Studies of deuteron and neutron cross-sections important for ADS research

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The collaboration Energy and Transmutation of Radioactive Waste uses different setups consisting of lead, natural uranium and graphite irradiated by relativistic protons and deuterons to study transmutation of radioactive materials by produced neutrons. The activation samples are used to determine integral of proton or deuteron beams and also produced neutron flux in different places of experimental set-ups. Unfortunately almost no experimental cross-section data for deuterons with GeV energies are available. The similar situation is also for threshold (n,xn) reactions of neutrons with higher energies. Therefore we carried out series of experiments devoted to determination of deuteron reactions on copper during uranium target QUINTA irradiations by deuterons with energies from 1 GeV up to 8 GeV. The cross-sections of various threshold reactions were studied by means of different quasi-monoenergetic neutron sources with possible energies from 14 MeV up to 100 MeV. Knowledge of such cross-sections is very important for all Accelerator Driven System studies.

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1. Introduction

Activation detectors are commonly used to measure integral flux of different particles during Accelerator Driven System (ADS) studies. Determination of proton or deuteron beam integral intensity during ADS irradiation is possible by means of mainly the aluminum or copper foils. Activation samples make also possible to measure neutron flux in different places of ADS set-ups. The samples are small and their placement inside set-up is very simple. Approximate determination of neutron spectra is possible by use of different materials and different threshold reactions. Significant voids in the cross-section libraries of different important reactions are the main problem of effective work with the activation detectors. This gap was the main reason for us to start series of cross-section studies.

2. Cross-sections of relativistic deuteron reactions on copper

We participate in ADS studies performed by "Energy and Transmutation of Radioactive Waste" (E&T RAW) international collaboration at JINR Dubna [1]. The accelerator Nuclotron is used as source of relativistic protons and deuterons for such studies. The aluminum and copper foils are used to determine beam integral during different experiments of our collaboration. Cross-sections of relativistic proton reactions with aluminum and copper are mostly known. Situation is very different for relativistic deuterons. Set of cross-sections for deuteron reaction ^{27}Al(d,3p2n)^{24}Na is very scare for energies higher than 200 MeV and experimental values of cross-sections for deuteron reactions on copper are completely missing. This was the reason why we started program of measurements of these cross-sections.

The measurements were performed during irradiations of QUINTA and GAMMA-3 set-ups of E&T RAW collaboration by means of deuteron beams from Nuclotron accelerator at JINR Dubna. Overall ten irradiations were performed during three sets of E&T RAW collaboration experiments (March 2011, December 2011 and March 2012). The integral intensities of deuteron beams were determined by production of ^{24}Na in aluminum foil, for more detailed description see [2]. The common irradiation of aluminum and copper foils made possible to determine cross-sections of production of different radioisotopes by reactions of relativistic deuterons with copper. The used copper foils have natural isotope composition (69.15 % of ^{63}Cu and 30.85 % of ^{65}Cu). Many radioisotopes with suitable decay time and gamma lines are produced during irradiation of copper by relativistic deuterons.

2.1 Activation method

Activation method was used to determine cross sections. Copper foil with size 10×10 cm with thickness 0.0128 cm was placed together with aluminum foil with the same size and thickness 0.0196 cm several meters far away from the experimental set-up in the direction to beam tube window. Both foils were irradiated by same beam portion in the same place. The distance was sufficient to neglect possible influence of back-scattered neutrons and other particles from irradiated set-up.

The yield (i.e., the number of nuclei activated in the foil during the whole period of the irradiation) of produced gamma-radioactive nuclei was determined with the help of gamma-spectroscopy. The foil was packed from original size to a smaller one with dimensions approximately 2.5x2.5x0.3 cm^3 for the spectroscopy measurement. Activated foils were measured by two or more detectors and also in more different geometries which were in the range from 4 cm up to 10 cm far from detector. More measurements were done to see and identify short lived and long lived radioisotopes. We started with short measurements (minutes) and ended with long measurements (days).

The detectors were calibrated using standard laboratory ^{54}Mn, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{228}Th, and ^{241}Am point sources which have several gamma-lines ranging from 80 keV up to 2610 keV. Evaluated calibration activities include correction on real coincidences for isotopes with more lines. Total number of points used for one calibration curve is more than 35.

Gamma-spectra were evaluated in the Deimos 32 code [3]. From the gauss-fit of the peaks we got also the statistical uncertainty. Total yield of ^{24}Na in the Al foil was determined according to the classic equation and all spectroscopic corrections were included, for details see [4]). The total uncertainty of the yield is gained by sum of statistical uncertainty, uncertainty of detector efficiency determination (mostly approximately 3 %) and different corrections uncertainties (mostly approximately 1 %).

The total beam intensity was determined by the activation analysis method using foil from ^{27}Al. The knowledge of ^{27}Al(d,3p2n)^{24}Na reaction cross-section is crucial for determination of absolute normalization of obtained copper reaction data.

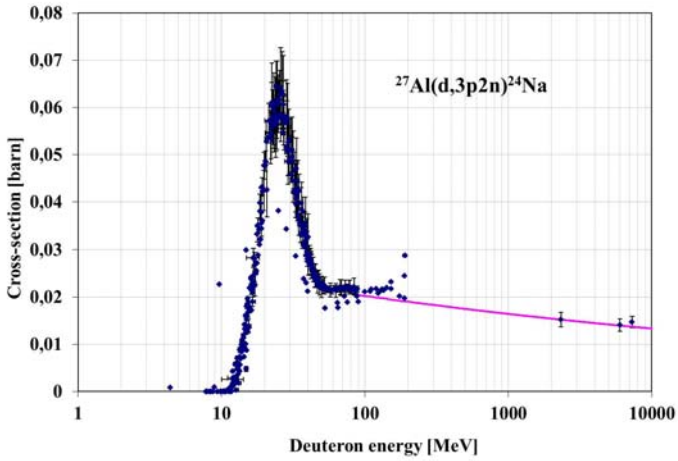
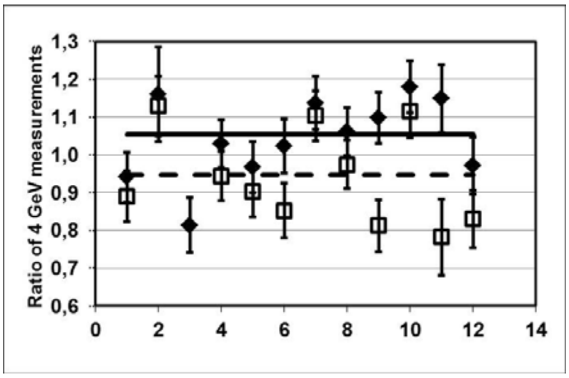


Fig. 1) Cross-section of ^{27}Al(d,3p2n)^{24}Na reaction as function of energy – experimental data from EXFOR and fit of the points.

2.2 Cross-sections of $^{27}Al(d,3p2n)^{24}Na$ reaction

Unfortunately, there are only three experimental cross-section values for $^{27}Al(d,3p2n)^{24}Na$ reaction in the GeV energy range. One value is from Dr. Bainaigs (15.25 ± 1.5 mbarn at 2.33 GeV) [5] and two are from Dr. Kozma (14.1 ± 1.3 mbarn at 6 GeV and 14.7 ± 1.2 mbarn at 7.3 GeV) [6], see Fig. 1.

We made a fit by function $\sigma = a \cdot E^b$ (linear in log scale of energy) between these three points and calculated the cross-section value for the two energies between the points from EXFOR database [7], we got a value of 16.4 mbarn for 1 GeV, a value of 14.5 mbarn for 4 GeV and a value of 13.6 mbarn for 8 GeV. The error is about 10% and we use this uncertainty for the cross-section in our calculations.



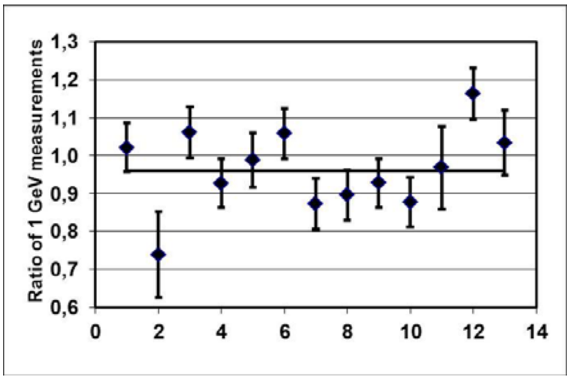


Fig. 2) Ratios of cross-sections obtained during three measurements with 4 GeV deuteron beam (left). Order of isotopes is ^{58}Co, ^{56}Co, ^{56}Mn, ^{52}Mn, ^{48}Sc, ^{44m}Sc, ^{57}Ni, ^{48}V, ^{47}Sc, ^{55}Co, ^{48}Cr and ^{43}K. Ratio of March 2011 and March 2012 is shown by empty boxes (single ratios) and dashed line (mean weighted value). Ratio of December 2011 and March 2012 is shown by full triangles and full line. Ratios of two measurements with 1 GeV beam (March 2012 / December 2011) are shown in the same manner in right figure.

2.3 Cross-sections of different radioisotope production by deuteron copper reactions

Gamma lines of the thirteen different radioisotopes (^{43}K, ^{43}Sc, ^{44m}Sc, ^{47}Sc, ^{48}Sc, ^{48}V, ^{48}Cr, ^{52}Mn, ^{56}Mn, ^{55}Co, ^{56}Co, ^{58}Co +^{58m}Co and ^{57}Ni) were identified in the obtained gamma spectra. Radioisotope ^{56}Co has the shortest half-life (2.6 hours) among them and radioisotope ^{58}Co the longest one (1700.6 hours). Radioisotopes ^{48}Sc (43.7 hours) and ^{48}V (383.4 hours) decay both to the same daughter nucleus ^{48}Ti. The analysis of the A=48 isobars is complicated also by the sequence of the decays ^{48}Cr → ^{48}V → ^{48}Ti.

Radioisotopes ^{43}K (22.3 hours) and ^{43}Sc (3.9 hours) decay both to the same daughter nucleus ^{43}Ca. The gamma line with energy 617.49 keV is produced only by ^{43}K decay and it is possible to use its intensity as norm to suppress contribution of ^{43}K to gamma line with energy

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372.8 keV during first few hours after end of irradiation. We will obtain yield of ^{43}Sc decay using 372 keV line after subtraction of ^{43}K contribution.

Similar analysis was done for decays of radionuclides ^{56}Mn (2.6 hours) and ^{56}Co (1854.5 hours). The gamma line with energy 1238.3 keV is produced only in ^{56}Co decay and it is possible to use it for calculation of contribution of ^{56}Co decay to 846.8 keV line during first hours after end of irradiation. The yield of ^{56}Mn is determined after subtraction of calculated contribution of ^{56}Co decay.

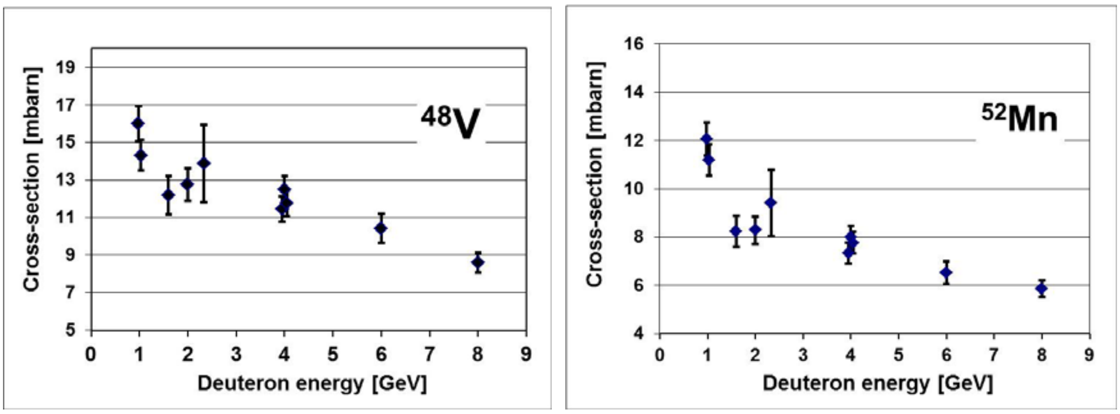

Fig. 3) Cross-sections of production of ^{48}V and ^{52}Mn by deuteron reaction on natural copper are shown.

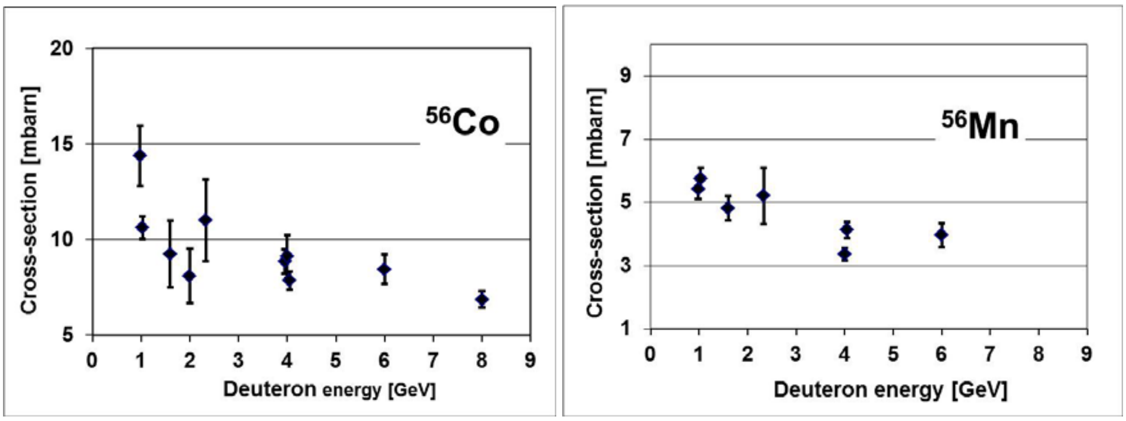

Fig. 4) Cross-sections of production of ^{56}Co and ^{56}Mn by deuteron reaction on natural copper are shown. These two radionuclides have the same daughter nucleus ^{56}Fe.

The statistical uncertainties, uncertainties of efficiency, spectroscopic corrections and subtractions are different for individual lines (radionuclides). The uncertainty of beam integral determination is same for all radionuclides measured in the same irradiation. We made irradiation by the 4 GeV deuteron beam three times and their systematic differences are within

10 %. Mean value of ratios between March 2011 and March 2012 results is 0.95(3) and mean value of ratios between December 2011 and March 2012 results is 1.05(2). Mean value of ratios between March 2012 and December 2011 results obtained during 1 GeV beam is 0.96(2). More detailed information is shown in Fig.2. It is consistent with expected uncertainty of beam integral determination.

Excitation functions of thirteen different radioisotopes were determined within the range of energies from 1 GeV up to 8 GeV. Several examples are shown in Fig. 3 and 4.

3. Cross-sections of neutron reactions

We routinely use threshold reactions in various materials to measure neutron flux during ADS studies. The usually used materials for neutron activation detectors are Al, Au, Bi, Co, In, Ta and Y. Unfortunately almost no experimental cross-section data for observed threshold (n,xn) reactions are available for higher neutron energies. Therefore we carried out series of experiments devoted to determination of neutron cross-sections of various threshold reactions using different quasi-monoenergetic neutron sources.

3.1 Quasimonoenergetic neutron sources

The quasi-monoenergetic neutron sources based on proton beam, lithium target and reaction $^{7}Li(p,n)^{7}Be$ are ideal tools for neutron reaction cross-section measurements. We used two such sources. The first neutron source is based on the cyclotron at Nuclear Physics Institute (NPI) in Řež with proton energy up to 38 MeV [8]. Flux density up to 10^{9} $cm^{-2}s^{-1}$ is possible. The second one is based on the cyclotron at The Svedberg Laboratory (TSL) in Uppsala with possible proton energy up to 200 MeV [9]. Flux density around 10^{5} $cm^{-2}s^{-1}$ is possible. We performed nine cross-section experiments 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.

3.2 Activation measurement of cross-sections

Activation method was used for cross-section determination. The same technique as in previous case is applied in these experiments. Yield of observed isotopes (products of the (n,xn) reactions) was calculated with respect to the various spectroscopic corrections - corrections on decay of the isotope between the end of irradiation and beginning of the measurement, correction on the intensity of the I_γ transition, correction on dead-time of the detector, correction on real γ-γ cascade coincidence, self-absorption correction, square-emitter correction (geometrical correction), detector efficiency, and beam instability correction. The background yield was also calculated, subtracted and cross-section was calculated by means of formulae in articles [11,12,13,14].

3.3 Examples of results

Seven irradiations with energies 22, 47, 59, 66, 72, 89 and 94 MeV were performed by means of quasi-monoenergetic neutron source at TSL in Uppsala. They were supplemented with irradiations on similar neutron source at NPI Řež with accurate neutron energies 17.5, 21.9, 30.4, 32.5 and 35.9 MeV. The Au, Al, Bi, In and Ta materials were studied in all irradiations. The yttrium was studied during last two years. Cross-sections of threshold reactions (n,2n), (n,3n) for these samples exist in EXFOR [7] up to 30 MeV, but no experimental data exist for higher energies or (n,xn) reactions of higher order.

Comparison between the data from EXFOR, TALYS [15] and results from the measurements from Řež and Uppsala was made. Examples of the results can be seen in following graphs (Fig. 5 and Fig. 6). Good agreement with TALYS 1.4 and EXFOR data (where exist) is observed for most of the isotopes.

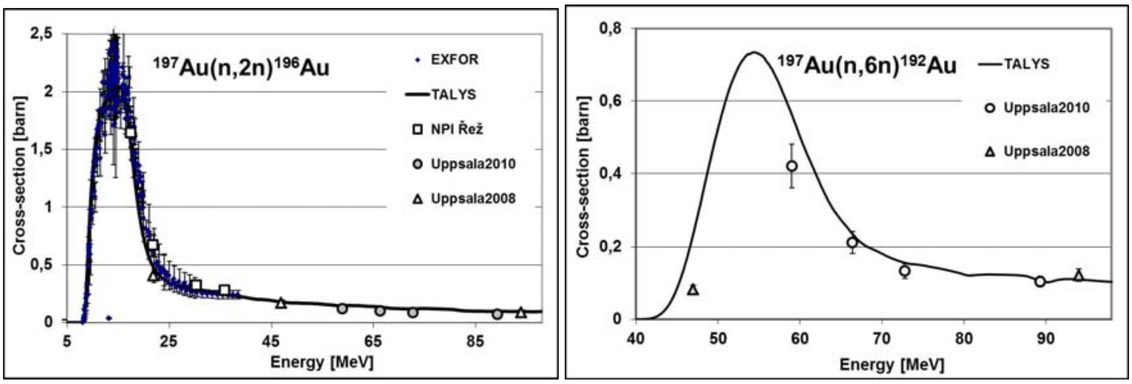

Fig. 5) Cross-sections of production of ^{196}Au and ^{192}Au by neutron reaction on gold are shown as examples of studied threshold reactions.

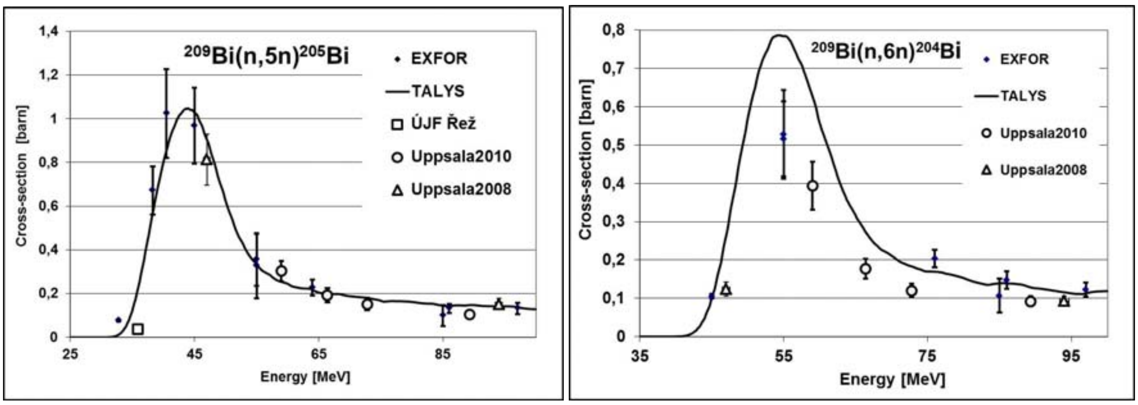

Fig. 6) Cross-sections of production of ^{205}Bi and ^{204}Bi by neutron reaction on bismuth are shown as examples of studied threshold reactions.

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We started detailed studies of neutron reactions with yttrium two years ago. The reaction $^{89}Y(n,3n)^{87}Y$ is very interesting. Prior to their further decay, the nuclei are produced either in their ground state with half-life of 79.8 hours, or in the 380.79 keV isomeric state with half-life 13.38 hours. 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 derived cross-sections of the $^{89}Y(n,3n)^{87}Y$ and $^{89}Y(n,3n)^{87m}Y$ reactions from set of gamma spectra measured in different times. Results of first test of such analysis made by means of irradiation at NPI Řež are referred in [13]. Results of all previous irradiations at NPI Řež and TSL Uppsala are presented in Fig. 7. We already started new set of studies of neutron threshold reactions on yttrium.

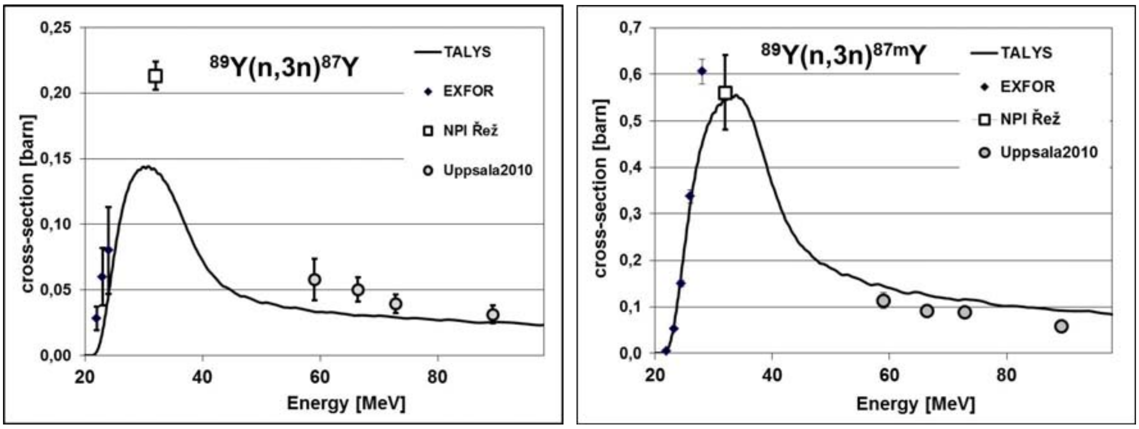

Fig 7) Studies of production of ^{87}Y and ^{87m}Y by neutron reaction on yttrium were started.

4. Conclusions

The activation method was used to determine cross-sections of relativistic deuterons with natural copper. The cross-sections of thirteen different radioisotopes production (^{43}K, ^{43}Sc, ^{44m}Sc, ^{47}Sc, ^{48}Sc, ^{48}V, ^{48}Cr, ^{52}Mn, ^{56}Mn, ^{55}Co, ^{56}Co, ^{58}Co and ^{57}Ni) were determined within the deuteron energy range from 1 GeV up to 8 GeV.

Original data on cross-sections of neutron threshold reactions were obtained for neutron energies above 17 MeV by means of quasi-monoenergetic $^{7}Li(p,n)^{7}Be$ neutron sources at TSL Uppsala and NPI Řež. The results are in good agreement with the cross-sections already published in EXFOR. Good agreement is mostly with calculation by means of TALYS 1.0 code.

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