

TABLE II
COMPARISON OF RESONANCE INTEGRALS FOR THE 192 eV
RESONANCE OF U²³⁸

U ²³⁸ :H	Temp. (°K)	μ	I_μ (barns)	$I_{\mu(0)}$ (barns)
1:1	0	1.57	0.172	0.172
	300	1.68	0.178	0.180
	600	1.74	0.184	0.188
	900	1.74	0.191	0.195
1:5	0	3.50	0.302	0.302
	300	3.82	0.329	0.335
	600	3.73	0.355	0.360
	900	3.53	0.380	0.381
1:10	0	4.92	0.393	0.393
	300	5.11	0.443	0.446
	600	4.70	0.488	0.484
	900	4.25	0.529	0.514
1:20	0	6.71	0.516	0.516
	300	6.25	0.608	0.602
	600	5.34	0.681	0.657
	900	4.62	0.742	0.700
1:100	0	11.0	0.983	0.983
	300	6.74	1.23	1.20
	600	5.07	1.37	1.30
	900	4.12	1.46	1.37

As expected, the trend in the interpolation parameter with increasing dilution is towards the "narrow-resonance" limit. Another trend which becomes apparent from a consideration of dilute mixtures is that μ moves back towards the "infinite-mass" value as the temperature increases.

The results as a whole show that, although the interpolation parameter μ may exhibit quite marked changes with temperature in some cases, the variation has a comparatively small effect on the effective resonance integral. Thus it appears that, in applying the formulas of Goldstein and Cohen, no significant error should result from the use of the 0°K value of μ in the calculation of the total effective resonance integral, except, perhaps, when determining the Doppler coefficient, for which a more thorough investigation may be needed.

It is interesting to note that, for the values of the parameters relevant to the resonances of Th²³² and U²³⁸ under practical conditions, the function $L(\theta, a_1, a_\lambda, x_0)$ has been found to be given within a few percent by the formula

$$L(\theta, a_1, a_\lambda, x_0) \simeq \frac{4}{\pi} J(\theta, a_1) J(\theta, a_\lambda) \tan^{-1} \frac{x_0}{c_1 + c_\lambda}, \quad (11)$$

which is exact in the limit as $\theta \rightarrow \infty$ and also in the limit as $x_0 \rightarrow \infty$.

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REFERENCES

1. R. GOLDSTEIN AND E. R. COHEN, *Nucl. Sci. Eng.* **13**, 132-140 (1962).

2. L. DRESNER, "Resonance Absorption in Nuclear Reactors." Pergamon Press, New York, 1960.
3. M. E. ROSE, W. MIRANKER, P. LEAK, L. ROSENTHAL, AND J. K. HENDRICKSON, WAPD-SR-506 (1954).
4. V. BELL, P. BUCKLER, AND I. PULL, Table of the function $J(\xi, k)$. Winfrith Computing Branch, U.K.A.E.A. (Unpublished).

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The Average Capture/Fission Ratio of U²³³ for Epithermal Neutrons

In thermal reactors using U²³³ as fuel somewhere around 10% of all absorption in the fuel may be expected to involve epithermal neutrons. While the capture/fission ratio α for thermal neutrons is known to be close to 0.10 (I) and while the fission, scattering, and total (i.e., fission plus capture plus scattering) cross sections have been studied as a function of energy (I), nevertheless there remains significant uncertainty as to the average capture cross section and capture/fission ratio for epithermal neutrons. If the latter ratio were sufficiently high the consequent effect on the neutron economy of a U²³³ thermal breeder could be serious. The purpose of the work described here was to compare the epithermal capture and fission values to the thermal values by the use of Cd ratios, radiochemical analyses of fission products, and mass-spectrographic ratios of U²³⁴/U²³³. Thereby an experimental α_{epi} for epithermal neutrons was evaluated, as were infinitely dilute resonance integrals for capture and fission, I_c and I_f , for U²³³.

Microgram quantities of U²³³ (prepared to be especially free of U²³⁴ by milking U²³³ from Pa²³³) were irradiated in both the LITR (ORNL Low Intensity Test Reactor) and ORR (Oak Ridge Research Reactor), both with and without 40 mil Cd filters surrounding them. The thermal fluxes were about 2×10^{13} and 1×10^{14} n/cm²-sec respectively. Both thermal and epithermal fluxes were monitored at the sample positions with cobalt in a dilute Co-Al alloy (containing 0.151% Co).

After irradiation, the uranium was separated from fission products and other impurities and analyzed mass-spectrographically for U²³⁴ produced. Analyses were made on the four fission products, 12.8-day Ba¹⁴⁰, 67-hr Mo⁹⁹, 54-day Sr⁸⁹ and 65-day Zr⁹⁵, by using standard procedures (2) with few modifications, in order to determine the number of fissions which occurred during irradiation.¹

¹The yield of I¹³¹ was also measured and found to give a resonance integral some 36% higher than that computed from Sr⁸⁹, Mo⁹⁹, and Ba¹⁴⁰. Since I¹³¹ is on the slope of the

The 2200 meters/sec cross section σ_0 and resonance integral I (above 0.54 ev),² used for Co were 37.0 and 75 b, respectively. The Maxwellian temperature was assumed to correspond to $E_m = 0.029$ ev. In the ORR irradiation used for measuring the capture parameters, the (Maxwellian flux)/(epithermal (1/E) flux per lethargy unit) was found to be 16.45³ and the integrated thermal flux-time, 3.65×10^{20} neutrons/cm². From the latter figure and the average observed U²³⁴/U²³³ mass ratio, the effective thermal (not 2200 meters/sec) cross section for capture was 61.3 b. By the use of Westcott's value (5) for "g", the thermal flux, the flux ratio, and the mass analyses for both filtered and bare samples, the following values were obtained for the 2200 meters/sec cross section and resonance integral for U²³³ capture

$$\begin{aligned}\sigma_0 &= 51.7 \text{ b} \pm 5\% \\ I \text{ (above 0.54 ev)} &= 147 \text{ b} \pm 5\%\end{aligned}$$

The 2200 meters/sec value may be compared with the World Consistent Set value of 53 ± 5.5 b or the U. S. value of 51 ± 5.5 (1). Westcott (5) quotes a value of the difference between the resonance integrals for absorption and fission (which should equal the capture integral) of 117 b; however this value being the difference between two large numbers may be subject to appreciable error.

By comparison of the fission product analyses in the bare and filtered samples and by using the monitored flux-time values, an average fission resonance integral of 865 ± 40 b was obtained for Ba¹⁴⁰, Sr⁸⁹, and Mo⁹⁹ analyses (6). This value compares favorably with the resonance integral of 900 b estimated from resonance data in BNL-325 (2nd ed.) and with the 820 b of Terasawa's computation (7) also using cross section vs. energy data.

Although direct measurements of the fission product yields of Sr⁸⁹, Mo⁹⁹, Ba¹⁴⁰, and Zr⁹⁵ for resonance neutron spectra have not been carried out, for the purpose of this study their values may be inferred. Variations of fission product yields in the thermal and resonance regions have generally been ascribed to varying contributions of symmetric and asymmetric fission in different levels of the fissioning nucleus (8). Ratios of asymmetric to symmetric fission for reactor epithermal neutrons,

$$R_{\text{epi}} = \frac{(A'_{\text{asym}}/A'_{\text{sym}})_{\text{epi}}}{(A'_{\text{asym}}/A'_{\text{sym}})_{\text{th}}}$$

have been measured for both U²³⁵ and U²³³ as 1.18 and 1.12 respectively (9). A change in this ratio from unity is primarily due to a change of the yield in the region of the valley of the fission product yield curve. The yield at the peak is essentially unperturbed. This is borne out by the constancy of the ratio of yields (10) of Mo⁹⁹/Ba¹⁴⁰ at both thermal and

fission product yield curve, its yield is more sensitive to symmetric-asymmetric competition than those fission products in the peak of the curve. For example, in U²³⁵, the 14 Mev neutron fission product yield of I¹³¹ is about 40% greater than the thermal yield (3).

² All resonance integrals mentioned here refer to the total reaction rate above the specified lower limit, i.e., no 1/v "tail" has been subtracted.

³ For conventions used, see Stoughton and Halperin (4); one difference is that the subscript zero has been dropped in the symbol for infinitely dilute resonance integral.

epithermal energies in U²³⁵. Since U²³³ has an even smaller value of R_{epi} than U²³⁵, the yields for peak fission products can similarly be expected to remain approximately constant for those resonances contributing to the fission resonance integral. Selected fission product yields in U²³³ have been measured for a fast neutron fission spectrum by Bonyushkan (11). The yields for Sr⁸⁹, Mo⁹⁹, and Ba¹⁴⁰ were found to be in essential agreement with thermal yields within the limits of error of the measurement. There appear to be no similar data available for Zr⁹⁵. In the present work the yield ratio of

$$\frac{Y(\text{Zr}^{95})_{\text{epi}}}{Y(\text{Zr}^{95})_{\text{th}}} \bigg/ \frac{Y(\text{Ni})_{\text{epi}}}{Y(\text{Ni})_{\text{th}}}$$

was found to be 1.14 where Ni = Sr⁸⁹, Mo⁹⁹, or Ba¹⁴⁰. In view of the internal agreement of the Sr⁸⁹, Mo⁹⁹, and Ba¹⁴⁰ and the direct evidence of their unaltered yields in a fast spectrum and the lack of data for Zr⁹⁵, we have not included the value derived from Zr⁹⁵ in the resonance integral average.

The ratio of the average capture/fission ratio for epithermal neutrons α_{epi} should be given by the ratio of the resonance integrals, or

$$\alpha_{\text{epi}}(\text{U}^{233}) = 147/865 = 0.170 \pm 0.017$$

The effect of this value of α_{epi} on thermal breeding may be seen by expressing the effective number of neutrons produced/neutron absorbed in fuel, η_{eff} , in terms of the thermal value η_{th} , the thermal and epithermal values of α (i.e., α_{epi} and α_{th}), and the fraction of absorptions in fuel which occur in the epithermal region f_{epi} . On the assumption that the number of neutrons emitted per fission (which is equal to $\eta(1 + \alpha)$) is nearly independent of the incident neutron energy over the range in question the η ratio becomes

$$\frac{\eta_{\text{eff}}}{\eta_{\text{th}}} = 1 - f_{\text{epi}} \left(\frac{\alpha_{\text{epi}} - \alpha_{\text{th}}}{1 + \alpha_{\text{epi}}} \right)$$

If a value of 0.100 (1) is taken for α_{th} , the above equation becomes

$$\begin{aligned}\eta_{\text{eff}}/\eta_{\text{th}} &= 1 - f_{\text{epi}} (0.060 \pm 0.015) \\ &= 0.994 \text{ for } f_{\text{epi}} = 0.1 \\ &= 0.988 \text{ for } f_{\text{epi}} = 0.2\end{aligned}$$

REFERENCES

1. D. J. HUGHES, B. A. MAGURNO, AND M. K. BRUSSEL, Eds., BNL-325, Suppl. 1 to Second Edition, Brookhaven National Laboratory (January 1960).
2. ORNL Master Analytical Manual, TID-7015 (1957).
3. Atomic Energy Report ANL-5800, pp. 13-14. (1958)
4. R. W. STOUGHTON AND J. HALPERIN, *Nucl. Sci. Eng.* **6**, 100-118 (1959).
5. C. H. WESTCOTT, "Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra." AECL-1101 (3rd ed., corrected) (November 1960).
6. S. KATCOFF, *Nucleonics* **18** (11), 201 (1960).
7. S. TERASAWA, Oak Ridge National Laboratory Report ORNL-2553 (August 20, 1958).
8. J. R. HUIZENGA AND R. VANDEBOSCH, in *Nuclear Reactions*, Vol. 2, P. M. ENDT AND M. DEMEUR, eds., Chap. II, p. 105, Interscience-Wiley, New York, 1962.

9. W. H. BURGUS, Atomic Energy Commission Report IDO-16797 (July 30, 1962) U. S. Dept. of Commerce, Washington 25, D. C., \$0.50; R. B. REGIER, W. H. BURGUS, AND R. L. TROMP, *Phys. Rev.* **163**, 1589 (1959).
10. G. A. COWAN, A. TURKEVITCH, AND C. I. BROWN, *Phys. Rev.* **122**, 1286 (1961).
11. E. K. BONYUSHKAN *et al.*, *At. Energ. (USSR)* **10**, 13 (1961).
12. R. L. MACKLIN, G. DESAUSSURE, J. D. KINGSTON, AND W. S. LYON, *Nucl. Sci. Eng.* **8**, 210 (1960).

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The $O^{16}(n,p) N^{16}$ Reaction Cross Section

The $O^{16}(n,p) N^{16}$ reaction produces the principal radioactivity in the coolant during operation of water-cooled reactors. High energy gamma rays accompany the decay of N^{16} . The cross section for this reaction has been the subject of several experimental investigations, the results of which have at times been in apparent agreement and at other times in apparent disagreement. It is the purpose of this note to summarize a review (1) of the available information concerning the cross section for this reaction with regard to application to water-cooled reactors.

The reaction cross section averaged over the fission neutron spectrum has been deduced from activation measurements obtained using water circulated through a reactor. The activation flux used by Henderson and Tunnicliffe (2) in arriving at a cross section of $18.5 \pm 1.5 \mu\text{b}$ is determined from a calculation of the average path length of a fission neutron in the cooling water channel of the reactor fuel element. Honeck has calculated this average path length using Monte Carlo techniques which account for self-shielding in the fuel element, thereby obviating the need for an assumed angular distribution of the neutrons entering the coolant channel. The results, as reported by the Neutron Cross Section Evaluation Group (3), are about one half as large as Henderson's, and would increase the cross section by about 2. When the integral activation cross section, of $19 \pm 5 \mu\text{b}$, reported by Roys and Shure (4) is

adjusted for the more recent information on the yield of 0.74 high energy gamma rays per N^{16} disintegration and the improved confidence of the magnitude of the $Al(n,\alpha)$ cross section which had been used as a monitor of the activation flux, the $O^{16}(n,p)$ value becomes $21 \pm 4 \mu\text{b}$.

Martin (5) had measured the excitation function of the $O^{16}(n,p) N^{16}$ reaction from 12.4 to 18 Mev. DeJuren, Stooksberry, and Wallis (6) have extended the range of the measurements, covering the region from 11 to 19 Mev with improved energy resolution. Their more accurate technique for monitoring the irradiation flux gives results lower than those of Martin by about a factor of 2. Their results exhibit a prominent resonance near 11.8 Mev previously unobserved. Similar measurements have been made between 12.6 and 16.3 Mev by Seeman and Moore (7).

In averaging the excitation function, it is important that the absolute value and shape of the fission neutron spectrum within several Mev of the reaction threshold of 10.23 ± 0.01 Mev be used. In the energy region above 10 Mev, experimental information on this spectrum is scanty. The results of Watt (8) and of Frye and Rosen (9) are given in Fig. 1 with arbitrary normalization at 7 Mev, with consistency noted even above 10 Mev. Also seen in Fig. 1 is the shape of the Watt spectrum ($e^{-E} \sinh \sqrt{2E}$) and that of the Cranberg spectrum ($e^{-E/0.965} \sinh \sqrt{2.29E}$), both normalized to the experimental results in the 3-4 Mev range. These shapes are consistent with the available experimental information in this region, with the Watt spectrum providing a somewhat better fit. The relative number of neutrons with energies greater than 10 Mev predicted by these representations differs by about 17%.

When the results of DeJuren, Stooksberry, and Wallis are weighted with the Watt spectrum, the average cross section is $19 \mu\text{b}$. When weighted with the Cranberg spectrum, the average cross section is $16 \mu\text{b}$. These results agree with the adjusted integral activation cross section of Roys and Shure, $21 \pm 4 \mu\text{b}$, to within the uncertainties of the experimental information.

REFERENCES

1. K. SHURE, "The $O^{16}(n,p) N^{16}$ Reaction Cross Section." WAPD-BT-25, p. 27 (May 1962).
2. W. J. HENDERSON AND P. R. TUNNICLIFFE, "The Production of N^{16} and N^{17} in the Cooling Water of the NRX Reactor." *Nucl. Sci. Eng.* **3**, 145 (1958).
3. S. MOORE, C. PORTER, AND R. SHER, "Neutron Cross Section Evaluation Group—Newsletter No. 2." BNL-634 (October 1960).
4. P. A. ROYS AND K. SHURE, "Production Cross Section of N^{16} and N^{17} ." *Nucl. Sci. Eng.* **4**, 536 (1958).
5. H. C. MARTIN, "Cross Sections for the $O^{16}(n,p) N^{16}$ Reaction from 12 to 18 Mev." *Phys. Rev.* **93**, 498 (1954).
6. DEJUREN, STOOKSBERRY, AND WALLIS, "Measurement of the $O^{16}(n,p) N^{16}$ Cross Section from 11 to 19 Mev." *Phys. Rev.* **127**, 1229 (1962).
7. K. W. SEEMAN AND W. E. MOORE, "The $O^{16}(n,p) N^{16}$ Cross Section from 12.6 to 16.3 Mev." KAPL 2214 (September 1962).
8. B. E. WATT, "Energy Spectrum of Neutrons from Thermal Fission of U-235." *Phys. Rev.* **87**, 1037 (1952).
9. CRANBERG, FRYE, NERESON, AND ROSEN, "Fission