2.1.2 Actinide Nuclear Data Evaluation for BROND and Beyond

V.M. Maslov

Radiation Physics and Chemistry Problems Institute
220109, Minsk-Sosny, Belarus, CIS

The neutron cross sections of minor actinides U, Pu, Am, Cm have been calculated in the energy range of 0.01 to 20 MeV. The optical cross sections were calculated with coupled channel model. Since in case of minors the fission data fit is virtually the only constraint for \((n,xn)\), \(x=1,2,3\) and \((n,\gamma)\) calculations, the theoretical tools employed were tested in case of consistent analysis of total, \((n,f)\), \((n,\gamma)\), \((n,n')\), \((n,2n)\) and \((n,3n)\) data for major actinides. The role of statistical model parameters testing is exemplified.

1. Introduction

Until recently the target of activity of Nuclear Data Evaluation Laboratory of the former Institute of Nuclear Engineering were mainly major actinides. The actinide data files prepared for BROND library are the following (the year of release in parentheses): \(^{239}\text{Pu}, \, ^{241}\text{Pu}, \, ^{242}\text{Pu} (1980), \, ^{240}\text{Pu} (1981), \, ^{235}\text{U} (1985), \, ^{236}\text{U} (1987), \, ^{238}\text{Pu}, \, ^{242}\text{Cm} (1988), \, ^{244}\text{Cm} (1989), \, ^{233}\text{U} (1991). Since then a number of methods, parameter systematics and calculation tools were developed\(^1\), which could be used for evaluation of minor actinide data. These data are urgent for actinide burner concept optimization, but unfortunately, with the rear exception \((^{242}\text{Cm}, \, ^{244}\text{Cm}\) for example\(^2,3\)) they were never produced as the files for major actinides. Virtually, only fission data, if any, are available for minor actinides, they would be used as a constraint for \((n,\gamma)\), \((n,n')\), \((n,2n)\) and \((n,3n)\) reaction cross sections.

In the present analysis the \(^{232-238}\text{U}, \, ^{236-244}\text{Pu}, \, ^{241-243}\text{Am}\) and \(^{242-248}\text{Cm}\) nuclides are covered. The total cross section, shape elastic and reaction cross sections were calculated with a coupled channel optical potential parameter systematics\(^4\). It was obtained when fitting total and differential scattering cross sections for major actinides. Although actual \(\beta_i\) and \(\beta_s\) deformation parameter values should be checked at least against \(S_0\) and \(S_1\) values.

The analysis was divided into two parts, i.e. 1) up to the emissive fission threshold and 2) above emissive fission threshold. In the first part total, reaction, elastic and inelastic scattering, direct inelastic scattering for 4 or 5 ground state band levels, capture and fission cross sections were calculated. Code STAT\(^5\) was used. In the second part fission, inelastic scattering, \((n,2n)\) and \((n,3n)\) cross sections were calculated with modified version of STAPRE\(^6\) code.

2. Cross sections below emissive fission threshold

The first "plateau" fission cross section analysis is accomplished within a Hauser-Feshbach formalism, the double-humped fission barrier model is used. The detailed description of the model is given elsewhere\(^7,8\). Here only the main points would be discussed.

The level densities of fissioning nuclides at inner and outer saddles as well as that of the residual nuclides were
calculated with a phenomenological model\(^9\):

\[ \rho(U, J, \pi) = K_{\text{rot}}(U, J) K_{\text{vib}}(U) \rho_{\text{qp}}(U, J, \pi) \]  

(1)

where \( K_{\text{rot}}(U, J) \) and \( K_{\text{vib}}(U) \) are factors of rotational and vibrational enhancement of level density and \( \rho_{\text{qp}}(U, J, \pi) \) is the quasi-particle level density, which was "renormalized" at low energies to fit the cumulative sums of low lying levels. The residual nuclides were assumed axially deformed, the main level density parameter \( a(B_n) \) was defined by fitting \( D_{\text{obs}} \), except those cases, like \(^{245}\)Pu and \(^{245}\)Cm compound nuclides, when resulting \( a(B_n)/A \) (A-mass number) looks like a spike as compared with neighbouring nuclide values. In such cases simple systematics for \( a/B \approx 0.484 - 0.00162/A \) was used. The liquid drop shell corrections were calculated with the liquid-drop mass parameters\(^10\). The asymmetries of fissioning nuclides at saddles were defined according to SCM calculations of fission barriers\(^11\).

Then the available fission data for U and Pu targets were analysed. Some peculiar features arise here for neutron-poor nuclides data \(^{232}\)U\((n,f)\) and \(^{236}\)Pu\((n,f)\). They exhibit a non-threshold cross section behaviour at low energies, which corresponds to rather low inner barrier heights (lower than the outer one), meanwhile, they gain axial symmetry at inner saddles\(^11\). As shown on the figs.1,2 the model employed is capable to reproduce also rather steep slope of \(^{232}\)U\((n,f)\) data\(^12\) above 2 MeV. The scatter of data on \(^{236}\)Pu\((n,f)\) prohibits more reasonable fitting of data, since at lower energies fission cross section values are as high as that of reaction cross section. In case of other U, Pu and Am nuclides the data are fitted rather well from 0.01 MeV up to 5 MeV.

In case of Cm targets the extreme paucity of fission data hinders an extensive analysis. In case of N-even Cm targets the bomb-shot data\(^16\) are consistent with data, measured with electron linac as a neutron source and lead slowing down time spectrometer\(^17\). The fig.3 shows the typical comparison of calculated and measured data for \(^{246}\)Cm\((n,f)\) data. Also with our approach we can't reproduce over-barrier structures in Cm bomb-shot data\(^16\).

Actually there is no adjustable parameter in neutron channel in case of most U and Pu targets. In case of minor actinides extreme care should be taken, so that the matching energy of continuum level density description would be lower than the energy, where the missing of low-lying levels occurs\(^18\). If the matching energy is too high, the significant part of inelastic cross section may be misinterpreted as elastic scattering cross section. This would be the case if CASTHY code approach is used. In convenient approach the fission channel parameters would be aberrated. Fig.4 shows what happens in case of \(^{243}\)Cm\((n,n')\) reaction if missing of levels in residual nuclide \(^{243}\)Cm above 0.4 MeV is ignored and matching energy of 1 MeV is adopted. The same happens in other cases, for example, \(^{241}\)Pu\((n,n')\) etc. The proposed approach fits the data on \((n,n')\) reactions for \(^{233}\)U, \(^{235}\)U and \(^{238}\)U, which justifies it's application for the minors.

Capture data for \(^{238}\)U and \(^{235}\)U provide an opportunity to develop the approach to predict \((n,y)\) reaction cross sections in case of even targets. The Poenitz data\(^19\) (see fig.5) show a fair agreement with JENDL-3 evaluation and our calculated curve up to
1 MeV. The radiation strength function $S_{T0}$ used is $9.5 \times 10^{-4}$. At higher energies JENDL-3 curve seems to underestimate the data, since it is a fit of some older Poenitz' data. Above 1 MeV incident neutron energy when calculating capture width the competition of $(n,\gamma f)$ and, more important, $(n,\gamma n')$ reactions should be included. In an opposite case calculated curve drastically overestimates the capture cross section. To resolve the remaining discrepancy (see fig.5) one should model the residual even-even nuclide $^{238}\text{U}$ level density at 1.3-2.8 MeV excitation energy, i.e. above pairing gap. Within pairing gap collective levels are modelled with a constant temperature model\textsuperscript{18}. Above pairing gap the two-quasi-particle state density could be modelled with a simple formulas\textsuperscript{20}. As a result the $(n,n')$ and, which is more important, $(n,\gamma n')$ competition will increase and we will get a fair agreement with data\textsuperscript{19}. The same effects are observed in case of $^{236}\text{U}(n,\gamma)$, but there are systematic discrepancies in measured $(n,\gamma)$ data around 1 MeV. The $(n,\gamma)$ cross section for N-odd fissile targets could be fitted with inclusion of $(n,\gamma f)$ reaction competition. The measured data are actually $\alpha=\sigma_{f}/\sigma_{f}$ data and above 0.1 MeV they are rather old\textsuperscript{21}. The $^{235}\text{U}(n,\gamma)$ cross section could be well reproduced, but in case of $^{235}\text{U}(n,\gamma)$ to reproduce the 'dip' in data\textsuperscript{20} above 0.6 MeV one needs a $S_{T0}$ value, which is ~50% lower than that fitting data\textsuperscript{21} at lower energies (see fig.6).

3. Cross sections above emissive fission threshold

For fixed statistical model parameters of residual nuclides, fissioning in $(n,nf)$, $(n,2nf)$ etc. reactions, the realistic trend of non-emissive fission cross section $\sigma_{f}$ is critical for reproducing the measured fission cross sections up to 20 MeV. The consistent description of the most complete set of measured data on $(n,f)$, $(n,2n)$ and $(n,3n)$ reaction cross sections for the $^{235}\text{U}$ and $^{238}\text{U}$ target nuclei and secondary neutron spectra for the latter target gives a strong ground to consider the estimate of $\sigma_{f}$ and contribution of the first neutron pre-equilibrium emission as fairly realistic. The model is described in detail elsewhere\textsuperscript{8,22}. At least, the new $^{232}\text{Th}(n,2n)$ data were reproduced fairly well without additional parameter variation\textsuperscript{23}, the fission cross section being the only constraint.

In case of N-odd, Z-even targets the method\textsuperscript{20} of resolving the well-known discrepancy between measured data on $^{239}\text{Pu}(n,2n)$ and convenient statistical model calculations could be applied. Near-threshold behaviour of the $(n,2n)$ cross section is interpreted as being due to jump-like excitation of two-quasi-particle states in residual even-even nuclide. The same effect is observed\textsuperscript{24} in case of $^{235}\text{U}(n,2n)$ data (see fig.7). The calculated $^{235}\text{U}(n,2n)$ cross section, governing the $^{232}\text{U}$ build-up in U-Th fuel cycle, is rather different from JENDL-3 (see fig. 8).

Almost no new data have appeared since previous analyses of U, Pu, Am and Cm data. The method of fission calculation\textsuperscript{22} proved reasonable in case of $^{232,238}\text{U}(n,f)$ data and $^{238,244}\text{Pu}$ data. The validity of the model for Z-odd target was demonstrated\textsuperscript{25} in case of consistent $^{239}\text{Np}(n,f)$ and $^{237}\text{Np}(n,2n)$\textsuperscript{24} data analysis\textsuperscript{25} and $^{241,243}\text{Am}(n,f)$ data analysis\textsuperscript{26}. For most of the nuclides there is a consistency between calculated and measured fission data. However the numerous discrepancies in calculated and JENDL-3 evaluated $(n,xn)$ cross sections being notified. The severe
discrepancy persists in case of $^{242}\text{Am}(n,f)$ above the onset of $(n,nf)$ reaction. The same kind of discrepancy still persists in case of $^{245}\text{Cm}(n,f)$. The analysis of the other existing evaluated Cm data shows the inadequacy between JENDL-3, ENDF/B-VI and statistical calculations with reasonable fission and neutron channel parameters.

4. Conclusion
I guess, that existing data base and knowledge data base are sophisticated enough to produce more justified neutron data for minor actinides, like Pa, U, Np, Pu, Am and Cm.

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References
Fig. 1

Fig. 2

Fig. 3

Fig. 4
Figure 5: 238U Capture Cross Section
- Poenitz, 1985
- JENDL-3
- Calculation
- Without (n,n') comp.
- Convenient (n,n') comp.

Figure 6: 233U Capture Cross Section
- Hopkins et al., 1962
- Calculation
- \( S_{10} = 833 \)
- \( S_{10} = 564 \)

Figure 7: 236U(n,2n) Cross Section
- Frehaut et al., 1980
- JENDL-3
- Calculation

Figure 8: 233U(n,2n) Cross Section
- JENDL-3
- Calculation