Measurement of the cross sections for the ^{232}Th(n,2n)^{231}Th reaction in the 6.745 to 10.450 MeV energy range

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A part of the excitation function of the ^{232}Th(n,2n) reaction was determined by the foil activation method for neutron energies of 6.745, 6.938, 7.190, 7.448, 7.697, 7.944, 8.457, 8.975, 9.445, 9.934, and 10.450 MeV. Neutron flux density was measured using reactions ^{238}U(n,f), ^{56}Fe(n,p), and ^{27}Al(n,α). Activity of the ^{231}Th residual nuclei was determined by their low energy γ transitions detected with a high purity Ge spectrometer. Results of this experiment are compared with renormalized literature data.

I. INTRODUCTION

Experimental investigation of the excitation function of the reaction ^{232}Th(n,2n) has both practical and theoretical importance. According to WRENDA (Ref. 1) these data are required to 10 MeV for the Th-U nuclear fuel cycle (control of the ^{232}U production). Since thorium may have a possible use in the blanket of fission-fusion systems, data are also necessary up to 15 MeV neutron energy. From a theoretical point of view this reaction is also interesting for investigations on the neutron emission and fission competition in heavy nuclei.

There are two systematical investigations on the excitation function in the 6.5—11 MeV interval (Refs. 2 and 3). Discrepancies as high as 25% occur between their results at about 10.5 MeV. (As numerical values of Ref. 2 had not been available to us, this comparison was made on a figure given in Ref. 4.) Our experiments supply data from 6.745 to 10.450 MeV using the activation method.

II. EXPERIMENTAL

Neutrons of energy from 6.745 to 10.450 MeV were produced via the ^{2}H(d,n)^{3}He reaction on the tandem accelerator EGP-10M of Fiziko-Energeticheskiy Institute (FEI) (Obninsk) using a 50 mm long target cell. Deuterium gas pressure was about 115 kPa. A (7.9±0.1) mg/cm^2 thick Mo foil served as entrance window. The end of the Ni cell had a Pt cover of 0.3 mm thickness to reduce background from (d,n) neutrons. The ion current was about 2 μA. The precision of the energy calibration of the generator is estimated to be ±7 keV.

The energy distribution of neutrons activating the samples was computed from the reaction kinematics on the basis of the deuteron energy loss in the window and gas as well as the differential cross section of the reaction.[5] The irradiation geometry was also taken into account. The mean energy of the neutrons was controlled in separate experiments. Resonances in the total cross section of the ^{12}C + n process at (5.369±0.003) and (6.293±0.003) MeV (Ref. 6) were used at different gas pressures in the target cell. The thickness of the entrance window of Mo was (8.2±0.1) and (16.4±0.2) mg/cm^2. A systematical shift of −(52±8) keV on average, compared to the above calculations, was observed when the results had been analyzed with stopping power values from Ref. 7. Using the results of Refs. 8 and 9 this discrepancy reduced to 24 keV, within the error limits quoted in Ref. 8. However, the precision of our energy calibration is much better. Therefore it seemed reasonable to increase the energy loss in the Mo foil by 2—4% to have agreement with the experiment, not exceeding the error limits. The mean neutron energies and the width of the calculated distributions (in 68% sense, symmetrized) are listed in Table I. The uncertainty in the mean values is about 8 and 17 keV at 7 and 10 MeV neutron energy, respectively.

Samples of metal thorium with a diameter of 18.5 mm and a thickness of about 0.3 mm (0.9—1 g) were used. The activity of the ^{231}Th nuclei was determined through their low energy γ rays ranging from 25.6 to 102.3 keV. These radiations were detected by the high purity Ge spectrometer of the Institute of Experimental Physics (IEP) (Debrecen).

Neutron flux density was measured by the reactions ^{56}Fe(n,p) and ^{27}Al(n,α) applying the foil activation technique and ^{238}U(n,f) using a flow-type fission chamber. All the foils including the fissionable layer were 19 mm in diameter. Reactions ^{115}In(n,n')^{115}In^{m} and ^{64}Zn(n,p) were also studied by the activation method; their results will be published elsewhere.

The monitor foils and the Th sample formed a 2.5 mm thick sandwich. It contained two Fe foils (of 0.35 mm thickness each) and two Al foils (0.2 mm) arranged symmetrically around the Th disc, one In (0.2 mm) and one Zn (0.5 mm) foil and one more Al foil to control the flux variation along the stack. A light aluminum sample holder fixed the sandwich to the end of the gas target at a distance of 39 mm at 0° direction. The fission chamber was touching the sandwich.

TABLE I. Results of the present measurements on the excitation function of the ^{232}Th(n,2n) reaction.

	^{232}Th(n,2n)^{231}Th			Neutron flux density, ϕ, 10^6 cm^{2}s^{-1}; cross section,[b] σ, mb					
E_n (MeV)	σn,2n (mb)[c]	$\phi\cdot\sigma$ (10^{-18} s^{-1})	$\bar{\phi}$ weighted average	^{238}U(n,f) σ	ϕ (± %)	^{56}Fe(n,p) σ	ϕ (± %)	^{27}Al(n,α) σ	ϕ (± %)
6.745±0.070	74±11	0.269(13)	3.640(4.2)	890	3.828(3.0)	26.7	3.961(3.7)	10.7	3.455(2.3)
6.938±0.070	209±13	0.855(5.7)	4.088(2.3)	930	4.070(1.0)	30.0	3.971(2.6)	14.5	4.597(3.3)
7.190±0.070	449±18	2.175(3.6)	4.839(1.7)	952	4.799(0.8)	33.2	4.844(3.7)	19.9	5.186(2.2)
7.448±0.070	827±37	3.020(4.3)	3.651(1.1)	978	3.683(1.2)	37.3	3.542(2.5)	26.2	3.593(4.3)
7.697±0.080	1021±29	5.184(2.3)	5.076(1.6)	984	5.044(2.8)	40.8	4.919(2.9)	32.7	5.187(2.1)
7.944±0.080	1386±57	4.839(4.0)	3.492(1.0)	990	3.480(1.0)	44.2	3.482(2.5)	39.7	3.680(3.5)
8.457±0.090	1649 67	8.069(2.7)	4.893(3.0)	1000	4.996(0.8)	51.9	4.575(2.4)	54.9	4.423(2.5)
	1671 59	8.256(3.5)	4.942(0.5)		4.945(1.0)		5.001(3.7)		4.880(2.1)
	[a]1660±33								
8.975±0.090	1904±79	7.272(3.0)	3.820(2.9)	998	3.987(1.8)	59.6	3.728(3.2)	69.5	3.603(3.0)
9.445±0.090	1978 55	11.071(2.7)	5.597(0.6)	982	5.670(2.5)	65.8	5.583(2.2)	80.6	5.561(2.1)
	1925 47	9.766(2.4)	5.074(0.3)		5.056(2.5)		5.105(2.5)		5.066(2.1)
	[a]1952±45								
9.934±0.100	2090±62	11.755(2.5)	5.625(1.6)	974	5.962(4.0)	72.1	5.594(2.4)	90.0	5.562(2.2)
10.450±0.110	2158±77	10.923(3.0)	5.061(1.9)	978	5.350(6.3)	79.8	5.269(4.0)	98.7	4.982(2.1)

[a]Accepted value, average of two irradiations.

[b]Reference 18 for all the three monitor reactions.

[c]The quoted errors do not include the estimated systematical error of ±3.7%.

Irradiations of 9 to 36 h were performed. Time variation of the neutron flux density was monitored by the fission chamber for the correction of the activities. Experiments at 8.457 and 9.445 MeV were repeated to check the reproducibility of our procedures. Additional irradiation of the sandwich with empty gas target was devoted to account for the (d,n) neutrons emerging from the structural materials at a deuteron bombarding energy of 7.834 MeV (which would correspond to the neutron energy of 10.450 MeV with deuterium gas in the target). Such an experiment was always performed for the ^{238}U(n,f) monitor.

III. EVALUATION OF THE EXPERIMENTAL RESULTS

A. Determination of the foil activities

The method of β-γ coincidences was used to determine the activity of the Al foils. The activity of the Fe (and In, Zn) and Al foils (in some cases for controlling the two techniques) was measured by a Ge(Li) γ spectrometer. Total losses of events from peaks caused by dead time and random pileup were determined using a pulse generator peak in the spectrum. The program GAMANAL (Refs. 10 and 11) was applied to the analysis of the spectra. Absolute full-energy peak and total efficiencies of the detector at various sample distances were determined by standard pointlike as well as calibrated ^{226}Ra sources 19 mm in diameter. Uncertainty of the efficiency curve, fitted by power functions, is 2—3% for the 200—1600 keV region. The estimated systematical error is 1.7%.

The activity of the Th samples was measured by a HP Ge detector. A plexiglass sheet of 5.4 mm thickness (on the Be window) prevented the crystal from overloading by β particles. The reaction residual nucleus, ^{231}Th, is the end product of the ^{235}U α decay, too. Using the idea of Perkin and Coleman[12] the efficiency calibration of the detector from 25 to 205 keV was carried out by ^{235}U samples (enrichment 99.3%) 19 mm in diameter with various thicknesses. (Some pointlike standard sources were also counted for control.) The self-absorption in the thick uranium and thorium samples is similar, therefore the precision of the measurements is not affected very much by the uncertainties of the attenuation correction. The reality of these corrections (controlled by attenuation experiments, too) is verified by the fact that the absolute photopeak efficiencies determined by ^{235}U samples of 0.3 and 0.024 g/cm^2 thickness have not shown any systematical variation as a function of the γ energy and agree well within the statistical errors (1—6 %). The activity of the ^{235}U samples, prepared from oxide powder, was calculated with data of Ref. 13.

Spectra of the Th and U samples were evaluated on a NOKIA LP-4900 analyzer used for acquisition. Activity of the irradiated thorium was determined by the weighted average of the values calculated from the intensity of the γ peaks of 25.64, 81.24—82.11 (complex), 84.21, and 102.27 keV. The weighted average of the activities, $\bar{A}$, and its standard deviation Δ were computed for N data of $A_i \pm \Delta_i$ as

$$\bar{A}=\frac{\sum_{i=1}^{N} A_i/\Delta_i^2}{\sum_{i=1}^{N} 1/\Delta_i^2};\quad \Delta^2=\frac{\sum_{i=1}^{N} (\bar{A}-A_i)^2/\Delta_i^2}{(N-1)\sum_{i=1}^{N} 1/\Delta_i^2}\,. \tag{1}$$

No systematical γ-energy dependence was found in the activities of a given sample. All the Th discs were counted before the irradiation to determine the intensity of the 81.30 and 84.48 keV lines originated from the ^{232}Th natural radioactivity. Both the uranium and thorium samples were measured also by the Ge(Li) spectrometer for intercalibrating the two systems.

Nuclear data (half-lives, decay schemes for the cascade coincidence corrections) and γ-branching ratios were taken from Refs. 14 and 15, respectively.

The self-absorption corrections (taking into account the variation of the detector efficiency and the activating neutron flux along the Th discs) were based on coefficients of Ref. 16.

Several (six to ten) γ spectra were taken for each Th and two to three for the other samples. Decay curve analysis on the basis of the least squares fitting procedure was carried out for all the activity measurements. No interference from fission products was observed when analyzing results for ^{231}Th γ peaks (energy resolution was 0.2—0.4 keV, FWHM).

B. Determination of the fission rates

The fission chamber, continuously fed by Ar gas, contained a 0.28 mg/cm^2 thick uranium layer (>99.99% ^{238}U). Its weight was certified with an accuracy of $\pm$2%.

The following corrections were applied to the measured fission events:

(i) losses in pulses due to the discriminator threshold, calculated from pulse height spectra (max 1.5%);

(ii) laboratory angular distribution of fragments (0.9—1.4 % as a function of the neutron energy);

(iii) fragment self-absorption in the layer (1.7% for all energies).

C. Corrections for the neutron field

The effect of the ^{2}H(d,np) neutrons on the fission chamber counts was determined by measuring the neutron spectrum with a time-of-flight spectrometer and using differential cross sections for this reaction and the ^{2}H(d,n) reactions from Refs. 17 and 5, respectively, for normalization. The reaction rates caused by the parasite neutrons were then calculated with the excitation function of the ^{238}U(n,f) reaction given in Ref. 18. The highest correction at a deuteron energy of 7.433 MeV (after the Mo window), when the activating neutrons have a mean energy of 10.450 MeV, is 19%, which is lower by about 20% than that calculated from data in Ref. 19.

Background neutrons from (d,n) reactions on the structural materials (window, beam stopper, etc.) were detected in experiments mentioned in Sec. II. The maximum of these corrections at 10.450 MeV neutron energy is 8.3% for ^{238}U(n,f) and about 1% for ^{56}Fe(n,p), ^{27}Al(n,α), and ^{232}Th(n,2n).

The reliability of all these corrections can be deduced from the flux densities determined by the $^{115}In(n,n')^{115}In^m$ reaction. In the high energy region (9.445–10.450 MeV) no systematical deviation from the flux given by the other reactions was found, although the total correction in this case exceeds 80% at 10.450 MeV.

It may be worth noting that the reaction $^{64}Zn(n,p)$ gave much lower (by about 32%) flux densities than the others, independent of the data base used.[4,20] In this case the activity was determined through the detection of the ^{64}Cu annihilation radiation using two Al stoppers of 1 mm thick each. A similar effect had been observed earlier when detecting $\pm\beta$ rays of the ^{64}Cu from the $^{65}Cu(n,2n)$ process. Perhaps decay data other than those of Ref. 14 ought to be used.

Variation of the neutron flux density with distance along the sandwich was about 3%/mm.

IV. RESULTS AND DISCUSSION

Results of our measurements are summarized in Table I and shown in Fig. 1.

A. Analysis of the results

Data for the monitor reactions[18] were selected on the basis of our earlier investigations.[21] The random errors, quoted in the table and depicted in the figure, are standard deviations (in 68% sense) and were constructed as follows.

Uncertainty of the reaction rate, $\phi\cdot\sigma$, of the process $^{232}Th(n,2n)$ contains standard deviation from the weighted averaging of the activities from the different γ rays (involving statistical and fitting errors) as well as a $\pm 2\%$ estimated random error of the efficiency of the HP Ge detector, added quadratically. The statistical error and the estimated uncertainty of the corrections, mainly for the $^2H(d,np)$ neutrons, were quadratically composed to calculate the error of the neutron flux density determined by the fission chamber. Statistical and fitting errors were combined with the estimated uncertainty of the Ge(Li) detector efficiency (2%) for the reaction $^{56}Fe(n,p)$. The same is valid for the β-γ coincidence measurements for the $^{27}Al(n,\alpha)$ monitor, but the detection efficiency and background were estimated to have an error of $\pm 1.5\%$. Errors of decay data were neglected. The accepted neutron flux densities in Table I were then calculated with weighted averaging [see Eq. (1)] the results of the three reactions.

The quoted error of the (n,2n) reaction cross section is a propagation of the constituents. Where two irradiations have been performed the accepted cross section is a simple average with an estimated error.

The listed errors, ranging from 1.5% to 13.7%, do not contain systematical ones, which can be estimated as follows. The systematical uncertainty of the Ge(Li) detector efficiency may be $\pm 1.7\%$ (cf. Sec. III A), while that of the β-γ experiments is supposed to be $\pm 1\%$. A $\pm 2\%$ error can be attributed to the results of the fission chamber (see Sec. III B). The final systematical error of the flux determination is expected not to exceed $\pm 2\%$ taking into account the correlation in the averaging process. The activity of the ^{231}Th nuclei may have a $\pm 1\%$ systematical error (mass of the U and Th samples, relative self-absorption). Thus the systematical uncertainty of our experiments is $\pm 2.2\%$ (composed quadratically). The precision of the monitor reaction cross sections (being correlated to an unknown extent) can be estimated to be $\pm 3\%$, on average. That is why an overall value of $\pm 3.7\%$ should be added quadratically to the figures quoted in Table I, giving precisions from 4% to 14.2%.

An obvious disagreement between the flux densities determined by the reactions $^{238}U(n,f)$ and $^{56}Fe(n,p)$ and the reaction $^{27}Al(n,\alpha)$ can be observed up to 7.190 MeV neutron energy. As regarding the early work performed by Schmitt and Halperin,[23] the excitation function of the $^{27}Al(n,\alpha)$ process may exhibit some structure in this region which has been smoothed out by recent compilations.

The repeated irradiations gave differences of 1.3% and 2.8% that would not contradict the arguments on the error estimations given above.

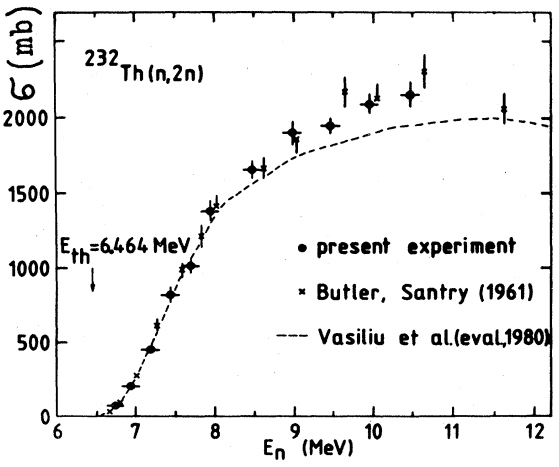

FIG. 1. Results of the present experiment for the excitation function of the reaction $^{232}Th(n,2n)$. Renormalized experimental data from Ref. 3 together with evaluation of Ref. 24 are also shown for comparison. (A systematical error of $\pm 3.7\%$ is not included in the plotted uncertainties of our results.)

B. Comparison to literature data

Results given by Butler and Santry,[3] renormalized with data for the $^{32}S(n,p)$ reaction from Ref. 22, are also shown in Fig. 1. Excellent agreement can be observed in this energy region. Although numerical results of Tewes *et al.*[2] have not been available, it can be stated by examining the figure in Ref. 4 that our measurements support the excitation function given by Butler and Santry. The compilation in Ref. 24, however, shows a different shape for this excitation function from 9.5 MeV (see Fig. 1). It underestimates the experimental values by about 13% at 10.5 MeV. This evaluation was obviously based on the results of Ref. 3, renormalization of which had probably been carried out by monitor data differing very much from those of Ref. 22.

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