REPORT ON EAF RELATED TOOLS

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Abstract

Progress of work on improved methods of nuclear model calculation for nuclear activation data, carried out at IFIN-HH since the Workshop on Activation Data – EAF 2001, is reported. In order to provide accurate predictions for the fast-neutron reactions cross sections of further interest for EAF-2003, no use of normalization or free parameters are allowed. Model calculations looking for completion of this requirement have been validated by analysis of activation cross sections based on (i) the unitary use of the common model parameters for different mechanisms, (ii) the use of consistent sets of input parameters which are determined by analyses of various independent experimental data, and (iii) the unitary account of a whole body of related experimental for isotope chains of neighboring elements.

I. INTRODUCTION

Improved nuclear model calculation methods for nuclear activation data have been carried out by using the exciton and the Geometry-Dependent Hybrid (GDH) semi-classical models for pre-equilibrium emission (PE) and the Hauser-Feshbach statistical model (SM) within the computer code STAPRE-H951. The points on which we have focused our work have been:

(i) description of the \((n,p)\) and \((n,\alpha)\) reaction excitation functions above the common 15 MeV value, which was recently proved\(^2\) to be not suitable provided by the usual semi-classic PE models;

(ii) the angular-momentum distribution of the nuclear-level density described, e.g. within the wide-used back-shifted Fermi gas (BSFG) model\(^3\), by the nuclear moment of inertia \(I\) which was found rather recently\(^4\) to be equal with only half of the rigid-body value \(I_r\) (usually it is still used, e.g. Ref.\(^5\), the value \(I=I_r\));

(iii) optical model potential (OMP) providing the \(\alpha\)-particle transmission coefficients, which is yet an open question of reaction cross-section calculations while the best approach for its description could be only a microscopic\(^6\) one.

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\(^1\)Report to the Workshop on Activation Data – EAF 2003, Prague, June 24-26, 2002.
\(^5\)S.F. Mughabghab and C. Dunford, Phys. Rev. Lett. 82, 4083 (1998)
The development in the meantime at IFIN-HH of a novel partial level-density formalism\(^7\), e.g. the recent IAEA Reference Input Parameter Library (RIPL)\(^8\), and an improved version of corresponding computer code PLD\(^9\) have contributed to progress on the above item (i), as it was shown during the Workshop on Activation Data – EAF-2001 and consequent papers\(^10,11\) on analysis of IRMM accurate measurements of fast-neutrons induced reactions on \(^{51}\)V. The main difference concerns the GDH specific account of the nuclear-density distribution by means of average quantities over the densities corresponding to the entrance-channel trajectory. Among other main assumptions of the model, no free parameter comes in but \(\alpha\)-particle pre-formation probability and the \(\alpha\)-particle state density \(g_\alpha = A/10.36\text{ MeV}^{-1}\). Moreover, the updated PLD formalism of the particle-hole level density has been extended to non-uniform level spacings while various effects could be also optionally taken into account (e.g. the pairing and shell corrections in addition to the Pauli effect, the surface effects in case of the first step of multistep processes, and the \textit{continuum} effect). This particle-hole state density with energy-dependent s.p.s. densities has finally been included within GDH model.

During the Workshop on Activation Data – EAF-2001 (6-7 Nov. 2000, CE de Cadarache) it was stated that extension of the model calculations, requested by the development of the next library version EAF-2003, is conditioned by their validation in various mass regions. It is why the above-mentioned consistent model calculations have been carried on also for the Mo as well as Ni isotopes. In the former case, in spite of the existence of a large amount of measured data, there are yet many discrepancies between even recent data sets, while three basic evaluations performed in the last decade at well-known laboratories show wide differences, e.g. up to ~50\% for the \((n,p)\) reaction\(^12,13\) and ~65\% for the \((n,\alpha)\) reaction\(^13,14\). Strong questions still exist also in the latter case, e.g. concerning the analysis of the \(^{58}\)Ni(\(n,\alpha\))\(^{55}\)Fe reaction\(^15\), while the \(^{58}\)Ni(\(n,p\))\(^{55}\)Mn reaction cross section - requested for EAF-2003 - has been proved recently\(^16\) to be uncertain within a factor of 2 - 4.

In order to obtain confident calculated cross sections under these conditions, we have had to enlarge the analysis of statistical model parameters\(^17\) carried out already in this respect concerning quantities which are most important also for calculation of the isomeric cross section ratios\(^5\). They have been next involved in nuclear model calculations taking care for compensation of opposite effects, e.g. due to various less accurate parameter values, through:
(a) unitary use of the common model parameters for different concerned mechanisms, and of
(b) consistent sets of input parameters determined by various independent data analysis, and
(c) unitary account of a whole body of related experimental data for isotope chains or neighboring elements.

Finally, concerning the above item (iii), microscopic calculations are involved for \(\alpha\)-particle OMP potentials, validated through description of experimental \((\alpha,\alpha)\) angular distributions.

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\(^17\)V. Avrigeanu, Reports on visits at EC/JRC/IRMM, 2000-03-01/04-30; -09-16/11-15 (unpublished).
II. STATISTICAL MODEL PARAMETERS

A. Nuclear Level Density

1. Low excitation energies

Since the isomeric cross section analysis is quite sensitive with respect to the spin distribution of the level density\(^5\), and there are large data basis as well as new EC/JRC/IRMM accurate measurements\(^18\) of neutron-induced reaction cross-sections for \(^{59}\)Co, Ni and Mo isotopes from threshold to 20 MeV, it has been found useful an extended level-density analysis in these mass ranges. The *nuclear-level density* BSFG parameters \(a\) and \(\Delta\) obtained previously\(^10,17\) for isotopes in the atomic-mass ranges \(A=40-58\) and \(79-111\) by fit of the recent experimental low-lying discrete levels (ENSDF file on the BNL-Brookhaven web site) and s-wave nucleon resonance spacings \(D_0\) (the corresponding RIPL file on the NDS/IAEA-Vienna web site), corresponding to the \(I/I_r\) values 0.5, 0.75, and 1, is extended up to \(A=70\). It has been also carried out for \(A=40-70\) and \(79-111\) by taking into account a variable ratio \(I/I_r\) between 0.5 for ground states and 0.75 at the neutron binding energy\(^19\) (Fig. 1), for further check of these assumptions through comparison of calculated and experimental isomeric cross section ratios\(^18\).

\[ \begin{align*}
    &\text{BSFG: } \frac{I}{I_r} = 0.5 \\
    &\text{BSFG: } \frac{I}{I_r} = 0.5-0.75 (E = 0-8 \text{ MeV}) \\
    &\text{BSFG: } \frac{I}{I_r} = 0.75 \\
    &\text{BSFG: } \frac{I}{I_r} = 1
\end{align*} \]

Fig. 1. Values of the level-density parameter \(a\) obtained by fit of the ENSDF and RIPL data.

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Fig. 2. Systematics of the BSFG level-density parameter $a$ for $A=2-254$ for variable $I$.

In the meantime the analysis of the nuclear-level density parameters $a$ and $\Delta$ of the BSFG model has been carried out, for the last assumption for $I$, for 343 isotopes in the range $A=20-254$ (Fig. 2). The same fit of the most recent experimental low-lying discrete levels and $D_0$ values was involved in this respect. Tables of parameters and fitted quantities will be made available on the IFIN-HH web site following the approval of Association EURATOM–NASTI Steering Committee.

2. Higher-energy excitation

The modified version of the code STAPRE-H95 is using also a changed formula for the nuclear level density at the excitation energies above the nucleon binding energy. The previous approach involved the use of the BSFG model for description of the nuclear level density at excitation energies lower than 12 MeV (which is the excitation region in which the corresponding parameters are obtained by fitting recent experimental low-lying discrete levels and s-wave nucleon resonance spacings) and a transition to a realistic analytical formula. In the meantime various related methods have been introduced, e.g. by Koning and Chadwick$^{20}$ which has been followed with a small change.

Thus, firstly we have considered the more recent formalism of Junghans et al.$^{21}$ for the higher excitation energies $E^*$, which is centered around the calculation of the entropy $S$. Second, the normalization has been applied for the effective level density parameter $a_{eff}=S/4E^*$. This takes into account that the involved experimental data - i.e. the nucleon-resonance spacings - are related actually to spin-dependent level densities. We have found the difference between the shell correction thus derived and the "microscopic" correction $E_{mic}$ which is the actual reconsidered parameter in order to describe the high-energy behavior of the level-density parameter, around the maximum value of 2 MeV. This change is thus similar to the one corresponding to various mass formulas involved within the usual procedure. It has also motivated the choosing of the zero-value for the shell-effect correction taken into account in the partial-level density involved in the PE calculation for the higher excitation energies (for references see Ref.$^{19}$).

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We have chosen the asymptotic value for the level-density parameter $a$ neither the form based on the analysis of neutron resonance data nor that corresponding to the Woods-Saxon potential but the value $A/9$ MeV$^{-1}$. It is rather close to the value calculated for the spectrum of s.p.s. of the Nilsson potential$^{22}$, and close to recent microscopic results around $A\approx50$ and $A\approx110$.

Finally, we have adopted a normalization of the effective excitation energy not to the energies of the odd-odd nuclei but to those of the odd-$A$ nuclei. A transition range from the BSFG formula description to the approach adopted for the higher energies has been chosen between the binding energy and $E^*=15$ MeV mainly in order to have a smooth connection. Thus a good description was obtained for both the $s$-wave nucleon resonance data and the known experimental level density data above 10 MeV, firstly for nuclei in the ranges $A=55-70$ and 104-114 (Fig. 3) and then$^{19}$ for lighter nuclei with $A=24-41$.

Fig. 3. Comparison of experimental and calculated level densities$^{19}$ for nuclei with $A=55-114$.

B. Phenomenological Optical Model Potentials

OMP analysis should be the first phase of any study of fast-neutron induced reactions on Ni isotopes in order to avoid too many uncertainties in usual Hauser-Feshbach statistical model calculations (e.g. Ref. 15). Since various discrepancies exist between the recent GELINA total neutron cross sections of \(^{58,60,61}\)Ni isotopes 23 and other data sets as well as neutron OMP predictions, the SPRT method 24 was involved for the analysis of some frequently used parameter sets. The OMP parameter sets of Kawano et al. 25 have been validated for the isotopes \(^{58,60,62,64}\)Ni, while parameters were modified for the isotopes \(^{59}\)Co and \(^{61}\)Ni (Fig. 4).

Fig. 4. Comparison of calculated and experimental neutron total cross sections for \(^{58,62,64}\)Ni and \(^{59}\)Co.

The OMP for calculation of proton transmission coefficients on the residual nucleus \(^{58}\)Co has been established through the analysis of the available \(^{59}\)Co(p,\(\gamma\))\(^{60}\)Ni and \(^{59}\)Co(p,\(n\))\(^{58}\)Ni reaction cross sections up to \(E_p=6\) MeV, and total proton reaction cross sections on \(^{59}\)Co at \(E_p=10-20\) MeV (Fig. 5). A similar analysis is shown in Fig. 6 for the case of the target nucleus \(^{93}\)Nb, used for the validation of the proton transmission coefficients involved of the fast neutron induced reactions on the stable Mo isotopes.

The validation of the OMP for calculation of \(\alpha\)-particle transmission coefficients has been favored by the special case of the reaction \(^{58}\)Ni(n,\(\alpha\))\(^{55}\)Fe at incident energies up to \(\sim 5\) MeV, where only discrete are important in all residual nuclei. By comparison of the calculated 26 and recent cross sections 26 it has been found that the real well diffuseness of the OMP of Ref. 27 should be changed to 0.71 fm.

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Fig. 5. Comparison of calculated and measured $(p, \gamma)$, $(p,n)$, and proton reaction cross sections for $^{59}$Co.

Fig. 6. Comparison of calculated and measured $(p, \gamma)$ and $(p,n)$ reaction cross sections for $^{93}$Nb.
C. The $\gamma$-ray Strength Functions

The $\gamma$-ray strength functions $f_{E1}(E_\gamma)$ which are used for the calculation of the $\gamma$-ray transmission coefficients, have been obtained by means of a modified energy-dependent Breit-Wigner (EDBW) model\textsuperscript{28,29}. Moreover, systematic EDBW correction factors $F_{SR}$ were extended from\textsuperscript{30} $A=83$-105 to $A=41$-82 by analyzing the experimental average radiative widths $\Gamma_{\gamma0}^{exp}$ of the $s$-wave neutron resonances, and assuming that $F_{SR}=\Gamma_{\gamma0}^{exp}/\Gamma_{\gamma0}^{EDBW}$ (bottom right of Fig. 7).

Since there are large discrepancies between measured recently at IRMM-Geel\textsuperscript{31,32} and the evaluated data within RIPL, the $f_{E1}(E_\gamma)$ thus obtained have been checked within calculations of capture cross sections. The calculated and experimental cross sections of the reactions $^{58}$Ni$(n,\gamma)^{59}$Ni, $^{60}$Ni$(n,\gamma)^{61}$Ni, and $^{59}$Co$(n,\gamma)^{60}$Co were compared in the neutron energy range from keV to 2-3 MeV. The RIPL values for $\Gamma_{\gamma0}^{exp}$ lead to $f_{E1}(E_\gamma)$ strength functions that are too large while recent IRMM and ORNL values\textsuperscript{33} provide good agreement with the capture data (Fig. 7).

Fig. 7. Comparison of calculated and experimental neutron capture cross sections for $^{58,60}$Ni and $^{59}$Co.

III. ACTIVATION CROSS SECTION CALCULATIONS

A. Completion of the Analysis for $^{51}$V

In the frame of the cooperation between IFIN-HH and EC/JRC/IRMM-Geel it was initiated the analysis of recent IRMM accurate measurements of fast-neutrons induced reactions on $^{51}$V. It has been represented by consistent model-calculations, completed within IFIN-HH, concerning firstly the reactions $^{51}$V($n,n'\alpha$)$^{47}$Sc, $^{51}$V($n,\alpha$)$^{48}$Sc, $^{51}$V($n,p$)$^{51}$Ti, $^{51}$V($n,2n$)$^{50}$V, and $^{50}$V($n,\alpha$)$^{47}$Sc.

Additional analysis of the sensitivity of the calculated activation cross sections in front of, e.g., nuclear-level density parameters (Fig. 8), proved the high effects of the uncertainties of experimental data considered for determination of these parameters (Fig. 8).

Fig. 8. Variation of the reaction $^{51}$V($n,p$)$^{51}$Ti cross section due to uncertainties of the low-lying levels of the nucleus $^{51}$Ti.

The check of the improved PE procedure has been carried out by means of the analysis of the neutron-, proton-, and $\alpha$-particle emission spectra produced by 14-15 MeV neutrons incident on $^{51}$V. The agreement of the calculated and the available experimental charge-particle spectra (Fig. 9) could be considered good, in the limit of the experimental errors.

Fig. 9. Comparison of experimental and calculated charge-particle emission cross sections for n+$^{51}$V.
Moreover, an increased importance has had the availability of a large data basis of neutron-emission spectra measured at PTB-Braunschweig for incident-neutron energies $E$ from 8 to 13.9 MeV. Their analysis has been carried on at the same time with that of the total neutron cross section for the target nucleus $^{51}$V (Fig. 10). The agreement between experimental and calculated pre-equilibrium and statistical emission is good. The high-energy end of the spectra could be also well described provided that the DWBA direct-interaction cross sections, already taken into account for reaction cross section decreasing, is added.

Another new point of this work is the complete calculation of the $^{51}$V($n,d$)$^{50}$Ti reaction excitation function by using an extrapolation of the Milano-group method for $\alpha$-particle pre-equilibrium emission (e.g.\cite{1}). The corresponding s.p.s. density at the Fermi level is assumed twice that of the $\alpha$-particles while a deuteron pre-formation probability also twice that for the $\alpha$-particles is found to describe the experimental deuteron-emission spectrum at 14.8 MeV.

Finally, the effect of uncertainties coming from both model assumptions and the independent data involved in model-parameter establishment can be decreased only by the unitary description of all available experimental activation cross sections for fast neutrons incident on $^{51}$V. It is shown in Fig. 11 while the corresponding input parameters and calculated cross sections are given within an EBDF-6 format file\cite{25}.

\begin{thebibliography}{99}
\bibitem{25} V. Avrigeanu, ENDF-6 format file for the material V51, IFIN-HH, Bucharest, Nov. 2001.
\end{thebibliography}
Fig. 11. Comparison of the experimental and calculated activation cross sections for fast neutrons on $^{51}$V

B. Fast Neutron Activation Analysis for $^{92,94-98,100}$Mo Isotopes.

The first step of this work has been the study of activation cross sections for reactions induced on $^{92}$Mo, i.e. $^{92}$Mo($n,p$)$^{92}$Nb$^m$, $^{92}$Mo($n,\alpha$)$^{98}$Zr$^{28,\infty m}$, $^{92}$Mo($n,2n$)$^{91}$Mo$^2m$, and $^{92}$Mo($n,n'p$)$^{91}$Nb$^m$, for which there is also a large amount of measured data. However, there are yet many discrepancies between even recent data sets, while three basic evaluations performed in the last decade at well-known laboratories show wide differences, e.g. up to $\sim$50% for the ($n,p$) reaction$^{12,13}$ and $\sim$65% for the ($n,\alpha$) reaction$^{13,14}$. 

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The comparison of the calculated and available experimental excitation functions of (n,p), (n,a), (n,2n), and (n,n'p+d) reactions (Figs. 12-15) proves a good agreement in the limit of experimental errors, supporting thus the PE approach involved firstly in the case of $^{51}$V.

On the other hand, new measurements of activation cross sections of Mo isotopes were performed at IRMM-Geel for neutron energies from 16 to 20.5 MeV at the same time with these reported calculations. Their comparison could be considered a blind exercise and thus proves the accuracy of calculations. Their forthcoming revision by taking into account also the

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new IRMM data will provide both (i) the usefulness of the activation cross sections at neutron energies just above the common value of 15 MeV, and (ii) the accuracy limits of the present calculations.

Fig. 13. Comparison of experimental and calculated neutron-activation cross sections for $^{94,95}$Mo.
Fig. 14. Comparison of experimental and calculated neutron-activation cross sections for $^{96,97}$Mo.

*Graph 1: $^{96}$Mo(n,p)$^{96}$Nb*

- Artemev (1980)
- Amemiya (1982)
- Atsumi (1984)
- Pepelnik (1985)
- Rahman (1985)
- Marcinkowski (1986)
- Ishida (1988)
- Li (1989)
- Gizardov (1997)
- Osman (1996)
- Molla (1997)
- Doczi (1998)
- STAPRE-H (2001)
- Smoothed calc.

*Graph 2: $^{97}$Mo(n,p)$^{97}$Nb*

- Fujino (1977)
- Chaturvedi (1980)
- Artemev (1980)
- Amemiya (1982)
- Pepelnik (1985)
- Rahman (1985)
- Molla (1986)
- Marcinkowski (1986)
- Iskender (1988)
- Osmani (1996)
- Molla (1997)
- Doczi (1998)
- STAPRE-H (2001)
- Smoothed calc.

*Graph 3: $^{96}$Mo(n,np+d)$^{95}$Nb*

- Atsumi (1984)
- Ikeda (1988)
- Haight (1981)

*Graph 4: $^{97}$Mo(n,np+d)$^{96}$Nb*

- Lu (1970)
- Rao (1979)
- Amemiya (1982)
- Marcinkowski (1986)
- Iskender (1988)
- Osaka (1988)
- Bostan (1997)
- STAPRE-H (2001)
- Smoothed calc.
Fig. 15. Comparison of experimental and calculated neutron-activation cross sections for $^{98,100}$Mo.
C. The Analysis of the $^{58}$Ni(n,$\alpha$)$^{55}$Fe reaction

The check and validation of the main statistical model parameters by detailed analysis of various data have provided the necessary accuracy for the final calculation of all activation cross sections\textsuperscript{12} for $^{58}$Ni and especially for $^{58}$Ni(n,$\alpha$)$^{54}$Mn reaction. These results (Fig. 16) support the latest experimental data of IRMM\textsuperscript{16} which are around twice lower than those of Iwasaki et al.\textsuperscript{37}

![Graph](image-url)

Fig. 16. Comparison of the $^{58}$Ni(n,$\alpha$)$^{55}$Fe and $^{58}$Ni(n,$\alpha$)$^{54}$Mn reaction cross sections above thresholds.

Moreover, it should be pointed out that a good agreement of the calculated and experimental data has been obtained\textsuperscript{12} also for the reactions $^{58}$Ni(n,$p$)$^{58}$Co\textsuperscript{m,g}, $^{58}$Ni(n,$\alpha$)$^{55}$Fe, $^{58}$Ni(n,2n)$^{57}$Ni, and $^{58}$Ni(n,$n^\prime$)$^{57}$Co, as well as $^{59}$Co(n,2n)$^{58}$Co\textsuperscript{m,g}, including the study of the isomeric cross section ratios. However, additional similar analysis of isomeric ratios for, e.g., A~90 would be useful for definite conclusions on the spin-dependence of the nuclear level density.

On the other hand, comparison of the $^{58}$Ni(n,$\alpha$)$^{55}$Fe and $^{58}$Ni(n,$\alpha$)$^{54}$Mn reaction cross sections just above the threshold (Fig. 6), the former being calculated by using various OMP in use for $\alpha$-particles, shows that the suitable $\alpha$-particles OMP is yet an open problem of the nuclear model calculations. It plays a similar role to the unexpected differences between various data sets.

IV. MICROSCOPIC REAL POTENTIALS FOR $\alpha$-PARTICLES

The double-folded (DF) microscopic real potentials\textsuperscript{38,39} for $\alpha$-particles should be validated through description of experimental ($\alpha$,\,$\alpha$) angular distributions, while a phenomenological imaginary part has to be considered as well as the corresponding dispersion relations to the real one. Use of the dispersion correction needs the energy dependence of the phenomenological imaginary OMP, which at least for A~100 nuclei and for the low incident

energies is missing\textsuperscript{40}. Therefore it has been necessary to establish a global parameterization of Wood-Saxon potential taking into account the strong energy dependence and nuclear structure effects of the imaginary part. The new parameterization (Fig. 17) describes satisfactorily the elastic scattering of $\alpha$-particles on A$\sim$100 target nuclei at low incident energies as 20-32 MeV. Actually there have been obtained three average parameter sets for various energy ranges, which are compared with the only one previous extrapolation to low energies\textsuperscript{27} and some particular data\textsuperscript{41}. Additional analysis are moreover necessary below 20 MeV in order to provide OMPs for nuclear data evaluation\textsuperscript{8}.

Concerning the basic input for DF calculations which are the colliding nuclei nuclear densities and the effective nucleon-nucleon (NN) interaction, the experimental $^4\text{He}(\alpha,\alpha)^4\text{He}$ angular distributions have been found quite useful for testing the real effective NN interactions (e.g., M3Y-Reid and M3Y-Paris). Due to the corresponding relative high reaction threshold no imaginary optical potential addition is necessary for their description at incident energies $E_{c.m.}<17.5$ MeV, so that unambiguous information concerning the effective NN interactions has been obtained\textsuperscript{42}. Various approaches for $\alpha$-particle density distribution have been also analyzed.

Moreover, since the IFMIF project requests nuclear-data evaluation for D incident on $^6,^7\text{Li}$ for D-energies up to 50 MeV, we have analyzed and proved\textsuperscript{38,43} the possibility to use the DF method for calculation of the nuclear potential for complex particles (e.g. $^2,^3\text{H},^3,^4\text{He}$) emitted in neutron induced reactions on medium nuclei. It could be equally used for evaluation of nuclear data for D on $^6,^7\text{Li}$, a corresponding proposal being already submitted.
V. CONCLUSIONS

In completion of the progress presented in this work it is proposed the analysis of isomeric cross section ratios of Mo isotopes, for improved knowledge of nuclear-level density spin distribution, and calculation of activation data requested for EAF-2003.

On the other hand, analysis of $\alpha$-particle elastic scattering at incident energies below 20 MeV on A~100 nuclei is planned in order to provide a basis for suitable OMP requested in nuclear data evaluation.