# Cross-section studies of relativistic deuteron reactions obtained by activation method.

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#### Abstract

The cross-sections of relativistic deuteron reactions on natural copper were studied in detail by means of activation method. The copper foils were irradiated during experiments with the big Quinta uranium target at Joint Institute for Nuclear Research (JINR) in Dubna, Russia. The deuteron beams with energies ranging from 1 GeV up to 8 GeV were produced by JINR Nuclotron. Residual nuclides were identified by the gamma spectrometry. Lack of such experimental cross-section values prevents the usage of copper foils from beam integral monitoring.

### **1** Introduction and motivation

The international collaboration "Energy and Transmutation of RadioActive Waste" (E&T RAW) at Joint Institute for Nuclear Research in Dubna (JINR Dubna), Russia, performed intensive studies of simple ADS set-ups irradiated by proton and deuteron beams during past years [1-6]. We use <sup>24</sup>Na production reaction in aluminium foils for beam monitoring. However, the foils have to be placed in large distance from irradiated set-up due to production of <sup>24</sup>Na also by spallation neutrons emitted from thick target studied. On the other hand, the production of radionuclides on copper monitor foils by deuteron beam is not affected by MeV spallation neutrons reactions and hence copper foils can be placed near the set-up under study. Therefore we measured partial crosssections of different radionuclides production by deuterons on copper. The obtained data will improve possibility to use copper monitors during ADS studies. The second main goal of these studies is to provide a database for evaluation of models used for prediction of the production of different radionuclides by relativistic deuterons in various fields of application.

An extensive set of experimental partial cross-section data is in EXFOR data base for different radionuclide production by relativistic proton beam on copper. However, on the other hand, there is only one partial cross-section value for such type of reaction for relativistic deuteron beam. The production of <sup>24</sup>Na on copper by 7.3 GeV deuterons was described by R. Brandt et al. [7]. An improvement of our knowledge of excitation functions of different radionuclide production on copper by relativistic deuterons is necessary.

### 2 Experimental method

The measurements were performed in the frame of E&T RAW collaboration during irradiations of QUINTA and GAMMA-3 set-ups with deuterons from Nuclotron accelerator at JINR Dubna [8,

9]. Typical irradiation lasted about ten hours. Sixteen irradiations were performed during five sets of experiments carried on from 2011 to 2013. Our new data were measured relatively to the monitoring reaction  ${}^{27}$ Al(d,X) ${}^{24}$ Na. The aluminium and copper foils had the same sizes (10×10 cm) and thicknesses of copper and aluminium were 0.0128 cm and 0.0196 cm, respectively. The used copper foils have natural isotope composition (69.15 % of  ${}^{63}$ Cu and 30.85 % of  ${}^{65}$ Cu). Both foils were placed in the same position. The distance from the set-up was sufficient to neglect the possible influence of neutrons and other particles produced by the set-up to the direction of the beam monitors.

Unfortunately, there are only three experimental cross-section values for  ${}^{27}Al(d,3p2n)^{24}Na$  reaction in GeV energy range. One value is from J. Bainaigs et al  $(15.25 \pm 1.5 \text{ mbarn at } 2.33 \text{ GeV})$  [10] and two are from P. Kozma et al  $(14.1 \pm 1.3 \text{ mbarn at } 6.0 \text{ GeV} \text{ and } 14.7 \pm 1.2 \text{ mbarn at } 7.3 \text{ GeV})$  [11]. The uncertainties of beam monitoring reaction cross-sections are the main source of systematic uncertainties of obtained absolute cross-section values. We made a fit between experimental points and calculated the cross-section value for the given energies between the points from database. We got value 16.4 mbarn for 1 GeV, value 15.4 mbarn for 2 GeV, value 14.5 mbarn for 4 GeV and value 13.6 mbarn for 8 GeV (error is about 10%).



**Fig. 1:** Cross-section of  ${}^{27}$ Al(d,3p2n) ${}^{24}$ Na reaction – experimental data taken from EXFOR data base and examples of point fits.

The activation method exploiting gamma-ray spectrometry was used for the cross-section determination. The foils were packed from original size to a smaller one with dimensions  $2.5 \times 2.5 \times 0.3$  cm<sup>3</sup> for the spectroscopy measurement. Several high purity germanium detectors were used. More different geometries were also used. Distances of measured sample from detector ranged from 4 cm up to 10 cm. Every radioactive sample was measured many times to detect and identify short lived and also long lived radioisotopes. First measurements were started only a few hours after end of irradiation and even very short lived radionuclides (half-lives only a few hours) were detected. The measured gamma spectra were analysed by the DEIMOS code [12]. The yield of activated material was calculated after identification of the isotope by means of gamma peaks. All necessary spectroscopic corrections and related sources of uncertainties were taken into account. All uncertainties were quadratically added according to the laws of uncertainty propagation. The cross-sections were determined taking into account the number of atoms in a sample and deuteron beam integral, see [13, 14] for details.

Gamma lines of more than twenty different radioisotopes were identified (for example <sup>24</sup>Na, <sup>42</sup>K, <sup>43</sup>K, <sup>43</sup>Sc, <sup>44</sup>Sc+ <sup>44m</sup>Sc, <sup>46</sup>Sc, <sup>47</sup>Sc, <sup>48</sup>Sc, <sup>48</sup>V, <sup>48</sup>Cr, <sup>51</sup>Cr, <sup>52</sup>Mn, <sup>54</sup>Mn, <sup>56</sup>Mn, <sup>52</sup>Fe, <sup>59</sup>Fe, <sup>55</sup>Co, <sup>56</sup>Co, <sup>57</sup>Co, <sup>58</sup>Co + <sup>58m</sup>Co, <sup>57</sup>Ni, <sup>61</sup>Cu, <sup>64</sup>Cu and <sup>62</sup>Zn). Some couples of radionuclides decayed to the same daughter nucleus and therefore it was necessary to analyze decay curves to distinguish separate radionuclides (for example <sup>43</sup>Sc and <sup>43</sup>K, <sup>48</sup>Sc and <sup>48</sup>V, <sup>56</sup>Mn and <sup>56</sup>Co). The complex analyzes of decay curves were necessary also in the case of decay sequences and study of isomeric state population (<sup>44m</sup>Sc). More detailed description of the used procedures is in [15, 16].



**Fig. 2:** Five measurements with 4 GeV deuterons were done. Ratio of cross-section determined during individual measurement and mean weighted average of all five measurements. Order of isotopes is <sup>58</sup>Co, <sup>56</sup>Co, <sup>52</sup>Mn, <sup>48</sup>Sc, <sup>44m</sup>Sc, <sup>57</sup>Ni, <sup>48</sup>V, <sup>47</sup>Sc, <sup>55</sup>Co, <sup>48</sup>Cr and <sup>43</sup>K. Used signs are: March 2011 – violet diamond, December 2011 – blue triangle, March 2012 – green square, December 2012 – red ring and March 2013 – red triangle.

More irradiations were done for some energy. For example, we obtained five independent values of cross-sections for deuteron energy 4 GeV (see Fig. 2). The systematic uncertainties of beam integral determination were estimated by means of these data. It is noted that all measurements are within 20 % range and within expected uncertainties of single irradiations. The similar situation, even better, was for other deuteron energies which were measured more times. The detailed analysis of different sources of systematic uncertainties was performed.

The more detailed study whether or not the given cross-sections are independent or do contain uncorrectable contributions from radioactive progenitors will be done in near future.

### **3** Obtained results

Examples of the new experimental data for deuteron reactions are presented in Figs 3-7 (left) together with EXFOR data for relativistic proton reactions (right figures). The excitation functions of lighter radionuclides (<sup>24</sup>Na, <sup>42</sup>K) production show sharp rise starting below 1 GeV and continuing to about 3 GeV (see Figs 3 and 4), above which slow decrease starts. Excitation functions of radionuclides between nucleon numbers 44 and 48 have constant value for energies

ranged from 1 to 3 GeV. The slow decrease is visible for higher energies. Decrease with rising energy is observable for excitation functions of radionuclides with nucleon number higher than 52 (see Fig. 5). There are set of partial cross-sections of proton production of different radionuclides on copper in EXFOR data base. The excitation functions of relativistic deuteron reactions on copper show very similar trends as excitation functions of relativistic proton reactions. The absolute values of the relativistic proton reaction partial cross-sections are lower by about 30 % than the same partial cross-sections for deuteron reactions.



**Fig. 3:** Cross-sections of <sup>nat</sup>Cu(d,x)<sup>24</sup>Na reaction measured by us, open triangle shows only one existing experimental value from EXFOR data base (left figure). Existing experimental cross-sections of <sup>nat</sup>Cu(p,x)<sup>24</sup>Na reaction induced by proton beam obtained from EXFOR data base (right figure).



**Fig. 4:** Cross-sections of  ${}^{nat}Cu(d,x)^{42}K$  reaction induced by deuteron beam measured by us (left figure). Existing experimental cross-sections of  ${}^{nat}Cu(p,x)^{24}Na$  reaction induced by proton beam obtained from EXFOR data base (right figure).

Some measured partial cross-sections have rather high values by approximately 30 mbarn (for example <sup>51</sup>Cr, <sup>54</sup>Mn, <sup>57</sup>Co, <sup>58</sup>Co and <sup>64</sup>Cu). Some values are about few milibarns (for example <sup>43</sup>K, <sup>43</sup>Sc, <sup>47</sup>Sc, <sup>52</sup>Mn, <sup>56</sup>Mn, <sup>56</sup>Co and others) and smallest values are up to fraction of milibarn (for example <sup>52</sup>Fe and <sup>62</sup>Zn). The production of <sup>62</sup>Zn is very special and interesting case. The proton from deuteron should be stopped at copper nucleus and only a few neutrons should be emitted in this case. The probability of such development is very small and the partial cross-section of <sup>62</sup>Zn production is very small, only about 0.6 mbarn.



**Fig. 5:** Cross-sections of  ${}^{nat}Cu(d,x){}^{54}Mn$  reaction induced by deuteron beam measured by us (left figure). Existing experimental cross-sections of  ${}^{nat}Cu(p,x){}^{54}Mn$  reaction induced by proton beam obtained from EXFOR data base (right figure).



**Fig. 6:** Cross-sections of  ${}^{nat}Cu(d,x){}^{59}Fe$  reaction induced by deuteron beam measured by us (left figure). Existing experimental cross-sections of  ${}^{nat}Cu(p,x){}^{59}Fe$  reaction induced by proton beam obtained from EXFOR data base (right figure).



**Fig. 7:** Cross-sections of  ${}^{nat}Cu(d,x){}^{62}Zn$  reaction induced by deuteron beam measured by us (left figure). Existing experimental cross-sections of  ${}^{nat}Cu(p,x){}^{62}Zn$  reaction induced by proton beam obtained from EXFOR data base (right figure).

## 4 Conclusions

In this work, the excitation functions of relativistic deuteron reactions on copper were studied. The partial cross-sections of more than twenty different radionuclide productions were determined. The residual nuclides investigated cover gamma emitting radionuclides with half-lives between 2 hours and almost 300 days. The shape of excitation function for light fragment production is initially sharply increasing up to 3 GeV and after wide maximum the excitation function starts to decrease slowly. Fragments with the nucleon number around 46 have constant value of partial cross-section up to deuteron energy 3 GeV and the slow decrease of cross-section value starts for higher deuteron energies. The excitation function is monotonically decreasing for nucleon number higher than 52. Our newly obtained data for <sup>24</sup>Na production partial cross-sections nicely agree with only one measured value of such cross-section placed in EXFOR data base. The shapes of the excitation functions for relativistic proton reactions are more than 20 % higher than for proton reactions

The new consistent set of experimental partial cross-sections measured by us will be used to test nuclear reaction models of high-energy reactions such as spallation and fragmentation. Mainly we want to test different models used by MCNPX. The obtained cross-sections will be also useful for monitoring of deuteron beam by copper foil.

The measurements of cross-sections of neutron threshold reactions with energies ranging from 10 MeV to 100 MeV are second main field of our studies. We use quasi-monoenergetic neutron sources based on the reaction on <sup>7</sup>Li target at Nuclear Physics Institute of ASCR in Řež and at The Svedberg Laboratory Uppsala. The last study of yttrium (n,xn) threshold reactions is described by Petr Chudoba in these proceedings and overview of our results obtained by us is in [13]. Both these studies were supported by EFNUDAT and ERINDA projects.

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## References

- [1] M. I. Krivopustov et al, First experiments with a large uranium blanket within the installation "Energy plus Transmutation" exposed to 1.5 GeV protons, Kerntechnik 68 (2003) 48
- [2] F. Křížek et al, The study of spallation reactions, neutron production, and transport in a thick lead target and a uranium blanket during 1.5 GeV proton irradiation, Czech. Journal of Physics 56 (2006) 243
- [3] A. Krása et al, Neutron production in a Pb/U-setup irradiated with 0.7-2.5 GeV protons and deuterons, Nucl. Instr. and Meth. in Phys. Res. A615 (2010) 70
- [4] M.I. Krivopustov et al, First results studying transmutation of <sup>129</sup>I, <sup>237</sup>Np, <sup>238</sup>Pu and <sup>239</sup>Pu in the irradiation of extended <sup>nat</sup>U/Pb-assembly with 2.52 GeV deuterons, Journal of Radioanalytical and Nuclear Chemistry, 279 (2009) 567
- [5] M. Zamani-Valasiadou et al, Performance of a Pb-spallation target surrounded by a Ublanket during irradiations with 1.6 and 2.5 GeV deuteron beams: Comparison with relativistic proton beams, Annals of Nuclear Energy 37 (2010) 241
- [6] J.J. Borger et al, Spatial distribution of thorium fission rate in a fast spallation and fission neutron field: An experimental and Monte Carlo study, Nucl. Instr. and Meth. in Phys. Res. A664 (2010) 70
- [7] R. Brandt et al, Enhanced production of 24Na by wide-angle secondaries produced in the interaction of relativistic carbon ions with copper, Phys. Review C45 (1992) 1194
- [8] W. Furman et al, Recent results of the study of ADS with 500 kg natural uranium target assembly QUINTA irradiated by deuterons with energies from 1 to 8 GeV at JINR NUCLOTRON, Proceedings of Science (Baldin ISHEPP XXI 086) http://pos.sissa.it/archive/conferences/173/086/Baldin%20ISHEPP%20XXI\_086.pdf
- [9] M. Suchopár et al, Monte Carlo simulations of natural uranium setups irradiated with relativistic deuterons by means of MCNPX code, Proceedings of Science (Baldin ISHEPP XXI
   091)
- http://pos.sissa.it/archive/conferences/173/091/Baldin%20ISHEPP%20XXI\_091.pdf
  [10] J. Banaigs et al, Determination De L, Intensite D, Un Faisceau De Deutons Extrait D"Un Synchrotron Et Mesure Des Sections Efficaces Des Reactions C-12(D,P2N)C-11 Et Al-27(D,3P2N)Na-24 a 2.33 GeV, Nuclear Instruments and Methods in Physics Research 95 (1971) 307-311
- [11] P. Kozma and V. V. Yanovski, Application of BaF2 scintillator to off-line gamma ray spectroscopy, Czech Journal of Physics, 40 (1990) 393
- [12] J. Frána, Program DEIMOS32 for gamma-ray spectra evaluation, Journal of Radioanalytical

and Nuclear Chemistry 257 (2003) 583

- [13] J. Vrzalová et al, Studies of (n,xn) cross-sections in Al, Au, Bi, Cu, Fe, I, In, Mg, Ni, Ta, Y, Zn by the activation method, Nucl. Instr. and Meth. in Phys. Res. A726 (2013) 84
- [14] O. Svoboda, Experimental Study of Neutron Production and Transport for ADTT, Disertation Thesis, Czech Technical University, Faculty of Nuclear Sciences and Physical Engineering, Prague (2011), [http://cie.wif.ece.org/www.meg/tengenut/cee/dialog/http://DUD\_Suchede.ndf]

[http://ojs.ujf.cas.cz/~wagner/transmutace/diplomky/PHD\_Svoboda.pdf]

- [15] V. Wagner et al, Studies of deuteron and neutron cross-sections important for ADS research, Proceedings of Science (Baldin ISHEPP XXI 090) http://pos.sissa.it/archive/conferences/173/090/Baldin%20ISHEPP%20XXI\_090.pdf
- [16] V. Wagner et al, Cross-section studies of important neutron and relativistic deuteron reactions, XX International School on Nuclear Physics, Neutron Physics and Aplications (VARNA 2013)