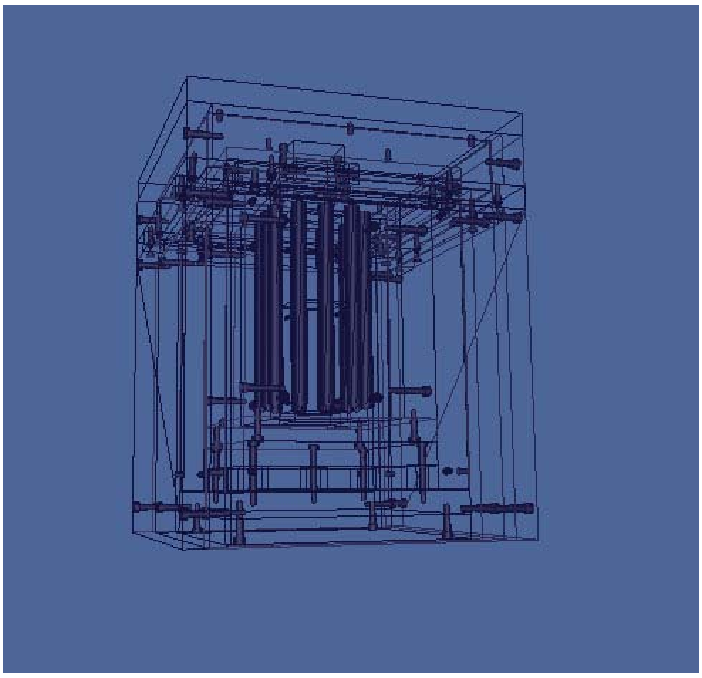

Fig. 2. View of positions of the neutron detectors inside the measuring chamber.

The fast delayed neutrons (~ 0.5 MeV) are slowed down in polyethylene layers and thermal neutrons are counted in ^{3}He detectors as a function of time. Details of the device and the data acquisition system were already described in [2] and [3].

2. Decay of delayed neutrons

Light elements posses usually the same number of neutrons and protons. Contrary to them, heavy elements are stable when the neutrons in a nucleus outnumber the protons. In order to achieve stability some products of fission emit neutrons. Such isotopes are called precursors of the delayed neutrons. Another way to equalize the number of protons and neutrons in the nuclei is the beta-decay.

The time decay of delayed neutrons from an individual isotope can be depicted as a sum of exponentials, characterized by decay-constants of its precursors:

$$S(t) = r_f \sum_{i=1}^{N} a_i \lambda_i \exp(-\lambda_i t)$$

where r_f is the number of fission reactions in the sample, N is the number of delayed neutron time groups, a_i is the abundance of the i-th group and λ_i is the decay-constant of the corresponding group. Among variety of possible mathematical representations of the delayed neutrons decay a six-group model has been commonly used, where the values of both the abundances and the decay-constants are free parameters, *i.e.*, calculated by the least square fit of six exponentials. A new concept was presented at an international workshop in Obninsk (Russia) in 1997. It had been proved that only a dozen or so precursors are involved in emission of over 80 % of delayed neutrons. Because of that, two eight-group models were proposed with fixed half-lives for any isotope [1]. Piksaikin's abundance weighted half-lives were chosen since they ensure a better fit. The values in question are presented in Table 1. The values of a_i are isotope dependent and change with energy of neutrons incident a fissionable material both in the free six-group and the constrained eight-group model [1]. Three initial neutron energies are customary used: thermal (0.0253 eV), fast (500 keV) and high (14 MeV).

Table 1. Piksaikin's half-lives for eight-group model [1].

group No.	precursor	$T_{1/2}$ (s)	abundance	group average $T_{1/2}$ (s)
1	^{87}Br	55.6	0.033	55.6
2	^{137}I	24.5	0.178	24.5
3	^{88}Br	16.3	0.111	16.3
4	^{138}I	6.46	0.046	5.21
	^{93}Rb	5.93	0.024	
	^{89}Br	4.35	0.101	
5	^{94}Rb	2.76	0.162	2.37
	^{139}I	2.30	0.046	
	^{85}As	2.08	0.107	
	^{98m}Y	2.00	0.088	
6	^{93}Kr	1.29	0.0048	1.04
	^{144}Cs	1.00	0.0070	
	^{140}I	0.86	0.0052	
7	^{91}Br	0.542	0.017	0.424
	^{95}Rb	0.378	0.049	
8	^{96}Rb	0.203	0.017	0.195
	^{97}Rb	0.170	0.0052	

3. DET-12 model for MCNP

In order to prepare a DET-12 Monte Carlo model, the device was modelled with the CAD program according to the technical design. Then the CAD model of DET-12 was exported to the "sat" format in order to covert it with the MCNP Visual Editor. The created file is a geometrical part of the MCNP input file. Usually modelling of geometry is the most laborious stage when preparing the input file. Unfortunately, the MCNP Visual Editor creates many identical, useless surfaces, which were removed manually and definitions of corresponding cells changed. Obviously, it is possible to run MCNP when the input file contains the same surfaces since the code deletes them automatically, but when their number is too large the geometrical structure becomes imperceptible for the user and it is difficult to introduce possible changes.

4. MCNP calculations

Prior to experiments, Monte Carlo calculations were performed to examine some aspects of proposed method. Three nuclides, pure ^{235}U, ^{238}U and ^{232}Th, were selected as possible materials of the irradiated sample. Additionally, sintered UO_2 pellet was modelled (^{234}U –

0.002%, ^{235}U – 0.316 %, ^{236}U – 0.014 %, ^{238}U – 99.668 %, bulk density $\rho = 4.74$ g·cm^{-3}).

The following energies of neutrons from tokamaks and stellarators can be considered: 14 MeV neutrons from the D-T reaction, 2.45 MeV neutrons from D-D reaction, and many of lower energy neutrons (scattered, slowed down, and thermalized in a construction and surrounding materials). For calculations the following energies of neutrons irradiating samples were chosen:

- thermal (Maxwell distribution at 293 K),
- fast 500 keV (or 600 keV in the case of ^{232}Th),
- and two high ones (2.45 and 14 MeV)

The calculations were performed for two arrangements. The first one was artificial: the sample in vacuum was irradiated by a mono-directional beam of neutrons (Fig. 3) and time-decay of neutron flux exiting the sample was calculated. In the second case some essential conditions of the real experiment were attempted to maintain. The fissionable sample was irradiated inside the device by the mono-directional beam of neutrons from the planar source placed below it (Fig. 4). The sample had always the same shape and dimensions: a cylinder of $h = 2r = 1.8$ cm.

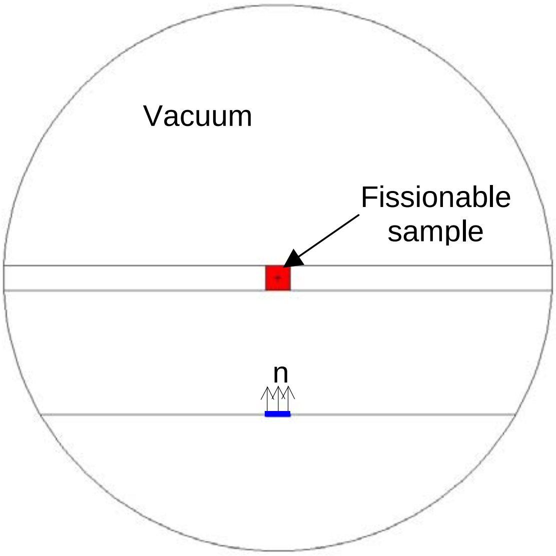

Fig. 3. MCNP model of the sample in vacuum. Neutrons are emitted monodirectionally from the blue segment.

All the computer simulations were carried out by means of the MCNP code [4]. Concerning the tablet materials, all cross-sections employed in the foregoing calculations were derived from the ENDF/B-VI library. Fission cross-sections for ^{235}U, ^{238}U and ^{232}Th at temperature of 300 K are presented in Fig. 5. The thorium sample was not irradiated with thermal neutrons. The ^{232}Th fission cross-section begins exactly at 500 keV and is on the order of 10^{-9} b while at 600 keV on the order of 10^{-5} b. It was decided to use 600 keV neutrons instead of 500 keV to irradiate the thorium samples in order to enhance reaction rate since such a small energy enlargement should not affect the decay of delayed neutrons.

The calculations were performed on the McRadiat computer cluster at IFJ PAN. The number of simulated histories in each case was $2 \cdot 10^{11}$.

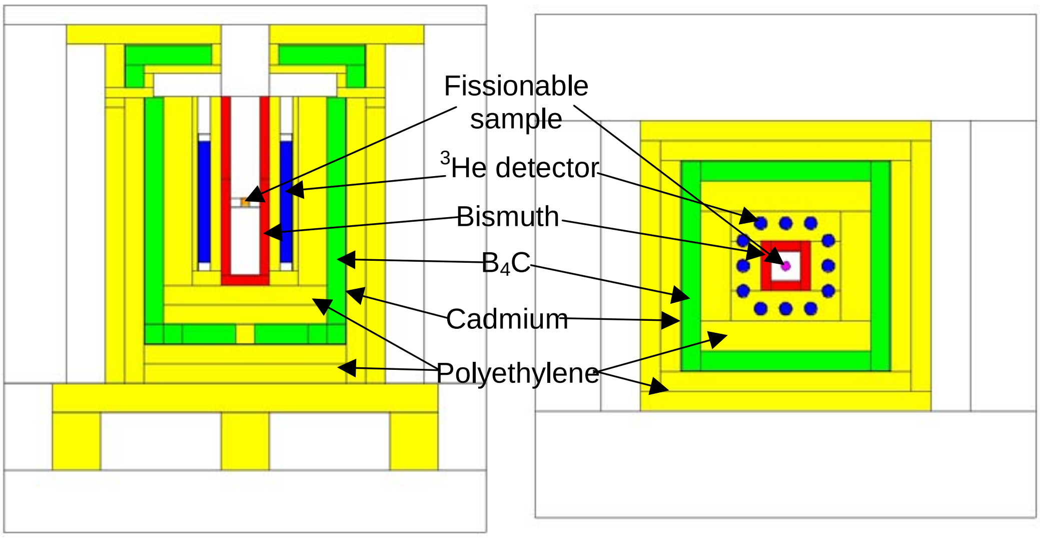

Fig. 4. MCNP model of the DET-12 device (vertical and horizontal cross sections). Helium detectors are drawn in navy blue. The sample is situated in the centre.

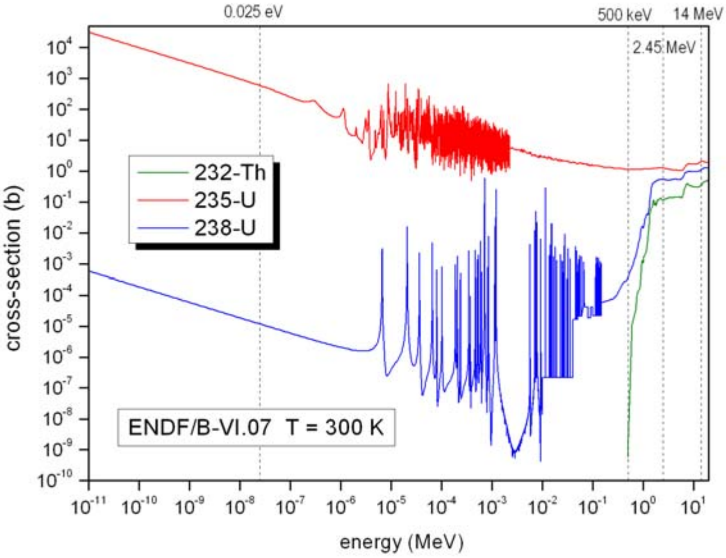

Fig. 5. Fission cross-sections for ^{235}U, ^{238}U and ^{232}Th from ENDF/B-VI.07 library at 300 K.

5. Results

5.1. Activation of the sample tablets and emission of the delayed neutrons

As said above, in the beginning pure physical effects were Monte Carlo modelled. The sample material tablets were irradiated by the primary neutron flux and the time distributions of delayed neutron fluxes Φ(t) emitted from the tablets were stored. The results $\Phi_1(t)$ normalized per one source neutron are shown in Figs 6–9.

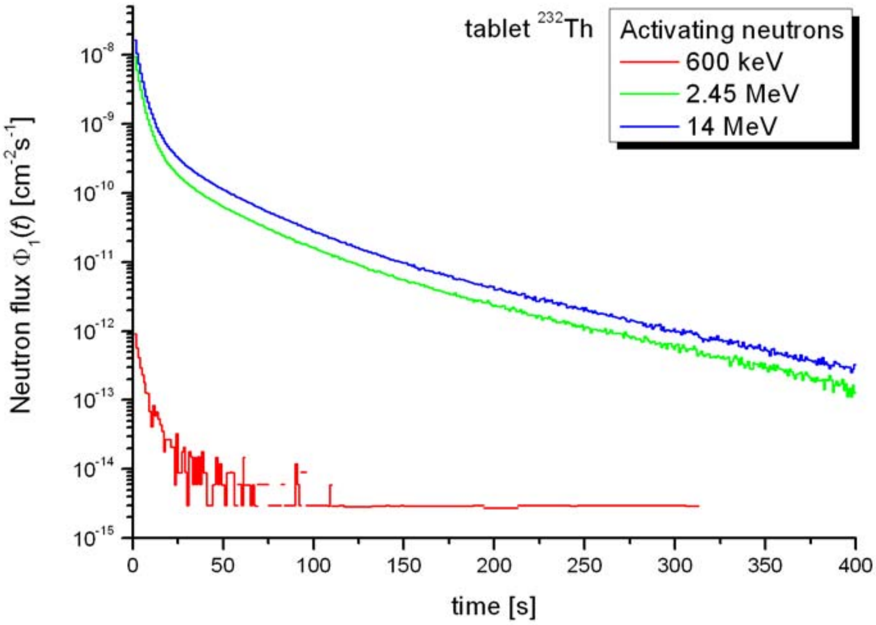

Fig. 6. Time distributions of delayed neutrons from the ^{232}Th tablet.

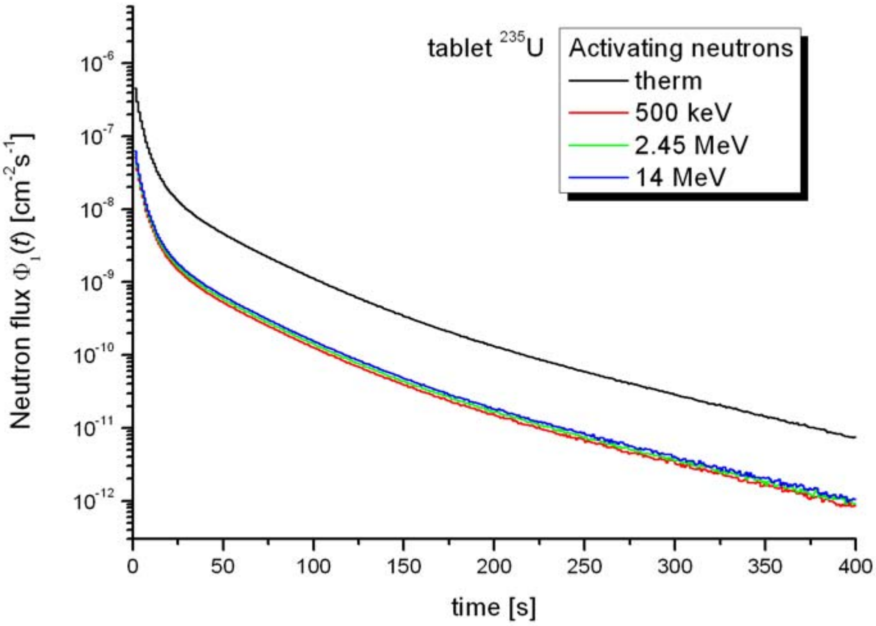

Fig. 7. Time distributions of delayed neutrons from the ^{235}U tablet.

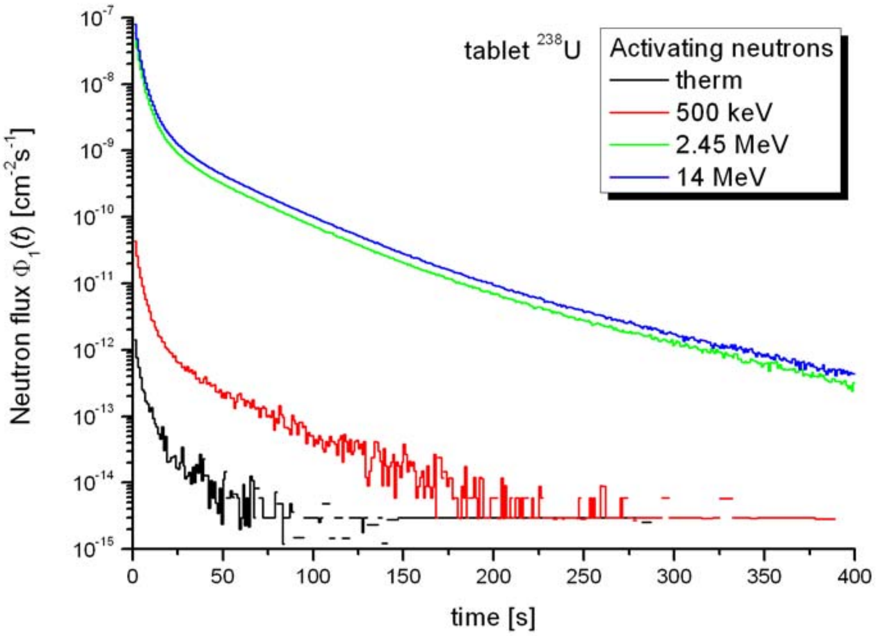

Fig. 8. Time distributions of delayed neutrons from the ^{238}U tablet.

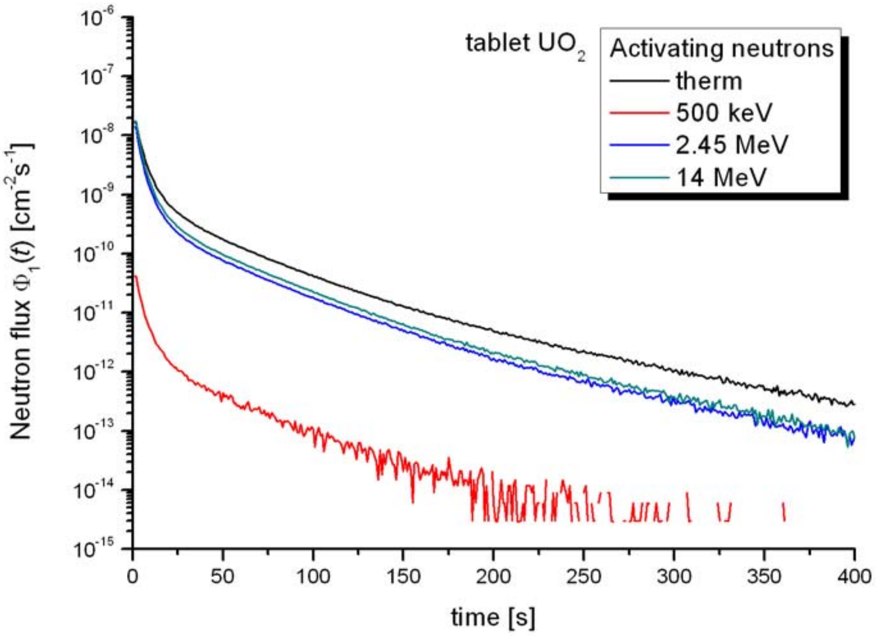

Fig. 9. Time distributions of delayed neutrons from the UO$_2$ tablet.

5.2. Emission of delayed neutrons from sample tablets and detection in DET-12

The second part of the MC simulations was carried out to investigate the response of DET-12 for the delayed neutrons emitted from the activated tablets. The resulting count rates $J_1(t)$ are shown in Figs. 10–13. They are also normalized per one source neutron.

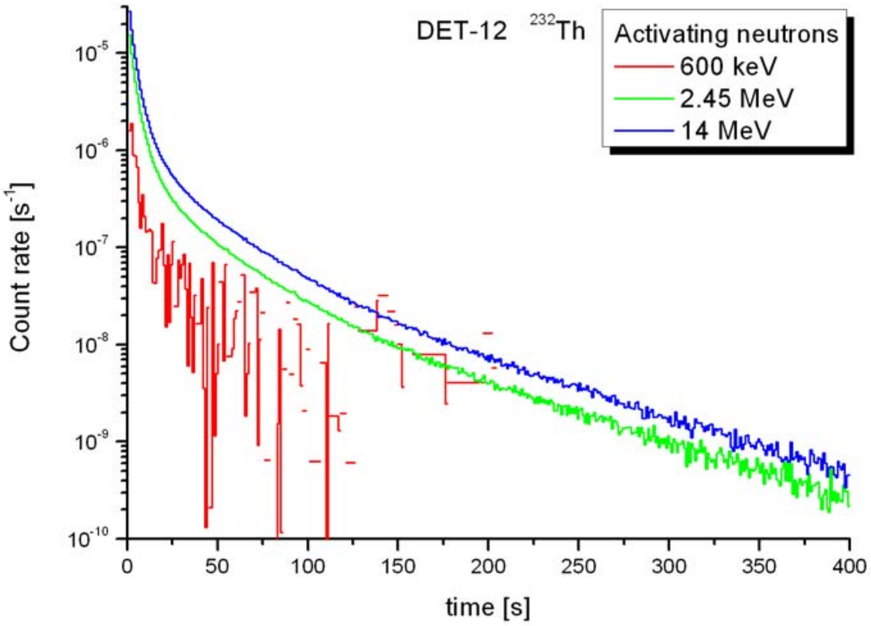

Fig. 10. Time distributions of the response of DET-12 for the delayed neutrons emitted from the activated ^{232}Th tablet.

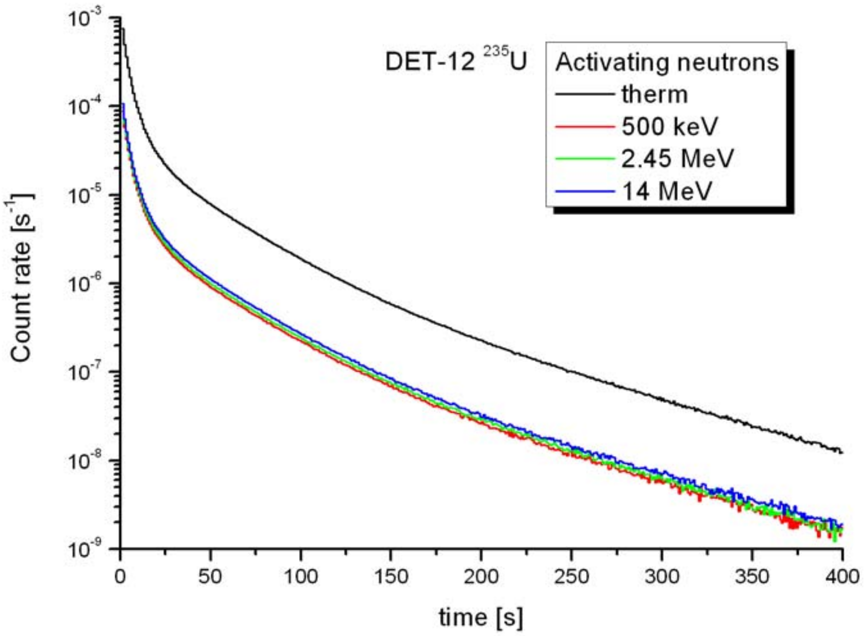

Fig. 11. Time distributions of the response of DET-12 for the delayed neutrons emitted from the activated ^{235}U tablet.

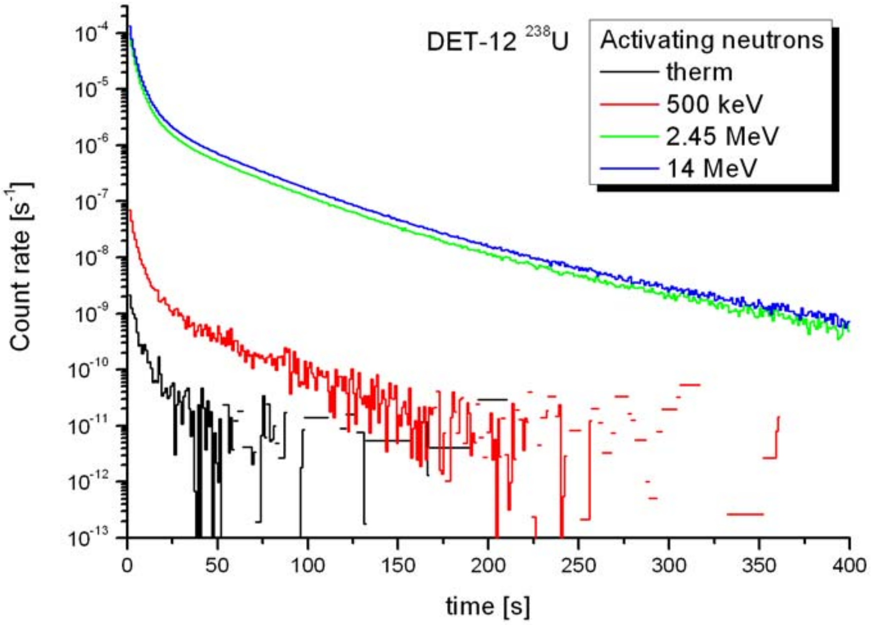

Fig. 12. Time distributions of the response of DET-12 for the delayed neutrons emitted from the activated ^{238}U tablet.

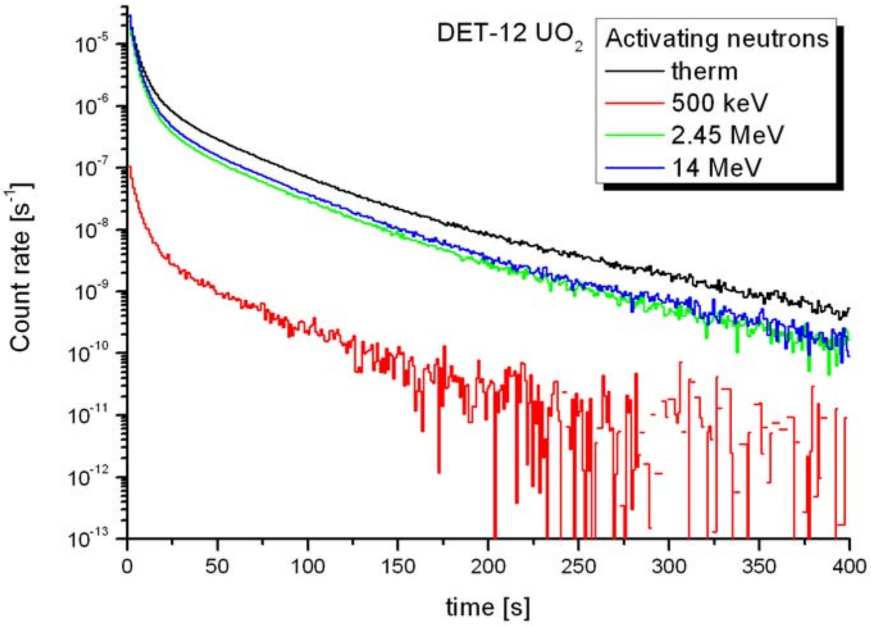

Fig. 13. Time distributions of the response of DET-12 for the delayed neutrons emitted from the activated UO_2 tablet.

5.3. Efficiency and reliability of delayed neutron detection in DET-12

Two problems are to be considered:

- a detection efficiency (understood as a number of counts in the detector set per neutron flux emitted from the activated tablet),
- possibility of distortion of the recorded die away curve in comparison to the shape of this curve for neutrons emitted from the tablet.

For this purpose the ratios of the time distributions of count rates $J_1(t)$ in DET-12 to the time distributions of the fluxes $\Phi_1(t)$ of delayed neutrons emitted from the tablets were calculated. The results $R = J_1(t)/\Phi_1(t)$ are presented in Figs. 14–17. As it can be seen from the previous calculations, for some energies the statistics is poor. For those energies the ratios are not taken into consideration. Horizontal straight lines indicate mean values of the ratio calculated over the corresponding time interval. All the calculated mean values are listed in Table 2.

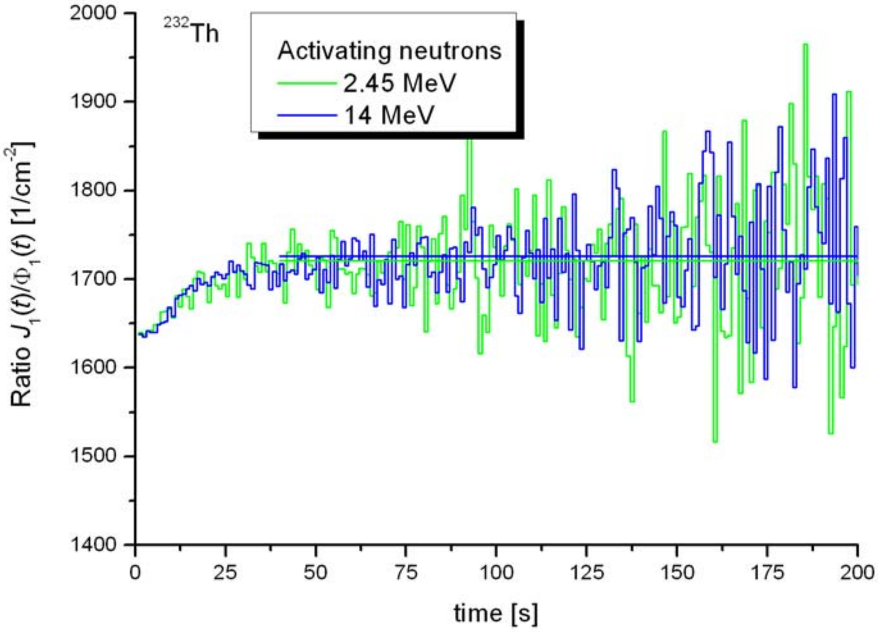

Fig. 14. Ratios of the time distributions of count rates in DET-12 to the time distributions of delayed neutrons emitted from the ^{232}Th tablet.

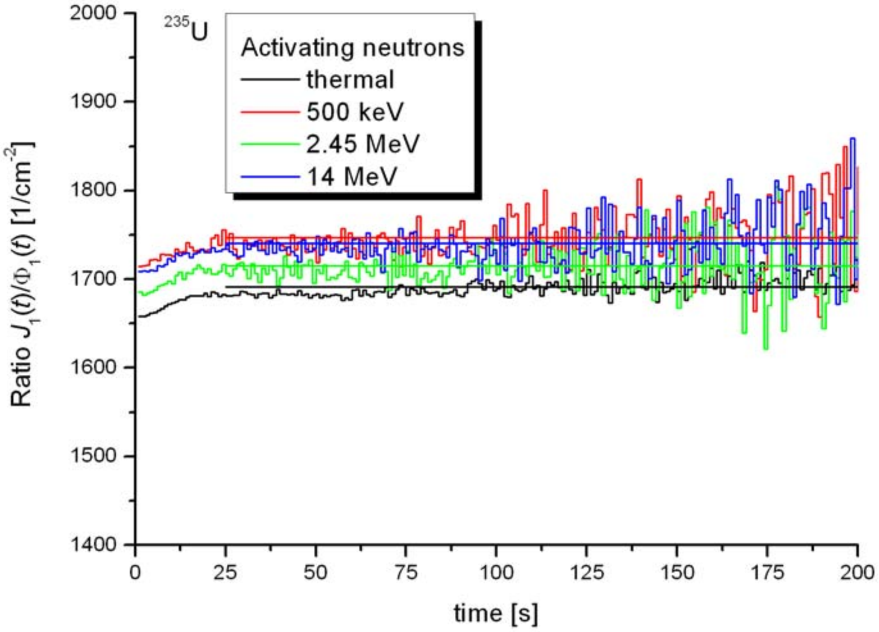

Fig. 15. Ratios of the time distributions of count rates in DET-12 to the time distributions of delayed neutrons emitted from the ^{235}U tablet.

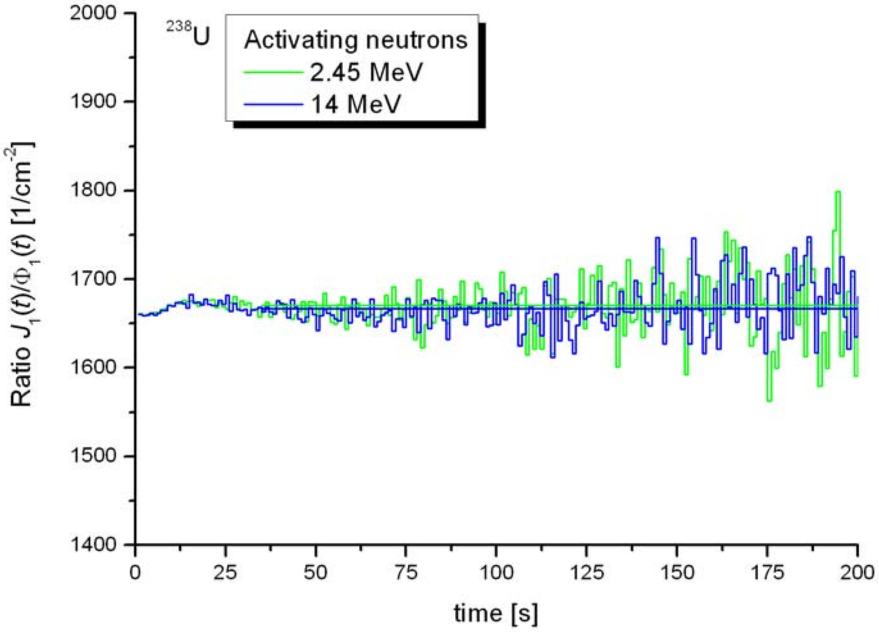

Fig. 16. Ratios of the time distributions of count rates in DET-12 to the time distributions of delayed neutrons emitted from the ^{238}U tablet.

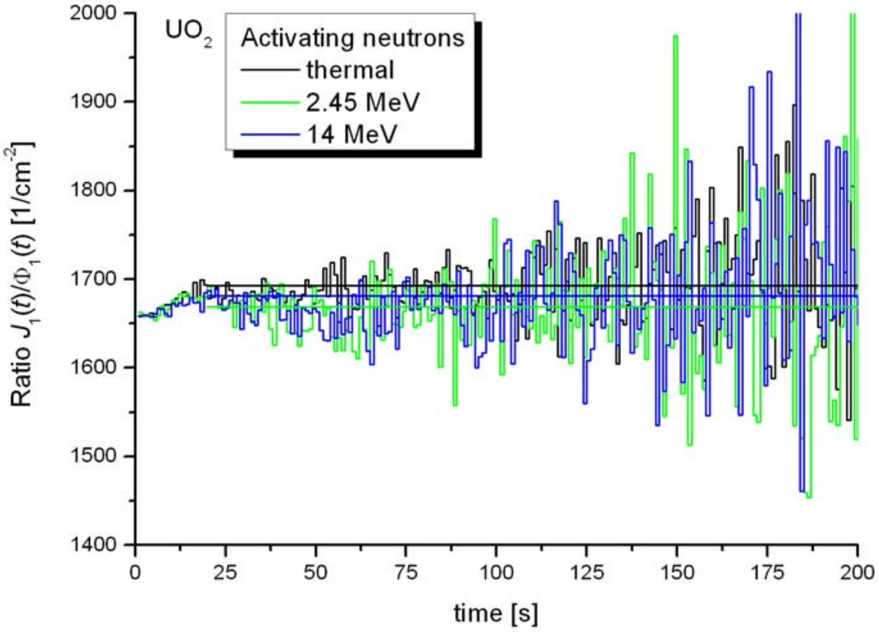

Fig. 17. Ratios of the time distributions of count rates in DET-12 to the time distributions of delayed neutrons emitted from the UO$_2$ tablet.

Table 2. Mean values of the ratios *R*(*t*) of the time distributions of count rates in DET-12 to the time flux distributions of delayed neutrons.

Sample	Mean ratio (DET-12 count rate)/(delayed neutron flux) [1/cm^{-2}] when sample is activated with initial neutrons of energy:			
	14 MeV	2.45 MeV	500 keV	thermal
^{232}Th	1730 ± 60	1720 ± 70	—	—
^{235}U	1740 ± 30	1720 ± 30	1750 ± 30	1690 ± 10
^{238}U	1670 ± 30	1670 ± 30	—	—
UO$_2$	1680 ± 70	1670 ± 70	—	1690 ± 50

6. Conclusions

The time distributions of delayed neutrons emitted from four samples of activated fissionable materials were calculated with the MCNP code. The tablets made of the pure ^{235}U, ^{238}U and ^{232}Th nuclides and the sintered UO$_2$ were employed. The samples were irradiated with the neutrons of four energies: thermal, 500 keV (600 keV for thorium), 2.45 MeV and 14 MeV. The sample tablets in void were irradiated by the mono-directional neutron beam and fluxes of neutrons exiting the samples were tallied in order to study the pure physical effect of generating and emission of the delayed neutrons. The actual experimental conditions of the neutron registration by means of the DET-12 device were also examined.

According to expectations, the isotopes of the high fission cross-sections for the specific energy provide a good statistics of the delayed neutrons.

For ^{232}Th and ^{238}U the emission coming from irradiation with slow neutrons is up to four-five orders lower than that from fast neutrons. A response for irradiation with 2.45 or 14 MeV neutrons cannot be distinguished. The delayed neutron emission after irradiation with thermal neutrons is high in case of ^{235}U (which has a high cross section for this reaction). This can suggest a method to estimate (at least roughly) an energy spectrum of primary neutrons if two types of sampling tablets were used, of ^{232}Th or ^{238}U and of ^{235}U simultaneously.

The detection efficiency of DET-12 does not depend neither on the kind of the isotope emitting the delayed neutrons nor on the energy of the neutrons irradiating the samples. It appears the delayed neutrons emitted from thorium, uranium and even oxygen are not distinguishable for the measuring chamber.

The set of the obtained experimental-like results delivers a good base to elaborate a method of analysis of registered time decays of the delayed neutrons for determination of the primary neutron intensity.

References

[1] G.D. Spriggs, J. Campbell and V.M. Piksaikin; *An 8-group delayed neutron model based on a consistent set of half-lives.* Progress in Nuclear Energy, Vol. 41, No. 1-4, pp.223-251 (2002).

[2] Drozdowicz K. et al., *Detection of the delayed neutrons from activation of fissionable materials in the neutron field at fusion-plasma devices*, Ann. Rept. 2008 of the Assoc. EURATOM-IPPLM, Warsaw 2009, 37-39 and CD-R 2_9_AReport2008.pdf, 15 pp.

[3] Drozdowicz K. et al., *Detection of the delayed neutrons from activation of fissionable materials in the neutron field at fusion-plasma devices*, Ann. Rept. 2009 of the Assoc. EURATOM-IPPLM, Warsaw 2010, 51 and CD-R 3_05_AR2009.pdf, 6 pp.

[4] X-5 Monte Carlo Team, *MCNP - A general Monte Carlo N-particle transport code, Version 5*, Los Alamos National Laboratory, LA-UR-03-1987, (2003).