

INTERNATIONAL NUCLEAR DATA COMMITTEE

TRANSLATION OF SELECTED PAPERS PUBLISHED IN NUCLEAR CONSTANTS 4, 1986

(Original Report in Russian was distributed as INDC(CCP)-268/G)

Translated by the IAEA

August 1989

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

INDC(CCP)-302/L

TRANSLATION OF SELECTED PAPERS PUBLISHED IN NUCLEAR CONSTANTS 4, 1986

(Original Report in Russian was distributed as INDC(CCP)-268/G)

Translated by the IAEA

August 1989

Reproduced by the IAEA in Austria August 1989

.

89-03525

<u>Contents</u>

Neutron Data Evaluation Activities of the Nuclear Data Centre V.N. Manokhin, A.I. Blokhin	5
Evaluation of Nuclear Data for Heavy Fissile Nuclei V.A. Kon'shin	.4
Absolute Measurements of the Fission Cross-Sections for Important Nuclides V.I. Shpakov	3
Measurement and Analysis of Radiative Capture Cross- Sections of Neptunium and Uranium Isotopes V.A. Tolstikov	1
Consistent Evaluation of Neutron Cross-Sections for the ^{242–244} Cm Isotopes A.V. Ignatyuk and V.M. Maslov5	53
Re-Evaluation of the Neutron Cross-Sections for Iron V.G. Pronyaev and A.V. Ignatyuk	;3
Evaluation of Cross-Sections for the Fission of Curium Isotopes for fast Neutrons B.I. Fursov, G.A. Kudyaev and G.N. Smirenkin	² 5

NEUTRON DATA EVALUATION ACTIVITIES OF THE NUCLEAR DATA CENTRE

V.N. Manokhin, A.I. Blokhin

The activities of the Obninsk Nuclear Data Centre of the State Committee on the Utilization of Atomic Energy concerning neutron data evaluation and the creation of a library of these data have covered the following areas:

- Establishment and development of a system of ENDF-format library service programs;
- Development of nuclear reaction theory and evaluation methods using theoretical models;
- Neutron data evaluation for structural materials and fission product nuclei;
- Organization of the analysis and expert examination of evaluated
 data in order to produce recommendations on their improved usage;
- Creation, checking and correction of evaluated data files.

The ultimate aim of this activity is the creation of a library of recommended evaluated neutron data files which might serve as the basis for an improved provision of constants for reactor calculations and other scientific and technical applications. The majority of the work was conducted in close co-operation with other laboratories of the Obninsk Power Physics Institute (PPI), and with institutes of the USSR and member countries of the CMEA.

Establishment of evaluated data library service programs

At present the Nuclear Data Centre possesses a large number of evaluated neutron data files in the ENDF/B, KEDAK, UKNDL and other formats. There follows a list of the foreign national evaluated nuclear data libraries received by the Nuclear Data Centre within the framework of the international data exchange:

	ENDF	B-IV		American National Library, Version 4
<u>Files</u> :				
		TAPE	401-411	General purpose
		TAPE	412	Data used in neutron dosimetry
		TAPE	413	Data used as standards in nuclear measurements
		TAPE	414-419	Fission product data
		TAPE	420-421	Gamma ray interaction data
	JENDI	L-1		Japanese National Library, Version 1, general
				purpose files
	JENDI	L-2		Ditto, Version 2
	JENDI	L-FP		Ditto, for fission products
	ENDL-	- 78		Livermore National Library, 1978 Version
	ENDL-	-82		Ditto, 1982 Version
	ENDL-	-84		Ditto, 1984 Version
	KEDAI	(-3		West German Library
	UKNDI			United Kingdom National Library
	ENDF	B-V		American National Library, Version 5

<u>Files</u>:

TAPE	511	Standards
TAPE	509,510,	
	541, 546	
	566	Fission product data
TAPE	531	Neutron dosimetry data
TAPE	532, 564	Activation cross-section data
TAPE	533,	Light element accumulation cross-section data
TAPE	514, 521	
	522, 565	Actinide data
INDL/V		International evaluated neutron data library
		compiled by the IAEA Nuclear Data Section for
		various elements and reactions

INDL/A	Ditto,	for	actinides	5				
IRDF/82	Ditto,	for	elements	and	reactions	used	in	reactor
	dosimet	try						

The ENDF/B, UKNDL and KEDAK formats are those most widely used for data presentation. To organize its work both in the preparation and the processing of evaluated neutron data, the Nuclear Data Centre installed those service programs most widely used for operating with ENDF/B-format data, and also created programs with additional functions [1].

There follows a list of the service programs and software systems installed on the EC-1033 computer in the Nuclear Data Centre, with a brief indication of the functions they perform:

PRINKED	Accessing of data by element, reaction etc. from
	KEDAK-format libraries
MERGER, PRINF,	
PRINTE	Ditto, for ENDF/B format
PRINUKN	Ditto, for UKNDL format
PRINKED	Compilation of information on the general content
	of KEDAK-format data files
SUMRIZ, LSTFCY,	
PRINF	Ditto, for ENDF/B format
PRINUKN	Ditto, for UKNDL format
RESEND, LINEAR,	
RECENT, SIGMA-1,	
NJOY	Calculation of cross-sections in the neutron energy
	resonance region, taking into account Doppler-
	broadening of levels
INTEND, GROPIE,	
NJOY	Calculation of unshielded group-averaged
	cross-sections

NJOY)Y Calculation of shielded group-averaged					
	cross-sections, and inelastic transition matrices					
INTER	Calculation of thermal cross-sections, g-factors,					
	resonance integrals					
TBL-1 (This program	1					
processes data obta	ined					
using the GROPIE an	ıd					
NJOY programs).	Presentation of group-averaged cross-sections in					
	tabular form.					
CHECKER, 1981 and						
1983 versions	Checking of data for ENDF/B-V format structure					

FIZKON, 1981 and

1983 versions

PSYCHE, 1981 and

```
1983 versions Checking of data for physical correlation of neutron data
```

CRECT, DICTION,

CRMAT Correction of formatted data

conformity

Using these programs the Nuclear Data Centre has developed technical means of processing requests for evaluated nuclear data which permit the accessing, processing and correction of data in the UKNDL, KEDAK and ENDF/B formats. To achieve this, we created a library of operation algorithms, consisting of catalogued source module sets written in FORTRAN or RL-I, and instructions for the preparation of input packages for each program. On request, the library can be made available to users together with various evaluated data sets.

The algorithm library not only permits the processing of evaluated neutron data in different ways, but will also promote wider use of such data in various applications.

Development of evaluation methods and evaluation

On the basis of an analysis of existing evaluations and experimental data, the Nuclear Data Centre re-evaluated the complete files of the structural materials chromium, nickel and iron. Apart from the files containing evaluations of the natural isotopic mixture, files of crosssections for separate isotopes have also been set up in the new version. In this version considerable attention has been given to cross-section resonance structure, and the evaluation of inelastic neutron scattering cross-sections for low-lying levels has been considerably improved. To this end, an opticalstatistical description was used whose parameters were determined from a correlated analysis of total neutron cross-sections, inelastic scattering cross-sections and the excitation cross-sections of the lowest collective levels of even-even nuclei. This approach allowed the contribution of direct scattering over the whole energy range to be taken into account, and also permitted correct evaluation of the excitation functions of the lowest levels of odd isotopes, for which direct experimental data do not exist.

An analysis was also conducted of neutron inelastic scattering and capture cross-sections for isotopes of zirconium, niobium and molybdenum. The results of the analysis are of interest for the parameter systematics of unified optical models and for a consistent description of the whole range of neutron cross-sections.

The Nuclear Data Centre and the Technical University of Dresden co-operated on the re-evaluation of total neutron cross-sections and capture cross-sections in the resolved resonance region for the natural silicon and lead files developed previously by the Technical University. New versions of the silicon and lead files have been created which take into account the results of this re-evaluation. When establishing the silicon file, the evaluation of the (n,p) reaction cross-section was taken into account.

In recent years the PPI has conducted an analysis of radiative capture cross-sections for the major fission products [2]. This demonstrated that for

the isotopes ^{95,97,98,100}Mo, ¹³³Cs and ¹⁴¹Pr no re-evaluation is necessary, since the recommendations of the JENDL-1 or ENDF/B-V libraries for them are fully satisfactory. For the isotopes ⁹⁹Tc, ^{101,102}Ru, ¹⁰³Rh, ^{105,107}Pd, ¹⁰⁹Ag, ¹²⁹I, ^{143,145}Nd, ^{147,149}Sm new evaluations of the mean neutron capture cross-sections for energies above 1 keV were performed. The neutron and radiation strength functions used for cross-section parametrization in the unresolved resonance region were determined on the basis of a statistical description of capture cross-sections and an analysis of mean resonance parameters. These data were amplified using resonance parametrization of cross-sections for resolved resonances.

For isotopes where no direct experimental data exist on fast neutron capture cross-sections and where only mean resonance parameter data are available, and also for isotopes where no data exist on cross-sections or on resonance parameters, the evaluations of fission product capture cross-sections were based on empirical systematics of radiation strength functions. On the basis of the above evaluations the Nuclear Data Centre has created complete ENDF-format fission product files.

For the isotopes 235 U and 238 U a correlated optical-statistical analysis was conducted of total neutron cross-sections, the fission crosssection, neutron spectra and excitation functions for (n,xm) reactions [3]. The level density description in the analysis takes into account shell, superfluid and collective effects, and also differences in the occurrence of such effects for equilibrium and transition deformations of fissile nuclei. The resultant parameters were used subsequently by the Nuclear Power Institute (NPI) of the Byelorussian SSR Academy of Sciences, Minsk, to correct neutron spectra and the (n,xm) reaction cross-sections for plutonium isotopes, and also to obtain a consistent evaluation of (n,2n) reaction cross-sections for 237 Np [4].

Creation of a recommended evaluated neutron data library

Using as a basis evaluations performed principally in the USSR (at the PPI and NPI), the Nuclear Data Centre has set up a recommended evaluated data library which includes files evaluated by the Nuclear Data Centre (chromium, iron, nickel, major fission products), by the NPI (235 U [5], plutonium isotopes [6]) and by the group constants laboratory of the PPI (deuterium [7], sodium, oxygen [8], 238 U [9]). The library includes the files for 6,7 Li evaluated in the PPI [10], the files evaluated jointly by the PPI and the Technical University of Dresden (silicon, lead), files recommended by the International Atomic Energy Agency (IAEA) [11], and several files from the JENDL [12, 13], ENDF [14], and UKNDL libraries.

Files were included in the library on the basis of recommendations made following analysis and expert examination of files by specialists from the PPI and other institutes. All the files were in ENDF/B-V format and were checked with the CHECKER and FIZCON programs for format conformity and physical correlation.

There follows a list of libraries which performed neutron data evaluations for certain isotopes included in this library.

IAEA	Hydrogen, 3 He, 10 B, 12 C, 237 Np
PPI	Deuterium, ^{6,7} Li, oxygen, sodium, chromium,
	50,52-54 Cr, iron, 54,56-58 Fe, nickel,
	58,60-64 238 Ni, U

NPI of the Byelorussian SSR Academy of Sciences ²³⁵U, ²³⁹⁻²⁴²Pu Technical University of Dresden and the PPI Silicon, ⁹³Nb, lead ENDF/B-IV Nitrogen UKNDL ^{241,243}Am In addition, there follows a list of libraries which performed neutron data evaluations for major fission products included in this library.

PPI 99_{Te} , $101,102,104_{Ru}$, 103_{Rh} , $105,107_{Pd}$, 109_{Ag} , 129_{I} , 131_{Xe} , 135_{Cs} , 144_{Ce} , $143,145_{Nd}$, 147_{Pm} , $147,149,151_{Sm}$ ENDF/B-V 141_{Pr} , $151,153_{Eu}$ JENDL-1 106_{Ru} , 133_{Cs} JENDL-2 95_{Mo}

Files included in the recommended evaluated neutron data library meet current standards. However, from the point of view of nuclear data accuracy requirements some evaluations need further refinement, in particular, the capture cross-sections of structural materials at energies below 1 MeV, the inelastic scattering excitation functions in the threshold region, the capture cross-sections of certain fission products, and the threshold reaction cross-sections for certain nuclei.

Expert analysis of the evaluations showed that to obtain more precise recommended neutron cross-sections for the most important reactor materials a further refinement of measuring techniques and theoretical neutron data analysis will be necessary.

REFERENCES

- [1] BLOKHIN, A.I., BULEEVA, N.N., KOLESOV, V.E. et al., Postanovka na EhVM EC-1033 kompleksa programm dlya raboty s bibliotekami otsenennykh nejtronnykh dannykh v formate ENDF/B (Installation on the EC-1033 computer of a software system for handling ENDF/B-format evaluated neutron data libraries) Vopr. at. nauki u tekh. Ser. Yad. konstanty 1 (55) (1984) 16.
- BELANOVA, T.S., GORBACHEVA, L.V., GRUDZEVICH, O.T. et al., "Sravnitel'nyj analiz otsenok sechenij radiatsionnogo zakhvata nejtronov dlya vazhnejshikh produktov deleniya" (Comparative analysis of evaluations of neutron radiative capture cross-sections for major fission products) In: Nejtronnaya fizika (Neutron Physics): (Proc. 6th All-Union Conf. on Neutron Physics, Kiev, 2-6 October 1983), Vol. 2, TsNIIatominform, Moscow (1984) 92; At. Ehnerg. <u>57</u> (1984) 243.

- [3] GRUZDEVICH, O.T., IGNATYUK, A.V., MASLOV, V.M., PASHCHENKO, A.B., "Soglasovannoe opisanie sechenij reaktsij (n,f'f) i (n,xn) dlya transuranovykh yader" (Consistent description of (n,f'f) and (n,xn) reaction cross-sections for transuranic nuclei) In: Nejtronnaya fizika (Neutron Physics): (Proc. 6th All-Union Conf. on Neutron Physics, Kiev, 2-6 October 1983), TsNIIatominform, Moscow (1984) 318.
- [4] IGNATYUK, A.V., KORNILOV, N.V., MASLOV, V.M., PASHCHENKO, A.B., Izomernoe otnoshenie i sechenie reaktsii ²³⁷Np(n,2n) (Isomeric ratio and cross-section of the ²³⁷Np(n,2n) reaction) (Proc. 15th Internat. Symp. on Fission Physics), Gaussig (1985).
- [5] KON'SHIN, V.A., ANTSIPOV, G.V., SUKHOVITSKIJ, E.Sh. et al., Otsenennye nejtronnye konstanty urana-235 (Evaluated nuclear constants for ²³⁵U), Nauka i tekhnika, Minsk (1985).
- [6] ANTSIPOV, G.V., KON'SHIN, V.A., SUKHOVITSKIJ, E.Sh., Yadernye konstanty dlya izotopov plutoniya (Nuclear constants for plutonium isotopes), Nauka u tekhnika, Minsk (1982).
- [7] NIKOLAEV, M.N., BAZAZYANTS, N.O., ZABRODSKAYA, A.S. et al., Nejtronnye dannye dlya dejteriya (Neutron data for deuterium), Obninsk, Analiticheskij obzor OB-114 (1980).
- [8] NIKOLAEV, M.N., ABAGYAN, L.P., BAZAZYANTS, N.O. et al., Nejtronnye dannye dlya kisloroda (Neutron data for oxygen), Obninsk, Obzor OB-77 (1977).
- [9] NIKOLAEV, M.N., ABAGYAN, L.P., KORCHAGINA, Zh.A. et al., Nejtronnye dannye dlya urana-238 (Neutron data for ²³⁸U), Obninsk, Obzor OB-45 (1978).
- [10] BONDARENKO, I.M., PETROV, Eh.E., Otsenka sechenij vzaimodejstviya nejtronov s yadrami ⁶Li dlya rascheta kerma-faktora (Evaluation of cross-sections of the interaction of neutrons and ⁶Li nuclei for kerma-factor calculations), Vopr. at. nauki i tekh., Ser. Yad. konstanty 3 (57) (1984) 35; BONDARENKO, I.M., PETROV, Eh.E., Otsenka sechenij vzaimodejstviya nejtronov s yadrami ⁷Li dlya rascheta kerma-faktora (Evaluation of cross-sections of the interaction of neutrons and ⁷Li nuclei for kerma-factor calculations), ibid. 44.
- [11] Nuclear data standards for nuclear measurements, IAEA, Vienna, Techn. Reports Series No. 227 (1983).
- [12] Summary of JENDL-2 general purpose file (NAKAGAWA, T., Ed.) NEANDC(J)-99/AU (1984).
- [13] KIKUCHI, Y., NAKAGAWA, T., MATSUNOBU, H. et al., Neutron cross-sections of 28 fission product nuclides adopted in JENDL-1, JAERI-1268 (1981).
- [14] ENDF/B Summary Documentation, ENDF-201 (1979).

.

EVALUATION OF NUCLEAR DATA FOR HEAVY FISSILE NUCLEI

V.A. Kon'shin

Significant progress has been made in recent years in the theoretical interpretation of neutron cross-sections. Neutron data have been evaluated using the coupled channel method, accurate models for level density and transitional fission states, and a multistep statistical model taking into account the possibility of pre-equilibrium decay, thus making further development possible in the evaluation of neutron cross-sections. In spite of the progress made, however, not all problems have been solved.

Application of the coupled channel method to evaluate neutron cross-sections for fissile nuclei

The application of the coupled channel method to the evaluation of nuclear data required a computer program which was fast enough to carry out detailed calculations for several nuclei. Ref. [1] describes the methods used by the present author to speed up the numerical solution of equations of the unified optical model. By modifying the computer code implementing the coupled channel method, it was possible to use this method for odd nuclei with a high spin value in the ground state - for example, $235_{U(7^{-}/2)}$. The potential parameters were optimized by the search program using the method of conjugate gradients, while the parameters were fitted to the experimental data not at individual points, but simultaneously over the 1 keV-20 MeV energy range. The experimental data from which the potential parameters were derived were evaluated values of s_0^{\prime} , s_1^{\prime} and σ_0^{\prime} in the energy region of several keV and those of the total cross-section σ_{\downarrow} in the energy range 1 keV-20 MeV. In addition to these data, highly reliable experimental data on the angular distributions of elastically and inelastically scattered neutrons were used, in which the contribution of the levels was clearly distinguished and the compound mechanism contribution disregarded. Optimizing the software

and speeding up the calculations made it possible to use the value χ^2 as a quantitative criterion for fitting the non-spherical potential parameters and to obtain optimal unified optical model parameters, common to the group of heavy nuclei. The use of these parameters enabled the existing experimental data on optical cross-sections for 238 U, 235 U, 239 Pu, 240 Pu and 232 Th to be described almost within the limits of experimental error [1].

On the basis of calculations made with the coupled channel method, an evaluation was performed of the optical cross-sections of neutron interaction with 235 U, 236 U, 238 U, $^{239-242}$ Pu nuclei in the energy range 1 keV-20 MeV. The difference between the theoretical and experimental values of the total cross-sections was no more than $\pm 2\%$ for the entire energy range under consideration (Fig. 1). A detailed comparison was made between differential elastic and inelastic neutron scattering cross-sections calculated using the unified optical model and existing experimental data for 238 U, 235 U and 239 Pu. The calculated cross-sections allowed for the contribution of neutron scattering through the compound nucleus, which is significant at energies below 4 MeV. It emerged that the old experimental data on angular





distributions of "elastic" scattering (i.e. the vast majority of data) could be correctly interpreted only if account were taken of the fact that they include an inelastic scattering contribution for at least the first two excited levels. Accordingly, evaluated data based on an expansion of the experimental data in Legendre polynomials significantly underestimate the anisotropy of elastic scattering. This conclusion can be drawn from Figs 2-4, which compare experimental and theoretical cross-sections (calculated using the coupled channel method and the statistical model) of the integral elastic interaction for 235 U and the angular distributions of scattered neutrons for 238 U.

The coupled channel method was developed for nuclei with rotational or vibrational level bands but no program has yet been devised to take account of the relationship between these bands. Work is being carried out on a computer program which will calculate the cross-sections of neutron interaction with nuclei in which the low-lying levels can be described as a mixture of rotational and vibrational states. This method was used to calculate the



Fig. 4 A comparison of theoretical and experimental [9] data on the excitation of 2350 level groups for inelastic neutron scattering. Sum of levels 103, 129.3 and 150.5 keV (curve 1); 103 and 129.3 keV (curve 2); 170.7, 171.4, 197.1 and 150.5 keV (curve 3); 170.7, 171.4 and 197.1 keV (curve 4); 225.4, 291.1, 294.7 keV (curve 5); 6 - total inelastic scattering cross-section ($\mathbf{F} - [6], \mathbf{F} - [9], \mathbf{F} - [10]$).

excitation functions of 238 U vibrational levels [11]. It emerged that the cross-section for the direct excitation of these levels was low (approximately $1 \cdot 10^{-31}$ m²), so that the calculation of the level excitation cross-section using compound nucleus formation yields a good description of the experimental data [11]. The advantages of the model under consideration should become more apparent in the case of structural materials nuclei.

Use of the statistical model of the nucleus to calculate and evaluate neutron cross-sections of fissile nuclei

The neutron permeability coefficients obtained by the coupled channel method were used in statistical nuclear model calculations. The accuracy of the neutron permeability calculation affects first of all the value of the compound nucleus formation cross-section and hence the reliability of the calculation and evaluation of both the total inelastic scattering cross-section and the cross-section for individual levels. It was shown in Ref. [1] that using permeabilities from the unified optical model in statistical model calculations and taking into account the direct excitation of lower levels lead to closer agreement for ²³⁹Pu with experimental data not only for lower levels but also for those whose excitation cross-sections are determined entirely by the decay of the compound nucleus.

Allowance for collective effects in level density was made using a method describing the mean characteristics of excited nuclei [12]. During the evaluation of neutron cross-sections for uranium and plutonium isotopes, a study was made of the influence of various level density models on the values of the cross-sections calculated, particularly σ_{nn} , and $\sigma_{n\gamma}$. Our calculations showed that the use of the traditional Fermi gas model for level density leads to a significant discrepancy between the calculated cross-sections $\sigma_{n\gamma}$ and the experimental data. The closest agreement in respect of the cross-sections $\sigma_{n\gamma}$ and $\sigma_{nn'}(E_q)$ for ²³⁸U and ²³⁹Pu over the whole energy range was obtained using the level density from the Fermi gas

model taking into account collective modes and the spectral factor in the form of two Lorentz lines. The main problem in measuring the ²³⁵ U neutron inelastic scattering cross-section is subtracting the fission spectrum from the total spectrum and the inelastic contributions of low-lying levels from the elastic peak. Armitage et al. determined the inelastic scattering cross-section of neutrons for level groups in U in an experiment with an insufficiently high energy resolution [9]. Figure 4 compares the experimental data from Ref. [9] with the theoretical data obtained in Ref. [1] for three level groups. It is clear from the figure that the integral theoretical and experimental data are in agreement within the limits of experimental error. The low energy resolution in the experiment in Ref. [9] prevents a closer comparison (it is unclear, for example, whether the level 150.5 keV was included in the experimentally measured value σ_{nn} , for the level group 100 < Q < 150 keV). It should be noted that the calculated total inelastic scattering cross-section in the range 1-2.5 MeV is somewhat higher than the experimental data in Ref. [9], which were obtained with a poor energy resolution (Fig. 4, curve 6); this confirms the assumption that the contribution from low-lying levels was included in the elastic scattering in these experiments.

For fissile odd nuclei, it is necessary to take into account the process $(n,\gamma f)$, which is particularly important in calculating the radiative capture cross-section, since it increases the spin and energy dependences of radiation widths. In calculating cross-sections using the statistical model, it is important not only to allow for competition between the $(n,\gamma f)$ and $(n,\gamma n')$ processes, but also to do so in the correct manner. Investigations have shown that it is insufficient to consider the emission of the first γ -quantum alone, as there is a definite probability of fission or inelastic scattering following the emission of several successive γ -quanta and, in addition, the probability of further γ -discharge cannot be disregarded if the excitation energy of the nucleus following emission of the γ -quantum is

greater than B. Competition between fission and inelastic scattering of γ -discharge was therefore considered only for two stages.

In accordance with the principle of detailed balance, the permeability values for excited nuclear states should be used as the neutron permeability coefficients when calculating decay processes in the compound nucleus. However, in practice these permeabilities are usually identical with the neutron permeabilities for the nuclear ground states obtained from the spherical optical model, in which the dependence T_n on the excitation energy of the nucleus is ignored.

The neutron permeability coefficients for excited nuclear states can be obtained using the unified optical model. It should be noted that differences in neutron permeabilities obtained from the spherical optical model and the coupled channel method become particularly significant as the orbital moment 1 increases, at which stage the values of T_n themselves decrease. This has a very great effect on the radiative capture cross-section calculation, since this cross-section is determined mainly by the contributions of channels with low neutron permeabilities, which compare weakly with the (n,γ) process.

Studies of this effect for the first two excited states of the ²³⁸U rotational band have shown that the values of the strength functions and, consequently, of permeabilities for different states vary widely, particularly at low energies of incident neutrons, and that the difference lessens as energy increases. The calculated values of the strength functions for ²³⁸U in the low energy region (approximately 1 keV) were: $s_0^{0+} = 1.16 \cdot 10^{-4}$, $s_1^{0+} = 1.94 \cdot 10^{-4}$, $s_0^{2+} = 1.03 \cdot 10^{-4}$, $s_1^{2+} = 1.89 \cdot 10^{-4}$, $s_0^{4+} = 0.79 \cdot 10^{-4}$, $s_1^{4+} = 3.74 \cdot 10^{-4}$. Calculations of the cross-section $\sigma_{n\gamma}$ for ²³⁸U indicated that the use of permeabilities for the excited states 2^+ and 4^+ from the unified optical model yield a much better description of the experimental data in the incident neutron energy range 0.3-0.8 MeV (Fig. 5).

A recent evaluation of the radiative capture cross-section for 236 U at the Nuclear Power Institute (NPI) of the Byelorussian SSR Academy of



Fig. 5 Influence of the difference in neutron permeabilities for the ground and excited states on calculated values of σ_{re} for 238 U: 1 - coupled channel method for ground state only; 2 - coupled channel method for ground and first two excited states. The experimental points are taken from Ref.[1].



Sciences confirms the experimental data in Refs. [13, 14]. These data are approximately 40% lower than previous experimental results. As shown by the calculations, it is hard to describe even these very low experimental data for the $\sigma_{n\gamma}$ cross-section with the same parameters over the entire 1-300 keV energy range, without making allowance for the difference in the strength function values for the ground and excited states (Fig. 6). Calculations using the coupled channel method give the following strength function values for 236 U: ${}^{0+}_0 = 1.156 \cdot 10^{-4}$, ${}^{0+}_1 = 1.74 \cdot 10^{-4}$, ${}^{2+}_0 = 1.0 \cdot 10^{-4}$, ${}^{2+}_1 = 1.54 \cdot 10^{-4}$, ${}^{4+}_0 = 0.78 \cdot 10^{-4}$, ${}^{4+}_1 = 3.0 \cdot 10^{-4}$.

Level density of deformed axisymmetric nuclei and parameter systematics

Nuclear level density plays a deciding role in all practical applications of the statistical model of nuclear reactions. The Fermi gas model has proved unable to describe the absolute value of the nuclear level density observed at an excitation energy equal to the neutron binding energy. This is clearly because the model is based on the concept of complete mixing of collective degrees of freedom in the excited nucleus, and takes no account of collective effects; however, these are included in the phenomenological model of level density developed in Ref. [12]. A model which takes account of the basic concepts of the structure of excited nuclei is a relatively simple and convenient tool for practical applications.

The superfluid nuclear model, which allows for collective modes, was used in papers evaluating nuclear data in the energy range above the discrete spectrum of levels. The constant temperature model is very useful for describing the density of low-lying levels in the range from 1.0 to 1.5 MeV:

$$\rho(U, J^{S_{L}}) = \frac{1}{\bar{T}_{n}} \exp\left[(U - E_{0})/\bar{T}_{n}\right] \left[(2J + 1)/2\sigma_{3\kappa cn}^{2}\right] \exp\left[-J(J + 1)/2\sigma_{3\kappa cn}^{2}\right] ,$$

where $E_0 = -n\Delta_0$; n = 1, 2, 3 for even-even, odd-even (even-odd) and odd-odd nuclei respectively; $\overline{T}_n = 0.385$ MeV; $\sigma_{exper.}^2 = 0.156A - 26.76$. The matching point U_c of the constant temperature model and the superfluid nuclear model is $U_c = 10.72 - n\Delta_0 - 0.028A$. The spin dependence parameter σ^2 is equal to $\sigma_{exper.}^2$ up to the excitation energies 1.2, 0.6 and 0.3 MeV for even-even, odd-even (even-odd) and odd-odd nuclei. For higher energies of up to U_c , the value of σ^2 is determined by linear interpolation between the values $\sigma_{exper.}^2$ and $\sigma_{\perp}^2(U_c) = F_{\perp}t$. In this

case, the dependence of the asymptotic parameter of level density \tilde{a} on the mass number A takes the form $\tilde{a} = 0.484A - 0.0016 A^2$. These level density models make it possible to reproduce the energy dependence of the experimental cross-sections for the (n,n'f) and (n,xn) reactions. The density of low-lying transitional fission states can also be described using the constant temperature model. Such an approximation of level density enables the cross-sections σ_f and σ_{n2n} in the threshold region to be described.

The level density in the range of low-lying states for transactinides (A = 225-254), on which no experimental data are available, is described with sufficient reliability using the mean parameters \overline{T} and E_0 . The level density of rare earth nuclei (A = 150-193) is accurately described by the constant temperature model with the parameter $\overline{T} = 0.1509 \cdot 10^{-2} A + 0.7473$ and the same values of E_0 as for transactinides. The parameter systematics obtained for the constant temperature model allow the observed neutron resonance density $\langle D \rangle_{obs}$ for transactinides and rare-earth nuclei to be described with an accuracy no worse than $\pm 50\%$. The difference between the theoretical and experimental data $\langle D \rangle_{obs}$, and also to the fact that some physical effects may not be properly taken into account. Thus, the error associated with the shell corrections affects the energy dependence of the level density ground parameter. A more reliable determination of the energy dependence of the contribution of collective effects is also required.

The results of the systematics make it possible in some cases to evaluate the reliability of the experimental data for $\langle D \rangle_{obs}$. Thus, it is very probable that the experimental values for $\langle D \rangle_{exper}$. for ²⁴⁵Pu and ²⁵³Cf have been underestimated by a factor of about 2.

Evaluation of fission and (n,xn) reaction cross-sections for actinides

The almost total absence of experimental data on the (n,xn) and (n,n') reaction cross-sections for transactinides makes it necessary to use theoretical models to evaluate neutron cross-sections. A consistent calculation of the cross-sections for these reactions is possible only if the calculation of fission cross-sections is sufficiently accurate. However, it has transpired that even if the experimental values of the fission cross-sections can be adequately reproduced, the theoretical values of (n,xn) reaction cross-sections can vary substantially.

This discrepancy is due to the difference in evaluations of the contribution from the first and second "chances" to fission. This problem can be resolved if a consistent analysis is made of existing experimental data. Such an analysis was carried out for 238 U and 235 U in Ref. [15], for which experimental data were obtained for the (n,f), (n,2n) and (n,3n) reactions, and for 238 U, for which experimental data were obtained for secondary neutron spectra. A consistent evaluation of (n,f) (n,2n), (n,3n) reaction cross-sections for $^{238-244}$ Pu was performed in Ref. [16].

By successively taking account of collective, superfluid and shell effects in the level density of neutron and fission channels, it was possible to reproduce the energy dependence of experimental fission cross-sections in the neutron energy range of up to the (n,n'f) reaction threshold (Fig. 7). The need to allow for deformations that disrupt the axial and mirror asymmetry of the fissile nucleus in the saddle configurations was shown, as was the need to use the correlation function in the transitional state $\Delta_f (\Delta_f = \Delta_0 + 0.08 \text{ MeV}).$

To describe (n,n'f) and (n,xn) reaction cross-sections in the threshold region, it is necessary to use the low-lying states so that the competition between fission and reactions with the emission of neutrons and γ -quanta can



10

c

Ē_n,

15

2,0

1,5

1,0

Ó





be taken into account. The constant temperature model with the appropriate parameters gives a fairly good description of the cumulative sum of levels N(E) of transactinides and can be used to describe the characteristics of low-lying states in the case of equilibrium deformation [1]. The density of transitional states of the fissile nucleus can also be approximated by the constant temperature model. This approximate description of transitional state level density in the second and third plateau region is possible because the fission parameters are averaged to a significant extent owing to the emission of prefission neutrons. In calculating (n,xn) reaction cross-sections above the threshold (n,n'f), account must be taken of the possibility of pre-equilibrium emission of the first neutron. The multistep statistical model which takes into account the laws of conservation of angular momentum and parity at all stages of nuclear decay, was used to describe (n,xn) reaction cross-sections [17]. Figure 8 shows the results of calculating (n,2n) and (n,3n) reaction cross-sections for Pu using this model.

Parametrization of secondary neutron spectra for ²³⁸U using the pre-equilibrium decay model made it possible to reproduce fission cross-sections of plutonium isotopes in the 1-20 MeV range and, consequently, to evaluate the (n,xn) reaction cross-sections. To overcome the difficulty of

clearly separating the influence on the fissionability of the nucleus of level density changes and the contribution of pre-equilibrium decay, it is important to obtain experimental data on the hard part of the inelastic scattering spectra of neutrons with an energy above 10 MeV.

Evaluation of mean parameters taking account of corrections for omitted levels

The average widths and, in particular, the mean level spacing <D>, must be known in order to calculate average fission and radiative capture cross-sections and self-shielding factors using the statistical model. This quantity may be obtained from data in the resolved resonance energy region with correction for the omitted levels. Owing to the preponderance of small neutron widths in the Porter-Thomas distribution, the correction for omitted weak levels may be significant.

Methods based on an evaluation of the number of omitted levels using the Porter-Thomas distribution employ the same assumptions and differ only in the degree of generalization. The most advanced of existing methods for introducing corrections for omitted levels, - those of Coceva et al. [18] and Froehner [19] - have been successfully incorporated into computer codes.

These methods are based on the use of the Porter-Thomas distribution for neutron widths. Their authors define in various ways the "distorted" Porter-Thomas distribution resulting from the omission of levels and to a much lesser degree attempt to use the "distorted" Wigner distribution. The two distributions were used together in a method developed in our laboratory [20]. It is likely that the inclusion of both distributions (Porter-Thomas and Wigner) will make it possible to take more accurate account of experimental conditions and make a more accurate adjustment for the omission of levels. The first calculations carried out by this method have shown that the mean level spacing <D> is approximately 10% lower than that obtained using the Froehner method (see Table).

6 <mark>- 11 - 1</mark>		Kethod o	fwork	
Nucleus	Parameter	<i>[</i> 18 <i>]</i>	<u>[]</u> [] [] [] [] [] [] [] [] [] [] [] [] []	[20]
235 _U	$< g\Gamma_n^o >, 10^{-5} eV$ < D >, 2B $< S_o >, 10^{-4}$	3,39 <u>+</u> 0,38 0,445 <u>+</u> 0,011 0,65 <u>+</u> 0,09	4,07 <u>+</u> 0,40 0,434 <u>+</u> 0,016 0,94 <u>+</u> 0,09	3,68±0,37 0,400±0,016 0,92±0,09
236 _U	$<\Gamma_n^{\circ}>, 10^{-3}$ eV <d>, 8B <math><s_0>, 10^{-4}</s_0></math></d>		1,76±0,20 15,1±0,5 1,16±0,20	1,64±0,25 14,1±0,5 1,16±0,18
240 _{Pu}	$< \rho \Gamma_n^0 >, 10^{-3}$ $< D >, 3B^{-3}$ $< S_0 >, 10^{-4}$	1,38±0,05 13,5±0,5 1,02±0,06	1,33±0,06 13,1±0,3 1,01±0,05	1,30 <u>+</u> 0,05 11,99 <u>+</u> 0,40 1,08 <u>+</u> 0,05
239 _{Pu}	$< g \Gamma_n^o >, 10^{-4} e^{V}$ $< D >, 5^B$ $< S_0 >, 10^{-4}$	2,51±0,15 2,17±0,04 1,16±0,05	2,51±0,10 2,17±0,04 1,16±0,05	

Comparison of mean parameter values obtained by various methods (resonance parameters evaluated by the NPI were used as reference parameters)

Application of the above methods in the analysis of evaluated data for 238 U has demonstrated that, if resonance parameters obtained in evaluations in Refs [21, 22] are used as the reference, then in the first case we have $<D_0>/<D_1> \approx 3$, and in the second $<D_0>/<D_1\approx 4$. This is due to the significant discrepancies in the two evaluations of the number of p-resonances and their widths.

Compilation of complete evaluated nuclear data files

Complete files of nuclear data constants ^{235,236}U and ²³⁹⁻²⁴²Pu in the energy range 10⁻⁵ eV-20 MeV were compiled using the above advanced methods of neutron data evaluation. Systems of evaluated nuclear constants compiled earlier were re-evaluated for plutonium isotopes in view of the appearance of new experimental data and more accurate theoretical models for calculating neutron cross-sections.

A comparison of the results of our evaluation of nuclear data with the ENDF/B-V evaluation reveals, among other things, a substantial difference in the ²³⁵U level excitation functions for inelastic neutron scattering (Fig. 9), because the contribution of direct processes is not taken into account in the ENDF/B-V data. There are also differences in the values of



Fig. 8 Comparison of data on (n,2n) and (n,3n) reaction cross-sections for (a) 238pu, (b) 239pu, (c) 240pu. Continuous curve-calculation results [16]; dashed curve - evaluation of ENDF/B-V; dot-dashed curve - evaluation from Ref.[19]; \mathbf{I} - experimental data from Ref.[18].

calculated compound contributions to the level excitation functions, as the method of calculating σ_{nn} , cross-sections, the level scheme and the method of calculating fission competition are not accurate enough in ENDF/B-V. The differences in level excitation cross-sections were, of course, also reflected in the total inelastic scattering cross-section. The results of our evaluation and that of the ENDF/B-V differ by a factor of 1.5-2 for the



Fig. 9 Comparison of results of the present evaluation and the ENDF/B-V data [23] for 235U excitation functions at three levels: (a) 46.21 keV, (b) 103.03 keV and (c) 171.36 keV for inelastic neutron scattering. Curves: 1 - evaluation [1]; 2 - ENDF/B-V evaluation; 3 - direct excitation process; 4 - compound process (3 and 4 from calculations in Ref.[1]).

 σ_{nn} , cross-section. The greater difference in the 0.1-5.0 MeV range is due to the inclusion of the contribution of direct processes to the low-lying states of 235 U, which were disregarded in the ENDF/B-V data. In the range of energies above 7 MeV, the higher value obtained for the σ_{nn} , cross-section in our evaluation is due to the inclusion of pre-equilibrium processes in that evaluation. Both of these effects lead to a hardening of the emitted neutron spectrum.

The angular distributions of inelastically scattered neutrons at low-lying levels are assumed to be isotropic in the ENDF/B-V library, whereas in the present evaluation they were calculated using the coupled channel method and proved to be anisotropic. The energy spectra of secondary neutrons in the ENDF/B-V data have a Maxwellian form, whereas in accordance with the pre-equilibruim decay model they are harder in the present evaluations.

A comparison of the most recent presentations with ENDF/B-V evaluated data for 235 U shows that the latter data are inadequate.

REFERENCES

- [1] KON'SHIN, V.A., ANTSIPOV, G.V., SUKHOVITSKIJ, E.Sh. et al., Evaluated neutron constants for Uranium-235, Nauka i tekh. Minsk (1985) [in Russian].
- [2] POENITZ, W.P., WHALEN, J.F., SMITH, A.B. et al., Total neutron cross-sections of heavy nuclei. In: Proc. of the intern. conf. on nuclear data for technology (Knoxville, 1979) USA (1979) 698-704.
- [3] SCHWARTZ, R.B., SCHRACK, R.A., HEATON, H.T., Total neutron cross-sections of 235 U, 238 U and 239 Pu from 0.5 to 15 MeV, Nucl. Sci. Eng., <u>54</u> (1974) 322-326.
- FOSTER, D.J.Jr., GLASGOW, D.W., Neutron total cross-sections at 2.5-15.0 MeV.
 Experimental. - Phys.Rev., <u>C3</u> (1971) 576-603.
- [5] CABE, I., CANCE, M., ADAM, A. et al., "Measure des sections efficaces totales neutroniques du carbone, du nickel, de l'uranium-235, de l'uranium-238 et de plutonium-239 entre 0.1 MeV and 6 MeV" (Measurement of total neutron cross-sections for carbon, nickel, uranium-235, uranium-238 and plutonium-239 between 0.1 MeV and 6 MeV), Nuclear Data For Reactors (Proc. Conf. Helsinki, 1970), Vol. 2, IAEA, Vienna (1970) 31-37.
- [6] KNITTER, H.H., ISLAM, M.M., COPPOLA, M., Investigation of fast neutron interaction with ²³⁵U, Z.Phys., <u>257</u> (1972) 108-123.
- SMITH, A.B., Elastic scattering of fast neutrons from ²³⁵U, Nucl.Sci. Eng., <u>18</u> (1964) 126-129.
- [8] KINNEY, W.E., PEREY, F.G., ²³⁸U neutron elastic scattering crosssections from 6.44 to 8.56 MeV, ORNL-4804, (1973).
- [9] ARMITAGE, B.N., FERGUSON, A.T.G., MONTAGUE, J.H. et al., "Inelastic scattering of fast neutrons by ²³⁵U", Nuclear Data For Reactors (Conf. Proc. Paris, 1966), IAEA, Vienna (1967) 383-392.

- [10] BATCHELOR, R., WYLD, K., Neutron scattering by ²³⁵U and ²³⁹Pu for incident neutrons of 2, 3 and 4 MeV: Rep. AWRE-055/69. Aldermaston (1969) 16.
- [11] EGAN, J.J., ARTHUR, E.D., KEGEL, G. et al., "A comparison of measured and calculated neutron inelastic scattering cross-sections for vibrational states from 680 to 1060 keV in ²³⁸U", Nuclear Data For Basic And Applied Science, (Conf. Proc.), USA, Santa Fe (1985).
- [12] IGNATYUK, A.V., ISTEKOV, K.K., SMIRENKIN, G.N., The role of collective effects in the systematics of nuclear level density, Yad. Fiz. (Soviet Journal of Nuclear Physics), <u>29</u> (1979) 875-883 [in Russian].
- [13] KAZAKOV, L.E., KONONOV, V.N., POLETAEV, E.D. et al., Measurement of radiative capture cross-sections for ²³⁶U and ¹⁹⁷Au nuclei in the energy range 3-420 keV, Vopr. at. nauki i tekh., Ser. Yad. konstanty 2 (1985) 44-49 [in Russian].
- [14] DAVLETSHIN, A.N., TIPUNKOV, A.O., TIKHONOV, S.V. et al., Cross-sections for the radiative capture of neutrons by ¹⁹⁷Au, ²³⁶U and ²³⁷Np, At. Ehnerg., <u>58</u> 3 (1985) 183-188 [in Russian].
- [15] GRUDZEVICH, O.T., IGNATYUK, A.V., MASLOV, V.M. et al., "Consistent description of the (n,n'f) and (n,xn) reaction cross-sections for transuranic nuclei" In: Neutron Physics: Proc. of 6th All-Union Conference on Neutron Physics, (Kiev, 2-6 October 1983 Vol. 2) TSNIIatominform, Moscow (1984) 318-323 [in Russian].
- [16] KONSHIN, V.A., KLEPATSKY, A.B., MASLOV, V.M. et al., "Possibilities of updating Pu fast neutron cross-sections with a theoretical analysis". In: [11].
- [17] UHL, M., STROHMAIER, B., STAPRE-Computer code, Rep. IRK-76/01, Vienna (1976).
- [18] COCEVA, C., STEFANON, M., Experimental aspects of the statistical theory of nuclear spectra fluctuations, Nucl. Phys, <u>315</u> (1979) 1-20.
- [19] FROEHNER, F.H., "Level density estimation with account of unrecognized multiplets applied to uranium and plutonium resonance data" (Proc. of the IAEA consultants meeting on uranium and plutonium isotope resonance parameters, Vienna, 1981), IAEA, Vienna (1981) 103-111.
- [20] PORODZINSKIJ, Yu.V., SUKHOVITSKIJ, E.Sh., Method for determining mean neutron widths and mean level spacing taking account of the experimental equipment's final resolution, Izv. Akad. Nauk Belor.SSR, Ser. Fiz.-Ehnerg. <u>1</u> (1986) 15-19 [in Russian].
- [21] DE SAUSSURE, G., OLSEN D.K., PEREZ, R.B. et.al., "Evaluation of the ²³⁸U neutron cross-sections for incident neutron energies up to 4 keV", Progress in Nuclear Energy, <u>3</u> (1979) 87-124.
- [22] NIKOLAEV, M.N., ABAGYAN, L.P., KORCHAGINA, Zh.A. et al., Neutron data for ²³⁸U, Obninsk, Power Physics (FEhI) Institute (1978) 241 [in Russian].
- [23] BHAT, M.R., Evaluated nuclear data file/B. Version V, MAT-1395, (1977).

ABSOLUTE MEASUREMENTS OF THE FISSION CROSS-SECTIONS FOR IMPORTANT NUCLIDES

V.I. Shpakov

Since 1975, the V.G. Khlopin Radium Institute in Leningrad and the Technical University of Dresden have been working together on absolute measurements of the fission cross-sections for the most important reactor nuclides. This paper reviews the results of measurements carried out in 1984-1985. The aim of this collaboration is to measure very accurate values for fission cross-sections at fixed neutron energies using the timecorrelated associated particle method, these being essential for greater accuracy and standardization of relative measurement data. This undertaking was endorsed by the decisions of an IAEA international consultants' meeting [1] which recommended that measurements using the time-correlated associated particle method should be carried out for as large a number of neutron energies as possible.

In Dresden, the measurements were carried out on the tandem accelerator at the Central Institute for Nuclear Research (Rossendorf). Measurements were made of fission cross-sections for 235 U at neutron energies of 4.45 and 18.8 MeV neutrons and for 239 Pu - at neutron energies of 4.8, 8.65 and 18.8 MeV neutrons. In the Radium Institute, the fission cross-sections for 233 U, 235 U, 237 Np and 239 Pu were measured on the Van de Graaff accelerator at 10wer neutron energies.

The time-correlated associated particle method used is well known, and was described in detail in Ref. [2]. Consequently, we need only note the main features of the method. The $D(d, n)^{3}$ He or $T(d, n)^{4}$ He reaction was used as the neutron source. The neutron-associated helions were registered by an associated particle detector. Its inlet aperture determines the cone of helions registered and, at the same time, sets the cone of neutrons correlated with them which are bombarding the fissile nuclide target under investigation. Fission events are registered in coincidence with associated

particles. If the neutron cone lies entirely within the boundaries of the fissile material target, the fission cross-section is defined by the simple expression $f = N_f / (N_c n)$, where N_f is the number of coincidences registered, N_c is the number of associated helions registered, and n is the number of nuclei of the fissile material per cm² of the target.

The principle behind this method is simple and it has definite advantages: there is no need to measure the neutron flux or - at least - the total associated particle integral, nor to measure solid angles with respect to the target and the detector, and there is no background due to scattered neutrons from side reactions etc. However, practical implementation of the method involves significant problems since associated particles must be registered against a strong background of scattered deuterons and charged particles produced by concomitant reactions in the neutron source and target materials, and in support and structural materials. Consequently, specific associated particle channels have to be set up for each neutron energy value.

For the measurements carried out at the Technical University with 4.45, 4.8 and 8.65 MeV neutrons, the $D(d, n)^3$ He reaction was used to produce neutrons. The target was a thin foil (approx. 1 mg/cm²) of deuterated polyethylene.

For the measurements with 18.8 MeV neutrons, the T(d, n)⁴He reaction was used to produce neutrons, and thin (approx. 1 mg/cm^2), flying, self-supporting titanium-tritium targets were employed. Modifications of the $(E-E_r)$ -telescope were used to register associated particles. The usual methods for analysing $E-E_r$ spectra could not be used owing to their lengthiness, and therefore the method of separating out helions described in Ref. [3] was applied. It involved forming a combination of the signals $E_g = (E + E_r)$ and $A_p = (aE + bE_r)$ and sorting them in the amplitude windows E_g and A_p . The corresponding range of values for a, b, E_g and A_p determined the optimal registration region in the two-dimensional spectrum $E-E_r$ for each neutron energy. This system ensured a maximum discriminator resolution
time of 100 ns, and effective registration of associated particles when the background in the channel does not exceed 2-5%. The background level was determined by replacing the deuterated polyethylene with normal polyethylene, or (where the target was being used) with titanium foil not saturated with tritium. The experiment methodology is described in detail in Refs [4] and [5].

In the Radium Institute, a deuterated polyethylene foil (thickness: $1-2 \text{ mg/cm}^2$) was used as a neutron source for the measurements. The neutrons emitted to the rear hemisphere were used, and helions emitted at small angles to the deuteron beam direction were registered. This produced a very high background of scattered deuterons. To remove this background and also the background of other charged particles, in particular the protons produced by the interaction of the deuteron beam with residual hydrogen in the target, the reaction products were magnetically separated. Helions were registered by a surface-barrier silicon detector of large area and small zone depth. The combination of magnetic separation and this type of detector reduced the background in the associated particle channel to a maximum of 0.1%. The background was determined by replacing the deuterated polyethylene with normal polyethylene. The remaining details of the experiment were the same for all measurements.

The position and profile of the neutron cone was constantly monitored during the experiment using a scintillation counter with a stilbene crystal and $(n-\gamma)$ -discrimination in coincidence with the associated particle detector.

Neutron losses in the cone due to scattering by target supports, windows and structural materials were calculated using a method comprising the solution of the inverse radiation transport problem [6]. The transport equations were solved by the Monte Carlo method.

Fissions were registered by fine multilayer ionization chambers, containing between three and six layers of fissile material in the various

experiments and operating in a pulsed current regime. The shallowness of the chamber (approx. 3 mm deep) permitted fragment pulses to be efficiently separated from alpha-particle pulses, and produced a suitably extensive plateau region. The efficiency of fission registration in this chamber was determined by counting losses due to discrimination in the counting channel and fragment absorption in the fissile material layer. Counting losses due to discrimination were determined by extrapolating the fragment pulse height spectrum to zero pulse height. Fragment absorption in the layer was calculated from the thickness of the layer, taking into account anisotropy and speed of transport as determined by the neutron pulse [7]. For this purpose the mean fragment path in the material was taken to be 7.5 mg/cm².

Corrections for random coincidences were worked out by analysing coincidence time spectra, and also by recording simultaneously the total number of coincidences and the number of random coincidences using the same set up.

The fissile material targets were prepared in the Radium Institute by high frequency sputtering and heat sputtering of the material onto eccentrically revolving substrates. With the exception of the ²³³U targets, isotopically pure materials obtained by the mass separator method were used to prepare the targets. The isotopic composition of the ²³³U targets was as follows (%): ²³²U - 0.003; ²³³U - 82.899 \pm 0.144; ²³⁴U - 0.332 \pm 0.014; ²³⁵U - 0.141 + 0.010; ²³⁸U - 6.628 + 0.038.

The number of nuclei in the target was determined by measuring the alpha activity in a small geometry device. The half-life values recommended in Ref. [8] were used when calculating the number of nuclei. Homogeneity of the active layer of the targets was monitored by scanning the surface using an alpha detector with a small solid angle, or by alpha counting while covering the surface of the target with diaphragms of varying diameter. Discrepancies in homogeneity did not exceed 1% in all the targets used.

Corrections, error components (%) and measurement results.

Isotope	Neutron energy WeV	Coincidence counting		Fission counting			Associated	Neutron cone		Fissile layers		1	1
		Statist- ics	Random coincid- ences	Correlated background	Extrapol- ation to zero	Fragment absorption	particle counting (background)	Neutron scatter- ing	Liftective thick- ness of the targe layer due to the cone aperture	Surface density	Inhomogen- eity of the layer	Total error	<i>б</i> ;. б
235 ₀	1.68	-/I.95 [%]	1.62/0.25	-/	3.95/0.50	I.95/0.85	0.05/0.02	0.35/0.40	0.12/0.06	-/0.93	-/0.72	2.52	(1.26+0.03)
235 ₀	4,45	-/1,26	1.40/0.17	-/-	I.IB/0.26	2,00/0,85	2,32/0,67	0,25/0,40	0 0,05/0,05	-/0,95	-/0.72	2,10	1,057+0,022
235 _U	3,8I	-/I,0I	2,62/0,21	1,72/0,04	1,67/0,16	1,73/0,75	5,62/1,35	0,44/0.40	0,12/0,08	-/0,93	-/0.72	2,25	1,995+0,045
237 _{EP}	1,90	-/1,9I	4,65/0,43	-/-	4,45/0,60	0,95/0,30	0,10/0,05	0,43/0,40	0,12/0,08	-/2.00	-/0,76	3,01	1,73+0,05
235 _{Fu}	I,92	-/2,10	9,06/0,60	-/-	2,03/0,76	0,63/0,30	0,10/0,05	0,38/0,40	0,12/0,08	I.00/I.00	-/0,65	2,72	2,01+0,05
239 ₇₁	4,8	-/1,27	0,64/0,II	-/-	1,50/0,31	1,21/0,46	2,35/0,56	0,25/0,40	0,06/0,05	-/1,00	-/0,69	2,00	I,740±0,035
235 ₃₂₂	Ε,65	-/I.0ô	1,85/0,17	-/-	1,0%/0,24	1,20/0,43	I,62/0,32	0,36/0,40	0,07/0,05	-/1,00	-/0,63	1,85	2,350-0,044
235 _{Pu}	18,8	-/2,52	4,55/0,63	0,54/0,13	2,57/0,85	1,30/0,39	5,92/1,74	0,34/0,40	0,12/0,08	-/I,00	-/0,88	З,55	2,487±0,085
255 0	1,94	-/2,94	13,3/1,1	-/-	I,8/0,5	0,99/0,3 (3,6/0,25)*3	0,1/0,05	0,49/0,40	0,12/0,08	-/2,00	-/0,35	3,6	1.93 <u>+</u> 0.07

Numerator - correction, denominator - error.

****** Contribution of other isotopes.



Fig. 1. Comparison of measurement results obtained for the ²³⁵U fission cross-section with evaluations contained in the following sources: ENDF/B-V; - UKNDL; 0 - ENDL-82; + - JENDL-II; - the present work.





Fig. 3. Comparison of measurement results obtained for the 237 Mp fission cross-section with evaluations in the following sources: ENDF/B-V; 0 - [10]; + - the present work.

Fig. 2. Comparison of measurement results obtained for the fission cross-section of 239 Pu with evaluations contained in the following sources: ENDF|B-V; ---- [9]; + - the present work.



Fig. 4. Comparison of measurement results obtained for the ²³³U fission cross-section compared with evaluations from the following sources: ENDF/B-IV; ----- UKNDL - DFN87B; + - the present work.

The corrections and corresponding error components in the measurement results, and the results themselves, are given in the Table.

A comparison of the results obtained for 235 U, 239 Pu, 237 Np and 233 U and the evaluations of various libraries is given in Figs. 1-4.

It is planned in the future to carry out absolute measurements on ²³⁸U using the time-correlated associated particle method. The fission cross-section of this nuclide is an accepted IAEA neutron standard, although absolute measurement data for it are practically non-existent.

The following persons were involved in carrying out the measurements: at the Radium Institute - I.D. Alkhazov, E.A. Ganza, L.V. Drapchinskij, V.A. Kalinin, S.S. Kovalenko, I.O. Kostochkin, V.N. Kuz'min, L.M. Solin, A.V. Fomichev; and at the Technical University - M. Josch, K. Merla, G. Musiol, H. Ortlepp, G. Pausch, C. Herbach.

REFERENCES

- [1] Proc. of the IAEA consultants meeting on ²³⁵U fast-neutron fission cross-sections, INDC(NDS)-146, Smolenice (1983) 13.
- [2] ARLT, R., et .al., In: Proc. of the intern. conf. on nuclear cross-sections for technology. NBS Spec. Publ. 594, Knoxville (1979) 990.
- [3] ARLT, R., et.al., In: Proc. of the advisory group meeting on nuclear standard reference data, IAEA-TECDOC-335, Geel (1984) 774.
- [4] HERBACH, C.M., et.al., Preprint 05-06-87 of TU-Dresden (1985).
- [5] HERBACH, C.M., et.al., Preprint 05-07-85 of TU-Dresden (1985).
- [6] DUSHIN, V.N., ZfK-382, Dresden (1979) 153.
- [7] ARLT, R., et.al., Preprint 05-05-79 of TU-Dresden (1979).

- [8] Nuclear data standards for nuclear measurements, IAEA Nuclear Standard File 1982: INDC/NEANDC, IAEA Vienna, (1983).
- [9] ANTSIPOV, G., BAKHANOVICH, L.A., ZHARKOV, V.F., et al., Evaluation of Nuclear Data for ²³⁹Pu, Preprint ITMO AN BSSR, No. 14 (1981) (in Russian).
- [10] DARRIEN, H., DOAT, J., FORT, E., Evaluation of ²³⁷Np neutron cross-sections, Rep. IAEA INDC(FR)-42/L (1980).

MEASUREMENT AND ANALYSIS OF RADIATIVE CAPTURE CROSS-SECTIONS OF NEPTUNIUM AND URANIUM ISOTOPES

V.A. Tolstikov

Accuracy requirements for the evaluated ²³⁸U radiative capture cross-section are very high. According to the US Nuclear Data Committee [1], the requirement is that the error should be no greater than 1.5% in the interval 10-300 keV, approximately 2% in the interval 0.3-1 MeV and up to 7% for the 1-10 MeV range. The same requirements clearly also apply to the experimental reference data.

A knowledge of neutron radiative capture cross-sections for 236 U and 237 Np is important both in order to understand the processes of fast neutron interaction with actinide nuclei and to obtain data for testing nuclear reaction models, and from the point of view of the technological processes involved in the fast reactor fuel cycle. In order to calculate the technological processes associated with the accumulation of α -active 236 Pu and 238 Pu, it is essential to know the evaluated capture cross-sections of 236 U to an accuracy of approximately 5% and of 237 Np to an accuracy of approximately 5% and of 237 Np to an accuracy of around 7-10%.

<u>Uranium-238</u>. Let us compare two widely used evaluations of $\sigma_{n,\gamma}^{238}$ U: BNAB-78 [2] (USSR) and ENDF/B-V (USA). Both evaluations were performed quite a long time ago, BNAB-78 in 1975-78 and ENDF/B-V in 1977. Owing to three factors - the further development of methods for evaluating cross-sections and their errors, the appearance of new experimental data, and advances in cross-section measurement methodology - it has become necessary to correct the earlier evaluations, to review the degree of reliability of the published data on which those evaluations were based, and to conduct precision measurements of $\sigma_{n,\gamma}^{238}$ U over a wide range of energies. For the purposes of technology, the required level of accuracy for the evaluated value of $\sigma_{n,\gamma}(E)^{238}$ U has not yet been attained. Initially, the BNAB evaluation used the values from Refs [3, 4] (BNAB-MIKRO), without any change.

The evaluated curve was drawn using the rational approximation method. The evaluation was then reviewed and the cross-sections σ_t , σ_{in} , $\sigma_{n,\gamma}$ were evaluated simultaneously for $E_n < 0.4$ MeV by the maximum-likelihood method using the statistical theory of nuclear reactions. In the overlap area the data matched the curve smoothly for $E_n < 0.4$ MeV. The following procedure was followed in the ENDF/B-V evaluation: a separate evaluation was made for four groups of data obtained by absolute methods or in relation to the (n,p) scattering cross-sections, $\sigma_{n,\gamma}$, ${}^{197}Au$, $\sigma_{n,f}$, ${}^{235}U$ and $\sigma_{n,\alpha}$, ${}^{10}B$. Final data for the recommended curve of $\sigma_{n,\gamma}$ (E) for ${}^{238}U$ were obtained by comparing and combining data from the four groups. Roughly speaking, both evaluations used a similar set of experimental reference data.

Let us consider the energy region $E_n < 0.4$ MeV, for which the greatest amount of experimental data is available. As the status of this range was analysed in detail in Refs [3-6], we shall merely note that a consistent theoretical description of the data was obtained for $\overline{\sigma}_t$, $\overline{\sigma}_{e1}$, $\overline{\sigma}_{n,\gamma}$ and $\overline{\sigma}_{in}$. The evaluated curve for $\sigma_{n,\gamma}(E)^{238}$ U in the 40-100 keV energy range is approximately 4-5% lower than the BNAB-78 evaluation [2], and below 40 keV it is 3-4% higher. It should also be noted that in order to agree with the BNAB-78 integral data for $E_n \leq 0.1$ MeV, the cross-section $\sigma_{n,\gamma}$ was reduced by 3-5% in relation to the BNAB-MIKRO data (evaluated experimental data). In the 0.1-0.4 MeV range the BNAB-78 and ENDF/B-V evaluations are similar.

Figure 1 compares these evaluations at 0.4 MeV. The comparison took into account experimental data from more recent papers which were not, or were only partly, included in the evaluations. It is clear from Fig. 1(a) that the BNAB-78 and ENDF/B-V evaluations show good agreement in the 0.4-1.5 MeV range. The maximum discrepancy in the range 0.5-0.9 MeV does not exceed 4%, the ENDF/B-V evaluation being higher. The data in Ref. [7] for $E_n < 1.1$ MeV are generally higher than the evaluations and in the region 1.38-1.52 MeV are somewhat lower. In the energy range 1.5-3 MeV (see Fig. 1(b)) the evaluations



 Fig. 1.
 238

 U neutron radiative capture cross-sections in the energy range 0.5-1.5 MeV (a), 1-5 MeV (b), 5-14 MeV (c).

 Data:
 • - [7]; ■ - [8]; * - [9, 10]; ---- BNAB-78 [2];

 ---- ENDF/B-V.

essentially coincide, as the set of data used for the evaluations was practically identical. The data in Ref. [7] for the energies 2.053 and 3.033 MeV are significantly higher than the evaluations. There is no reliable theoretical calculation for this energy range, and careful experimental study is therefore required to establish the degree of accuracy of this result. The general feeling is that the experimental data in early papers for the range $E_n > 2$ MeV (mostly obtained by the activation method) are too high because insufficient account was taken of scattering effects. In the 7-14 MeV range (see Fig. 1(c)) no experimental data are available and the nature of the evaluated curve is determined by the value of the cross-section assumed for 14 MeV by extrapolating smoothly from the 4-7 MeV region.

The BNAB-78 file includes extrapolation to 2.6 mb at 14 MeV. In the light of Ref. [11], this figure appears too high: the experimental capture cross-sections in the range of mass numbers 50-240 at 14 MeV are of the magnitude of approximately 1 mb. Accordingly, the data in the BNAB-78 file for the 5-15 MeV range should be revised downwards. The following conclusions can be drawn from the above.

- 1. The status of experimental data and the evaluations based on them for the 1.5-7 MeV energy range is unsatisfactory. Precision measurements of 238 U capture cross-section are required for practical purposes and in order to test methods of calculating actinide cross-sections, which will be difficult to obtain experimentally in the near future. For the 1-7 MeV energy range detailed measurements of $\sigma_{n,\gamma}^{238}$ U in steps of no more than 100 keV are desirable in order to detect possible non-monotonicities in the cross-section. Further improvement of measurement methods is therefore essential.
- 2. For the 0.3-1 MeV energy range the activation method at its present stage of development is already potentially capable of measuring $\sigma_{n,\gamma}^{238}$ U with an error of less than 3%. Other methods cannot yet achieve such accuracy.

- 3. For $E_n < 0.3$ MeV it is essential that new series of independent measurements of $\sigma_{n,\gamma}^{238}$ U and of the shielding coefficients be performed by various groups of experimenters using the time-offlight method so that consistent experimental data with low error levels can be obtained. The exacting demands of the power engineering sector will only be met once a set of experimental data is available in which the data have been obtained by various methods, have a wellfounded and stated error of approximately 3%, and are consistent within the error limits.
- 4. The BNAB-78 and ENDF/B-V files do not provide an adequately substantiated evaluation of the errors associated with evaluated data in various energy ranges. New evaluations should do so and should include a table of errors along the lines of those contained in the IAEA standards file e.g. for σ_f^{235} U.

<u>Uranium-236</u>. The status with respect to ²³⁶U was partially studied first in Ref. [12], and subsequently in Ref. [13]. The situation is unusual because, up to 1970, there existed only two papers showing poor agreement in the overlapping neutron energy ranges ($E_n = 0.3-4$ MeV) [14, 15]. Reference [16] was published in 1970 ($E_n = 0.1-20$ keV). These papers served as the basis for the establishment of the evaluated cross-section $\sigma_{n,\gamma}(E)^{236}U$ in the ENDF/B-V evaluation. It should be noted that Refs [14, 15] employed the activation method, and Ref. [16] the time-of-flight method.

In the past five years new measurements have been performed using the following methods: slowing-down time in lead [17], activation [18-21] and time-of-flight in a pulsed Van de Graaff accelerator [22]. Also, strength functions have been determined for 236 U using the statistical theory of nuclear reactions, and these have been compared with the strength functions of 238 U and 232 Th [23]. Using the rational approximation method, an evaluated curve was produced on the basis of experimental data, and this



+ -[30]; 7_ - [31]; ----- - [32]; --_ - averaged ENDF/B-V evaluations in the 74-group system of constants with constant spectrum.

served as the basis for one of the evaluations of $\sigma_{n,\gamma}(E)^{236}U$ [24]. The current status of $\sigma_{n,\gamma}^{236}U$ for neutrons in the energy range 0.001-4 MeV is shown in Fig. 2. It should be noted that numerical data from Ref. [16] had by this time become available.

Without going into the different characteristics of the experimental methods used, it is worth noting that our first measurements of $\sigma_{n,\gamma}^{236}$ U by the activation method [18] already produced results 50-60% lower than in

the ENDF/B-V evaluation based on Refs [14, 15]. Later measurements relative to various standards [Au(n, γ), (n-p) scattering] using various normalization methods (absolute and in terms of thermal cross-sections) fully confirmed this conclusion [19].

The data in Refs. [18-21], together with Ref. [17], form a group which is systematically lower than the ENDF/B-V evaluation over the entire energy range with the exception of 1-4 keV, where the average data from ENDF/B-V are similar (1-3 keV) to or 20% lower (3-4 keV) than the data in Ref. [17]. For neutron energies of 4-53 keV, the ENDF/B-V data (the experimental data in Ref. [16]) are on average 20-25% higher than the data in Ref. [17]. These discrepancies are within two root-mean-square errors of the experiments (10% for Ref. [16] and 5% for Ref. [17]). The energy dependences of the cross-sections also diverge somewhat. In our view, the question of the shielding of cross-sections in the low-energy region and their absolute normalization should be considered more carefully.

The discrepancies in the energy range above 200 keV are much greater (exceeding 50%). It is possible that again the reason for these differences is systematic errors associated with normalization and the allowance made for the influence of scattered neutrons. On the other hand, theoretical calculations [18, 23] have shown that the group of data with the lowest cross-sections is preferable as it is described by the calculation parameters $\tilde{\Gamma}_{\gamma}^{\text{res}}$ and \tilde{D}^{res} , which are within their error limits. In addition, the (radiative and p-neutron) strength functions obtained by comparing theory and experiment are in good agreement with the strength functions of nuclei having the same parity and a similar number of nucleons (neutrons) - 238 U and 232 Th. In contrast, the description of the group with higher data values requires a \tilde{D} value which is roughly twice as low.

There are no data for $E_n > 1.2$ MeV, apart from one early paper [14]. However, preliminary data for 0.96 and 3.3 MeV which we obtained incidentally in an experiment on the measurement of fission product yields clearly show

that the cross-sections in Ref. [14] are approximately twice as high as they should be.

In 1985, Ref. [22] was published, in which $\sigma_{n,\gamma}^{236}$ U were measured in the 3-420 keV range by the time-of-flight method. In the overlap area (0.17-0.4 MeV), the data from this paper more or less duplicate those from our paper [19] (activation method). In the 6-50 keV range the data from Ref. [22] are close to those in Ref. [16]. In the range above 6 keV they deviate sharply from the data in Refs [16, 17].

It can be concluded that, for the neutron energy range above 1 MeV, new experiments need to be conducted by various methods, including repeat measurements by the activation method. Repeat measurements are also required in the energy range below 0.15 MeV, particularly in the range below 30 keV.

<u>Neptunium-237</u>. Figure 3 shows the data available to us on the $^{237}Np(n,\gamma)$ reaction. The first paper on this reaction – activation data [29] in the neutron energy range 0.15–1.5 MeV – appeared only in 1967. The ENDF/B-V evaluation was carried out using data published in 1976 [28] (activation method, $E_n = 0.1 \pm 3$ MeV). The data from Ref. [19] obtained relative to the ¹⁹⁷Au(n, γ) reaction cross-section and the hydrogen scattering cross-section, are in good agreement with the results of Ref. [28]. The data in Ref. [27], with the exception of the value for 330 keV, differ from the ENDF/B-V evaluation by 50% at energies above 600 keV.

The results in Ref. [29] are clearly overestimated. This may be partly due to normalization. However, the systematic increase in the discrepancy as neutron energy increases may be due to insufficient allowance for 237 Np fission and the influence of scattered neutrons. The data in Refs [30] and [31] (for which we have no numerical results or detailed description) for neutron energies above 150 keV differ greatly. For these energies the results in Ref. [30] are close to the activation results. An analysis of the data in Fig. 3 shows that there is a pressing need for further series of experiments for energies below 200 keV and above 1.5 MeV.

An analysis of data on $\sigma_{n,\gamma}^{238}$ U shows that even the existence of several papers with a demonstrably high accuracy of results is no guarantee of obtaining evaluated data within the required error limits.

Special mention should be made of the present status of knowledge regarding the cross-sections $\sigma_{n,\gamma}^{237}$ Np for velocities of 2200 m/s. The maximum variation in these data is 17% of the average value (187-158 b [33, 34]). The evaluations also fluctuate between 181 [35] and 169 b [32, 36]. New precision measurements and an appropriate analysis of all published data are required.

REFERENCES

- [1] Compilation of requests for nuclear data. January 1983: BNL-NCS-51572. US Department of Energy (1983).
- [2] ABAGYAN, L.P., et al., Group constants for reactor and shielding calculations, Energoizdat, Moscow (1981) (in Russian).
- [3] VINOGRADOV, V.N., DAVLETSHIN, A.N., KRIVTSOV, A.S., et. al., Evaluated average capture cross-sections for ²³⁸U in the energy range 0.001-7 MeV. In: Nuclear Physics Research in the USSR, 22 (1976) 4 (in Russian).
- [4] VINOGRADOV, V.N., DAVLETSHIN, A.N., PLATONOV, V.P., Radiative capture of fast neutrons by uranium-238. In: Neutron Physics: Proc. 3rd All-Union Conference on Neutron Physics, Kiev, 9-13 June 1975, Part 4, TSNIIatominform, Moscow (1976) 104 (in Russian).
- [5] NIKOLAEV, M.N., ABAGYAN, L.P., KORCHAGINA, Zh.A., et al., Neutron data for uranium-238, Part 1: Analytical review A-34 (OB-45), Power Physics Institute, Obninsk (1978) (in Russian).
- [6] MANTUROV, G.N., LUNEV, V.P., GORBACHEVA, L.V., Evaluation of neutron data for ²³²Th and ²³⁸U in the unresolved resonance region. In: Neutron Physics: Proc. 6th All-Union Conference on Neutron Physics, Kiev, 2-6 October 1983, Vol. 2, TsNIIatominform, Moscow (1984) 231 (in Russian).
- [7] FAWCETT, L.R., POENITZ, W.P.Jr., SMITH, D.L., Measurement of the fast neutron capture cross-section. In: Nucl. cross-sections for technol.: Proc. of the Intern. conf. of nuclear cross-sections for technology (Knoxville, 1979). Washington (1980).
- [8] DAVLETSHIN, A.N., TIKHONOV, S.V., TIPUNKOV, A.S., TOLSTIKOV, V.A., Measurement of neutron radiative capture cross-sections for ²³⁸U and ¹⁹⁷Au relative to the proton elastic scattering cross-section, At. Ehnergiya <u>48</u> 2 (1980) 87 (in Russian).
- [9] PANITKIN, Yu.G., TOLSTIKOV, V.A., Neutron radiative capture in ²³⁸U with energies of 1.2-4 MeV, ibid., <u>33</u> (1972) 782 (in Russian).

- [10] PANITKIN, Yu.G., STAVISSKIJ, Yu.Ya., TOLSTIKOV, V.A., Measurement of neutron radiative capture cross-sections for uranium-238 in the energy range 0.024-1.1 MeV, Proc. of All-Union Conference on Neutron Physics, Part 1, Kiev (1972) 321 (in Russian).
- [11] DRAKE, D., BERGQVIST, I., McDANIELS, D.K., Dependence of 14 MeV radiative neutron capture on mass number. Phys. Letters (1980), <u>36B</u>, N 6, 557.
- [12] DAVLETSHIN, A.N., TIPUNKOV, A.O., TOLSTIKOV, V.A., On the discrepancy of two groups of experimental data on the fast neutron radiative capture cross-sections for uranium-236, INDC Report, INDC/P (83-49) (Brazil, Rio de Janeiro, May 1983) (in Russian).
- [13] TOLSTIKOV, V.A., MANOKHIN, V.N., Status of neutron radiative capture data for ²³⁶U and ²³⁷Np. Transactinium isotope nuclear data (1984): Proc. of the third advisory group meeting on transactinium isotope nuclear data (Uppsala, 21-25 May 1984). Vienna: IAEA (1985) 323-328.
- [14] BARRY, J.F., BUNCE, I.L., PERKIN, J.L., The radiative capture cross-section of ²³⁶U for neutrons in the energy range 0.3-4.0 MeV. Proc. Soc. (1961) <u>78</u> N 503 801.
- [15] STUPEGIA, D.C., HEINRICH, R.R., MACLOUD, I.H., Neutron capture cross-sections of ²³⁶U. J. Nucl. Energy (1961) parts A/B <u>15</u> 200.
- [16] CARLSON, A.D., FRIESENHAHN, J.F., LOPEZ, W.M., FRICKE, M.P., The ²³⁶U neutron capture cross-section. Nucl. Phys. (1970) <u>A141</u> 577-594.
- BERGMAN, A.A., MEDVEDEV, A.N., SAMSOMOV, A.E., et al., Measurement of 236U neutron radiative capture cross-section in the energy range 0.1-50 keV. Vopr. at. nauki i tekh. Ser. Nuclear Constants, 1(45) (1982) 3-7 (in Russian).
- [18] GRUDZEVICH, O.T., DAVLETSHIN, A.N., TIKHONOV, S.V., et al., ²³⁶U neutron radiative capture cross-sections in the energy range 0.15-1.1 MeV ibid 2(51) (1983) 3-15 (in Russian).
- [19] DAVLETSHIN, A.N., TIPUNKOV, A.O., TIKHONOV, S.V., TOLSTIKOV, V.A., 197Au, 236U and 237Np fast neutron radiative capture cross-sections, At. Ehnergiya, <u>58</u> 3 (1985) 183-188 (in Russian).
- [20] DAVLETSHIN, A.N., TIPUNKOV, A.O., TIKHONOV, S.V., et al., Results of measurements of $\sigma_{n,\gamma}^{236}$ U relative to $\sigma_{n,\gamma}^{197}$ Au in the neutron energy range 0.16-1.15 MeV. In: Neutron Physics: Proc. 6th All-Union Conference on Neutron Physics, Kiev, 2-6 October 1983 <u>2</u>, TSNIIatominform, Moscow (1983) 181-184 (in Russian).
- [21] DAVLETSHIN, A.N., TIPUNKOV, A.O., TIKHONOV, S.V., et al., Results of σ_n, γ^{236} U relative to σ_n, γ^{236} U in the neutron energy range 0.16-1.15 MeV, ibid. 164 (in Russian).
- [22] KAZAKOV, L.E., KONONOV, V.N., POLETAEV, E.D., et al., Measurements of neutron radiative capture cross-sections for ²³⁶U and ¹⁹⁷Au in the energy range 3-420 keV, Vopr. at. nauki i tekh. Ser. Nuclear Constants, 2 (1985) 44 (in Russian).

- [23] LUNEV, V.P., MANTUROV, G.N., TIPUNKOV, A.O., TOLSTIKOV, V.A., Evaluation of ²³⁶U neutron cross-sections in the unresolved resonance region. See [20] Vol. 1, 97 (in Russian).
- [24] BADIKOV, S.A., GAJ, E.V., RABOTNOV, N.S., et al., Evaluation of 236 U capture cross-section in the energy range 1 keV \leq En \leq 1.1 MeV on the basis of the Padé approximation, ibid., 106.
- [25] DAVLETSHIN, A.N., TIPUNKOV, A.O., TIKHONOV, S.V., et al, Measurement results for $\sigma_{n,\gamma}^{237}$ Np relative to $\sigma_{n,\gamma}^{197}$ Au in the neutron energy range 0.16-1.15 MeV, ibid., Vol. 2 189 (in Russian).
- [26] DAVLETSHIN, A.N., TIPUNKOV, S.V., et al., Measurement results for $\sigma_{n,\gamma}^{237}$ Np relative to $\sigma_{n,p}$ in the neutron energy range 0.16-1.15 MeV., ibid., 185 (in Russian).
- [27] TROFIMOV, Yu.N., NEMILOV, Yu.A., 237 Np radiative capture cross-section for E_n = 0.3-1.9 MeV, ibid., 142 (in Russian).
- [28] LINDNER, M., NAGLE, R.I., LANDRUM, J.H., Neutron capture cross-sections from 0.1 to 3 MeV by activation measurements, ibid., (1976) <u>59</u> 381.
- [29] STUPEGIA, D.C., SMITH, M., KEEDY, C.R., The capture cross-section of ²³⁷Np. Nucl. Sci. and Engng (1967) <u>29</u> N 2 218.
- [30] WESTON, L.W., TODD, I.H., Neutron capture cross-section of neptunium-237. Nucl. Sci. and Engng (1981) <u>79</u> 184.
- [31] HOFFMAN, M.M., et al., Bull. Amer. Phys. Soc. (1971) <u>16</u> 1181, see also [36].
- [32] MANN, F.M., SCHENTER, R.E., et al., ENDF/B-V data file for ²³⁷Np (MAT 1337). BNL (1979).
- [33] EBERLE, S.H., et al., Project Actiniden. Rep. KFK-1456 Karlsruhe (1971).
- [34] YUROVA, L.I., POLYAKOV, A.A., RUKHLO, V.P., et al, Experimental determination of radiative capture thermal cross-sections and resonance integrals for ²³⁰Th, ²³¹,²³²,²³³Pa, ²³⁶U, ²³⁷Np, In: Neutron Physics: Proc. 6th All-Union Conference on Neutron Physics, Kiev, 2-6 October 1983, Vol. 2, TsNIIatominform, Moscow (1984) 177 (in Russian).
- [35] DERRIEN, H., DEAT, J.P., FORT, E., LAFORD, D., Evaluation of ²³⁷Np neutron cross-sections in the energy range from 10⁻⁵ eV to 14 MeV: Rep. INDC(FR)-42/L. Cadarache, September 1980.
- [36] DERRIEN, H., FORT, E., Proc. Intern. conf. naeutron cross-section and technology, October 22-26 1979. Knoxville (1980) 872.

CONSISTENT EVALUATION OF NEUTRON CROSS SECTIONS FOR THE $^{242-244}$ cm isotopes A.V. Ignatyuk and V.M. Maslov

It is necessary to know the neutron cross-sections for curium isotopes in order to solve the problems of the external fuel cycle. Experimental information on the cross-sections is very meagre and does not satisfy requirements. Moreover, existing evaluations of neutron cross-sections in the ENDF, JENDL and INDL libraries differ substantially, and these differences are especially large for the fission and (n,2n) reaction cross-sections. This situation requires a critical review of the entire set of evaluations of the neutron cross-sections for curium.

In the energy region up to the threshold of the (n,n'f) reaction the differences between the evaluations are due principally to the normalization of the calculated cross-sections to the various experimental data on the neutron fission cross-section [1-7] or on fissility in reactions with charged particles [8]. Above the threshold the discrepancies in the evaluations are due mainly to differences in the determinations of the contributions of emissive fission. The details of the theoretical models on which these evaluations are based demonstrate the need to analyse existing experimental data on the basis of a stricter theoretical approach which takes into account the whole range of contemporary concepts about the optical-statistical characteristics of deformed heavy nuclei. Existing phenomenological systematics of observed fissilities of actinides [9] and the semi-empirical descriptions of the multiple neutron emission cross-sections based on them [10] can but partly satisfy the practical requirements of neutron data evaluation. The justification for the approximations which go into such systematics also requires more consistent theoretical calculations of cross-sections.

In order to calculate the neutron cross-sections, we used the statistical approach, which takes into account the pre-equilibrium emission of

neutrons in the initial stages of compound nucleus formation and the maintenance of the angular moment at all stages of decay of the compound nucleus. The parameters of the pre-equilibrium neutron evaporation model were obtained on the basis of the consistent description of neutron spectra, the (n,2n) and (n,3n) reaction and also the neutron fission cross-sections for 238 U [11], for which the fullest experimental information is available for all the data considered. It has been shown that such an approach also ensures a good description of the neutron cross-sections for the neighbouring odd 235 U isotope [11].

The level density in the neutron and fission channels was calculated with the use of the phenomenological model, which consistently takes into account shell, superfluid and collective effects [12]. The model parameters for the neutron channel were determined from the systematics obtained by combined analysis of the neutron resonance density and the cumulative sums of low-lying levels [13]. It has been shown that for all actinides the density of the low-lying levels can be described satisfactorily by the constant temperature model with parameter T = 0.388 MeV common to all actinides and odd-even differences determined by the value of the correlation function $\Delta_{o} = 12/\sqrt{A}$ MeV. The selection of the level density parameters for the fission channel is considered in detail in Ref. [14], where, using neutron reactions as an example, we demonstrated the need for taking into account the collective effects associated with the non-axial deformation of fissile nuclei on the internal barrier and the mirror deformation on the external barrier. In that case, the correlation functions of the fission channel Δ_{0} + 0.08 will be systematically higher than the similar values in the neutron channel, while the shell corrections needed to describe the observed fission cross-sections at the internal hump $\delta \epsilon_{A} = 2.5$ MeV and at the external hump $\delta \epsilon_{\rm B} = 0.6$ MeV will remain practically unchanged for the whole uranium and plutonium isotope chain [14]. Such an evaluation of shell corrections is in good agreement with the phenomenological systematics of the

two-humped fission barrier parameters for actinides, and consequently the parameters in question can also be used in the calculations of the neutron fission cross-sections for curium isotopes.

The analysis of the neutron cross-sections in curium isotopes is complicated by the fact that the experimental data obtained for nuclear-explosion neutrons [2, 4, 5] exhibit poor agreement with each other both in absolute value and in the energy dependence of the cross-sections. They also fit poorly into the existing systematics of the isotopic dependence of fission cross-sections at the first plateau [15, 16]. In these circumstances. as reference cross-sections the authors used, for the first plateau, the evaluations of the 3-MeV neutron fission cross-sections for curium obtained within the framework of the consistent systematics of fissilities of actinides in neutron reactions and in reactions with charged particles [9]. Such reference cross-sections make it possible to determine the parameters of the fission channel for the whole curium isotope chain, and further calculations of the energy dependence of the fission cross-sections and also the (n,2n) reaction cross-sections are no longer associated with any variations in the parameters. To calculate the cross-sections for compound nucleus formation and the corresponding neutron transmission coefficients, we used the non-spherical optical model with the potential parameters recommended in Ref. [17].

The results of theoretical calculation of the fission cross-sections, together with available experimental data [1-7], are shown in Fig. 1. For comparison we have also given the results of the various evaluations. From the data presented it will be seen that there is a considerable difference between evaluations both in absolute value and in the description of the energy dependence of the fission cross-sections. Substantial differences exist also between the experimental data (see Fig. 1,c) so that the fission cross-section evaluations based on the systematics of fissility of nuclei by charged particles are preferable [9]. It should be noted that, although we







used such systematics only for the neutron energy of 3 MeV, no substantial differences occur between the theoretical calculations and the systematics of the fission cross-sections [9] for the second and the third plateau region (see Fig. 1,a-c). We can therefore regard the theoretical calculations and the phenomenological systematics of the neutron fission cross-sections for the nuclei [9] as sufficiently consistent for the whole region of neutron energies above 3 MeV.

Figure 2 shows the results of the present calculations and the set of available evaluations of the (n,2n) reaction cross-sections. For this reaction there are no experimental data, and the differences between the various evaluations are a direct consequence of the models on which they are based. Since in all evaluations the sum of cross-sections is normalized practically always to the cross-section for compound nucleus formation, a considerable part of the differences in the evaluations of the (n, 2n)reaction cross-sections is due directly to discrepancies between the evaluations of the fission cross-sections and, in the neutron energy region above 14 MeV, also to discrepancies between the evaluations of (n,3n) reaction cross-sections. The non-physical nature of the energy dependence of the (n,2n) reaction cross-sections in the evaluations of the ENDL and ENDF libraries is obvious. At the same time, the evaluations of Ref. [10], which used a model conceptually close to our approach, have a cross-section energy dependence similar to our calculations, and the differences in the absolute value of the cross-sections are due to the error of the evaluations of the fissility of nuclei and to the more approximate modelling of the cross-sections for compound nucleus formation.

Because of the above differences in the evaluations of the (n,2n) reaction cross-sections, the evaluations of this reaction for uranium and plutonium isotopes deserve attention. Figure 3 shows available evaluations of the (n,2n) reaction cross-sections in the region of their maximum value, i.e. for neutron energies of 10-12 MeV. We have given the results of our





calculations of the cross-sections for 246 Cm and 248 Cm, together with those for the light isotopes of curium. The analysis of the isotopic dependence of the cross-sections for compound nucleus formation [17] shows only comparatively small variations in these cross-sections for the incident neutron energies considered. Hence it can be concluded that changes in the (n,2n) reaction cross-sections at the maximum are determined almost entirely by changes in the fissility of nuclei. Since for all uranium, plutonium and curium isotopes the fissility decreases monotonically the heavier the isotopes are, the (n,2n) reaction cross-sections should exhibit the inverse dependence - a monotonic increase in the cross-sections. This result is, of course, confirmed by the experimental data available for 235 U and 238 U [11]. In the consistent theoretical calculations a monotonic isotopic increase in the (n,2n) reaction cross-sections is observed for all actinides (see Fig. 3), but the ENDF/B-V and JENDL-II evaluations do not show this trend, indicating inconsistency between the evaluations for the different isotopes.

A similar pattern of isotopic changes appears also in the (n,3n) reaction cross-sections. However, these need not be discussed in the present work since for the curium isotopes considered the (n,3n) reaction cross-section is very small (not greater than 20 mb for 244 Cm).

The results of theoretical calculations of the main neutron reaction cross-sections for curium isotopes, together with the results of the phenomenological systematics of the fission cross-sections [9] based on an analysis of the fissility of transactinides in reactions with charged particles, indicate the unsatisfactory nature of the majority of the evaluations included in the INDL/A files. Existing evaluations cannot be recommended to neutron data users for one single isotope. A considerable amount of work must be carried out on the revision of the cross-section evaluations in the neutron energy region above 1 MeV. It is evident that the necessary degree of reliability of such evaluations can be ensured only by the use of coherent theoretical models which are consistent with the whole set of



Fig. 3. Isotopic dependence of cross-sections $\sigma_{n,2n}^{\lambda}$ at the maximum according to the data of: Present work; $x = ENDP/B-V; \Box = JENDL-II; O = (10)$.

concepts relating to neutron reaction mechanisms and with the statistical description of the properties of the competing decay channels of fissile nuclei. We hope that the results of the present work will provide the necessary basis for the practical re-evaluation of the neutron cross-section files for $^{242-244}$ Cm.

REFERENCES

- [1] KOONTZ, P., BARTON, D., in: Neutron Cross-Sections and Technology <u>1</u> (1968) 597.
- [2] MOORE, M.S., KEYWORTH, G.A., Phys. Rev. C<u>3</u> (1971) 1656.
- [3] FOMUSHKIN, Eh.F., et al., Yad. Fiz. <u>17</u> (1973) 24.
- [4] SILBERT, M.G., Rep. LA-6239 (1976).
- [5] FOMUSHKIN, Eh.F., et al., Yad. Fiz. <u>31</u> (1980) 39.
- [6] WHITE, R.M., BROWNE, J.C., in: Nuclear Data for Science and Technology (BROCKHOFF, K.N., Ed.), Reidel Publ. Conf. (1982) 218.
- [7] VOROTNIKOV, P.G., et al., Yad. Fiz. <u>40</u> (1984) 1141; At. Ehnerg. <u>57</u> (1984) 61.

- [8] BRITT, H.C., WILHELMY, J.B., Nucl. Sci. and Engng <u>72</u> (1979) 222.
- [9] KUPRIYANOV, V.M., FURSOV, B.I., SMIRENKIN, G.N., Yad. Fiz. <u>39</u> (1984) 281.
- [10] BYCHKOV, V.M., PLYASKIN, V.I., Yad. Fiz. 34 (1981) 684.
- [11] GRUDZEVICH, O.T., IGNATYUK, A.V., MASLOV, V.M., PASHCHENKO, A.B., in: Neutron Physics: Proceedings of the Sixth All-Union Conference on Neutron Physics, Kiev, 2-6 October 1983, <u>2</u>, TsNIIatominform, Moscow (1984) 318 (in Russian).
- [12] IGNATYUK, A.V., ISTEKOV, K.K., SMIRENKIN, G.N., Yad. Fiz. 29 (1975) 875.
- [13] ANTSIPOV, G.V., KON'SHIN, V.A., MASLOV, V.M., in: Proc. of Consultants Meeting on Uranium and Plutonium Isotope Resonance Parameters, INDC-129/GJ, Vienna (1981) 329.
- [14] IGNATYUK, A.V., KLEPATSKIJ, A.B., MASLOV, V.M., SUKHOVITSKIJ, E.Sh., Yad. Fiz. <u>42</u> (1985) 569.
- [15] BEHRENS, J.W., HOWERTON, R.J., Nucl. Sci. and Engng 65 (1978) 464.
- [16] SMIRENKIN, G.N., FURSOV, B.I., in: Vopr. At. Nanki i Tekh., Ser. Yad. Konstanty No. 2 (1985) 31 (in Russian).
- [17] LAGRANGE, Ch., Rep. INDC(FR)-56/L (1982).

,

RE-EVALUATION OF THE NEUTRON CROSS-SECTIONS FOR IRON

V.G. Pronyaev and A.V. Ignatyuk

In spite of the abundance of evaluated neutron cross-section files for iron and its isotopes, there is a need to continue the work of evaluating the cross-sections of this very important structural material for two reasons: first, the results have been published both of new measurements which have significantly extended the resolved resonance region [1] and of measurements of the capture area under the individual resonances [2-5]. Secondly, none of the available evaluations offers good-quality data for the calculation of systems with different spectra (for example, fast reactors and fusion reactors).

Evaluation of cross-sections in the resonance energy region

The main problems that arise during the evaluation of cross-sections in the region of resolved and unresolved resonances and the methods used to solve them in the present evaluation can be described in the following manner. The total cross-section structure, which determines their self-1. shielding and which was obtained in experiments with a high resolution, should be retained in the evaluated data file. This is required because the upper boundary of the resolved s-resonance region for $\frac{56}{r}$ Fe (E_r^{max} = 850 keV) and ⁵⁴ Fe (E_r^{max} = 500 keV) was taken to be appreciably higher than for the p- and d-waves (350 and 200 keV, respectively, for ⁵⁶ Fe and ⁵⁴ Fe). Since the ENDF-V format does not formally allow for one isotope, the inclusion of different boundaries of resolved resonance regions with different spin characteristics, we used the so-called pseudo-isotope method for cases where the resonance regions for different orbital waves were considered to belong to different pseudo-isotopes with an isotopic content equal to unity. This did not require any change in the processing programs.

2. The cross-sections in a sufficiently extended region of resolved reasonances (850 keV for s-neutrons in 56 Fe) were described with a single

potential scattering radius and without using any background cross-sections except in cases where their presence was due to the requirements of data presentation in the ENDF-V format (for example, the contribution of the inelastic scattering cross-section to the total cross-section for ⁵⁷Fe, which has a low inelastic scattering threshold). It was found that this could be achieved by introducing some number of distant resonances and using the Reich-Moore formalism to calculate the cross-sections from the resonance parameters. In order to illustrate this, we show in Fig. 1 the total crosssection for ⁵⁶ Fe obtained from the resonance parameters of the present evaluation within the framework of the Reich-Moore formalism and the multilevel Breit-Wigner formula. As will be seen, there are marked differences even in the cross-section averaged over sufficiently wide groups. The differences in the forms of individual resonances and in the minima of cross-sections are still greater. As a rule, the Reich-Moore formalism yields a description which is a good approximation of the detailed behaviour of the total cross-section. Moreover, the resonance parameters for the nuclei of structural materials are themselves obtained from the transmission data mainly using the Reich-Moore formalism.

3. The average resonance parameters for calculations of cross-sections in the unresolved resonance region were evaluated first by averaging the parameters in the resolved resonance region and then were corrected by the EVPAR program [6] with allowance for the possible differences in level density with respect to parity. The contribution of the f-wave to the capture cross-section is appreciable in the unresolved resonance region for the individual isotopes 56,58 Fe and it was taken into account effectively by somewhat overstating the average radiative capture widths for the d-wave. The value of this contribution was chosen in such a way as to describe existing experimental data on the average cross-sections in this region.

4. The cross-sections for natural iron in the resonance energy region are obtained in a consistent manner by summing the contributions from the





individual isotopes without introducing any additional background cross-section components.

The selection of parameters for the individual isotopes and the evaluation procedure for the average resonance parameters are described in greater detail in Ref. [7].

Comparison of the results of various cross-section evaluations in the resonance region

The neutron total and capture cross-sections in the resonance energy region for a natural mixture of iron isotopes, which are taken from various evaluations, are compared in Fig. 2. For ease of comparison, the cross-sections were averaged over 74 energy groups. Except in the 1-6 keV region and in some groups above 100 kev, the total cross-sections agreed with each other to within 5-10%. The background in the total cross-section was 10-30% of the value of the whole cross-section in the region up to 100 keV for the JENDL-II evaluation and was small for the evaluation from the ENDF/B-IV library, while there was no background in the evaluation of the present paper. The upper boundary of the resonance region given in these files differs noticeably. It is 60 keV for the file from the ENDF/B-IV library, 250 keV for the JENDL-II file and 850 keV for the file from the present evaluation.

The differences in the total cross-section values for some groups with energies above 100 keV are due to the fact that for the calculation of cross-sections the JENDL-II library used the multi-level Breit-Wigner formula with the addition of a smooth background selected on the basis of available experimental data on the total cross-section for a natural mixture of iron



Fig. 2. Comparison of the results of the different evaluations of the total cross-section (a) and the capture crosssection (b) for natural iron: the continuous curve represents the evaluation of the authors of the present study; the short dashes represent the JENDL-II data and the long dashes the ENDF/B-IV data.

isotopes. In the present evaluation the total cross-section for this mixture was obtained in a consistent manner on the basis of the evaluated cross-sections for the individual isotopes; in this case, during the pointwise calculation of cross-sections for the two major isotopes (^{54,56}Fe) we used the Reich-Moore formalism, whose advantages were discussed above.

The evaluated total cross-sections for iron from the ENDF/B-IV library are based on experimental data obtained up to 1972.

The most noticeable differences in the capture cross-section between the present evaluation and that of JENDL-II are observed in the 6-40 keV region, where the latter evaluation is systematically higher by 20-60%. The reason for this difference can be understood from an analysis of the resonance parameters which make the largest contribution to the capture cross-section in this region. As will be seen from Table 1, the radiation widths of the

Table 1

E _r , keV	Isotope	l	I	Γ _n ,eV	Γ r ,eV	$ag \frac{\Gamma_n \Gamma_r}{\Gamma_t}, eV$
7,8	54 _{Pe}	0	0,5	1160 (1040)	I,74 (2,5)	0,1008 (0,1447)
11,18	54 _{Fe}	I	I,5 (0,5)	3,85 (7,7)	0,4 (2,5)	0,042 (0,110)
14,45	54 _{Fe}	I	0,5	I,4 (I,4)	1,12 (2,5)	0,036 (0,055)
22,82	56 _{Pe}	I	0,5	0,25I (0,27)	0,54 (0,54)	0,1576 (0,1656)
27,74	56 _{Fe}	0	0,5	1474 (1420)	1,06 (1,40)	0,9745 (1,287)

Parameters of resonances determining the capture cross-section for iron in the 6-30keV region for the present evaluation in comparison with the data from the JENDL-II library (in brackets)

Remarks. E_r is the resonance energy, Γ_n and Γ_{χ} are the neutron and radiation widths of resonance, $ag(\Gamma_n \Gamma_{\chi}/\Gamma_t)$ is the capture area with allowance for isotopic content; I is the spin of the nucleus and 1 the orbital moment.

resonances of 54 Fe in the JENDL-II evaluation are ascribed the same average width of 2.5 eV, while the radiation width of the s-resonance of 56 Fe situated at 27.74 keV is 30% higher than that from the present evaluation, which is based on the latest measurement results [2, 5]. The capture cross-section in the evaluated ENDF/B-IV library for energies above 60 keV is determined by experiments with a low resolution performed before 1970.

Evaluation of cross-sections in the fast neutron energy region

When evaluating neutron cross-sections for energies above 850 keV, particular attention was paid to attaining a consistent description of the total elastic cross-section and the level excitation functions for inelastic scattering without introducing any additional physically unjustified renormalizations. Such an approach imposes higher requirements on the selection of parameters, and primarily, the optical potential parameters, which need to provide a satisfactory simultaneous description of the total cross-section, the cross-section for inelastic processes and the elastic



Fig. 3. Comparison of the results of the different evaluations with available experimental data on the total elastic crosssection and the cross-section for the inelastic processes of interaction of neutrons with natural iron: continuous curve - evaluation of the authors of the present study; short dashes - JENDL-II data; long dashes - ENDF/B-IV; dot-dash curve - CJD. Experimental data for the total cross-section obtained with a high resolution and averaged over energy groups with a width of 0.5 or 1 MeV: 0 - [8]; • - [9]; x - [10]; o - [11].

scattering cross-section. The parameters were selected with the help of the ABAREX program, which uses the optical-statistical approach to cross-section calculations. The parameters found enabled us to describe the available experimental data on the total cross-section and the cross-section for inelastic processes in the 2-20 MeV interval with an accuracy of 2-3% (Fig. 3). In regions where the evaluated cross-sections have a resonance structure the results presented are averaged over energy groups (histograms). The experimental data for the inelastic cross-sections are taken from studies included in the EXFOR library.

As regards the description of the elastic scattering cross-section, the following should be noted. In Fig. 3 the light and dark circles indicate, respectively, the elastic scattering cross-sections obtained as the difference between the total cross-section known with 2-4% accuracy and the inelastic cross-section measured with a high accuracy (light circles), and by integration of the angular distribution of elastic scattering (dark circles). In the latter case, the accuracy of determination of the elastic scattering cross-section clearly does not exceed 10% because the cross-section for small angles, which makes the main contribution to the integral cross-section, is known with a relatively low accuracy. As can be seen from the figure, the scattering cross-section determined by integrating the angular distributions lies, on an average, 10% below the same cross-section obtained by subtraction. It should be noted that the experiments to determine the inelastic cross-sections were performed in the mid-1950s and have not been repeated since. Nevertheless, together with the total cross-sections, they have been used as reference cross-sections in the present evaluation.

For comparison, we also give the results of other evaluations in Fig. 3. The greatest difference is observed in the case of the inelastic cross-sections in the 3-14 MeV region, which is attributable to the difference in the absorption cross-section predicted by the optical models with different parameters used in the various studies.

The results of the evaluation of the excitation functions for individual levels and their groups and of the corresponding angular distributions at an initial neutron energy of 5 MeV are compared in Figs 4 and 5 with the latest experimental data and the results of other evaluations. The present evaluation is based on the cross-sections obtained in the optical-statistical approach, with direct processes taken into account by the coupled-channel method. The level excitation function for 0.847 MeV in the energy region from the threshold to 3.3 MeV, which contains marked cross-section fluctuations, was evaluated on the basis of well-known





Excitation functions for individual or groups of levels of natural iron during inelastic neutron scattering. Evaluated data: continuous curve – evaluation of authors of present study; short dashes – JENDL-II data; long dashes – ENDF/B-IV; dot-dash curve – CJD-1 data. Experimental data: $\phi - [9]; \phi - [12];$ + – [13].



Fig. 5.

Angular distributions of inelastic scattering of neutrons with an initial energy of 5 MeV and with excitation of individual levels or groups for natural iron. Continuous curve – evaluation of authors of present study. Experimental data from Ref. [13] for neutrons with initial energy, MeV: $\oint -5; \oint -5.04;$ $\oint -5.44; \oint -5.56.$
experimental data [14, 15]. It should be pointed out that experimental data on inelastic scattering are very limited and often contradictory. For this reason, in their evaluation an appreciably greater role is played by consistent model approaches based on a consistent description of a wide range of reactions.

In evaluating the secondary neutron spectra of the (n,2n) and (n,n') reactions the following assumptions were used:

- The contribution of the direct processes to the continuous level spectrum region has a plateau-like shape, and the contribution of the compound processes corresponds to the evaporation model with the dependence of the nuclear temperature on the initial neutron energy as in the Fermi-gas model;
- The competition of gamma photons with neutrons at the second cascade can be neglected. Then, having the evaluated integral cross-sections for the (n,2n) and (n,n') reactions, we can derive the contribution of each reaction mechanism to the continuous primary neutron emission spectrum. The results of this simplified approach to the evaluation of spectra are shown in Fig. 6.

The most appreciable differences are observed in the evaluation of the radiative capture cross-section (Fig. 7). In the present evaluation we used the calculations of the capture cross-section in the optical-statistical approach, taking into account the contribution of direct and semi-direct processes. The structure in the capture cross-section at an energy below 4 MeV is due to the competition of the inelastic scattering channels. Need for new measurements

Because of the problems which arise during the evaluation of the neutron cross-sections for isotopes of iron and its natural mixture, it is desirable to carry out the following measurements:

- The total cross-section for ⁵⁶Fe and its natural mixture in the 1-6 keV region;





$$\oint - \left[16\right]; \quad \oint - \frac{d^2 \overline{\sigma}(90^\circ)}{dE'_n d\Omega} 4\pi \text{ from } [17]; \quad \sim - [18]; \quad \oint - [19]; \quad + - \frac{d^2 \overline{\sigma}(60^\circ)}{dE'_n d\Omega} 4\pi \text{ from } [20]; \\ \mathcal{N} - \frac{d^2 \overline{\sigma}(45^\circ)}{dE'_n d\Omega} 4\pi \text{ from } [21].$$

- The sum of the cross-sections for inelastic processes in the interval above 3 MeV;
- The capture cross-section for energies above 1 MeV;
- The (n,pn) and (n, an) cross sections, for which there are practically no experimental data.



Fig. 7. Comparison of the results of the different evaluations for the neutron radiative capture cross-section in natural iron: continuous curve – evaluation of the authors of the present study; short dashes – JENDL-II data; long dashes – ENDF/B-IV; dot-dash curve – CJD-1 data.

In addition, it is necessary to obtain non-contradictory data on neutron inelastic scattering with excitation of individual discrete levels and their groups. These measurements will markedly improve the reliability of evaluated data for the cross-sections and energies considered.

The comparison made between the evaluated cross-sections from different libraries and the results of the present evaluation shows that the latter has the following advantages:

- It retains the resonance structure of the cross-sections up to
 850 keV;
- The capture cross-section in some groups has changed appreciably, owing to the appearance of new measurements;
- In the resonance region of energies the files do not have any background in the cross-sections, except where this is due to format requirements;
- The evaluation of most cross-sections is based on the use of physical models with parameters chosen so as to describe numerous experimental data. There is no physically unjustified renormalization of cross-sections based only on the desire to improve the description of experimental data on individual cross-sections;

- The file for natural iron was obtained from the files for
 - individual isotopes, allowance being made for their content in the natural mixture.

The evaluation made in this study can serve as the basis for preparing constants for both fast and fusion reactor calculations.

REFERENCES

- 1. Cornelis B.N., Mewissen L., Poortmans F. In: Nucl. data for sci. and technol.: Proc. of the Intern. conf. (Antwerp, 1982). Hollund, 1983, p.135.
- 2. Brusegan A., Corvi P., Rohr G. e.a. Ibid., p.127.
- 3. Corvi F., Brusegan A., Buyl R. e.a. Ibid., p. 131.
- 4. Rohr G., Brusegan A., Corvi F. e.a. Ibid., p.139.
- 5. Wisshek K., Kappler F. In: Proc. of the IAEA consultants meeting on nuclear data for structural materials; INDC(NDS)-152/L, 1984, p.46.
- Manturov G.N., Lunev V.P., Gorbacheva L.V., Problems of Atomic Science and Technology, Ser. Nuclear Constants No. 1 (50) (1983) 50 (in Russian).
- 7. Pronyaev V.G., Ignatyuk A.V., ibid. 3 (1986) 13.
- 8. Carlson A.D., Cerbone R.J. Nucl. Soi. and Engng, 1970, v.42, p.28.
- 9. Perey F.G.J., Love T.A., Kinney W.E. Rep. ORNL-4823, 1972.
- 10. Cierjacks S., Forti F., Kopsch D. e.a. Rep. KPK-1000, 1968.
- 11. Schwartz R.B., Schrack R.A., Heaton H.T. Hep. NBS-MONO-138, 1974.
- 12. Littler A., Nardini J., Couchell G.P. Rep. EXFOR-10519, 1975.
- 13. Kinney W.E., Perey F.G.J. Rep. ORNL-4515, 1970.
- 14. Kinney W.E., Perey P.G.J. Nucl. Sci. and Engng, 1977, v.64, p.418.
- 15. Smith A., Guenter P. Ibid., 1980, v.73, p.186.
- 16. Towle J.H., Owens R.O. Nucl. Phys., 1967, v.A100, p.257.
- 17. Biryukov N.S., Zhuravlev B.V., Kornilov N.V., et al., Yad. Fiz. 19 (1974) 1190.
- 18. Vonach H., Chalupka A., Wenniger F., Staffel G. Rep. ZFK-382, 1979, p.159.
- 19. Hermadorf D., Meister A., Sassonoff S. c.e. Rep. 2PK-277(U), 1975.
- 20. Sal'nikov O.A., Lovchikova G.N., Kotelnikova G.V. et al., Nuclear Constants 7 (1973) 102 (in Russian).
- 21. Takachashi A., Fukusawa M., Yanagi Y., Yamamoto J., OKTAVIAN: Rep. A-83-08 (1984).

EVALUATION OF CROSS-SECTIONS FOR THE FISSION OF CURIUM ISOTOPES BY FAST NEUTRONS

B.I. Fursov, G.A. Kudyaev, G.N. Smirenkin

Experimental measurements of fission cross-sections for curium isotopes tend to be inaccurate, with wide discrepancies between data from different authors. This is mainly due to measurement problems arising from the high probability of α -decay and spontaneous fission of curium isotopes. The unsatisfactory state of the experimental data and difficulties with the theoretical description of the dependence $\sigma_f(E_n)$ lead to the wide discrepancies in fission cross-section evaluations. In the author's opinion, no evaluation of the fission cross-sections in the fast neutron range can currently be recommended for use in the neutron energy range 0.1-20 MeV. Only new and more accurate experimental data can form a basis for improved evaluations. At present such data, as given in Ref. [1], are available only for ²⁴⁵Cm. The quality of these results with regard to accuracy, detail and width of the energy range studied is such that the data in Ref. [1] supersede the evaluation of the 245 Cm fission cross-section for fast neutrons. However, for other curium isotopes too a more realistic evaluation can be obtained from existing experimental data, and that was the purpose of this work.

The special features of this approach to evaluating the fission cross-sections of curium isotopes are as follows:

- A broad range of experimental data are considered, not only those obtained for (n,f) reactions, but also those for the direct reactions (³He,df), (³He,tf), (t,pf), (d,pf) and (p,p'f) from Britt et al;
- A uniform approach is adopted for the selection and co-ordination of experimental results;
- The evaluation is based on the systematics for the of cross-sections and other characteristics of the fission of transuranium nuclei developed in Ref. [2].

Evaluation method

It is impossible to calculate the fission cross-sections in the near-threshold region of neutron energies with acceptable accuracy owing to incomplete knowledge of the fission channel spectrum, particularly for even-even fissile nuclei. However, it is precisely at low energies (no more than 1 MeV) that experimental data on fission cross-sections and fissilities for curium isotopes have been studied in most detail. Far less data are available for higher neutron energies, even though conditions are more suitable for application of the theory. With increasing excitation energy the number of accessible fission channels grows exponentially, individual features of nuclei are obliterated, and, with the above difficulty resolved, a statistical description becomes appropriate.

Theoretical and phenomenological analyses of fission probability show that in the region above the threshold ($E_n \approx 2-5$ MeV) the average fissility of heavy nuclei < P_f > is weakly dependent on the energy. As a result, there are so-called "plateau" regions in the energy dependence $P_f(E)$. The author's evaluation exploits this fact: at neutron energies of 2 MeV or more it is assumed that $P_f(E_n) = P_f^0 = \text{const for all curium}$ isotopes.

The dependence of the value of P_f^o on the nucleon composition of the nuclei is investigated thoroughly in Refs [2, 3]; it is shown that for transuranium nuclei (uranium - einsteinium) the set of values of P_f^o can be described with a root-mean-square error of approximately 15% by a simple physical model which the authors will refer to below as the P_f^o systematics. By using results from the systematics for evaluating the fission cross-section of curium isotopes as reference values in the plateau region $(E_n = 2-5 \text{ MeV})$, it is possible to avoid serious errors in analysing inconsistent experimental data.

In order to achieve a consistent treatment and description of data on the neutron fission cross-section $\sigma_f(E_n)$ and on the fissility of nuclei

in direct reactions $P_f(E)$, information is required on the neutron cross-section for compound nucleus formation, which makes it possible to compare data on the values of P_f and σ_f : $P_f(E) = \sigma_f(E_n)/\sigma_c(E_n)$, if $E = E_n + B_n$, and conversely, $\sigma_f(E_n) = \sigma_c(E_n)P_f(E_n + B_n)$. In this paper we have used calculation results for $\sigma_c(E_n)$ based on the coupled channel model with parameters for ²⁴⁶Cm from the data in Ref. [4]. For other target nuclei with mass number A, we assumed $\sigma_c^A(E_n) = \sigma_c^{246}(E_n)(A/246)^{2/3}$. In the fission of a compound nucleus with A in the neutron energy range above 6 MeV, the (n,n'f) reaction is included, and above 12 MeV the (n,2nf) reaction, in which nuclei with A - 1 and A - 2, respectively, undergo fission. If data on the fissility $P_f(E)$ in the excitation energy range from the fission threshold to the plateau are known for the isotope chain A, A - 1, A - 2, the total fission cross-section in the energy range above the thresholds for the (n,nf) and (n,2nf) reactions, i.e. for neutron energies above 5 MeV, can be calculated.

For the spectrum of emitted neutrons $N(\epsilon) = \epsilon/T^2 \exp(-\epsilon/T)$ a value $T_1 = T_2 = T = 0.5$ MeV, which is similar to the data in Ref. [5], was adopted.

The evaluation of the fission cross-section for each curium isotope was carried out in the following manner:

1. The set of experimental data on the fission probability $P_f(E)$ obtained from neutron experiments and in direct reactions was analysed for the chain of fissile curium nuclei with A = 241-249 over the entire energy range studied. Individual experimental

results were rejected as the shape of the energy dependence was inconsistent with other more reliable data or with physical conceptions based on a comparison with other nuclei of the same parity;

- 2. The selected experimental values of $P_f(E)$ were normalized over the 2-4 MeV neutron energy range to the value of P_f^0 from the systematics in Ref. [2]. As a rule, the displacements of data due to the normalization did not exceed the experimental error;
- 3. The set of normalized experimental values was approximated by rational functions (PADE-2 program). As a result of this approximation, a set of functions $P_f(E)$ from the fission threshold to the plateau region ($E_n = 2-5$ MeV) was obtained for all compound nuclei of curium with mass numbers A = 241-249. At lower energies the value of $P_f(E)$ was based on the smoothed experimental dependence, while for higher energies (up to 20 MeV) this value was taken as $P_f^0 = \text{const [2]}$;
- 4. On the basis of the relationships in Ref. [1], the dependence $\sigma_f(E_n)$ was calculated over the neutron energy range 0.1-20 MeV for curium target nuclei with A = 242-248.

Discussion of results

 $\frac{242}{\text{Cm}}$ (Fig. 1(a)). The data from Ref. [6] are multiplied by 1.23 for consistency with the systematics in Ref. [2]. In the fission threshold region they show satisfactory correspondence with the neutron data in Ref. [7] (measured with a statistical error of 15-20%), which were energy-averaged and increased by 9%. The present evaluation is higher than the others shown in Fig. 1(a) in the region below the threshold for the (n,nf) reaction; in the neutron energy range above 7 MeV it has the lowest values. The value $\sigma_{\rm f} = 2.86 \pm 0.30$ b for a neutron energy of 14.5 MeV from Ref. [8] was reduced by 5.5% owing to the change in the standard $\sigma_{\rm f} (\frac{235}{U})$. All the same, it exceeds the value of the cross-section for compound nucleus formation $\sigma_c = 2.68$ b [4] and may still be overestimated.

The fission cross-sections in the region above the thresholds for the (n,nf) and (n,2nf) reactions were calculated using the data in Ref. [6] for the compound nuclei 242 Cm and 241 Cm (coefficients for normalization to systematics 1.09 and 1.08, respectively).

 $\frac{243}{Cm}$ (Fig. 1(b)). The energy-averaged data from Ref. [9] and the results from Ref. [6] show satisfactory agreement when made consistent with the systematics by multiplying by 0.95 and 1.07, respectively. The data in Ref. [16] differ sharply in the shape of the energy dependence from the data in Refs. [6, 9], and for this reason have been rejected. The ENDF/B-V evaluation based on these results is probably incorrect for the neutron energy range below 8 MeV.

 $\frac{244}{\text{Cm}} \text{ (Fig. 1(c))}. \text{ The evaluation is based on neutron data from}$ Refs. [10-12] with coefficients for normalization to the systematics [2] of 1.02, 1.11 and 1.06, respectively. In the fission threshold region, the data from Ref. [11] are energy-displaced by -100 keV for better agreement with Ref. [10] in the threshold region. For a neutron energy of 14.5 MeV, the value $\sigma_{\rm f} = 3.1 \pm 0.3$ b [8] is reduced to up-to-date values of the halflives and standard. Nevertheless, this modified value $\sigma_{\rm f} = 2.93 \pm 0.30$ b, as in the case of 242Cm, appears to be overestimated ($\sigma_{\rm c} = 2.69$ b [4]).

 $\frac{245}{\text{Cm}}$ (Fig. 1(d)). The data from Ref. [1] give an evaluation of the fission cross-section for 245 Cm. The calculated curve does not increase the accuracy and reliability of these experimental data, which were measured with an error of 3-5%. Unfortunately, we do not have the numerical values from Ref. [1], apart from one point for a neutron energy of 14 MeV. Figure 1(d) shows data taken from a graph in Ref. [1], and also experimental results from Ref. [10], which are energy-averaged and reduced by 10%. The results from [1] and [10] then agree well with each other and with the systematics [2] for neutron energies of 3 MeV. It is clear from the diagram that all existing



1 0,1

•,•

1

5

10

(d)

En, MeV

15

 $\frac{242}{C_{E}} (\epsilon), \frac{243}{C_{E}} (\epsilon), \frac{244}{C_{E}} (\epsilon), \frac{245}{C_{E}} (d), \frac{246}{C_{E}} (e), \frac{247}{C_{E}} (f), \frac{246}{C_{E}} (g). Baluation: _____ - present work; _____ - ___ IEDL/4$ $(Italy); _____ - SIEDL-11(Japan); _____ EDLI (FRG, Israel); ______ - ___$ $IEDF/F-Y (USA). Experimental points from Refs: <math>\mathfrak{g} - /2/$ (value of systematics); $\Delta_{-} / c/7; \ \Box - /7/; \ \nabla - /6/; \ O - /2/; \ O - /10/; \ \Delta - /6, 11/; \ \nabla - /12/; \ A - /12; \ 4 /13, 147; \ \mathfrak{e} - /15/$





En, MeV



evaluations of the fission cross-section for 245 Cm in the range 0.1-20 MeV must be rejected.

 $\frac{246}{\text{Cm}}$ (Fig. 1(e)). The data from Ref. [11] agree with the systematics [2] in the plateau region and are used in the evaluation without renormalization. The energy is reduced by -100 keV, as in the case of 244 Cm. The results from Ref. [10] are energy-averaged and reduced by 7%, which increased their agreement with the data from Ref. [11]. The present evaluation occupies an intermediate place over the entire energy range in comparison with the others.

 $\frac{247}{Cm}$ (Fig. 1(f)). The results from Ref. [10] were rejected as they differ by a factor of about 2 from the systematics [2] and contradict the data in Refs. [13, 14] on the energy dependence of P_f(E). The present evaluation is based on the results of Refs [13, 14] for energies below 2 MeV and on calculated data following the systematics [2] for higher energies. Our evaluation is very different from the JENDL-11 and ENDF/B-V evaluations for the entire neutron energy range below 10 MeV.

 $\frac{248}{Cm}$ (Fig. 1(g)). The evaluation is based on results from Ref. [15] (normalization coefficient 1.12), as well as data from Ref. [10], which are reduced by 4%. It is possible that this renormalization slightly distorted the energy dependence in the neutron energy range 1-2 MeV.

The algorithm used in the present evaluation to describe the experimental data, and to predict them in those neutron energy regions for which no data are available, was the same for all the isotopes considered, and physically consistent. The authors believe that such an approach makes for progress in evaluating cross-sections for fission of curium isotopes by fast neutrons despite the significant renormalization of contradictory data required in the first plateau region and the simplification of the model to describe the fission cross-section at high energies.

The error in the recommended curves in the energy range below 6 MeV is evaluated at approximately 10-15% (20% for 242 Cm, 247 Cm). In the range above 6 MeV, the error increases by a factor of about 1.5.

New and more reliable measurement methods and successful theoretical analysis of the experimental information is required for further study of $\sigma_f(E_n)$.

REFERENCES

- WHITE, R.M., BROWNE, J.C., in: Nucl. data for sci. and technol.: Proc. Intern. of conf. (Antwerp, 1983) (1983) 218.
- [2] KUPRIYANOV, V.M., SMIRENKIN, G.N., FURSOV, B.I., Yad. Fiz. <u>39</u> 2 (1984) 281.
- [3] KUPRIYANOV, V.M., ISTEKOV, K.K., FURSOV, B.I., SMIRENKIN, G.N., ibid., <u>32</u> 2 (1980) 355.
- [4] LAGRANGE, Ch., Rep. NEANDC(E) "L" (1982) 228.
- [5] KNITTER, H.H., BUDTZ-JURGENSEN, C., In: [1] 744.
- [6] BRITT, H.C., WILHELMY, J.B., Preprint LA-UR-79-614 (1979).
- [7] VOROTNIKOV, P.E., DMITRIEV, S.V., MOLCHANOV, Yu.D., et al., Yad. Fiz. 40 5 (1984) 1141.
- [8] FOMUSHKIN, Eh.F., GUTNIKOVA, E.K., ZAMYATNIN, Yu.S., et al., ibid., <u>5</u> 5 (1967) 966.
- [9] SILBERT, M.C., Rep. LA-6239-MS (1976).
- [10] MOORE, M.S., MEYWORTH, G.A., Phys. Rev. C, <u>3</u> 4 (1971) 1656.
- [11] FOMUSHKIN, Eh.F., NOVOSELOV, G.F., VINOGRADOV, Yu.I., et al., Yad. Fiz., <u>31</u> 1 (1980) 39.
- [12] KOONTZ, P.G., BARTON, D.R. Proc. conf. neutron cross-sections and technology, Washington, D.C., Vol. 1 (1968) 597.
- [13] BRITT, H.C., GAVRON, A., GOLDSTONE, P.D., WILHELMY, J.B., Phys. Rev. Letters, Vol. 38 (1977) 1457.
- [14] BACK, B.B., HANSEN, Ole, BRITT, H.C., GARRETT, J.D., Proc. Symp. of phys. and chem. of fission. Vienna, IAEA, Vol. 1 (1974) 25.
- [15] FOMUSHKIN, Eh.F., NOVOSELOV, G.F., VINOGRADOV, Yu.I., et al., Yad. Fiz. <u>36</u> 1 (1982) 582.
- [16] FULLWOOD, R.R., DIXON, D.R., LOUGHED, R.W., Rep. LA-4220 (1970) 157.