DR-1748 ORNL/TM-6161 (ENDF-260)



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Evaluation of the ²³²Th Neutron Capture Cross Section Above 3 keV

G. de Saussure R. L. Macklin

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ORNL/TM-6161 (ENDF-255) Dist. Category UC-79d

Contract No. W-7405-eng-26

Neutron Physics Division

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G. de Saussure and R. L. Macklin*



*Physics Division.

Date Published - December 1977

Prepared by the OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee 37830 operated by UNION CARBIDE CORPORATION for the DEPARTMENT OF ENERGY

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ABSTRACT

This memo describes an evaluation of the ²³²Th neutron capture cross section in the neutron energy range from 3 keV to 20 MeV. Most existing differential measurements are reviewed, and some data are renormalized to current values of the standards. Several experimentally determined sets of average resonance parameters are also discussed.

From 3 to 50 keV the evaluated cross section is described by a set of average statistical resonance parameters. Above 50 keV the evaluated capture cross section is a smooth curve which follows the trend of the most recent measurements.

The evaluated capture cross section is compared with many measurements and uncertainty estimates are given.

I. INTRODUCTION

The representation of the 232 Th cross sections in the present version of ENDF/B, Version IV, is probably inadequate for the calculation of reactor performance. Indeed, most of the Version IV Th evaluation was taken unchanged from the 1966 Version I evaluation of Wittkopf *et al.*¹ and, hence, ignores the results of several measurements done in the past 10 years.²

This note describes an evaluation of the 232 Th neutron capture cross section above 3 keV, which is part of a larger effort at ORNL and elsewhere to reevaluate the cross sections of Th and 233 U.

In the next section we review the direct measurements of the ²³²Th capture cross section above 2 keV. Several measurements are renormalized to current values of the standards; the results of other measurements are rejected.

In Section III we review several experimental determinations of the 232 Th average resonance parameters and we evaluate a best set of parameters for the neutron energy range 0 to 50 keV.

In Section IV we evaluate the capture cross section above 3 keV. Below 50 keV, the cross section should be represented by "statistical parameters" so that the self-shielding may be properly computed; in that region the best parameters evaluated in Section III appear to yield an acceptable compromise between several inconsistent direct capture measurements. Above 50 keV the evaluated capture cross-section

is a smooth curve which follows the trend of the most recent measurements.

In Section V we attempt to estimate the errors in the evaluated cross section and discuss some of the assumptions used.

II. REVIEW OF THE DIRECT CAPTURE MEASUREMENTS ABOVE 2 keV

We have examined some 20 measurements, reported over a period of more than 30 years and listed in Table I.³⁻²⁴ We will discuss first the measurements done with "24 keV" neutrons from Sb-Be sources or from Fe-filtered beams, then the activation measurements done with monoenergetic neutrons produced by charged particle reactions using an electrostatic accelerator, finally the measurements done by the time-of-flight technique or by the lead-slowing-down technique.

A. Measurements at 24 keV

Some values of the 24 keV capture cross section for 232 Th as well as for 127 I, 197 Au and 238 U are compared in Table II.

The measurements of Belanova^{5,11,19} were done with Sb-Be neutrons and by the spherical shell transmission method. The differences between the 1958 and 1964 values, for both ²³²Th and ²³⁸U, exceed six times the errors quoted. The self-shielding and multiple scattering corrections to the measurements are important, must be obtained by a Monte Carlo calculation and are sensitive functions of the poorly known neutron transport parameters around 24 keV. It is beyond the scope of our evaluation to recompute these corrections.²⁵ On the other hand, it is difficult to choose between the value with a small error reported in 1958 and that with a much larger error reported in 1964, and it does not make much sense to average between two values so inconsistent. For these reasons, we decided to ignore the result of the spherical shell transmission method altogether.

The activation measurements of Macklin *et al.*⁴ and of Chaubey and Sehgal¹⁸ were also done with Sb-Be neutrons, and were done relative to the ¹²⁷I capture cross section which was taken to be .82 \pm .06 b, following a separate measurement of Macklin.⁴ Macklin *et al.* determined the amount of ²³³Th produced by detecting with a NaI crystal the .310 MeV gamma radiation from the ²³³Pa daughter; they assumed .9 γ rays of .310 MeV per disintegration and apparently did not correct for the fact that approximately 50% of the transitions are internally converted and, hence, could not have been detected.²⁶ If we correct the value reported by Macklin *et al.* for the reduction in efficiency due to the internal conversion, we obtain a cross-section value near 1 b., which is considerably higher than all the other reported values. We further note that the cross-section obtained by Macklin *et al.*⁴ for Au is also larger by a factor of two than the presently accepted value. For these reasons we decided to also ignore the ²³²Th measurement of Macklin *et al.*

Chaubey and Sehgal measured the Th activation by detecting the β -rays from ²³³Th in a thin window beta counter. Since more than 97% of the ²³³Th β -rays are either to the ground state or to very low energy states of ²³³Pa, there is no important efficiency correction.²⁶ We also note that the value of the Au capture cross-section reported by Chaubey

Year	Authors	Energy Range (keV)	Points	Comments
1946	Linenberger and Miskel ³	3-390	7	Activation
1957	Macklin, Lazar and Lyon ⁴	24	1	Sb-Be Source, Activation
1958	Belanova ⁵	25.,220.,830	3	Spherical Shell Transmission
1958	Leipunski <i>et al.</i> ⁵		1	Activation
1958	Perkin, O'Connor and Coleman ⁷	14500.	1	Activation
1959	Barry, O'Connor and Perkin ⁸	300-1200	10	Activation
1959	Hanna and Rose ⁹	100-1230	13	Activation
1960	Block and Slaughter ¹⁰	up to 6.	10	Time-of-flight
1961	Belanova ¹¹	220.	1	Spherical Shell Transmission
1961	Stavisskii <i>et al</i> . ¹²	30-964	25	Activation
1962	Miskel <i>et al.</i> ¹³	32-3970	26	Accivation
1963	Tolstikov <i>et al.</i> 14	5.5-102.	10	Activation
1963	Moxon and Chaffey ¹⁵	3-143.	98	Time-of-flight
1963	Stupegia, Smith and Hamm ¹⁶	191-1170	22	Activation
1964	Macklin, Pasma and Gibbons ¹⁷	10-60	136	Time-of-flight
1965	Chaubey and Sehgal ¹⁸	24	1	Sb-Be Source, Activation
1966	Belanova $et \ al.$ ¹⁹	24	1	Spherical Shell Transmission
1971	Lindner <i>et al</i> . ²⁰	121-2730	30	Activation
1971	Forman et al. ²¹	up to 30.	28	Time-of-flight
1971	Stavisskii <i>et al</i> . ²²	up to 35.	10	Lead-slowing-down Spectrometer
1976	Yamamuro <i>et al.²³</i>	24	1	Fe-filtered beam
1977	Macklin and Halperin ²⁴	2-800.	(95)	Time-of-flight

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Table I. Direct Measurements of the ²³²Th Capture Cross Section above 2 KeV

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¹²⁷ I	¹⁹⁷ Au	^{2 3 2} Th	^{2 3 8} U	Author (year)	Method
	57+ 03	.457±.004	.572±.007	Belanova $(1958)^5$ Belanova $(1964)^{19}$	Sb-Be, Shell Transmission
(.82±.06)	1.12±.11	.5±.1	.61±.061	Macklin <i>et al.</i> $(1957)^4$	Sb-Be, Activation
(.82±.06)	.5±.035	.48±.05		Chaubey and Sehgal $(1965)^{18}$	Sb-Be, Activation
.75±.045 .769	.68±.05 .664	.49±.04	.50±.035 .491	ENDF/B-IV	fe-filtered beam (evaluation)

Table II. Neutron Capture Cross Sections near 24 keV

Values in parenthesis were taken as standard.

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and Sehgal agrees much better with present estimates than did that of Macklin *et al*. Hence, we accept the value of Chaubey and Sehgal as given.

The measurements of Yamamuro *et al.*²³ were done with 24 keV neutrons from an electron Linac filtered through 15 cm of iron.²⁷ The prompt capture gamma rays were detected by two C_6F_6 scintillators, using the pulse height weighting technique to determine the absolute capture probability. The flux was measured by using a thick ¹⁰B sample. The ²³²Th capture cross section reported by Yamamuro *et al.* agrees well with that of Chaubey and Sehgal. However for I-127 and Au-197, there are small discrepancies between the values reported by the two groups; this suggests that the good agreement for ²³²Th may be somewhat fortuitous. It must be remembered, too, that the energy distribution of the Fe-filtered neutrons is not exactly that of the Sb-Be source.

B. Activation Measurements with Neutrons from Particle Reactions

1. Linenberger and Miskel (1946)³

This was probably the first 232 Th capture cross-section measurement in the keV range. The neutrons were produced by the Li(p,n) and D(d,n) reactions, the capture cross-sections at 7 neutron energies were measured relative to the 235 U fission cross-section. The cross-section ratio was normalized at thermal energies using neutrons from a graphite thermal column. The results, as reported, are listed in the first three columns of Table III. Col. 4 has the ratio of 232 Th capture cross-section (col. 3) to the 235 U fission cross-section (col. 4 has the ratio of 232 Th capture cross-section (col. 3) to the 235 U fission cross-section

(1) E	(2) 0 e ²⁵	(3) σ(Th)	(4) Ratio	(5) Renormalized	(6) σ _ε ²⁵	(7) σ_,(Th)
(keV)	і (b)	nγ (b)			(b)	пү (b)
3±3	6.32	.958	.152	.174	5.00	.870
$16\pm_{7}^{12}$	2.80	.568	.203	.233	2.48	.578
$33\pm_{20}^{35}$	2.44	.574	.235	.270	1.98	.535
$67\pm\frac{40}{35}$	2.10	.414	.197	.226	1.74	.393
$98\pm\frac{35}{30}$	1.94	.363	.187	.215	1.57	.338
195± ⁵ 20	1.65	.278	.168	.193	1.30	.252
390± ⁵ 20	1.46	.167	.114	.131	1.19	.156

Table III. Renormalization of the Data of Linenberger and Miskel³

(4) is normalized to a thermal ratio³ 6.03/545 = .01106(5) is renormalized to a thermal ratio²⁸ 7.4/582.8 = .01270(6) From M. R. Bhat³² (7) Obtained using col. (5) and σ_f^{25} from (6). Errors estimated at $\pm 10\%$.

thermal. In col. 5 we give the same ratio renormalized to a thermal value 7.4/582.8 = .01270. This last ratio was obtained by using the cross sections at .0253 eV recommended in the 1973 Edition of ENL-325.²⁸ Since the Wescott g-factors²⁹ for ²³⁵U fission³⁰ and ²³²Th capture^{2,31} near room temperature are of the order of .98, the ratio of the cross sections at .0253 eV is within 2% of the ratio averaged over a thermal spectrum. In col. 6 of Table III, we list ENDF/B-V ²³⁵U fission cross-sections obtained from an evaluation of M. R. Bhat.³² The renormalized ²³²Th capture cross-sections are listed in the last col. of Table III. No errors are reported for the measurement, but in a plot of their data, the authors show 10% error bars. This probably represents their estimate of the errors.

2. Miskel, Marsh, Lindner and Nagle (1962)¹³

The Th activation was determined by measurement of the β -activity of its 27.4-day daughter 233 Pa, with a calibrated end-window proportional counter. The neutron flux was measured with a 235 U fission chamber, and the 235 U fission cross-sections reported by Allen and Henkel 33 were used. In Table IV we show how we have renormalized the reported 232 Th capture cross section from the 235 U fission cross-section of Allen and Henkel to that recently evaluated by Bhat. 32 . The authors have estimated their maximum error to be 10%.

3. Lindner, Nagle and Landrum (1975)²⁰

In this experiment the Th activation was measured by detecting the 233 Pa β -activity in a counter calibrated with a source of 233 Pa in equilibrium with a known quantity of 237 Np. The neutron flux was

<u></u>	• • • • • •				
(1) F	(2) (Th)	(3) (²⁵	(4) Ratio	(5)	(6) (Th)
Ц	σηγ	ſ	Kallo	⁰ f	nγ
(keV)	(mb)	(b)		(b)	(mb)
32±9	819	2.66	.308	2.00	616
42±11	615	2.46	.250	1.94	485
59±13	409	2.21	.185	1.78	329
69±4	363	2.10	.173	1.72	297
84±16	350	1.97	.178	1.56	277
118±18	306	1.76	.174	1.526	265
112±11	217	1.79	.121	1.54	187
176±21	204	1.56	.131	1.36	178
240±40	177	1.45	.122	1.254	153
247±24	219	1.44	.152	1.25	190
255±10	141	1.43	.0986	1.24	122
430±23	148	1.27	.117	1.17	136
580±70	153	1.21	.126	1.12	142
710±15	145	1.18	.123	1.095	135
790±21	157	1.18	.133	1.098	146
850±20	112	1.17	.0957	1.119	107
870±80 .	148	1.17	.126	1.14	144
990±50	133	1.28	.106	1.213	128
1000±90	138	1.26	.110	1.214	133
1610±40	99.7	1.30	.0767	1.26	96.6
1790±80	84	1.30	.0646	1.275	82.4
2000±90	61	1.31	.0466	1.284	59.8
2720±40	28	1.27	.0220	1.240	27.3
3000±40	23.1	1.24	.0186	1.224	22.8
3650±70	15.1	1.21	.0125	1.185	14.8
3970±70	13.5	1.20	.01125	1.165	13.1

Table IV. Renormalization of the Data of Miskel $et \ al.$ ¹³

(3) σ_{f}^{25} from R. L. Henkel³³ (5) σ_{f}^{25} from M. R. Bhat³²

(6) $\sigma_{n\gamma}^{-}$ (Th) corrected to the 1976 Values of the σ_{f}^{25} Standard

Errors estimated at $\pm 10\%$

determined with a 235 U fission chamber, using the ENDF/B-IV fission cross-sections.³⁴ There may be a 5% systematic uncertainty associated with the absolute counting system. We have not renormalized those data, though, in the energy range of the measurement (.1 to 2.7 MeV) the preliminary ENDF/B-V values of the 235 U fission cross sections are in places as much as 4% below the Version IV values.^{34,35}

4. Barry, O'Connor and Perkin (1959)⁸

In this experiment the neutron flux was monitored by a Hanson-McKibben type long counter. The 232 Th activation was measured relative to the activation of a similar sample of 238 U, and the 232 Th capture cross-section was normalized at 600 keV relative to the 238 U capture cross-section assumed to be 132 mb. The errors were estimated to be about 8%. In Table V we have renormalized the values of this measurement to the ENDF/B-IV value of the 238 U capture cross-section at 600 keV (114.7 mb).

5. Hanna and Rose (1958)⁹

This measurement was done before 1955. The activity produced in a 232 Th sample by a flux of fast neutrons was compared with that produced in a related sample by thermal neutrons. The thermal neutron flux was determined by activation of gold and measurement of its absolute disintegration rate by coincidence counting. The fast neutron flux was measured by a proton recoil proportional counter. The ratio of the 232 Th capture cross-section to that of 197 Au for thermal neutrons was taken from the work of Crocker³⁶ and of Myasicheva *et al.*³⁷ Those authors report a value .0741 + .0011 for this ratio. This is consistent

E	$\sigma_{n\gamma}(Th)$	Renormalized
(keV)	(mb)	(mb)
300+90	206+17	179+15
400±80	190±11	165±10
500±75	172±11	149±10
600±73	195±12	16 9 ±10
700±69	187±12	162±10
800±67	172±11	149±10
900±66	173±10	150±9
1000±65	120±8	104±7
1100±64	120±8	104±7
1200±62	118±8	103±7

Table V. Renormalization of the Data of Barry et al.⁸

Standard deviations are given; systematic errors are estimated at 8%.

with the presently accepted value²⁸ (7.40/98.8 = .0749). Hence, no renormalization of the values of Hanna and Rose is required. The errors reported are between 8 and 10%.

6. Stavisskii and Tolstikov (1961)¹²

This activation measurement was done relative to the capture crosssection of 127 I, and, in effect, originally normalized at thermal energies to a value of the 127 I thermal capture cross-section 5.66 b. 38 In Table VI, we have renormalized the 232 Th capture cross-sections as reported in ref. 38 to ENDF/B-IV values of the 127 I standard.

7. Tolstikov, Sherman and Stavisskii (1963)¹⁴

This activation measurement was made relative to the ${}^{10}B(n,\alpha)$ cross-section, assumed to be 1/v in the neutron energy range 1-200 keV. The 232 Th cross section was normalized to a value of 390 ± 59 mb obtained in the measurement of Stavisskii and Tolstikov. 12 In Table VII, we have renormalized the cross section to a value of $.49 \pm .04$ b at 24 keV, from the measurements of Chaubey and Sehgal¹⁸ and Yamamuro *et al.*²³

8. Stupegia, Smith and Hamm (1962)¹⁶

In this activation measurement, the 233 Th produced was determined by detecting the 233 Th β -rays in a calibrated end-window proportional counter. The neutron flux was measured with a 235 U fission chamber. The authors do not report what values of the 235 U fission cross-section were used, but those were probably³⁹ the values of the 1965 Edition of BNL-325.⁴⁰ With this assumption, we have renormalized the 232 Th capture cross-sections to the ENDF/B-V values of the 235 U fission cross sections,

		·			
(1)	(2)	(3)	(4)	(5)	
E	$\sigma_{n\gamma}(Th)$	σ _{nγ} (I-127)	σ _{nγ} (I-127)	σ _{nγ} (Th)	
(keV)	(mb)	(mb)	(mb)	(mb)	
30±14	596±54	920	691	406±55	
41±14	547±55	800	590	367±53	
55±14	569 <u>+</u> 34	670	502	308±46	
76±15	403 <u>+</u> 26	530	396	273±34	
100±15	330 <u>+</u> 18	400 .	309	232±30	
133±18	274 ± 14	285	250	218±27	
185±22	218 <u>+</u> 16	233	220	187±23	
221±22	214±11	200	185	180±27	
296±27	198±14	175	147	158±20	
339±51	167±12	157	. 150	145±20	
313±23	168±7	167	.154	141±16	
403±24	173±11	145	140	151±18	
331±23	161 <u>±</u> 6	162	151	135±15	
422±24	166±8	141	138	148±17	
490± 25	150±4	130.5	128	134 <u>+</u> 16	
513±26	164±7	128	126	146 <u>+</u> 17	
580± 26	167±5	119	120	152 <u>+</u> 18	
605±27	156±12	116.5	117	142 <u>+</u> 20	
677±28	165±10	105	109	155±19	
696±29	167±7	103	107	158 <u>+</u> 28	
770±28	173±10	88	98	188±24	
865±10	167±10	82	. 88	163 <u>+</u> 20	
883±30	153±7	81	87	153±17	
964± 31	149±5	81	84	141 <u>+</u> 17	

Table VI. Renormalization of the Data of Stavisskii and Tolstikov¹²,³⁸

(2) Thorium cross section measured

(3) I-127 cross section standard used

(4) I-127 ENDF/B-IV

(5) Renormalized Th-cross section: .908 x (col. 2) x (col. 4)/(col. 3). The factor .908 correction is employed in ref. 38.

(1)	(2)	(3)	
E	σ _n γ	σ	
(keV)	(mh)	(mb)	
5.5±4.7	1648±272	1038±171	
11.5±4.8	1112±183	701±115	
16.7±4.9	807±141	509±89	
19.7±5.0	809±141	509±89	
25.7±5.1	764±124	480±78	
30.9±5.2	616±117	388±74	
. 40.7±5.4	591±109	372±69	
51.1±5.6	370±62	233±39	
71.5±6.1	390±60	246±38	
102.1±6.7	262±45	165±28	

Table VII. Renormalization of the Data of Tolstikov et al.¹⁴

The data reported by Tolstikov *et al.*,¹⁴ col. 2, were normalized to a value 390 mb at 71.5, from the measurement of Stavisskii *et al.*¹² The data of col. 3 are renormalized to an evaluated value of 490 ± 40 mb at 24 keV.

(1)	(2)	(3)	(4)	(5)	
Е	$\sigma_{n\gamma}$ (Th)	$\sigma_{\rm f}^{25}$	σ_{f}^{25}	$\sigma_{n\gamma}(Th)$	
(keV)	<u>(m</u> b)	(b)	(b)	(mb)	
191±38	217±15	1.42	1.32	202±15	
290±36	183±13	1.32	1.22	169±13	
394±36	164±11	1.25	1.19	156±11	
482±35	167±11	1.23	1.14	155±11	
491±37	193±13	1.22	1.14	180±13	
493±36	178±12	1.22	1.14	166±12	
493±36	182±12	1.22	1.14	170±12	
523±50	180±17	1.21	1.14	170±17	
590±36	188±13	1.20	1.11	174±13	
684±38	220±15	1.20	1.10	202±15	
689±39	197±14	1.19	1.10	182±14	
705±50	210±20	1.19	1.09	192±20	
785±37	213±14	1.17	1.10	200±14	
791±32	196±13	1.17	1.10	184±13	
809±50	180±17	1.17	1.10	169±17	
885±38	176±12	1.20	1.16	170±12	
887±43	195±13	1.20	1.16	189±13	
978±38	165±12	1.24	1.21	161±12	
988±38	152±11	1.25	1.21	147±11	
1086±39	152±11	1.27	1.22	146±11	
1091±39	156±10	1.27	1.22	150±10	
1170±43	152±11	1.27	1.23	147±11	
	(1) E (keV) 191 \pm 38 290 \pm 36 394 \pm 36 482 \pm 35 491 \pm 37 493 \pm 36 493 \pm 36 523 \pm 50 590 \pm 36 684 \pm 38 689 \pm 39 705 \pm 50 785 \pm 37 791 \pm 32 809 \pm 50 885 \pm 38 887 \pm 43 978 \pm 38 988 \pm 38 1086 \pm 39 1091 \pm 39 1170 \pm 43	$\begin{array}{cccc} (1) & (2) \\ E & \sigma_{n\gamma}^{}(Th) \\ \hline (keV) & (mb) \\ \hline 191\pm 38 & 217\pm 15 \\ 290\pm 36 & 183\pm 13 \\ 394\pm 36 & 164\pm 11 \\ 482\pm 35 & 167\pm 11 \\ 491\pm 37 & 1.93\pm 1.3 \\ 493\pm 36 & 178\pm 12 \\ 493\pm 36 & 182\pm 12 \\ 523\pm 50 & 180\pm 17 \\ 590\pm 36 & 188\pm 13 \\ 684\pm 38 & 220\pm 15 \\ 689\pm 39 & 197\pm 14 \\ 705\pm 50 & 210\pm 20 \\ 785\pm 37 & 213\pm 14 \\ 791\pm 32 & 196\pm 13 \\ 809\pm 50 & 180\pm 17 \\ 885\pm 38 & 176\pm 12 \\ 887\pm 43 & 195\pm 13 \\ 978\pm 38 & 165\pm 12 \\ 988\pm 38 & 152\pm 11 \\ 1086\pm 39 & 152\pm 11 \\ 1091\pm 39 & 156\pm 10 \\ 1170\pm 43 & 152\pm 11 \\ \end{array}$	$\begin{array}{c cccccc} (1) & (2) & (3) \\ E & \sigma_{n\gamma} (Th) & \sigma_{f}^{25} \\ \hline (keV) & (mb) & (b) \\ \hline 191\pm 38 & 217\pm 15 & 1.42 \\ 290\pm 36 & 183\pm 13 & 1.32 \\ 394\pm 36 & 164\pm 11 & 1.25 \\ 482\pm 35 & 167\pm 11 & 1.23 \\ 491\pm 37 & 193\pm 13 & 1.22 \\ 493\pm 36 & 178\pm 12 & 1.22 \\ 493\pm 36 & 182\pm 12 & 1.22 \\ 493\pm 36 & 182\pm 12 & 1.22 \\ 523\pm 50 & 180\pm 17 & 1.21 \\ 590\pm 36 & 188\pm 13 & 1.20 \\ 684\pm 38 & 20\pm 15 & 1.20 \\ 689\pm 39 & 197\pm 14 & 1.19 \\ 705\pm 50 & 210\pm 20 & 1.19 \\ 785\pm 37 & 213\pm 14 & 1.17 \\ 791\pm 32 & 196\pm 13 & 1.17 \\ 809\pm 50 & 180\pm 17 & 1.17 \\ 885\pm 38 & 176\pm 12 & 1.20 \\ 978\pm 38 & 165\pm 12 & 1.20 \\ 978\pm 38 & 165\pm 12 & 1.24 \\ 988\pm 38 & 152\pm 11 & 1.27 \\ 1091\pm 39 & 156\pm 10 & 1.27 \\ 1170\pm 43 & 152\pm 11 & 1.27 \\ \end{array}$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$

Table VIII. Renormalization of the Data of Stupegia et al. 16

 235 U fission cross sections of col. 3, from Ref. 40, of Col. 4, from Ref. 32; col. 5 has the renormalized 232 Th capture cross sections.

as evaluated by Bhat.³² This renormalization is shown on Table VIII.

C. Measurements by Time of Flight

1. Block and Slaughter (1960)¹⁰

The capture cross section was measured up to 6 keV with a ²³²Th sample of 588 b/atom in a large liquid scintillator tank located on a 11.5 m flight path at the ORNL fast chopper. Only preliminary values were given, not corrected for multiple scattering or resonance selfshielding. Because final data have never been reported, the results of this measurement will be disregarded.

2. Moxon and Chaffey (1963)¹⁵

The measurement was done at the Harwell Linac, the capture gamma rays were detected with a "Moxon-Rae" detector on a 30 m flight path; the neutron spectrum was determined by measuring the capture γ -rays from the ${}^{10}B(n,\alpha)$ reaction, assumed to be 1/v. The normalization was done with the "black-resonance technique". We have not renormalized these data.

3. Macklin, Pasma and Gibbons (1963)¹⁷

The measurement was done with a Moxon-Rae detector at the ORNL 3-MeV Van de Graaff, relative to the Ta capture cross-section which was taken as $8.6 \ E^{-.697}$, where E is the neutron energy in keV and where the cross section is given in barns.⁴¹ The capture cross sections of several nuclei were measured, but the data on ²³⁸U and ²³²Th were never published because the high activity due to the associated radioactive decay chains produced large backgrounds which made the reduction of the data unreliable. Preliminary values transmitted to the NNCSC in

(1) E (keV)	(2) $\frac{\sigma_{n\gamma}(Th)}{\sigma_{f}^{25}}$	(3) ^σ f ²⁵ (b)	(4) σ _{nγ} (Th) (mb)	
34.6	0.20±0.03	1.977	395±60	•
24.2	0.21±0.03	2.127	447±70	
17.3	0.24±0.03	2.39	574±60	
12.5	0.25±0.03	2.75	688±70	
. 9.5	0.26±0.03	3.025	786±80	
7.5	0.25±0.03	3.165	790±80	· · · ·
6.0	0.27±0.03	3.60	970±100	
5.0	0.27±0.03	4.00	1080±110	
4.0	0.28±0.03	4.40	1230±125	
3.2	0.29±0.03	5.00	1450±150	

Table IX. Renormalization of the data of Stavisskii $et \ al.^{22}$

The 235 U fission cross-section values of col. 3 were taken from the work of Bhat. 32

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1964 have been much used, but will be ignored in this evaluation. (For 238 U, these preliminary data are approximately 20% higher than ENDF/B-IV)

4. Forman, Schelberg, Warren and Glass (1971)²¹

The ²³²Th capture cross-section was measured from 20 eV to 30 keV, using neutrons from an underground nuclear explosion (Physics-8), a 250 m flight path, modified Moxon-Rae detectors for the γ -cascade measurement and the ⁶Li(n, α) reaction for the neutron flux determination. The absolute uncertainties are estimated to be <u>+</u> 15%. The capture cross section was normalized to a capture area for the level at 129 eV corresponding to a neutron width of 3.5 <u>+</u> .2 mV.

5. Macklin and Halperin (1977)²⁴

The ²³²Th capture cross-section was measured from 2.5 to 800 keV, using neutrons from ORELA. The pulse height weighting method was used with small liquid scintillators to measure the prompt gamma ray energy release following neutron capture. The neutron flux was measured with a thin Li⁶ glass detector, using a prescription of Uttley slightly modified for the Li⁶(n, α) cross section.⁴² The data were normalized at low energy by the saturated resonance technique.

6. Stavisskii, Tolstikov, Chelnokov, Samsonov and Bergman (1971)²²

This measurement was done with a lead-slowing down pile. The capture cross-section of 232 Th was measured relative to the 235 U fission cross-section, and normalized at low energy. In Table IX, we have renormalized the results of the measurement to the ENDF/B-V values of the 235 U fission cross-sections, as evaluated by M. R. Bhat. 32

Several of the measurements of the ²³²Th capture cross-section below 1 MeV, renormalized as described, are compared in Fig. 1 to 4. It is clear that even after some poorly documented data have been rejected, and the remaining data have been renormalized, there are systematic discrepancies of the order of 50% in the magnitude of the cross-section and considerable differences in shape. One could hope that, as experimental techniques improve, the more recent measurements would converge towards the real values, but this does not seem to be the case: below 30 keV, for instance, the two most recent measurements of Forman *et al.* $(71)^{21}$ and Macklin and Halperin $(77)^{24}$ bracket the values of all other measurements. From 600 keV to 800 keV, the cross sections of Macklin and Halperin increase with energy whereas those of Lindner *et al.*²⁰ (76) decrease with increasing energy.

III. AVERAGE ²³²Th RESONANCE PARAMETERS

Table X lists some experimental values of the 232 Th average resonance parameters for s-wave and p-wave neutrons. The values of Ashgar *et al.*,⁴³ Ribon⁴⁴ and Rahn *et al.*⁴⁵ were obtained by statistical analysis of the parameters of the low energy resolved resonances. The values of Seth *et al.*⁴⁶ Uttley *et al.*⁴⁷ and Camarda⁴⁸ were derived from an analysis of transmission curves through several thicknesses of 232 Th, for neutrons in the keV energy range. We have not included in Table X older Columbia University⁴⁹ and Harwell values,⁵⁰ which we consider superseded by the more recent values. We have not included either the values of the average parameters obtained by Forman *et al.*²¹ and by Macklin and Halperin²⁴ by fitting their capture data, since the corre-



Fig. 1. Comparison of several measurements of the 232 Th(n, γ) cross section with the present evaluation in the range 3 to 50 keV. The data have been renormalized as described in the text.



Fig. 2. Comparison of several measurements of the 232 Th(n, γ) cross section with the present evaluation in the range 3 to 50 keV. The data have been renormalized as described in the text.



Fig. 3. Comparison of several measurements of the 232 Th(n, γ) cross section with the present evaluation in the range 10 keV to 4 MeV. The data have been renormalized as described in the text.







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Table A.	Comparison	OT	Measured	Average	Resonance	Parameters	TOL	10

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	10 ⁴ s ⁽⁾	Γ _{nr} (0)	Γ _γ	D,	$10^{4}S^{(1)}$	а	10 ⁴ S ⁽¹⁾
		mV	mV	eV		10 ⁻¹³ cm	(a=8.3610 ⁻¹³ cm)
Ashgar <i>et al</i> . $(66)^{43}$.80±.17	1.36	21.53±.77	18.6			
Ribon (69) ⁴⁴	.89±.11	1.51	21.8±1	17±1	1.4±.5	8.60	1.48±.5
Rahn et al. (72) ⁴⁵	.84±.08	1.4±.15	21.2±.3	16.7	∿.9	8.67	∿1.
Seth <i>et al.</i> $(64)^{46}$	1.2±.5				.5±.25	8.91	.6±.25
Uttley et al. $(66)^{47}$	(.8)				1.64±.24	8.295	1.62±.24
Camarda (74) ⁴⁹	(.85)		(21.2)	(16.8)	1.5±.4	8.6	1.59±.4

s-wave parameters in parenthesis were assumed by Uttley $et \ al$. and by Camarda to derive p-wave strength functions from the measurements.

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l	J ^P	D eV	Γ nr mV	Γ _Υ mV
0	1/2+	17±1	1.51±.15	21.3±.3
1	1/2	17	2.72±.34	21.3±3
	3/2	8.5	1.36	21.3
2	3/2 ⁺	8.5	.75	21.3
	5/2 ⁺	5.7	.5	21.3

Table XI. Evaluated Average Resonance Parameters for ²³²Th

The values of D are proportional to $(2J + 1)^{-1}$. For a given, parity, the values of Γ are equal. The uncertainties in the resonance parameters are correlated as described in the text.

sponding capture cross-sections have already been considered. The result of a recent measurement by Kobayashi *et al.*⁵¹ is mostly sensitive to the value of the effective scattering radius, and hence is not very helpful in evaluating average strength functions. The s-wave parameters given in parentheses in Table X were assumed by Uttley *et al.*⁴⁷ and by Camarda⁴⁸ in deriving the corresponding p-wave parameters. The p-wave strength function S¹ is approximately inversely proportional to the square of the nuclear radius, a; since the several authors use different values for a, the corresponding p-wave strength functions have been corrected to the ENDF/B value of a.⁵² Those corrected values are listed in the last column of the Table.

We shall disregard the strength functions given by Seth *et al.*,⁴⁶ because (1) for both ²³⁸U and ²³²Th, those authors obtain values of the p-wave strength function at least a factor of two samller than those reported by others, and (2) Seth *et al.* give insufficient information about their experiment and its analysis to evaluate the validity of their results.

In Table XI, we list evaluated resonance parameters for the unresolved energy range 3 to 50 keV. These parameters were obtained as follows:

(1) The s-wave average reduced neutron width and average spacing of Ribon⁴⁴ were adopted in preference to those of Ashgar *et al.*⁴³ and Rahn *et al.*⁴⁵ for reasons that are discussed in some detail in Derrien's 1974 evaluation.⁵³ The reduced neutron widths of Ashgar *et al.* and of Rahn *et al.* and the average spacing of

Rahn et al. are consistent with our adopted value.

- (2) Our evaluated capture width is a weighted average of the values reported by Ashgar et al., Ribon and Rahn et al. This average capture width is assumed to be independent of parity and of angular momentum.
- (3) We have obtained a p-wave strength function as the weighted average of the consistent values reported by Ribon, Uttley *et al.* and Camarda. $(10^4 S^1 = 1.6 + .2).$
- (4) To derive p-wave and d-wave average parameters, we have assumed that the level density was proportional to 2J + 1 and independent of parity. We have further assumed that the strength function was only parity dependent (i.e., the same for s-wave and d-wave neutrons).

IV. EVALUATION OF A "BEST" CAPTURE CROSS SECTION ABOVE 3 keV

Fig. 5, from the paper of Macklin and Halperin,²⁴ illustrates the behavior of the ²³²Th capture cross section in the region from 2.5 to 10.4 keV. We see on the figure that above the resolved resonance region (which we take here at 3 keV) there is still considerable resonance structure. This structure becomes relatively less important as the energy increases, because the Doppler effect broadens the peaks, and because the spacing between large peaks decreases as the p-wave levels become more important. Around 50 keV, the room temperature Doppler width becomes comparable with the p-wave level spacing (5 eV) and the cross-section is expected to be much smoother.

Just above the resolved region, the cross-section must be represented by unresolved resonance parameters so that resonance self-shielding may

be properly computed. At high energies, the cross section may be represented by a smooth curve. The transition between the unresolved resonance representation and the smooth cross section representation is conveniently taken at 50 keV, because, as stated above, the cross sections are expected to be smooth above that energy, and the first inelastic level of 232 Th being at 50 keV, the average cross sections below that energy can be represented by a particularly simple formalism, as the sum of s, p and d wave contributions.⁵² (The contribution of neutrons of higher angular momentum is clearly negligible below 50 keV)

In Fig. 1 and 2, we compare the average capture cross-section computed with the evaluated average resonance parameters of Table XI with several of the data discussed in Section II. The capture cross section was computed by the ENDF/B formula given on p. D 15 of reference 52; Table XII lists values of the average capture cross-section computed at several energies. The data shown on the figures are renormalized as indicated in Section II, and the time-of-flight data of Moxon and Chaffey¹⁵ and Macklin and Halperin²⁴ have been averaged over convenient energy intervals for the clarity of the figures.

The computed average cross section is in good agreement with the lead-slowing-down data of Chelnikov *et al.*²² At 24 keV, the computed cross-section, 501 mb, is in very good agreement with the values reported by Chaubey and Sehgal,¹⁸ 480 \pm 50 mb, and by Yamamuro *et al.*²³ 490 \pm 40 mb. The data of Tolstikov *et al.*¹⁴ shown in Fig. 2, were normalized to a value 490 mb at 24 keV, as explained in Section II; those data also agree reasonably well with the computed cross section. The differences

E(keV)	<o<sub>ny (b)></o<sub>
3.0	1.278
5.0	.982
10.0	.730
15.0	.618
20.0	.545
25.0	.491
30.0	.450
35.0	.416
40.0	. 389
45.0	.366
50.0	.346

Table XII. Evaluated Average Capture Cross Section in the Unresolved Resonance Range

between the computed cross section and the activation data of Linenberger $et \ al.^3$ and Miskel $et \ al.^{13}$ is not thought to be very significant, because the activation measurements are fairly old, and the incident neutrons have a wide and poorly known energy spectrum.

The time-of-flight data of Forman $et \ al.^{21}$ are systematically higher than our computed cross-section, by 10 to 20%. On the other hand, the time-of-flight data of Moxon and Chaffey¹⁵ and Macklin and Halperin²⁴ are systematically lower than our computed cross-section with a discrepancy of the order of 30% below 5 keV, decreasing with increasing energy up to 40 keV or so, where the two sets of data agree with the calculated crosssection. Those discrepancies are difficult to understand. At the same time that they measured the ²³²Th capture cross section, Moxon and Chaffey^{15,54} also measured the ²³⁸U capture cross section. The ²³⁸U cross sections were later revised upward by Moxon, because he found a better method to estimate the resonance self-protection correction.⁵⁵ This revision, for ²³⁰U, was of the order of 10% below 6 keV and 4% from 6 to 10 keV, becoming negligible above that energy. Presumably a similar revision should have been done on the ²³²Th data which have a resonance structure comparable to that of ²³⁸U. A correction in that direction would considerably improve the agreement between our computed cross-section and the Moxon Chaffey data.

The discrepancy between the data of Forman *et al.*²¹ and those of Macklin and Halperin²⁴ is extremely puzzling: Both groups have performed area analysis of the large s-wave resonances. Forman *et al.* have done this analysis in the range 1 to 2 keV using Ribon's⁴⁴ values of Γ_n and find an average radiation width of 20.9 mV. Macklin and Halperin have

done their analysis in the range 2.6 to 4 keV, using Rahn's⁴⁵ values of Γ_n , which are essentially consistent with those of Ribon, and find an average radiation width of 19.8 mV. Since both groups find essentially the same average value of the radiation width, they presumably measure the same area under the large s-wave levels, and the 40% difference between the cross sections obtained by the two groups must be due to different estimates of the cross section inbetween the large resonances.

Fig. 5 suggests that too much background could have been substracted from the data of Macklin and Halperin. For instance, in the interval 3480 to 3500 eV, the capture cross section seems to be almost zero (perhaps even negative). In such an interval, there sould be 3 to 4 p-wave levels, since the average p-wave spacing is of the order 5.7 eV, and if we believe the parameters of Table XI, the average contribution of those levels should be of the order of .4 b. However, even if we were to displace the "zero-line" by .4 b., the average cross section of Macklin and Halperin near 3.5 keV would become 1.06 b which is still 30% below the value of 1.43 b of Forman *et al.*

It seems that somewhat "unreasonable" modifications of the average resonance parameters shown on Table X would be required to fit either the data of Forman *et al.*, or those of Macklin and Halperin. Since the parameters evaluated in the preceeding section yield an average cross section which is intermediate between those two measurements, and which is consistent with the measurement of Chelnokov *et al.*,²² we shall adopt the cross section computed with those average parameters as the "best cotimate" of the average cross section below 50 keV.

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Fig. 5. 232 Th(n, γ) effective cross section at several energies. This figure is reproduced from the paper of Macklin and Halperin (ref. 24).

Above 50 keV, calculations of the average capture cross-section must take into account the contribution of higher angular moments and the poorly known competition of the inelastic scattering. Rather than use such a calculation, we have evaluated the capture cross section from 50 keV to 1 MeV by tracing a smooth curve, splitting the difference between the data of Macklin and Halperin²⁴ and those of Lindner et al.²⁰ Those two measurements are the most recent and appear to us as being the most reliable. Above 1 MeV, our evaluation follows the measurements of Lindner et al.²⁰ and of Miskel et al.¹³ which from 1 to 4 MeV are in good agreement. Above 4 MeV, the only measurement available is that of Perkin et $al.^7$ at 14.5 MeV. We believe that the value of 1 mb at 14 MeV, suggested by the work of Qaim,⁵⁶ is probably more correct than the value of 5.2 + .8 mb at 14.5 MeV given by Perkin *et al*. We note that for 238 U, the value given by Perkin et al., 3.3 + .5 mb, is also considerably higher than that later measured by Drake $et \ al.$,⁵⁷ or that evaluated in ENDF/B-IV, 1 mb. The evaluated cross section above 50 keV is given in Table XIII and illustrated in Figs. 3, 4 and 6.

V. ESTIMATED UNCERTAINTIES IN THE EVALUATED CAPTURE CROSS SECTION

From the technique used in deriving the resonance parameters, which describe the evaluated average capture cross section below 50 keV, we estimate the following uncertainties:

- 6% uncertainty in the level spacings, correlated over the spin and parity values
- (2) 10% uncertainty in the reduced neutron widths of the positive parity levels

E(keV)	σ _{nγ} (b)
50.0	.346
110.0	.190
190.0	.150
0.00	.135
430.0	.135
600.0	.150
700.0	.160
800.0	.160
900.0	.150
1600.0	.10
2000.0	.060
4000.0	.0130
4500.0	.010
10,000.0	.0020
Ī4,000.0	.0010
20,000.0	.00050

Table XIII. Evaluated Capture Cross Section above 50 keV

The interpolation between points should be log-log i.e. $\ln(\sigma_{n\gamma})$ is linear in $\ln(E)$.



Fig. 6. $^{2\,3\,2}\text{Th}(n,\gamma)$ cross section. Comparison of this evaluation with ENDF/B-IV.

- (3) 12.5% uncertainty in the reduced neutron widths of the negative parity levels
- (4) 0.3 mV uncertainty in the average capture width of the positive parity levels
- (5) 3 mV uncertainty in the average cpature width of the negative parity levels

We note that a number of assumptions were made in deriving the parameters, as stated in Section III, and we made the further implicit assumptions that the neutron widths and level spacing followed the usual distribution laws and that their average values did not vary significantly with energy below 50 keV. We did not raise the uncertainties to account for those assumptions.

We also note that the unresolved resonance parameters representation, with which we describe the average cross section, is a statistical representation; hence, at any energy or over any energy group, the actual cross section may be very different from the computed average cross section, even if the average resonance parameters are correct.

From 50 keV to 1 MeV, we estimate $a \pm 15\%$ uncertainty correlated over the entire energy range. This is considerably larger than the systematic uncertainties claimed by most of the measurers, but considering that there is a 20% systematic difference between the two most recent measurements,^{20,24} it seems unjustified to reduce the evaluated uncertainty below the values given.

Above 1.5 MeV, we estimate a \pm 30% uncertainty correlated from 1.5

to 20 MeV. Such a large value results from the small number of independent measurements. However, in that energy region the cross section decreases rapidly and the sensitivity of most reactor calculations to the cross sections above 1.5 MeV is expected to be small.

VI. CONCLUSIONS

We have evaluated the capture cross-section of ^{2 32}Th from 3 keV to 20 MeV. Below 50 keV, the evaluation is in terms of unresolved resonance parameters. Those parameters were derived from an analysis of the resolved resonances from measurements of the total cross section, hence, presumably they will also yield a reasonable representation of the total cross section.

In Figs. 3 and 4, we show comparison of our evaluated cross section with various experimental data. In Fig. 6, we compare our evaluation to ENDF/B-IV, MAT 1296. In the Appendix, we give the evaluation in ENDF/B format. There are large systematic discrepancies among the different data sets, which exceed the uncertainties quoted by the experimenters. These discrepancies lead us to assign uncertainties of \pm 15% to \pm 30% to the evaluated cross sections.

VII. ACKNOWLEDGMENTS

The authors would like to acknowledge helpful discussions with M. R. Bhat (NNCSC) and R. W. Peelle (ORNL) concerning the current values of the standards. L. Forman (LASL) and W. S. Lyon (ORNL) supplied unpublished information about their measurements. M. A. Bjerke, J. E. White and R. Q. Wright (ORNL) provided several ENDF/B average cross-sections and set the evaluation in the ENDF/B-Format. C. W. Fu, K. Gwin and J. Halperin (ORNL) gave particularly valuable advice.

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APPENDIX

Partial Listing of MAT = 7296 $[^{232}$ Th, Modification of ENDF(B-IV)]

1. UNRESØLVED RESØNANCE PARAMETRES. FILE I.MT=151

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