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Evaluation of activation nuclear data in the energy region 150 MeV to 1 GeV

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Abstract. An updated and extended version of the Intermediate Energy Activation File IEAF-2001 is currently under development. It will contain neutron-induced activation and transmutation cross sections for target nuclides Z = 1 to 84 and neutron energies up to 1 GeV. Below 150 MeV, the cross section data are evaluated on the basis of nuclear model calculations making use of experimental data whenever available. Above 150 MeV to 1 GeV, the evaluation is based on intranuclear cascade model calculations. This work describes the evaluation and preparation of the data in the upper energy region 150 MeV to 1 GeV. The analysis and identification of best models and tools for the cross section calculation was performed using all experimental data for nucleon induced reactions available up-to-date in EXFOR. Intranuclear cascade models used for comparison are all those included in MCNPX and CASCADE/INPE codes. A thorough comparative analysis of model calculations for more than 4000 experimental points corresponding to approximately 1000 reactions on target nuclei with atomic numbers from 6 to 83 was performed using different statistical and correlation criteria. The models giving the best agreement with experimental data in each target nuclei mass range were identified and used for subsequent calculation of neutron-induced cross sections for 684 nuclei in the energy range 150 MeV–1 GeV. The resulting cross section data were processed in the ENDF-6 data format making use of the MT = 5 (neutron, anything) option with the excitation functions stored in files section MF = 3 and the product nuclide vectors in MF = 6 (LAW = 0) to allow processing of the data with the NJOY code.

1 Introduction

An increasing research activity related to transmutation of radioactive nuclear waste in Accelerator-Driven Systems (ADS) and fusion-based systems requires the precise knowledge of production cross sections for a wide range of stable and unstable nuclei produced under the irradiation by intermediate and high energy particles. These activation data serve as a basis for evaluating the transmutation potential of dedicated installations as well as for studying the radiation endurance of structural materials in such installations. In addition, such data may find an application in the nuclear astrophysics when estimating production rates of heavy isotopes.

The majority of nuclear data libraries contain data for neutron-induced reactions up to 150 MeV. It emphasizes the activities on updating and extending of the Intermediate Energy Activation File IEAF-2001 [1]. Previous version of the library contained the neutron-induced reaction data up to 150 MeV, while the new one will contain neutron-induced activation and transmutation reaction cross sections for target nuclei Z = 1 to 84 and neutron energies up to 1 GeV. Below 150 MeV, the cross section data were obtained using nuclear models and available experimental data. At neutron energies from 150 MeV to 1 GeV, the evaluation is based on intranuclear cascade model calculations. The extension of the upper energy limit to 1 GeV significantly increases the time necessary for calculation and evaluation of cross sections since the number of open nuclear reaction channels rapidly grows. This report describes the evaluation and preparation of the data in the energy region 150 MeV to 1 GeV.

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2 Methods

2.1 Library structure

The upper energy part of the forthcoming version of Intermediate Energy Activation File represents the library with neutron activation data at primary neutron energies from 150 MeV to 1 GeV. The library consists of 684 files for nuclei from ^{1}H to ^{210}Po containing total reaction cross sections and multiplicities of nuclides produced in neutron-induced reactions.

2.2 Nuclear models to be compared

At present, to describe the processes of interactions of intermediate and high energy nucleons with matter various quasi-classical models are used. As a rule, the range of model applicability is defined by the energy of primary particle or the mass of target nucleus. The intranuclear cascade models, as a rule, handle interactions from GeV energy region down to 150–200 MeV [2], followed by various preequilibrium models based on exciton de-excitation [3]. Final de-excitation of the nucleus is described by the Hauser-Feshbach [4] or Weisskopf-Eving [5] statistical models. For nuclei that might undergo fission, the de-excitation process is characterized by the competition of the particle evaporation and the fission, the latter is described by corresponding high-energy approaches [6, 7].

A general-purpose Monte Carlo transport code MCNPX v.2.5.0 [8] was used to calculate production yields and cross sections. The code optionally may calculate singular nuclear interactions (i.e., without transport of particles) by using various combinations of intranuclear cascade, preequilibrium, evaporation and fission models. Fast cascade stage is described by Bertini [9], ISABEL [10] and INCL4 [11] models which may be combined with multistep preequilibrium model. The statistical processes are handled by Dresner evaporation [12], ABLA [13] fission-evaporation and ORNL [6] and RAL [7] fission models. In addition, the CEM2k model [14] (or its successor CEM03 [15] replacing it in the last version of MCNPX 2.6.c) including all reaction stages might be chosen.

The code CASCADE/INPE [16] based on the Dubna intranuclear cascade model [17] treats all the reaction stages in a manner similar to CEM2k. This code has also been chosen for comparative analysis.

The statistical comparative analysis of various model combinations from MCNPX and CASCADE/INPE has been performed targeting to identify the optimal model combination for neutron activation data calculations depending on the mass of target nucleus. The analysis has been based on all available in EXFOR [18] experimental data on neutron- and proton-induced production cross sections and yields and involved the minimization of deviation criteria of calculated data from experimental ones. It should be pointed out, that at energies of interest, the majority of experimental data are proton-induced. However, as a rule, neutron- and proton- induced cross sections are close to each other at intermediate and high energies. Thus the use of proton experimental data to verify models for neutron calculations is expedient.

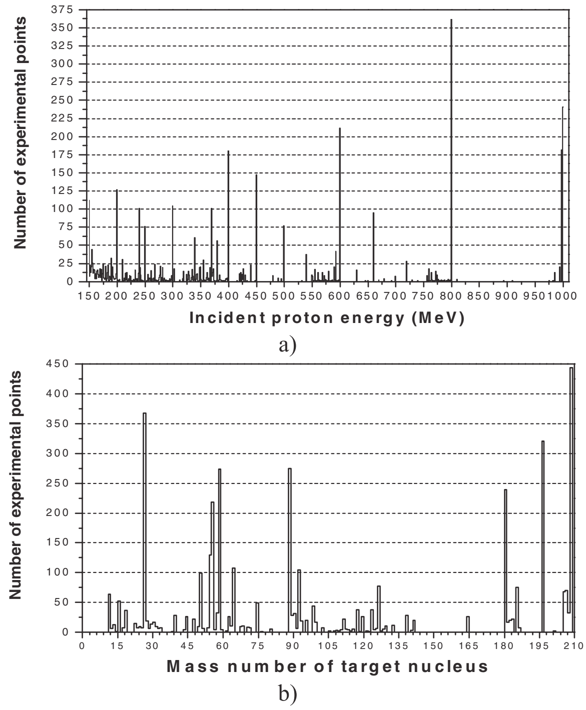

Fig. 1. Experimental points distributions over primary proton energy (a) and target mass number (b).

2.3 Experimental database and deviation criteria

The comparative analysis was performed for 3999 proton experimental points taken from EXFOR for more than 1000 (p,xnypza) nuclear reactions with targets ranging from Z = 6 to 84 and energies 150–1000 MeV.

The distributions of experimental datasets over primary proton energy and target mass number are shown in figure 1. The most informative energy region is turned out to be between 150 and 450 MeV (60% from the total number of experimental points) as well as separate energy points 500, 600, 660, 800 and 1000 MeV. As for target masses, the highest numbers of experimental data points were found for ^{27}Al and ^{208}Pb (368 and 444, accordingly).

The criteria to minimize were chosen to be the linear combination and the product of the two following deviation criteria:

$$F = 10^{\sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\lg\sigma_i^{\exp} - \lg\sigma_i^{calc}\right)^2}} \tag{1}$$

where N stands for the total number of experimental points. $\sigma_i^{\exp}$ σ_i^{calc} are the experimentally measured and calculated cross sections, and

$$H = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\frac{\sigma_i^{\exp} - \sigma_i^{cqlc}}{\Delta\sigma_i^{\exp}}\right)^2} \tag{2}$$

where $\Delta\sigma_i^{\exp}$ represents the experimental error.

In such case the F criterion allows to adequately and reliably estimate the correlation closeness between calculated and experimental data, while H accounts for experimental error.

3 Results and discussion

3.1 Results of deviation criteria analysis

To choose optimal model depending on target nucleus mass, the whole experimental dataset was split on the approximately equal mass subsets. Each diapason comprises about 400 experimental points. For each point, the production yield cross sections were calculated using all eight model combinations. Next step of analysis represented the calculation of deviation factors for each reaction. The linear combination of F and H criterions was formed by summing them up with weights equal to the values of each factor for all models in the considered target mass range. The products of F and H with the same weights were calculated as well. When calculating H the experimental point with zero on non-available experimental error was expunged.

The results of the analysis are shown in figure 2. For the light nuclei (A < 6) the analysis was not performed due to restricted applicability of the majority of calculation models.

The CASCADE/INPE code might calculate production cross sections for targets with A ≥ 12. Thus the MCNPX was applied to calculate neutron activation cross sections in this mass range. Moreover, for A < 4 the code uses interpolation tables to calculate cross sections such as (n,p), (n,d), (n,t), (n,^{3}He), (n,α). The production yields of residual nuclei were obtained using these reaction cross sections.

The best calculation models for each target mass range producing cross sections most close to experimental ones are summarized in table 1. The subsequent calculation of neutron activation data in the energy range 150 MeV to 1 GeV was performed according to this table.

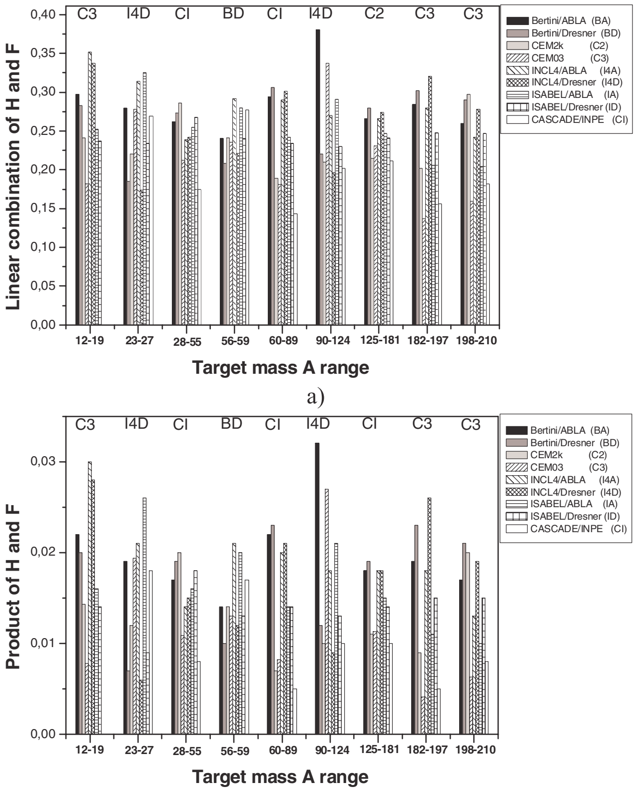

Fig. 2. Values of linear combination of normalized H and F factors (a) and product of normalized H and F factors (b) depending on target nucleus mass A range. The best model giving the minimum value of factor for each range is highlighted on the top of the plot.

Table 1. Calculation models recommended from statistical analysis.

Target nuclei range	Recommended model
^{1}H – ^{4}H	MCNPX interpolation tables
^{6}Li – ^{22}Ne	ISABEL/Dresner+CEM03
^{23}Na – ^{27}Al	INCL4/Dresner
^{28}Mg – ^{55}Co	CASCADE/INPE
^{56}Cu – ^{59}Ni	Bertini/ Dresner
^{60}Fe – ^{89}Zr	CASCADE/INPE
^{90}Sr – ^{124}Xe	INCL4/ Dresner + CASCADE/INPE
^{125}Sn – ^{181}Re	CEM2K
^{182}Hf – ^{210}Po	CEM03

Prior to calculations of neutron activation data, the significant efforts were directed to evaluating the reasonable ratio of accuracy to computational time. It is obvious that the higher energy of particle and mass of the target the longer time is needed to calculate all reaction open channels. Taking into account the computer park power dedicated for calculations, the statistics of 1,000,000 primary neutron interactions has chosen for each target nuclei. Only those cross sections obtained with statistical error less than 20% ("questionable" results according to MCNP/MCNPX terminology [19]) have been written to final files.

Table 2. Integral normalized values of $MMQ - COR_N$ for considered models for the whole target mass range for different models*.

Value of D	BA	BD	C2	C3	I4A	I4D	IA	ID	CI
				Calculation model					
40%	0.059	0.100	0.131	0.172	0.115	0.117	0.105	0.115	**0.259**
80%	0.097	0.114	0.101	0.164	0.151	0.106	0.131	0.100	**0.201**

*Abbreviations for models are the same as used for figure 2.

4 Results of correlation and least squares comparisons

In addition to statistical analysis the correlation and least squares tests have been performed.

The basis for comparison comprises proton experimental data for target nuclei ranging from Z = 6 to Z = 84 in the energy region 150 MeV to 1 GeV compared against cross sections calculated with all 9 considered model combinations. Due to a large amount of significantly heterogeneous experimental data (both on a number of data for different reactions and on degree of homogeneity inside each considered subrange) the additional analysis and selection has been performed with the help of the following criterion:

$$D = \left| \frac{\sigma_i^{calc} - \sigma_i^{\exp}}{\sigma_i^{\exp}} \right|. \tag{3}$$

The cross section data whose value of D exceeds 80% and 40% have been rejected. The selected estimations allowed excluding from consideration those calculated data where there are significant discrepancies from the experimental values resulting in the two datasets for D = 80% (2124 experimental points from 249 nuclear reactions) and D = 40% (1624 experimental points for 191 reactions).

The MATHCAD package [20] was used for analysis. The data have been represented as vectors of experimental Y_0 and calculated Y cross sections (each j-th model ($j = 1, 2, \ldots 8$) had corresponding vector Y). The standard deviations and covariations were calculated for each model using MATHCAD operators $stdev(Y_j)$ and $cvar(Y_0,Y_j)$. In addition, the vector MMQ_COR_i with components equal to products of inverse squared deviations and correlations of experimental and calculated data for each reaction was calculated (the maximum of each component of MMQ_COR_i characterizes the "best" agreement of experimental and calculated data with account of their correlation).

Since not for all the models and reactions the data have been available, the special regulating procedures have been applied to account for this fact.

For the frequency-based analysis the values of these vectors have been normalized to unity for each j-th reaction.

The MMQ_COR_N values obtained as a sum over all j-th reactions normalized to unity for each considered model is shown in table 2.

In general, the results of this analysis coincide with those shown in table 1, with exception that in the mass range corresponding to fission products (^{90}Sr – ^{124}Xe) CASCADE/INPE code reveals the best agreement with experimental data.

It should be pointed out that the basic criterion for above comparison is least squares deviation method, while the correlation data give the valuable information in the case of least squares deviation method gives close results. The frequency-based analysis is stable to initial data due to inclusion of integral characteristics of these data.

4.1 Neutron-induced data calculation and representation

The neutron induced production cross sections have been calculated with the step of 5 MeV in the energy region 150 to 250 MeV, with the step of 25 MeV between 250 and 600 MeV and with the step of 50 MeV above 600 MeV up to 1 GeV. All 684 files for isotopes from ^{1}H to ^{210}Po were recorded in ENDF-6 format [21] using MF = 3, MT = 5 and MF = 6, MT = 5 representation (i.e. keeping the representation the same as for previous version IEAF-2001). The section MT = 5 of the file MF = 3 contains the total nonelastic reaction cross sections while section MT = 5 of the file MF = 6 (LAW = 0) lists the energy dependent product yields per incident neutron for each reaction product. This scheme with minor changes allows the data to be processed by NJOY code [22] for subsequent application in the activation calculations.

5 Conclusions

The work aimed at analysing and defining the best models and tools for the neutron activation cross section calculation in the energy region 150 MeV to 1 GeV. The analysis was performed using all experimental data for nucleon induced reactions available up-to-date in EXFOR. Intranuclear cascade models used for comparison were all those included in MCNPX and CASCADE/INPE codes. A thorough comparative analysis of model calculations for about 4000 experimental points corresponding to approximately 1000 reactions on target nuclei with atomic numbers from 6 to 84 was performed using different statistical and correlation criteria. Models giving the best agreement with experimental data in various target nuclei mass ranges were identified and used for subsequent calculation of neutron-induced cross sections for 684 target nuclei in the energy range from 150 MeV to 1 GeV. Resulting cross section data were processed in the ENDF-6 data format.

The updated Intermediate Energy Activation File containing neutron induced activation data up to 1 GeV might become valuable tool for activation related research.

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