TESTING CAPTURE CROSS-SECTIONS OF FISSION PRODUCTS IN REACTIVITY PERTURBATION EXPERIMENTS

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ABSTRACT

Central reactivity worths for twelve fission product nuclides ($^{95,97,98,100}Mo$, ^{103}Rh, ^{105}Pd, ^{141}Pr, $^{143,145}Nd$, ^{149}Sm, ^{153}Eu) were measured on 15 fast critical assemblies. The measured central reactivity worths normalized by ^{10}B were compared with calculational values. Necessary corrections (on heterogeneity of media, self-shielding of media and that of a sample and others) were introduced to ensure adequate conditions for the comparison. The IPPE recommended library of fission product capture cross-sections was tested by this comparison and some corrections to these cross-sections were proposed. A satisfactory agreement of the obtained experimental data with STEK and CFRMF experiments was observed in case of similar conditions. The evaluated experimental information was used for updating БНАБ group cross-section library (БНАБ-90).

INTRODUCTION

In 1983-1988 in the Institute of Physics and Power Engineering (IPPE, Obninsk) the experiments were carried out on a series of fast critical assemblies to test capture cross-sections of most important stable fission products(FP) accumulated in a fast reactor core.

On the whole 15 cores with dioxide and metallic fuel (both U and U-Pu) were investigated. The spectral range of assemblies included all fast reactor types of possible interest. Most of the experiments were carried out on БФС facility, some of them - on КБР facility and one of the experiments on - SEG facility (Zfk,Dresden).

Central reactivity worths (CRW) of FP samples were measured by a periodical criticality perturbation method. A list of investigated FPs' and their contributions into a total absorbtion of all fission products is given in Table 1 according to /1/. The investigated nuclides are responsible for approximately 36% of the total FPs' absorbtion.

DESCRIPTION OF THE INVESTIGATED ASSEMBLIES

БФС and КБР critical assemblies consist of tightly packed vertical stainless steel tubes arranged to a triangle lattice. The tubes were filled with pellets of fuel and other materials. The

Table 1

The contribution of investigated nuclides to total absorbtion of fission products for БН-1600 spectrum.

Nuclide	Contribution,%	Nuclide	Contribution,%
^{95}Mo	1.5	^{109}Ag	2.7
^{97}Mo	2.9	^{141}Pr	0.9
^{98}Mo	1.2	^{143}Nd	2.3
^{100}Mo	0.9	^{145}Nd	3.4
^{103}Rh	5.5	^{149}Sm	3.4
^{105}Pd	9.9	^{153}Eu	1.3

outer diameter of the tube is 50 mm, the wall thickness is 1 mm, the lattice pitch is 51 mm.

The structure of the SEG assembly was similar to that of БФС assemblies. Pellets of the same size were placed into vertical channels arranged in an aluminium block with the same lattice pitch.

More detailed description of the assemblies is given in /2,3/. A homogeneous concentrations of core materials are presented in Table 2. Some characteristics of investigated critical assemblies are given in Table 3.

The CRW measurements were carried out in the intertube gaps near the central tube of БФС and КБР cores and in the special graphite column in the center of SEG.

CALCULATION APPROACH AND ANALYSIS OF EXPERIMENTAL RESULTS

The measured CRW of FPs (marked below as ρ^1) were normalized by CRW of ^{10}B (ρ^{10}), whose absorbtion cross-section is well known and depends on the energy according to 1/v law.

An energy dependence of capture cross-sections for most of the FPs in the investigated energy range is similar to that of ^{10}B. That is why the use of boron as a standard for interpretation of reactivity measurement results allows to exclude some of systematic errors arising in the calculational procedure of heterogeneous corrections, corrections on spectrum and self-shielding factors perturbation et al.

A one-dimensional spherical model for a calculation of the critical assemblies was used. The calculations of assemblies were carried out by КРАБ-1 code /4/ using P_1- and S_n-approximations with

Table 2

Homogeneous concentrations of elements in the assemblies
($*10^{22}$ nucl./cm^2).

Assembly	^{235}U	^{238}U	^{239}Pu	O	Na	Fe	Cr
БФС-45А-1	0.182	0.668	0	1.299	0.768	1.045	0.287
БФС-45Б-1	0.181	0.668	0	1.715	0.785	1.328	0.338
БФС-47-5	0.003	0.624	0.170	1.254	0.741	1.153	0.318
БФС-49-1	0.005	1.263	0.172	2.537	0	0.558	0.149
БФС-49-2	0.004	0.969	0.132	1.948	0	0.525	0.141
БФС-49-3	0.005	1.250	0.169	2.512	0	0.559	0.143
БФС-49-4	0.004	0.966	0.131	1.940	0.382	0.629	0.171
БФС-51-1	0.379	0.661	0	0	1.000	1.120	0.305
БФС-52-1	0.197	0.690	0	1.354	0.517	1.362	0.375
БФС-55-1	0.005	1.274	0.142	0	0.621	1.652	0.459
КБР-10	0.048	0.006	0	0.110	0	4.480	1.247
КБР-12	0.051	0.265	0	0.634	0	3.816	1.524
КБР-13	0.049	0.206	0	0.521	0	0.722	4.024
КБР-15	0.026	0.003	0	0.059	0	0.454	5.246
SEG-V	0.173	0.307	0	0	0	0	0
БФС-45А-1	0.134	0.271	0.023	0.038	0.011	0.002	0
БФС-45Б-1	0.246	0	0	0	0	0	0
БФС-47-5	0.149	0.222	0.025	0.038	0.012	0.001	0.007
БФС-49-1	0.070	0.414	0.012	0.064	0.006	0.001	0.007
БФС-49-2	0.066	0.320	0.011	1.494	0.005	0.001	0.005
БФС-49-3	0.075	0.399	0.011	0.128	0.005	0.123	0.008
БФС-49-4	0.080	0.319	0.013	0.050	0.006	0.001	0.005
БФС-51-1	0.143	0.328	0.024	0.007	0.011	0	0
БФС-52-1	0.175	0.697	0.030	0.026	0	0	0
БФС-55-1	0.215	0.204	0.036	0.037	0.017	0.001	0.006
КБР-10	0.583	0	0.098	0.151	0	0	0.163
КБР-12	0.064	0.102	0.011	0.016	0.005	0	0
КБР-13	0.094	0.048	0.016	0.005	0.008	0	0
КБР-15	0.059	0.015	0.010	0.003	0	0	0
SEG-V	0	1.381	0	5.608	0	0	0.159

Notes: *) R - for БФС ^{240}Pu
- for КБР Mo-nat.
- for SEG B-nat.

Table 3

Some integral parameters of assemblies.

Assembly	Fuel	Enrich-ment,%	Share of neutrons in spectrum with $E_n \leq 10$ keV,%	ρ^{10}/ρ^{235} calc. ($*10^{-2}$)	ρ^{1}/ρ^{235} exp. ($*10^{-3}$)
БФС-45А-1	UO_2	21	4.7	-110	175 ± 3
БФС-45Б-1	UO_2	21	5.3	-110	213 ± 3
БФС-47-5	$(PuU)O_2$	21	4.2	- 88	55 ± 2
БФС-49-1	$(PuU)O_2$	12	5.4	- 66	- 2 ± 1
БФС-49-2	$(PuU)O_2$	12	13.1	- 92	77 ± 1
БФС-49-3	$(PuU)O_2$	12	11.3	-102	42 ± 1
БФС-49-4	$(PuU)O_2$	12	6.7	- 72	24 ± 1
БФС-51-1	U	36	0.4	- 72	316 ± 3
БФС-52-1	UO_2	22	4.6	-110	250 ± 5
БФС-55-1	PuU	10	2.4	- 60	- 93 ± 2
КБР-10	UO_2	90	16.0	-154	----
КБР-12	UO_2	16	15.1	-135	----
КБР-13	UO_2	19	13.0	-129	- 15 ± 1
КБР-15	UO_2	90	16.7	-185	----
SEG-V	U	36	18.4	-162	- 4 ± 1
STEK-500	U	90	6.2	-146	----
STEK-1000	U	90	13.5	-253	----
STEK-2000	U	90	18.5	-348	----
CFRMF	U	93	7.8	108*)	----

Notes: 1. *) σ^{10}/σ^{235}- absorbtion to fission cross-sections ratio;
2. ρ^{1}, ρ^{10}, ρ^{235}- central reactivity worths of hydrogen, ^{10}B and ^{235}U correspondingly.

АРАМАКО-80 26-group constants preparation on the basis of БНАБ-78 nuclear data library /5/. During FPs CRW calculation the group cross-sections and self-shielding factors were used after deriving from evaluated neutron data files prepared in the IPPE /6/ and condensing into group constants by means of ГРУКОН code /7/.

A heterogeneous structure of assemblies was taken into account by means of a cell code НЕЕРС /8/. A neutron and adjoint flux in a flat layers model of real cell were calculated by the first collision probability method using the integral transport approximation with resonance self-shielding calculations procedure proposed by T.Tone /9/. A self-shielding for samples of different thickness used in the experiments was assessed by the rational Wigner approxi-

Table 4

Description of samples for reactivity perturbation.

Material	Main isotope content,%	Material	Main isotope content,%
^{95}Mo	95.4	^{109}Ag	99.4
^{97}Mo	91.7	85%$^{141}PrO_2$+	
	-	+15%$^{141}Pr_2O_3$	100
^{98}Mo	95.9	$^{143}Nd_2O_3$	83.2
^{100}Mo	96.1	$^{145}Nd_2O_3$	84.8
^{103}Rh	100	$^{149}Sm_2O_3$	96.9
^{105}Pd	93.8	$^{153}Eu_2O_3$	99.2

mation /10/. A bilinear corrections which are understood as resonance self-shielding decrease in the surrounding medium due to neutron scattering in the sample were also made. The uncertainties of the heterogeneity and sample self-shielding corrections were also evaluated experimentally. They are given in this paper together with the statistical ones.

EXPERIMENTAL SAMPLES

Samples of FPs and ^{10}B used in the experiments were of different thicknesses. Types of chemical compounds of investigated nuclides and contents of the main isotopes are given in Table 4. The contribution of oxygen to a sample worth was measured by means of Al and Al_2O_3 samples. The influence of isotopic admixtures in samples was calculated. The worth of stainless steel cladding of the samples was measured in an additional experiment.

At a preliminary stage during comparing the experimental and calculational ratios of CRW $r^i=\rho^i/\rho^{10}$ for some FPs ($^{95,97,98}Mo$, ^{141}Pr, $^{143,145}Nd$, ^{149}Sm) an essential scattering of discrepancies between experiments and calculations $\delta^i=(r_e^i-r_c^i)*100/r_e^i$ was observed in assemblies with similar spectra (a spectrum characteristic in this paper, ε - is a share in the spectrum of neutrons with energy below 10 keV).

A hypothesis of moisture presence in some samples was proposed for explanation of this fact. The main reasons for taking this hypothesis into consideration were smooth dependence of δ^i on ε and absence of δ^i dependence on hydrogen to ^{235}U CRW ratio (ρ^1/ρ^{235}). A further analysis of measured CRW ratios and an additional

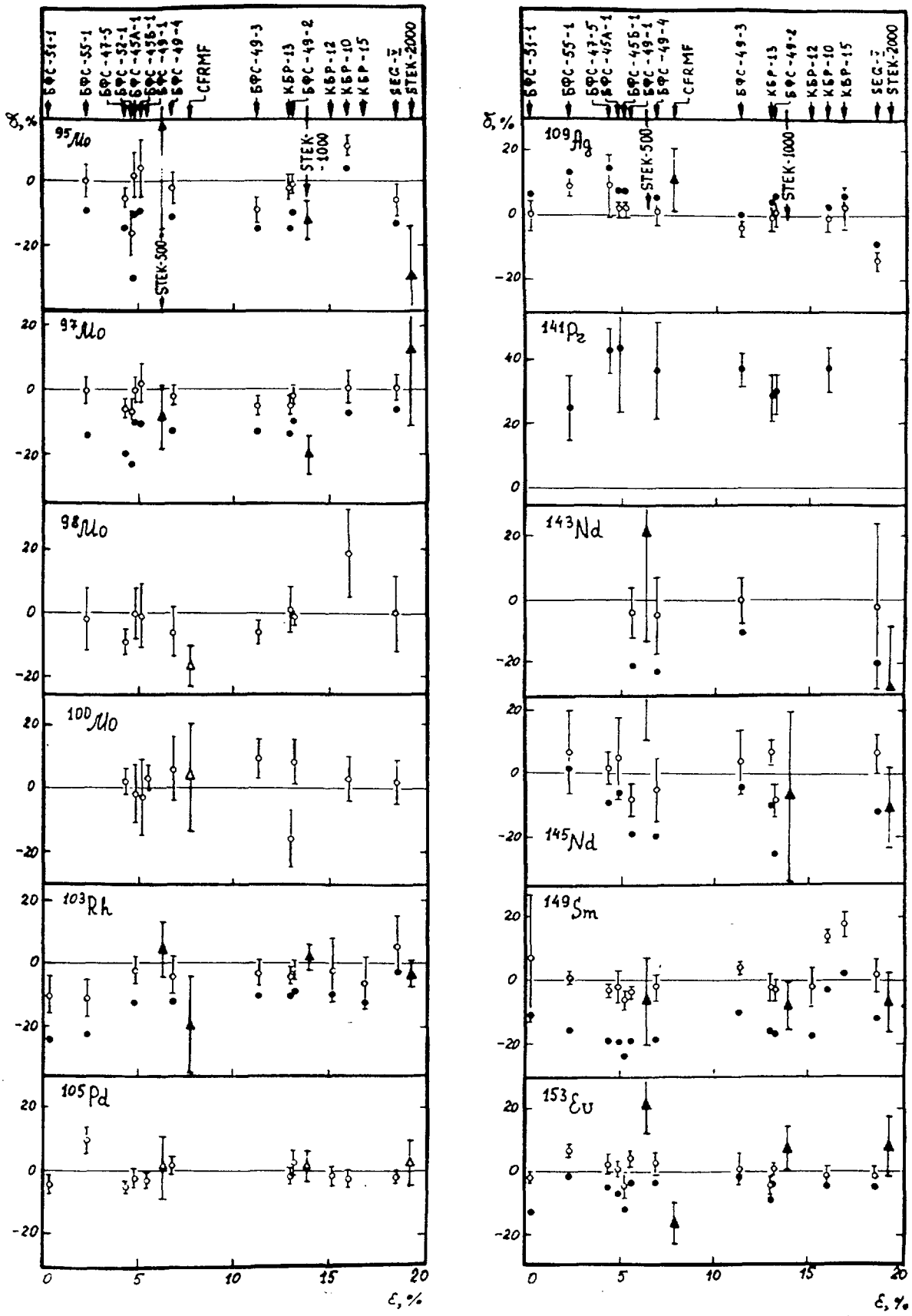

Fig. 1. Discrepancies δ^i between experimental and calculational ρ^i/ρ^{10} values.

experimental information on ratios of hydrogen and oxygen CRW to ^{235}U one (ρ^1/ρ^{235} and ρ^{16}/ρ^{235}) for entire set of assemblies allowed to confirm this hypothesis, to determine a mass content of moisture in the samples and introduce necessary corrections to the experimental ρ^i/ρ^{235} ratios. The control points for this assessment were the experimental data obtained on the assemblies with with $\rho^1/\rho^{235}\simeq 0$ and with essentially different ε (SEG-V and БФС-49-1). The reliability criterion of moisture content determination in the samples can be an agreement of the results obtained with "wet" IPPE samples of ^{149}Sm and "dry" ZfK ones /12/. This method of moisture determination was confirmed by a patent of the Federal Republic of Germany /13/.

COMPARISON OF EXPERIMENTS AND CALCULATIONS. CROSS-SECTIONS CORRECTION.

In addition to comparison of experimental data from БФС, КБР and SEG with calculational ones on the basis of evaluated data files /6/ capture cross-sections correction of FPs was carried out. The capture cross-sections for a number of FP nuclides were corrected to provide their agreement with the experimental data from БФС, КБР and SEG assemblies. An attention was paid to avoid contradictions with existing differential measurements of capture cross-sections.

The extent of agreement between measured ρ^i/ρ^{10} and those calculated on the basis of data recommended in /6/ as well as corrected capture cross-sections are shown in Figure 1.

The energy dependences of FP capture cross-sections resulting from different evaluations including those proposed in this paper are shown in Figures 2-11.

The general information on the results of FPs capture cross--sections testing is given in Table 5. The discrepancies between calculated and measured values of ρ^i/ρ^{10} are presented as an average discrepancy for entire set of assemblies: $\delta^i=(1/N)\Sigma\delta^i_n$ (N is a number of investigated assemblies).

Values of proposed cross-section correction can be found both from Figures 2-11 and in the last coloumn of the Table 5 (j is the energy group number in 26 - group approximation).

The evaluated uncertainties of tested and corrected capture cross-sections of investigated nuclides are given in Table 6. It should be noted that more detailed analysis of the obtained integral data was carried out in papers /12,22,23/.

The experiments on criticality perturbation of assemblies by a small samples were carried out earlier in Netherlands (STEK), Sweden

Table 5

Capture cross-sections testing for fission products on a fast critical assemblies.

Nuclide	Version /6/	Average uncertainty of capture cross--section %	Number of investigated assemblies	Discrepancy δ^i,% in comparison with		Recommendations on correction of version /6/
				version /6/	correction result	
^{95}Mo	JENDL-2	15	11	-13 ± 3	-3 ± 2	-(10-18)% j=6-13
^{97}Mo	JENDL-2	15	11	-13 ± 2	-2 ± 1	-(8-26)% j=6-12
^{98}Mo	JENDL-2	20	10	- 2 ± 3	-2 ± 3	JENDL-2
^{100}Mo	JENDL-2	20	10	+ 1 ± 2	+1 ± 2	JENDL-2
^{103}Rh	IPPE	15	10	-12 ± 2	-4 ± 1	-(5-23)% j=3-14
^{105}Pd	IPPE	10	11	- 1 ± 1	-1 ± 1	IPPE
^{109}Ag	IPPE	10	12	+ 5 ± 2	+1 ± 2	+(2-14)% j=6-14
^{141}Pr	ENDF/B4	10	8	+36 ± 2	----	contradicts to differ. data
^{143}Nd	IPPE	20	4	-18 ± 3	-3 ± 1	+(5-28)% j=6,7,15 -(3-17)% j=8-14
^{145}Nd	IPPE	20	9	-12 ± 3	+1 ± 2	+(1-17)% j=6-8 -(0-26)% j=9-15
^{149}Sm	IPPE	20	14	-14 ± 2	+2 ± 2	JENDL-1
^{153}Eu	ENDF/B4	15	12	- 6 ± 1	+1 ± 1	-(2-11)% j=6-14

Table 6

The evaluated uncertainties of fission products capture cross-sections as the result of testing.

Nuclide	Energy range (keV)		
	1 - 100	100 - 800	800 - 1400
^{95}Mo	5	8	15
^{97}Mo	6	12	20
^{98}Mo	4	6	10
^{100}Mo	3	5	8
^{103}Rh	4	9	15
^{105}Pd	4	6	15
^{109}Ag	3	6	10
^{143}Nd	10	10	15
^{145}Nd	10	10	15
^{149}Sm	7	7	10
^{153}Eu	3	5	8

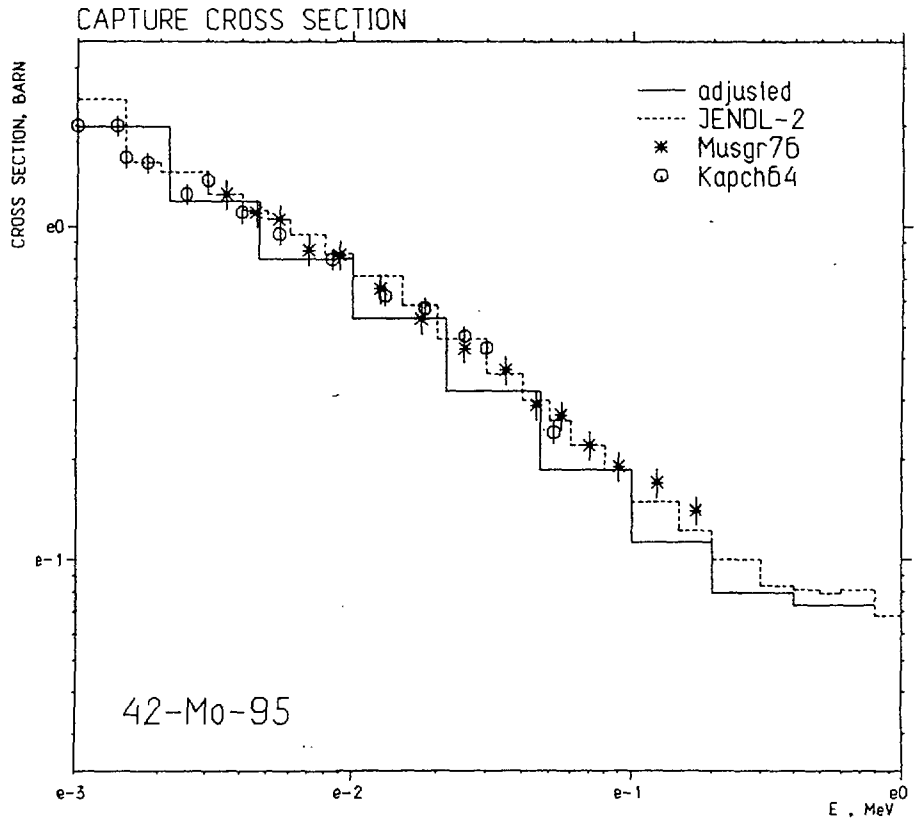

Fig.2. A radiative capture cross section of the molybdenum-95.

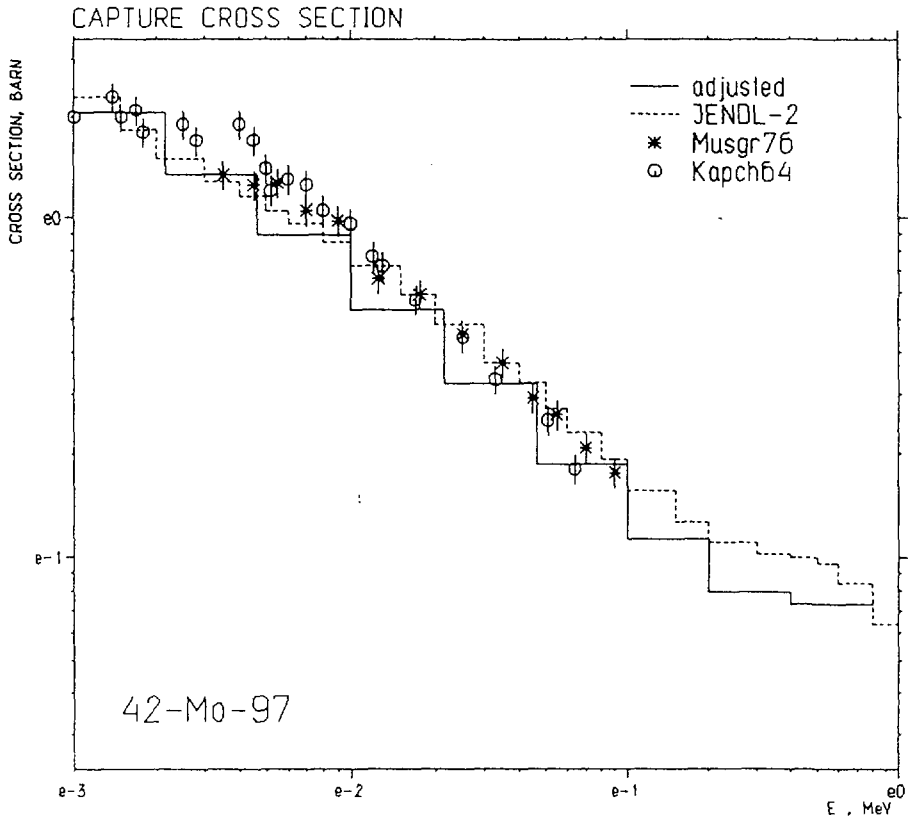

Fig.3. A radiative capture cross section of the molybdenum-97.

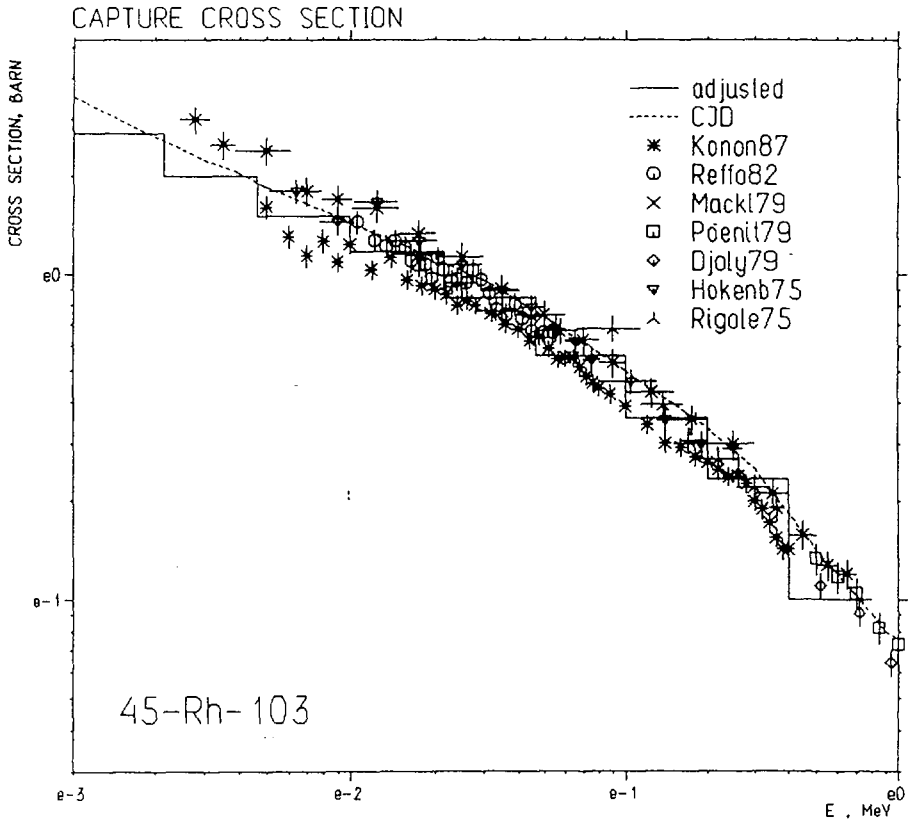

Fig.4.A radiative capture cross section of the rhodium-103.

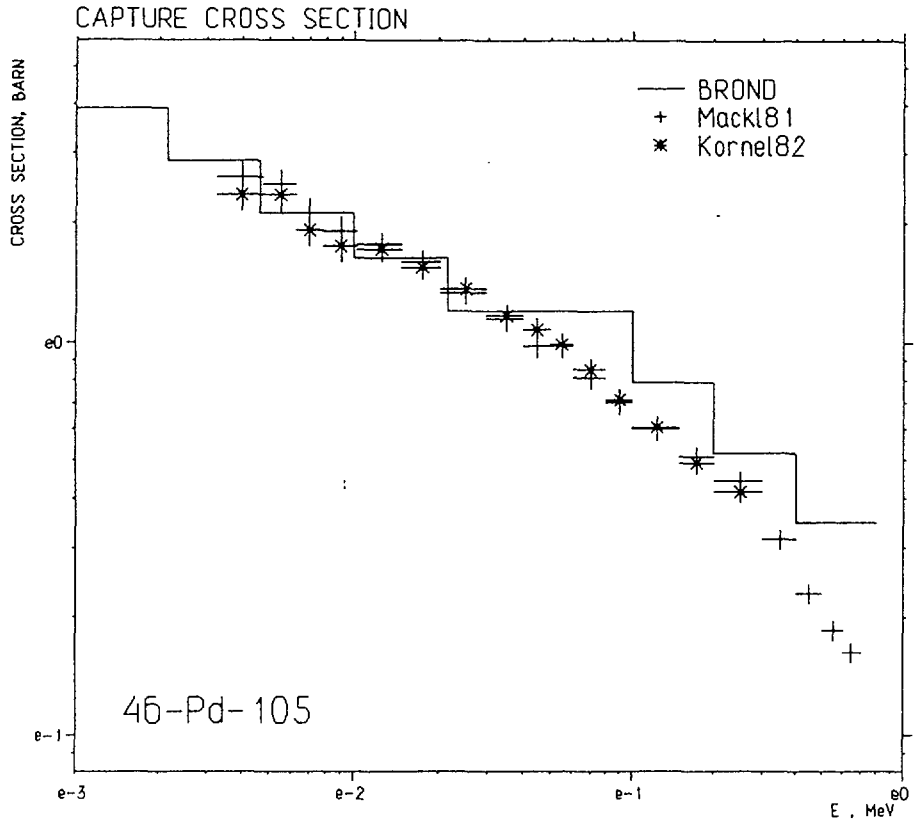

Fig.5. A radiative capture cross section of the palladium-105.

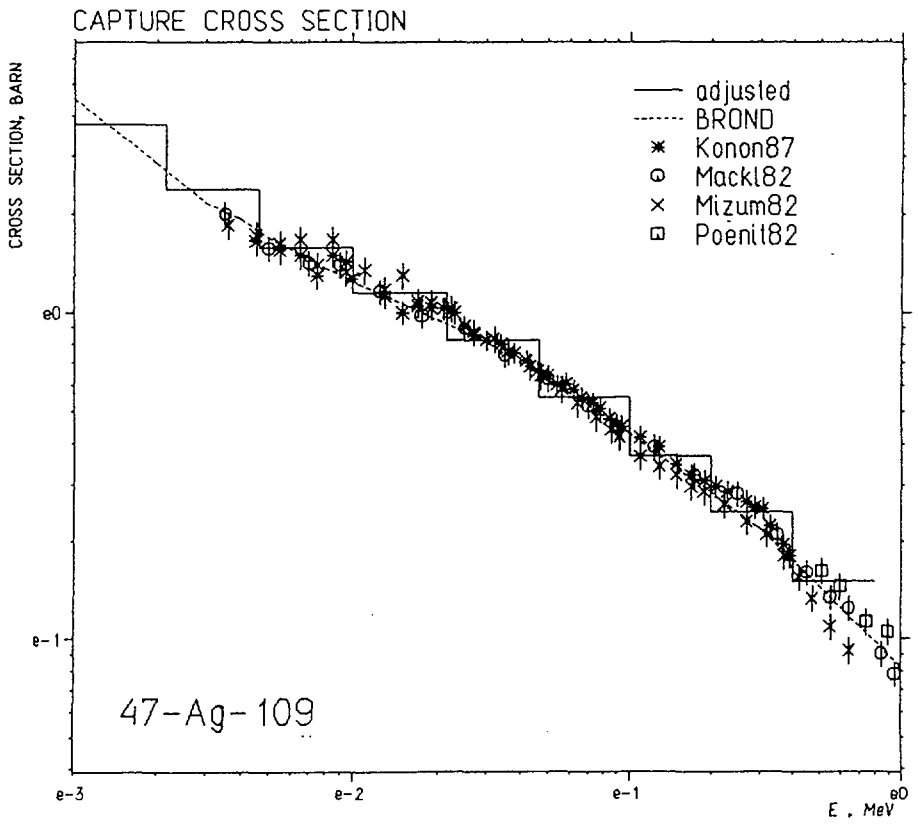

Fig.6. A radiative capture cross section of the silver-109.

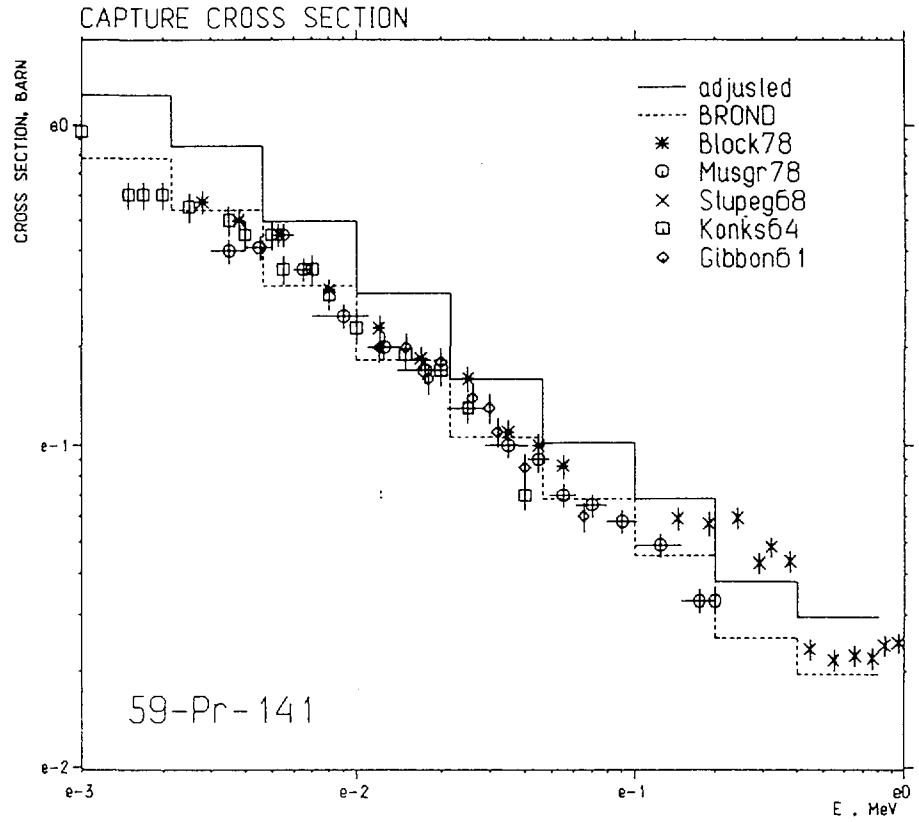

Fig.7. A radiative capture cross section of the praseodymium-141.

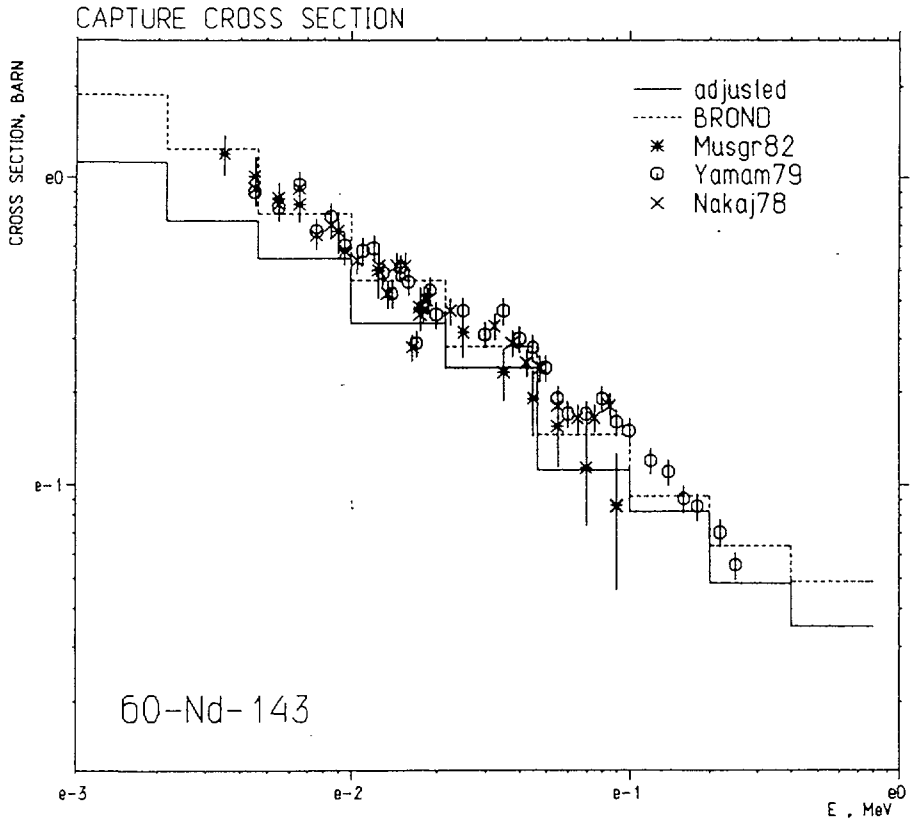

Fig.8. A radiative capture cross section of the neodymium-143.

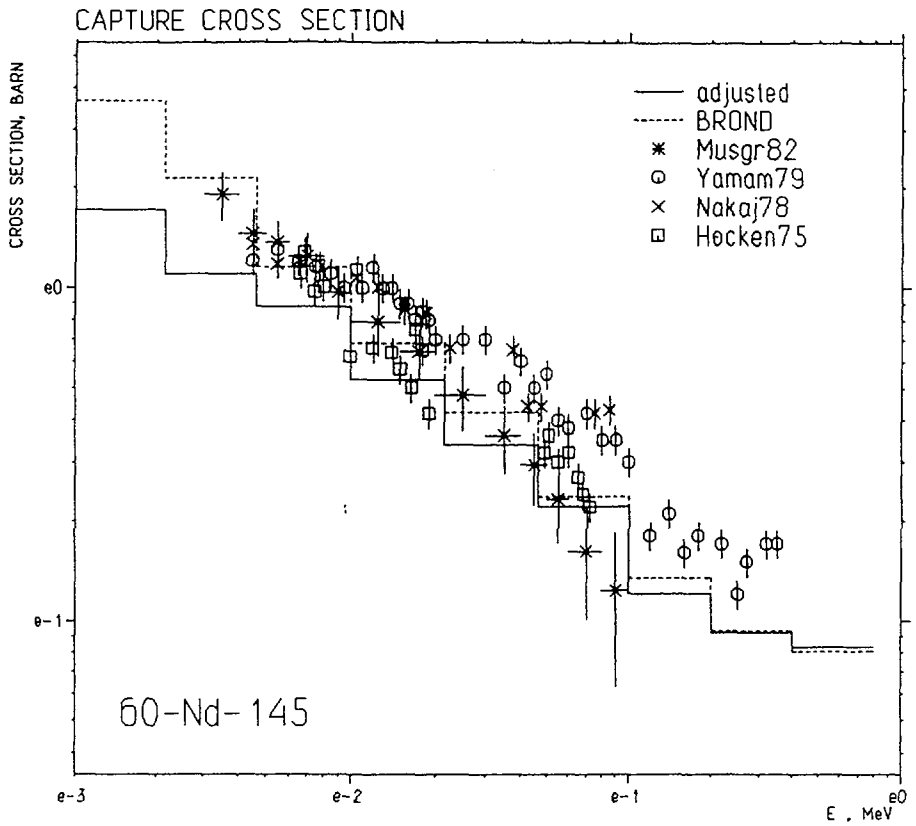

Fig.9. A radiative capture cross section of neodymium−145.

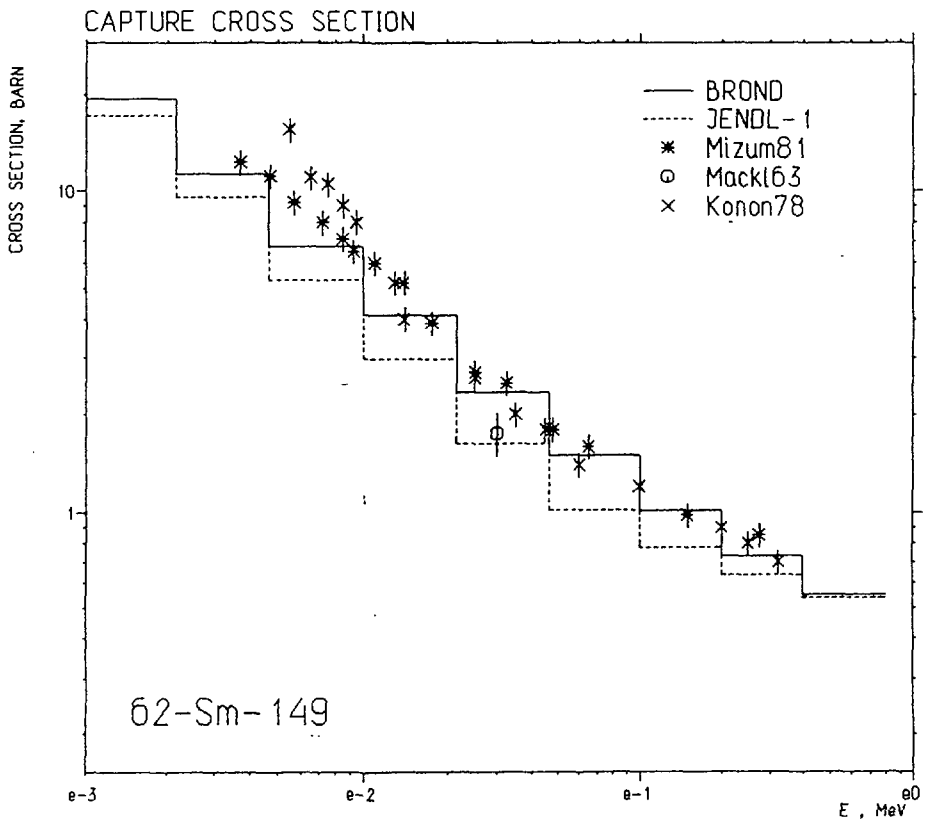

Fig.10. A radiative capture cross section of the samarium−149.

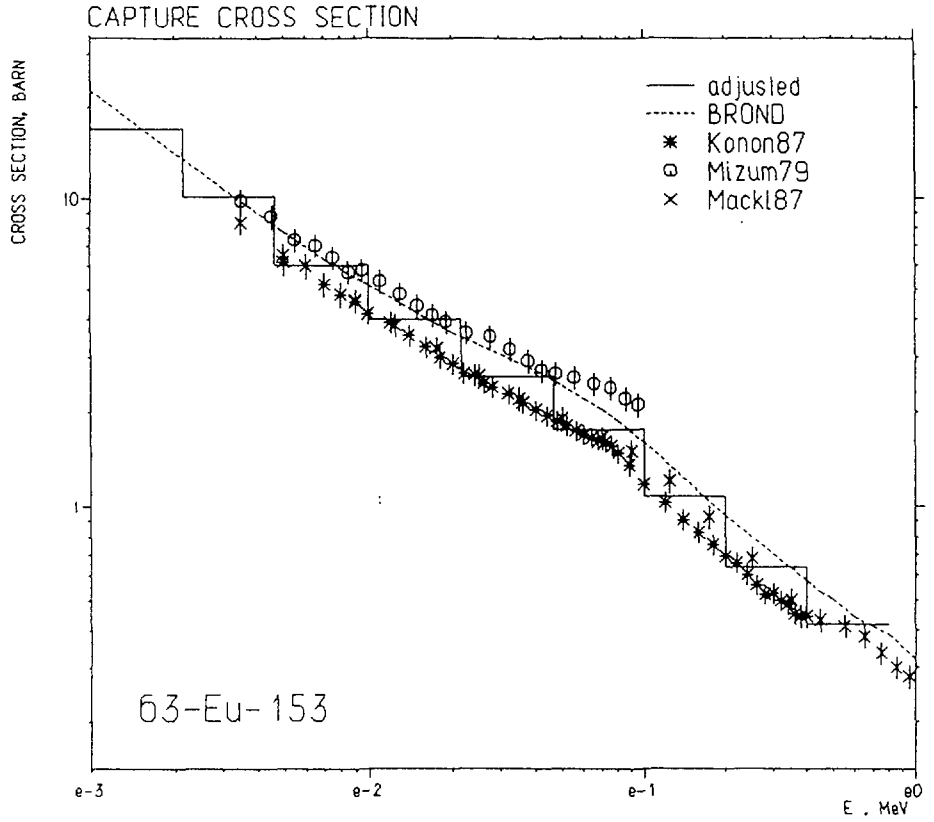

Fig.11. A radiative capture cross section of the europium-153.

(FR0), France (ERMINE, MASURCA) /14-20/. Moreover, the average capture cross-sections of FPs were measured in USA on CFRMF assembly by activation method /21/.

It was of interest to compare the results of БФС, КБР and SEG measurements with other ones. Unfortunately, we have not enough information about experiments on FR0, ERMINE and MASURCA. Nonetheless, the comparison proved to be possible with STEK and CFRMF data.

The structure of STEK could be easily presented by the flat layers model like БФС, КБР and SEG. This fact allowed to use our calculational methods for assessment of experiments on these assemblies. Spectral characteristic ε (a share of neutrons in the spectrum with energy below 10 keV) for STEK assemblies covers the entire range of БФС, КБР and SEG assemblies. But the most suitable cores from the point of view of comparison with our results are STEK-500, -1000 and -2000 whose spectrum is harder then that of other STEK cores. It is also true for CFRMF assembly. Some integral parameters of these assemblies are also given in Table 3.

The agreement of STEK, CFRMF, БФС, КБР and SEG results can be demonstrated by Figure 1.

Since for obtaining corrected cross-sections only the integral data of this paper were used the comparison of STEK and CFRMF

results in Figure 1 is only made with evaluated data /6/ that is enough for assessment of the experimental data agreement.

CONCLUSIONS

The FP capture cross-sections testing on the basis of results from the experiments on БФС, КБР and SEG critical assemblies allowed to improve essentially the reliability and accuracy of the neutron data. No corrections are required for the capture cross-sections of even molibdenium isotopes because of good agreement of experimental and calculational CRW. For ^{141}Pr the data of CRW measurements require to increase capture cross-sections in the region of 1 keV - - 1 MeV by about 50-60%. This contradicts essentially with recommendation made on the basis of differential measurements. So, we do not propose any corrections for this isotope.

The agreement of STEK and CFRMF data with results of this paper in most cases can be considered to be satisfactory.

The evaluated experimental information about capture cross--sections of the investigated nuclides was documented and included in the computer library of macroexperiments LEMEX /24/. This information together with the results of testing recent versions of neutron data for reactor materials was used for updating БНАБ group cross-section library (БНАБ-90).

REFFERENCES

1. Schenter R.,England T. ENDF/B-5 Fission product cross--section evaluation.-In: Proc. Spec. Meeting on Neutron Cross--Sections of Fission Product Nuclei. 1979, Bologna, Italy; NEANDC(E), 209"L", p.253.

2. Казанский Ю.А.,Дулин В.А.,Зиновьев В.П. и др. Методы изучения реакторных характеристик на критических сборках БФС. М.: томиздат, 1977.

3. Dietze K.,Fahrmann K.,Huttel G.,Lehmann E. Neutron data check for structural materials by reactivity measurements in a fast facility with energy independent adjoint flux.- Kernenergie, 1986, Bd 29, s.401.

4. Савоськин М.М.,Маркелов И.П.,Барыба М.А. и др. Аннотация пакета программ КРАБ-1. -Вопросы атомной науки и техники. Сер.: Физика и методы расчета ядерных реакторов, 1984, вып.6(43), с.44.

5. Абагян Л.П.,Базазянц Н.О.,Николаев М.Н.,Цибуля А.М. Групповые константы для расчета реакторов и защиты. М.: Энергоиздат, 1981.

6. Игнатюк А.В.,Кравченко И.В.,Мантуров Г.Н. Библиотека рекомендованных оцененных нейтронных сечений для важнейших продуктов деления ядер. -Вопросы атомной науки и техники. Сер.: Ядерные константы. 1987, вып.1, с.3.

7. Синица В.В. Программа расчета групповых констант на основе библиотек оцененных нейтронных данных. -Вопросы атомной науки и техники. Сер.: Ядерные константы, 1984, вып.5(59), с.34.

8. Бедняков С.М. Реализация метода оценки интегральных экспериментов в условиях стенда БФС: Препринт ФЭИ-2114, 1990.

9. T.Tone. -Journal on Nuclear Science and Technology, -1975, v.12, p.467.

10. Белл Д.,Глесстон С. Теория ядерных реакторов. Пер. с англ. под ред. В.Н.Артамкина - М.: Атомиздат, 1974.

11. Дулин В.А. Оценка точности групповых расчетов возмущений критичности реакторов. -Вопросы атомной науки и техники. Сер.: Ядерные константы, 1983, вып.1(50), с.63.

12. Бедняков С.М.,Мантуров Г.Н.,Дитце К. Тестировка сечений захвата некоторых лантаноидов-продуктов деления в экспериментах на быстрых критсборках. -Вопросы атомной науки и техники. Сер.: Ядерные константы, 1990, вып.4, с.91.

13. E. Lehmann,G.Huttel,K.Dietze,S.M.Bednyakov.- Patentschrift DD 294098 A5.

14. Klippel H.,Smith J. The coupled fast-thermal critical facility STEK.- RCN-209, 1974.

15. Veenema J.,Janssen A. Small sample reactivity worths of fission product isotopes and some other materials measured in STEK.- - ECN-10, 1976.

16. Fast Reactor Programme Second Quarter 1973 Progress Report.- RCN-190, 1973.

17. Fast Reactor Programme Third Quarter 1973 Progress Report.- - RCN-199, 1973.

18. Fast Reactor Programme Fourth Quarter 1973 Progress Report.- RCN-207, 1974.

19. Anderson T. Integral measurements of fission product reactivity worths in some fast reactor spectra.- AE-428, 1971.

20. Martin-Deidier L. Mesure integrale de la capture des produits de fission dans les reacteurs a neutron rapides. CEA-R- -5023, 1979.

21. Harker Y.,Anderl R. Integral cross-sections measurements on fission products nuclides in fast reactor fields.- In: Proc. Spec. Meet. on Neutron Cross-Sections of Fission Product Nuclei, Bologna, Italy, Dec. 12-14, 1979, p.5.

22. Бедняков С.М.,Боховко М.В.,Мантуров Г.Н.,Дитце К. Уточнение сечений захвата ^{103}Rh, ^{105}Pd, ^{109}Ag и ^{153}Eu на основе анализа экспериментов на быстрых сборках. -Атомная энергия, 1989, т.67, вып.3, с.199.

23. Бедняков С.М.,Мантуров Г.Н.,Дитце К. Уточнение сечений захвата изотопов молибдена на основе анализа экспериментов на быстрых сборках. -Атомная энергия, 1990, т.69, вып.1, с.31.

24. Коробейникова Л.В.,Мантуров Г.Н.,Цибуля А.М. Библиотека оцененных макроскопических экспериментов на быстрых критсборках и реакторах. -В кн.: Нейтронная физика. М.: ЦНИИатоминформ, 1988, т.1, с.305.