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ABSTRACT

The neutron radiative capture cross-sections for thorium-232 in the 0.82-2.44 MeV energy range were measured relative to $\sigma_{n,\gamma}$ for ^{197}Au and $\sigma_{n,f}$ for ^{235}U by the activation method. The neutron source was the reaction T(p,n)^{3}He, produced on the EhG-5 electrostatic accelerator of the Nuclear Research Institute of the Academy of Sciences of the Ukrainian SSR. The induced activity was recorded with a Ge(Li) detector from the gamma peak of ^{233}U with E = 312 keV. The neutron radiative capture cross-section for gold-197 relative to $\sigma_{n,f}$ for ^{235}U was determined at the same time.

The ecological consequences of using conventional fuels (coal, fuel oil), the greenhouse effect and pollution of the environment with combustion products (including radioactive ones) point to the need to switch to safe nuclear power. Public opinion in the civilized world has recently shown a trend in favour of nuclear power stations. However, as we know, thermal reactors do not solve the energy problem in the long term. Therefore work is continuing on the philosophy and design of safe breeder reactors, the raw material for the production of ^{239}Pu in which is ^{238}U. Progress has been achieved in this area, notably as regards the supply of nuclear data for breeder design.

However, in nature there exist considerably greater reserves of another nuclear raw material, namely thorium. The capture of neutrons in thorium leads to the formation of another fuel - ^{233}U. However, the fund of technological experience and data (in particular on neutron capture) which has been accumulated for the uranium-plutonium cycle, does not yet exist in this case.

In economically developed countries (United States, France, Germany, etc.) extensive national programmes have been initiated on the thorium cycle. These include investigation of nuclear data in differential and integral experiments. Such a programme is also being conducted in the USSR and the present work was carried out in that context. It was performed for the purpose of obtaining more reliable data on $\sigma_{n,\alpha}$ for ^{232}Th, because our preliminary measurements resulted in lower values for ^{232}Th capture cross-sections than those given in the ENDF/B-V evaluation.

Method of measurement

The radiative capture cross-section for ^{232}Th was measured in relation to the cross-sections $\sigma_{n,\gamma}$ for ^{197}Au and $\sigma_{n,f}$ for ^{235}U. The relationship between the measured cross-section and the standard $\sigma_{n,f}$ for ^{197}Au is expressed as follows:

$$\sigma_{n,\gamma}\,(<E_n>) = \sigma^{st}\left(<E_n>^{st}\right) \cdot \frac{\eta(E_\gamma^{st})}{\eta(E_\gamma} \cdot \frac{N_{\gamma o}}{N_{\gamma o}^{st}} \cdot \frac{N_{nei}^{st}}{N_{nei}} \cdot \frac{f(\lambda,t)}{f(\lambda^{st},t^{st}} \cdot \frac{C_\varphi^{st}}{C_\varphi} \cdot \frac{I^{st}}{I} \quad . \qquad (1)$$

Here, the terms with the superscript "st" relate to gold and those without relate to thorium. $<E_n>$ is the mean energy of neutrons hitting the sample; η is the efficiency of the Ge(Li) detector in recording gamma rays of energy E_γ of the corresponding sample; N_{nei} is the number of activated nuclei in the sample; $f(\lambda,t)$ is the time factor reducing $N_{\gamma o}$ to the moment of termination of irradiation; and C_φ is a correction for inconstancy of the neutron flux with time.

$N_{\gamma o}$ is calculated by the formula:

$$N_{\gamma o} = \frac{B \cdot N_{\gamma}}{T_x} , \quad (2)$$

where N_{γ} is the area of the gamma peak over the time of measurement of the activity; B is the correction for neutrons scattered on the experimental rig and in the laboratory; T_x is a correction for the "dead" time of the recording channel.

I is determined by numerical integration:

$$I = (1 + \beta \cdot D) \int_{\Delta E} N(E) dE , \quad (3)$$

where ΔE is the energy range of neutrons hitting the sample; $(1 + \beta D)$ is a factor allowing for the energy dependence of the cross-section in the range ΔE; N(E) is the energy spectrum of neutrons from the source hitting the sample within the corresponding solid angle. More detailed information on the method of measurement used and the relevant references are to be found in Refs [1, 2].

In the case of measurement of $\sigma_{n,\gamma}$ for ^{232}Th relative to $\sigma_{n,f}$ for ^{235}U the relationship (1) has a similar form but the terms with the superscript "st" refer to the fission chamber. In this case the value $f(\lambda^{st}, t^{st}) = 1$, N_{γ} is the number of fission events recorded over the irradiation time, and η^{st} is the efficiency of recording of fission events by the chamber.

A few comments regarding the value $<E_n>$. The method used for processing the results of the experiment enables us to obtain the cross-section for a specific neutron energy $<E_n>$ using a non-monochromatic neutron source. This is determined by the expression from Ref. [1]:

$$<E_n> = \int_{\Delta E} E \cdot N(E)dE / \int_{\Delta E} N(E)dE. \qquad (4)$$

The values $<E_n>$ and $<E_n^{st}>$ are practically identical for the Au-Th pairing and differ considerably for the U-Th pairing. These values are given in the table of results of the experiment. The values of $\pm \sqrt{D}$ are also given, where D is the dispersion of the spectrum N(E).

Experiment

The neutron source was the reaction $T(p,n)^3He$ performed on the EhG-5 electrostatic accelerator of the Nuclear Research Centre of the Academy of Sciences of the Ukrainian SSR. Titanium-tritium targets with a thickness of 1 mg/cm^2 were used (142 keV at the reaction threshold). The target was cooled by compressed air.

The experimental assembly for irradiating the samples is similar to that used in Refs [1, 2]. The samples were fixed close together on the target assembly in a special holder ~ 4 cm from the neutron source. The thorium samples (ThO_2) were packed in stainless steel containers 0.01 cm thick. The diameter of the samples was 2 cm, the thickness of the gold sample 0.02 cm, that of the thorium sample 0.07 cm and the mass of the samples was 1.0 g.

The flow-type fission chamber has a thin walled casing (0.01 cm) made of stainless steel, the mass of the layer of ^{235}U is 1.277 mg, the thickness of the layer is 0.405 mg/cm^2 and the enrichment is 99.99%. The distance between the layer and the neutron source was around 5.5 cm. The correction for inconstancy of the neutron flux with time was determined using the readings of the monitor (BF_3-counter).

The induced activity in the gold sample was recorded with a Ge(Li) detector from the $^{198}H_\gamma$ peak with E_γ = 411.8 keV ($T_{½}$ = 2.696 days), and in the thorium sample from the

^{233}U peak with $E_\gamma = 311.9$ keV ($T_{½} = 27.0$ days). The samples were rigidly attached to the surface of the detector. The information required for processing the results of the measurements (thermal neutron activation cross-section, half-lives, gamma-ray yield probability) was taken from Refs [3 and 4]. The electronic part of the experimental set-up was controlled and the experimental results were processed with the aid of a SM-1420 computer.

Measurement of recording efficiency

In this section, for convenience of assessing the results of measurements of efficiency, we shall use different notations: η_{312}, η_{412}, and η_{fc} are the gamma-ray recording efficiencies for the thorium sample, the gold sample and fission events in the fission chamber; N_{Th}, N_{Au}, and N_U are the numbers of activated nuclei in the thorium and gold samples and of uranium-235 nuclei in the active layer of the fission chamber; $\sigma_{n,\gamma}(Th)$, $\sigma_{n,\gamma}(Au)$ and $\sigma_{n,f}$ are respectively the neutron interaction cross-sections for thorium, gold and uranium-235; and N_{312}, N_{412} and N_{fc} are the number of counts for the thorium and gold samples and the fission chamber. The symbols with superscript "th" refer to thermal flux irradiation and those with no superscript refer to fast flux irradiation. The thermal flux irradiations necessary for determining the efficiencies were performed in channels of the VVRM (modernized water water reactor) reactor of the Nuclear Research Institute of the Academy of Sciences of the Ukrainian SSR.

In processing the results of measurements of η_{312}, η_{412} and η_{fc}, corrections were introduced for attenuation of the thermal neutron and gamma-ray flux in the samples and the walls of the containers, and the necessary time corrections were also made.

The gamma-ray recording efficiencies η_{312}, η_{412} with a Ge(Li) detector were determined using a set of standard OSGI-3-2 gamma sources. Since the samples have a diameter of 20 mm, while the standard sources are practically point-like, the samples were placed 50 cm from the detector during measurement of the efficiency. At this distance the activated sample may be regarded as point-like.

The dependence of efficiency on gamma-ray energy in the 0.24-0.66 MeV range was determined from the nine gamma peaks of the ^{133}Ba, ^{152}Eu and ^{137}Cs sources. Using the efficiency values obtained, the absolute activities of the gold and thorium samples irradiated with thermal neutrons were measured. Then measurements were made of the activities of the same samples in the same position on the surface of the Ge(Li) detector which was used for measuring the activities of the samples irradiated on the accelerator. From the results of these measurements we calculated the efficiencies η_{312} and η_{412} : $\eta_{412} = 3.398 \cdot 10^{-2}$ $\pm$ 3.4% and $\eta_{312} = 2.148 \cdot 10^{-2} \pm 3.5\%$. Here the quantum yields employed were $P_{312} = 0.36$ and $P_{412} = 0.9555$.

The fission event recording efficiency η_{fc} was determined from the recording of the pulse amplitude spectrum for the fission chamber irradiated with a known thermal flux. The flux was determined from the induced activity of the gold sample irradiated in the same flux. A value of $\eta_{fc} = 0.979 \pm 3\%$ was obtained. It should be noted that this value refers to a certain selected pulse-amplitude recording threshold which was allowed for in processing the results of the experiment carried out on the accelerator. In calculating the value η_{fc}, we used the following thermal neutron cross-sections:

$$\sigma^{th}_{n,\gamma}(Au) = 98.86 \pm 0.3\%, \ \sigma^{th}_{n,f} = 582.26 = \pm 0.2\%$$

The radiative capture cross-sections were calculated from formula (1) using the above values of the efficiencies η_{312}, η_{412} and η_{fc}. However we wish to draw attention to certain features of the results for $\sigma_{n,\gamma}(Th)$ obtained in relation to different standard cross-sections (we shall omit those terms in formula (1) which are of no significance for our treatment). For the standard $\sigma_{n,\gamma}$(Au) the cross-section $\sigma_{n,\gamma}$(Th) is proportional to the values:

$$\sigma_{n,\gamma}(Th) \approx \left(\frac{N_{312} \cdot \sigma_{n,\gamma}(Au)}{N_{412}}\right) \cdot \frac{\eta_{412}}{\eta_{312}} \cdot \frac{N_{Au}}{N_{Th}} \cdot \tag{5}$$

The values dependent on neutron energy are shown in brackets. For the standard $\sigma_{n,f}$, taking into account the procedure described above for measuring the value η_{fc}, we have such a relationship:

$$\sigma_{n,\gamma}(Th) \approx \left(\frac{N_{312} \cdot \sigma_{n,f}}{N_{fc}}\right) \cdot \frac{\eta_{412}}{\eta_{312}} \cdot \frac{N_{Au}}{N_{Th}} \cdot \frac{N_{fc}^{th}}{N_{412}^{th}} \cdot \frac{\sigma_{n,\gamma}^{th}(Au)}{\sigma_{n,f}^{th}} \cdot \tag{6}$$

It should be noted that expression (6) does not depend on the number of uranium-235 nuclei in the active layer of the fission chamber but on N_{Au}, which can be determined with fewer systematic errors. At the same time there is a connection between expressions (5) and (6) through the efficiencies which leads to common systematic errors for the values of $\sigma_{n,\gamma}$(Th) measured relative to both standards.

A few comments here regarding possible systematic errors.

The method used for determining the ratio η_{412}/η_{312} from the same dependence $\eta(E_\gamma)$ reduces the possible systematic errors.

The value of the efficiency η_{412} was checked by the following method. The activity of the gold sample irradiated by thermal neutrons was also measured on another Ge(Li) detector (in the Physics and Power Engineering Institute), for which the gamma-ray recording efficiency with E_γ = 412 keV had been determined through measurement of the absolute activity of the sample by the $4\pi\beta$-γ coincidence method. The numbers of active ^{198}Au nuclei recorded in the first and second measurements at the moment of termination of radiation were equal to: $N_1 = 8.04 \cdot 10^8 \pm 3.4\%$ and $N_2 = 7.92 \cdot 10^8 \pm 1.7\%$, i.e. a difference of 1.5%.

The data obtained when the thorium sample and the fission chamber are irradiated in the same thermal flux enable the value of $\sigma_{n,\gamma}$(Th) relative to the standard $\sigma_{n,f}$ to be obtained by yet another method. In this case we obtain such a relationship:

$$\sigma_{n,\gamma}(Th) \approx \left(\frac{N_{312}\sigma_{n,f}}{N_{fc}}\right) \cdot \frac{N_{fc}^{th}}{N_{312}^{th}} \cdot \frac{\sigma_{n,\gamma}^{th}(Th)}{\sigma_{n,f}^{th}} \; . \tag{7}$$

It clearly differs considerably from relationship (6) in the part not dependent on fast neutron energy. We would stress that relationship (7) is independent of the quantity of thorium and uranium nuclei participating in the reactions. The cross-sections $\sigma_{n,\gamma}$(Th) for E_n = 1030 keV calculated from relationships (6) and (7) were equal to 108.0 ± 5.6 mb and 108.9 ± 4.6 mb respectively. The value of $\sigma_{n,\gamma}^{th}$ (Th) = 7.40 b ± 1.1%.

From the information supplied it may be concluded that the values in relationships (5) and (6) that are independent of fast neutron energy do not introduce significant systematic errors.

Corrections for scattered neutrons

For any detector the correction B in relationship (2) for scattered neutrons causing an additional effect in the detector is determined in accordance with the model:

$$N_\gamma = N_{\gamma o} + N_{bs} + N_{br} \; . \tag{8}$$

Here N_γ is the measured effect from the detector; $N_{\gamma o}$ is the effect caused by neutrons striking the detector directly from the source; N_{bs} is the effect due to neutrons scattered by elements of the experimental rig; and N_{br} is the room background.

Having transformed expression (8), we obtain relationship (2) (without correction for "dead" time of the recording channel):

$$N_{\gamma o} = N_\gamma \left(1 - \Delta N_{bs} - \Delta N_{br}\right) = N_\gamma \left(B_\gamma - \Delta N_{br}\right) = N_\gamma B. \qquad (9)$$

The relationship between the new terms introduced and those used above requires no further explanation.

As follows from relationship (9), the correction B consists of two components: B_γ and ΔN_{br}.

The value of ΔN_{br} was measured experimentally in the form of the ratio of the effects due to the detector irradiated in two positions: (1) at a distance of ~ 2 m from the neutron source; and (2) in the standard position 4-5 cm from the source. When the neutron energy was varied in the range 0.8-2.4 MeV, the values of ΔN_{br} varied in the range 0.065-0.105 for gold samples, and 0.040-0.025 for the fission chamber, whilst for the thorium samples the contribution of neutrons scattered in the room was zero. The error of measurement of values of ΔN_{br} was 0.02-0.03.

The value of B_γ, which takes into account the effect of neutrons scattered by parts of the experimental assembly, was determined by calculation. The calculations were performed using the set of BRAND programs [5]. The geometrical configuration of the experimental assembly was reproduced almost exactly in the calculation. The mass of the assembly is 297 g. It should be noted that additional calculations showed that the main effect is produced by elements of the assembly located 5-6 cm from the neutron source; in our case these elements have a mass of 70 g. The chemical composition of the assembly amounts to 15 elements. Data on the kinematics of the reaction $T(p,n)^3He$ were taken from Ref. [6]. In a simulation of the history of a neutron the energy varies from maximum to 5 eV. The statistical error of the calculation amounted to ~ 0.3%.

Figure 1 shows the results of calculating the correction B (combining the results of experiment and calculation) for three detectors in the neutron energy range investigated. The sum error of the value B amounted to 2% for the thorium samples, not more than 3.1% for the gold samples, and 2.1% for the fission chamber.

Owing to the presence in the cooling air of oil particles from the air compressor during the irradiation process, drops of oil appeared on the gold sample. An approximate estimation was made of their effect: with increase in energy of the irradiating neutrons, the increase in the activity of the gold sample ranges from 2.4 to 1.7%. However this information was not employed in processing the results of the measurements.

Results of the measurements

The results of the experiments performed to measure the radiative capture cross-section for thorium-232 are given in the Table and in Fig. 2. The Table shows the standard cross-sections employed (ENDF/B-6 library) and their errors (in the authors' commentary it is indicated that these values are not definitive).

The errors in $\sigma_{n,\gamma}$ for thorium-232 shown in the table were calculated by quadratic addition of the aforementioned errors in the values in formula (2) and the errors caused by statistical scatter in the recording of radiation. It should be noted that the evaluations of random errors are possibly slightly too high.

It was indicated above that the systematic errors in the experimental results should not be very great. In fact the systematic difference existing between the cross-sections obtained relative to the two standards does not exceed the random errors indicated in the table (varying from -3.5% to +6.4%). None the less it may be assumed that this difference can be caused both by systematic errors in calculating the corrections B and systematic errors in the reference cross-sections.

Figure 2 shows the evaluated data for $\sigma_{n,\gamma}$ of ^{232}Th from two libraries (the most recent being JENDL-3 of 1987). Our results are closer to the JENDL-3 evaluation but are systematically 1.5-14% below it.

The last column of the table shows the values of $\sigma_{n,\gamma}$ for ^{197}Au measured in relation to $\sigma_{n,f}$ for ^{235}U. The corresponding values of the mean neutron energy are given in the first column of the table. The formula used for the calculations is similar to relationship (7).

In Fig. 3 our results are plotted together with the evaluated data from the ENDF/B-6 library. On the whole our data are higher than the evaluated data (apart from the point at E_n = 2.04 MeV which is 7% lower), but agree with them within the error limits of the experimental and evaluated data.

Conclusions

The evaluated data for radiative capture cross-sections (including those for thorium-232) are still tending towards reduced values as the amount of experimental data increases. The results obtained by us correspond to this trend. Evidently it is desirable to carry out additional experiments in the energy range concerned, using other methods especially considering the inadequate accuracy of our results.

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[7] Report JAERT 1319 (1990).

Table

Results of measurements of neutron radiative capture cross-sections for thorium-232 and gold-197 (all errors are indicated in per cent)

№№	Data from the ENDF/B-6 library				Results of measurements			
	Gold-197		Uranium-235		$\sigma_{n,\gamma}$, mb			
					Thorium-232			Gold-197
	$\langle E_n \rangle$, keV	$\sigma_{n,\gamma}$, mb	$\langle E_n \rangle$, keV	$\sigma_{n,f}$, b	$\langle E_n \rangle$, $\sqrt{D}$ keV	rel. to $\sigma_{n,\gamma}$	rel. to $\sigma_{n,f}$	rel. to $\sigma_{n,f}$
1	813,3	89,0±2,1	819,9	1,113±0,6	813,3 35,3	138,5±7,6	143,5±6,3	92,2±4,4
2	1030	79,4±2,5	1038	1,196±0,6	1030 33	112,7±6,5	113,4±5,9	80,4±3.4
3	1533	67,2±1,8	1543	1,237±0,6	1533 30	91,0±6,3	96,2±5,9	70,9±3,75
4	2036	5[illegible],6±2,1	2048	1,271±0,6	2036 30	58,8±6,2	55,8±5,9	48,2±3,7
5	2435	35,2±3,0	2450	1,253±0,6	2435 31	36,8±7,1	34,6±6,3	35,6±3,7

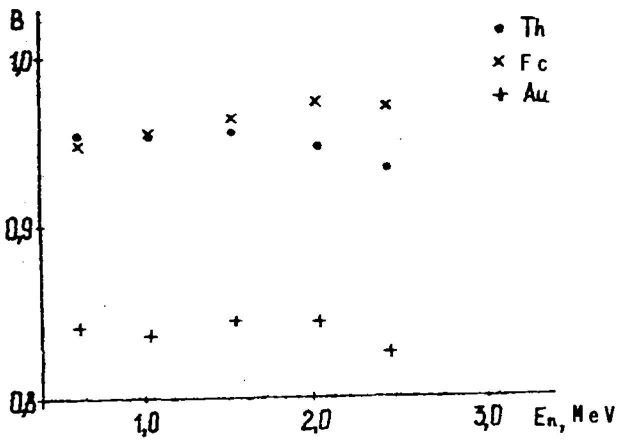

Fig. 1. Corrections for scattered neutrons for samples of ThO_2, Au and the fission chamber.

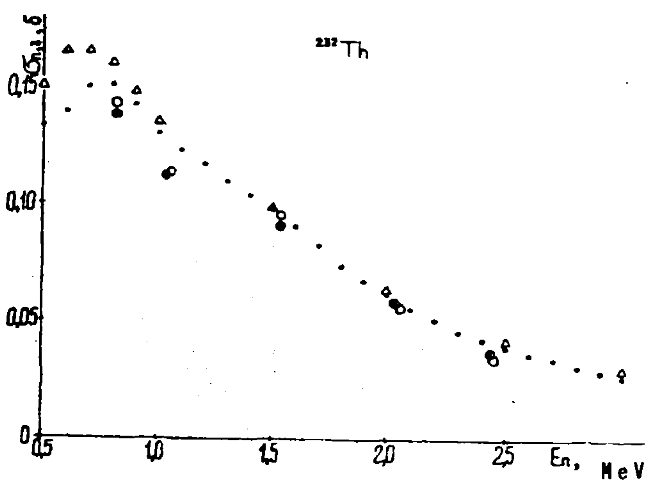

Fig. 2. Neutron radiative capture cross-sections for thorium-232. Experiment: ⊗ - relative to $\sigma_{n,\gamma}$ of ^{197}Au; O - relative to $\sigma_{n,f}$ of ^{235}U. Evaluated data: * - JENDL-3 [7]; Δ - ENDF/B-6.

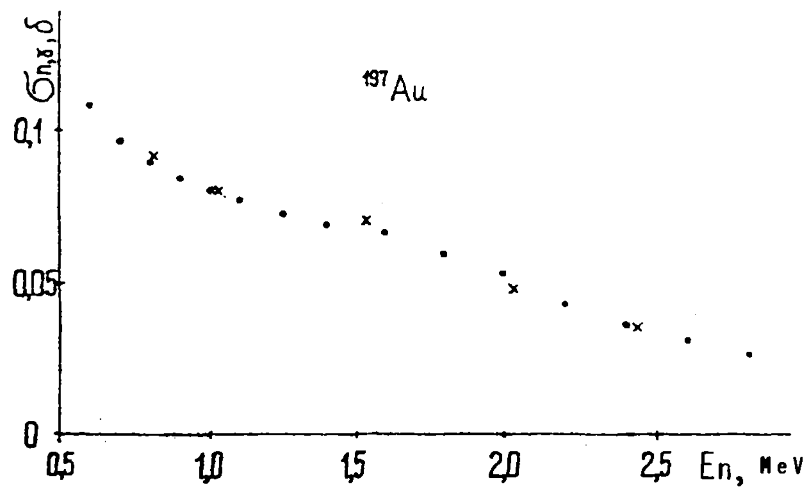

Fig. 3. Neutron radiative capture cross-section for gold-197. Experiment: x - relative to $\sigma_{n,f}$ of ^{235}U. Evaluated data: * - ENDF/B-6.