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MEASUREMENT OF THE CROSS SECTIONS FOR THE ^{238}U(n,2n) AND ^{232}Th(n,2n) REACTIONS IN THE 13.5 - 14.8 MeV ENERGY RANGE

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Abstract

(n,2n) reaction cross sections have been determined for ^{238}U and ^{232}Th by using the activation method. Neutrons were produced in D-T reaction by bombarding a TiT target with analyzed deuteron beam from a Cockcroft-Walton accelerator. Neutron energy was varied by changing the emission angle to the deuteron beam. The activities of the ^{237}U and ^{231}Th residual nuclei were measured by a Ge(Li) and a HP Ge gamma-spectrometer, respectively. Several standard reactions were used for the determination of the neutron flux density. An overall precision of 3.5 - 4.5 % was estimated and a 1.4 - 3.5 % reproducibility was achieved. Comparison of the recent results to literature data and a brief analysis of the monitor reactions are given.

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1. INTRODUCTION

The knowledge of (n,2n) reaction cross section of fissionable nuclei at 14 MeV are required by the future hybrid reactor technology and the non-destructive analytical methods for safeguards purposes. An other field is the preequilibrium model calculations and the description of the competition between neutron emission and fission.

There were some 31 data from 12 experiments for the reaction ^{238}U(n,2n) in the 13 to 15 MeV interval. Most of them have been determined out by the activation method [Po54,Ph56,Ro57,Gr58,An58,Pe61, Ma69,La73,Ac75,Ve78,Go87] precision of which ranges from 5 to 15 % . The neutron multiplicity technique has given an independent tool for the measurement [Fr74] having greater uncertainties (10-25%). However, the normalized cross sections show discrepancies of about 30 % , generally. It amounts to 50 % in the 13.9 - 14.1 MeV range (Fig. 2). Analysis of the experiments and a compilation of the excitation function have already been published in our paper [Ko82].

Less data are available for the ^{232}Th(n,2n) reaction around 14 MeV. The activation technique has been used in 7 experiments [Ph56, Te60,Zy60,Bu61,Pe61,Pr61,Ka79] to measure 23 points with uncertainties of 5 - 20 % . Although the discrepancies are not much higher (25-30 %) than the quoted errors the shape of the excitation function seems to be not well determined (Fig. 3). This is more clear when analyzing the cross sections from threshold up to 20 MeV [Ko90]. The latest work has been carried out in 1979 [Ka79].

2. EXPERIMENTAL

The experimental conditions for the reactions ^{238}U(n,2n) and ^{232}Th(n,2n) are summarized in Tables I and II, respectively.

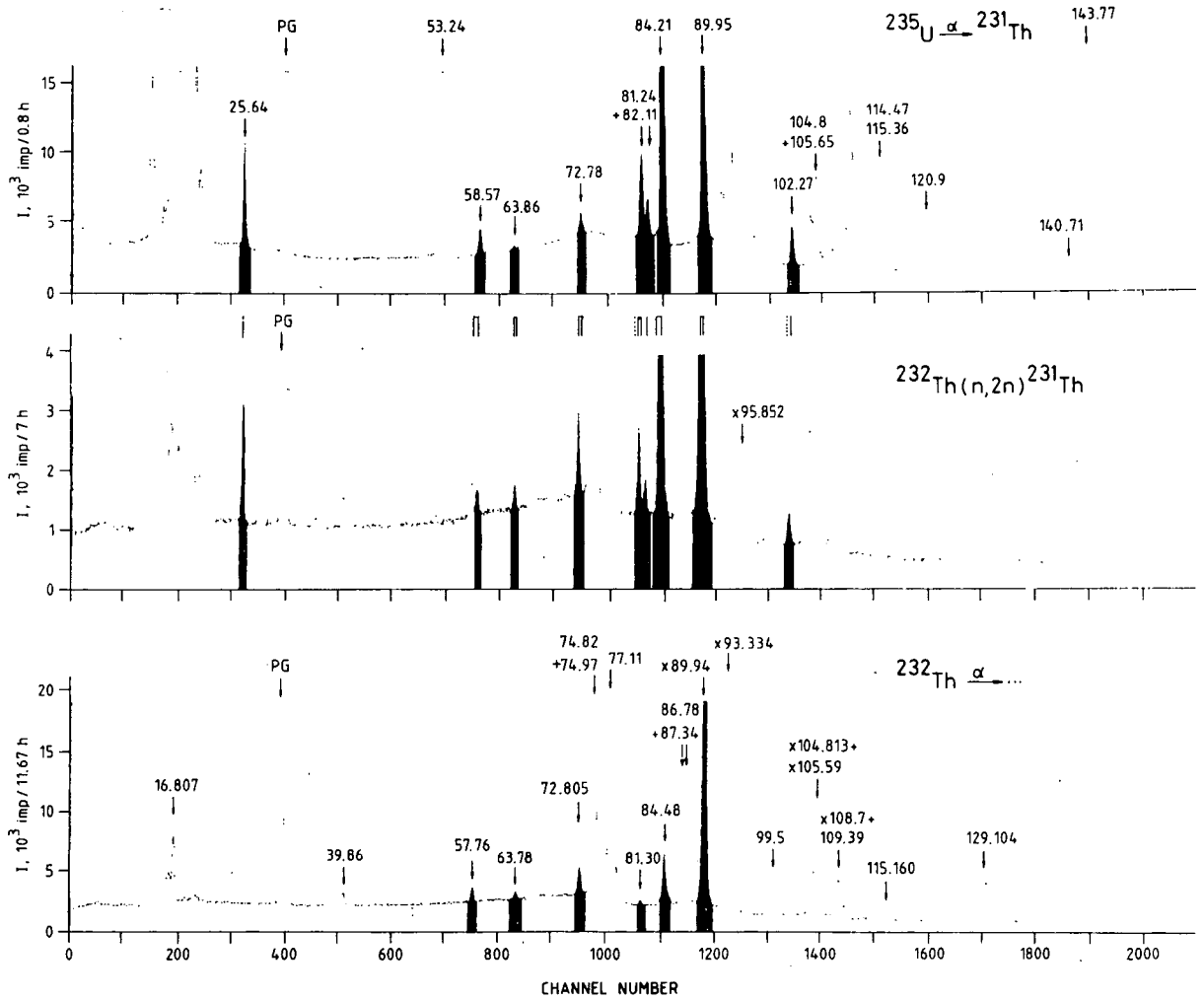

Fig.1. Method for the activity measurement of ^{231}Th

Top: Gamma-lines of a ^{235}U sample. Center: Spectrum of an irradiated ^{232}Th target (cooling time 14.6 h).

Bottom: Gamma-lines from the decay of thorium and its daughters. (PG: pulser, X: X-rays)

Analysis of the high discrepancies in the measured cross sections around 14 MeV shows some technical problems of the experiments. That is why special attention was paid to the circumstances as follows.

(a) Calculation of the mean energy (and dispersion) of the neutrons.

(b) Determination of the flux density by several reactions to avoid possible systematical errors, and to estimate the effect of background neutrons.

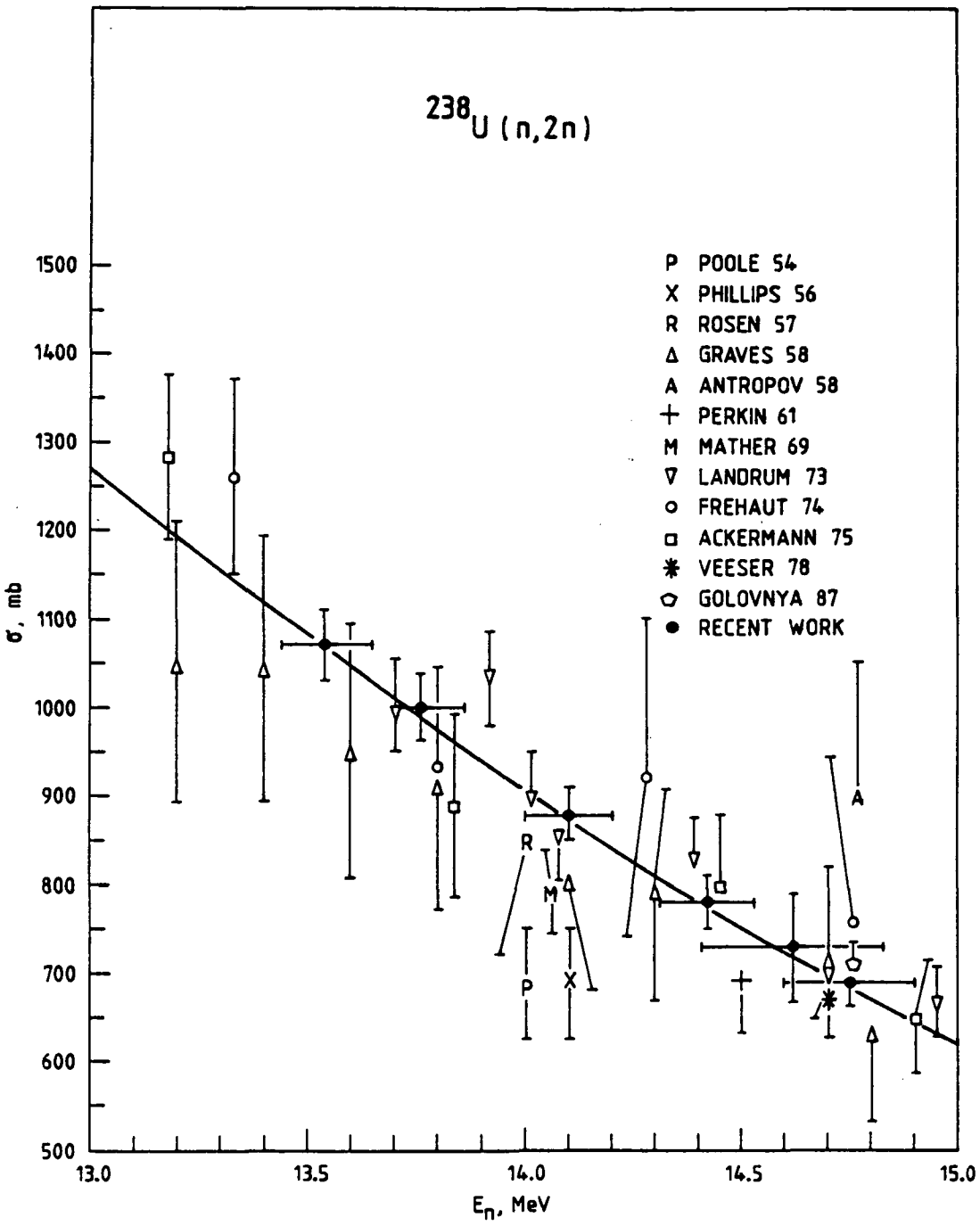

Fig. 2. Excitation function of the $^{238}U(n,2n)^{237}U$ reaction in the 13 - 15 MeV neutron energy range

References for the experimental data: [Po54, Ph56, Ro57, An58, Pe61, Ma69, La73, Fr74, Ac75, Ve78, Go87]. Solid line: Pade-approximation of the excitation function from threshold to 19 MeV [Ko82].

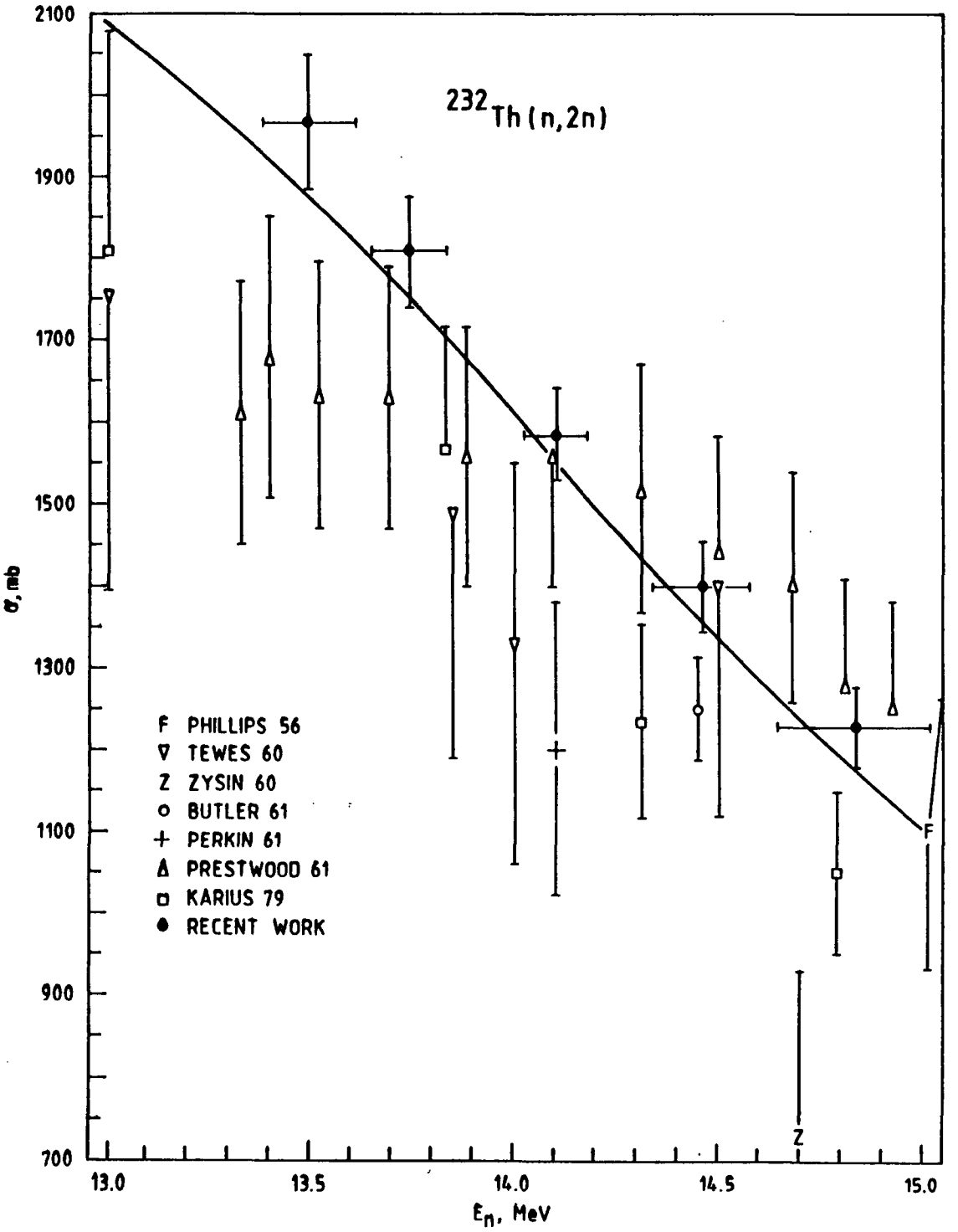

Fig. 3. Excitation function of the $^{232}Th(n,2n)^{231}Th$ reaction from 13 to 15 MeV

References for the experimental data: [Ph56, Te60, Zy60, Bu61, Pe61, Pr61, Ka79]. Solid line: Pade-approximation of the excitation function from threshold to 20.5 MeV [Ko90].

Table I. Details for the cross section measurements of the 238-U(n,2n) reaction

EXPERIMENT/SAMPLE:	1/Db	2/Db	3/Db	4/Db	4/Obn
		magnetically analyzed deuteron beam			
PARAMETERS:					
Deuteron energy:	(120 +/-8) keV	(175 +/-5) keV	(175 +/-5) keV	(135 +/-5) keV	
TiT t. assembly:	thick steel/water c.	thick steel/water c.	thick steel/water c.	thin Aluminum wobbler/air cooling	
Target-sample dist.:	5 mm	6 mm	70 mm	65 mm	
Sample cladding:	0.5 mm Cd	1 mm Cd	1 mm Cd	No	No
position, deg:	0	45	0 +60 +90 +120 -150 -60 -90 -120	-60 -90 -120 -148	0 60 90 120 148
weight (oxid):	2.64 g	0.9967 g	1.6 g	1.6 g	2.7 g
thickness:	1.47 mm	0.67 mm	1.3 mm	1.3 mm	1.6 mm
diameter:	19.1 mm	19.0 mm	19.0 mm	19.0 mm	18.4 mm
U:O ratio:	0.85:0.15	0.85:0.15	0.85:0.15	0.855:0.145	0.90:0.10
238-U content:	0.992745 (natural)	0.992745	0.992745	0.992745	0.9999
Duration of irrad.:	29.25 h	1.25 h	2.0 h	11.0 h	
Flux monitor:	6LiI(Eu), stilben	6LiI(Eu)	6LiI(Eu)	238U(n,f) fission chamber	
measurement:	27Al(n,a):Ge(Li)	27Al(n,a):Ge(Li)	115In(n,n'):Ge(Li)	115In(n,n'), 27Al(n,alpha):Ge(Li)	
			63,65Cu(n,2n):4Pi-beta	238U(n,f):fission chamber (0 deg)	
			27Al(n,a),(n,p):4Pi-beta	27Al(n,p):Ge(Li) (0 deg)	
237-U act.meas.period:	46 d	29 d	63 d	50 d	
acquisition:	1 - 4 h	1 - 15 h	0.3 - 24 h	1.5 - 24 h	
Ge(Li)-spectra:	27	7	8*14=112	9*10=90	
det.-samp.dist.:	50 mm	30 mm	1.4 mm	30 mm	

(c) Activity measurements with precisely calibrated Ge(Li) and HP Ge gamma-detectors as well as 4Π beta-counters.

(d) Corrections (self-absorption in high atomic number elements, counting losses, interferences of gamma-lines, chemical composition of the targets).

Energy calibration of the accelerator has been carried out by reactions ^{11}B(p,gamma) and D(p,gamma) [Cs87,Sz89]. Calculation of the energy distribution of the neutrons from the D-T source was performed by taking into consideration of deuteron stopping in the TiT-layer, the position (0°, 60°, 90°, 120°, 150°) and geometry of the samples (19 mm in diameter, thickness of 0.3-1.6 mm). The uncertainty of the average energy (Tables VI and VII) contains, combined quadratically,

Table II. Details of the 232-Th(n,2n) experiment

PARAMETERS:	DATA:
Deuteron energy:	(200 +/-5) keV (analyzed beam)
TiT t. assembly:	thin Aluminum wobbler/air cooling
Target-sample dist.:	65 mm
Sample cladding:	No
position, deg:	0 60 90 120 150
weight (metal):	0.82 - 0.84 g
thickness:	0.3 mm
diameter:	19.0 mm
232-Th content:	1.0
Duration of irrad.:	9.42 h
Flux monitor:	6LiI(Eu); 238U(n,f) fission ch.
measurement:	27Al(n,a), 56Fe(n,p): Ge(Li) 238U(n,f):fission ch. (0 deg)
231-Th act.meas.period:	8 d
acquisition:	3.4 - 11 h
HP Ge-spectra:	5*5=25
det.-samp. distance:	11.2 mm (with 5.4 mm plexiglas abs.)

Table III. Nuclear data of the reactions used for flux measurement

Reaction	Isotopic abundance % (a)	Effective threshold MeV	Half-life (a)	Radiation, particles	Gamma-energy keV	Gamma-intensity % (b)
115-In(n,n')	95.7	0.339	4.486 h	gamma	336.23	45.8
238-U(n,f)	99.2745	0.80	prompt	fragment		
27-Al(n,p)	100	3.0	9.45 m	gamma	843.76	71.8
					1014.4	28.2
56-Fe(n,p)	91.72	5.0	2.579 h	gamma	846.754	98.89
					1810.72	27.2
					2113.05	14.3
27-Al(n,alpha)	100	6.0	14.97 h	gamma	1368.5	100
					2753.9	99.9
65-Cu(n,2n)	30.83	10.06	12.701 h	positron	(511.0)	18.4 (c)
				beta		38.0 (c)
63-Cu(n,2n)	69.17	11.02	9.74 m	positron		97.8
				beta		2.2

References: (a) Chart of the Nuclides [Wa84],
(b) Reus and Westmeier [Re83],
(c) Martin and Blichert-Toft [Ma70] and Vonach [Vo82].

Table IV. Nuclear data for the (n,2n) reactions

	238-U(n,2n)		232-Th(n,2n)		
Reaction threshold:	6178 keV	(a)	6467 keV	(a)	
Half-life of final nucl.:	6.75 d	(b)	25.52 h	(b)	
Energy and intensity of gamma-lines:	* 208.0 keV	21.8 %	* 25.64 keV	12.8	14.5 %
	267.5	0.73	58.57	0.42	0.48
	332.4	1.21	63.86	-	0.023
		(c)	* 72.78	0.22	0.25
			* 81.24	0.744	0.89
			* 82.11	0.341	0.40
			* 84.21	5.6	6.6
			89.95	0.84	0.90
			93.02	0.0446	-
			99.28	0.118	0.12
			* 102.27	0.38	0.41
				(c)	(d)

* Used for activity determination.

References: (a) Wapstra and Audi [Wa83]
(b) Chart of the Nuclides [Wa84]
(c) Gunnink [Gu75]
(d) Reus and Westmeier [Re83]

one standard deviation of the mean, effect of the multiple scattering of deuterons in the layer (50 keV) and the estimated error of the deuteron energy (5-8 keV). To reduce the computations on the distributions use was made of a series expansion of the non-relativistic kinematics for the neutron energy giving 1-2 keV uncertainty up to 2 MeV deuteron energy [Ra90]. An overall check of the neutron energies was a method using relative cross section measurements on the excitation function of (n,2n) reactions giving residual nuclei ^{89}Zr and ^{92m}Nb [Cs86].

Several monitor reactions have been used to determine the neutron flux density. Nuclear data used for the evaluation are shown in Table III. Reaction ^{238}U(n,2n) was a testing ground to analyze the neutron

Table V. Error estimates for the measured cross sections

	238-U(n,2n)		232-Th(n,2n)	
	random error, %	systematical error, %	random error, %	systematical error, %
FLUX MEASUREMENT				
activity:	0.5-0.9		0.5-1.3	
nuclear data:	1.0	1.0	1.0	1.0
Detector efficiency:	1.0 Ge(Li)	1.5	1.0 Ge(Li)	1.5
			1.0 HPGe	1.7
ACTIVITY OF FINAL NUCLEUS				
sample position, selfabs.:	1.5	1.0	1.2	1.5
weight:		0.7		0.1
peak area:	0.4-1.0		0.4-0.8	
gamma-intensity:		1.0		0.0
OVERALL ERRORS:	3.5-4.5		3.6-4.0	
REPRODUCIBILITY:	1.4-3.5		not measured	

field around different target assemblies (thick stainless steel holder for Mo-backed TiT target with water cooling, light-weight wobbler tube of aluminum with direct mounting of the target and air cooling, source sample distances from 5 to 70 mm, samples with and without Cd-cladding, see Table I). Effects of the background neutrons have been observed through the "apparent flux densities" depending on the threshold of the monitor reactions [Ra79,Ra90]. Corrections for the activity originating from these neutrons were calculated using the "flux - threshold energy" curves. High amount of low energy neutrons was detected by the ^{115}In(n,n')^{115m}In reaction when using the thick target assembly (Table VI).

The neutron flux was measured, in some cases, also by the ^{238}U(n,f) reaction using an argon filled flow-type fission chamber [Na80]. Thickness of the uranium layer was 0.287 and 0.190 mg/cm^2 for the experiments ^{238}U(n,2n) and ^{232}Th(n,2n), respectively. They

Table VI. Analysis of the experiments for the 238-U(n,2n) reaction

angle: (deg)	En MeV	+-dEn	238U(n,2n) σ mbarn	+-dσ	27Al(n,a) (a) Φ cm-2.s-1 1.E6	σ mb	EXP.	27Al(n,p) (b) Φ 1.E6	σ mb	115In(n,n') (h) Φ 1.E6	σ mb	65Cu(n,2n) (c) Φ 1.E6	σ mb	(d) Φ 1.E6	σ mb	63Cu(n,2n) (c) Φ 1.E6	σ mb	(e) Φ 1.E6	σ mb
150:-	13.51	0.12	1072	49	50.6	125.6	3/Db	55.5	81.5	80.7	78.3	41.7	830	47.0	736	50.5	400	48.9	413
148:+	13.55	0.10	1053	38	40.7	125.5	4/Obn			40.1	77.3								
148:-	13.55	0.10	1087	39	42.0		4/Db			40.0	77.3								
	13.54	0.11	1070	39	accepted														
120:+	13.75	0.10	1005	46	49.9	123.3	3/Db	53.1	79.6	76.6	72.0	40.5	870	45.7	772	52.2	411	46.6	459
+	13.76	0.08	982	35	41.3	123.3	4/Obn			41.2	72.0								
-	13.76	0.08	1021	36	44.3		4/Db			44.9									
-	13.76	0.10	997	45	52.3		3/Db	57.2	79.6	86.1		42.6	872	48.1	773	53.8	439	51.2	461
	13.76	0.10	1001	36	accepted														
90:+	14.08	0.10	868	31	30.9	120.4	4/Obn			39.1	65.2								
-	14.08	0.10	891	32	42.5	120.4	4/Db			41.5	65.2								
-	14.10	0.19	895	41	41.0	120.4	3/Db	45.0	77.0	80.5	65.0	32.7	930	37.3	816	42.9	487	40.4	517
+	14.13	0.11	864	39	54.9	120.4	3/Db	56.8	76.7	81.6	64.6	44.0	933	50.2	818	54.8	491	51.6	521
	14.10	0.10	879	31	accepted														
0:	14.41	0.20	753	90	8.0	117.4	1/Db												
60:-	14.40	0.16	782	36	44.2	117.6	3/Db	50.1	74.7	80.9	61.8	35.6	990	41.8	843	49.0	524	45.7	562
+	14.42	0.10	773	28	41.9	117.2	4/Obn			43.3	61.7								
-	14.42	0.10	794	29	44.9		4/Db			45.9									
+	14.45	0.12	778	36	55.6	116.5	3/Db	59.5	74.3	80.6	61.4	43.9	1000	51.8	847	58.9	535	55.4	569
	14.42	0.11	782	28	accepted														
45:	14.62	0.21	732	58	4530	114.0	2/Db	accepted											
0:	14.73	0.15	685	24	43.1	113.5	4/Obn	44.3	72.2	45.0	60.1								
					(f) 42.5	1232:	238-U(n,f)												
					(g) 44.0	1190:	238-U(n,f)												
	14.80	0.17	696	32	58.2	112.5	3/Db	63.5	71.6	85.8	59.8	45.2	1070	55.6	870	64.7	585	61.5	616
	14.76	0.15	689	24	accepted														

References for the monitor cross sections: (a) Kornilov et al. [Ko84] and Tagesen, Vonach [Ta81]
(b) ENDF/B-IV [Ma75]
(c) IRDF [Ma87]
(d) Vonach et al. [Vo68]
(e) Lapenas [La75]
(f) ENDF/B-IV [Ma75]
(g) ENDF/B-V Mod.2. [Bö87]

Table VII. Measured cross sections for the 232-Th(n,2n) reaction

angle: (deg)	En MeV	+-dEn	232Th(n,2n) σ mb	+-dσ	<Φ> cm-2 s-1 1.E6	27Al(n,a) (a) Φ 1.E6	σ mb	56Fe(n,p) (b) Φ 1.E6	σ mb	238U(n,f) Φ 1.E6	σ mb
150:	13.49	0.12	1967	79	22.9	22.6	125	23.6	115	-	
120:	13.74	0.09	1807	69	26.3	26.0	124	26.7	114	-	
90:	14.10	0.08	1585	57	26.5	26.3	121	26.8	113	-	
60:	14.46	0.12	1400	55	26.7	26.3	117	27.4	109	-	
0:	14.84	0.19	1231	47	24.0	23.8	113	24.6	104	23.0	1244 (c)
										23.8	1201 (d)
										23.4	1224 (e)

$$\langle\Phi\rangle = 0.67*\Phi(n,a) + 0.33*\Phi(n,p)$$

References for the monitor cross sections:
(a) Kornilov et al. [Ko84] or Tagesen and Vonach [Ta81] and Vonach [Vo83]
(b) Bychkov et al. [By82] or Lapenas [La75]
(c) ENDF/B-IV [Ma75]
(d) ENDF/B-V Mod.2. [Bö87]
(e) Lapenas [La75] or Sowerby et al. [So74]

were made from U highly depleted in ^{235}U. The following corrections were applied to the measured fission events: losses in pulses due to the discriminator threshold calculated from pulse height spectra, laboratory angular distribution of fragments, and fragment self-absorption in the layer. Monitoring of the time variation of the neutron flux was performed either by the fission chamber or a ^{6}LiI(Eu) or stilben detector. These data were then used to compute the corrections for the initial activities of different half-lives.

Activity of the residual nuclei for the ^{238}U(n,2n), ^{115}In(n,n'), ^{27}Al(n,p), ^{27}Al(n,alpha) and ^{56}Fe(n,p) reactions have been determined by a Ge(Li)-detector of 40 cm^{3} having energy resolution of about 3 keV

(fwhm) at 1000 keV. Photo (total absorption) peak efficiency was measured by point-like standards as well as calibrated extended sources of ^{226}Ra (with daughters), ^{235}U and ^{182}Ta [Na74,Ra90]. Geometry dependence of the efficiency was also investigated in details (distance from the detector surface and axis). Total detection efficiency was determined for the corrections of true coincidence losses. Random pile-up and dead time were calculated using a pulser. This technique has been verified in an international intercomparison experiment [Me85]. A HP Germanium detector of 1.4 cm^3 with 0.5 keV resolution at 100 keV was applied to analyze low energy gamma-rays from ^{231}Th. Its Be-window of 0.127 mm was covered by a 5.4 mm thick plexiglas to prevent the crystal from beta-particles. The detector efficiency has been measured for extended sources of 19 mm in diameter, only. The activity measurements with the two spectrometers were cross-correlated by ^{226}Ra and ^{235}U sources. The self-absorption corrections were calculated with coefficients from tables in Ref. [St70] and verified by absorption experiments. Inhomogeneous activation and the variation of the efficiency along the sample were also taken into consideration.

Activity of the Al and Cu foils has also been determined by 4Π-beta counting using a flow-type proportional counter. The efficiency was measured by calibrated sources. Self-absorption was determined experimentally. Separate experiment was devoted to check the activity measurements carried out by gamma-spectrometry and beta-counting. An agreement within +- 0.7 % was found.

Uranium samples were produced by pressing oxide powder. The chemical composition was checked by gamma-spectrometry for cases quoted as "Db" in Table I. They were of natural isotopic abundance. Discs noted as "Obn" were made from uranium highly depleted in ^{235}U. Their uranium content was verified by X-ray diffractometry and chemical

methods together with gamma-spectrometry. Samples of metal thorium were used in the other experiment.

Several gamma-spectra were taken as a function of the elapsed time (Tables I and II). The initial activity was evaluated from the computer analysis of decay curves giving possibility to observe interferences as well as to check the half-life. Nuclear data for the reactions studied are listed in Table IV.

Activity of ^{237}U was determined through its 208 keV gamma-line. Depending on the energy resolution of the Ge(Li)-detectors there may be some interferences to this peak. Gamma-rays of 205.3 keV energy from the decay of ^{235}U may be taken as a background line to be measured before the irradiations. Fission product of ^{149}Nd with a 211 keV peak has a half life of 1.73 h being easily separable in time. Reaction ^{238}U(n,gamma) results in 209.7 keV photons (3.3 %) with a half-life of 2.35 d after the beta-decay of ^{239}U (23.5 m) to ^{239}Np. Its effect depends on the slow neutron background varying strongly with the experimental conditions. This contribution was determined by decay curve analysis of the complex peak and by the help of the 277.6 keV line (14 %) of ^{239}Np.

Low energy gamma-lines of ^{231}Th have been measured by the HP Ge detector. The absolutization of the activity determination suffers from two difficulties. (i) Branching ratios given in the literature have 10 - 20 % uncertainties even for the most intense transitions (Table IV), and (ii) the self-absorption is very hard to calculate for such low energy photons (its values for the lines of 25.64 and 102.27 keV are 0.061 and 0.81, respectively, for the present samples). That is why the method developed by PHILLIPS [Ph56] for beta-counting has been adopted for gamma-spectrometry utilizing the same lines after the alpha-decay of ^{235}U and following the beta-decay of ^{231}Th. Thus absolute intensities are based on the radioactivity of ^{235}U and the

determination of the self-absorption reduces to relative values for uranium and thorium. To decrease the possible systematical errors further we evaluated the ^{231}Th activity using six gamma-lines in the 25.64 - 102.27 keV region (Table IV). Interferences originate from peaks due to the decay of ^{232}Th and its daughters. This problem was again solved by acquisition of spectra before the irradiations together with the analysis of decay curves. The principle of this method is shown by the gamma-spectra in Fig.1.

Precision of the measured cross sections is analyzed in Table V. All data are expressed in one standard deviation (confidence level of 68 %) and its relative value in per cent. The overall errors were quadratically combined from the random and systematical components. Reproducibility was estimated from experiments 3 and 4 on uranium. Its values are in good agreement with the estimated uncertainties.

3. RESULTS OF THE EXPERIMENTS

Our measured cross sections as well as literature data for comparison are shown in Figs. 2 and 3 for uranium and thorium, respectively. The results are summarized in Tables VI and VII. Here are also listed the details of the final calculations including the neutron flux densities and the monitor cross sections. These values together with the nuclear data in Tables III and IV make it possible to renormalize our results by compilators. We have also evaluated the excitation functions of these (n,2n) reactions [Ko82,Ko90].

^{238}U(n,2n)^{237}U

Experiments 1 and 2 were of single energy irradiations having the lowest and highest neutron fluences. Detailed investigation on the

neutron field was performed in measurement 3 where the possible problems of the scattered neutrons were observed. The last irradiation was devoted to check the earlier results using better experimental conditions.

Effect of background neutrons is clearly seen as higher "apparent flux" given by the reaction ^{115}In(n,n') in experiment 3. It was reduced drastically by the light weight aluminum target assembly used in the last measurement. Neutron flux densities determined by the reactions ^{27}Al(n,alpha) and ^{238}U(n,f) are in good agreement using data from Kornilov et al. [Ko84] or Tagesen and Vonach [Ta81] for (n,alpha) and ENDF/B-IV [Ma75] or ENDF/B-V Mod.2. Dosimetry File [Bö87] for (n,f) (see Tables VI and VII). This is also true for ^{27}Al(n,p) in the last experiment. The fluxes were increased by low energy neutrons when using the stainless steel target system. There is some discrepancy for the other monitors. Cross sections of VONACH et al. [Vo68] and LAPENAS [La75] for the ^{65}Cu(n,2n) and ^{63}Cu(n,2n) reactions, respectively, resulted in better agreement for the neutron fluxes than the recent compilations in the BOSPOR and IRDF files [Ma87]. More than 15 per cent discrepancy may be noted between the results obtained by data of the BOSPOR library [Ma87] and VONACH [Vo68] for ^{65}Cu(n,2n). Better agreement was found for ^{63}Cu(n,2n) with data of the IRDF and LAPENAS but other compilations would give much higher differences. These problems may be explained by the nuclear characteristics of the residual nuclei of the (n,2n) reactions on the copper isotopes (half-life, energy of the beta-particles, positrons, branching ratios) affecting the precision of detection. Therefore, these reactions can not be used as standards.

Cross sections of the ^{238}U(n,2n) reaction were calculated using fluxes determined by the ^{27}Al(n,alpha) reaction in all cases. The reason is that their thresholds are very similar thus the unreliable

data of the copper reactions did not make it worth while to correct for a very small energy difference, and this monitor has the least uncertainty in the compilations. Values quoted as "accepted" in Table VI are weighted averages where the neutron energy interval is not too wide compared to the width of the distribution. Since the agreement of the results is good and the characteristic precision is determined mainly by the uncertainties listed in Table V the quoted errors could be the minimum of the components.

Cross sections of the recent work are compared to other measurements in Fig.2. The agreement with most of the data is generally good within the experimental errors. Some of the old experiments [Po54, Ph56,Ma69] giving single values at 14.0 - 14.1 MeV (irradiations probably at 90° to the deuteron beam) are systematically lower. The trend determined by our data has a slope (-315 mbarn/MeV) higher than that of GRAVES et al. [Gr58] but is similar to those determined by LANDRUM et al. [La73], FREHAUT and MOSINSKI [Fr74] and ACKERMANN et al. [Ac75]. The recommended excitation function is represented by the solid line drawn through the points. It was calculated by the Pade-approximation method [Vi87] applied to all the experimental data from threshold to 19 MeV [Ko82]. The uncertainty of this curve may be about 3 % in the 13.5 - 15.0 MeV interval.

^{232}Th(n,2n)^{231}Th

The irradiation has been performed in the "low scattering" arrangement (cf. Tables I and II). Details of the results are summarized in Table VII. Comparison of our measurement with others from the literature is shown in Fig.3. Neutron flux density was determined by the reactions ^{27}Al(n,alpha) and ^{56}Fe(n,p). It was completed by the ^{238}U(n,f) process at 0°. The agreement of the (n,alpha) and (n,f) monitors is excellent. Data for the (n,alpha) reaction were taken from compilations made by

KORNILOV et al, [Ko84] or TAGESEN and VONACH [Ta81] and VONACH [Vo83] giving the same results. Cross sections in ENDF/B-V Mod.2. and IRDF [Ma87] are higher by 2 - 3 % and lower by 1 - 5 %, respectively, in the 13 - 15 MeV region. Excitation function of BYCHKOV et al. [By82] or LAPENAS [La75] was chosen for the ^{56}Fe(n,p) reaction. Systematical differences may be observed between the reactions (n,alpha) and (n,p) throughout the energy region considered resulting in higher flux densities with the latter. Their values in the order of appearance in Table VII are 4.4, 2.7, 1.9, 4.2 and 3.4 per cent giving an average of 3.4 % . Data from the files ENDF/B-V Mod.2. or IRDF would increase the discrepancy up to 6.5 % . The final flux was calculated from average with weights for the (n,alpha) and (n,p) reactions of 0.67 and 0.33, respectively, reflecting their relative precision.

Data measured by KARIUS et al. [Ka79] are systematically lower by about 15 % than ours. There are some cases, especially from 14.1 to 14.8 MeV, where local agreement may be seen with data of TEWES et al. [Te60] and PRESTWOOD and BAYHURST [Pr61]. Cross section at 14.1 MeV in the last work, having a neutron flux absolutization by the associated particle method at 90°, agrees excellently with our point. These two curves decline mainly at energies achieved in backward angles. Single measurement of PHILLIPS at 15 MeV [Ph56] fits fairly well to our trend. The present results show the highest slope of -534 mbarn/MeV for the excitation function in the 13.5 - 14.8 MeV region. A similar value could be extracted from data of KARIUS et al. [Ka79] in the same interval. Other measurements [Te60,Pr61] would give -260 mbarn/MeV being significantly lower than the slope of the (n,2n) reaction on ^{238}U of much higher fissility.

The solid line in Fig. 3 is again a result of the Pade-approximation performed for the excitation function from threshold to 20.5 MeV [Ko90]. The high energy region should have been based on work of

KARIUS et al. [Ka79] giving low values around 14 MeV. Cross sections measured earlier by us with the same technique [Ra85], together with those of BUTLER and SANTRY [Bu61] have appropriately determined the 6.75 - 11 MeV region but there is an interval from 11 to 13 MeV filled mainly by the very questionable points of TEWES et al. [Te60]. That is why the line in Fig. 3 goes lower than our measured values. Some data have been neglected from the fitting procedure to maintain the physics of the excitation function reproduced also by statistical and preequilibrium calculations [Ja77,Ka79,Ra90,Ra90a].

4. CONCLUSIONS

The present results confirmed the general trend of the $^{238}U(n,2n)$ reaction cross sections around 14 MeV determined by some earlier measurements and greatly reduced its uncertainty. A quite new shape was the result of our measurement for the $^{232}Th(n,2n)$ reaction. Relative error of 3.5 % has been achieved as a minimum applying the conventional activation method. It may be reduced to about 3 % in the cases where the chemical composition of the sample is defined better and the self-absorption is not too high. The main problem of this technique is to find appropriate standard reactions, having thresholds in wide energy range, for the determination of the neutron spectral density. These monitors should have consistent excitation functions tested in various neutron fields.

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