Measurement of Neutrons in Different Pb/U Setups Irradiated by Relativistic Protons and Deuterons by means of Activation Samples

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Abstract. The collaboration Energy and Transmutation of Radioactive Waste uses different setups consisting of lead, uranium and graphite irradiated by relativistic protons and deuterons to study transmutation of radioactive materials by produced neutrons. Our group measured spatial distribution of neutrons by means of activation samples during the assembly irradiation by the JINR Nuclotron beams. We also present results of simulations using MCNPX code and their comparison with obtained experimental data. We use Au, Al, Bi, In and Ta foils as activation detectors, but unfortunately almost no experimental cross-section data for observed threshold (n,xn) reactions are available for higher neutron energies. Therefore we carried out series experiments devoted to determination of neutron cross-sections of various threshold reactions using different quasi-monoenergetic neutron sources.

1. The spallation neutron production and transmutation of actinides and fission products

Accelerator Driven Transmutation Systems (ADTS) based on a subcritical nuclear reactor driven by an external spallation neutron source are being investigated with increasing interest in the last three decades. The transmutations by very intensive fast and resonance neutron fields require more precise knowledge, particularly about emitted and moderated neutrons, residual nuclei and transmutations reactions. The studies of simple or more complex model set-ups simulating ADTS features are necessary. The collaboration Energy and Transmutation of Radioactive Waste is an international project that investigates neutron production by spallation reactions and their transport as well as transmutation of actinides and fission products in corresponding neutron fields.

Different set-ups are used to simulate properties of future ADTS. Lead or uranium targets of such set-ups are irradiated by proton or deuteron beams generated by the JINR Dubna accelerators. The first type of set-ups is simple lead target surrounded by paraffin or graphite moderator (GAMMA 1 and GAMMA 3). More complex set-up consists of lead target, natural uranium blanket and polyethylene biological shielding. Such set-up is named as “Energy plus Transmutation” and the systematic set of

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experiments with irradiation by proton and deuteron beams with different energies was performed [1]. Last year, experiments with new big uranium target KVINTA were started.

The measurements of the spatial distribution of neutron field inside such set-ups represent an important part of described studies. The activation detectors of neutrons are used for determination of neutron intensities and energies. The big advantages of this type of neutron detectors are their small size and their simplicity of usage.

2. The measurement of neutron reaction cross-sections

Unfortunately, almost no experimental cross-section data for most of the observed threshold (n,xn), (n,p) and (n,α) reactions are available for used activation detector materials and higher neutron energies. This is main motivation for the cross-section measurements done by means of two quasi-monoenergetic neutron sources based on proton beam, lithium target and reaction 7Li(p,n)7Be. The first neutron source is based on the cyclotron at Nuclear Physics Institute (NPI) at Řež with proton energy up to 38 MeV [2] and the second one is based on the cyclotron at The Svedberg Laboratory (TSL) in Uppsala with possible proton energy up to 200 MeV [3]. We performed five cross-section measurements exploiting the neutron source at NPI in Řež and seven irradiations by means of the neutron source at TSL in Uppsala using different energies of produced neutrons from 17 up to 94 MeV.

We studied Al, Au, Bi, In, Ta, Y and I materials usually in the form of thin foil. The activation method was used for determination of cross-sections. The first results were published in [4] and [5].

2.1. The sources of uncertainties

The very detailed analysis of all possible sources of statistical and systematic uncertainties is the most important condition for obtaining good quality results. The cross-section σ is determined by means of equation:

\[
\sigma = \frac{N_{\text{yield}} \cdot S \cdot A}{N_n \cdot N_A}
\]

where S is area of the foil, A is molar weight and N_A is Avogadro number. Then first source of possible uncertainties is connected with the neutron number N_n determination. The yield of radioisotope N_{yield} produced in the studied reaction is obtained by gamma spectroscopy using equation (2):

\[
N_{\text{yield}} = \frac{S_p \cdot C_{\text{abs}}(E) \cdot t_{\text{real}} \cdot 1 - e^{(-t_{\text{live}})} \cdot \lambda \cdot t_{\text{yr}}}{I_{\gamma} \cdot e_p(E) \cdot COI \cdot C_{\text{area}} \cdot m_{\text{foil}} \cdot 1 - e^{(-t_{\text{live}})} \cdot 1 - e^{(-t_{\text{yr}})}}
\]

where S_p is the gamma peak area, C_{abs} is self-absorption correction, I_{\gamma} is gamma line intensity, e_p(E) is detector efficiency, COI is correction for real coincidences, C_{area} is square-emitter correction, t_{real}/t_{live} is dead time correction, m_{foil} is the sample mass and the last two fractions represent decay during cooling and measurement and decay during irradiation. Then the second source of uncertainties is connected with accuracy of the different described gamma spectroscopy corrections and parameters.

The last source of uncertainties is connected with the neutron spectrum of the used source. Such sources are quasi-monoenergetic and their neutron spectrum contains beside the main neutron peak also a lower continuum. This background is negligible problem in the case of neutron energies which are near to the studied reaction threshold. The accurate subtraction of such background is very important if the cross-sections measurement is performed within neutron energies, which are much higher than the energy threshold.

We have prepared systematic studies of different neutron reaction cross-sections on yttrium. This monoisotopic material is very useful as a high energy neutron activation detector. It has many useable threshold (n,xn) reactions and we use such activation detectors very intensively during our ADTS
studies. But the experimental information about cross-sections for almost all reactions is very scarce, see EXFOR [6].

![Figure 1](image.png)  

Figure 1) Number of radioactive nuclei $^{88}$Y determined by means of gamma line with energy 898 keV, first eight values (side A was near to detector) and last seven values (side B was near to detector).

As part of this preparation we made methodical experiment by means of the NPI neutron source. The yttrium foil and gold foil (a common material with detailed knowledge of its properties) were irradiated by neutrons with energy 32.5 MeV obtained using proton beam with energy 35 MeV. The irradiation took 22 hours and high activities of both samples were obtained. Gamma measurements were performed for different distances of sample (15, 23, 33, 53, 70, 93 and 173 mm) to the detector and several measurements (between 6 and 15) were taken for each distance. The sample was turned to the opposite side after every measurement to analyze if there is an influence of geometry on measurements with different side nearer to detector. Magnitudes of the different gamma spectroscopy corrections are quickly decreasing with increasing distance to the detector. But it is necessary to use short distances in the case of low activity samples and to know accuracy of correction determination.

The yttrium foil was thicker and not fully homogenous. This was the reason why there was small difference between measurements with different side nearer to the detector for the nearest distances to the detector. But the difference is smaller than 1.4 % for second nearest position and within statistical error of single measurement, see figure 1. No such difference was observed in the case of thinner and homogenous gold foil. A methodical study of the uncertainties concerning the determination of other corrections was performed. The biggest influence has uncertainty of efficiency determination which is about 3 %.

2.2. The study of population of ground and isomeric $^{87}$Y states

The only two threshold reactions $^{89}$Y(n,2n)$^{88}$Y and $^{89}$Y(n,3n)$^{87}$Y are possible for neutron energy 32.5 MeV. The first reaction has the threshold energy 11.6 MeV and influence of the neutron background is important. The amount of $^{88}$Y nuclei produced by background is about 65 %. The cross-sections needed for background influence determination were calculated by means of the deterministic code TALYS [7] and the spectra of neutron source described in [2] were used. The last cross-section from EXFOR data base is for neutron energy 28 MeV. We obtained cross-section for new energy region.

The second reaction $^{89}$Y(n,3n)$^{87}$Y is very interesting. Prior to their further decay, the nuclei are produced either in their ground state of half-life of 79.8 hours, or in the 380.79 keV isomeric state of half-life 13.38 hours, see figure 2. The isomeric state decays by gamma transition to the ground state with probability 98.4 %. The beta decay of this state is within our accuracy negligible.
We made many measurements in different times and we obtained detailed description of isomeric state decay and population and decay of $^{87}$Y ground state, see figure 3. From the described data, we derived cross-sections of the $^{89}$Y(n,3n)$^{87}$Y and $^{89}$Y(n,3n)$^{87m}$Y reactions. Advantage is threshold of this reaction which is 21 MeV in this case. This is the reason why the amount of $^{87}$Y nuclei produced by the neutron background is relatively very small - about 7%.

Number $N_1$ of nuclei in isomeric state is described by equation (3):

$$N_1 = N_0 e^{-\lambda_1 t}$$

(3)

where $N_0$ is number of nuclei in isomeric state just after end of irradiation and $\lambda_1$ is decay constant of isomeric state. Number $N_2$ of nuclei in ground state is described by equation (4):

$$N_2 = \left( N_{02} + \frac{\lambda_1}{\lambda_1 - \lambda_2} \cdot N_{01} \right) e^{-\lambda_2 t} - \frac{\lambda_1}{\lambda_1 - \lambda_2} \cdot N_{01} e^{-\lambda_1 t}$$

(4)

where $N_{02}$ is number of nuclei in ground state just after end of irradiation and $\lambda_2$ is decay constant of ground state. The influence of decay and population of both states during irradiation is necessary to include for obtaining the cross-sections of $^{87}$Y ground and isomeric states production. The cross-section of isomeric state is about 2.8 times larger than the cross-section of ground state, see figure 4.
Figure 4) Cross-sections of $^{87}$Y ground and isomeric states production, experimental data marked by points and TALYS calculations are described by lines. Our data are preliminary.

3. Conclusions

The activation neutron detectors represent a convenient tool for the determination of the spatial distribution of a neutron field. The measurements of cross-sections in wide range of energies are needed to increase the usability of such detectors. We have used two quasi-monoenergetic neutron sources to obtain needed cross-sections. The methodic measurement of neutron reactions on yttrium for energy 32.5 MeV tested the possibility to obtain excitation functions of such reactions. The origins and magnitudes of the uncertainties were studied in detail.

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