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New Precision Measurements of the ^{235}U(n,γ) Cross Section

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The neutron capture cross section of ^{235}U was measured for the neutron incident energy region between 4 eV and 1 MeV at the DANCE facility at the Los Alamos Neutron Science Center with an unprecedented accuracy of 2-3 % at 1 keV. The new methodology combined three independent measurements. In the main experiment, a thick actinide sample was used to determine neutron capture and neutron-induced fission rates simultaneously. In the second measurement, a fission tagging detector was used with a thin actinide sample and detailed characteristics of the prompt-fission γ-rays were obtained. In the third measurement, the neutron scattering background was characterized using a sample of ^{208}Pb. The relative capture cross section was obtained from the experiment with the thick ^{235}U sample using a ratio method after the subtraction of the fission and neutron scattering backgrounds. Our result indicates errors that are as large as 30 % in the 0.5-2.5 keV region, in the current knowledge of neutron capture as embodied in major nuclear data evaluations. Future modifications of these databases using the improved precision data given herein will have significant impacts in neutronics calculations for a variety of nuclear technologies.

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Our present poor experimental understanding of radiative capture on fissile nuclei is caused by the difficulty in separating capture γ-rays from the large fission fragment γ-ray decay background. Current estimates of the capture cross section uncertainty in the 1 keV - 1 MeV region are as high as 15% or even more [1, 2]. Discrepancies between the nuclear data libraries ENDF/B-VII.1[1] and JENDL-4.0[2] are as large as 30% in the 0.5-2.5 keV region for ^{235}U [3]. Uncertainties of this size need to be reduced for a number of applications in nuclear technology, including design of advanced reactors. In addition, present uncertainties in capture cross sections impact our understanding of the criticality of uranium systems as well as understanding transmutation rates for ^{236}U production.

Measurements of ^{235}U capture cross sections are complicated by γ-ray background originating from neutron-induced fission. Typically, fission fragment detectors are employed to identify the neutron-induced fission reaction and remove it from the analysis of capture data. This requires actinide samples be thin enough to achieve a high fission fragment detection efficiency. On the other hand the thin sample compromises the counting statistics of the measurements. As a result, the residual spectrum remains contaminated with both prompt fission γ-rays and delayed γ-rays that follow β-decay of fission products. In addition, neutron scattering background and the backgrounds associated with the neutron beam facilities need to be subtracted. This removal of several background components results in large uncertainties and, if not performed accurately, ultimately leads to systematic errors. In this work we present the results of a new experimental approach to determine the relative neutron capture cross section for ^{235}U using the Detector for Advanced Neutron Capture Experiments (DANCE) [4, 5]. The new method combines three independent measurements to achieve a high precision in neutron capture cross section determination: *1) Measurement with a thick sample.* The neutron-induced reaction rates from thick samples are sufficiently large to obtain good counting statistics up to 1 MeV of incident neutron energy, *2) Measurement with a thin sample inside a 4π Parallel Plate Avalanche Counter (PPAC).* This allows "tagging" of all γ-rays associated with fission and thus permits a detailed subtraction of the fission contribution from the thick sample measurement. *3) Measurements of the neutron scattering background using a ^{208}Pb sample.*

A key feature of the current approach is the fact that in the experiment with the thick sample, the DANCE data for large γ-ray multiplicities M_γ>6 include only prompt-fission γ-rays from the neutron induced-fission reaction. This is because excited fission fragments populate higher angular momentum states than those formed in neutron capture and the resulting cascade of γ-rays produces a region of higher multiplicity and higher total energy release than for capture events. The neutron-induced fission reaction rates obtained in measurements with the PPAC and the thick sample for M_γ>6 are shown in Fig. 1 demonstrating the agreement within a statistical errors of experiments. Therefore, the overall fission γ-ray spectrum obtained in coincidence with the PPAC detector can be normalized to the thick target data in this "clean" γ-ray multiplicity region and then subtracted from the thick target spectrum including those regions having lower γ-ray emission multiplicity where neutron capture is present. Furthermore, using an experimental

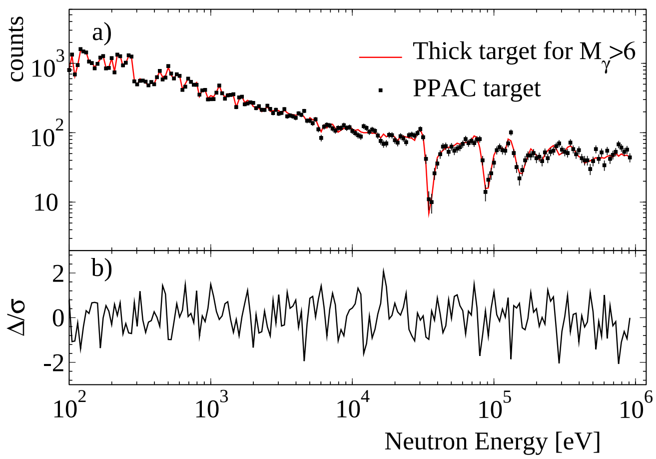

FIG. 1. (Color online) a) Comparison of ^{235}U(n,f) reaction counts from experiments with PPAC (squares) and the thick target (red line) as a function of incident neutron energy between 100 eV and 1 MeV. b) Difference of the two spectra Δ divided by the statistical uncertainty σ of the measurements.

ratio of capture-to-fission reaction rates, the neutron flux is removed from the analysis, as described next.

Capture and neutron-induced fission cross sections, $\sigma_{n\gamma}$ and σ_{nf}, at a particular neutron energy E_n can be determined from the experimental data using a following relation:

$$\sigma_{n\gamma(nf)} = \frac{M}{N_A \rho_s} \frac{N_{n\gamma(nf)}}{\varepsilon_{n\gamma(nf)} \Phi S} , \tag{1}$$

where N_A is Avogadro's number, M is the molar mass, ρ_s the areal density of the target, $N_{n\gamma(nf)}$ is the number of measured neutron capture (neutron-induced fission) events per eV per second, $\Phi(E_n)$ is the neutron flux, S is the illuminated target area and $\varepsilon_{n\gamma(nf)}$ is the total efficiency for detecting capture γ-rays (fission γ-rays) after applying data reduction cuts on the event multiplicity and the total γ-ray energy E_{sum} gated around the Q-value of the (n,γ) reaction.

If the rates $N_{n\gamma}$ and N_{nf} are measured simultaneously in one experiment, a ratio approach removes common errors. The ratio of the two reaction rates is proportional to a ratio of corresponding cross sections,

$$\alpha = \frac{\sigma_{n\gamma}}{\sigma_{nf}} = \frac{N_{n\gamma}}{N_{nf}} \frac{\varepsilon_{nf}}{\varepsilon_{n\gamma}}, \tag{2}$$

and the neutron flux is removed from the analysis completely. The rate of detected neutron capture events $N_{n\gamma}$ as a function of neutron incident energy can be described as:

$$N_{n\gamma} = N_{n\gamma}^{raw} - \varepsilon_{nfcut} N_{nf} - N_{bck}, \tag{3}$$

where $N_{n\gamma}^{raw}$ is a total rate of events for data reduction cuts that maximize the capture signal to other background components, N_{nf} is the detected neutron-induced fission rate, ε_{nfcut} represents the amount of neutron-induced fission background present in the $N_{n\gamma}^{raw}$ neutron capture rate and N_{bck} is the rate of scattering background in the $N_{n\gamma}^{raw}$ spectrum. It is important to note that detected rates N_{nf} and $N_{n\gamma}^{raw}$ are obtained using different data reduction cuts and the ε_{nfcut} variable is extracted to calculate accurately the neutron-induced fission component for the data reduction cuts that maximize the capture signal.

When using a ratio method we obtain the following relations:

$$\frac{N_{n\gamma}}{N_{nf}} = \frac{N_{n\gamma}^{raw} - \varepsilon_{nfcut} N_{nf} - N_{bck}}{N_{nf}} = \frac{N_{n\gamma}^{raw}}{N_{nf}} - \varepsilon_{nfcut} - B, \tag{4}$$

where $B = N_{bck}/N_{nf}$ is the scattering background component, and ε_{nfcut} represents the amount of fission component left in the raw neutron capture rate spectrum $N_{n\gamma}^{raw}$. Further, using equations (2) and (4), we obtain a relation for the relative neutron capture cross section:

$$\sigma_{n\gamma}^{rel} = A_{n\gamma} \frac{N_{n\gamma}}{N_{nf}} \sigma_{nf}, \tag{5}$$

where σ_{nf} is the known neutron-induced fission cross section and $A_{n\gamma}$ is a normalization constant. In this approach, σ_{nf} is assumed to be known with high precision, which is true for most of the actinides. In the resonance region, σ_{nf} needs to be accurately broadened taking into account the broadening function of the moderator and DANCE detector. The code SAMMY7 [6] is used for this purpose. Finally, the self-shielding and scattering corrections do not need to be applied when using the ratio approach. It is however important to make an estimate of the sample thickness when the scattering corrections start to play a major role, as they can cause the ratio method to fail.

We used the methodology described above to determine the capture cross section of ^{235}U. The thick-target experiment used a 26 mg/cm^2 self-supporting sample of 94% enriched ^{235}U. The thickness of the target did not invalidate the ratio approach and the scattering corrections did not influence the ratios by more than 1%. For the thin-target experiment, we used a 99.9 % enriched ^{235}U sample (130 μg/cm^2) installed inside the PPAC that provided a trigger when the neutron-induced fission reaction occurred. Preliminary results and details of the PPAC detector can be found in [7, 8]. Neutron scattering background was measured using a 99% isotope enriched ^{208}Pb sample of $\sim$ 120 mg/cm^2.

A crucial part of the analysis is the removal of the (n,f) and neutron scattering component from the thick target data to obtain the (n,γ) rates. In order to extract neutron capture rates, a specific data reduction cut has to be applied on the M_γ vs E_{sum} data to maximize the signal-to-background for the neutron capture cascades. The best results were obtained for $M_\gamma = 3 - 5$ and

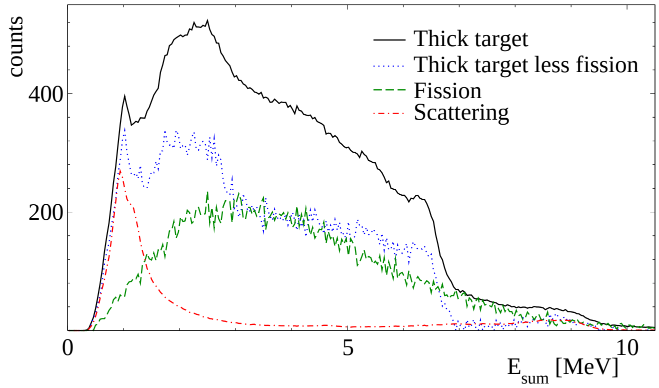

FIG. 2. (Color online) Total γ-ray energy spectra obtained for neutron incident energies E_n=0.2-10 keV and γ-ray multiplicities M_γ=3-5: black solid line - ^{235}U thick target, green dashed line - the fission background component, blue dotted line - spectrum after the fission background removal, red dot-dashed line - the scattering background normalized in the region $E_{sum} = 7.5 - 10$ MeV. (see text)

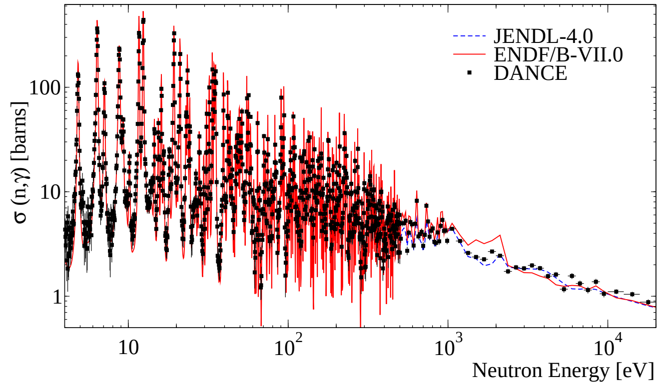

FIG. 3. (Color online) ^{235}U(n,γ) cross section measured at DANCE (black squares) compared to ENDF/B-VII.1 (red line) and JENDL-4.0 (blue dashed line) for incident neutron energies between 4 eV and 20 keV.

$E_{sum} = 5.7 - 6.7$ MeV. Applying such a cut on the thick target data leaves a portion of neutron-induced fission background in the spectra. The PPAC data, normalized to thick target data for M_γ>6, is used to determine this amount.

The scattering background was measured using the ^{208}Pb target at the same incident neutron energies as in the ^{235}U measurements. The neutron capture cross section of ^{208}Pb is very small and most of the DANCE data in this measurement originates from the scattered neutrons capturing in the materials of the DANCE array. To determine the neutron scattering background present in the thick ^{235}U target data we normalized ^{208}Pb data to rates inside the gate E_{sum}=7.5-10 MeV. More details on the scattering background subtraction can be found in [9].

The background components are subtracted for every incident neutron energy bin independently, because both (n,f) and scattering background depend on the neutron incident energy. The different components of the background that are present in the neutron capture rates spectra are shown in Fig.2 for neutron incident energies E_n=0.2-10 keV, where the total γ-ray energy spectra of different components are shown for γ-ray multiplicity M_γ=3-5. The black solid line in Fig. 2 shows the spectrum for the ^{235}U thick target, the green dashed line shows the spectrum of the fission background component, the blue dotted line shows the spectrum after the fission background removal and the red dot-dashed line shows the scattering background normalized in the region E_{sum} = 7.5-10 MeV to the spectrum shown in blue.

In order to obtain the final neutron capture cross section from the ratio $N_{n\gamma}^{raw}/N_{nf}$ we need to multiply it by the neutron-induced fission cross section. We used

	$I = \int \sigma_{n\gamma} dE_n$ [eV barns]				
E_n [keV]	DANCE	ENDF-B/VII [1]	JENDL-4 [2]	[11]	[12]
0.5 - 0.6	447±11	534[+19.5]	386[-13.7]	506	562
0.6 - 0.7	458±12	495[+8.1]	370[-19.2]	481	449
0.7 - 0.8	472±13	490[+3.8]	387[-18.0]	513	475
0.8 - 0.9	371±11	440[+18.6]	353[-4.9]	444	397
0.9 - 1.0	446±14	505[+13.2]	441[-1.1]	542	482
1.0 - 1.1	447±14	509[+13.9]	452[+1.2]	522	463
1.1 - 1.2	366±14	414[+13.1]	369[+0.8]	395	332
1.2 - 1.3	299±13	341[+14.1]	269[-10.0]	372	267
1.3 - 1.4	261±13	304[+16.5]	237[-9.2]	304	225
1.4 - 1.5	251±13	356[+41.8]	242[-3.6]	301	254
1.5 - 2.5	2312±49	3087[+33.5]	2121[-8.3]	-	-

TABLE I. Integral cross sections obtained at DANCE for neutron incident energy between 0.5 keV and 2.5 keV. ENDF-B/VII.1 and JENDL-4.0 values are shown with the deviations from DANCE results I_{EVAL}/I_{DANCE}-1 in [%]. The experimental results from [11] and [12] are shown also.

SAMMY 7.0 to obtain the optimal fit to experimental ^{235}U(n,f) data, using ENDF/B-VII.1 resonance parameters and the known broadening function of DANCE. Finally, the capture cross section is obtained from experimental data combining equations (4) and (5), where N_{nf} rates are obtained using M_γ>6 data and $N_{n\gamma}$ rates are obtained using M_γ=3-5, E_{sum}=5.7-6.7 MeV and backgrounds removal as described above in detail. A normalization constant $A_{n\gamma}$ is obtained in the region of incident neutron energies between 45 and 100 eV using ENDF/B-VII.1 data with $\int_{45eV}^{100eV} \sigma_{n\gamma} dE_n$=837.8 eVbarns.

The results are shown using black squares in Fig. 3 in the neutron energy region from 4 eV to 20 keV and in Fig. 4 for the neutron energy region from 1 keV to 1 MeV. Our results (black squares) are compared to ENDF/B-VII.1 (red line) and JENDL-4.0 (blue line). In the resolved resonance region, DANCE data agree very well (within

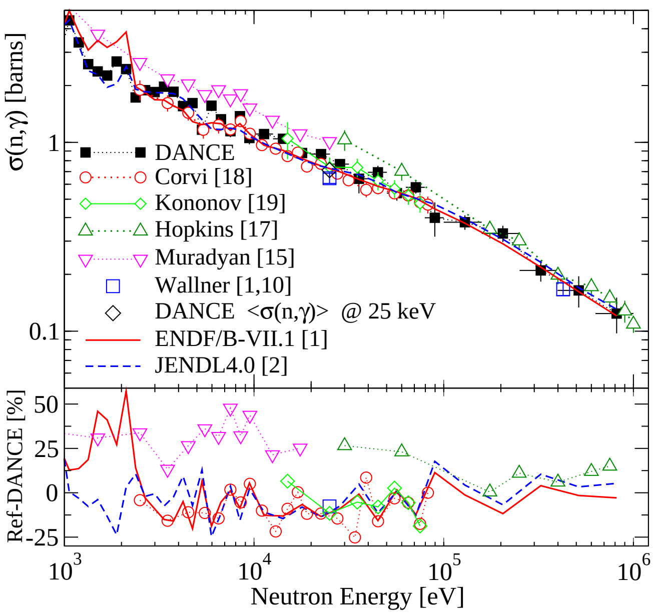

FIG. 4. (Color online) (Top)^{235}U(n,γ) cross section measured at DANCE (black squares) compared to ENDF/B-VII.1 (red line) and JENDL-4.0 (blue dashed line) and available experimental data for incident neutron energies between 1 keV and 1 MeV.(Bottom) A difference in [%] between available data and results of this work.

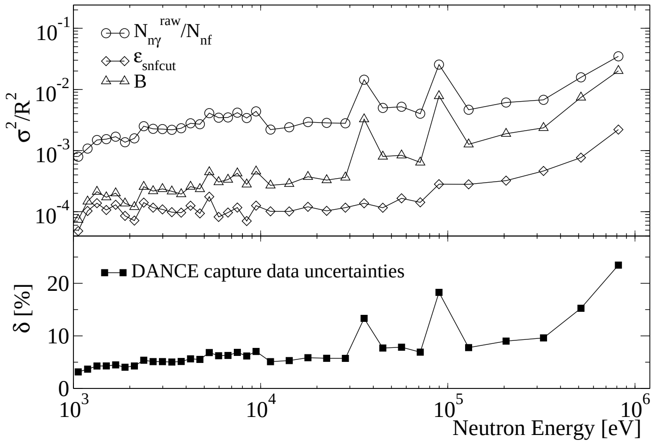

FIG. 5. (Top) A variance σ^2 of variables in equation 4 contributing to a total variance of the ratio $R = N_{n\gamma}/N_{nf}$ - see text for details. (Bottom) Relative uncertainties of ^{235}U(n,γ) cross section obtained at DANCE.

0.5%), however starting from 100 eV, deviations from evaluated data are observed. We observe that ENDF/B-VII.1 values are consistently higher than our measurement. Between 0.5 keV and 1 keV ENDF/B-VII.1 values are ∼10-15 % higher and in the interval from 1 keV to 2.5 keV ∼30 % higher. On the other hand, JENDL-4.0 is lower than our results, where the largest discrepancy of ∼20% is observed between 0.5-0.8 keV. Integral cross sections for neutron incident energy between 0.5 keV and 2.5 keV are compared to evaluations and experimental data in Table I.

Between 10 keV and 30 keV, DANCE cross sections are ∼10% larger than both ENDF/B-VII.1 and JENDL-4.0. Significant discrepancies are observed among other measurements. Neutron flux at DANCE is attenuated by Al material in the beam at 35 keV and 80 keV and as a result, increased uncertainties (up to 20 %) are observed in our data at these energies. Above 100 keV, experimental data and evaluations agree well with the DANCE results. Finally, a recent activation measurement of Wallner et al. [10] using an Accelerator Mass Spectrometry (AMS) technique (empty squares in Fig. 4) is compared to DANCE results. Because of the integral nature of the AMS result, we calculated a weighted average of our cross section over the pseudo-Maxwellian neutron flux provided by Wallner [10]. Our integral cross section of 0.70±0.06 is to be compared with 0.646±0.040 obtained in [1, 10]. An insufficient neutron energy resolution in our data prevents a comparison to the 426 keV result obtained by AMS.

The uncertainties of all experimental variables in equations(4)-(5) are propagated using the standard error propagation formulas. Variances of $(N_{n\gamma}^{raw}/N_{nf})$, ε_{nfcut} and B, from equation(4) are shown in Fig.5(top) relative to the ratio squared $R^2 = (N_{n\gamma}/N_{nf})^2$. For the final capture cross section uncertainty determination in equation (5), we include systematic uncertainties of 0.5 % and 1 %, for the normalization constant $A_{n\gamma}$ and ENDF-B/VII.1 neutron-induced cross section σ_{nf}, respectively. The relative experimental uncertainties of the data extracted in this work are shown in Fig.5(bottom).

In summary, a new experimental method was developed that enabled the determination of the rates of neutron capture and neutron-induced fission simultaneously in an experiment using a thick 26 mg/cm^2 ^{235}U target and the DANCE detector array. Neutron-induced fission and neutron scattering backgrounds were accurately subtracted using data from two independent measurements, one using a thin ^{235}U sample inside a 4π PPAC fission tagging detector, and one with a ^{208}Pb sample, respectively. The high precision relative capture cross section of ^{235}U was obtained in the incident neutron energy region between 4 eV and 1 MeV. and the cross section was normalized to ENDF/B-VII.1 in the neutron incident energy region from 45 eV to 100 eV. Significant discrepancies as large as 30 % are reported, especially in the region between 1 keV and 2.5 keV. Our new measurement has significantly improved the knowledge of neutron capture cross section on ^{235}U. We expect that our measurement technique can be applied at other laboratories, for example by CERN, and Rensselaer Polytechnic Institute, who are striving to measure actinide neutron capture cross sections accurately. A more detailed report on our work is in preparation. This report also expands on other aspects of our measurement that have led to unique data,

for example the total γ-ray energy spectra for fission and for capture γ-rays shown in Fig. 2. This work benefited from the use of the LANSCE accelerator facility. This work was performed under the auspices of the U.S. DOE at Los Alamos National Laboratory by the Los Alamos National Security, LLC under Contract No. DE-AC52-06NA25396 and at Lawrence Livermore National Laboratory by the Lawrence Livermore National Security, LLC under Contract No. DE-AC52-07NA27344.

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[1] M.B. Chadwick at al., Nucl. Data Sheets, 112, Issue 12, 2887-3152 (2011)
[2] K. Shibata et al., J. Nucl. Sci. Technol.. 48(1), 1-30 (2011)
[3] P. Talou et al., Nucl. Data Sheets, 112, 3054 (2011)
[4] M. Heil et al., Nucl. Instrum. Methods A**459**, 229 (2001)
[5] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, Nucl. Sci. Eng. **106**, 208 (1990)
[6] N. M. Larson, Oak Ridge National Laboratory report ORNL/TM-9179/R7 (2006).
[7] T. A. Bredeweg et al., Proc. of the Int. Conf. on Nucl. Data for Science and Technology 2007, Vol 1, 2008, p.607
[8] M. Jandel et al. , Proc. of the 4th Int. Conf. on Fission and Properties of Neutron-Rich Nuclei, 2007, Sanibel Island, Florida, World Scientific, p. 440
[9] M. Jandel et al., Phys. Rev. C**78**, 034609 (2008)
[10] A. Wallner et al., J. Korean Phys. Soc. **59**, 1410 (2011); Private Communication (2012)
[11] G. de Saussure, R. Gwin, L. W. Weston, and R. W. Ingle, ORNL/TM-1804, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory, Oak Ridge, TN (1967)
[12] R. B. Perez, G. de Saussure, and E. G. Silver, Nucl. Sci. Eng. 52, 46 (1973)
[13] L. C. Leal, H. Derrien, N. M. Larson, R. Q. Wright, ORNL/TM-13516, Lockheed Martin Energy Research Corp., ORNL, Oak Ridge, TN (1997)
[14] J. M. Wouters et al., IEEE Transactions on Nucl. Sci., Vol. **53**, No 3., 880 (2006).
[15] G.V.Muradyan et al., 1980 Kiev, Vol.2, p.119 (1980)
[16] G.de Saussure et al., 1966 Paris, Vol.2, p.233(48)(1966)
[17] J.C.Hopkins et al., Nucl. Sci. Eng., 12, 169 (1962)
[18] F.Corvi et al., NEANDC(E)-232, 5 (1982)
[19] V.N.Kononov et al., At. Energy, 38, 81 (1975)