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ARTICLE

"Proton Activation Data File" to Study Activation and Transmutation of Materials Irradiated with Protons at Energies up to 150 MeV

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To satisfy growing needs in nuclear data at intermediate energies, the Proton Activation Data File (PADF) has been prepared. It contains 418,575 excitation functions of nuclear reactions for 2,355 target nuclei from Mg to Ra at proton energies up to 150 MeV. The data are available in the PADF for stable and unstable target nuclei including isomeric targets with the half-life more than one second. Cross-sections included in the PADF were obtained by using the TALYS code, the modified ALICE code, and the available experimental data.

KEYWORDS: protons, nuclear data file, intermediate energies, evaluated data, cross-sections, excitation functions, activation, radionuclide production

I. Introduction

The evaluation and compilation of data for nuclear reactions induced by intermediate energy protons is important for a wide range of applications including the activation study for advanced nuclear systems, like accelerator-driven systems and the study of production of radionuclides used in medicine and industry. Evaluated proton data files such as ENDF/B-VII,[1)] JENDL-HE,[2)] and JEFF-3.1[3)] contain the information for a few tens of stable nuclides, which number keeps growing, but, still is not enough for a complete and detailed study of activation and transmutation of the materials irradiated with intermediate energy protons. To satisfy growing needs in nuclear data at intermediate energies, the Proton Activation Data File (PADF) has been composed. It contains calculated and evaluated excitation functions of nuclear reactions for the target nuclei from Mg to Ra at proton energies up to 150 MeV. Data are available in the PADF for stable and unstable target nuclei with half-life more than one second.

Data for the Proton Activation Data File were obtained using the TALYS code,[4)] the ALICE/ASH code,[5)] and the existing experimental data. Before the forming of the PADF, the preliminary work has been done to define nuclear models and approaches providing the best agreement with experimental nuclide yields. The comparison with the experimental data has been made using nuclear models implemented in TALYS and ALICE/ASH. Approaches having minimal values of deviation factors[6)] at the description of the experimental data were applied for cross-section calculations for the PADF. Additional corrections have been made for calculated cross-sections using the available experimental data.

II. Brief Description of Methods Used to Obtain Cross-Sections for PADF

1. Calculations Using Nuclear Models

Calculations of cross-sections for (p,x) reactions including the radiative capture were performed using nuclear the models implemented in the TALYS code and the ALICE/ASH code.

The TALYS code involves calculations by the pre-equilibrium exciton model[7)] and the Hauser-Feshbach model.[8)] The phenomenological model from Ref. 9) was used for the description of the pre-equilibrium complex particle emission from nuclei. The contribution of direct processes in proton inelastic scattering was calculated using the ECIS code[10)] integrated in the TALYS code. The coupled channel model or DWBA was selected by TALYS for calculations using the available information about nuclear level schemes.[4)] The macroscopic phenomenological model was used to describe collective excitations in the inelastic channel. The model is discussed in detail in Ref. 4).

The geometry dependent hybrid model (GDH)[11)] and the Weisskopf-Ewing model[12)] were applied for numerical calculations with the ALICE/ASH code. Intranuclear transition rates were calculated using the effective cross-sections of nucleon-nucleon interactions in nuclear matter. Corrections were made to the GDH approach for the treatment of effects in peripheral nuclear regions.[5,13)] The simulation of the multiple precompound emission was done according to Ref. 11). The numbers of neutrons and protons for initial exciton state were calculated using realistic nucleon-nucleon interaction

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Table 1 The values of deviation factors,[6,26,28,29] which quantify the difference between experimental cross-sections for proton induced reactions (p,x) including radiative capture at energies up to 150 MeV, cross-sections calculated by TALYS and cross-sections included in PADF

Deviation factor	Before the evaluation	After the evaluation (PADF)
$\left(\frac{1}{N}\sum_{i=1}^{N}\left(\frac{\sigma_i^{\mathrm{exp}}-\sigma_i^{\mathrm{calc}}}{\Delta\sigma_i^{\mathrm{exp}}}\right)^2\right)^{1/2}$	122.0	4.69
$\frac{1}{N}\sum_{i=1}^{N}\frac{\sigma_i^{\mathrm{calc}}}{\sigma_i^{\mathrm{exp}}}$	1.71	0.975
$\frac{1}{N}\sum_{i=1}^{N}\left\lvert\frac{\sigma_i^{\mathrm{exp}}-\sigma_i^{\mathrm{calc}}}{\sigma_i^{\mathrm{exp}}}\right\rvert$	1.02	0.124
$10^{\left(\frac{1}{N}\sum_{i=1}^{N}[\log(\sigma_i^{\mathrm{exp}})-\log(\sigma_i^{\mathrm{calc}})]^2\right)^{1/2}}$	2.15	1.47

The number of experimental points (N) is equal to 19,253.

cross-sections in nucleus.[5] The exciton coalescence pick-up model[14,15] and the knock-out model were used for the description of the pre-equilibrium complex particle emission. Parameters of models are described in details in Refs. 5, 16–18).

The reaction cross-sections used in TALYS and ALICE/ASH calculations were obtained by the optical model with parameters from Ref. 19). In the TALYS code, the transmission coefficients for neutrons and protons were calculated with the optical potential from Ref. 19) and those for composite particles (d, t, ^{3}He, ^{4}He) with optical model parameters described in Ref. 4). Parameters of the optical potential applied for the description of the secondary particle emission in the ALICE/ASH code were discussed in Ref. 16).

The preliminary work has been done for the comparison of available experimental radionuclide yields in neutron and proton induced reactions with calculations by TALYS and ALICE/ASH using various nuclear models for the nuclear level density calculations. Three various approaches were used to obtain the nuclear level density in TALYS calculations. In the first approach, the level density was calculated by the Fermi gas model[20] supplemented by the "constant temperature" model at low excitation energies. In the second approach, the same model was used, but, with the explicit description of the vibrational and rotational enhancement.[4] In both cases, different systematics for the asymptotic value of the nuclear level density parameter and the shell damping parameter were used.[4] The third approach implements the results of the microscopic calculations performed by Goriely and coauthors.[21–23] Calculations by the ALICE/ASH code were carried out using the Fermi gas model with the nuclear level density parameter A/9, the model from Ref. 20), and the superfluid nuclear model.[24,25]

More than 17,000 experimental radionuclide yields in neutron induced reactions for target nuclei from Al to Bi at energies above 0.1 MeV[6,26] and about 19,000 experimental points for proton induced reactions for targets from Mg to Bi at energies up to 150 MeV[27] were used for the comparison with calculations. The comparison was done using various statistical criteria.[6,26,28,29] Results of the comparison[6,26,27] showed a definite advantage of the Fermi gas model without an explicit description of the collective enhancement[4] implemented in the TALYS code and the superfluid model[24,25] included in the ALICE/ASH code comparing with other approaches. These two models for the calculations of the nuclear level density were used to obtain the cross-sections for the Proton Activation Data File.

The TALYS code was used to obtain the reaction cross-sections for stable nuclei and unstable nuclei with the half-life more than ten minutes. To reduce the time of computation, cross-sections for targets with the shorter decay half-life were obtained by the ALICE/ASH code. Calculations for isomeric targets were performed using the TALYS code.

2. Use of Experimental Data

The experimental cross-sections available in EXFOR for proton induced reactions (p,xnypzα) including (p,γ) were used for the fitting and correction of calculated excitation functions. It involved 19,253 independent (non-cumulative) residual yields for 735 nuclear reactions from 1434 EXFOR data sets for 162 target nuclei. In most cases, the correction concerned the shift and the normalization of theoretical curves providing the minimal χ^2 value. If the shape of calculated excitation function and the trend of experimental data differed completely, the approximation of data was performed by rational functions. In the present version of the file, the correction of calculated cross-sections was done for the sum of yields of residuals in isomeric and ground state.

Table 1 shows the results of the comparison of the experimental data and cross-sections calculated using the TALYS code and the comparison of measured cross-sections with the evaluated data from the PADF. The comparison was done using 19,253 experimental points from the various measured data sets involving 162 target nuclei from Mg to Bi and incident proton energies from the minimal available energy up to 150 MeV. Deviations from the experimental data are significantly reduced by the present evaluation. **Figure 1** shows the ratio of cross-sections calculated by the TALYS code without any corrections. The agreement of the evaluated data

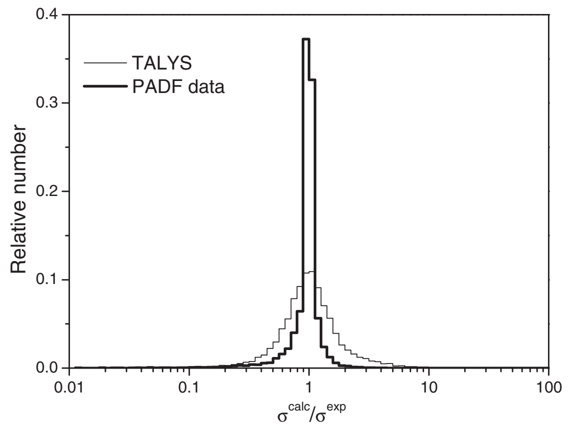

Fig. 1 The ratio of cross-sections calculated by TALYS to measured cross-sections (thin solid line) and the ratio of corrected and evaluated cross-sections from PADF to measured cross-sections (thick solid line)

Corrections involved 19,253 experimental points for 735 nuclear reactions and 162 target nuclei from Mg to Bi at the incident proton energy up to 150 MeV from 1434 EXFOR data sets. Points on the graph are combined by histograms for the best view.

with the experiments is noticeably better than that with the results of the calculation. An observed big difference between evaluated and measured data (ratio $\sigma^{calc}/\sigma^{exp}$ less than 0.5 and more than 2 in Fig. 1) is mainly originated from the difference of results of measurements obtained by the various authors for the same nucleus and the nuclear reaction. **Figure 2** shows examples of calculated cross-sections and evaluated cross-sections from the PADF. The non-smooth structure of evaluated cross-sections in Fig. 2 results from the fitting of the experimental data.

III. Content of PADF

The file is written in the ENDF-6 format.[30)] The file MF 3 and the section MT 5 were used to record the sum of cross-sections for individual reactions. Yields of residuals are written in the MF 6 file and the MT 5 section. The number of neutron deficient isotopes for each element, included in the PADF, did not allow the use of the common rule for material definition from Ref. 30). In the PADF, target nuclei are identified by the MAT numbers taken from the radioactive data file JEFF-3.1/RDD file.[3)]

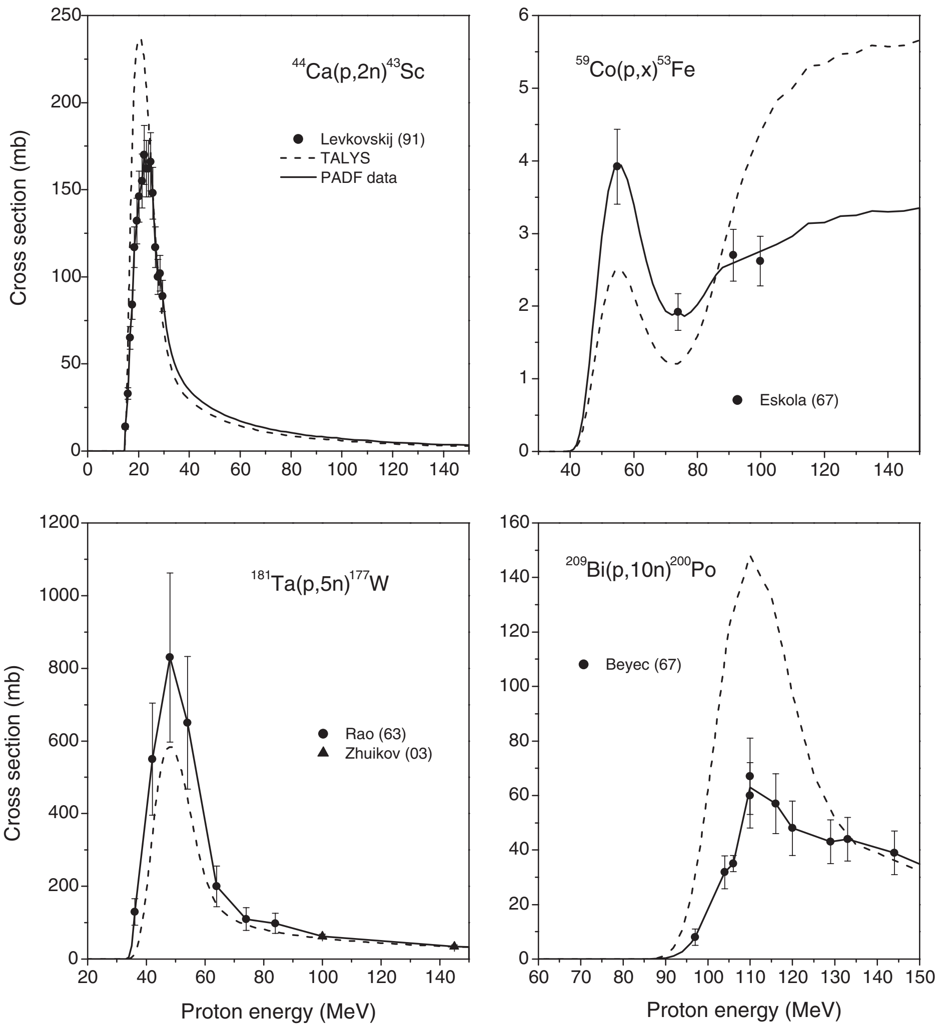

Fig. 2 Examples of cross-sections calculated by TALYS (dashed line) and evaluated data from PADF (solid line) Experimental data are taken from EXFOR.

The Proton Activation Data File contains 418,575 excitation functions for nuclear reactions at proton energies up to 150 MeV. Data are available for 2,355 stable target nuclei and unstable target nuclei with half-life ($T_{1/2}$) more than 1 s with atomic numbers between 12 and 88. The calculation of cross-sections for stable target nuclei and unstable nuclei with the half-life more than 10 min was performed using the TALYS code. Cross-sections for target nuclei with the decay half-life $1\,\text{s} < T_{1/2} < 10\,\text{min}$ were obtained using the ALICE/ASH code. The isomeric residuals are presented in the PADF for nuclei with $T_{1/2} > 1$ s. Also, the PADF includes calculated production cross-sections for the secondary neutrons, protons, deuterons, tritons, ^{3}He nuclei, α-particles, and γ-rays.

IV. Results

The Proton Activation Data File (PADF) has been composed. It contains 418,575 excitation functions of nuclear reactions for 2,355 target nuclei from Mg to Ra at proton energies up to 150 MeV. The data are available for stable and unstable target nuclei including isomeric targets with the half-life more than one second.

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References

1) P. Obložinský, "Recent advances in the ENDF/B Library," *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Santa Fe, New Mexico, USA, September 26–October 1, 2004, 165 (2004), http://www.nndc.bnl.gov/exfor4/endf00.htm.

2) Y. Watanabe, T. Fukahori, K. Kosako *et al.*, "Nuclear Data Evaluations for JENDL High-Energy File," *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Santa Fe, New Mexico, USA, September 26–October 1, 2004, 326 (2004), http://wwwndc.tokai-sc.jaea.go.jp/ftpnd/jendl/jendl-he-2004.html.

3) A. J. Koning, O. Bersillon, R. A. Forrest *et al.*, "Status of the JEFF Nuclear Data Library," *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Santa Fe, New Mexico, USA, September 26–October 1, 2004, 177 (2004), http://www.nea.fr/dbforms/data/eva/evatapes/jeff_31/.

4) A. J. Koning, S. Hilaire, M. C. Duijvestijn, *TALYS-0.64. A Nuclear Reaction Program. User Manual*, NRG Report 21297/04.62741/P FAI/AK/AK, Nuclear Research and Consultancy Group (NRG) December 5, 2004 (2004).

5) C. H. M. Broeders, A. Yu. Konobeyev, Yu. A. Korovin *et al.*, *ALICE/ASH–Pre-Compound and Evaporation Model Code System for Calculation of Excitation Functions, Energy and Angular Distributions of Emitted Particles in Nuclear Reactions at Intermediate Energies*, FZKA 7183, Forschungszentrum Karlsruhe, May, 2006 (2006), http://bibliothek.fzk.de/zb/abstracts/7183.htm.

6) C. H. M. Broeders, A. Yu. Konobeyev, L. Mercatali, "Global comparison of TALYS and ALICE code calculations and evaluated data from ENDF/B, JENDL, FENDL, and JEFF files with measured neutron induced reaction cross-sections at energies above 0.1 MeV," *J. Nucl. Radiochem. Sci.*, **7**, 1 (2006).

7) A. J. Koning, M. C. Duijvestijn, "A global pre-equilibrium analysis from 7 to 200 MeV based on the optical model potential," *Nucl. Phys.*, **A744**, 15 (2004).

8) W. Hauser, H. Feshbach, "The inelastic scattering of neutrons," *Phys. Rev.*, **87**, 366 (1952).

9) C. K. Kalbach Walker, *PRECO-2000: Exciton Model Preequilibrium Code with Direct Reactions*, March, 2001 (2001), http://www.nndc.bnl.gov/nndcscr/model-codes/preco-2000/index.html.

10) J. Raynal, *Notes on ECIS94*, CEA-N-2772, Service de Physique et Techniques Nucleaires (1994).

11) M. Blann, H. K. Vonach, "Global test of modified precompound decay models," *Phys. Rev.*, **C28**, 1475 (1983).

12) V. F. Weisskopf, D. H. Ewing, "On the yield of nuclear reactions with heavy elements," *Phys. Rev.*, **57**, 472 (1940).

13) Yu. A. Korovin, A. Yu. Konobeyev, P. E. Pereslavtsev, "Database development for analysis of accelerator-driven systems," *Progr. Nucl. Energy*, **40**, 673 (2002).

14) A. Iwamoto, K. Harada, "Mechanism of cluster emission in nucleon-induced preequilibrium reactions," *Phys. Rev.*, **C26**, 1821 (1982).

15) K. Sato, A. Iwamoto, K. Harada, "Pre-equilibrium emission of light composite particles in the framework of the exciton model," *Phys. Rev.*, **C28**, 1527 (1983).

16) A. I. Dityuk, A. Yu. Konobeyev, V. P. Lunev *et al.*, *New Advanced Version of Computer Code ALICE-IPPE*, INDC(CCP)-410 (1998).

17) C. H. M. Broeders, A. Yu. Konobeyev, "Evaluation of ^{4}He production cross-section for tantalum, tungsten and gold irradiated with neutrons and protons at the energies up to 1 GeV," *Nucl. Instr. Meth. Phys. Res.*, **B234**, 387 (2005).

18) C. H. M. Broeders, A. Yu. Konobeyev, "Phenomenological model for non-equilibrium deuteron emission in nucleon induced reactions," *Kerntechnik*, **70**, 260 (2005).

19) A. J. Koning, J. P. Delaroche, "Local and global nucleon optical models from 1 keV to 200 MeV," *Nucl. Phys.*, **A713**, 231 (2003).

20) A. V. Ignatyuk, G. N. Smirenkin, A. S. Tishin, "Phenomenological description of the energy dependence of the level density parameter," *Sov. J. Nucl. Phys.*, **21**, 255 (1975).

21) S. Goriely, F. Tondeur, J. M. Pearson, "A Hartree–Fock Nuclear Mass Table," *Atomic Data and Nuclear Data Tables*, **77**, 311 (2001).

22) P. Demetriou, S. Goriely, "Microscopic nuclear level densities for practical applications," *Nucl. Phys.*, **A695**, 95 (2001).

23) T. Belgya, O. Bersillon, R. Capote *et al.*, *Handbook for Calculations of Nuclear Reaction Data, RIPL-2*. IAEA-TECDOC-1506, IAEA, Vienna (2006), http://www-nds.iaea.org/RIPL-2/.

24) A. V. Ignatyuk, K. K. Istekov, G. N. Smirenkin, "Role of collective effects in the systematics of nuclear level densities," *Sov. J. Nucl. Phys.*, **29**, 450 (1979).

25) A. V. Ignatyuk, R. Capote, *Nuclear Level Densities, Handbook for Calculations of Nuclear Reaction Data, RIPL-2*, IAEA-TECDOC-1506, 85 (2006), http://www-nds.iaea.org/RIPL-2/handbook.html.

26) C. H. M. Broeders, A. Yu. Konobeyev, L. Mercatali, "Uncertainty in cross-section calculations for reactions induced by neutrons with energy above 0.1 MeV," *Kerntechnik*, **71**, 174 (2006).

27) C. H. M. Broeders, A. Yu. Konobeyev, L. Mercatali, *An Extensive Assessment of the Predictive Capabilities of Different Nuclear Models for the Calculation of Proton Induced Reac-*

tion Cross-Sections up to 150 MeV. Actinide and Fission Product Partitioning and Transmutation, Ninth Information Exchange Meeting, Nîmes, France, 25–29 September 2006 (2006), http://inrwww.fzk.de/nimes2006_mercatali_paper.pdf.

28) R. Michel, R. Bodemann, H. Busemann *et al.*, "Cross sections for the production of residual nuclides by low- and medium-energy protons from the target elements C, N, O, Mg, Al, Si, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Sr, Y, Zr, Nb, Ba and Au," *Nucl. Instr. Meth. Phys. Res.*, **B129**, 153 (1997).

29) C. H. M. Broeders, A. Yu. Konobeyev, "Improvement in simulation of equilibrium particle emission using intranuclear cascade evaporation model," *Nucl. Instr. Meth. Phys. Res.*, **A550**, 241 (2005).

30) M. Herman (ed.), *ENDF-6 Formats Manual*, BNL-NCS-44945-05-Rev. Document ENDF-102, National Nuclear Data Center, Brookhaven National Laboratory (BNL) (2005).