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How to Use Benchmark and Cross-section Studies to Improve Data Libraries and Models

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Abstract. Improvements of the Monte Carlo transport codes and cross-section libraries are very important steps towards usage of the accelerator-driven transmutation systems. We have conducted a lot of benchmark experiments with different set-ups consisting of lead, natural uranium and moderator irradiated by relativistic protons and deuterons within framework of the collaboration “Energy and Transmutation of Radioactive Waste”. Unfortunately, the knowledge of the total or partial cross-sections of important reactions is insufficient. Due to this reason we have started extensive studies of different reaction cross-sections. We measure cross-sections of important neutron reactions by means of the quasi-monoenergetic neutron sources based on the cyclotrons at Nuclear Physics Institute in Řež and at The Svedberg Laboratory in Uppsala. Measurements of partial cross-sections of relativistic deuteron reactions were the second direction of our studies. The new results obtained during last years will be shown. Possible use of these data for improvement of libraries, models and benchmark studies will be discussed.

1. Introduction

Highly accurate reaction cross-section data are needed for planning of future nuclear technologies as fast breeder reactors, accelerator-driven transmutation systems or fusion reactors. Different models and codes were developed to describe and design present and future nuclear systems. Accurate measurements with simple and more complex set-ups which can simulate future nuclear technologies are needed for benchmarks and improvement of the models and codes. For this purpose the beams of neutrons and other particles in rather broad range of energies provided by different neutron sources and accelerators are used.

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2. Relativistic deuteron reactions on copper

The experimental cross-sections of relativistic deuteron reactions on copper are almost completely missing. Situation is very different for relativistic protons. An extensive set of experimental partial cross-section data can be found in the EXFOR database for different radionuclide production in this proton case. Therefore, we have started a set of studies of relativistic deuteron reactions on copper. We used the deuteron beam produced by JINR Dubna Nuclotron accelerator. The activation method and the gamma-ray spectrometry were used for determination of partial production cross-sections. Beam integral was determined by means of ^{24}Na production in the aluminium monitor [1,2,3].

Overall 38 different radioisotopes were identified by their respective gamma lines with half-life span from less than 1 hour to hundreds of days and their independent or cumulative production cross-sections were measured using activation method. Most of our measured cross-sections are cumulative (namely ^{22}Na , ^{24}Na , ^{28}Mg , ^{38}S , ^{39}Cl , ^{43}K , ^{47}Ca , ^{43}Sc , ^{48}V , ^{48}Cr , ^{49}Cr , ^{51}Cr , ^{52}Mn , ^{56}Mn , ^{52}Fe , ^{59}Fe , ^{55}Co , ^{56}Co , ^{57}Co , ^{56}Ni , ^{57}Ni , and ^{61}Cu). We have determined independent partial cross-sections for ^7Be (appropriate isobars are not produced), ^{42}K (half-life of ^{42}Ar is 32.9 years), ^{44}Sc and $^{44\text{m}}\text{Sc}$ (half-life of ^{44}Ti is 60 years), ^{46}Sc (^{46}Ca is stable), ^{47}Sc (half-life of ^{47}Ca is 4.5 days and we used early measurements), ^{48}Sc (^{48}Ca is stable), ^{54}Mn (^{54}Cr is stable), ^{58}Co (stable isobars around) and ^{60}Co (stable isobars around). The independent cross-sections have been determined also in the case of ^{65}Ni , ^{64}Cu , ^{62}Zn , and ^{65}Zn , where other appropriate isobars are not produced.

The excitation functions were obtained. Representative set of the new experimental data for deuteron reactions are presented in Fig. 1 and 2 together with proton reaction data from the EXFOR database. The change of excitation function shape with increasing nucleon number of the produced radionuclide is nicely visible. The shape of deuteron reaction excitation functions is similar to proton ones but absolute values of the deuteron reaction cross-sections are by about 30-40 % larger than proton ones. Exception is the case of ^7Be production where cross-sections of deuteron reactions are about two times higher. The only one measured cross-section value of deuteron production of ^{24}Na on copper collected in the EXFOR database [4] agrees nicely with our new data (see Fig. 1a).

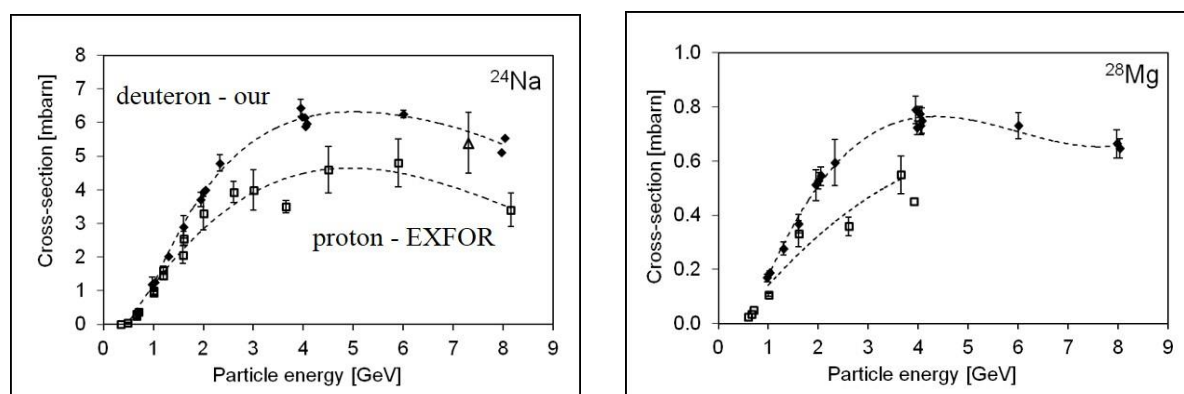


Figure 1. (a) Excitation function of $^{\text{nat}}\text{Cu}(\text{d},\text{x})^{24}\text{Na}$ reaction (b) Excitation function of $^{\text{nat}}\text{Cu}(\text{d},\text{x})^{28}\text{Mg}$ reaction. Our experimental data for deuterons are compared with EXFOR data for protons. Examples of light radionuclide production. Measured value of the ^{24}Na production cross-section existed in EXFOR database (signed by triangle) agrees with our data.

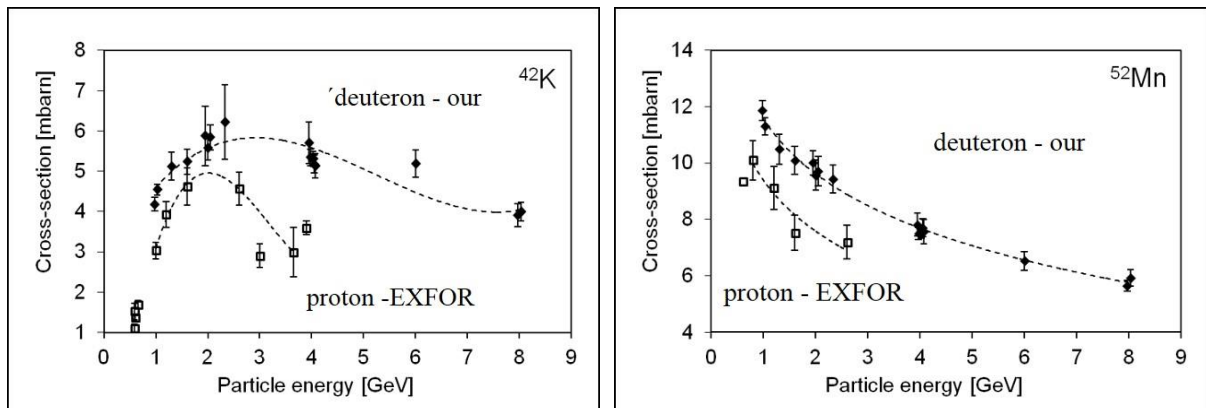


Figure 2. (a) Excitation function of $^{nat}\text{Cu}(d,x)^{42}\text{K}$ reaction, example of medium heavy radionuclide production (b) Excitation function of $^{nat}\text{Cu}(d,x)^{52}\text{Mn}$ reaction, example of target like radionuclide production. Our experimental data for deuterons are compared with EXFOR data for protons.

The obtained excitation functions were compared with simulation done by the Monte Carlo code MCNPX version 2.7.0 [5] with two intra-nuclear cascade-evaporation models INCL-ABLA and LAQGSM included in the code. The experimental data are better fitted with the LAQGSM model than the INCL-ABLA model in most cases. The simulated excitation functions are generally similar in shape but can differ significantly from the experimental data in absolute value in certain cases. Selected examples of our measured cross-sections for deuterons together with excitation functions for deuteron reactions simulated in the MCNPX code are displayed in Fig. 3 and 4. We see that the accuracy of the experimental data make possible to distinguish which model describes excitation functions better and the obtained experimental values shown possibility for improvements of models.

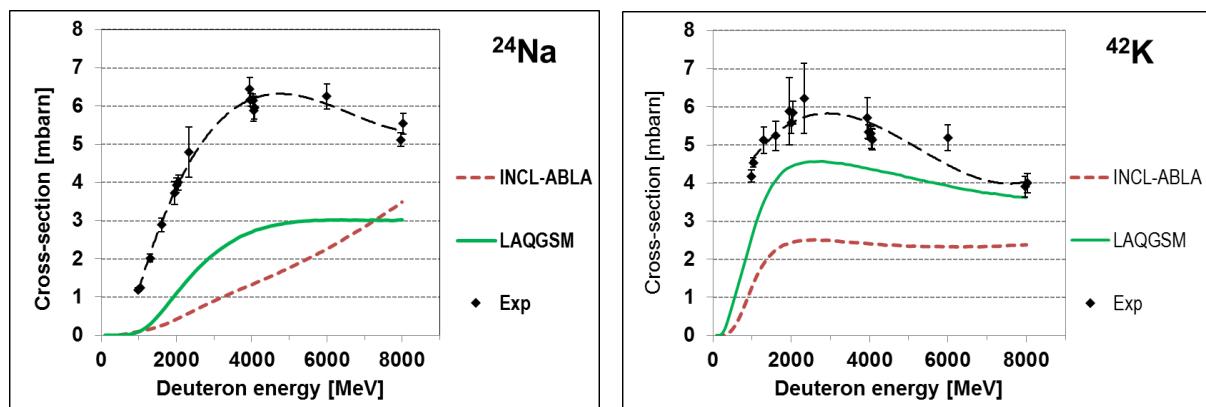


Figure 3. (a) Excitation function of $^{nat}\text{Cu}(d,x)^{24}\text{Na}$ reaction (b) Excitation of $^{nat}\text{Cu}(d,x)^{42}\text{K}$ reaction. The experimental data are compared with the model predictions (INCL-ABLA and LAQGSM are used)

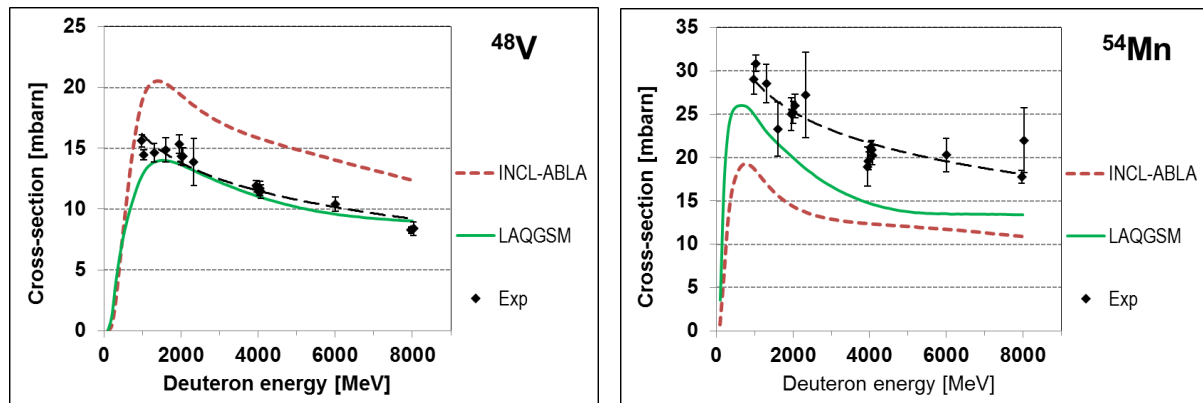


Figure 4. (a) Excitation function of $^{nat}\text{Cu}(d,x)^{48}\text{V}$ reaction (b) Excitation of $^{nat}\text{Cu}(d,x)^{54}\text{Mn}$ reaction. The experimental data are compared with the model predictions (INCL-ABLA and LAQGSM are used).

3. Excitation functions of neutron reactions

Activation detectors are a very useful tool for measurement of neutron flux. We use intensively neutron threshold detectors during our studies of accelerator driven transmutation systems in JINR Dubna [6,7]. Experimental cross-section data of used reactions are very scarce for energies above 30 MeV. The neutrons with energies above 100 MeV are produced during our experiments using the relativistic proton and deuteron beams of JINR Dubna accelerators. The threshold reactions enable to measure flux of such neutrons (see Fig 5). The decay products of radionuclides produced by reactions with very high threshold are necessary to measure in some cases. The decay chains $^{197}\text{Au}(n,9n)^{189}\text{Au}$ (0.5h) \rightarrow ^{189}Pt (10.9h) \rightarrow ^{189}Ir (13.2d) \rightarrow ^{189}Os and $^{197}\text{Au}(n,10n)^{188}\text{Au}$ (0.5h) \rightarrow ^{188}Pt (10.2d) \rightarrow ^{188}Ir (41.5h) \rightarrow ^{188}Os , or $^{209}\text{Bi}(n,10n)^{200}\text{Bi}$ (0.6h) \rightarrow ^{200}Pb (21.5h) \rightarrow ^{200}Tl (26.1h) \rightarrow ^{200}Hg , $^{209}\text{Bi}(n,11n)^{199}\text{Bi}$ (0.45h) \rightarrow ^{199}Pb (1.5h) \rightarrow ^{199}Tl (7.4h) \rightarrow ^{199}Hg and $^{209}\text{Bi}(n,12n)^{198}\text{Bi}$ (0.17h) \rightarrow ^{198}Pb (2.4h) \rightarrow ^{198}Tl (5.3h) \rightarrow ^{198}Hg are examples of such possibilities. The production of such radionuclides by direct neutron reactions for so heavy nuclei as gold or bismuth is negligible. One example of measurement by means of bismuth activation detectors is shown on Fig. 6.

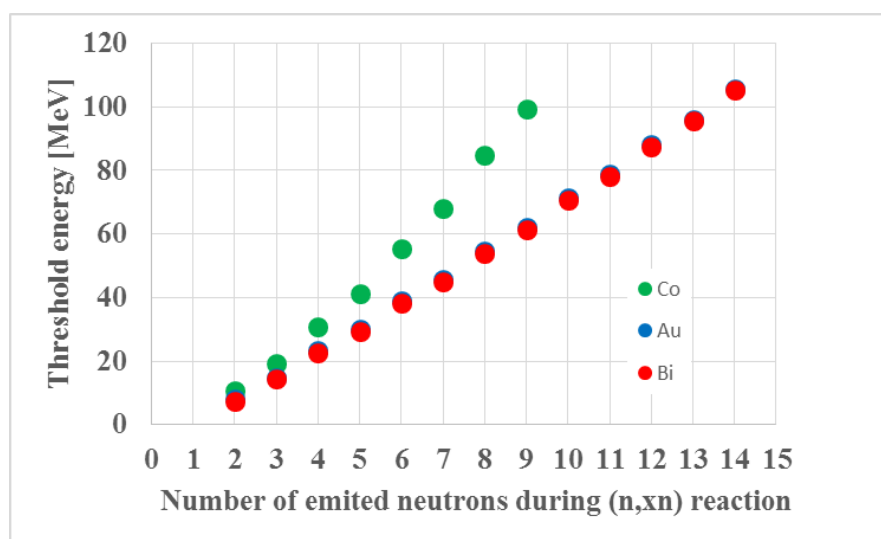


Figure 5. The energy thresholds of different (n,xn) reactions on three materials (cobalt, gold a bismuth).

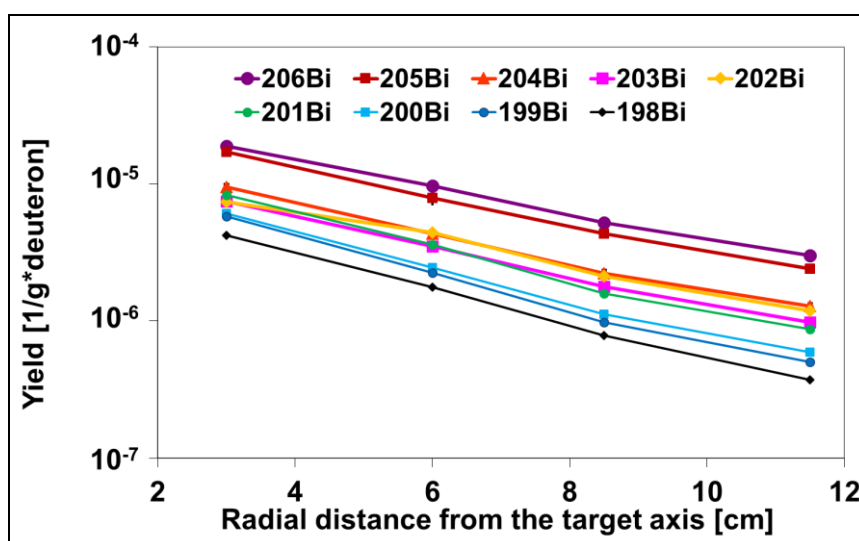


Figure 6. Example of spatial distribution of different radionuclides production on bismuth activation samples. Threshold reactions up to (n,12n) and threshold above 80 MeV were measurable during irradiation of the E+T set-up by 4 GeV deuterons. Distance of activation detectors from the front of the target was 11.4 cm

This was reason why we started extensive studies of (n,xn) reactions on materials used as activation detectors [8,9]. We used two different quasi mono-energetic neutron sources based on the reaction on ^7Li target. We performed experiments by means of the neutron source at Nuclear Physics Institute of ASCR in Řež and the neutron source at The Svedberg Laboratory of Uppsala University. These two neutron sources made possible to cover very broad range of neutron energies from 14 MeV up to 100 MeV. We used activation method of cross-section determination and detailed description of experiments, used techniques and discussion of all uncertainty sources are in [8,9]. The example of results for one high threshold reaction (n,7n) is on Fig 7. Our results enable to test different versions of TALYS code [10]. The Fig 8 shows that changes between the different versions of TALYS code are very small for low threshold reaction but there are big differences between the different versions for high threshold reaction.

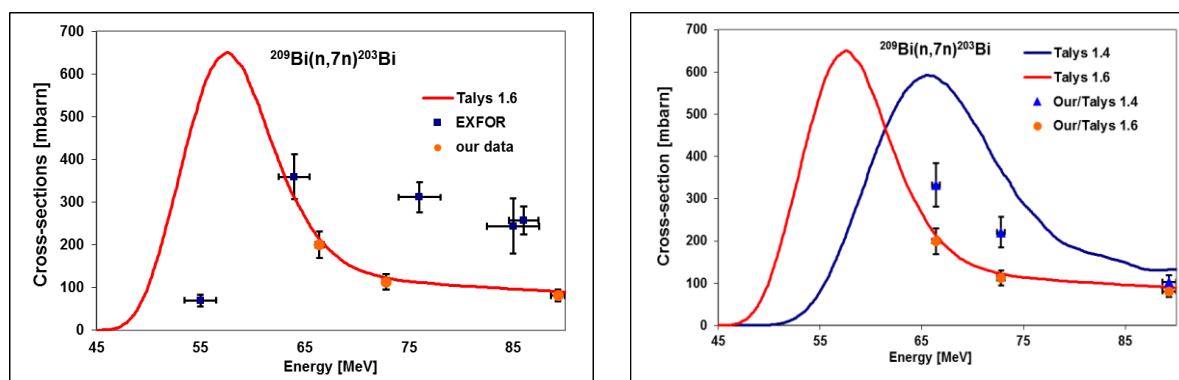


Figure 7. (a) Our data obtained during Uppsala experiment at 2010 are compared with EXFOR data and TALYS 1.6 results. (b) Comparison of predictions obtained by means of two different version of TALYS code and also results of subtractions of background estimated by these two different version of TALYS code.

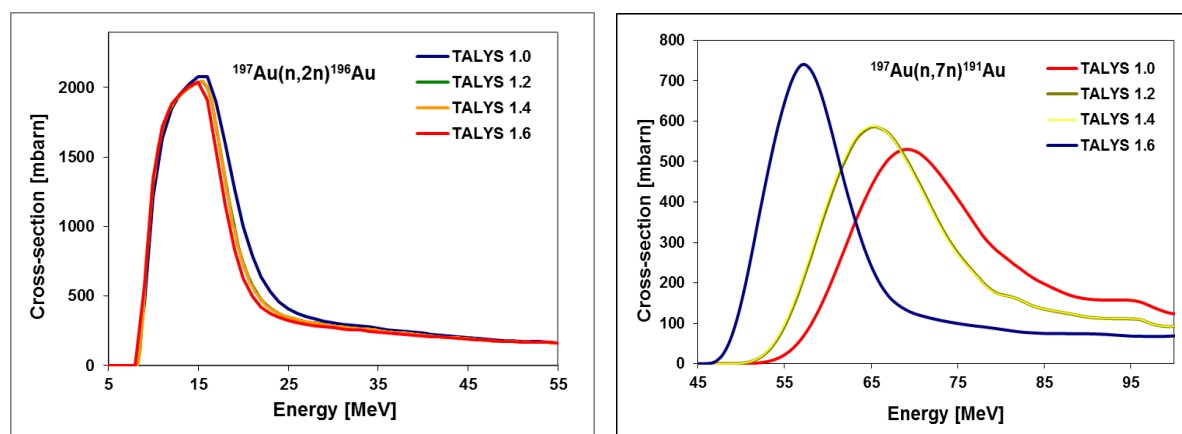


Figure 8. (a) The time evolution of TALYS code, for the low threshold reaction (n,2n) are only very small changes (b) Different situation is for the high threshold reaction (n,7n).

4. Conclusion

Accuracy of experimental cross-section data obtained in our experiments enables to test and improve models and codes (Monte Carlo and deterministic). They enable beam integral determination besides aluminium foil also by more than three dozen monitoring reactions available when copper foil is used during our accelerator-driven systems studies and also more accurate determination of high energy neutron fields inside studied set-ups by the activation detectors.

Acknowledgements

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