Investigations of a Resonance Structure of Neutron Cross-
Sections using the Multiplicity Spectrometry Technique

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Abstract. It is known that uncertainties in the calculation of characteristics of power nuclear
reactors such as critical sizes, power density field, reactor life time etc are caused mainly
(about 40 %) by uncertainties in the knowledge of neutron constants of reactor materials.
Allowed errors on neutron constants for all basic reactor materials and fission products are
about 1-5 %. Required errors on constructional materials should be in the range of 5-10 % for
\( \sigma_t \), \( \sigma_s \), \( \sigma_{\gamma} \) and from 2 to 12 % for self-shielding factors. The required accuracy on neutron
constants for the majority of reactor materials is not achieved up to now and consequently it is
necessary to continue experimental research in this direction. For example, such an accuracy
could be achieved using the multiplicity spectrometry technique of the gamma-rays. The
description of this method, its features and several experimental results will be presented.

1. Introduction
The information on nuclear data is needed for tuning of different theoretical models of nuclear
reactions and a structure of nuclei. More precise definition is important for realization of different
transmutation projects and for increasing of a nuclear safety of energetic atomic reactors. Modern
calculations of fast reactors require a detailed knowledge of nuclear data, especially of neutron
characteristics of fissile and constructional materials. Also new accurate measurements of the neutron
cross-sections are needed for construction of the nuclear reactors of type BREST, BN-800 etc [1].
These trends and objectives are closely related to the improvement of estimated nuclear data of basic
world data libraries. The world requirements of precision on nuclear data are 1-5 % for basic reactor
and fissile materials, 3-7 % for constructional materials and 5-10 % for fission products.

2. The Multiplicity Spectrometry technique
It is possible to achieve precisions mentioned above on neutron cross-sections using the multiplicity
spectrometry technique. The multiplicity spectrometry of gamma-quanta and neutrons, emitted by
exited nuclei, has been developed to provide wide investigations of interactions of neutrons with
nuclei [2]. In contrast to separate experiments in which the same parameters are determined
independently from each others, an experiment with such a setup will provide considerably more
complete information together with separate quantities and spectra and will be possible to obtain all
possible correlations between them. It also allows to obtain neutron cross-sections and their ratios with a high accuracy and to study different channels of decays of excited nuclei.

Examining the probability of some nuclear process as a function of these parameters we obtain the corresponding spectra e.g. the multiplicity spectrum /A(\nu)/. The multiplicity spectrum is a distribution of number of particles emitted in a process of de-excitation namely gamma-quanta. It illustrates a probability of emitting one particle, two or more. Here we are talking about the total multiplicity of neutrons and gamma-quanta \( \nu=\nu_n + \nu_\gamma \), emitted as a result of interactions of slow neutrons with nuclei. In this case, the multiplicity spectrum \( A(\nu) \) consists of well-separated maxima, corresponding to scattering, radiative capture and fission processes /see figure1/.

![Figure1. A sketch of the multiplicity spectrum with measurements of scattering, radiative capture and fission processes.](image)

In the scattering only a single neutron is detected, so that the \( A(\nu) \) is everywhere equal to zero except at the point \( \nu=1 \) so the first maximum corresponds to the scattering process. Approximately 4 gamma-quanta are formed with the radiative capture, i.e. the capture \( A_\gamma(\nu) \) is concentrated in the region \( \nu\sim4 \). For fission the total number of gamma-quanta and neutrons is \( \sim10 \). Thus the third maximum corresponds to the fission multiplicity spectrum \( A_f(\nu) \).

Such multiplicity spectra could be registered by a \( 4\pi \)-multisectional detector with an efficiency close to 100 %. A sample under investigation is placed in the center of this detector. A number of detected k-fold coincidences of signals form different sections of the detector gives directly the multiplicity spectra \( A(\nu)=N(k) \). For this purpose a number of sections in the detector should be larger than a maximum number of emitted particles while an efficiency of detection of each particle by the detector is considered to be close to 100 %.

3. Detector systems.

At the Laboratory of Neutron Physics of the Joint Institute for Nuclear Research over many the technique base has been established. As a result we have two multisectional detectors which have been used for measurements of partial cross-sections and their ratios for many materials. The IBR-30 reactor has served as a neutron source [3]. During the measurements it has been operated at a mean power level of 10 kW with a neutron pulse width of 4 \( \mu \)s and a pulse repetition rate of 100 Hz.

3.1. The multisectional scintillation detector on the base of NaI(Tl) crystals.

The multisectional detector “ROMASHKA” consists of 16 scintillation sections made from NaI(Tl) (figure2). Each section of the detector is optically connected with its photomultiplier. A total volume of the detector is 36 l. A geometrical efficiency is about 90 %. A registration efficiency of single gamma-quanta at the energy of 1 MeV is about 85 % in the absence of its registration in several sections of the detector [4]. Using such a setup it is possible to measure the multi-parameter spectrum of a dependency of a number of events on the time-of-flight of neutrons (or a number of time channel),
on multiplicity coincidence of signal in the sections, on energy-release in each section of the detector and in the whole detector.

This spectrometer has the following important features:
(a) the geometrical arrangement,
(b) the actual performance of the individual detector elements,
(c) the associated electronics and data acquisition system, and
(d) the response of the system to input γ-cascades including the effect of crystal-to-crystal scattering and the response to neutrons.

3.2. The multisectional liquid scintillation detector “Parus”.

Another experimental setup allows to go down in energy to 2 eV (figure 3). The detector setup consists of two tanks. The total volume is 80 l. Each tank is divided into 8 sections separated from each other by the aluminum glare shield. Each section is viewed by a photomultiplier. A mixture of 100 g of scintillation additions, methilborate and tholuol is used as a scintillation liquid. The registration efficiency of gamma-quanta is approximately 30 %, the energy resolution for gamma-quanta at the energy of 662 keV (Cs line) is about 30 %.
4. Some experimental results.

4.1. Molybdenum.

Molybdenum belongs to reactor constructional material with a high melting temperature 2620°C. The following reaction

\[ ^{98}\text{Mo}(n,\gamma)^{99}\text{Mo} \rightarrow ^{99m}\text{Tc} \]  

is of interest for reactor construction, where Mo is widely used like a constructional material owing to its high resistance to high temperatures and to relatively low neutron capture cross-section. It is important that a product of the reaction mentioned above is \(^{99m}\text{Tc}\) radionuclide having ideal characteristics for its usage in radiopharmaceutical preparations for the diagnostic and medical treatment of different organs (\(T_{1/2}=6.006\) h, \(E=140.5\) keV, yield of gamma-quanta is 89.6 %, absence of \(\beta\)-rays)[5]. Also isotopes of Mo are accumulated in a large amount at a burning of a nuclear fuel and on 10-20 % determine a “parasitic” capture of neutrons from all fission products in fast reactors. This fact influences the duration of a campaign of nuclear reactors [6].

To obtain neutron cross-sections for Mo we have carried out measurements of the time-of-flight spectra using the multisectional scintillation detector “Romashka” on the basis of NaI(Tl) crystals (already described in subsection 3.1). During the measurements the Romashka setup has been installed at a distance of 500 m from the active zone of the IBR-30 reactor. The NaI(Tl) detector has operated in the multiplicity coincidence spectrometry conditions. The metal disk of Mo with a thickness of 0.4 mm or 0.00281 at/b and with a diameter of 80 mm has been used as a radiator-sample [7]. The multiplicity coincidences spectra have been registered from the 1st up to 15th multiplicities.

After background components had been subtracted and the time-of-flight spectra had been normalized, one determined the quantity pulses in the energy groups of the BNAB constants system [8] over the energy region from 20 eV up to 10 keV. Analogous measurements were realized for thin \(^{238}\text{U}\) sample with a thickness of 0.25 mm or 0.00121 at/b and with a diameter of 80 mm and used as a “standard” for determination of group capture cross-sections for Mo. The neutron capture cross-section for Mo can be determined by the following relations:

\[ <\sigma^U_C>_{i}^{Mo} = S^U_S^{Mo} \cdot \frac{(nd)^U}{(nd)^{Mo}_i} \cdot \frac{N^{Mo}_i}{M^{Mo}_i} \cdot \frac{M^U_i}{N^U_i} <\sigma^U_C>_{i}^{U} \]  

where: \(M^U\) and \(M^{Mo}\) - monitor coefficients for the \(^{238}\text{U}\) and Mo, \(S^U\) and \(S^{Mo}\) areas, \(n\) and \(n^{Mo}\) - thickness of radiator samples, \(d\) – diameters of the samples, \(N_i\) - counts per i-th channel.

After combining the energy intervals the \(^{98}\text{Mo}\) group capture cross-sections were determined according to formula (2). The group capture cross-sections for the \(^{98}\text{Nb}\) and \(^{238}\text{U}\) were obtained by the calculation with the help GRUCON code [9] on the basis of the estimated data libraries. A comparison of the experimental group capture cross-sections of Mo with other available experimental data and with the estimated values is shown in figure 4.
As one can see from figure 4 the experimental data are in good agreement not only with the estimated ones [10,11] but also with the other experimental results [12,13].

4.2. Thorium.

$^{232}$Th as well as $^{238}$U is a basic reactor material. Thorium is not a fissile nucleus but it absorbs slow neutrons. The following decay chain leads to the formation of $^{233}$U.

\[
\begin{align*}
  n + ^{238}U & \rightarrow ^{239}U \rightarrow ^{239}Np \rightarrow ^{239}Pu \\
  \beta^-, 23 \text{ min} & \beta^-, 2.3 \text{ days}
\end{align*}
\]

\[
\begin{align*}
  n + ^{232}Th & \rightarrow ^{233}Th \rightarrow ^{233}Pa \rightarrow ^{233}U \\
  \beta^-, 22 \text{ min} & \beta^-, 27 \text{ days}
\end{align*}
\]

$^{233}$U is a perfect nuclear fuel, supporting chain reaction. Each neutron absorbed by $^{239}$Pu or by $^{235}$U gives 2.03-2.08 new neutrons and by $^{233}$U-2.37. If they use the thorium cycle in nuclear reactor then Pu and other transuranium elements are accumulated much less in comparison with the uranium fuel cycle.

It is well known that the precise measurement of the capture cross-sections for Th is impeded by the high gamma background arising from its natural decay chain. Commonly, this gamma background is reduced by chemically removing the daughter products responsible for this background or by raising the pulse discrimination level. In the first case the measurements have to be carried out very quickly after a preparation of the sample because the daughter products build up again. The second method has a disadvantage that some of the gamma-rays resulting from the neutron capture are discriminated out which leads to a deterioration of the signal-to-background ratio and loss of experimental information.

To avoid such difficulties we have used the multiplicity spectrometry technique. The neutron capture cross-sections of $^{232}$Th were measured in the neutron energy range from 2 eV to 10 keV by using the neutron time-of-flight (tof) method at the IBR-30 pulsed neutron booster of JINR [14]. The gamma rays originating from neutron capture were detected by 16 sections of liquid scintillator located at a distance of 121 m from the neutron source (see subsection 3.2).

With this detector we have performed measurements of tof spectra for thin metal sample of Th (0.2 mm or 0.00665 at/b) and powder of U$_3$O$_8$ (0.000498 at/b) with 99.9 % of $^{238}$U. We have carried out a detailed analysis, including subtraction of background component, definition of intervals for energy group in the energy range from 2 eV to 10 keV and for low resolved resonance. The capture cross-sections were determined in compliance with the known cross-section of $^{238}$U by analogy with $^{98}$Mo.
using the expression (2). These capture cross-sections are presented in figure 5 in comparison with the evaluated data. As one can see from this Figure the experimental results agree very well with the evaluated ones within the experimental errors.

![Figure 5](image1)

**Figure 5.** The group capture cross-sections for $^{232}$Th.

Also we have extracted capture cross-sections for several low resolved resonances. A comparison of experimental data with the evaluated ones is shown in figure 6.

![Figure 6](image2)

**Figure 6.** The capture cross-sections for $^{232}$Th for low resolved resonances.

This comparison points to very good agreement between two sets of data.

5. Conclusion.
The presented experimental material demonstrates great possibilities of the gamma-ray multiplicity spectrometry for the investigation of exited fissionable and constructional nuclei. In future we are planning to build a similar multisectional detector system with a larger number of crystals and to measure neutron cross-sections for most important materials using the IREN neutron source at FLNP JINR [15].
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