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RADIATIVE CAPTURE-TO-FISSION CROSS-SECTION RATIO FOR PLUTONIUM-239 IN THE NEUTRON ENERGY REGION BELOW ~ 50 keV

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The authors present the results of measurements of the parameter $\alpha(E)$ (neutron radiative capture-to-fission cross-section ratio) carried out with a spectrometer making use of slowing-down time in lead. The characteristic feature of this method is that the specimen and the detector are placed in an isotropic neutron field which is not altered as a result of scattering of the neutrons by nuclei of the specimen and the detector material. For normalizing the energy dependence of $\alpha(E)$ use is made of a satisfactorily thermalized neutron spectrum obtained with a graphite prism.

Introduction

Many laboratories are systematically measuring the parameter $\alpha(E)$ for ^{293}Pu, which to a considerable extent determines the breeding factor of the nuclear fuel used in fast reactors. Estimates of $\alpha(E)$ from total and fission cross-sections based on experiment indicate that the accuracy attainable in deriving it is not better than 30% [1]. Hence direct measurements are most important ones.

The difficulty involved in direct measurement of $\alpha(E)$ is separation of the radiative capture and fission events, as both processes are accompanied by the emission of prompt gamma radiation. Furthermore, in methods entailing time-of-flight measurements there is a great deal of uncertainty in the experimental values due to scattering by the specimen nuclei and to the difficulty of measuring the background level.

The method employing a neutron spectrometer based on the slowing-down time in lead [2, 3] enables us to measure the parameter $\alpha(E)$ at neutron energies below ~ 50 keV. The basic feature of this method is that the specimen and the detector are placed in an isotropic neutron field which is not altered as a result of scattering of the neutrons by the nuclei of the specimen and the detector material. The low gamma background of the spectrometer means that we can record gamma rays with a gas proportional counter, in which the efficiency with which the gamma rays are recorded is proportional to their energy [4, 5].

Method of measurement, detectors and equipment

The specimen studied and the gamma detector were placed in the measuring channel of the lead moderator, and the dependence of the detector count $I_\gamma(t)$ on the slowing-down time was measured:

$$I_\gamma(t) = \text{const.}\left[\varepsilon_c\sigma_c(E) + \varepsilon_f\sigma_f(E)\right]\Phi(t) = \\ = k_\gamma\left[\sigma_c(E) + \beta\sigma_f(E)\right]\Phi(t) \tag{1}$$

where $\beta = \varepsilon_f/\varepsilon_c$ is the ratio of the efficiencies with which the fission and capture events are recorded from the prompt gamma rays; $\Phi(t)$ is the neutron flux; $k_\gamma(\bar{n}_x, M_x, \varepsilon_c^x)$ is the normalizing factor, depending on the effective thickness of the specimen $\bar{n}_x$, the monitor count M_x and the efficiency with which the capture event is recorded for the nuclei of the given specimen. The gamma detector measurements alternated with measurements of the count from the fission chamber $I_f(t)$ containing layers of ^{239}Pu:

$$I_f(t) = k_f\sigma_f(E)\Phi(t) \tag{2}$$

where $k_f(\bar{n}_K, M_K, \varepsilon_K)$ is the normalizing factor, dependent on the effective thickness of the layer in the chamber $\bar{n}_K$, the monitor count M_K, and the chamber efficiency ε_K.

Dividing (1) by (2) we get the fundamental expression for parameter $\alpha(E)$:

$$\alpha(E) = \frac{\sigma_c(E)}{\sigma_f(E)} = \frac{k_f \cdot I_\gamma(t)}{k_\gamma \cdot I_f(t)} - \beta \tag{3}$$

To record the prompt gamma rays for fission and radiative capture use was made of a gas proportional counter [4, 5] in which the recording efficiency for gamma rays is proportional to their energy. In this case the recording efficiency for the capture and fission events is a weak function of the variation in the gamma ray spectrum, and is defined only by the total gamma ray energy per interaction event under consideration:

$$\varepsilon_c = \text{konst.} \cdot B_n\ ; \quad \varepsilon_f = \text{konst.} \cdot E_f \tag{4}$$

where B_n is the neutron binding energy in the nucleus, and E_f is the total energy of the gamma ray cascade per fission act. It follows from the above equalities (4) that the normalizing constants k_f/k_γ and β, which are characteristics of the detectors, are constant within the energy range under investigation.

The amplifying channel of the detectors contained a broad-band anti-saturation UIS-2 amplifier. The time dependence of the detector count was analysed by the measurement and recording centre of the P.N. Lebedev Institute of Physics of the USSR Academy of Sciences (FIAN) [6]. In view of the fact that the time resolution for the spectrometer was ~ 15%, the analyser contained sets of channels with a width of 0.25 μsec at the beginning and 64 μsec at the end of the cycle.

The low resolving power of the spectrometer prevented us from reliably determining the normalizing constants from resonances with known values of $\alpha(E_0)$. Hence normalization from the thermal values was the basic operation. For this purpose we made measurements in a graphite prism, which was placed near a lead "cube" and fed with a pulsed neutron burst from the lead [7], using, in turn, the fission chamber (ΔI_f^{th}), the specimen under investigation in the gamma counter (ΔI_γ^{th}), and a non-fissionable standard ($\Delta I_{\gamma эт}^{th}$):

$$\Delta I_f^{th} = k_f \sigma_f^{th} \Delta\Phi^{th} \tag{5}$$

$$\Delta I_\gamma^{th} = k_\gamma \left(\sigma_c^{th} + \beta \sigma_f^{th}\right) \Delta\Phi^{th} \tag{6}$$

$$\Delta I_{\gamma эт}^{th} = k_\gamma^{эт} \sigma_{c\, эт}^{th} \Delta\Phi^{th} \tag{7}$$

where σ_f^{th}, σ_c^{th}, $\sigma_{cэт}^{th}$ are thermal fission and radiative capture cross-sections averaged over the neutron spectrum in the graphite prism; $\Delta\Phi^{th}$ is the thermal neutron flux.

The relations (6) and (7) are dependent, since the condition in (4) is fulfilled for the proportional gamma counter, and for the specimens with identical geometry we get:

$$\frac{k_\gamma}{k_\gamma^{эт}} = \frac{\bar{n}_x M_x \varepsilon_c^x}{\bar{n}_{эт} M_{эт} \varepsilon_c^{эт}} = \frac{\bar{n}_x M_x B_n^x}{\bar{n}_{эт} M_{эт} B_n^{эт}} = C_1 \tag{8}$$

After simple transformations, (5), (6) and (7) give us the normalizing constants:

$$\beta = \frac{\sigma_{c>T}^{th}}{C_1\,\sigma_f^{th}}\;\frac{\Delta I_\gamma^{th}}{\Delta I_{\gamma>T}^{th}} - \alpha^{th} \tag{9}$$

$$\frac{\kappa_f}{\kappa_\gamma} = (\alpha^{th} + \beta)\,\frac{\Delta I_f^{th}}{\Delta I_\gamma^{th}} \tag{10}$$

where $\alpha^{th} = \sigma_c^{th}/\sigma_f^{th}$ is the thermal radiative capture-to-fission cross-section ratio averaged over the neutron spectrum.

<u>Measurements and processing of results</u>

In the lead moderator of the spectrometer we made the measurements at a neutron burst frequency of 312.5 and 625 Hz, the burst lasting ~ 0.5 μsec, and in the graphite prism at a frequency of 312.5 Hz, the burst lasting ~ 2 μsec.

Measurement of the signal from the specimen was alternated with measurement of the gamma background (i.e. of the background specimen in the gamma counter) and the natural background from the specimen, together with measurement with a fission chamber. The results for the lead moderator were processed with allowance being made for activation of the specimen (~ 1%) and the contribution made by "recycled" neutrons (~ 0.5%), and after corrections had been introduced for counting errors (not more than 2%) [4, 5]. In the processing of the measurements obtained for the graphite prism allowance was made for activation of the specimen and reduction of the flux due to absorption by the specimen.

Measurements were made with the proportional gamma counter for PuO_2 specimens (~ 1.8% ^{240}Pu) with different effective thicknesses ($\bar{n} = 4.4 \times 10^{21}$; 4.2×10^{20} and 1.7×10^{20} at/cm^2). For purposes of normalization, we also made measurements with the graphite prism for specimens of ^{107}Ag and ^{109}Ag ($\bar{n} = 4.3 \times 10^{21}$ at/cm^2) and specimens of ^{197}Au ($\bar{n} = 1.8 \times 10^{21}$ at/cm^2). The fission signal was measured by means of ionization fission chambers containing 1.6 and 12 mg of $^{239}PuO_2$.

Processing of the relative path of the energy dependence $\alpha(E)$, obtained with specimens of different thickness, showed that the resonance self-shielding effect becomes considerable in the case of the specimen with $\bar{n} = 4.4 \times 10^{21}$ at/cm^2 for energies below ~ 8 keV. For the specimens with $\bar{n} = 4.2 \times 10^{20}$ and 1.7×10^{20} at/cm^2 no difference in the relative path of the energy dependence $\alpha(E)$ was observed within the limits of the statistical measurements over the energy range above 0.2 keV; this suggests that the dependence of the measurement results on the thickness is only slight for the given specimens. This result is borne out by evaluations of the resonance self-shielding effect made by Dresner [8], on the basis of assumptions in which the geometry of the cylindrical specimen and the isotropy of the neutron flux are taken into account.

Since the signal/background ratio for the specimen with $\bar{n} = 4.2 \times 10^{20}$ at/cm^2 (~ 25% for energies of 0.2-2.0 keV; ~ 20% in the energy region 2-8 keV, and ~ 10% above ~ 8 keV) is considerably better than for the one with $\bar{n} = 1.7 \times 10^{20}$ at/cm^2, the principal measurements in the lead moderator were made with a thicker specimen. Results of measurements with the specimen $\bar{n} = 4.4 \times 10^{21}$ at/cm^2 were only used over the energy range ~ 8-15 keV, where resonance self-shielding is small, but the signal/background ratio (~ 20-15%) is better than for the specimen with $\bar{n} = 4.2 \times 10^{20}$ at/cm^2. Above 15 keV the signal/background ratio deteriorates as the background in the vicinity of the neutron burst increases. Furthermore, because of the unstable position of the neutron burst in relation to the commencement of the cycle, the mean square spread of the measurement data is broad and amounts to ~ 15% for $\alpha(E)$; this largely reduces the reliability of the data above ~ 15 keV when $\alpha(E)$ is small.

Results and discussion

The thermal cross-sections used to determine the normalizing constants are shown in Table 1. The absolute error due to normalization is given in Table 2 as a function of $\alpha(E)$.

Table 1

Thermal cross-sections used for normalization

Element	Values at E = 0.0253 eV	f	Values for Maxwellian spectrum
^{239}Pu	$\sigma_f = 741.6 \pm 3.1$ barn [9]	1.055	$\sigma_f^{th} = 780.2$ barn
	$\alpha = 3659 \pm 0.0039$ [9]	1.088	$\alpha^{th} = 0.3982$
^{197}Au	$\sigma_c = 98.6 \pm 0.3$ barn [10]	1.005	$\sigma_c^{th} = 99.1$ barn
$^{107, 109}$Ag	$\sigma_c = 63.6 \pm 0.6$ barn [10]	1.004	$\sigma_c^{th} = 63.8$ barn

Note: $\sigma_x^{th} = f\sigma_x$ (E = 0.0253 eV), where f is the Westcott factor [11].

The measurements in the graphite prism gave the value $\beta = 0.80 \pm 0.06$, which corresponds to the mean total energy of the cascade of prompt fission gamma rays $E_f \approx 5.12$ MeV.

The results of our measurements of the energy dependence of $\alpha(E)$ for ^{239}Pu are shown in Table 3, and also in the figure in the form of a comparison with data obtained by other authors.

Table 2

Absolute error in $\alpha(E)$ due to normalization

$\alpha(E)$	$\pm\Delta\alpha$	$\alpha(E)$	$\pm\Delta\alpha$
0.4	0.06	1.0	0.10
0.6	0.07	1.2	0.11
0.8	0.08		

Note: The normalization error is basically determined by the measurement statistics in the graphite prism (1-2%), the error in the absolute weight of the specimens (~ 4%), and the uncertainty in the dependence of the gamma counter efficiency on the gamma ray energy (~ 4%).

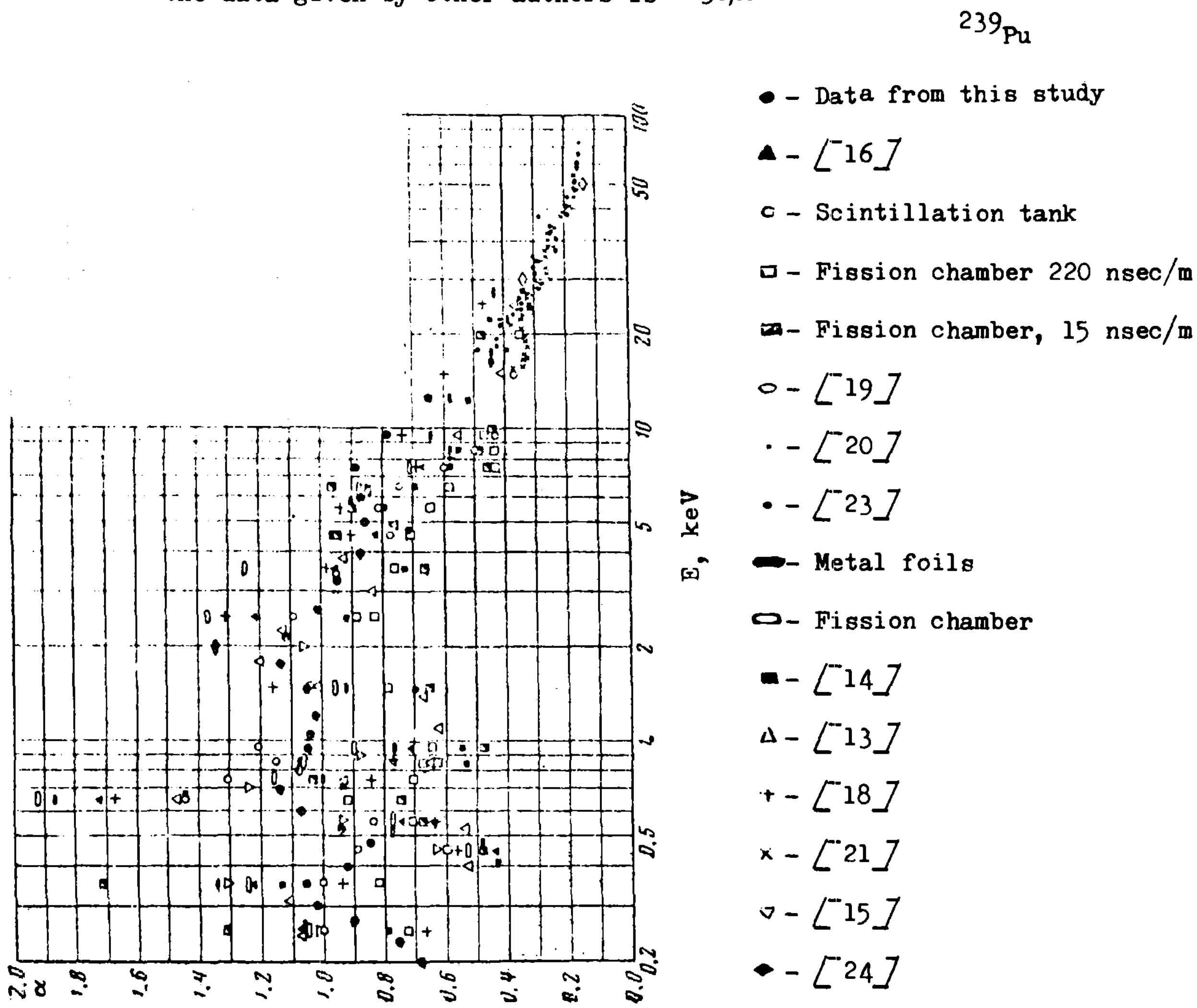
The data obtained in studies made by the time-of-flight method with linear accelerators [12-15], with the neutron spectrometer of a cyclotron [16], and in a fast pulsed reactor [17], together with data [18] obtained experimentally during an underground nuclear explosion, have been averaged over the energy ranges, the midpoint of each interval being shown by the corresponding point in the figure.

For purposes of comparison, the figure also contains data obtained in experiments carried out on pulsed Van de Graaff generators [19-21], and with photoneutron sources [22, 23], as well as data from measurements using a scandium filter in a reactor beam [24].

As can be seen from the figure, the spread of the $\alpha(E)$ values for ^{239}Pu is fairly broad. The discrepancy found in the data obtained by one group of authors, though they applied different procedures [12, 17], should be noted.

On the average, the results of the present work in the energy region up to ~ 6 keV do not disagree with the sum total of the data, if we take into consideration the low resolving power of this method. For energies above ~ 9 keV better agreement is observed with the data of Farrel and

Auchampaugh [18]. In the energy region ~ 6-9 keV the disagreement with the data given by other authors is ~ 30%.



(Figure caption) Energy dependence of α(E) for ^{239}Pu. The data obtained with the scintillation tank (detector) and fission chambers for 220 nsec/m and 15 nsec/m are taken from Ref. [17]. Those obtained with the use of metal foils and a fission chamber are from Ref. [12].

It is important to note in conclusion that when evaluating the recommended α(E) values for ^{239}Pu we must take into account the resonance self-shielding effect, since for specimens thicker than ~ 4×10^{20} at/cm^2 the effect of their thickness may be quite considerable in different experiments.

Table 3

Numerical values of fission and radiative capture cross-sections and α(E) for ^{239}Pu

E, keV	α(E)	σf(E), barn [7]	σc(E), barn	E, keV	α(E)	σf(E), barn [7]	σc(E) barn
12.5	0.65 ± 0.13	1.65 ± 0.10	1.1 ± 0.2	0.92	1.05 ± 0.11	5.7 ± 0.3	6.0 ± 0.9
9.5	0.79 ± 0.13	1.74 ± 0.10	1.4 ± 0.3	0.80	1.08 ± 0.11	6.1 ± 0.3	6.6 ± 1.0
7.5	0.89 ± 0.13	1.88 ± 0.11	1.7 ± 0.3	0.70	1.13 ± 0.11	6.7 ± 0.3	7.6 ± 1.2
6.0	0.87 ± 0.16*	2.06 ± 0.11	1.8 ± 0.3	0.60	1.07 ± 0.10	7.7 ± 0.4	8.2 ± 1.4
5.0	0.86 ± 0.13	2.26 ± 0.12	1.9 ± 0.4	0.53	0.94 ± 0.10	8.4 ± 0.4	7.9 ± 1.2
4.0	0.88 ± 0.13	2.37 ± 0.13	2.1 ± 0.4	0.47	0.85 ± 0.14*	8.9 ± 0.5	7.6 ± 1.2
3.2	0.95 ± 0.12	2.61 ± 0.13	2.5 ± 0.5	0.40	0.92 ± 0.09	9.4 ± 0.5	8.6 ± 1.5
2.6	1.01 ± 0.12	2.80 ± 0.14	2.8 ± 0.5	0.35	1.05 ± 0.09	10.5 ± 0.6	11.0 ± 1.8
2.17	1.12 ± 0.12	3.14 ± 0.16	3.5 ± 0.6	0.30	1.02 ± 0.09	12.3 ± 0.6	12.5 ± 2.0
1.77	1.16 ± 0.12	3.7 ± 0.2	4.3 ± 0.7	0.27	0.90 ± 0.09	13.8 ± 0.7	12.4 ± 2.0
1.47	1.05 ± 0.16*	4.3 ± 0.2	4.5 ± 0.7	0.23	0.76 ± 0.09	15.8 ± 0.7	12.0 ± 1.9
1.23	1.03 ± 0.12	5.1 ± 0.3	5.2 ± 0.8	0.20	0.60 ± 0.09	16.5 ± 0.8	11.4 ± 1.8
1.05	1.04 ± 0.11	5.6 ± 0.3	5.8 ± 0.8				

Note: Here we show the statistical error in the results for α(E) measurements, but the total error is given in the case of values marked with an asterisk.

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