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A SEARCH FOR POSSIBLE STRUCTURE IN THE ^{238}U(n,f) CROSS SECTION NEAR 2.3 MeV

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Abstract—The shape of the ^{238}U(n,f) cross section was measured relative to the ^{237}Np(n,f) cross section over the neutron energy region 1.9–2.6 MeV to search for possible gross structure in the ^{238}U cross section. Forty-two measurements were made in this energy interval with ~ 24 keV energy resolution and a statistical accuracy of $< 1\%$ using the ^{7}Li(n,p)^{7}Be reaction as the neutron source. No structure was observed. Absolute measurements were made at 2.150 and 2.453 MeV to normalize the shape data.

INTRODUCTION

The neutron flux measurements necessary for absolute cross section determinations are difficult to perform, so the trend has been to make very accurate cross section measurements of a few reactions and then use these as standards for other measurements. The ^{238}U(n,f) reaction is prominent among those reactions considered for this purpose, as it has a number of desirable characteristics. It is, for example, a fission reaction with a large cross section which allows the use of a simple, efficient and stable detector. The material is also readily available and samples are easy to prepare. However, the cross section of a standard should vary smoothly with energy in the region of interest as otherwise the effective cross section may depend on the energy resolution of the measurement. There is still some question concerning the ^{238}U fission cross section in this respect. Several years ago, some high-resolution measurements of the ^{238}U(n,f) reaction were reported by Blons *et al.* (1976) which showed significant structure from threshold to 3 MeV neutron energy. Much of this structure was small in amplitude and had peak separation of the order of 50 keV, but a more prominent feature was observed at about 2.3 MeV where the cross section changed by over 10%. The size of this "hole" is shown in Fig. 1. Cierjacks (1977) observed that the presence of such a structure made the usefulness of ^{238}U as a standard doubtful to some extent since the requirement that a standard should possess a smooth excitation function would not be fulfilled. Much of this structure, particularly that above 2 MeV, was not apparent in the other data available at that time due to poor energy resolution and widely-spaced data points. Later measurements by Difilippo *et al.* (1980) showed some structure between threshold and 3 MeV but, even after taking the difference in energy resolution into account, there was little indication of a "hole" at 2.3 MeV.

Recently, Poenitz (1987), in the context of a review of the ^{238}U data, observed that the structure in the ^{238}U fission cross section was not confirmed and the shape was not well established. Furthermore, while the resolution of the fine structure would require very good resolution, the "hole" near 2.3 MeV could be observed with much more modest resolutions. In view of the importance of the ^{238}U fission cross section, he recommended that a set of measurements be undertaken to confirm or deny that particular feature.

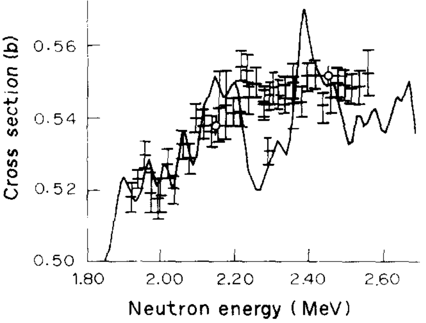

Fig. 1. The ^{238}U fission cross section derived from the ratio measurements: ○, based on the absolute ratio measurements; +, based on the shape measurements normalized to the absolute measurements; –, the data of Blons *et al.* (1976) smoothed with the resolution function of Fig. 2.

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This note describes a series of measurements of the ^{238}U fission cross section relative to ^{237}Np with an energy resolution of ~24 keV for the purpose of determining the shape of ^{238}U(n, f) excitation function in the neutron energy region of 1.9–2.6 MeV. The measurement was specifically made to see if there was any structure in this energy region that would have an impact on the use of ^{238}U(n, f) as a standard reaction in measurements of modest resolution. Measurements of the absolute fission cross section ratio were made at two energies, but with much broader energy resolution, in order to normalize the shape data. Since both the ^{237}Np and ^{238}U fission cross sections are possible secondary standards, such an intercomparison is fundamentally useful.

EXPERIMENTAL METHOD

Most aspects of the experimental system have been described elsewhere, particularly in Meadows (1983b, 1988). Briefly, the relative fission rates of pairs of ^{238}U and ^{237}Np samples were measured by placing them back to back in a double ionization chamber located on and perpendicular to the axis of the neutron source. The separation of the two samples was ~ 0.25 mm, while the neutron source–sample distance was 6.1 cm. The shape of the cross section ratio was measured between 1.9 and 2.56 MeV neutron energy using samples $^{238}_{60}$U and $^{237}_{79}$Np, with the ^{238}U sample in the 0° position and the ^{237}Np sample facing the neutron source. Measurements were made over the entire region in steps of ~20 keV; then additional measurements were made at random energies between 2.2 and 2.36 MeV. All shape measurements were made with a continuous beam of 10–12 μA and an energy resolution of 18–30 keV.

The ^{237}Np fission cross section was chosen as a reference rather than the more conventional ^{236}U because ^{237}Np(n, f) is a threshold reaction. Thus, a large correction for fissions induced by low energy room-return neutrons was not needed. This removed the necessity of using a pulsed neutron source in order to eliminate the room return by time correlation. In addition, the use of a continuous beam resulted in higher average beam currents, and consequently required much less accelerator time. This assumes, of course, that there is no structure in the ^{237}Np(n,f) cross section that could interfere with the measurement. The survey of the ^{237}Np data by Cierjacks (1977), and the partial survey of more recent data by Meadows (1983a), show no evidence of structure between 2 and 3 MeV; however, it may have been obscured by broad energy resolution and widely-spaced data points. Certainly, the high-resolution measurements of Plattard *et al.* (1976) show no structure below between ~0.2 and 2 MeV. If there is structure in the ^{238}U and ^{237}Np fission cross sections, it is unlikely that they will match in energy and amplitude, so an absence of structure in the cross section ratio will still be evidence for an absence of structure in either cross section.

The normalization measurements were made at two energies with ~ 100 keV resolution, using three pairs of samples at each energy. A pulsed and bunched charged particle beam was used to obtain a pulsed neutron source, and fast timing techniques selected those fissions suitably correlated with the neutron burst. This eliminated most of these few fissions that were caused by prompt neutron scattering from the floor, walls and air. The differences in sample geometry, neutron transmission, and the effect of momentum transfer on the detector efficiency were eliminated to first-order by making measurements with both sample orientations and then averaging the two measurements.

The individual fission fragment spectra were corrected for losses in the sample, including momentum transfer and fragment angular distribution effects, and extrapolated to zero channel to correct for those fissions lost below the detector bias level. Corrections were made to the fission cross section ratio for the minor isotopes in the ^{238}U samples for fissions produced by neutrons from the ^{7}Li(p,n)^{7}Be* reaction and from neutron scattering in the detector and neutron source assemblies. These corrections are discussed in greater detail in Meadows (1983b).

NEUTRON SOURCE AND ENERGY RESOLUTION

Protons, accelerated by the Argonne 8-MeV Tandem Dynamitron Accelerator to energies ranging from 3.6 to 4.2 MeV, impinged on a thin Li metal target and produced neutrons by the ^{7}Li(p, n)^{7}Be reaction leading to the ground state of ^{7}Be. A secondary reaction, leading to the first excited state of ^{7}Be, was also possible at these energies, and it produced about 10% of the total neutron yield. The proton energy was controlled by a 90° analyzing magnet and slit feedback system that was calibrated by comparing the observed thresholds of the ^{7}Li(p,n)^{7}Be, ^{11}B(p,n)^{11}C and ^{27}Al(p,n)^{27}Si reactions with those given by Marion (1966) and Beckner *et al.* (1961).

Targets for the neutron source were prepared by evaporating a thin layer of Li metal onto an 0.25 mm-thick Ta plate. Target thickness was controlled by the weight of Li evaporated, but the final target thickness, in terms of proton energy loss in the Li layer was determined by measuring the energy dependence of

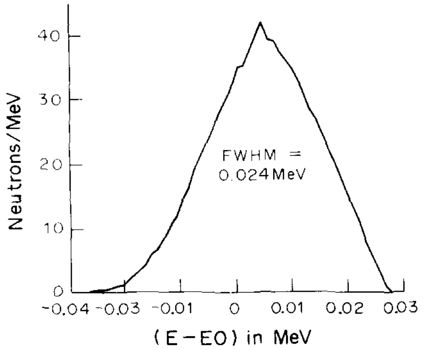

Fig. 2. A typical energy resolution function for the neutrons used in the measurement of the neutron energy dependence of the ^{238}U/^{237}Np fission cross section ratio.

the 0 neutron yield just above the ^{7}Li(p,n)^{7}Be threshold. The deposit thickness usually increased with use, so the targets were replaced after a few hours when the energy loss had increased by 20–30%. The target thickness was measured at the beginning and end of the period of use, and it was assumed that the thickness increased linearly with use. The energy distribution of the primary source reaction neutrons that were incident on the sample was calculated from the incident proton energy, the angular distribution of the source reaction, the thickness of the Li metal target, and the source–sample geometry. The energy resolution used for the shape measurements (full-width-at-half-maximum) ranged from 18 to 30 keV, with an average value of ~24 keV. The variation was due to the spread in the initial Li target thickness plus the increase in target thickness with use. A typical resolution function is illustrated in Fig. 2. Much broader energy resolution was used for the normalization measurements, where the function was nearly rectangular and about 105 keV wide. These deposits were too thick to be measured by the 0 yield method, so the energy loss in the Li layer was estimated from the weight of Li evaporated.

In order to test the energy scale and target measurement procedures, a transmission measurement was made across the narrow C resonance near 2 MeV using a thin Li target. The location of the resonance was determined to be 2.079 keV, as compared with the reported value of 2.077 ± 0.002 keV (Mughabghab and Garber, 1973). The thickness of the Li target, based on the shape of the transmission curve, was 21 keV as compared with the 24 keV determined from the 0 neutron yield.

SAMPLES

All the samples had either been used in earlier measurements or were made from the same material as samples used in those measurements, and details as to their preparation and assay are summarized by Meadows (1983b, 1988). The preparation of the material used for the ^{237}Np samples is detailed in Meadows (1983a).

The isotopic composition and weights of the samples used are given in Table 1. All samples, with the exception of $^{238}_{60}$U, were prepared by electro-deposition of 2.54 cm dia deposits onto 0.13 mm-thick Pt plates, with subsequent heating to a temperature high enough to convert them to U_3O_8 or Np_2O_3. $^{238}_{60}$U was deposited onto a 0.25 mm-thick, stainless-steel plate and was heated to a much lower temperature. As a result, the form of the deposit is a mixture of hydrated oxides, with an overall composition approaching $UO_4 \cdot H_2O$. The specific activities of the sample materials were calculated using the isotopic compositions in Table 1 and the half-lives recommended in Reich (1985), and the sample weights were then determined by low-geometry α-counting. New α-counts were made specifically for the present measurement, so the sample weights of Table 1 are not identical with those of Meadows (1988). The differences are negligible, with the exception of the $^{238}_{60}$U sample where it is −0.8%.

ERRORS AND RESULTS

The principal sources of experimental error for the normalization measurements are listed in Table 2, which also gives an indication of their magnitude and of their correlation. A more detailed discussion of errors associated with fission ratio measurements is given in Meadows (1987). The individual measurements were combined using the methods described by Smith (1981, 1987) and the correlation coefficient for the results at the two energies was found to be 0.70. Only relative errors were important for the shape measurement. As a test the data was fitted with a polynomial that was second-order in E_n^2; then χ^2, based on the statistical counting errors, was calculated. When the divergent point at 2.90 MeV was discarded, χ^2 was 1.1, indicating that the scatter in the shape data was almost entirely due to the statistical counting errors.

The results of the absolute measurements are listed in Table 3. The shape measurements were normalized to the absolute measurements and are given in Table 4. Both sets of data were converted to ^{238}U fission cross sections using the ^{237}Np fission cross sections from ENDF/B-V (1979, 1981).

The purpose of the absolute ratio measurements in this work was to normalize the shape determinations, but they can also be used to test the internal con-

Table 1. The weights of the samples and their isotopic composition in mol %

Sample No.	Isotope number 234	235	236	237	238	Weight of element (mg)
Np-237-76	—	—	—	100	—	1.775
Np-237-79	—	—	—	100	—	0.656
U-238-60	←	<0.6 ppm	→	—	100.0	1.946
U-238-210	1.012	2.369	0.0001	—	96.49	1.180
U-238-211	1.012	2.369	0.0001	—	96.49	1.699
U-238-217	1.012	2.369	0.0001	—	96.49	0.439

Table 2. Summary of errors for the absolute ratio measurements

Error	Magnitude (%)
Uncorrelated error	0.7–0.9
Thickness correction	
Method[a]	0.3
Sample[b]	0.2–1.3
Extrapolation correction	
Method[a]	0.3
Sample[b]	0.4–0.6
Scattering correction[a]	0.6
Half-lives	
^{234}U[c]	0.12
^{238}U[d]	0.11
^{237}Np[a]	0.47
Isotopic analysis[c]	0.4
Second neutron group[a]	0.0–0.3
α-count[b]	0.3–0.4

[a] Fully correlated for all measurements.

[b] Fully correlated for all measurements with a particular sample.

[c] Fully correlated for samples U-238-210, 211 and 217.

[d] Applies only to sample U-238-60.

sistency of evaluations and other data sets. In Table 5 this work is compared with ENDF/B-V and with experimental data sets, where the ^{238}U and ^{237}Np fission cross sections were measured relative to ^{235}U by the same experimental group. The agreement is generally good, considering the experimental errors.

The primary purpose of these measurements was to search for significant structure in the ^{238}U fission cross section between 1.9 and 2.56 MeV neutron energy, particularly in the vicinity of 2.3 MeV. In this region, the high-resolution measurements of Blons *et al.* (1976) showed a fine structure of small narrow peaks with an average separation of ~35 keV, and also a "hole" near 2.3 MeV that was over 100 keV wide with a cross section change of more than 10%. These data have been smoothed with the resolution function shown in Fig. 2 and plotted in Fig. 1 for comparison with the present measurements. The fine structure is greatly reduced, but the "hole" at 2.3 MeV remains a pronounced feature. A certain regularity in the data from the present measurement suggests the residual fine structure in the smoothed data of Blons *et al.*

Table 3. The results of the normalization measurements

Sample No.	Ratio	Statistical error (%)	Total error (%)	σ^{a} ^{238}U (barn)
$E_n = 2.15$ MeV $\Delta E_n = $ (FWHM) $= 0.112$ MeV				
U-238-60/Np-237-79	0.3161	0.66	2.07	0.5341
U-238-210/Np-237-76	0.3214	0.50	1.71	0.5432
U-238-211/Np-237-76	0.3132	0.60	2.01	0.5396
Average	0.3190	—	1.36	0.5390
$E_n = 2.453$ MeV ΔE_n(FWHM) $= 0.106$ MeV				
U-238-60/Np-237-70	0.3268	0.59	2.09	0.5549
U-238-210/Np-237-76	0.3263	0.55	1.77	0.5540
U-238-217/Np-237-79	0.3248	0.73	1.37	0.5515
Average	0.3256	—	1.00	0.5529

[a] The ^{238}U fission cross sections are based on the ^{237}Np fission cross sections from ENDF/B-V (1979, 1981).

Table 4. The results of the shape measurement

E (MeV)	Resolution (FWHM) (MeV)	Ratio[a]	Statistical error (%)	$\sigma(^{238}U)$[b] (barn)
1.920	0.023	0.3121	0.77	0.5181
1.938	0.024	0.3131	0.81	0.5204
1.957	0.027	0.3184	0.73	0.5298
1.976	0.030	0.3112	0.74	0.5195
1.995	0.031	0.3104	1.11	0.5178
1.999	0.022	0.3106	0.92	0.5187
2.019	0.022	0.3155	0.74	0.5274
2.038	0.023	0.3109	0.74	0.5202
2.058	0.023	0.3152	0.74	0.5327
2.077	0.026	0.3152	0.73	0.5286
2.096	0.027	0.3216	0.74	0.5399
2.116	0.028	0.3202	0.81	0.5383
2.139	0.022	0.3183	0.77	0.5360
2.158	0.025	0.3240	0.76	0.5466
2.177	0.024	0.3241	0.76	0.5471
2.196	0.027	0.3205	0.76	0.5417
2.216	0.028	0.3281	0.76	0.5546
2.218	0.023	0.3206	0.76	0.5418
2.235	0.029	0.3272	0.77	0.5533
2.239	0.023	0.3227	0.76	0.5459
2.260	0.018	0.3250	0.74	0.5499
2.278	0.024	0.3222	0.77	0.5451
2.280	0.020	0.3218	0.73	0.5444
2.290	0.021	0.3141	0.74	0.5312
2.297	0.025	0.3230	0.75	0.5465
2.307	0.024	0.3241	0.76	0.5486
2.319	0.022	0.3228	0.76	0.5464
2.319	0.022	0.3235	0.77	0.5476
2.336	0.026	0.3237	0.74	0.5481
2.337	0.023	0.3224	0.76	0.5458
2.356	0.026	0.3256	0.89	0.5507
2.359	0.020	0.3215	1.30	0.5446
2.380	0.020	0.3216	0.76	0.5448
2.398	0.022	0.3256	0.77	0.5519
2.420	0.018	0.3247	0.74	0.5523
2.438	0.021	0.3206	0.76	0.5435
2.458	0.023	0.3222	0.82	0.5465
2.479	0.019	0.3245	0.75	0.5503
2.499	0.020	0.3238	0.75	0.5495
2.518	0.019	0.3232	0.75	0.5489
2.538	0.023	0.3225	0.75	0.5476
2.558	0.021	0.3248	0.91	0.5514

[a] The ratios are normalized to the absolute measurements in Table 3.

[b] The ^{238}U fission cross sections are based on the ^{237}Np fission cross sections from ENDF/B-V (1979, 1981).

(1976), but the "hole" is not observed. At this resolution, the cross section varies smoothly with energy and shows no gross structure. There is a divergent point at 2.290 MeV, but measurements ~10 keV above and below that energy agree with the mass of the data. Most of the available data from other experimenters cannot show such structure, as the resolution is too broad or the data points are too widely spaced. However, Difilippo *et al.* (1980) have reported white source measurements over this energy region with resolution at 2 MeV of ~25 keV, which is comparable to the present work. Their data shows little evidence of the "hole" at 2.3 MeV.

Table 5. Comparison of the absolute $^{238}U/^{237}Np$ fission cross section ratios with other data sets

	E (MeV)	Ratio	Error (%)	Ratio to this work
This work	2.150	0.3190	1.2	—
	2.453	0.3256	1.0	—
Stein *et al.* (1968)	2.150	0.3117	3.4	0.977
	2.453	0.3140	3.4	0.964
Behrens and Carlson (1977)				
Behrens *et al.* (1982)	2.150	0.3342	2.2	1.048
	2.453	0.3343	2.2	1.026
Meadows (1972, 1985)	2.150	0.3128	1.5	0.980
	2.453	0.3207	1.2	0.985
Meadows (revised (1983b)	2.150	0.3176	1.6	0.996
	2.453	0.3237	1.2	0.994
ENDF/B–V (1979, 1981)	2.150	0.3182	—	0.980
	2.453	0.3178	—	0.976

To summarize, absolute measurements of the $^{238}U/^{237}Np$ fission cross section ratio were made at 2.150 and 2.453 MeV and compared with other data sets. Measurements of the shape of the $^{238}U(n,f)$ cross section were made relative to ^{237}Np between 1.9 and 2.6 MeV with ~24 keV neutron energy resolution. At this resolution, the cross section was found to vary smoothly with energy and showed no gross structure near 2.3 MeV. However, the fine structure would not have been observed. It may be concluded that the $^{238}U(n,f)$ excitation function can be an adequate standard in this energy region provided that averages are made over intervals >25 keV.

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