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Analysis and Evaluation of Experimental Data on the Value of Alpha for Plutonium-239

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ANALYSIS AND EVALUATION OF EXPERIMENTAL DATA ON THE VALUE OF ALPHA FOR PLUTONIUM-239

V.N. Kononov and E.D. Poletaev

ABSTRACT

Analysis and evaluation of experimental data on the value of alpha for plutonium-239. Experimental data on the value of alpha for plutonium-239 (microscopic experiments) in the range of neutron energies from 0.1 to 1000 keV are used to obtain estimated values for alpha and the covariance matrix.

Introduction

The neutron radiative capture cross-section for plutonium-239 is one of the most important constants for making calculations on the reactor physics and engineering economics of fast-neutron reactors. In the majority of microscopic experiments, however, the measurements give the value of alpha - the ratio of the radiative capture to the fission cross-sections. In the determination of the group cross-sections from the results of microscopic experiments, therefore, the value of α is usually calculated, and the capture cross-section is then obtained from a combination of the estimated values of α and the fission cross-section. The value of α for plutonium-239 has been estimated in the well-known work by Sowerby and Kon'shin [1] and Kon'shin et al. [2], carried out in 1971 and 1973. Since then, the list of experimental papers has grown considerably and it therefore seems desirable to make a new evaluation of this quantity from all the experimental data which have been published up to the present time [3-25]. In our analysis of the experiments and evaluation of the data, we have considered it important to calculate not only the errors in the estimated values but also their correlation properties - an aspect which has hardly been considered in previous estimates of the value of α.

Methods for measuring the value of α and the principal sources of error

The experimental work on the measurement of α can be divided into three major groups:

(1) Experiments with resonance-neutron spectrometers, using different types of pulsed neutron source and a moderator;

(2) Experiments with pulsed electrostatic generators;

(3) Experiments with quasi-monoenergetic neutron sources which have an intensity which is constant in time.

The division is based both on the region of neutron energies in which each group is used and on the general methodological features which are responsible for the systematic errors. At the same time, all the experimental methods for measuring the value of α have a number of common characteristics and it is these which we shall discuss first.

None of the reported experiments measures α directly: instead, they determine the quantity $\gamma = N_\gamma/N_f$, which in the simplest case is connected with α by the relationship

$$A_\gamma \simeq \alpha + B,$$

where N_γ and N_f are the measured count rates in the channels which record the capture and fission gamma-rays and the fission events; and A and B are constants of the measuring equipment. Thus the accuracy in the measurement of α depends very strongly on the absolute value of the constant B associated with the method. This varies between 0.3 and 2.5 in the different experiments and essentially characterizes the sensitivity of a given method to the value of α. It can easily be seen that, on the one hand, an error in determining B leads to a systematic error in the final value of α which increases rapidly as the absolute value of α gets smaller (as is typical of the $\alpha(E_n)$ behaviour for plutonium-239 at neutron energies above ~10 keV). On the other hand, the error in the value of α which arises during the reduction of the raw spectra (subtraction of the background, for example) and from other effects, increases approximately as $(1 + B/\alpha)$.

The value of the constant B is also a determining factor in the choice of a method for obtaining the absolute value of α. For values of B ≃ 0.3, such as were achieved in the three experiments with pulsed electrostatic generators [15-19], the use of a high-efficiency technique for recording fission events by means of a scintillation tank loaded with cadmium or gadolinium made it possible to get absolute measurements of α to the acceptable accuracy of ~10%.

In most other experiments, the measurements are relative ones and the equipment constants are determined by normalization to "reference" quantities. These reference quantities are the values of α for a few well-resolved resonances [3, 4, 7-9, 14], the fission and absorption cross-sections in the range 0.05-0.4 eV [10-12], and the values of α for thermal-neutron spectra [6, 13, 25] and in the fast-neutron region at E_n = 30 keV [20, 21]. In some papers the equipment constants were measured partly by experiment. These normalization methods have of course both advantages and disadvantages. Thus, when the results are normalized to resonance values of α, a wide range (from 0.5 to 9) is encompassed, in spite of their relatively low accuracy. However, there is a danger here that the difference in the resonance blocking effects between the normalization region and the region where the principal measurements are made and the degree of correctness with which this factor is taken into account, can lead to additional systematic errors. This danger is particularly acute, for example, in Ref. [8], where a "thick" sample was used in the gamma-ray recording channel and a thin sample of plutonium was used in the fission chamber of the fission channel. Normalization to the epithermal region, which was used in the work of the Oak Ridge Group [10-12], leads to highly-reliable reference cross-section, but covers a significantly narrower range of α values (0.4-0.6). This disadvantage also extends to experiments where the normalization uses the value of α in the thermal-neutron spectrum.

It should be noted that whenever relative measurements are normalized, it is assumed that the efficiency of the detector system is constant over the entire range of neutron energies investigated. However, this important condition is not satisfied in all experiments for measuring α since some of the detector systems used are sensitive, for example, to changes with neutron energy in the properties of the capture and fission processes (the capture and fission gamma-ray spectrum, the total energy of the fission gamma-rays, the average number of fission neutrons, and so on) used to record the capture and fission events.

For gamma-ray detectors these difficulties are most important in systems which are based on the use of small-volume NaI(Tl) and stilbene crystals [6, 9, 14, 20, 21, 25], whose efficiency is proportional to the multiplicity of gamma-rays and hence is extremely sensitive to changes in the capture and fission gamma-ray spectra. Similar problems also arise in the use of large liquid scintillation detectors employed under coincidence conditions between the two halves of the detector [8, 10-12] and also with an increased recording threshold in total-energy detectors [5] and with screening of the sample by lead to improve the effect-background ratio.

In experiments where the fission events are recorded in terms of the fission neutrons [3, 5-7, 14-19, 21, 25], it is possible to get a sensitivity to changes in $\bar{\nu}$ with the energy of the incident neutrons. We may note that this sensitivity is strongest when use is made of small-volume detectors [3, 5, 6, 14, 21, 25] in which $\varepsilon_f \sim \bar{\nu}$ and variations in $\bar{\nu}$ have a direct effect on the result.

In addition to these factors, the efficiency can also vary with the neutron energy as a result of the sensitivity of the detectors and recording equipment to overloading. This makes it necessary to take special precautions [17] or to introduce appropriate corrections (which might be quite large; ~13% in [12]).

In many experiments for measuring α, quite large samples of plutonium have been used. This means that the necessary corrections have to be made to the results; the corrections include those for resonance self-screening and multiple scattering of neutrons in the sample, for absorption of the capture and fission gamma-rays in the sample, for neutron multiplication in the sample, and so on. In a number of experiments it is important to allow for the correlation in the direction of emission of the fragments and neutrons resulting from fission and the variation of the average number of neutrons with the energy of the incident beam. All this leads to additional systematic errors, and the importance of most of the corrections increases as α decreases so that the systematic errors related to difficulties in the methods of introducing the corrections turn out to depend on the neutron energy.

A simple consideration of the main characteristics of the methods for measuring α for plutonium-239 shows that all the experiments so far carried out are indirect and require from the authors a comprehensive analysis of the experimental conditions and the introduction of a large number of corrections. In this connection, an important criterion of the reliability of the data in any particular work is the presence of a detailed analysis of the experiment and of the errors, and evidence that a number of independent measurement cycles have been made, preferably with a change in the experimental conditions.

We shall now consider some special features of the three groups of experiments for measuring α.

Quite a large number of experiments have already been carried out on resonance-neutron spectrometers, covering the range of neutron energies from 0.1 keV to several tens of keV and recently up to 200 keV. In all these experiments, the most important problem is that of allowing for the background in the gamma-ray recording channels; in our opinion, this problem has not yet been fully treated. The background in a gamma-channel, apart from that due to radioactivity and cosmic radiation, is due to an effect which cannot in principle be eliminated, namely the scattering of neutrons in the sample itself. The special feature of resonance-neutron spectrometers is that only a small fraction ($\sim 10^{-2}$) of the neutrons which intersect the sample come within the range of neutron energies which is of interest. By far the largest fraction of the neutrons, namely the fast neutrons, constitute the principal source of the varying background - which is most difficult to determine.

The varying background has two components: the instantaneous background whose recording coincides in time with the passage of the relevant neutrons through the sample, and the delayed background which is recorded by the detector a certain time (the "wander" time) after the corresponding neutrons have passed through. The relation between the components is different for different types of detector, but we can state that in the majority of experiments with neutron energies above ~1-5 keV, the total varying background forms more than 50-80% of the total count in the gamma-ray recording channel. At first sight this is an unexpected result, but small-volume detectors have a varying-background level which is even higher than that for large liquid scintillation detectors [11].

The accepted procedure for measuring the varying background by means of "black" resonance filters does not allow the instantaneous background to be observed and does not give a true picture of the delayed background since the placing of a filter in the neutron beam leads to an attenuation of the fast-neutron flux which is the main source of the background. The determination of the background by extrapolation to zero filter thickness is incapable of giving sufficient accuracy. In our opinion the only way to get a correct picture of the delayed background is to carry out the experiment with filters placed constantly in the neutron beam. The instantaneous component of the background can be measured to an acceptable degree of accuracy in experiments which use a scatterer which is approximately equivalent to the sample, provided that resonance filters are placed constantly in the beam. Unfortunately, it does not appear from the description of the experimental procedures that this simple technique has been used in the measurement of α for plutonium-239.

In addition to the effects related to the scattering of fast neutrons in the sample, a varying background in resonance-neutron spectrometer measurements of α also arises from the delayed fission gamma-rays produced in the sample by fast neutrons and from gamma-rays from the decelerating target. These effects further complicate the analysis of the experimental results and also lead to considerable overloading of the detectors.

In view of the relatively low sensitivity of the majority of the methods used (high value of the constant B), these difficulties in making an exact determination of the varying component of the background in resonance-neutron spectrometer experiments at energies above ~1-5 keV probably represent the principal cause of the large systematic errors in this range of neutron energy measurements and produce the large discrepancy (up to ~50%) observed in the data obtained by this method for $E_n > 5$-10 keV.

Specially important amongst the studies carried out on resonance-neutron spectrometers is that by Muradyan et al. (IAE) [9] and Gwin et al. (Oak Ridge) [12]. In these experiments, the sample was in the form of layers of plutonium in a fission chamber and the capture events were separated by anti-coincidence with the fission events recorded by the fission chamber. Thus, in addition to the use of a thin sample, there is the possibility in these experiments of achieving a small value of B. However, as a result of the low intensity of the neutron source at IAE, it was not possible in Ref. [9] to realize the full possibilities of the experimental arrangement. In Gwin's experiments, which were carried out over a number of years on the OPELA accelerator, it proved possible to extend considerably the measured range of neutron energies, but unfortunately the constant B was still quite high ($B \simeq 0.7$) because of the low efficiency of the fission chamber.

Two types of measurement have been made on pulsed electrostatic generators: absolute measurements which use a large liquid scintillation detector [15-19] and relative measurements with small-volume detectors [20, 21]. In contrast to the resonance-neutron spectrometer experiments, the value of the true background is determined directly during the actual measurements. Moreover, as a result of the high repetition frequency of the neutron pulses in comparison with the neutron lifetime in the detector, the delayed background, which is here the major component, is largely averaged out and varies very little over the entire range of neutron energies. Thus, despite the high background level of the liquid scintillation detectors, associated with the high efficiency of the neutron and gamma-ray detection - the principal feature from the point of view of absolute measurements - it is possible to get an acceptable accuracy in the value of α.

It should be noted that experiments for the absolute measurement of α by means of large liquid scintillation detectors [15-19], which are based on a common principle but differ appreciably in their actual realization, show good agreement amongst themselves, except for the point at $E_n = 64 \pm 15$ keV in Ref. [15].

Pulsed electrostatic generator measurements of the relative value of α, in contrast to those with large liquid scintillation detectors, can be carried out over short flight paths. It has therefore been possible to make measurements in the range of neutron energies ~5-10 keV.

The third group of experiments (with quasi-monoenergetic neutrons) includes those based on a spherical geometry with photo-neutron sources [22-24] and relative measurements involving normalization to the thermal region carried out in filtered reactor beams [25]. The derivation of values for α from experiments with a spherical geometry involves certain difficulties connected with the fact that it is necessary to take into account the resonance nature of the cross-sections in considering the passage of the neutrons through the fairly thick spherical samples which are used. These measurements of α with filtered beams involve the adjustment of the results to the thermal region and so the data in Ref. [25] give important additional information on the value of α in the fast-neutron region, although for E_n = 2 keV the interpretation of these data requires an exact knowledge of the neutron spectrum of the source because of the detailed structure in α.

We have considered the most important features - together with their associated errors - in the existing methods for measuring α. Taken together, these determine the experimental accuracy in deriving the value of α. It can be seen from what we have said that, in addition to the statistical accuracy and the random errors (for which there exists a standard and formalized evaluation technique), there are also other errors, which are inherent in all the methods and which are, in fact, the determining factors. These are the errors connected with subtraction of the background, normalization, equipment problems and the introduction of different types of correction. These errors are of a systematic nature. Unfortunately, no general approach towards an objective estimate of these errors has been developed. It is possible therefore that the authors of the majority of the experimental papers do not pay proper attention to this very important problem in the treatment of the estimated data. As a result, in the reduction of the estimated data and of the errors and their correlation properties, we have been guided by the ideas we have developed about the various experimental methods, and this fact is to some extent reflected in the preceding analysis.

Evaluation of the data on the value of α for plutonium-239 in the range of neutron energies from 0.1 keV to 1 MeV

Neutron energies from 0.1 to 10 keV

The best work, from the point of view of experimental design and the level of experimental technique, is that of the Oak Ridge group [12]. We have therefore used their data as the basis of the estimated curve in this energy range. The data from all other sources has been given an overall "weight" of one half.

Neutron energies near 30 keV

By using the data from three absolute measurements [15, 16, 19] and also from two papers by the Oak Ridge group [11, 12], we have obtained an average value $<\alpha>_{30}$ for the neutron-energy range 20-40 keV of $<\alpha>_{30} = 0.315$. The average was derived by means of "weights" which were inversely proportional to the square of the quoted errors: the exception was Ref. [15], where the "weight" was reduced. The value of $<\alpha>_{30}$ includes the renormalized data from the relative measurements on pulsed electrostatic generators [20, 21] with allowance for the special features of each experiment. We considered it unjustified to renormalize the results of the absolute measurements as was done, for example, in the estimate in Ref. [2].

Neutron energies from 10 to 100 keV

The estimate in this region used the data from the absolute measurements of Refs [15, 16, 19], the renormalized data from the relative measurements of Refs [20, 21] and also the data from the Oak Ridge group [11, 12]: the "weight" of the last two results for neutron energies above 50 keV was reduced because of the possible large error in the measurement of the background level. The estimate took into account the results of filtered beam-experiments [25].

Neutron energies from 0.1 to 1 MeV

The estimate here is based on the results obtained with pulsed electrostatic generators for absolute and relative measurements, and the results of experiments on filtered beams.

The results for the range 0.1 keV-1 MeV are given in Table 1; Fig. 1 shows all the experimental data for energies above 10 keV which were used in deriving the estimate. A comparison of the present results for α in plutonium-239 with those of other estimates known to us [1, 2, 26, 27] is given in Fig. 2. Satisfactory agreement can be seen overall and also from the comparison of the values of $<\alpha>$ averaged over the core spectrum of the Baker reactor [28] (Table 2). However, in the region $E_n > 50$ keV there is an appreciable difference between our estimate and that of ENDF/B-IV [26], of the Japanese [27] and of Sowerby and Kon'shin [1].

Figure 3 shows the capture cross-sections for plutonium-239 derived from the various estimates. The data in the present paper were obtained by a combination of our estimated values of α and the σ_f estimated by Sowerby et al. [29]. As expected, the observed difference in the estimates of α are fully reflected in the radiative-capture cross-section values. The wide maximum and the wave structure in $\sigma_{n\gamma}(E)$ for E_n = 100-400 keV are not characteristic of the behaviour of capture cross-sections in heavy odd nuclei. The theoretical curve obtained by Lynn [30] is also shown in Fig. 3 and in the main it confirms the tendency for a smooth change in the capture cross-section with energy that we have derived.

Table 3 gives the estimated values of α in the representation of the 26-group system of constants [31] and also the cross-section $\sigma_{n\gamma}$ obtained from the use of data on the fission cross-section from the estimate in [29].

From a study of the experimental conditions, we have analysed and evaluated the various non-correlating components of the errors in the estimated values of α. For neutron energies above ~10 keV we have used as our basis the structure of the experimental errors in the absolute measurement of α with a pulsed electrostatic generator. This structure has been analysed in some detail in [19]. For lower energies we have taken the information on the error structure obtained from an analysis of the work of the Oak Ridge group [19]. We have thus obtained a correlation matrix for the errors in the estimated values of α. This matrix, together with the total estimated error, is given in Table 3. From this matrix we have obtained the error in the value of $\langle \alpha \rangle$ averaged over the spectrum of the standard reactor. This is given in Table 2. For purposes of comparison, this Table also shows the error in $\langle \alpha \rangle$ obtained on the assumption that the error correlation matrix is diagonal (denoted by *).

In order to get the covariance matrix of the radiative capture cross-section, we also have to know the covariance matrix of the estimated values of the fission cross-section. However, since the error in α represents the main contribution to the error in the capture cross-section, the correlation matrix given in Table 3 presumably gives an adequate picture of the main features of the correlation matrix for the capture cross-section.

In conclusion, the authors wish to thank A.I. Voropaev for useful discussions.

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Table 1

Estimated values of the quantity α for plutonium-239 in the neutron energy range from 0.1 keV to 1 MeV

E_n, keV	α	E_n, keV	α
0,I - 0,2	0,863	20 - 25	0,369
0,2 0,3	0,936	25 - 30	0,3I7
0,3 - 0,4	I,I6I	30 - 35	0,300
0,4 - 0,5	0,50I	35 - 40	0,274
0,5 - 0,6	0,736	40 - 45	0,258
0,6 - 0,7	I,497	45 - 50	0,24I
0,7 - 0,8	0,973	50 - 60	0,209
0,8 - 0,9	0,8I8	60 - 70	0,I83
0,9 - I,0	0,724	70 - 80	0,I68
I - 2	0,880	80 - 90	0,I62
2 - 3	I,020	90 - I00	0,I55
3 - 4	0,779	I00 - 200	0,I34
4 - 5	0,850	200 - 300	0,I08
5 - 6	0,825	300 - 400	0,093
6 - 7	0,787	400 - 500	0,082
7 - 8	0,624	500 - 600	0,07I
8 - 9	0,547	600 - 700	0,060
9 - I0	0,563	700 - 800	0,050
I0 - I5	0,547	800 - 900	0,040
I5 - 20	0,4I2	900 - I000	0,030

Table 2

Comparison of the estimated values of $<\alpha>$ for plutonium-239 averaged over the neutron spectrum of the standard Baker reactor (without allowance for resonance blocking)

Paper	$\langle\alpha\rangle$	Ratio to $<\alpha>$ from this paper
BHAB-70 [28]	0,300	I,007
Sowerby-Kon'shin [1]	0,307	I,030
Kon'shin [2]	0,303	I,0I7
ENDF/B-IV [26]	0,304	I,020
ENDF/B-III [12]	0,299	I,003
Present paper	$0{,}298 \pm \begin{cases} 0{,}020 \\ 0{,}009 \end{cases}$ *)	

Table 3

Group values of the quantity α and of the radiative capture cross-section for plutonium-239

i	E_n, keV	α	Correlation matrix of the errors in α													$\sigma_{n\gamma}$, barn
5	800-I400	0,029±0,008	I													0,05±0,0I
6	400-800	0,068±0,0I2	0,93	I												0,II±0,02
7	200-400	0,I02±0,0I3	0,90	0,89	I											0,I5±0,02
8	I00-200	0,I34±0,0I5	0,87	0,87	0,87	I										0,20±0,02
9	46,5-I00	0,I84±0,02I	0,70	0,70	0,7I	0,7I	I									0,30±0,04
I0	2I,5-46,5	0,304±0,028	0,36	0,38	0,42	0,42	0,48	I								0,49±0,05
II	I0-2I,5	0,483±0,045	0,27	0,29	0,3I	0,29	0,33	0,36	I							0,84±0,08
I2	4,65-I0,0	0,7I4±0,064	0,28	0,30	0,32	0,30	0,32	0,36	0,45	I						I,47±0,I4
I3	2,I5-4,65	0,904±0,075	0	0	0	0	0,I4	0,3I	0,49	0,46	I					2,63±0,24
I4	I,0-2,I5	0,889±0,073	0	0	0	0	0,I4	0,3I	0,49	0,46	0,87	I				3,7I±0,33
I5	0,465-I,0	0,827±0,068	0	0	0	0	0,I4	0,3I	0,49	0,46	0,87	0.87	I			6,89±0,6I
I6	0,2I5-0,465	0,930±0,077	0	0	0	0	0,I4	0,3I	0,49	0,46	0,87	0,87	0,87	I		I2,0±I,06
I7	0,I00-0,2I5	0,868±0,072	0	0	0	0	0,I4	0,3I	0,49	0,46	0,87	0,87	0,87	0,87	I	I6,4±I,45

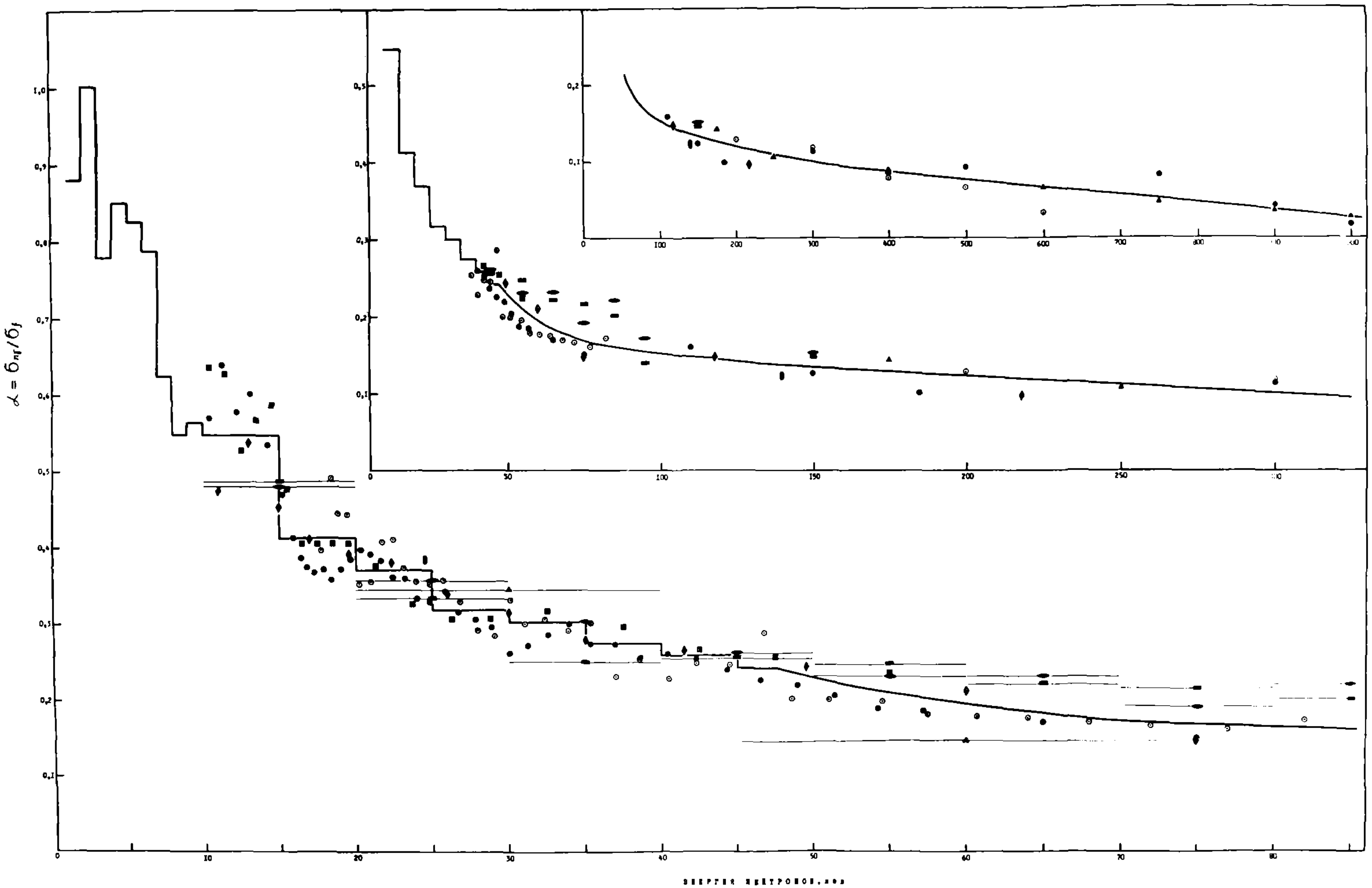


Fig. 1.: Energy dependence for α for plutonium-239.

- [11]; - [12]; - [15]; - [16]; - [19]; - [20]; - [21]; - [25];

– estimate from present paper.

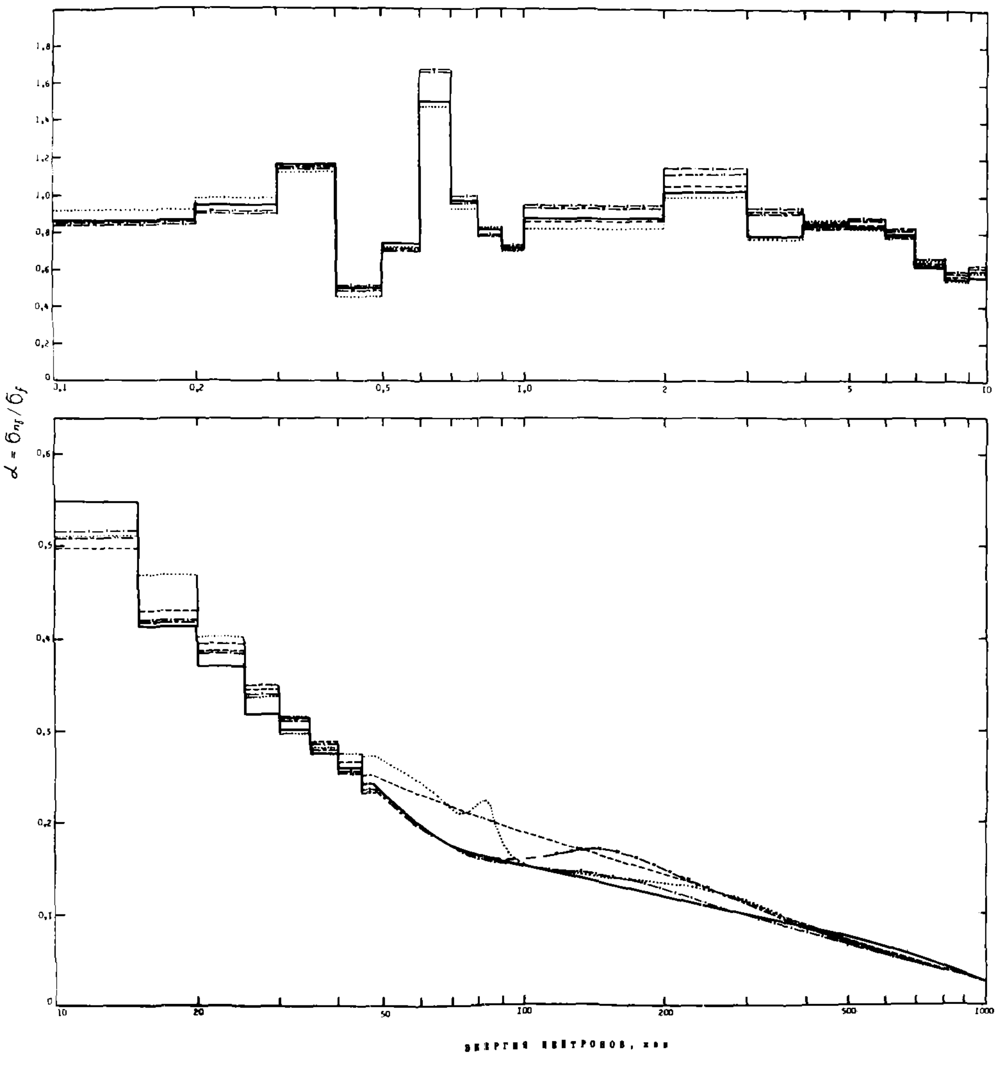
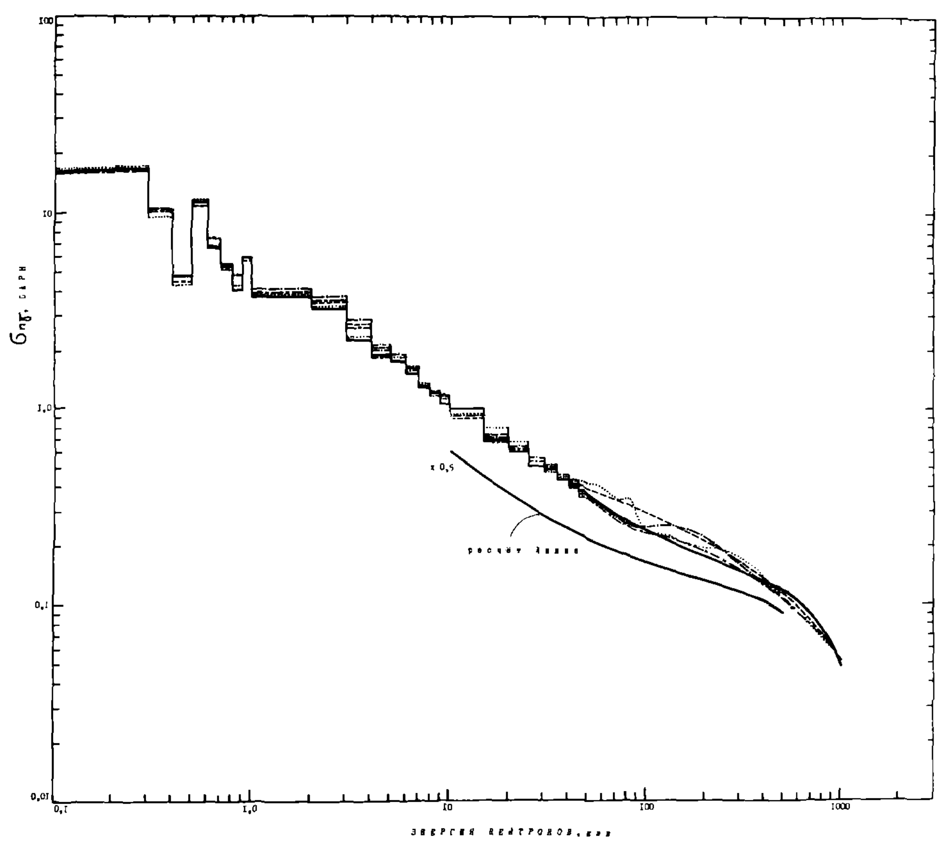


Fig. 2.: Value of α for plutonium-239 in the neutron energy range from 0.1 keV to 1 MeV.

——— - present paper; —·— - Sowerby-Kon'shin [1]; —··— - Kon'shin et al. [2]; .... - ENDF/B-IV [26]; ------ - Kawai [27].



<u>Fig. 3.</u>: Neutron radiative capture cross-section for plutonium-239 in the energy range from 0.1 keV to 1 MeV.

——— - present paper; —'— - Sowerby-Kon'shin [1];

—''— - Kon'shin et al. [2]; .... - ENDF/B-IV [26];

- - - - - Kawai [27]; ▬▬▬ - calculation by Lynn [30].