On the Accuracy of Different Theoretical Models in the Prediction of Neutron Induced Reaction Cross-Sections at Intermediate Energies

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Abstract

An extensive assessment of the predictive capabilities of different nuclear models in the reproduction of experimental activation and transmutation cross-sections for neutron induced reactions is presented. Experimental EXFOR data have been processed and treated in order to systematically analyze all the available measurements for target nuclei from ^{27}Al to ^{209}Bi with energy of the projectile above 0.1 MeV. Experimental data have been compared with the correspondent simulations performed by means of the TALYS code and the ALICE/ASH code using different models for the description of the nuclear level densities at equilibrium states, these being both phenomenological and microscopic ones. The comparison between measurements and calculations is quantified by means of different statistical deviation factors, which are given as functions of the target nuclei mass number and of different channels. Recommendations are provided to the users on the best combinations of codes and models to optimize the accuracy of the simulations. Furthermore, the calculation of a of a "mean model error" deviation factor has also been performed, making the results of this work also useful for the construction of covariance matrixes of "model deficiencies" for a wide number of nuclides.

KEYWORDS: Nuclear models, Cross-sections, TALYS, ALICE/ASH

1. Introduction

The accurate simulation of neutron interactions with matter represents an essential requirement for a wide range of applications from the basic science up to the design of advanced nuclear energy systems. In recent years, considerable improvements have been achieved in the capabilities of theoretical models (implemented in different codes) to predict cross-section values. However, despite such big efforts in the field of cross-section theory, a comprehensive and satisfactory methodology to accurately calculate all the nuclear cross-sections for any target nuclei over the widest energy range is not available and a systematic assessment of the predictive capabilities of the most advanced nuclear models and computational tools which have direct relation in the generation of nuclear data files is still needed. This paper deals with the evaluation of the uncertainty associated to the calculation of activation and transmutation cross-sections for

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neutron induced reactions. The predictive power of different nuclear models relative to the description of the nuclear level densities at equilibrium states as implemented in the TALYS code and in the ALICE/ASH code has been assessed throughout a statistical and systematic comparison between theoretical results and experimental data over the target nuclei mass number range 27<A<209.

2. Theoretical Calculations

2.1 The TALYS and ALICE/ASH Codes

In this work all the calculations have been performed by means of the TALYS code and the ALICE/ASH code [1-2]. The investigation of the performance of these simulation tools is motivated by their extensive use within the international community for the generation of nuclear data files.

As far as the TALYS code, the pre-equilibrium particle emission is described using the two-component exciton model [3]. The model implements new expressions for internal transition rates and new parametrization of the average square matrix element for the residual interaction obtained using the optical model potential of Koning et el. [4]. The phenomenological model is used for the description of the pre-equilibrium complex particle emission [5]. The contribution of direct processes in inelastic scattering is calculated using the ECIS-97 code incorporated in TALYS. The equilibrium particle emission is described by means of the Hauser-Feshbach model.

The ALICE/ASH code is a modified and advanced version of the ALICE code [6]. The geometry dependent hybrid model (GDH) is used in the description of the pre-equilibrium particle emission from nuclei [7]. Intranuclear transition rates are calculated using the effective cross-section of nucleon-nucleon interactions in the nuclear matter. The number of neutrons and protons for initial exciton states is calculated using realistic nucleon-nucleon interaction cross-sections in nucleus. The exciton coalescence model and the knock-out model are used for the description of the pre-equilibrium complex particle emission [8]. The equilibrium emission of particles is described by the Weisskopf-Ewing model without detailed consideration of angular momentum.

2.2 The Calculation of Nuclear Level Densities

For the purposes of the present work, both the TALYS and the ALICE/ASH codes have been used with default values of input parameters, with the exception of the parameters describing the particular model used for the nuclear level densities description. In particular, six different level density models have been considered, corresponding to the input parameters *ldmodel* equal to 1, 2 or 3 and *ldopt* equal to 0, 4 and 5 in the TALYS code and in the ALICE/ASH code respectively. Detailed information on these models can be found in [9-13]. The main features can be summarized as follows:

a. *ldmodel*1: Fermi gas model with the energy dependent level density parameter $a(U)$ without explicit description of the collective enhancement.
b. *ldmodel*2: Fermi gas model with the energy dependent level density parameter $a(U)$ with explicit description of the rotational and vibrational enhancement.

c. *ldmodel*3: Microscopic model based on the results of microscopic calculations performed by Goriely et al. using the Hartree-Fock-BCS model.
d. *ldopt*0: Fermi gas model with the dependent level density parameter $a=A/9$.
e. *ldopt*4: Fermi gas model with the energy dependent level density parameter $a(U)$.
f. *ldopt*5: Superfluid nuclear model.

In the following we will refer to the results of the calculations performed with the different models with the following notation: *ldmodel*1=IST1, *ldmodel*2=IST-C, *ldmodel*3=G, *ldopt*0=FG, *ldopt*4=IST2, *ldopt*5=SF.

In order to avoid inconsistencies between the results of the TALYS and the ALICE/ASH calculations caused by different values of total nonelastic cross-sections, the ALICE/ASH calculated cross-sections have been normalized on the values of nonelastic cross-sections calculated by the TALYS code.

3. Experimental Data

Experimental data to be compared with the calculated results have been extracted from EXFOR and presented in the C4 format by means of a processing procedure performed using the X4TOC4 code [14]. All the (n,xnypzα) reactions available have been considered with the exception of (n,γ), (n,np), (n,d) and (n,^{3}He). Out-dated and superceded measurements, measurements for targets containing natural mixture of isotopes as well as data for reactions with metastable products were also excluded from the consideration. As a result of this selection criterion the experimental data taken into consideration can be summarized as follows:

  ➤ Projectile: neutron
  ➤ Projectile energy range: $0.1 \div 64.4$ MeV
  ➤ Target range: $13 < Z < 83$
  ➤ Reactions considered: (n,n'), (n,p), (n,α), (n,t), (n,2n), (n,nα), (n,2p), (n,pα), (n,2α), (n,3n), (n,4n) and other reactions denoted as (n,x)
  ➤ Total number of experimental points (Z,A,E): 17937

As far as the energy distribution of the experimental points (Fig. 1), 30% of the total measurements considered are relative to the projectile energy in the range $14 \div 15$ MeV.

In order to quantify the quality of model calculations when comparing calculated cross-sections with experimental ones, four different point-wise deviation factors have been calculated as follows:

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

$$R = \frac{1}{N} \sum_{i=1}^{N} \frac{\sigma_i^{calc}}{\sigma_i^{exp}} \tag{2}$$

$$D = \frac{1}{N} \sum_{i=1}^{N} \left| \frac{\sigma_i^{exp} - \sigma_i^{calc}}{\sigma_i^{exp}} \right| \qquad (3)$$

$$F = 10^{\left(\frac{1}{N} \sum_{i=1}^{N} \left[\log(\sigma_i^{exp}) - \log(\sigma_i^{calc}) \right]^2 \right)^{1/2}} \qquad (4)$$

where σ_i^{exp} and $\Delta\sigma_i^{exp}$ are the measured cross-section and its uncertainty, σ_i^{calc} is the calculated cross-section and N is the number of experimental points.

To estimate the uncertainty in the calculated cross-sections a covariance matrix has been proposed, which takes into account the contribution to the uncertainty due to the failure of the model used for the calculations [15]. The matrix, which defines the "model deficiencies", is constructed using the mean model error δu extracted from the reproduction of experimental data by a given reaction model as follows:

$$M_{i,j}^{(def)} = C_{i,j} (\delta u)^2 \, \sigma_i^{calc}(E_i) \, \sigma_j^{calc}(E_j) \qquad (5)$$

where E_i and E_j are kinetic energies of primary particles and coefficients C_{ij} are defined in [15]. The square of the mean model error is used in the present work as an additional factor to estimate the quality of model calculations, as follows:

$$L = (\delta u)^2 = \frac{\sum_{i=1}^{N} w_i \left(\frac{\sigma_i^{calc} - \sigma_i^{exp}}{\sigma_i^{calc}} \right)^2}{\sum_{i=1}^{N} w_i} \qquad (6)$$

where:

$$w_i = \left(\frac{\sigma_i^{calc}}{\Delta\sigma_i^{exp}} \right)^2$$

Figure 1: Energy distribution of the experimental points.
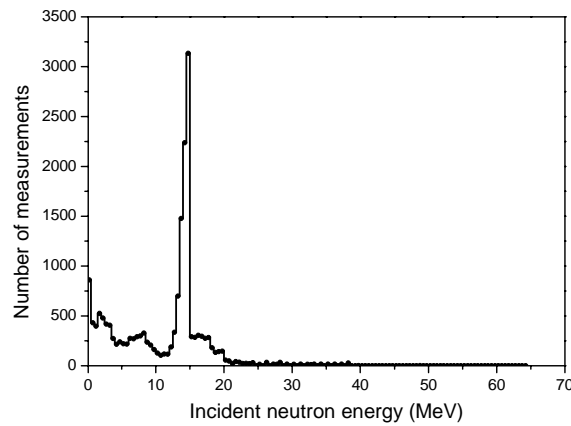
4. Results and Discussion

4.1 Comparison of Experimental Data with Nuclear Model Calculations

The limited volume of the paper does not allow to present the detailed information on the deviation factors calculated for all individual nuclei. Tables 1 through 3 provide a summary of the main findings of our work which can be used as preliminary recommendations on the use of various nuclear models implemented in the TALYS and ALICE/ASH codes.

When averaging the factors of Eq. (1-4,6) calculated for the target nuclei from ^{27}Al to ^{209}Bi over the entire energy range and over all the channels, the minimal value of the H factor is the one obtained using the IST1 model in the TALYS code (Table 1). In addition, Table 1 provides also results relative to two different mass number ranges of the target nuclei. Approximately, the division corresponds to the dominate contribution of equilibrium (A < 120) and precompound (A > 120) processes in the (n,p) and (n,α) reactions, which together represent nearly 58% of the total experimental points considered in this work. The use of the TALYS code with the IST1 model applied for the nuclear level density calculation shows the best results for A < 120. On the contrary, the minimal H value in the target mass number range A > 120 corresponds to the use of the ALICE/ASH code with the superfluid model (SF).

Table 1: Deviation factors for nuclei from ^{27}Al to ^{209}Bi calculated using the TALYS and ALICE/ASH codes.

Factors	TALYS			ALICE/ASH		
	IST1	IST-C	G	FG	**IST2**	**SF**
Targets with atomic mass number 27 ≤ A < 209						
H	<u>10.35</u>	30.60	12.61	16.18	28.87	13.78
R	1.26	1.60	1.29	1.05	0.79	1.00
D	0.50	1.05	0.57	0.53	0.64	0.52
F	2.09	2.88	2.14	2.81	18.31	3.55
L	0.14	0.59	0.21	0.29	0.60	0.23
Number of points	17296	17270	17295	17136	17050	17122
27 ≤ A < 120						
H	<u>10.33</u>	29.34	12.01	17.50	31.38	14.88
R	1.25	1.57	1.27	1.06	0.78	1.01
D	0.50	1.06	0.56	0.56	0.68	0.56
F	2.10	2.97	2.15	2.93	22.39	3.76
L	0.13	0.55	0.18	0.29	0.60	0.24
Number of points	14467	14441	14466	14313	14277	14304
120 ≤ A ≤ 209						
H	10.45	36.39	15.31	6.15	7.38	<u>5.44</u>
R	1.32	1.77	1.38	1.03	0.84	0.95
D	0.50	0.95	0.58	0.36	0.42	0.34
F	2.03	2.41	2.08	2.19	4.42	2.49
L	0.27	0.77	0.44	0.14	0.29	0.13
Number of points	2829	2829	2829	2823	2773	2818

In Fig. 2 the H and R deviation factors are plotted as functions of the target nuclei mass number. As far as the TALYS calculations, one can observe that the IST1 and the G models are somehow in competition, while the IST-C model appears to be the less accurate. In the case of the ALICE/ASH calculations, the SF model is the one with the best capability to extensively predict experimental data. Furthermore, when comparing the best model of TALYS with the best model of ALICE/ASH (Fig. 3) we observe the IST1 and the SF models perform better for light (A $\sim \leq 140$) target nuclei and heavy (A $\sim \geq 140$) target nuclei respectively.

In the case of the deviation factors calculated as average by single reaction channel (Table 2), the use of the TALYS code provides the minimum values for nuclei with A < 120 with the exception of the (n,t) channel. In the range A > 120 the ALICE code provides generally a better accuracy.

Table 2: The H deviation factor calculated for different channels.

Reaction	TALYS			ALICE/ASH		
	IST1	IST-C	G	FG	IST2	SF
Targets with atomic mass number $27 \leq A < 120$						
(n,n')	12.77	12.48	12.79	13.00	16.71	13.00
(n,2n)	13.56	14.94	13.32	31.48	60.77	22.62
(n,3n)	13.35	3.04	15.27	11.62	6.24	11.67
(n,p)	8.22	28.07	9.31	10.93	19.38	12.74
(n,α)	7.91	44.57	13.76	10.71	11.23	10.50
(n,t)	20.52	30.62	21.04	5.12	5.70	5.09
Others	7.20	4.83	7.98	9.98	15.62	10.52
All reactions	10.33	29.34	12.01	17.50	31.38	14.88
$120 \leq A \leq 209$						
(n,n')	2.17	2.62	2.22	2.11	5.68	2.52
(n,2n)	3.81	4.33	3.96	5.09	7.60	4.94
(n,3n)	4.65	4.76	5.22	12.49	10.92	5.98
(n,p)	17.80	23.60	18.29	32.81	6.99	6.53
(n,α)	11.56	96.20	33.74	5.45	6.64	5.66
(n,t)	41.81	103.70	42.07	4.03	4.08	4.03
Others	4.80	4.56	5.63	9.04	8.88	6.91
All reactions	10.45	36.39	15.31	6.15	7.38	5.44

4.2 Comparison of Experimental Data with Evaluated Data

This work is mainly devoted to the assessment of the uncertainty associated to the calculation of neutron induced cross-sections using modern theoretical approaches. However, a comparison between experimental data and evaluated data has also been performed in order to quantify the gain in accuracy due to the evaluation process with respect with the uncertainty due to nuclear model calculations. Measured cross-sections have been compared with the most complete and modern evaluated data sets. When analyzing results relative to this comparison (Table 3) one may observe that the data from JEFF-3.0/A (European Activation File [16]) present minimal values of deviation factors when compared with other libraries. The comparison in between

factors of Table 1 with the ones of Table 3 shows generally a certain gain in accuracy in the description of experimental data presented by evaluations in the case of the JEFF-3.0/A, ENDF/B-VI and JENDL.3.3 libraries.

In order to investigate the gain in accuracy one may additionally obtain in the case of nuclear model calculations, a procedure for the optimization of the H factor has been applied, consisting in the minimization of the calculated excitation function multiplied by a coefficient which was assumed to vary in the range 0.5÷1.5. In Table 4 we summarize the results of this optimization relative to the (n,p) and (n,α) reactions. Corrected values of H are significantly lower (by about 40%) with respect to the original ones obtained using the TALYS code with the IST1 model. Furthermore, after the correction the calculated cross-sections seem to globally better reproduce experimental data than the JEFF-3.0/A evaluated data.

Table 3. Deviation factors for different mass range of target nuclei calculated using evaluated cross-sections from different nuclear data libraries.

Factors	ENDF/B-VI.8	FENDL-2/A	JEFF-3.0/A	JENDL-3.2	JENDL-3.3
Targets with atomic mass number $27 \leq A < 120$					
H	8.13	76.26	7.05	24.42	8.28
R	1.09	2.17	1.23	1.83	1.69
D	0.26	1.34	0.44	1.02	0.88
F	1.48	2.10	1.91	2.05	2.03
L	0.06	0.87	0.06	0.43	0.08
Number of points	10497	12591	12542	13802	13516
$120 \leq A \leq 209$					
H	14.12	6.29	6.10	7.45	7.40
R	1.34	1.14	1.11	1.19	1.19
D	0.54	0.33	0.26	0.38	0.38
F	2.30	2.03	1.94	2.22	2.22
L	0.41	0.14	0.14	0.19	0.19
Number of points	1693	2571	2548	1836	1902

Table 4. The H deviation factor resulting from the minimization procedure (corrected) compared with the ones relative to the comparison of experimental data with nuclear model calculations and with evaluations

Factor	TALYS		JEFF-3/A
	IST1	Corrected	
(n,p) reaction			
H	9.63	5.31	6.97
Number of points	6216	6216	614
(n,α) reaction			
H	7.96	4.77	6.34
Number of points	3846	3846	4375

Figure 2: H and R deviation factors as functions of the target nuclei mass number in the case of TALYS and ALICE/ASH calculations
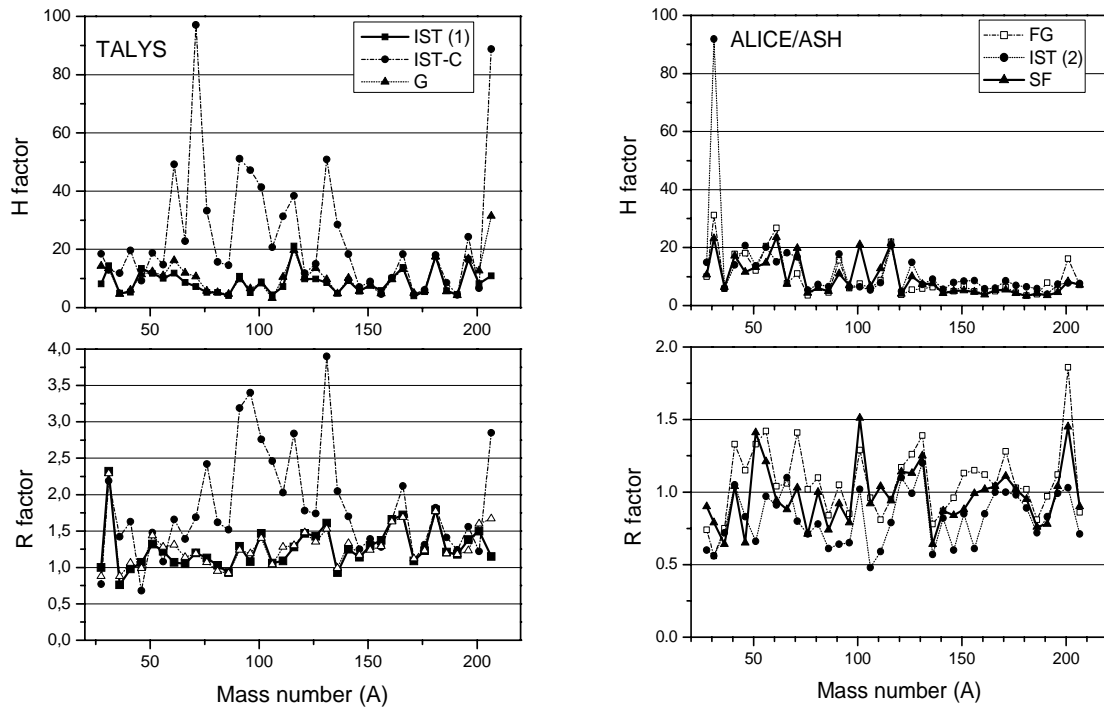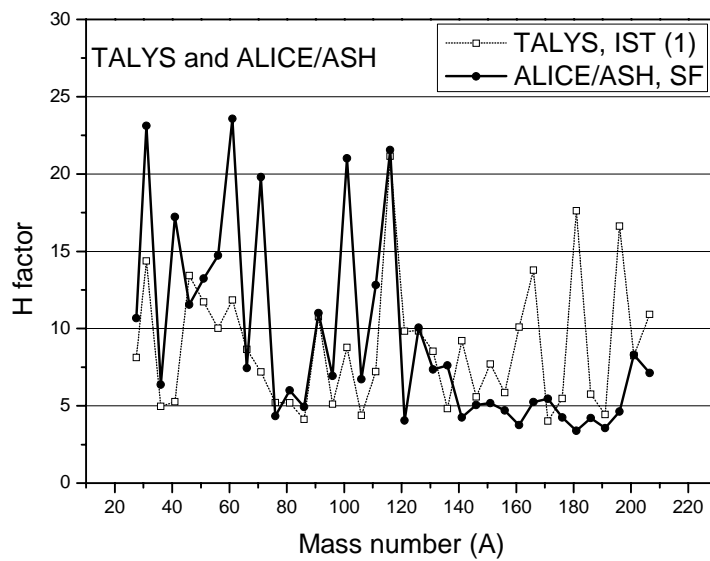

Figure 3: The H factor as function of the target nuclei mass number in the cases of IST1 and SF calculations

5. Conclusion

We have presented a large scale comparison between experimental and calculated data relative to activation and transmutation cross-sections for neutron induced reactions in the energy range 0.1÷64.4 MeV. The comparative analysis was performed for target nuclei from ^{27}Al to ^{209}Bi. This range of nuclides includes important structural materials used in fission reactors, fusion units as well as in advanced nuclear energy systems (i.e. ADS's). Different models for the description of the nuclear level densities as available in the TALYS code and in the ALICE/ASH code have been used in the calculations and recommendation have been provided on the best combination of models and codes in order to optimize the accuracy of the simulations, as for different target nuclei as for different channels. The gain in accuracy due to the evaluation process with respect with the uncertainty due to nuclear model calculations has also been assessed by means of comparison between the most common and recent versions of data libraries and the set of experimental data. Finally, a pointwise "mean model error" deviation factor has been evaluated, which can be used for the construction of covariance matrixes of "model deficiencies".

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