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Final Report of Research Contract 14485

Resonance and Fast Neutron Induced Fission Cross Sections of Americium and Curium Nuclides

Third-year Progress Report of Research Contract 14485

(under the Coordinated Research Project on Minor Actinide Neutron Reaction Data (MANREAD))

Prepared by

A.A. Alekseev[1], A.A. Bergman[1], A.I. Berlev[1], E.A. Koptelov[1],
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ABSTRACT

The neutron induced fission cross sections of Am and Cm isotopes were measured relative to ^{239}Pu in the neutron energy range from 1 eV to 20 keV at the INR RAS lead slowing down spectrometer LSDS-100. The fission resonance integrals were also estimated using the measured cross section data. The results have been compared with the available experimental and evaluated data. This analysis has shown the present status of the measured fission cross sections and the necessity to revise the evaluated cross sections libraries for the minor actinides.

January 2012

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Most of the minor actinides are the highly radioactivity nuclides. So, the minute samples of actinides and intensive neutron fluxes are needed to measure the fission cross sections of these nuclides. The fission cross sections of the ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{248}Cm, as well as ^{241}Am, ^{242m}Am and ^{243}Am were measured with a Lead Slowing Down Spectrometer **(**LSDS-100) installed at the INR RAS proton Linac (Moscow Meson Factory - MMF). Joint team of experimentalists was named INR – IPPE (Troitsk-Obninsk). LSDS was successfully used for cross section measurements of the nuclides that cannot be investigated by other methods.
The experimental method has been described in the different papers [1 - 8]. Spectrometer LSDS-100 has a form of prism 3.3 m length, 1.62 m width and 1.79 m height. Spectrometer was assembled from 100 blocs of high purity lead (99.996%). Total mass was 100 t. The proton beam parameters were following: the primary energy was equal to 209 MeV, the current pulse width was 1-2 μs, the repetition rate was 50 Hz, and current in the peak was 10-15 mA. The electronics allowed time and amplitude analyses of the fission events during the time range up to 18 ms. Fission events were detected by the multi-sectional fast fission ionization chambers being placed into the operating channels of the LSDS-100 at the distance of 82 cm or 120 cm from centre of the neutron source.
The fissile samples (6 mm in diameter) were prepared using the separated isotopes of high purity just before the measurements. The isotopic composition of the samples is listed in Tables 1 and 2. The Am and Cm fission cross sections were measured relatively to the ^{239}Pu(n,f) cross section. A mass of the high purity (99.9966%) reference ^{239}Pu sample was 17-20 μg. The mass ratios of the fissile materials were determined from their α-activity using semiconductor spectrometer with 1% uncertainty.

Table 1. The masses of Cm samples (m) and their isotopic composition (% at).

sample	mass, μg	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	^{248}Cm
^{243}Cm	4.26	**95.65**	3.79	0.057	**0.48**	0.100	**----**
^{244}Cm	4.51	0.18	**99.32**	0.09	0.56	**0.007**	**----**
^{245}Cm	8.44	**----**	1.110	**98.48**	0.405	**----**	**----**
^{246}Cm	13.0	**----**	0/207	0.0165	**99.53**	0.137	0.10
^{247}Cm	0.335	**----**	2.23	0.78	12.56	**73.84**	10.59
^{248}Cm	3.57	**----**	1.46	0.022	1.3	0.229	**96.98**

Table 2. The masses of Am samples (m) and their isotopic composition (% at).

sample	mass, μg	^{241}Am	^{242m}Am	^{243}Am
^{241}Am	6.41	**~100**	$< 3\cdot10^{-4}$	$< 3\cdot10^{-4}$
^{242m}Am	3.44	13.768	**80.865**	5.36
^{243}Am	4.70	0.0045 ±0.002	0.0035 ±0.002	**99.92**

<u>Cm isotopes</u> [2–7]. The all Cm samples (Table 1) contain the impurities of neighbour isotopes. So their fission cross sections have been measured at the same time and the absolute value of the fission cross section has been calculated as a result of the joint analysis. The normalization constants take into account the uncertainties in the basic cross section (~3%), in the Cm/Pu atom ratios and in the ratio of the Cm/Pu fission detection efficiency. The systematic uncertainty due to the normalization was evaluated as 4.0% for ^{246}Cm, ^{247}Cm and 3.4% for ^{248}Cm. The received data in tabulated form have been placed in the EXFOR database.

The average reduced cross section values $\langle \sigma \sqrt{E} \rangle$ of the curium isotope x are obtained from the measured time spectra *N(t)* of fission events using the relation:

$$\langle \sigma_x(E)\sqrt{E} \rangle = (N_x / N_9)_{E=\overline{E}(t)} \cdot (n_9 / n_x) \cdot (\varepsilon_9 / \varepsilon_x) \cdot \langle \sigma_9(E)\sqrt{E} \rangle, \qquad (1)$$

where $\langle\ \rangle$ – an averaging sign on the actual neutron energy E in the vicinity of the mean neutron energy $\overline{E}(t)$ at the moment *t* of the slowing down time; index *x*=9 corresponds to the ^{239}Pu standard sample; n_x is the number of atoms in the sample *x*; ε_x is the detection efficiency for fission fragments in sample *x*; $\langle \sigma_9(E)\sqrt{E} \rangle$ is the average reduced cross section of ^{239}Pu from the ENDF/B-VII.0 evaluation. The ratio $N_x(t)/N_9(t)$ is calculated from the measured time spectra, the value (n_9/n_x) is determined in the α-activity experiment, and $(\varepsilon_9/\varepsilon_x)$ is found from the analysis of the measured amplitude spectra with taking into account corrections on the fission fragments losses in the fissile layers.

In this work, the determination of the functions $\overline{E}(t)$ and $\langle \sigma_9(E)\sqrt{E} \rangle$ were done in the framework of the LSDS computer model (based on Monte-Carlo calculations). The model parameters have been deduced from the condition of the optimal fitting of the measured time spectra for ^{235}U and ^{239}Pu with the simulated time spectra calculated using the ENDF/B-VII files for the standards. The model allowed to reproduce all observed structures in ^{239}Pu fission cross section up to epithermal neutron energies ($t \leq 6000$ μs).

The model calculations show that the mean neutron energy $\overline{E}(t)$ depends on the slowing down time *t* as

$$\overline{E}(t) = K/(t+t_0)^2 + e_T, \qquad (2)$$

where parameter e_T varies from 20 meV to *kT* as a function of *t*. In the case of $^{244, 246}$Cm the slowing down constant *K* is equal to 169.2 keV μs^2 and t_0 = 0.35 μs; for ^{243}Cm case *K*=168.8 keV μs^2, t_0 = 0.85 μs.

The threshold sensitivity of LSDS-100 was estimated from the measurement of the ^{238}U fission cross sections (total operating time was 11 h) in the maximum of the 0.72 keV resonance (σ_m = 1.78 ± 0.24 mb). The measured value ($m\sigma_f \cong 1$ mg mb) was close to the one available in data library.

<u>^{243}Cm</u> [3]. The time spectrum *N(t)* has been measured for the neutrons with corresponding energies in the range E=0.03 eV to 20 keV. The total operating time of measurements was 45 h. Due to the low detection efficiency (about 60%) the absolute value of the fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ was determined by the normalization to the ENDF/B-VII data at the neutron energies 29 – 62 meV. The correction was applied at 11% admixture of ^{239}Pu daughter atoms formed due to α-decay of ^{243}Cm. No corrections have been done for negligible contributions from ^{244}Cm and ^{246}Cm isotopes presented in the ^{243}Cm sample.

Analysis of the fission cross section shape in the epithermal energy range allowed the estimate of Westcott factor as *g(T)* = 0.985 for the temperature $T = 293.6$ *K*. The ENDF/B-VII evaluation for $\sigma(E_T)\sqrt{E_T}$ is based on the experimental value at the thermal point E_T with *g(T)* = 1. After corresponding correction, the new ENDF/B-VII value for $\sigma(E_T)\sqrt{E_T}$ was estimated as 102.1 ± 4.6 b eV$^{1/2}$, compare with 98.7 b eV$^{1/2}$ – the old ENDF/B-VII value.

The final ^{243}Cm cross section data after their renormalization are shown in Fig. 1. Their systematic uncertainty due to the normalization procedure is 2% and uncertainty in the $\langle \sigma_9(E)\sqrt{E} \rangle$ values is less than 5%. The relative statistical uncertainty depends on the neutron energy and is equal to 2.2 – 1% in the range 14 keV – 1.5 eV, 2 – 6.8% in the range 1 – 0.29 eV, 2.3 – 9.8% in the range 0.28 eV–43 meV and it goes up to 55% at E = 30 meV.
In order to compare LSDS-100 results with the available experimental data of the Los Alamos group (bomb shot data) [9] and the ENDF/B-VII, these data were averaged with the Gaussian resolution function having a half-width of distribution $\Delta E = 0.35E$. As seen in Fig.1, the LSDS-100 data match to the broadened Los Alamos data, especially in the range 20 -50 eV. By this, the known resonance structure has been confirmed. The resonance peak at 72 eV in our data has less strength than resonance in the bomb shot data and in the evaluated data. The JEFF-3.1 data show strong resonance at $E \cong 16$ eV but it practically not seen in the ENDF/B-VII data. The peak in our data has an intermediate strength. It will be noted that JEFF-3.1 does not point out the presence of any resonance at $E \cong 27$ eV which is seen in other works.
In the range E < 10 eV, where the experimental fission data are absent, the LSDS-100 data are ~25% below ENDF/B-VII data but the cross section shape is the same. Remarkable peaks at E~ 7 eV (due to two resonances at 6.15 eV and 7.21 eV), E ~ 2.31 eV and at E ~0.67 eV were observed.

^{244}Cm [4]. The time spectrum was measured in the neutron energy range from $E = 0.1$ eV to 20 keV. The total operating time was 45 h. The absolute values of the fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ have been calculated by using Eq. 1 with the normalization constant $(n_9/n_4)\cdot(\varepsilon_9/\varepsilon_4) = 4.11 \pm 0.17$. The ratio $(\varepsilon_9/\varepsilon_4)$ was found to be 1.07 ± 0.04. It is a result of analysis of the measured amplitude spectra and calculation of the detection efficiency (ε_4=0.91 ± 0.03). The systematic uncertainty due to the normalization is equal to 4.1%. The cross section values have been corrected for contribution from daughter nuclei of ^{240}Pu formed in the α-decay of ^{244}Cm, and for impurities of the principal isotopes ^{243}Cm and ^{245}Cm in the ^{244}Cm sample.
The results of the fission cross-section measurement of ^{244}Cm are shown in Fig. 2. The statistical accuracy of the data is 4.12% for $E \geq 5.6$ eV in the average (the best is 1.9%, the worst - 11.4%). The relative uncertainty is increased from 12% to 40% from high to low energies.
Our data have confirmed all the known resonance structures at E >5 eV and are in good agreement with the LANL data [10]. The LANL data were obtained by using time-of-flight method (TOF) and neutrons were produced in underground nuclear explosion (Physics 8). For comparison with the LSDS-100 data, the LANL results were broadened according to the LSDS–100 energy resolution. Excluding the range 5–20 eV, the LSDS–100 values are notably below the values obtained with the LSDS–75 assembly RINS [11]. The ENDF/B-VII evaluation is based on the RINS data in the range E > 1 keV. The discrepancy between the RINS results and the ENDF/B-VII data in E <5 eV region is explained by the isotopic impurities of ^{245}Cm and ^{243}Cm.

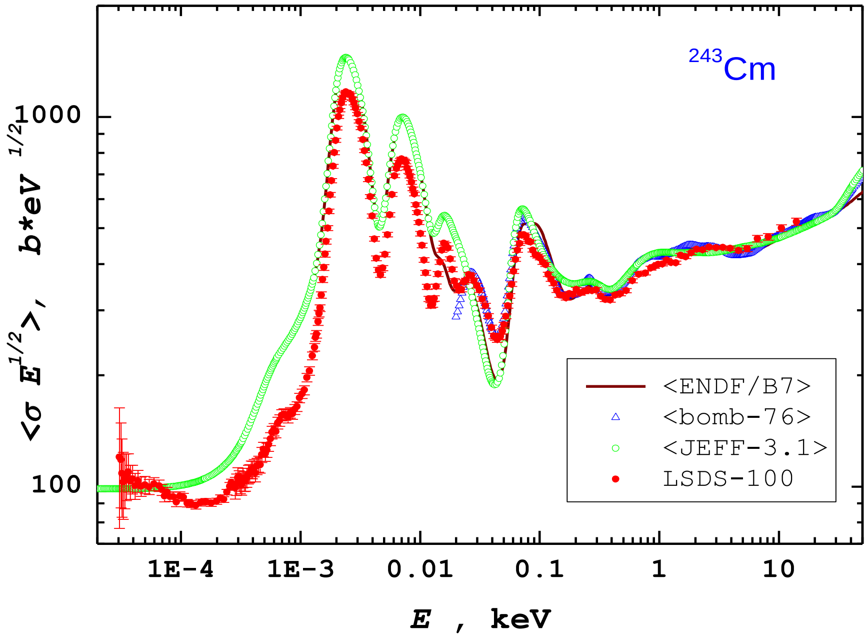

Fig. 1. Reduced fission cross section $\langle\sigma(E)\sqrt{E}\rangle$ of ^{243}Cm vs. the neutron energy E.

Designations: ● – LSDS-100 data [4]; (△) – "bomb" data [9]; solid line – ENDF/B-VII data; (○) – JEFF–3.1 data. The "bomb" data and the recommended data are averaged with the LSDS-100 resolution function.

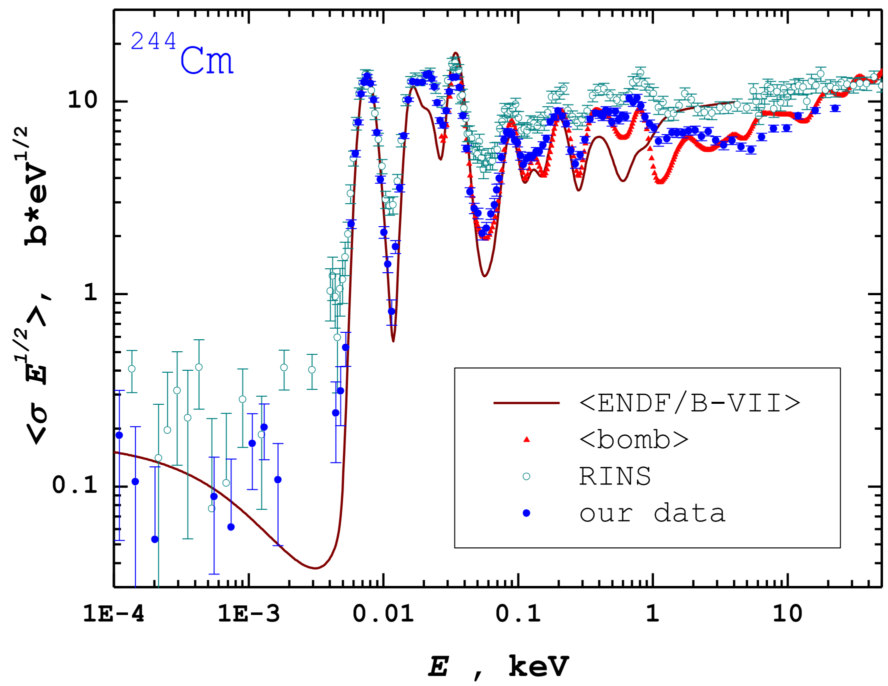

Fig. 2. Reduced fission cross section $\langle\sigma(E)\sqrt{E}\rangle$ of ^{244}Cm vs. the neutron energy E.

Designations: ● – LSDS-100 data [3]; ▲ – LANL data [10]; ○ – RINS data [11]; solid line - ENDF/B-VII data. The LANL and the ENDF/B-VII data are averaged with the LSDS-100 resolution function.

^{245}Cm [2]. The results for epithermal neutrons are shown in Fig. 3. The value of the thermal cross section recommended in ENDF/B-VII (σ_T = 2149 b) is based on the TOF data (2143 ± 58 b) from Livermore [12]. The weighted average value at 25.3 meV for reactor data is equal to $<\sigma_T>$ = 1971 ± 33 b which is 8% less than ENDF/B-VII value. The ENDF/B-VII value for $\sigma_T\sqrt{E_T}$ is 342±9 b eV$^{1/2}$ at the energy 25.3 meV. The extrapolation of the LSDS–100 data to the thermal point gives the value 344 ± 8 b eV$^{1/2}$.

Available data for the ^{245}Cm fission cross section in resonance energy range are shown in Fig. 3. The ENDF/B-VII evaluation is based on the TOF data of Livermore (E<63 eV) [12, 13]. For neutrons with E > 20 eV, the Physics 8 bomb shot experiment [10] resulted with some remarkable data. To compare with the LSDS-100 result, all the former data were averaged with the LSDS-100 spectrometer resolution function. For neutrons with energy below 200 eV the LSDS-100 data agree well with other data on absolute values of the cross-sections as well as on its energy dependence. The good agreement between our data, the bomb shot data and LSDS-44 KIAE data [14] demonstrates the drawback of the ENDF/B-VII evaluation which does not reproduce the intermediate structure in the cross section observed at 100 –200 eV energy range and near 8 eV. Our data are also 12–17% higher than the ENDF/B-VII values in the energy range E_n = 2–20 keV.

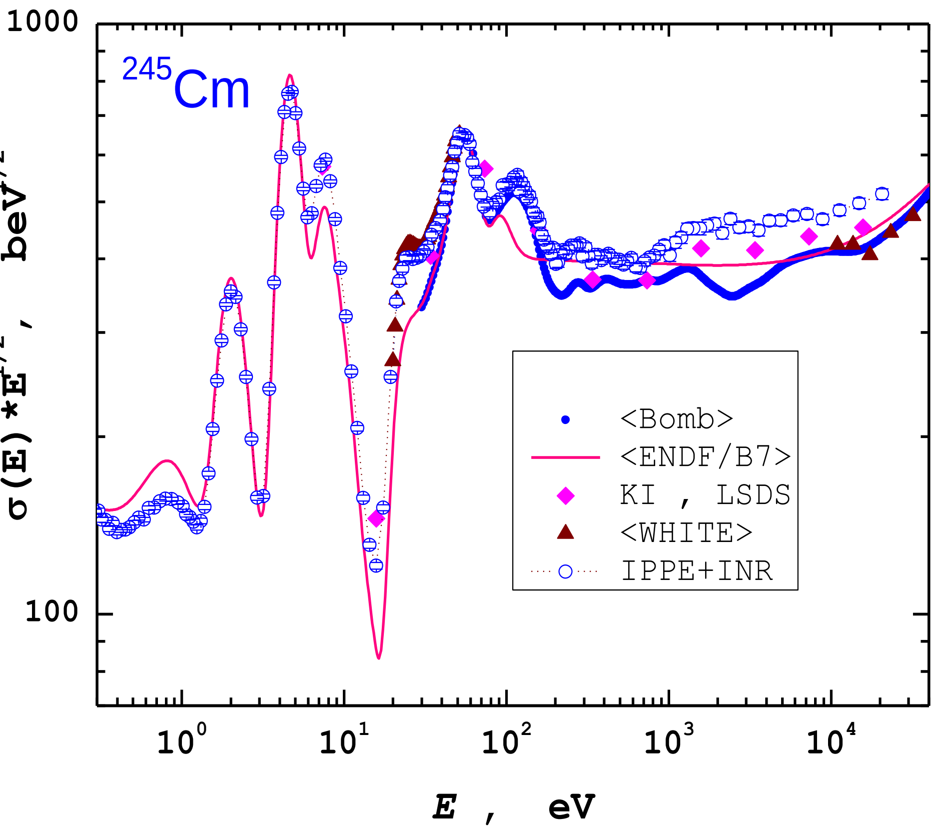

Fig. 3. The reduced fission cross section $\langle\sigma(E)\sqrt{E}\rangle$ of ^{245}Cm from 0.5 eV to 20 keV.

Designations: **○** – LSDS-100 data [3]; solid line **—** ENDF/B-VII; ◆ – LSDS-44 [12]; ▲ – [13];
• – [10]. The ENDF/B-VII data and data [10, 13] are averaged taking into account the LSDS-100 spectrometer energy resolution.

^{246}Cm [5,6]. The total operating time of measurements was 47 h. The time spectrum contains a constant background component due to the spontaneous fission, which dominates in the range $t > 450$ μs. In this range the statistical uncertainty exceeds 50 %. The statistical accuracy is ~6 % for $t < 50$ μs in the average (the best is 3.0%, the worst is 7.7%). In the range $t = 50 - 200$ μs the relative uncertainty changes between 5% for resonance peaks and 36% for cross section valleys.

The absolute values of the fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ have been calculated using Eq. 1 with the normalization constant $(n_9/n_6)\cdot(\varepsilon_9/\varepsilon_6)=2.25 \pm 0.08$. The ratio $(\varepsilon_9/\varepsilon_6)$ has been found equal to 1.07 ± 0.04 and the detection efficiencies are $\varepsilon_6=0.91 \pm 0.03$ and $\varepsilon_9=0.97 \pm 0.02$. The systematic uncertainty due to the normalization is equal to 4.0%. The cross sections $\langle \sigma\sqrt{E} \rangle$ are plotted in Fig. 4.

The data have been corrected for the contribution from high fissile isotopes ^{243}Cm and especially from ^{247}Cm in the ^{246}Cm sample. The contribution from ^{247}Cm dominates in the range $E < 3$ eV.

In the range $E = 0.71 - 2.63$ eV the data have been combined into a single group. The accuracy of the cross section value in this energy group is ~50% and is determined by a separation of dominant contribution of ^{247}Cm resonance at 1.24 eV. In the range $E = 0.0615 - 0.68$ eV the measured average cross section is negative. The upper limit of the 95% confidence interval for this range is equal to 0.038 b eV$^{1/2}$ being the sensitivity threshold for these measurements most probably.

As it can be seen in Fig. 4, the LSDS–100 data are between the RINS data [11, 15] and the bomb shot results of LANL group [10]. All known resonance structures have been confirmed. The ENDF/B-VII evaluation is based on the RINS data in the range above 3 eV. Our data in the range $E > 200$ eV are about 1.4 times lower than the RINS data. Above 3 eV the average accuracy of the RINS values is ~10%, whereas the best one is ~7%. At lower energies the uncertainties are 30 − 50%. The best accuracy of the LANL data is equal to 11% and in the average it is less than 20%.

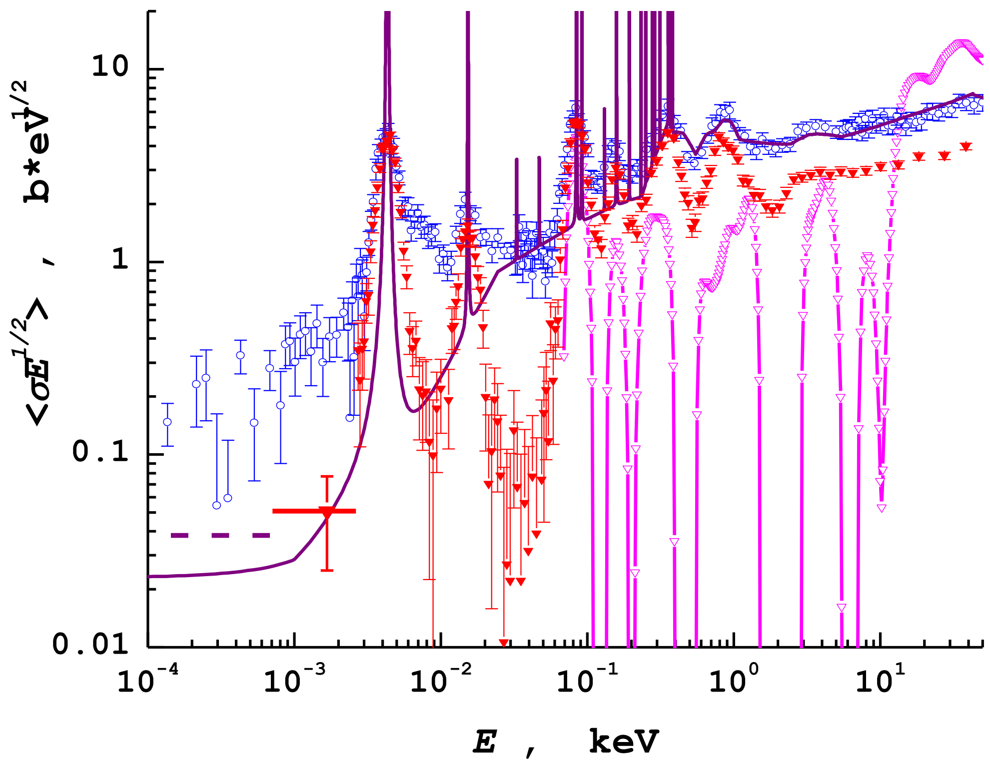

Fig.4. Reduced fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ of ^{246}Cm vs. the neutron energy E.

Designations: (▼) – LSDS-100 data; dash line (- -) – top 95% confidence level for our data; (○) – RINS data [11, 15]; solid line – ENDF/B-VII data; (▽) – LANL data [10], averaged on the Gaussian with a width $\triangle E$=*0.28E.*

We observed almost fully resolved resonances below 85 eV unlike the RINS data. It seems that the existence of resonances at 33 eV and 47 eV assumed in the ENDF/B-VII evaluation cannot be justified. Our data do not contradict to the ENDF/B-VII data in the energy range below 3 eV. The results of the fission cross-section measurement in the range E = 0.14 eV − 20 keV confirm the main conclusions of our earlier work [5] for this nuclide. In the present experiment the total number of detected events was about 3.3 times more than earlier. The statistical accuracy is 2.5 - 4% for E < 250 eV and in the resonance peaks.

^{247}Cm [6]. The results of the fission cross-section measurement are shown in Fig. 5 in the range $E =$ 0.03 eV − 20 keV. The statistical accuracy is 1.5 - 3% for E > 1 eV and 3 - 10% for E < 1 eV. Within error limits our data are in good agreement the RINS data [15] and the LANSCE time-of-flight data [16]. Our data show a more striking resonance structure than the RINS data. In the range $E =$ 0.03 – 0.1 eV the average LSDS-100 cross section is $\langle \sigma\sqrt{E} \rangle = 19.0 \pm 0.4$ b eV$^{1/2}$. It is some higher than the ROSFOND-2010 estimate (17.7 b eV$^{1/2}$) in the thermal point and it differs noticeably from the JEFF-3.1 recommendation (13 b eV$^{1/2}$).

A satisfactory agreement is found between our data, the RINS data, the LANSCE data and the ROSFOND estimate until E=150 eV. For higher energies our data are found to be situated below and the discrepancy is about 17% at 20 keV. But at the same time our data are in agreement with the averaged data of the Physics 8 experiment [10].

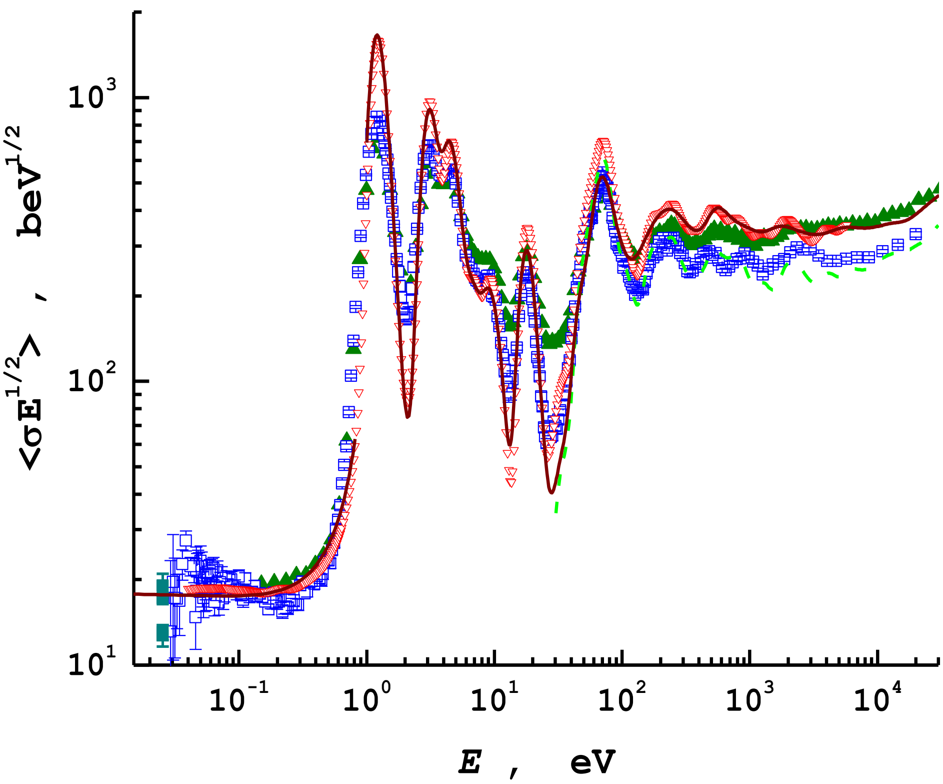

Fig. 5. Reduced fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ of ^{247}Cm vs. neutron energy E.

Designations: (◘) – LSDS-100 data; (▲) — RINS data [15] ; (▽) – LANSCE data [16]; (■) – thermal point data; solid line (——) – ROSFOND-2010 evaluation; dashed line (– –) – Physics 8 (LANL) data [10]. The data of LANSCE, LANL and ROSFOND are averaged on the LSDS-100 resolution function.

^{248}Cm [6]. The cross sections $\langle \sigma \sqrt{E} \rangle$ are plotted in Fig. 6. The relative uncertainty of the data varies with the energy from 20 to 100% for E < 20 eV and to 1% for the resonance peak at 77 eV. Further it rises up to 11% at E ~1 keV and falls away to 4% at E ~10 keV.
In the range E = 150 eV – 1 keV the LSDS-100 data are found to fall apart from the RINS data [11] and from the ROSFOND-2010 data being broadened according to the LSDS–100 energy resolution. The Physics 8 data [10] are in disagreement both with RINS and our data in this range. In the energy interval 10 – 30 keV the discrepancy between the data disappears.
The cross section analysis has been made for eight low lying resonances up to 238 eV. Within one and half error the results agree with the RINS values and the LANL ones for the resonances 26.9, 76.1, 98.95 and 237.9 eV. An excellent agreement has been found for the strong resonance at 76.1 eV.

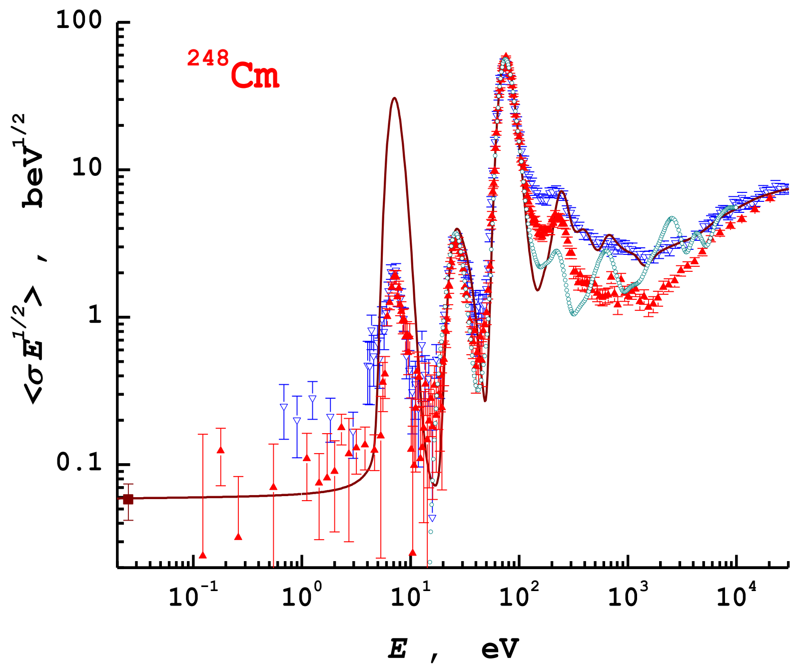

Fig. 6. Reduced fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ of ^{248}Cm vs. the neutron energy E.

Designations: (▲) – IPPE+INR data; (∇) – RINS data [11]; (■) – thermal point data; (◦◦◦) – LANL (Physics 8) data [10]; solid line (—) – ROSFOND-2010 evaluation. The LANL and ROSFOND data are averaged on the LSDS-100 resolution function.

Am isotopes.

^{241}Am. An attempt of the measurement with the ^{241}Am sample (6.41 μg) has been made. However during analysis of the measured time spectra it was found that the fissile layer contains 0.6% of ^{235}U impurity atoms. So for the ^{241}Am sample only qualitative results were obtained. Our data show a more striking resonance structure than the KULS data [17] and do not contradict to JENDL-4.0 cross section trend in general.

^{242m}Am [2]. The results of the fission cross section measurements in the epithermal neutron energy range are shown in Fig. 7a. The averaged value weighted on all data at 25.3 meV is equal to 6408 ± 212 b that agree with the ENDF/B-VII recommended value (6416 b) and is close to the time-of-flight (TOF) data of the Livermore group 6329 ± 320 b [18]. The averaged value for $\sigma_T \sqrt{E_T}$ is 1020 ± 34 b·eV$^{1/2}$. In the energy range 30 – 50 meV the LSDS–100 data are in a good agreement with

the data of the Japan group [19] (where LSDS-40, thermal neutron facility and TOF method were used) and the TOF data of Oak Ridge [20]. The extrapolation of the LSDS–100 data to the thermal point gives the value 1087 b·eV$^{1/2}$ with statistical uncertainty 9 b·eV$^{1/2}$.
Available data on the ^{242m}Am fission cross section in the resonance energy range are shown in Fig. 7b. The ENDF/B-VII evaluation is based on the TOF data of Livermore. There is a systematical discrepancy (~20%) between these data and the TOF data of Oak Ridge. For comparison with the LSDS-100 results these data were averaged with LSDS-100 resolution function. The Japanese data (LSDS–40) [19] are in agreement with TOF data at $E > 80$ eV but at $E < 1$ eV they are closer to the Livermore data [18].
The LSDS-100 results lay between the data [18] and [20] in the energy interval E = 20 eV – 1 keV. For E > 8 keV our results follow the data of [18]. There is a visible peak at 4.5 keV in the results of Japanese measurements but it is not displayed in the other data. It needs to mention that the LSDS–40 energy resolution is lower than LSDS–100 one.

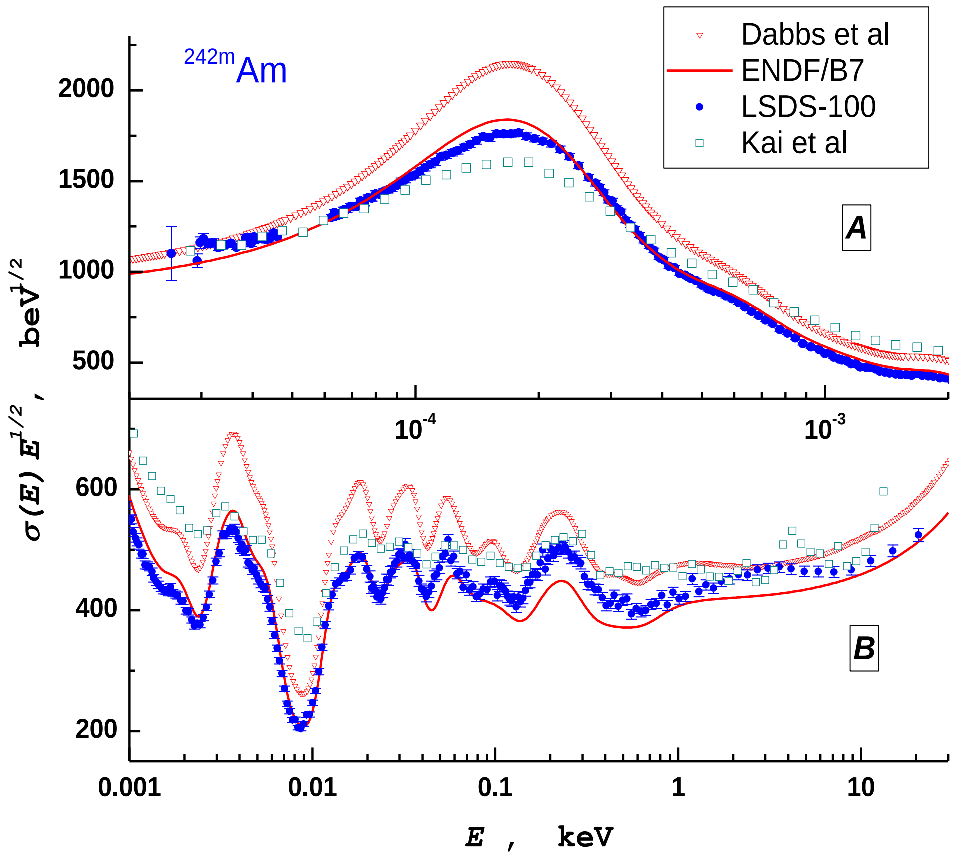

Fig. 7. The reduced fission cross section $\left\langle \sigma(E)\sqrt{E} \right\rangle$ of ^{242m}Am from 0.8 eV to 30 keV.

Designations:
● – LSDS-100 data; solid line (—) – ENDF/B-VII data; ▽ – [20]; □ – [19]. The ENDF/B-VII data and Oak-Ridge data [20] are averaged with account of LSDS–100 energy resolution.

^{243}Am [8]. The time spectrum has been measured in the range corresponding to the neutron energies from E = 0.2 eV to 11 keV. The absolute value of the fission cross section has been found by normalization to the contribution of ^{239}Pu daughter atoms admixture in the measured ^{243}Am time spectrum of fission events. The systematic error due to the normalization is equal to 4.3%. The reduced fission cross-section $\langle \sigma\sqrt{E} \rangle$ is shown in Fig. 8. The statistical accuracy is 4 - 8% for E > 0.8 eV and is about 100% a E ~ 0.3 eV.

It has been found that in the range E > 1 eV our data fall apart with the KULS data [21]. Our data show a better energy resolution than KULS value. There is a visible peak at 708 eV in the results of Japanese measurements but it is not found in our data. The background cross-section level in LSDS–100 data is noticeable lower than in KULS data.

In the range E< 60 eV the resonance structure observed in the LSDS-100 data correlates with the resonance area distribution measured in Geel [22]. Our data are much more close to the JENDL-4.0 evaluation than ENDF/B-VII ones. In the range from 20 eV to 200 eV all the recommended data are in contradiction with our data.

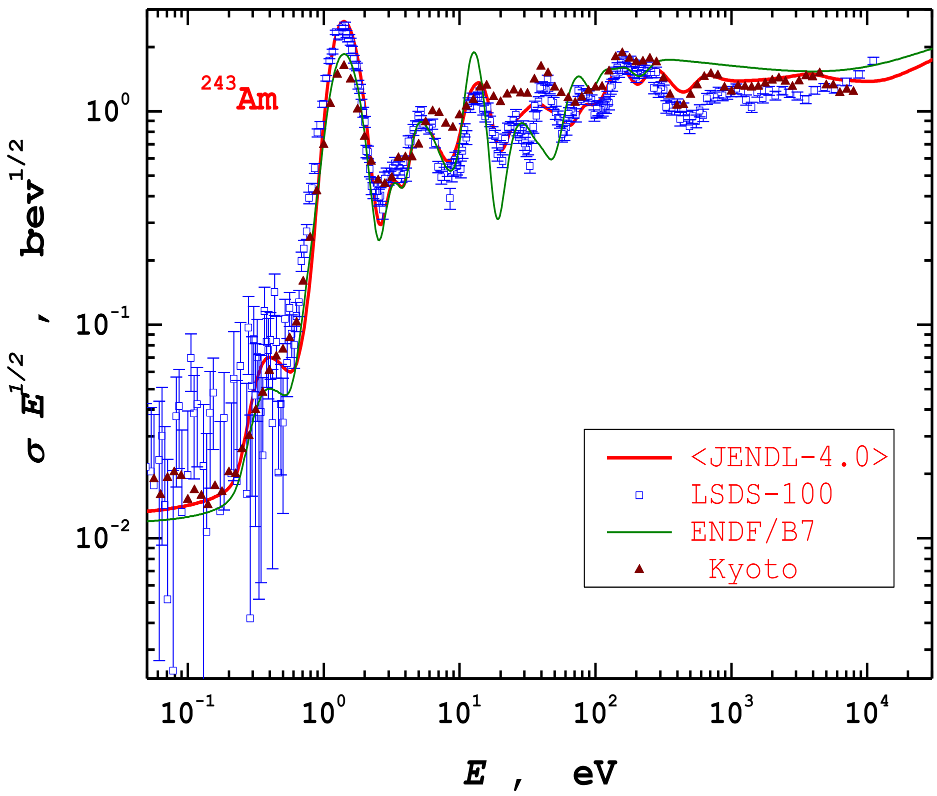

Fig. 8. Reduced fission cross-section $\langle \sigma\sqrt{E} \rangle$ of ^{243}Am vs. neutron energy E.

Designations: (□) – our data; (▲) – KULS data; solid line: thin (—) – ENDF/B-VII data and thick (—) – JENDL-4.0 data averaged with the LSDS-100 resolution function.

Fission resonance integrals

The LSDS-100 data allow calculate the fission resonance integral

$$I_f = \int_{E_1}^{E_2} \sigma_f(E)\, dE/E,$$

where $E_1 = 0.5$ eV and $E_2 = 20$ keV. The results of calculations are presented in Table 3 together with the results of reactor measurements [23, 24] and with the recommended values for the cases $E_2 = 20$ MeV and $E_2 = 20$ keV. The relative uncertainty of the reactor is not better than 10% for minor actinides.

For nuclides having a high fissility (odd nuclei) the values I_f depends slightly on the energy E_2. In the case of even-even Cm isotopes the dependence is strong enough and the values I_f falls more than two times when the upper border E_2 varies from 20 MeV to 20 keV.

The LSDS-100 data for isotopes ^{244}Cm, ^{246}Cm, ^{248}Cm show strong variations of I_f versus atomic mass (Table 3) as opposed to the reactor data [23]. Our data are in agreement with the recommended values for ^{244}Cm and ^{248}Cm. But for ^{246}Cm the recommended value (ENDF/B-VII) is ~2.8 times larger than our result.

Table 3. Fission resonance integrals I_f (b) for ^{235}U, ^{239}Pu and for minor actinides.

Target nucleus	Evaluation $E_2 = 20$ MeV	Experiment	Evaluation $E_2 = 20$ keV	LSDS-100
^{235}U	274.98	275±5 [24]	264.74	
^{239}Pu	301,4	328 ± 22 [23]	289.4	
^{241}Am	14.75 *ENDF/B-VII*	14.7±0.45 [24]		
	14.66 *ROSFOND*	27.7±1.6 [23]		
	13.3* *JENDL-4.0*	22.5±1.7 [24]		
^{242m}Am	1528 *ENDF/B-VII*	2260 ± 200 [23]	1512 *ENDF/B-VII*	
^{243}Am	7.50 *ENDF/B-VII*	8.45±0.45 [22]	2.436 *JENDL-4.0*	2.46±0.12
	7.845 *JENDL-4.0*	17.1±1.3 [24]	$E_2 = 10$ keV	$E_2 = 10$ keV
^{243}Cm	1548 *ENDF/B-VII*	1575±157 [24]	1531 *ENDF/B-VII*	1180±60
^{244}Cm	13.36 *ENDF/B-VII*	13.4 ± 1.5 [23]	5.864 *ENDF/B-VII*	6.14 ± 0.31
^{245}Cm	799.4 *ENDF/B-VII*	805 ± 80 [23]	785.3 *ENDF/B-VII*	809 ± 33
^{246}Cm	10.29	13.3 ± 1.5 [23]	4.86	1.734 ± 0.10
^{247}Cm	1130 *ROSFOND*	730 ± 70 [23]	1016 *ROSFOND*	889 ± 36
	612 *JEFF-3.1*			
^{248}Cm	10.0 *ROSFOND*	13.1 ± 1,5 [23]	3.84 *ROSFOND*	3.61 ± 0,20
	17.5 *JEFF-3.1*			
	16.32 *ENDF/B-VII*			

In the case of ^{248}Cm the low values of the full integral I_f (E_2 = 20 MeV) recommended by the ROSFOND estimate are borne out by our data. The ENDF/B-VII recommendations and the JEFF-3.1 ones are seemed too large.
Our data for ^{243}Am (in the range from E_1 = 0.5 eV to E_2 = 11 keV) are in good agreement with the JENDL-4.0 recommendation and is in contradiction with the reactor measurements [23].
For nuclides having a high fissility there are not noticeable contradictions between the results of the reactor measurements and the differential ones. In the case of ^{247}Cm the LSDS-100 data are in good agreement with the RINS data [15] and differ slightly from the LANSCE data [16]. The ROSFOND-2010 recommendation bases on the LANSCE data and is some higher than our value. The reactor data [23, 24] are slightly below them. The JEFF-3.1 estimate is seemed too low.

Resonance parameters

The resonance analysis allowed to determine the fission area $A_f = (\pi/2)\sigma_0\,\Gamma_f$ of the resonance, where σ_0 is the total peak cross section in the resonance and Γ_f is the fission width. The parameters A_f were sought by the non-linear least square method. The fit was made with the theoretic cross section as a sum of the Breit-Wigner cross sections being broadened according to the LSDS–100 energy resolution.
Our results for the resolved low-energy resonances of ^{244}Cm at 7.67 eV and 16.8 eV are in good agreement with the RINS data [11] and ENDF/B-VII evaluation. The results for 35 eV, 86 eV and 96 eV resonances match the LANL data [10]. For 22.85 eV resonance our value exceeds the bomb shot result almost at 1.5 times but this resonance is at lower limit of the energy range for the bomb shot measurements.
Our results for the resolved low-energy resonances of ^{246}Cm at 4.32 eV and 15.33 eV do not contradict to the RINS data. The difference between the data is no more than the 2σ error. The resonances at 84.4 eV and 91.8 eV are not resolved in this experiment, but their total area agrees with the LANL data [10] and with the RINS data within the limits of the 2σ error. We observed almost fully resolved resonances below 85 eV unlike the RINS data. So it seems that the hypothesis about the resonances at 33 eV and 47 eV assumed in the ENDF/B-VII evaluation failed. It will be noted that our data do not contradict to the ENDF/B-VII data in the range below 3 eV.
The areas A_f have been found for 15 low lying resonances of ^{247}Cm below 19.25 eV. For ten of them the uncertainty of the A_f value is less than 10%, for the resonances 1.23, 3.16, 4.66, and 18.0 eV it is 4,5%, for the resonances 7.94, 10.16, and 11.2 eV it is more than 25%.
Within the error limits our data are in agreement with the RINS data [15] for the resonances 9.44 and 18.03 eV, for the resonance 1.23 eV our value A_f is placed between the RINS values and the LANSCE ones [16]. For the resonance 4.66 eV our value is four times larger than the RINS value and it is some less than the LANSCE value. As soon as our data are some higher than the ROSFOND-2010 recommendation in the epithermal range so our value of the negative resonance area is found ~1.5 times larger.
The cross section analysis has been made for eight low lying resonances of ^{248}Cm below 238 eV. Within one and half error the results match the RINS values [11] and the LANL ones [10] for the resonances 26.9, 76.1, 98.95, and 237.9 eV. An excellent agreement has been found for the strong resonance at 76.1 eV.
It was found a strong discrepancy (about 18 times) between our values and the ROSFOND-2010 estimate for the resonance 7.25 eV. It will be noted that this estimate describes well the thermal point and the resonances at E=26.8 and 75 eV.

The fission resonance area A_f has been sought for 17 resonances of ^{243}Am. For five ones of them the *a posteriori* errors are less than 10%. In general our results are in agreement with the Geel values [22] and the JENDL-4.0 ones.

The analysis of the obtained values A_f gives a new information about the fission resonance width Γ_f. It was calculated by using *a priori* values of the parameter σ_0 based on the known experimental data and on the recommended estimates.

A high sensitivity of the LSDS-100 spectrometer has been confirmed by the result for the resonance 35 eV of ^{248}Cm. The value Γ_f is found to be equal 9.8 ± 5.1 μeV (the ^{248}Cm target mass is 3.6 μg) in comparison with the RINS value 70 ± 20 μeV [11] for the target mass 31 μg.

Conclusion

In conclusion it will be noted that the results obtained with the LSDS-100 spectrometer have essentially enhanced the information field on the neutron induced fission cross sections and the fission resonance parameters for the minor actinides. The results demonstrate also the level of our knowledge of the measured fission cross sections and show the need in the revision of the cross sections libraries on minor actinides.

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Resonance and Fast Neutron Induced Fission Cross Sections of Americium and Curium Nuclides

3-rd Year Progress Report RC-14485
"Resonance and Fast Neutron Induced Fission Cross Sections of Americium and Curium Nuclides"

Coordinated Research Project:
Minor Actinide Neutron Reaction Data (MANREAD)

Report was performed by research assistants of (SSC RF IPPE, Obninsk):
A.S. Egorov, B.I. Fursov, B.F. Samylin, V.S. Shorin

Engineers who take part in experiments (INR RAS, Troitsk):
A.A. Alekseev, A.A. Bergman, A.I. Berlev, E.A. Koptelov

Work Plan for the period from 2009-10-09 to 2011-09-30 (according Research Contract 14485) included:

1. Radiochemical preparation and making of fissile samples from the separated isotopes of Am-241, Am-243, Cm-247, Cm-248 in the form of thin oxide layers on Al backings and determination of the numbers of the fissile nuclei in each sample using alpha-spectrometry methods. *Performed.*

2. The multi-sectional fast fission chambers preparing, examination of detector electronics and experimental data acquisition system. *Performed.*

3. Modernization of the 100 t Lead Slowing Down Spectrometer (LSDS) based on the 209 MeV proton beam of Moscow Meson Factory (MMF): experimental investigation, calibration and numerical simulation of the LSDS characterizations. *Performed.*

4. Measurements Am-241, Am-243, Cm-247, Cm-248 fission cross sections in the neutron energy range from 1 eV to 20 keV using LSDS-100. *Performed.*

5. The experimental information processing to obtain the nuclear data – Am-241, Am-243, Cm-246, Cm-247, Cm-248 fission cross sections in 1 eV-20 keV energy range. *Performed.*

The main measured experimental results and comparison with evaluations are presented below.

Neutron–induced Fission Cross Sections of $^{241, 243}$Am and $^{246, 247, 248}$Cm isotopes

A.A. Alekseev, A.A. Bergman, A.I. Berlev, E.A. Koptelov,
A.S. Egorov, B.F. Samylin, B.I. Fursov, V.S. Shorin

The neutron induced fission cross sections of ^{241}Am, ^{243}Am, ^{246}Cm, ^{247}Cm and ^{248}Cm were measured relative to ^{239}Pu (n, f) cross section for neutron energies below 20 keV by using of the INR RAS lead slowing down spectrometer LSDS-100. The fission resonance integrals were calculated. The results have been compared with the available data.

It is known that the intensive neutron fluxes are needed to measure the fission cross sections of the nuclides of the minor actinides group. In this work the fission cross sections of ^{241}Am, ^{243}Am, ^{246}Cm, ^{247}Cm and ^{248}Cm were measured with a lead slowing down spectrometer LSDS-100 (SVZ-100) installed at the INR RAS proton Lynac (“Moscow Meson Factory”).
The experimental method has been described in the papers [1 - 8]. The proton beam parameters are following: the primary energy is equal to 209 MeV, the current pulse width is 1 – 2 µs, the repetition rate is 50 Hz, the current in the peak ~10÷14 mA. The electronics allow a time and an amplitude analysis of fission events during the time range up to 18 ms. The fission events were detected by the multi-sectional fast fission ionization chambers being placed into the operating channels of the LSDS-100 at the distance 82 cm ($^{246-248}$Cm) and at 62 cm (^{241}Am, ^{243}Am) from neutron source centre. The total operating time of measurements is about 60 h.
The fissile samples were prepared using the separated isotopes of high purity just before irradiation. The isotopic composition of the samples is listed in Table I. The mass of the fissile matter was determined on its α-activity using semiconductor spectrometer. In the measurements the ^{239}Pu(n, f) cross-section (ENDF/B-VII.0) was used as a standard.
The mass of a high purity (99.9966%) ^{239}Pu sample was 22.4 µg for Cm isotopes run and 16.3 µg for Am isotopes run. The ratio of the Cm α-activity to the ^{239}Pu one was determined with 1% uncertainty.

Table I. The masses of Cm samples and their isotopic contents.

Target	Mass, µg	Content, atom parts					
		^{240}Pu	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	^{248}Cm
^{246}Cm	13	0.00122	$9.6\cdot10^{-4}$	$1.65\cdot10^{-4}$	**1.000**	0.00138	0.001
^{247}Cm	0.335 ± 0.01	0.00745	0.0228	0.0106	0.1701	**1.000**	0.1434
^{248}Cm	3.57 ± 0.11	0.0080	0.0070	0.00023	0.0134	0.00236	**1.000**

At the measurement moment the isotopic content (at. %) of the ^{243}Am sample (4.70 ± 0.14 µg) was: ^{241}Am – 0.0045 ± 0.002, ^{242}Am – 0.0035 ± 0.002, ^{243}Am – 99.925 ± 0.003. Due to α- radioactive decay the sample included a noticeable admixture (0.0670%) of the strong fissile isotope ^{239}Pu.
The cross section values have been corrected for contribution from the isotope impurities and for the α-decay daughter nuclei in the samples under study. The mean neutron energy and the ^{239}Pu fission cross section $\langle \sigma_9(E)\sqrt{E} \rangle$ averaged on the LSDS resolution function and their time dependences were calculated in the framework of the LSDS computer model. The energy resolution of the LSDS-100 spectrometer $\Delta E/E$ is equal to 0.35 at the neutron energy E=15 eV, 0.53 at E=1.0 eV and 0.78 at E=10 keV for proton pulse width 1 µs.

^{241}Am. An attempt of the measurement with the ^{241}Am sample (6.41 µg) has been made. But during the analysis of the measured time spectra it was found that the fissile layer contains 0.6% of ^{235}U impurity atoms. Thus only qualitative results for the ^{241}Am sample were obtained. Our data show a more striking resonance structure than the KULS data and do not contradict to JENDL-4.0 cross section trend in general.

^{243}Am. The time spectrum has been measured in the range corresponding to the neutron energies from E = 0.2 eV to 11 keV. The absolute value of the fission cross section has been found by normalization to the contribution of ^{239}Pu daughter atoms admixture in the measured ^{243}Am time spectrum of fission events. The systematic error due to the normalization was considered to be constant − 4.3%. It took into account the uncertainties in the basic cross section $\langle \sigma_9(E)\sqrt{E} \rangle$ (~3%). The reduced fission cross-section $\langle \sigma\sqrt{E} \rangle$ is shown in Fig. 1. The statistical accuracy is 4 - 8% for E > 0.8 eV and is about 100% at E ~0.3 eV.

It has been found that in the range $E > 1$ eV our data fall apart with the KULS data [9]. Our data show a better energy resolution than the KULS value. There is a visible peak at 708 eV in the results of Japanese measurements but it was not found in our data. The background cross-section level in the LSDS–100 data is noticeably lower than in the KULS data.

In the range E < 60 eV the resonance structure observed in the LSDS-100 data correlates with the resonance area distribution measured in Geel [10]. Our data are much more close to the JENDL-4.0 evaluation than to ENDF/B-VII. In the range from 20 eV to 200 eV all the recommended data are in contradiction with our data.

The LSDS-100 data allow to calculate the fission resonance integral I_f from E_1 = 0.5 eV to E_2 = 11 keV. It has been found to be 2.46 ± 0.12 b that is in a good agreement with the JENDL-4.0 value (2.436 b) but is in contradiction with the reactor measurements [11].

Using the LSDS-100 data the fission resonance area A_f and the fission width Γ_f have been sought for 17 resonances by the non-linear least square method based on the Bayes approach. For five ones of them the *a posteriori* errors are less than 10%. In general our results (see Table II) are in agreement with the Geel values and the JENDL-4.0 ones.

Cm isotopes [6, 7]. The all Cm samples (Table I) contain impurities of neighbour isotopes. So their fission cross sections have been measured at the same time and the absolute value of the fission cross section has been calculated as a result of the joint analysis by solving of several equations simultaneously. The normalization constants take into account the uncertainties in the basic cross section $\langle \sigma_9(E)\sqrt{E} \rangle$ (~3%), in the Cm/Pu atom ratios and in the ratio of the Cm/Pu fission detection efficiency. The systematic uncertainty due to the normalization was evaluated as 4.0% for ^{246}Cm, ^{247}Cm and 3.4% for ^{248}Cm. The received data in tabulated form have been submitted in the EXFOR database [12].

^{246}Cm. The results of the fission cross-section measurement in the range E = 0.14 eV − 20 keV confirm the main conclusions of our earlier work on this nuclide [5]. In the present experiment the total number of detected events was about 3.3 times more than earlier. The statistical accuracy is 2.5 - 4% for E < 250 eV and in the resonance peaks. The results of the last experiment have been included in EXFOR as Entry 41533.002.

The resonance integral being calculated from the LSDS-100 data within the limits from 0.5 eV up to 20 keV was found to be equal I_f =1.734 ± 0.10 b. The ENDF/B-VII.0 value is ~2.8 times larger than our result.

^{247}Cm. The results of the fission cross-section measurement are shown in Fig. 2 in the range E = 0.03 eV − 20 keV. The statistical accuracy is 1.5 - 3% for E > 1 eV and 3 - 10% for E <1 eV. Within error limits our data are in good agreement with RINS [13] and LANSCE time-of-flight data [14]. Our results show a more striking resonance structure than the RINS data.

In the range E = 0.03 – 0.1 eV the average LSDS-100 cross section is $\langle \sigma \sqrt{E} \rangle =$ 19.0±0.4 b eV$^{1/2}$. It is some higher than the ROSFOND-2010 estimate (17.7 b eV$^{1/2}$) in the thermal point and it differs noticeably from the JEFF-3.1 recommendation (13 b eV$^{1/2}$). Note that the ENDF/B-VII.0 fission cross section is adopted in ROSFOND-2010.

A satisfactory agreement is found between our, RINS and LANSCE data and the ROSFOND estimate below E = 150 eV. For higher energies our data are found being placed below and the discrepancy is about 17% at 20 keV. But at the same time our data are in agreement with the averaged data of the Physics 8 experiment [15].

The resonance integral being calculated on the LSDS-100 data within the limits from 0.5 eV up to 20 keV is found to be equal I_f =889±36 b, this is close to the ROSFOND value -1016 b. The JEFF-3.1 (610 b) estimate seems too low.

The areas A_f have been found for 15 low lying resonances until to 19.25 eV. For ten of them the A_f value uncertainty is less than 10%, for the resonances 1.23, 3.16, 4.66 and 18.0 eV - 4.5%.

^{248}Cm. The cross sections $\langle \sigma \sqrt{E} \rangle$ are plotted in Fig. 3. The relative uncertainty of the data varies with the energy from 20 – 100% for E < 20 eV to 1% for the resonance peak at 77 eV. Further it rises up to 11% at E ~1 keV and falls away to 4% at E ~10 keV.

In the range E = 150 eV – 1 keV the LSDS-100 data are found to fall apart from the RINS data and from the ROSFOND-2010 data being broadened according to the LSDS–100 energy resolution. The Physics 8 data are in disagreement both with RINS and our data in this range. In the range 10 – 30 keV the discrepancy between the data disappears.

The resonance integral being calculated on the LSDS-100 data within the limits from 0.5 eV up to 20 keV found to be equal I_f =3.61 ± 0.20 b that agree with the ROSFOND value (3.842 b) and differ from the JEFF-3.1 and ENDF/B-VII estimates. Note that the JENDL-3.3 fission cross section is adopted in ROSFOND-2010 after a few modifications.

The cross section analysis has been made for eight low lying resonances up to 238 eV. Within one and half error the results agree with the RINS and LANL values for the resonances 26.9, 76.1, 98.95 and 237.9 eV. An excellent agreement has been found for the strong resonance at 76.1 eV.

The results demonstrate also the level of our knowledge of the measured fission cross sections and show the necessity to revise the evaluated cross sections libraries for the minor actinides.

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Table II. Fission resonance area A_f (mb•eV) of low-lying s- wave neutron resonances for ^{243}Am and their relative errors (%), δ^{ap} - *a priory* value and δ - *a posteriori* one.

E, eV	Reference			Error	
	JENDL-4.0	Geel [10]	LSDS-100	δ	δ^{ap}
-2	687.0		74.0		
0.419	23.88		23.5±5.9	25	70
0.983	46.94		196.5±12	6	70
1.356	1338	1083±14	1290±26	2	20
1.744	611.6	479±11	444.2±20	4.5	20
3.14	52.31	51.8±5.9	60.5±9.1	15	20
3.424	277.3	274.6±12.1	351.5±18	5	20
5.125	685.2	678.5±20.1	742.9±30	4	20
6.554	348.2	344.8±15.9	282.1±30	10.5	20
7.863	215.7	213.6±13	169.5±25	15	20
9.314	163.3	161.7±13	140.8±25	18	20
10.314	204.9	202.9±15.3	160.4±29	18	20
12.877	700.9	694±29.3	588.9±53	9	20
13.152	327.0	161±10.1	288.5±46	16	30
15.404	462.8	458.3±27.8	430.1±48	11	20
16.21	620.	613.9±33.4	511.6±51	10	25
21.115	165.4		189.9±51	27	80

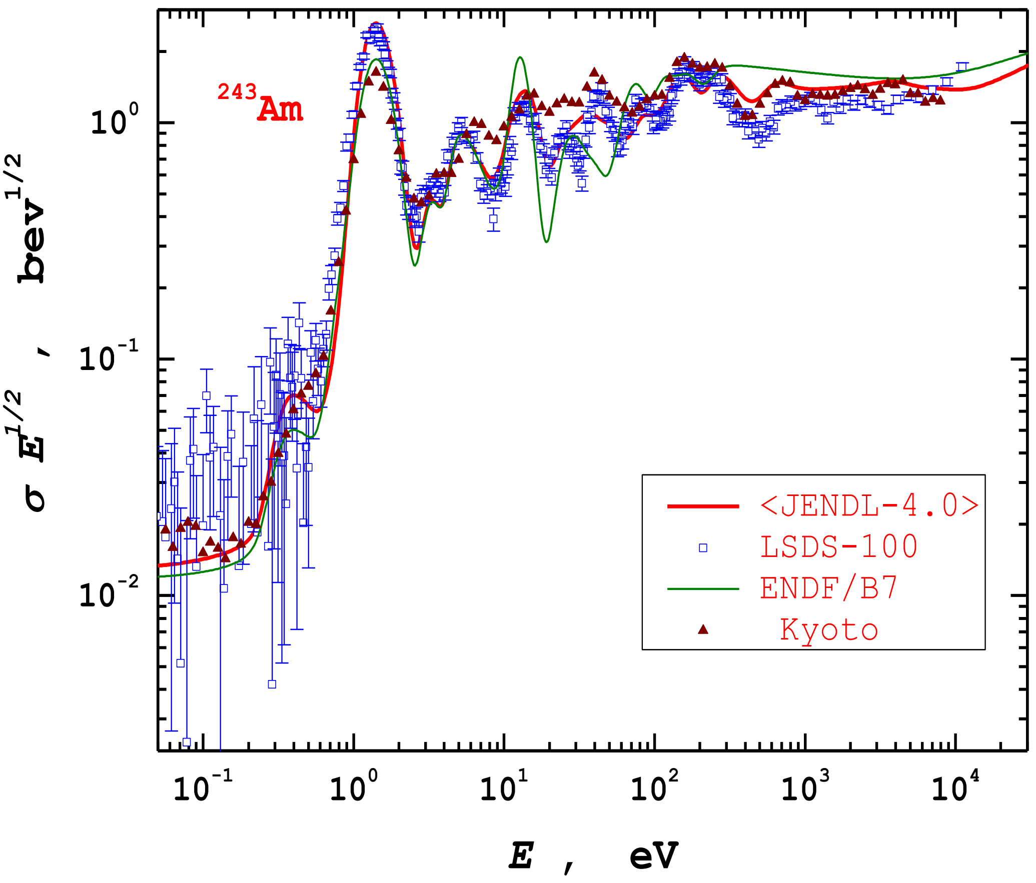

Fig. 1. Reduced fission cross-section $\left\langle \sigma \sqrt{E} \right\rangle$ of ^{243}Am versus the neutron energy E.

Designations: (∀) – our data; (▲) – KULS data; solid line: thin (—) – ENDF/B-VII.0 and thick (—) – JENDL-4.0 data averaged on the LSDS-100 resolution function.

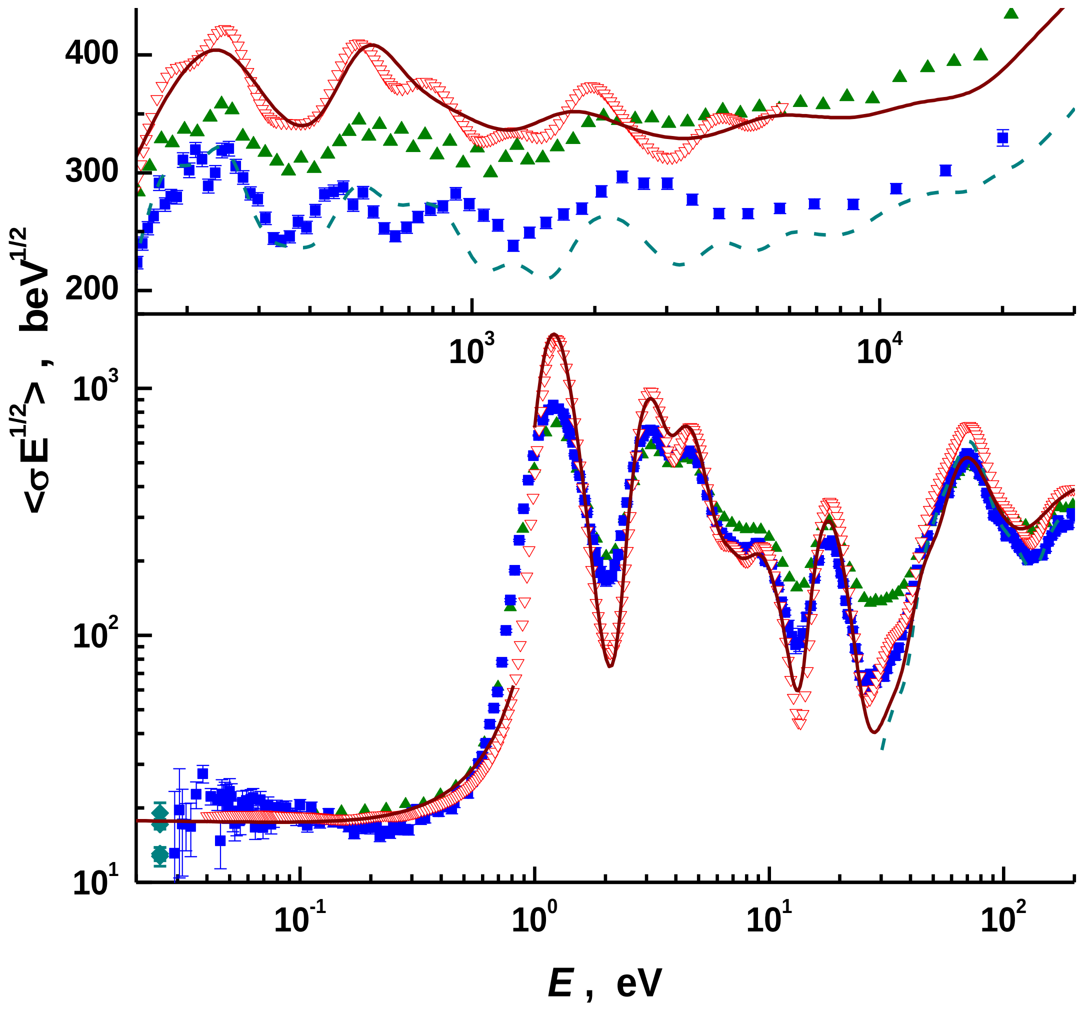

Fig. 2. Reduced fission cross section $\langle \sigma(E)\sqrt{E} \rangle$ of ^{247}Cm vs. the neutron energy *E*.

(▫) – IPPE+INR data; (▲) — RINS data; (▽) – LANSCE data; (♦) – thermal point data; solid line (—) – ROSFOND-2010 evaluation; dashed line (– –) – Physics 8 (LANL) data. The data of LANSCE, LANL and ROSFOND are averaged on the LSDS-100 resolution function.

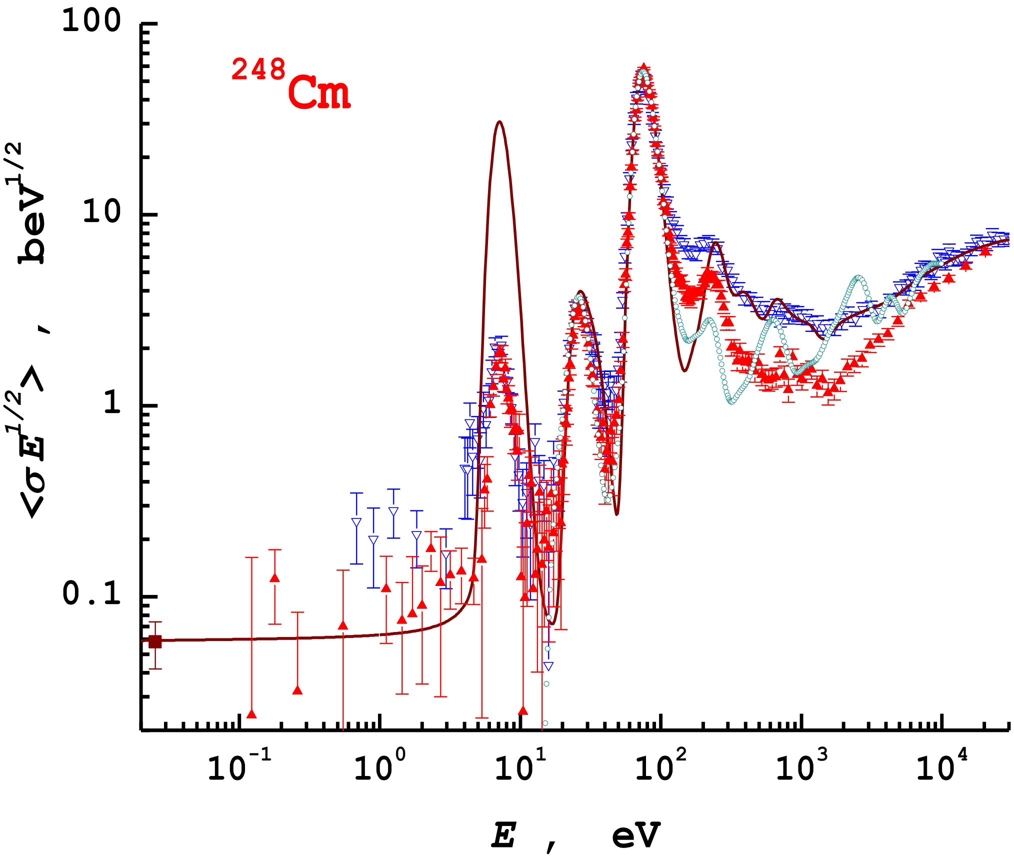

Fig.3. Reduced fission cross section $\left\langle \sigma(E)\sqrt{E} \right\rangle$ of ^{248}Cm vs. the neutron energy E.

Designations: (▲) – IPPE+INR data; (▽) – RINS data; (■) – thermal point data; (ooo) – LANL (Physics 8) data; solid line (——) – ROSFOND-2010 evaluation. The LANL data and the ROSFOND ones are averaged on the LSDS-100 resolution function.

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