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Neutron Absorption Cross Section of Uranium-236

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Engineering Physics and Mathematics

Neutron Absorption Cross Section of Uranium-236

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ABSTRACT

U-236 neutron absorption was measured as a function of neutron time-of-flight from 20 eV to 1 MeV. The neutron flux was monitored with a ^{6}Li glass scintillator. Average cross sections from 3 keV to 1 MeV were derived. Estimated uncertainties were less than 5% below 600 keV and increased to 9.5% at 1 MeV. Resonance parametrization from 20 eV to a few keV remains to be done.

1. INTRODUCTION

U-236 has a half-life of 23 My, so any incorporated in the material of the solar system 4.7 billion years ago has decayed to undetectable levels, breaking up into natural ^{232}Th and ^{4}He. It is formed in considerable quantities as a compound nucleus at >5 MeV excitation in nuclear reactors from neutron absorption by ^{235}U. Most of it breaks up promptly into fission products and neutrons. As the residual ^{236}U builds up in reactor fuel it absorbs more neutrons with very little subsequent fission. Thus it becomes a sort of poison which must be allowed for in reactor designs, particularly in designs where the fuel has a long life or is recycled. The neutron absorption properties of ^{236}U as a function of neutron energy are similar to those of the naturally occurring ^{238}U, with s-wave resonances spaced about 15 eV apart. Above a few keV, absorption by p-wave resonances becomes significant and above 100 keV or so d-wave contributions must also be considered.

Most American and British measurements date from the 1960s. Since 1982 several Russian measurements have been reported.[1–3] A recent review[4] points up the incompatibility between the old and new measurements of average neutron absorption and capture, particularly for neutron energies above a few keV. Davletshin *et al.*[3] specifically call for new measurements giving a broad overlap with the energy ranges covered in the diverse earlier experiments. A comparison of measurements and evaluations has recently been compiled at Brookhaven National Laboratory.[5] The world request list WRENDA 87/88[6] shows two requests for ^{236}U neutron capture cross sections in energy ranges 1 keV to 1 MeV and 500 eV to 1.4 MeV, respectively.

The present measurements cover a neutron energy range 20 eV to 1 MeV, but only the average absorption cross section from 3 keV to 1 MeV is covered in the present report. At the lower energies it is planned to fit resonance parameters to the data.

2. U-236 SAMPLE

The ^{236}U used in the present experiments originated from electromagnetic isotope separations in the Calutron facility at the Oak Ridge National Laboratory (ORNL) in mid-1967.[7] About 12 g of product was designated as Batch 201C. Part of the batch was sent to Los Alamos National Laboratory (LASL) where it was converted to metal and rolled into foils for use in superconductivity experiments.[8] A delay in these experiments allowed Drs. J. Smith and M. Tinkle of LASL to loan the ^{236}U foils to ORNL in 1988 for neutron absorption measurements at the Oak Ridge Electron Linear Accelerator (ORELA) pulsed neutron source.

Two of the foils were selected for use, weighing 0.7009 g and 1.5517 g, respectively. The uneven blackish-blue surface coatings hinted at extensive oxidation during storage. The two foils were mounted in the neutron beam one above the other in a plane perpendicular to the beam. After the neutron cross-section measurements were completed, microscopic examination of a small slice showed about 80% of the thickness retained a metallic luster. The average density of a foil was determined by a classical immersion technique to be 17.1 g/ml. Assuming either UO_2 or U_3O_8 and metal gave weight fractions of uranium averaging 0.985 ± 0.002, in close agreement with the microscopic observation. The thickness of the foils varied appreciably but averaged about 0.28 mm. The rather irregular outlines of the pieces were traced to determine the total area. Two independent attempts gave 450 mm^2 and $\sim$500 mm^2. The average volume and thickness gave an area of 470 mm^2.

In addition to mass spectrometric analyses a spark source optical spectrum was searched for elemental impurities, and a germanium gamma-ray spectrum was searched for radioactive contaminants. Results are shown in Table 1. As ^{228}Th was indicated in the last of these tests, the neutron absorption yield at 22 eV and 23.5 eV was searched to place an upper limit of 0.5% on possible ^{232}Th contamination. Most of the ^{228}Th activity is attributed to decay of ^{232}U.

Table 1. Sample analyses

Mass spectrograph				Optical spectrograph		Gamma spectroscopy	
Mass	Atom %[a]	Atom %[a]	Uncertainty	Element[b]	$\mu g/g$		
232	c	c		Al	30	^{137}Cs	trace
233	0.011	0.011	0.001	Ca	50	^{228}Th	0.12 μ Ci[d]
234	0.0008	0.0008	0.0001	Cl	30	^{235}U	~0.15%
235	0.201	0.204	0.002	Cr	20	^{241}Am	~1 ppm
236	99.726	99.714	0.003	Cu	20		
238	0.061	0.070	0.001	Fe	200		
				K	10		
				Mg	30		
				Mn	5		
				Na	20		
				Ni	10		
				P	1		
				V	1		
				Zn	5		
				Zr	50		
					482		

[a] Different pieces of the measured sample.

[b] Other elements were not detected or upper limits were set at <3 $\mu g/g$.

[c] Less than 2 ppm and not detected.

[d] Corresponds to about 5 ppb ^{232}U left in the sample at present.

3. EXPERIMENTAL PROCEDURES

Neutron absorption was measured at the ORELA pulsed neutron time-of-flight facility. Prompt gamma rays were detected by a pair of fluorocarbon scintillators. The apparatus and procedure were used previously for measurements on tungsten[9] and tantalum.[10] Pulse height weighting was used to measure total gamma energy release. Dividing this by the excitation energy available in the compound nucleus per captured neutron gives the number of neutrons captured. Dividing by the number of ^{236}U atoms in the sample and the incident neutron flux leads to an effective neutron capture cross section. As found earlier for ^{241}Am by Weston and Todd,[11] this prompt gamma-ray detection method has essentially the same efficiency for capture and fission events, so the total response corresponds to the sum of the two processes i.e., the absorption cross section.

The metal sample was mounted between the detectors, facing the neutron source and at an effective flight path of 40121 mm. Data taking was accomplished in 208 hours of ORELA beam time. The ORELA operating conditions were 800 seven nanosecond pulses per second and a beam power of 11-kW. Cross-section calibration was done by the saturated 4.9-eV gold resonance method as usual.[12] Corrections applied include electronic dead-time loss and amplifier gain standardization, average background from long-lived radioactivity, average scattered neutron backgrounds, gamma energy loss in the sample, average resonance self-protection and neutron scattering before capture in the sample.

4. EXPERIMENTAL RESULTS

The trace of ^{235}U (0.2%) in the sample contributed between 0.8% and 1.8% of the absorption yield at various energies. The ^{235}U absorption component was parametrized to 10% based on ENDF/B-V fission and capture cross sections and subtracted from the sample yield to get the ^{236}U cross section. Figure 1 shows the overall result in broad histogram averages. The data are listed in Table 2. Figure 2 shows finer steps from 100 keV to 200 keV together with the statistical uncertainty of counting at each step. The sharp drop in one bin near 160 keV is unexpected but is not due to a single data channel as seen in Fig. 3 where the sample yield at the full 1-ns resolution of the measurement from 155 keV to 165 keV is displayed. The neutron energy range from 3 keV to 100 keV in 1/4 keV steps is shown in Fig. 4 as an indication of the extent of fluctuations about an average cross section. Finally, Fig. 5 shows the sample yield at the 8-ns time-of-flight bin width used below 7.92 keV. The regions between peaks encompass the average yield of the ^{235}U contaminant, shown as a straight line, indicating that the background subtraction procedure has been successful. The average cross sections and statistical standard deviations have been submitted to the Nuclear Data Center at Brookhaven National Laboratory for distribution.

Table 2. Average ^{236}U neutron absorption

Energy interval keV	Cross section mb	Uncertainty[a] ±mb
3 – 4	1336	52
4 – 6	1122	44
6 – 8	1025	40
8 – 10	874	35
10 – 15	724	32
15 – 20	647	28
20 – 30	549	24
30 – 40	491	21
40 – 60	418	19
60 – 80	317	14
80 – 100	264	12
100 – 150	219	10
150 – 200	195	9
200 – 300	178	9
300 – 400	160	8
400 – 500	162	8
500 – 600	168	8
600 – 700	188	9
700 – 800	238	13
800 – 900	316	19
900 – 1000	420	40

[a]Statistical uncertainties are less than 1% standard deviation in the intervals shown. The systematic uncertainties shown here are derived by summing in quadrature the components listed in Table 3.

Table 3. Uncertainty estimates

Source	Effect (± %)
Average constant and time dependent backgrounds	~3.
Oxide effects	0.2
Sample area (gamma-ray attenuation)	0.1
Saturated resonance calibration	2.4
Flux monitor efficiency	<3.4[a]
Neutron scattering and absorption by the sample	0.3
Efficiency for fission gamma rays (±10%)	0.2[b]
^{235}U in sample	<0.2
Uncompensated instrumental drifts	0.4
Combined quadrature at 100 keV	4.6
Combined quadrature at 600 keV	5.0
Combined quadrature at 850 keV	6.0
Combined quadrature at 1000 keV	9.5

[a] Increases from 0.1 at 3 keV to 3.4 at 1 MeV (Ref. 10).

[b] For neutron energies <500 keV. ~3% at 850 keV, ~8% at 1 MeV.

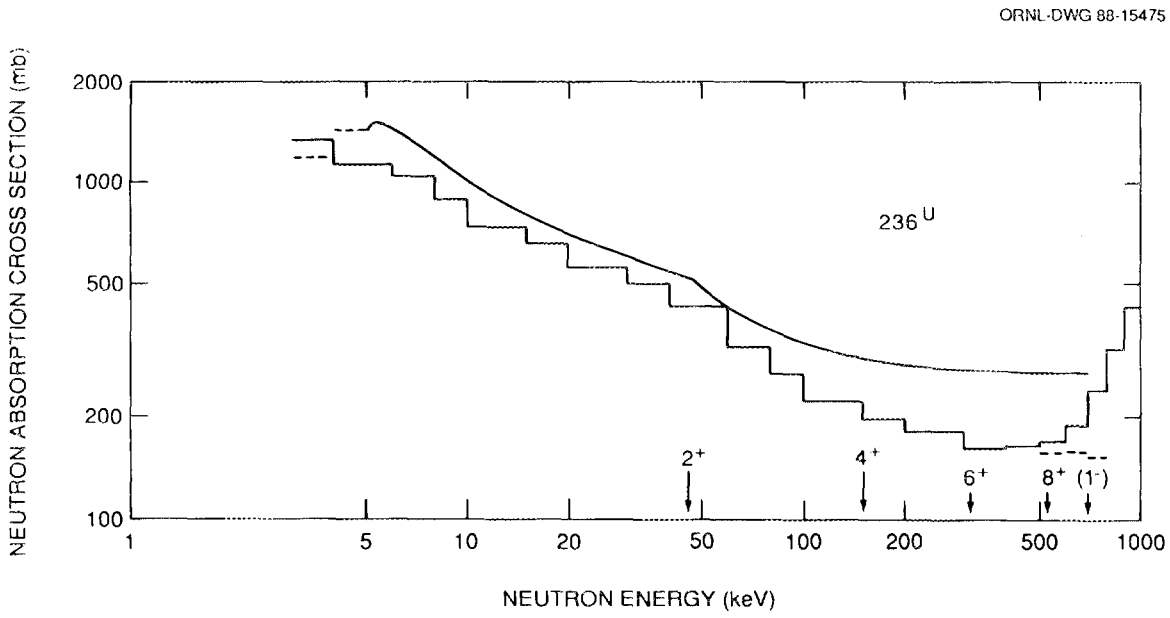

Fig. 1. Average cross sections from 3 keV to 1 MeV. The solid histogram approximates the present data whose statistical standard deviations are less than 1% for the energy intervals shown here. The dashed histograms from 3 keV to 5 keV and the smooth curve approximate the ENDF/B-V evaluation. The dashed histogram from 500 keV to 700 keV is derived by subtracting the ENDF/B-V fission component from the measured absorption cross section. Thresholds for inelastic neutron scattering are indicated by arrows and competition above the lowest level is apparent in the data.

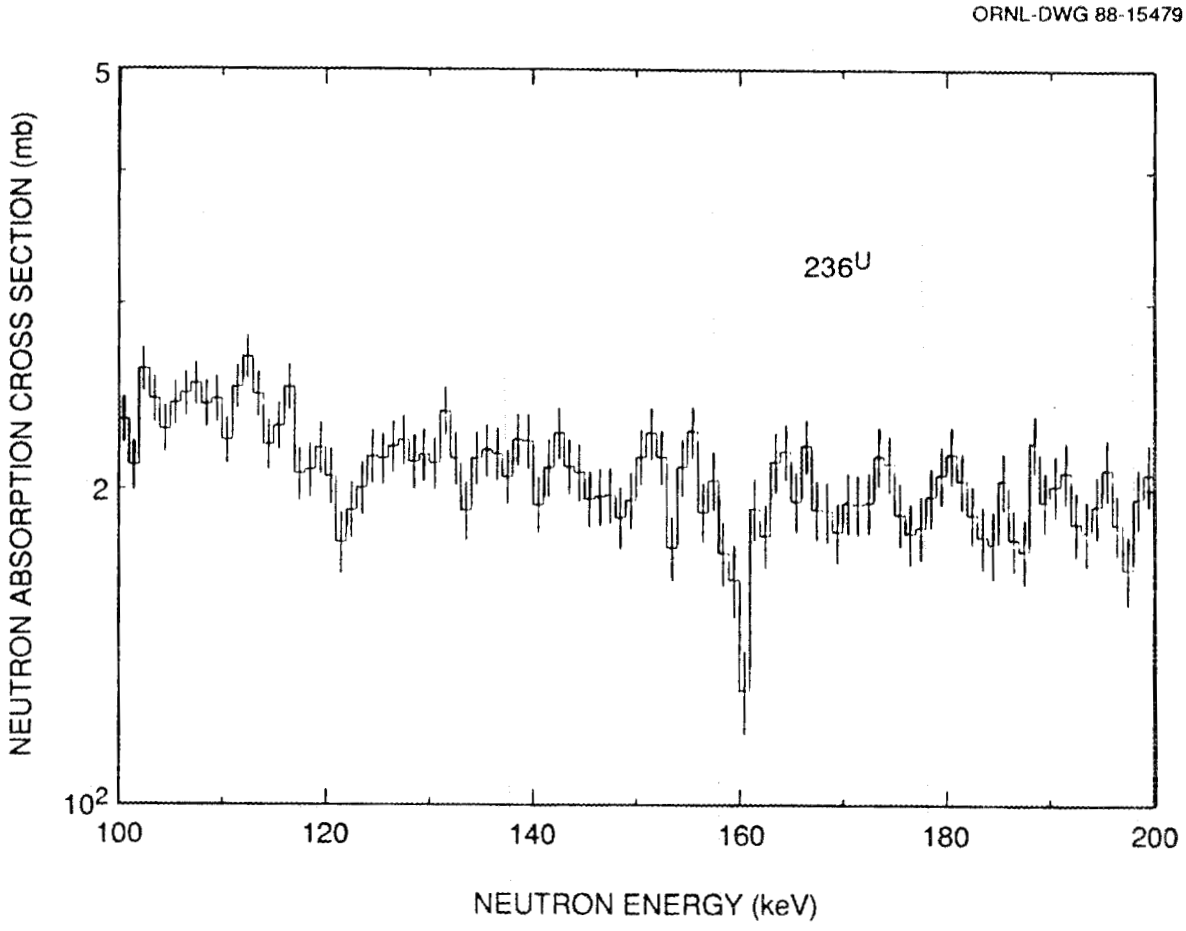

Fig. 2. Average cross sections from 100 keV to 200 keV. The error bars indicate the statistical standard deviations of counting in each 1 keV bin.

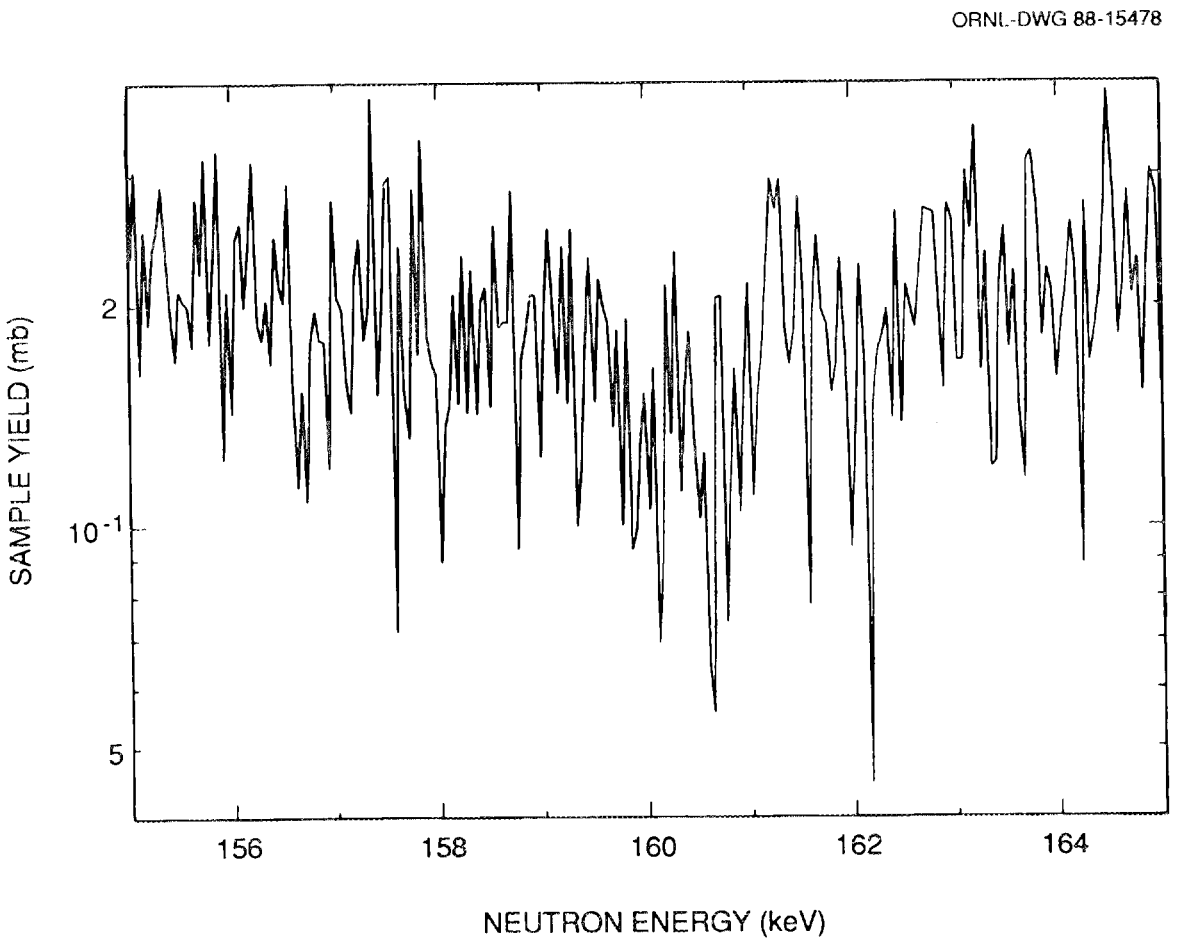

Fig. 3. Sample yield from 155 keV to 165 keV. The data are shown at the 1 ns time-of-flight intervals at which they were recorded, without averaging. The full width at half maximum of the experimental resolution function at 160 keV was about 1/2 keV.

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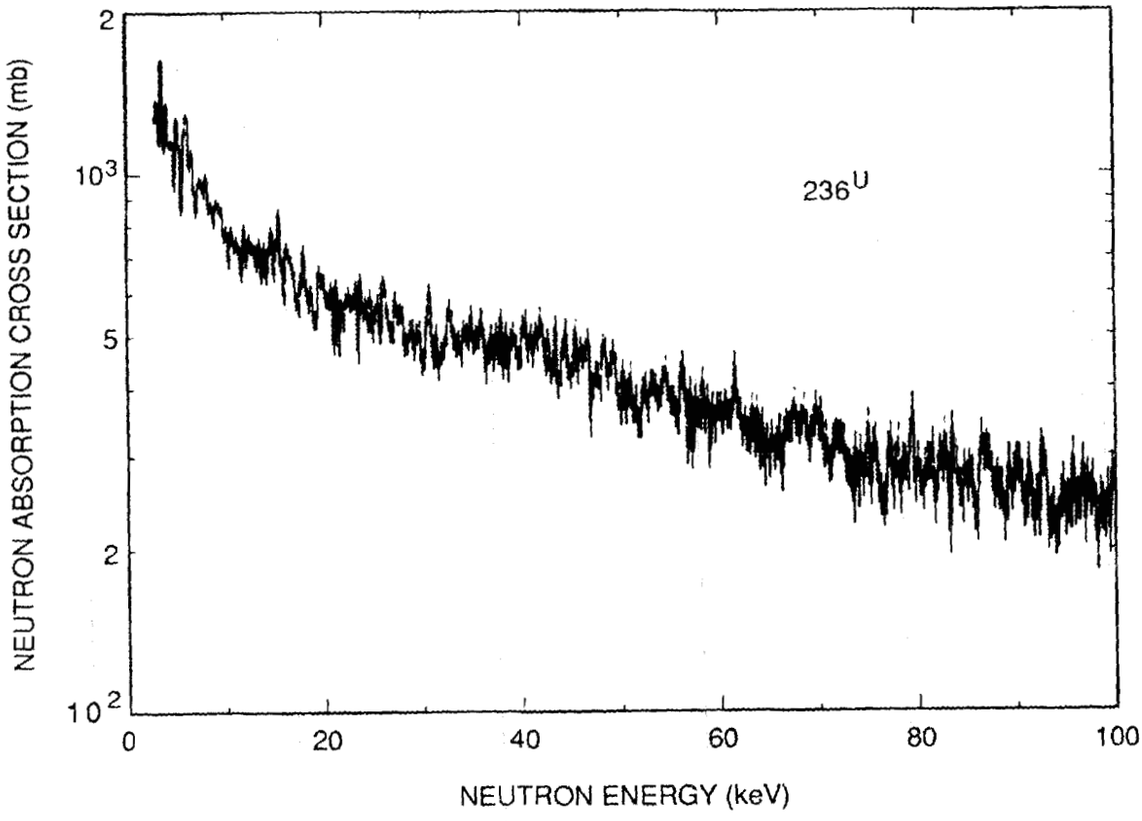

Fig. 4. Average cross sections from 3 keV to 100 keV. The error bars indicate the statistical standard deviations of counting in each 1/4 keV bin. Each bin, on average, should contain 17 s-wave resonances and many more small p-wave and d-wave contributions.

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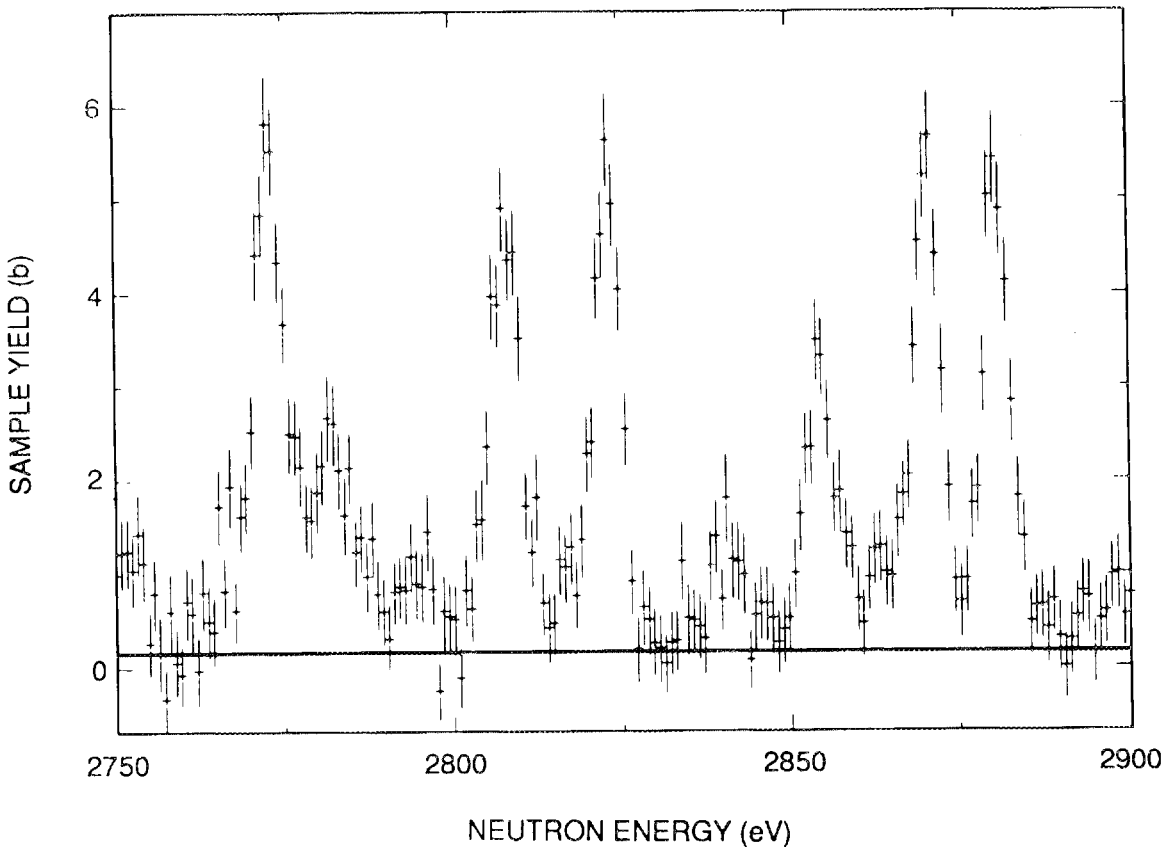

Fig. 5. Sample yield from 2.75 keV to 2.9 keV. The data are shown at the 8 ns time-of-flight intervals at which they were recorded, without averaging. The full width at half maximum of the experimental resolution function at 2.9 keV was about 4 eV. The solid line represents the average contribution from the 0.2% ^{235}U in the sample.

5. UNCERTAINTIES

The purity of the sample was remarkably good, considering that it had to be retrieved from reactor fuel containing a wealth of radioactive fission products. The detectable radioactivity remaining came from decay of ^{241}Am and ^{228}Th, the latter attributable to ^{232}U decay. This sample radioactivity was over five times higher than the background experienced with non-radioactive samples but well within the counting rate capabilities of the measuring apparatus. It was sampled a millisecond after each accelerator burst and averaged 416 counts per second.

The sample appeared black, indicating extensive oxidation which could add up to 15% to the weight. There was also uncertainty as to the oxide compound present. Fortunately the oxide densities correlate inversely with the oxide content, so the final uncertainty in the metal content was only $\pm 0.2\%$. The oxygen also has a direct effect in scattering neutrons, especially at the 434-keV resonance, and a small background correction was made for its effect. The very irregular sample area was $\sim$470 mm^2 but has little effect on the average cross section.

The saturated resonance intercalibration technique has been used with several suitable isotopes in the past, showing $\pm 1\%$ agreement for holmium, gold and ^{238}U.[13] Recently Bergman *et al.*,[1] have observed thermal neutron capture gamma-ray spectra using a barium fluoride scintillator. Comparing a number of elements to ^{236}U they found its spectrum most similar to that of gold. While calibration at the 5.45-eV resonance of ^{236}U might be preferable to gold, the present samples were not adequate for that purpose.

The extension of the neutron flux shape from the 4.9 eV intercalibration energy to 1 MeV[14] involves both the ^{6}Li (n, α) cross section up to 70 keV and the ^{235}U fission cross section above 3 keV. Uncertainty components involved in using the ENDF/B-V files and an experimental intercomparison of the lithium glass scintillator with a ^{235}U fission chamber have been discussed and tabulated previously.[10]

The process of inelastic neutron scattering followed by capture at lower energy has been a problem in some measurements. For a thin sample it can be corrected for with little resultant uncertainty. This correction, for a sample twice as thick as our ^{236}U, has been discussed earlier.[15] The uncertainty introduced by the smaller corrections for neutron scattering and self-protection in the present sample is estimated to be 0.3%.

The assumption of equal detection efficiency for capture and fission gamma rays[11] seems reasonable, but an uncertainty of $\pm 10\%$ for it also seems reasonable. Over most of our energy range the fission to capture ratio is quite small, averaging $\sim$1.6% in the resolved resonance range. Above 500 keV the "fission barrier" is approached and the fission cross section exceeds the capture cross section above $\sim$800 keV. The resultant uncertainty in our absorption cross section is $\sim$3% at 850 keV and $\sim$8% at 1 MeV. We have indicated a separation of the capture component from 500 keV to 800 keV as shown by dotted lines on Fig. 1.

Instrumental uncertainties, primarily gain drift of the electronics and phototubes have been tabulated previously.[16] The gain drifts observed before, between and after our experimental runs using long-lived radioactive sources totaled 1.3%. It was corrected for by linear interpolation and the resultant uncertainty in the cross-section results is estimated as ±0.2%.

6. DISCUSSION

The present results as seen in Fig. 1 average 28% below the ENDF/B-V evaluation[17] in the energy range 4 keV to 800 keV. The evaluation was based on data and calculations up to 1967. Competition with inelastic neutron scattering to the 2+ level can be seen above the threshold at 45 keV. Similar competition near 150 keV is not expected to be significant. The dashed curve from 500 keV to 800 keV results from subtracting the ENDF/B-V fission cross section from our absorption cross section to indicate the course of the capture cross section. Recent Russian data for ^{236}U capture, activation, and absorption[1-3] do not appear to conflict with the present results. Fluctuations in the average absorption are shown in Figs. 2 and 4. The low value at 160 keV is not expected but is not due to a single data channel as seen in Fig. 3. Individual resonance peak structure is shown in Fig. 5 to indicate the lack of residual background in the sample yield data file.

7. CONCLUSIONS

The present results for the average ^{236}U neutron absorption cross section cover a broad energy range, 3 keV to 1 MeV, overlapping the range of earlier measurements. The values found are distinctly lower than those in the ENDF/B-V evaluation, in agreement with more recent experiments. This work should partially satisfy two official requests[6] for ^{236}U neutron capture cross-section measurements.

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