REPORT ON EAF RELATED TOOLS*

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Progress of work on improved methods of nuclear model calculations for nuclear activation data carried out at IFIN-HH since the start of the activity of the OECD-NEA WPEC subgroup 19 is reported. In order to provide accurate predictions of further interest for EAF-2003, no use of normalization or free parameters are involved. Model calculations have been validated by analysis of activation cross sections of the all V, Co, Ni and Mo stable isotopes.

1. 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 an updated version of the computer code STAPRE-H95¹. Basic points of our work have been:

(i) **consideration of partial-wave PE effects** able to provide suitable description of \((n,p)\) and \((n,\alpha)\) reaction excitation functions above 15 MeV, at variance with usual PE models²;

(ii) **angular-momentum distribution of the nuclear-level density** given, e.g. within the back-shifted Fermi gas (BSFG) model³, by the nuclear moment of inertia \(I\) (found recently⁴ to be only half of the rigid-body value \(I_r\), while the value \(I=I_\text{r}\) is still widely used, e.g.⁵);

(iii) **optical model potential (OMP) providing the \(\alpha\)-particle transmission coefficients**, which is still an open question especially if a microscopic approach is not involved⁶.

The progress on the above item (i) was favored by development in the meantime at IFIN-HH of a novel partial level-density (PLD) formalism⁷, e.g. the recent IAEA Reference Input Parameter Library (RIPL)⁸, and improved version of corresponding computer code PLD⁹. The

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⁴S.F. Mughabghab and C. Dunford, Phys. Rev. Lett. 82, 4083 (1998)


⁶* * * The Model Code Meeting, May 21-22, 2002, EC/JRC/IRMM, Geel, Belgium.


first results were obtained in the case of the $^{51}$V activation cross section analysis\textsuperscript{10,11}. Work concerning the item (ii) has been done in connection with the main tasks of the former Subgroup 1 of the WPEC of OECD/NEANSC, concerning the $(n, \alpha)$ reaction and the nuclear level density which were underlined as generic problems. Since possible reasons for the well-known differences among the evaluated $(n, \alpha)$ cross sections have been considered\textsuperscript{12}

1. competition of other channels,
2. alpha-particle optical model potential,
3. level density, and
4. pre-formation factors in the pre-equilibrium emission (PE) model,

the actual model calculations have been validated by analysis of activation cross sections of the isotopes $^{51}$V, $^{59}$Co, $^{58,60,61,62,64}$Ni and $^{92,94,95,96,97,98,100}$Mo by using

(a) unitary use of the common model parameters for different concerned mechanisms,
(b) consistent sets of input parameters determined by various independent data analysis,
(c) unitary account of a whole body of related experimental data for the above-mentioned isotope chains or neighboring elements.

Compensation of opposite effects due to various less accurate parameter values has been thus avoided. This is particularly important since the first measurement at LASL–Los Alamos and comprehensive study\textsuperscript{13,14} of the alpha-production by neutrons on $^{58,60}$Ni as well as $^{59}$Co from threshold to 50 MeV have not supported any of the previous evaluations while latest analyses still consider the large uncertainties in the $(n, \alpha)$ cross sections as a general problem, in conjunction also with LD approaches\textsuperscript{15,16}. Thus LD was the first subject of our major interest\textsuperscript{17,18}, while for the above item (iii) we have firstly used or adjusted the phenomenological OMP proved able to describe alpha emission\textsuperscript{19}, and also considered\textsuperscript{20} the semi-microscopic calculation using the double-folding method for the real potential (e.g. Ref.\textsuperscript{21}) as the only proper treatment\textsuperscript{6} of this matter.

Actually recent measurements\textsuperscript{22,23} at IRMM-Geel and FZ-Jülich of cross-sections for other channels in competition with $(n, \alpha)$ reactions, especially at incident energies between 14 and 21 MeV, have made possible enlarged analysis\textsuperscript{23,24,25} of the calculated fast-neutron activation of $^{59}$Co and $^{58,60,61,62,64}$Ni isotopes. The same approaches have been involved in the case of $^{92,94,95,96,97,98,100}$Mo activation, and comparison of calculated and available experimental

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\textsuperscript{20}M. Avrigeanu, this workshop.
\textsuperscript{22}A. Fessler and S.M. Qaim, Radiochim. Acta 84, 1 (1999).
excitation functions of \((n,p)\), \((n,\alpha)\), \((n,2n)\), and \((n,n'p+d)\) reactions proves a good agreement\textsuperscript{11} in the limit of experimental errors. On the other hand, new measurements of activation cross sections of also Mo isotopes were performed at the same time\textsuperscript{26} at IRMM-Geel for neutron energies from 16 to 20.5 MeV. The comparison with them of the calculated values could be considered a \textit{blind exercise}, while in this report is given a revision of the Mo calculated data by taking into account also the latest IRMM data. However, firstly are briefly mentioned various independent data analyses used for fixing a consistent sets of input parameters, other than the LD and \(\gamma\)-ray strength functions \(f_{E1}(E_\gamma)\) described previously\textsuperscript{18} also for the A\textadd{=}90-100.

2. Optical Model Potentials

The calculation of the neutron transmission coefficients was carried out through the analysis of the OMP for the interaction of neutrons with the target nuclei\textsuperscript{\textcopyright,\textdegree,100} Mo, for energies up to 20 MeV, by using the SPRT method\textsuperscript{27}. This detailed analysis is necessary since the neutron OMP used in Hauser-Feshbach calculations should provide realistic total reaction cross sections over a wide incident energy range, as well as a reasonable description of the emission of low-energy neutrons in statistical processes. On the other hand, additional questions have concerned the molybdenum isotopes which show a rapidly changing character from that of the essentially spherical \textsuperscript{\textdegree}Mo (with a filled \(\text{g}_{9/2}\) neutron shell) to nuclei making a transition from collective vibrators to the deformed rotors. Consequently, as the isotopic mass and collectivity of the Mo isotopes increase, discrepancies appear for description of the nucleon scattering by means of the spherical optical-statistical and even direct-reaction models.

For the OMP analysis of the neutron total cross sections available at BNL we have firstly derived broad energy-averages over 200 keV of the data for the representative isotopes \textsuperscript{\textdegree,\textdegree\textdegree}Mo and \textsuperscript{98}Mo. They were compared with several optical model predictions\textsuperscript{28,\textdegree,30,31}, of which the OMP obtained earlier\textsuperscript{28,\textdegree,29} at ANL was based on the data measured up to 5.5 MeV and involved with good results\textsuperscript{32} in the analysis of data of neutron inelastic scattering from molybdenum isotopes obtained recently\textsuperscript{33} at IRMM. Our comparison of the corresponding calculated and available experimental\textsuperscript{34} neutron \(s\) - and \(p\)-wave strength functions and the potential scattering radius \(R'\) led to the conclusion that the spherical OMP of Lagrange\textsuperscript{31} provides similar results to the more complex vibrational and dispersive models, recently developed by Smith\textsuperscript{30}. On the other hand, this potential was involved previously\textsuperscript{35} (Figs. 3-4) in IFIN-HH analysis of differential elastic scattering cross sections for \textsuperscript{\textdegree,100}Mo at neutron energies of 7, 9 and 11 MeV, and inelastic scattering on low-lying excited levels of \textsuperscript{\textdegree,\textdegree,\textdegree,98,100}Mo. However, since the total neutron cross sections are overestimated at energies below 1 MeV (Fig. 1), the OMP of Smith\textsuperscript{30} for natural molybdenum was used within this range.

\begin{footnotesize}
\begin{enumerate}
\item \textsuperscript{27}J.P. Delaroche, Ch. Lagrange, and J. Salvy, IAEA-190 (IAEA, Vienna, 1976), vol. 1, p. 251.
\item \textsuperscript{28}A.B. Smith, P.T. Guenther, and J. Whalen, Nucl. Phys. A\textbf{244}, 213 (1975).
\item \textsuperscript{30}A.B. Smith, J. Phys. G \textbf{26}, 1467 (2000).
\item \textsuperscript{32}T. Kawano, Y. Watanabe, and M. Kawai., J. Nucl. Sci. Tech. (Japan), \textbf{35}, 519 (1998).
\item \textsuperscript{34}S.F. Mughabghab, M. Divadeenam, N.E. Holden, \textit{Neutron Cross Sections} (AP, New York, 1981), v. 1.
\end{enumerate}
\end{footnotesize}
Fig. 1. Comparison of calculated and experimental neutron total cross sections for $^{92,94,96,98,100}$Mo.
The calculation of the proton transmission coefficients was based on the analysis of the interaction of protons with the target nucleus $^{93}$Nb, taking firstly into account the proton reaction data for energies up to 5.5 MeV. In addition, there have been calculated and compared with the experimental data the cross sections for the reaction $^{93}$Nb$(p,n)^{93}$Mo in the incident energy range of 2-6 MeV. Thus, we have used the OMP of Johnson et al.\textsuperscript{36} with the energy dependence suggested by Lagrange\textsuperscript{37} and the values of the depth $W_D$ for the imaginary part of the OMP given by the smooth curve of the anomalous dependence $W_D(A)$ found by Flynn et al.\textsuperscript{38}. On the other hand, we have taken the advantage of new $^{93}$Nb$(p,\gamma)^{94}$Mo reaction cross-section measurements below 5 MeV reported in the meantime\textsuperscript{39} and used them for additional validation of the proton OMP involved in our calculation. Our present results (Fig. 2) solved the discrepancies reported\textsuperscript{39} for the model description of the $(p,\gamma)$ data above 3.5 MeV, at the same time with a good agreement obtained with the $(p,n)$ reaction data.

The OMP for calculation of $\alpha$-particle transmission coefficients was established previously\textsuperscript{19} by analysis of experimental $(n,\alpha)$ reaction cross sections just above the effective Coulomb barrier.

### 3. The $\gamma$-ray Strength Functions

The $\gamma$-ray strength functions $f_{\text{E1}}(E_\gamma)$ 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{40,41}. Systematic EDBW correction factors $F_{\text{SR}}$ were established\textsuperscript{18} for A=41-105 by analyzing the experimental average radiative widths $\Gamma_{\gamma0}^{\text{exp}}$ of the s-wave neutron resonances, and assuming that $F_{\text{SR}}=\Gamma_{\gamma0}^{\text{E1}}/\Gamma_{\gamma0}^{\text{EDBW}}$. The $f_{\text{E1}}(E_\gamma)$ thus obtained were finally checked by calculation of capture cross sections of $^{93}$Nb and $^{92,94,98,100}$Mo nuclei (Fig. 3).

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Fig. 3. Comparison of calculated and experimental neutron capture cross sections of $^{93}\text{Nb}$ and $^{92,94-98,100}\text{Mo}$ isotopes.
4. The Distorted Wave Born Approximation (DWBA) method

The collective enhancement of the direct scattering cross section due to the low-energy surface vibrations of quadrupole and octupole multipolarity, included by means of the DWBA method and deformations parameters are generally considered. However, anomalous enhancement of DWBA cross sections near the threshold of inelastic scattering was reported around $A \sim 100$ when global OMP parameters were used. Moreover, since it was often claimed that DWBA is inappropriate for evaluation of fission-product data, recent measurements as well as DWBA and coupled-channels (CC) analyses were performed for $^{98,100}$Mo.

Thus Kawano et al. found that the problem comes from the optical potential used, i.e. when an adopted OMP to DWBA calculation is physically reasonable, differences between the calculated cross sections with the DWBA and those with the CC theory are small. Finally, it was concluded that the DWBA is an appropriate method to evaluate cross sections of inelastic scattering from the Mo isotopes, by using the ANL “regional” parameter set. This achievement should be even more correct in the case of Lagrange OMP based on SPRT analysis.

Following also Kawano et al., we have calculated the direct inelastic scattering only for the one-phonon states (2$^+$ and 3$^-$) of the Mo isotopes. while cross sections for two-phonon triplet (0$^+$, 2$^+$, and 4$^+$) are expected to be small. The deformation parameters $\beta_2$ and $\beta_3$ for the even-even target nuclei were taken from latest tabulations. For the odd-A isotopes we have adopted, as values for excitation energies and deformation parameters of collective states, the average between corresponding values for the neighboring even-even Mo isotopes. Finally, total direct inelastic scattering cross sections (Fig. 4) were obtained by using the DWUCK4 code.

![Fig. 4](image)

Fig. 4. Ratios of direct inelastic scattering cross section to neutron reaction cross section for $^{92,94-98,100}$Mo.

5. Fast neutron activation analysis of the $^{92,94-98,100}$Mo isotopes

Completion of the previous analysis has taken into account also the activation measurements performed at the same time at IRMM for incident energies from 16 to 20.5 MeV.

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Fig. 5. Comparison of experimental and calculated fast-neutron cross sections for $^{92}$Mo.
On the other hand, the main change with respect to previous analysis concerned the PLD formalism\cite{7,9} including surface effects, and a value of 40 MeV for the Fermi energy. A slight change concerned also the pre-formation probability considered for the deuteron PE emission, described by using the Milano-group method for the $\alpha$-particle PE emission (e.g. Refs. within\cite{1}).
Fig. 7. Comparison of experimental and calculated neutron-activation cross sections for $^{95,96}\text{Mo}$. The corresponding single-particle state density at the Fermi level has been assumed twice that of the $\alpha$-particles while a deuteron pre-formation probability of 0.5-0.6 has been found now to describe the experimental deuteron-emission spectra at 14.8 MeV. It can be compared with the value 0.18 used for the pre-formation probability considered in this work for the $\alpha$-particles PE emission.

Fig. 8. Comparison of experimental and calculated neutron-activation cross sections for $^{97,98}\text{Mo}$. 

$^{97}\text{Mo}(n,p)^{97}\text{Nb}$

$^{98}\text{Mo}(n,p)^{98}\text{Nb}$

$^{97}\text{Mo}(n,np+\alpha)^{96}\text{Nb}$

$^{98}\text{Mo}(n,np+\alpha)^{95}\text{Zr}$

$E_n$ (MeV) vs. $\sigma$ (b) for $^{97,98}\text{Mo}$.
Fig. 9. Comparison of experimental and calculated neutron-activation cross sections for $^{100}$Mo.
The first step of this work has been the study of activation cross sections for reactions induced on $^{92}\text{Mo}$, i.e. $^{92}\text{Mo}(n,p)^{92}\text{Nb}^m$, $^{92}\text{Mo}(n,\alpha)^{89}\text{Zr}^m$, $^{92}\text{Mo}(n,2n)^{91}\text{Mo}^m$, and $^{92}\text{Mo}(n,n'p)^{91}\text{Nb}^m$ (Fig. 5), for which there is also a large amount of measured data but 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$^{46,47}$ and $\sim$65% for the $(n,\alpha)$ reaction$^{47,48}$. The agreement between calculated and the available experimental data could be considered good in the limit of experimental errors.

However, the PE effects are also lowest in the case of the lightest stable isotope $^{92}\text{Mo}$. For the validation of this mechanism they are more important the isotopes $^{98,100}\text{Mo}$ (Figs. 8-9) and especially the $(n,2n)$ reaction on the heaviest isotope. The correct description of the recent experimental data around the maximum of this reaction excitation function (Fig. 9) can be considered positive in this respect.

Furthermore, the comparison of the calculated and available experimental excitation functions of $(n,p)$, $(n,\alpha)$, $(n,2n)$, and $(n,n'p+d)$ reactions on the target nuclei $^{94,96-98}\text{Mo}$ (Figs. 6-8) proves a good agreement in the limit of the experimental errors. It is supported thus the PE approach involved firstly in the case of $^{51}\text{V}$, namely the effect of the thresholds for various partial waves contributions to the PE processes$^{11}$ which are shown by the calculated cross sections (small crosses in Figs. 5-9) before applying a smoothing procedure (thick solid curves). They are compared with the results of the previous analysis$^{11}$, where it was shown the same structure, as well as with the predictions$^{16}$ of THALYS.

The quite schematic model adopted for the deuteron PE emission could be the reason of some discrepancies between the calculated and experimental $(n,n'p+d)$ reaction cross sections. The assumption of the $d$-particle state density closely related to the phenomenological $\alpha$–particle state density$^{49}$ $g_{\alpha}=A/10.36\text{ MeV}^{-1}$ could be questionable especially concerning the energy dependence. Unfortunately the present analysis shows similar numbers of favorable and doubtful cases in this respect, and further work would be useful.

Finally one may consider that actual revision of these calculations by taking into account also the new IRMM data proved both

(i) the usefulness of the activation cross sections at neutron energies just above the common value of 15 MeV, and

(ii) the prediction power as well as the accuracy limits of the present calculations, mainly related to the decay schemes in the case of the isomer ratio calculations.

The latter point concerns the low-lying discrete levels where missing experimental branching ratios or even any $\gamma$-decay data for some levels may hardly affect the isomeric cross-section calculated values. The $\gamma$-ray strength functions $f_{E1}(E_\gamma)$ description has received an additional validation in the case of the analysis of the $^{94}\text{Mo}(n,2n)^{93}\text{Mo}^m$ reaction, where the 16th excited state is the isomeric one and there is no feeding of it from the higher discrete levels. Unfortunately only in such limit cases one may avoid the use of the low-lying level decay schemes, while they remain critical in the general case. The magnitude of this effect could be the subject of further analysis.