# **Thermal Neutron Capture Cross Sections\***

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The Oak Ridge pile oscillator has been used for a survey of the thermal neutron capture cross sections of the elements. Sixty-nine elements from atomic number 3 to 82 have been measured by comparison with gold, whose capture cross section is known from other experiments. From the internal consistency of the measurements, the care in selecting samples, and the accuracy of the value for gold, these values are estimated to be accurate within 5 percent. A comparison is made with the results of other pile oscillator measurements and of other methods such as time-of-flight and activation.

#### THERMAL NEUTRON CAPTURE

#### **Previous Measurements**

**C**YSTEMATIC measurements of the interactions of neutrons with the elements were made almost as soon as the techniques of working with neutrons were known. The first measurements were simple transmission experiments used to determine total cross sections, that is, the sum of scattering and capture cross sections. Since then several methods have been devised for measuring the scattering and capture cross sections separately.

There are several compilations of nuclear data which include thermal neutron capture cross sections. Most charts or tables of nuclear properties which include neutron cross sections contain no evaluation of the probable accuracy of the values. However, in a report by Way and Haines<sup>1</sup> of the thermal cross sections of the 81 stable elements from hydrogen to bismuth known in 1948, estimates of the accuracy are given. Of the 75 elements whose total cross sections had been reported, 16 were probably known within 10 percent; for 15 of the elements there was only one reported experiment. Scattering cross sections had been reported for 61 elements, with 10 of them probably known within 10 percent; for 25 elements only one experiment had been reported. Thermal capture cross sections were known for 73 elements, with 10 of them probably known within 10 percent; for 6 elements only one experiment had been reported, and for 12 elements the cross sections were known only from activation measurements.

With the advent of neutron chain-reacting piles, it became possible to measure the poisoning effects of neutron absorbing materials. Introduction of a neutron absorbing substance into a pile decreases the pile reactivity, and in order to keep the power constant the control rods must be displaced. By proper calibration this displacement may be used as a precision measure of the capture cross section. In this way Anderson et al.<sup>2</sup> found the capture cross sections for several elements. About 39 elements in all were measured by Wattenberg by this method, but only four values have been published outside the project reports. The pile oscillator method for determining capture cross sections was developed as a logical refinement of the aforementioned method.

Since 1948 capture cross sections have been measured at several laboratories. From the Oak Ridge National Laboratory, values for 69 elements are included in this report, in the main with an estimated accuracy of 5 percent. These measurements were performed with the pile oscillator which employs the local depletion of neutrons by the sample; the values were included in the volume "Nuclear data."<sup>3</sup> The Argonne National Laboratory has recently reported values for 53 elements<sup>4</sup> using a pile oscillator that effects a measurable change in the pile reactivity. With the same method the laboratory at Harwell has reported values for 40 elements.<sup>5</sup>

### **Pile Oscillator Method**

The ORNL pile oscillator measures the thermal neutron capture cross section of a sample by moving the sample so as to pulse periodically the neutron density within the pile. The theory of pile oscillations has been previously reported by Weinberg and Schweinler,<sup>6</sup> and the instrument has been described by Hoover et al.<sup>7</sup> It consists of a mechanical system which oscillates a sample back and forth about once a second through an annular, neutron-sensitive ionization chamber, which is located in a constant and intense neutron flux in the graphite reflector of the pile. The local depletion of neutrons around the capturing sample

<sup>\*</sup> Based on a dissertation presented in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the Uni-

 <sup>&</sup>lt;sup>1</sup>K. Way and G. Haines, AECD 2138, Thermal Neutron Cross Sections for Elements and Isotopes (Oak Ridge, Tennessee, 1948; Technical Information Branch, AEC); also published as Document No. PB-95632, Office of Technical Services, Department of Commerce, Washington, D. C.

<sup>&</sup>lt;sup>2</sup> Anderson, Fermi, Wattenberg, Weil, and Zinn, Phys. Rev. 72, 16 (1947).

<sup>&</sup>lt;sup>3</sup> "Nuclear data," Natl. Bur. Standards (U.S.) Circ. 499 (1950). <sup>6</sup> "Nuclear data," Nati. Bur. Standards (U. S.) Circ. 499 (1950).
 <sup>4</sup> Harris, Muehlhause, Rasmussen, Schroeder, and Thomas, Phys. Rev. 80, 342 (1950).
 <sup>6</sup> F. C. W. Colmer and D. J. Littler, Proc. Phys. Soc. (London) 63, 1175 (1950).
 <sup>6</sup> A. M. Weinberg and H. C. Schweinler, Phys. Rev. 74, 851 (1950).

<sup>(1948).</sup> 

<sup>&</sup>lt;sup>7</sup> Hoover, Jordan, Moak, Pardue, Pomerance, Strong, and Wollan, Phys. Rev. 74, 864 (1948).



FIG. 1. Scattering and absorption signals (schematic). A (on) and B (off) are the mechanical rectification points. The cross hatched area represents the signal.

depends on the absorption by the sample and can be used to compare absorptions. This transient depletion pulses the ionization chamber current, and these pulses are amplified, and integrated by using the rectified pulses to charge a condenser whose voltage can then be measured. The ratio of the capture cross sections for two samples is found from the weights which experimentally give the same voltage response. In practice all samples were compared with gold, whose capture cross section is rather well known.

The voltage pulse from the ionization chamber is primarily caused by the capture of neutrons in the sample. It is found, however, by oscillating a piece of graphite which has negligible absorption, that a signal is generated by the effects of scattering. Comparison of the scattering and absorption signals (Fig. 1) shows that the scattering signal is opposite in sign, has a steeper rise, and reaches its maximum at an earlier time than the capture signal. This can be explained if one assumes that the scattering signal arises chiefly from neutrons that would have passed through the central hole of the ionization chamber without being detected but instead are deflected into the chamber by the sample which blocks the channel. By phasing the rectifying switch so that the signal integration occurs only during the interval indicated between the two vertical lines, the galvanometer deflection because of a scatterer can be made very small. With 5.5 g of carbon, corresponding to 132 mm<sup>2</sup> of scattering area and 0.13 mm<sup>2</sup> of capture area, the adjustment was such that equal capture and scattering areas in the sample gave measured signals in the ratio of about 500 to 1. Under these conditions corrections for scattering need be made

only for samples with capture cross sections of a fraction of a barn.

Although thermal neutrons are known to predominate in the region in which the sample is oscillated, a test was made to determine whether neutron capture by resonances above about 1 ev would give a measurable response. The ratio of nonthermal to thermal neutrons at the position of the ionization chamber was determined by measuring the relative activity of two identical 5-mil-thick indium foils, irradiated with and without a cadmium cover. The ratio of activities was 35, indicating that indium captured 35 thermal neutrons to 1 epi-cadmium neutron, about a 3 percent effect for the 1.44-ev resonance, while a regular pile oscillator measurement of indium gave a capture cross section that agreed to about 2 percent with the values found by activation and by velocity selection methods. For higher energy resonances this effect would be even less. Those elements with resonance capture peaks at less than 0.5 ev are so indicated in Table II, because for them the values of the capture cross section are really averages over the pile neutron energy spectrum. For the other elements, which absorb thermal neutrons as  $1/(E)^{\frac{1}{2}}$ , the dependence of cross section on neutron energy is the same for all, and the particular neutron energy spectrum of the ORNL pile oscillator does not affect the measurement.

The random variations in the readings arise mainly from the variability of the pile neutron flux, and possibly from "noise" in the ionization chamber. The amplifier was checked with a simulated pulse and found to be quite constant. With the average neutron flux at a monitored point in the pile remaining constant within about  $\frac{1}{2}$  percent, and with no unusual experimental activity around the pile, the flux at the experimental point is constant within about 1 percent. Short period fluctuations, which can affect the oscillator measurements, cannot be controlled. Empirically the long period constancy of the pile oscillator was established by remeasuring a sample. Short period variations were noted only by seeing that successive two-minute readings during a ten-minute run showed no large variations. For ordinary samples variations corresponding to no more than 0.1 mm<sup>2</sup> of capture area were permitted. With some weakly absorbing samples it was not possible to introduce a large sample because of volume limitations, and to maintain the same accuracy it was necessary to use longer periods of measurement with the pile held especially constant. In particular, one special sample of 0.07-mm<sup>2</sup> cross section showed only a 30 percent range of variation in several measurements made on different days.

#### Samples

The beryllium boat which carried the samples imposed a size limitation of  $8 \times 8 \times 64$  mm<sup>3</sup>. The samples were made to cover the full length of the boat and nearly the full width, but the thickness varied with the capture

cross section. For nine-tenths of the samples the thickness was less than that corresponding to 3 percent neutron capture, and no sample had a capture thickness greater than 5 percent. For very strong absorbers the sample was diluted to decrease the possibility of lumps of large capture thickness; sodium carbonate was a satisfactory diluent. The effect of self-protection was about as large as the capture thickness, but the standard gold foils had nearly the same capture thickness as the samples, and no error results in the comparison except for the different shapes of the sample and the gold. Because some of the samples required a wrapper, all the samples were wrapped with a 200-mg piece of 1-mil aluminum foil. The foil could be used as a tube to enclose the powdered samples, as a simple wrapper around solid samples, or as a flat piece to cover the comparison gold foils. The few liquid samples and chemically reactive samples were sealed in quartz tubes and then covered with aluminum.

The chemical states of the samples were chosen so that the foreign atoms had low capture cross sections; thus the oxides ( $\sigma_c$  of oxygen < 0.001 barn) or fluorides ( $\sigma_c$ of fluorine = 0.01 barn) as well as the elemental form were usually used. The composition with respect to strongly absorbing impurities had also to be established. If a sample has an assay of 98 percent, it does not follow that the measurement of the cross section is in error by only two percent. The elements reported here possess capture areas varying from 14,000 mm<sup>2</sup>/g to 0.05 mm<sup>2</sup>/g, and for a weak absorber as little as one part per million of strongly absorbing impurity is serious. As an example, about 25  $\mu$ g (micrograms) of boron are detectible with the pile oscillator; thus the boron impurity should be less than 10  $\mu g$  per sample or an accurate analysis for boron must be made if a correction is to be calculated. In the range of sample weights that were used, from 100 mg to 5 g approximately, the boron tolerance is from 100 ppm to 2 ppm, respectively.

Samples of the commoner materials were chosen from commercial reagent grade chemicals and most of them were analyzed spectrographically at this laboratory for metallic impurities. Chlorine is the only important nonmetallic impurity that could not be detected spectrographically, the rare earths are not easily detected but they are not generally expected as impurities, and boron may escape detection in amounts that are significant because of its large capture cross section. Normally dry compounds were chosen to avoid dilution of the sample by adsorbed water and carbon dioxide. They were heated, dried, and stored in a desiccator. At least two samples of different origin and preferably of different composition were used for each element.

The rare earths, including scandium and yttrium, are a special case because of their large capture cross sections and because of the difficulties of purifying them in sufficient amounts. Only for three of them-lanthanum, cerium, and neodymium-were two very pure samples available. For the others, the rare earths

TABLE I. Capture cross sections of gold for neutrons at 2200 m/sec.

Refer- ence	Total	Scatter	Cap- ture	Method				
a	104 b	9 b	95 b	Modulated cyclotron (Los Alamos)				
ь	103.8	10.8	93.0	Mechanical velocity selector (Argonne)				
c	100			Mechanical velocity selector (Argonne)				
d	101			Mechanical velocity selector (Argonne)				
8	103.5	7.6	95.9	Crystal spectrometer (ORNL)				
f		6.5		Comparison with graphite (Argonne)				
g		7.5		Neutron diffraction (ORNL)				

McDaniel, Sutton, Lavatelli, and Anderson, Phys. Rev. 72, 729 (1947).
 A. Wattenberg, AEC Report ANL-4174 (1949) (unpublished).
 T. Brill and H. V. Lichtenberger, Phys. Rev. 72, 585 (1947).
 E. Fermi, quoted by Seren, Friedlander, and Turkel, Phys. Rev. 72, 888 (1970).

(1947).
 • L. W. Cochran, AEC report ORNL-481 (1949) (unpublished).
 • E. Bragdon, AEC Report CP-2305 (1944) (unpublished).
 • C. G. Shull and E. O. Wollan, Phys. Rev. 81, 527 (1951).

produced by column extraction methods at the ORNL (by Darwin Harris) and at Iowa State College, were the only ones deemed satisfactory. Their purities are estimated by a study of the extraction techniques, including the use of radioactive rare earth tracers, and verified to the limit of the spectrographic method.

Zirconium and hafnium are similar to the rare earths for difficulty of separation. Because of its large capture cross section relative to zirconium, hafnium is significant if it is present to more than 0.1 percent in the zirconium. This required the preparation of special samples and the development of new analytical methods. Zirconium metal can be made purer than its compounds with respect to impurities other than hafnium, and the final value of the capture cross section in this report is from measurements on metal samples. The measurement of hafnium offered no difficulties.

The ORNL pile oscillator has been used to make measurements of the thermal capture cross sections of 69 of the first 83 elements. Two elements-Tc and Pm exist only in small quantities that are neither stable nor of any standard isotopic mixture. Five elements are the inert gases-He, Ne, A, Kr, and Xe-whose low density precludes placing sufficient weight in the limited volume of the pile oscillator. Five elements-Be, C, O, F, and Bi-have capture cross sections less than 0.02 barn but scattering cross sections several hundred times larger; any correction for the scattering of neutrons with so large a ratio of cross sections would be an uncertain one. One element-H-not only has a large ratio of scattering to capture but produces slow neutrons by elastic collisions with fast ones, thus counteracting in part its capture of slow neutrons. Gold, the standard for these measurements, was measured by others using direct methods.

## Standards

Because the capture cross sections in Table II were determined from ratios with the capture cross section for gold, they are no more accurate than the value used for gold. If 95 b per atom for gold for neutrons at 2200

	<b>ORNL</b> pile oscillator				Comparison data			
Substances examined	Z	Element	Final value	Estimated error (%)	Pile oscillators Argonne <sup>a</sup> Harwell <sup>b</sup>		Trans- mission	Activ- ation®
Li <sub>2</sub> CO <sub>3</sub> ; LiIO <sub>3</sub>	3	Li	67	5		65.5	72.8 <sup>d</sup>	
BF <sub>8</sub> ; B <sub>2</sub> O <sub>8</sub> ; Borax	5	В	730	5	(710)	(710)	05° 745ª 704 <sup>f</sup> 722≊ 703°	
NaCN; KNO3; NaNO3 Na; Na2CO3; NaF	7 11 12	N Na Ma	1.86 0.47	5 5 10	0.52	1.76 0.50 0.057	2.15 <sup>d</sup>	0.63
Al SiO <sub>2</sub>	12 13 14	Al Si	0.22 0.10	5 20	0.22 0.16	0.037 0.212 0.16	0.23 <sup>d</sup>	0.21
P red; P <sub>2</sub> O <sub>5</sub> S KCl; NaCl; PbCl <sub>2</sub>	15 16 17	P S Cl	0.15 0.47 32.7	10 10 5	0.51 31.5	0.193 0.49 31.8	0.62 <sup>d</sup>	0.23
K; KF; K2CO3 CaF2; CaO; CaCO3	19 20	K Ca	2.05 0.41	5 5	2.11 0.43	1.89 0.40		
$Sc_2O_3$ Ti: TiO <sub>2</sub>	21 22	Sc Ti	11.8 5.8	5 5	31.8 5.88	5.0		22
$V_2O_5$ ; VC	23	V	4.7	5	4.93	4.4	3 4 7 d	4.50
Mn; Mn <sub>3</sub> O <sub>4</sub> ; Mn oxide	24	Mn	12.8	5	12.3	12.8	14.2 <sup>d</sup>	10.7
Fe; Fe <sub>2</sub> O <sub>3</sub> Co; Co <sub>3</sub> O <sub>4</sub>	26 27	Fe Co	2.39 34.2	5 5	2.48 35.7	2.4 38.2	2.21 <sup>d</sup> 40.5 <sup>d</sup> 39.0 <sup>h</sup>	22.4
Ni; NiO Cu	28 29	Ni Cu	4.5 3.57	5 5	4.37 3.71	4.8 3.6	4.87ª 3.42	2.56
$Zn; ZnO Ga_2O_3$	30 31 32	Zn Ga	1.00 2.77	5 5 10	1.09 3.10 2.64	1.09	2.21 <sup>d</sup>	2.16
As; $As_2O_3$	33	As	4.14	5	5.46	4.9		4.2
Se; SrSe Br; KBrO <sub>3</sub> Rb <sub>2</sub> CO <sub>3</sub>	34 35 37	Se Br Rb	12.1 6.4 0.70	5 5 10	12.2 8.83	11.4 8.7	6.40 <sup>d</sup>	6.6 0.55
SrO; SrCO <sub>3</sub>	38 30	Sr V	1.16	5 10	1.53	1.35		1 24
$2^{\circ}$ Zr; ZrO <sub>2</sub> Nb; Nb <sub>2</sub> O <sub>5</sub> Mo: MoO <sub>2</sub>	40 41 42	Źr Nb Mo	0.18 1.06 2.40	10 5 5	$\sim 0.4$ 1.51 3.04	0.250 1.26 2.95	0.6 <sup>d</sup>	~1
Ru; RuO <sub>2</sub> Rh	44 45	Ru Rh	2.46 150	5 5	6.30 172	-,,,,	2.47 <sup>d</sup> 141 <sup>f</sup> 150°	149
Pd Ag	46 47	Pd Ag	6.6 60	5 5	10.3 90	10.0	57.2 <sup>d</sup> 59.2 <sup>e</sup>	70.7
Cd In	48 49	Cd In	3500 191	res <sup>i</sup> 5			2300 <sup>d</sup> 191 <sup>f</sup> 189°	190
Sn; SnO <sub>2</sub> Sb; Sb <sub>2</sub> O <sub>3</sub>	50 51	Sn Sb	0.58 5.3	5 5	0.72 8.15	0.70 7.6	4.05 <sup>d</sup>	4.9
I; NaI; KI	52 53	Ie I	4.5	5 5	5.82 9.23	7.4	7.1 <sup>d</sup>	6.25
$Cs_2CO_3$ ; $CsBr$	55 56	Cs Ba	29.0 1.05	5	35.8	1 25	0.71	25.6
$\begin{array}{c} \text{BacO}_3, \text{ BaF}_2\\ \text{La; La}_2\text{O}_3\\ \text{CaO}_2\end{array}$	57 58	La Ce	8.8 0.80	5 10	9.01	< 0.92		8.4
$Pr_6O_{11}$ Nd <sub>2</sub> O <sub>3</sub>	59 60	Pr Nd	11.2 44	5 5	52	<b>\0.72</b>		10.1
Sm <sub>2</sub> O <sub>3</sub> Eu oxalate	62 63	Sm Eu	10600 4200 35000	res res	~8200	27600		
$\operatorname{Tb}_{4}O_{7}$	65 66	Tb Dv	33000 44 890	10 res	30300	37000		
$Ho_2O_3$ Er <sub>2</sub> O <sub>2</sub>	67 68	Ho Er	64 166	5				59.6
$Tm_2O_3$ Yb <sub>2</sub> O <sub>3</sub>	69 70	Tm Yb	118 36	5 10				106 ≥34
Lu <sub>2</sub> O <sub>3</sub> HfO <sub>2</sub>	71 72	Lu Hf	108 102	5 5	171	110		106.9
$\begin{array}{c} Ta; Ta_2O_5\\ W; WO_3 \end{array}$	73 74	Ta W	21.3 17.7	5 5	36 24	21.2 20.5	19.0 <sup>d</sup> 17.2 <sup>d</sup>	20.6

TABLE II. Thermal neutron capture cross sections (in barns).

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	ORNL pile oscillator				Comparison data			
Substances examined	Z	Element	Final value	Estimated error (%)	Pile os Argonneª	cillators Harwell <sup>b</sup>	Trans- mission	Activ- ation®
Re; KReO4	75	Re	84	5	120		100 <sup>d</sup>	85.0
Os; OsO4	76	Os	14.7	5			17 <sup>d</sup>	
Ir	77	Ir	440	5	$\sim 470$		405ª	467
Pt	78	Pt	8.1	5	13.5	15.0	6.5 <sup>d</sup>	
Au	79	Au	95	ī į	$\sim 157$			
HgO: HgI	80	Hg	341	res	$\sim 380$		405 <sup>d</sup>	
Tl	81	Τĩ	3.27	5	3.65		3.8d	2.28
Pb	82	Pb	0.19	5	0.16	0.28	0.18 <sup>d</sup>	

TABLE II.—Continued.

<sup>a</sup> Argonne (reference 4).
 <sup>b</sup> Harwell (reference 5).
 <sup>c</sup> Seren, Friedlander, and Turkel, Phys. Rev. 72, 888 (1947).
 <sup>d</sup> Columbia University modulated cyclotron, variously reported by Dunning, Rainwater, Havens, Wu, and Melkonian: Phys. Rev. 70, 136, 154 (1946); 71, 05, 105, 174 (1947); 72, 634 (1947); 73, 733, 963 (1948); 75, 1296 (1949); 76, 1750 (1949).

(1949); 70, 1750 (1949).
 <sup>e</sup> Argonne mechanical velocity selector: Fermi, Marshall, and Marshall, Phys. Rev. 72, 193 (1947): Fermi quoted in Can. J. Research A25, 26 (1947); Fermi quoted in Phys. Rev. 72, 888 (1947).

m/sec is not correct, the list would have to be modified accordingly. The value of 95 barn is an estimated best value based on the cross sections determined by several investigators and listed in Table I. Some recent work done elsewhere indicates that 93 barn is a better value, but the calculated effect of the strong capture resonance at 4.9 ev with the near thermal neutron spectrum used with the pile oscillator is enough to make the "effective" value for gold 94 barn or 95 barn, and thus the values in Table II need not be altered. The pile oscillator work at other laboratories used 710 barn per atom for boron as the standard for comparison; here the preparation of boron samples of about 1 mg that showed no selfprotection and were accurately weighed was discarded in favor of the gold foil standard samples.

## Accuracy

The thermal neutron capture cross sections reported here have an estimated error of about 5 percent with some exceptions noted in Table II. Measurements on a single sample were reproducible within 2 percent, except for some weak absorbers where the largest usable sample neutron capture areas, for example-magnesium, silicon, and phosphorus, were less than the 2 mm<sup>2</sup> required for good precision. Consideration of response of the pile oscillator to processes other than the capture of thermal neutrons, the accuracy of the value for the gold standards, and, perhaps most important, whether the sample was well identified with respect to all its constituents, together with the reproducibility of results led to the estimate of accuracy.

The pile oscillator measurements at other laboratories involve a change of pile reactivity and thus include some resonance absorption along with the predominant <sup>1</sup> Cornell University modulated cyclotron: Bacher, Baker, and McDaniel, Phys. Rev. 69, 443 (1946); B. D. McDaniel, Phys. Rev. 70, 832 (1946); W. B. Jones, Jr., Phys. Rev. 72, 362 (1947); R. R. Meijer, Phys. Rev. 75, 773 (1949).

<sup>173</sup> (1949).
<sup>6</sup> Los Alamos modulated cyclotron: Sutton, McDaniel, Anderson, and Lavatelli, Phys. Rev. 71, 272 (1947).
<sup>b</sup> Oak Ridge crystal spectrometer: Bernstein, Dial, Stanford, and Stephenson, Phys. Rev. 75, 1302 (1949).
<sup>1</sup> Res indicates a neutron capture resonance at less than 0.5 ev.
<sup>1</sup> The value of 95 barn for gold (for incident neutrons of 2200-m/sec velocity) was the standard for this work.

thermal absorption. For some of their values the Argonne group indicates it made a correction for the resonance capture. The values of both other laboratories, when compared with the ORNL results, should be equal if there was no resonance contribution or the correction was made, greater if there was an uncorrected resonance contribution. The Argonne group indicates an accuracy of better than 5 percent in its values; the Harwell group has published only tentative values.

For further comparison, the capture values determined by activation methods and by transmission methods using monoenergetic neutrons are included in Table II. Seren et al. state an estimated probable error for their activation cross sections of 20 percent, although their work seems in general to be better than that. Theirs is the largest single report of such cross sections; other activation measurements are to be found in the Bureau of Standards Circular, "Nuclear data."<sup>3</sup> The curves relating neutron transmission with energy have been compiled but not the values of the breakdowns into scattering and capture. The accuracy of the capture values from such work varies, but all such values for 1/v absorbers are included in Table II. Other methods of measuring neutron capture such as diffusion, comparison of absorption in a standardized apparatus, and danger coefficients, are in no single published compilation of cross-section values. They are not tabulated here for comparison because the accuracies are generally either unknown or low, and for many of them there is no knowledge of the average neutron energy used in the measurements.

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