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The Resonance Absorption of Uranium Metal and Oxide

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THE RESONANCE ABSORPTION OF URANIUM METAL AND OXIDE.

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Summary:

The resonance integrals for uranium metal and uranium oxide have been determined for a 1/E flux. The following results were obtained

$$\text{Metal RI} = 2.95 + 25.8\sqrt{\frac{S}{M}}$$

$$\text{Oxide RI} = 4.15 + 26.6\sqrt{\frac{S}{M}}$$

The oxide value agrees with the expression found earlier at this laboratory. But the result for the metal is 4.5 % larger than the earlier one.

In addition, the resonance absorption in a R1 fuel rod has been compared with that for a cadmium-covered rod placed in an approximate cell boundary flux. The former came out 3 % larger than the latter. A comparison of the fuel rod absorption with that for a 1/E flux yields a corresponding figure of 7 %. The neutron flux was monitored below the lowest resonance in uranium.

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The resonance absorption of uranium metal and oxide.

E. Hellstrand and G. Lundgren

I. Introduction

The aim of the present investigation was twofold. Firstly, recent measurements by Smith, Hardy, Klein and Mitchell (1) of the relative epicadmium absorption in uranium metal and oxide have given a larger value of metal to oxide resonance absorption than that calculated from formulas earlier obtained here (2). Thus a renewed study of the effective uranium resonance integral was considered well motivated.

Secondly, one generally aims at measuring the resonance absorption in a 1/E flux and published values of resonance integrals are, more or less correctly, claimed to be valid for such a neutron spectrum. However, the energy dependence of the neutron flux at the surface of a fuel rod in a reactor lattice may strongly deviate from 1/E, as demonstrated for instance by the calculations of Bigham and Pearce (3). A separate study has therefore been made of how the resonance absorption for a R1 fuel rod in a lattice position compares with that for a cadmium-covered rod of the same diameter placed in an approximate cell boundary flux.

II. Measurement of the resonance integrals for uranium metal and uranium oxide

1. Method

The method of measurement resembles the one used in the study of thorium metal (4). The calibration in barns was obtained by means of the dilute resonance integral of gold and the thermal cross sections for gold and uranium (for the old measurements (2) the dilute resonance integral for uranium was used). The arrangement for the irradiations is illustrated in Fig. 1. The neutron flux was monitored with cadmium-covered gold foils. They were placed about 120 mm above the samples to avoid screening effects from the uranium resonances.

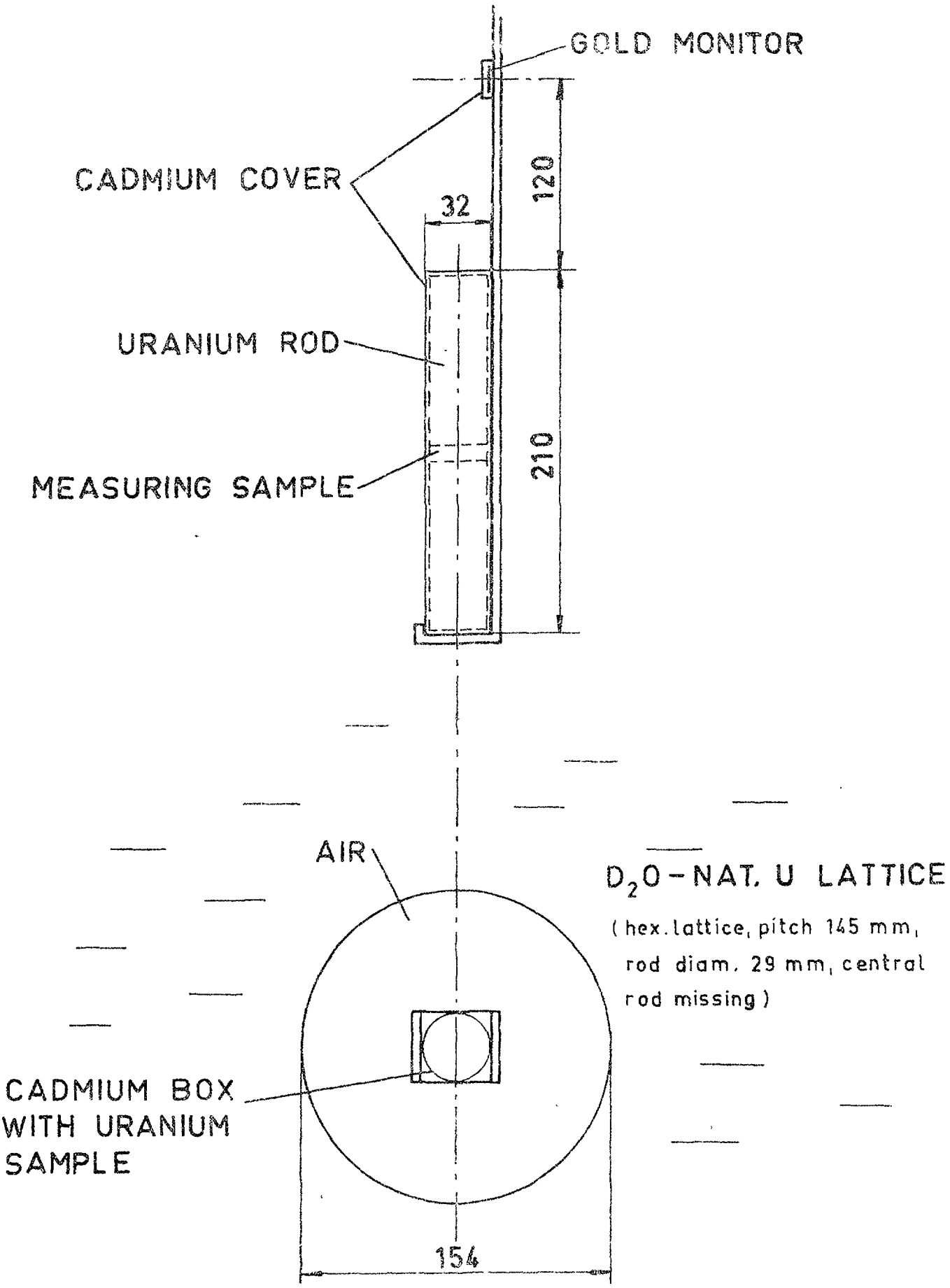

Fig. 1. Experimental arrangements

As the linear dependence of RI on $\sqrt{\frac{S}{M}}$ has been well established for the range of S/M of most interest only two rod diameters were used. These were for metal 28 mm and 10 mm and for oxide 17 mm and 12.5 mm.

The uranium samples consisted of cylindrical disks (2 and 4 mm thick for the two metal rods and 4 and 18 mm for the oxide rods) placed between 100 mm long end-pieces of the same diameter. In order to fix the position of the disks the bars were wrapped in thin aluminium foils. The assembly was then enclosed in a 220 mm long cadmium tube of 32 mm diameter and 0.8 mm wall thickness. The irradiations were performed in the central channel of the reactor R1 at maximum flux position. For all the measurements the reactor power was 4 kW and the irradiation time 20 min.

The same arrangement was also used for the gold foil irradiations. The foils were thus placed inside cadmium tubes of the same dimensions as those used for the uranium. The foils were made of a lead gold alloy containing 0.1 wt % gold, and the foil thickness was 0.1 mm. The selfscreening of the 4.9 eV resonance in the foil is estimated as about 1.5 % for an isotropic flux.

The calibration of the detector equipment for relating the gold and uranium activities was performed by irradiating a thin foil of depleted uranium surrounded by a pair of lead-gold foils in the thermal column of the reactor. Thin foils of aluminium were placed between the uranium and lead-gold foils in order to prevent fission fragments from reaching the latter. The U^{235} content in the uranium foils was 0.04 %, and the fission product activity was thus small and could be corrected for. In order to get activities of the same order of magnitude as in the central channel measurements the reactor power was in this case chosen as 100 kW and the irradiation time as 30 min.

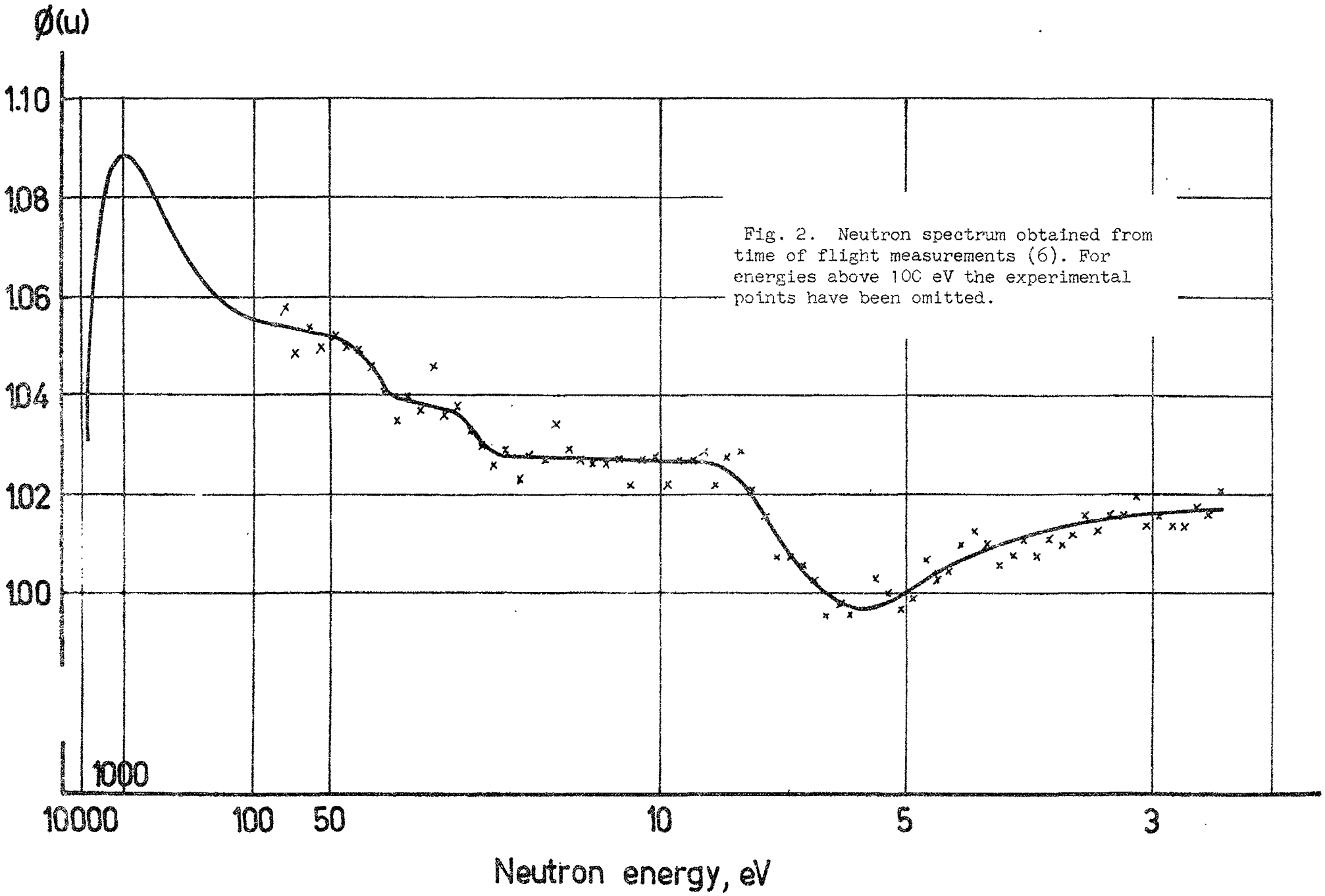

Fig. 2. Neutron spectrum obtained from time of flight measurements (6). For energies above 100 eV the experimental points have been omitted.

2. Activity measurements

The samples were dissolved in 50 % aqeous solution of nitric acid and the volume was so chosen in each case that the uranium concentration was the same for all solutions. Samples of equal volume were taken from these solutions and, three days after the irradiations, the 106 keV γ-line from the neptunium decay was measured with the aid of a γ-scintillation spectrometer. The contribution to the activity from fission products was corrected for using the results of earlier experiments involving chemical separation (2).

A correction for the natural activity of uranium was obtained from measurements on unirradiated samples. The activities from the lead-gold and the monitor gold foils were measured with β-scintillation counters.

3. Various effects

a) 1/v contribution

To obtain the resonance integrals the 1/v contribution to the capture has to be subtracted. From the calculations by Stoughton et al. (5) we estimate a cadmium cut-off energy of (0.50 ± 0.04) eV, which gives $\sigma_{1/v} = (1.20 \pm 0.05)$ b. The cadmium thickness of the tubes was 0.8 mm and the height-to-diameter ratio was about 7.

b) Energy dependence of the flux

To correct for the non-1/E dependence of the neutron flux the curve of ϕ (u) as a function of energy obtained by Johansson et al. (6) has been used. In a slightly modified form the curve is reproduced in Fig. 2. Defining γ as $\frac{\Sigma \phi (u_i) RI_i}{\Sigma RI_i}$ the actual resonance absorption for this spectrum may be written as γ RI. RI_i is the contribution to the total resonance integral from an individual resonance or a group of resonances as calculated for

instance by Adler, Hinman and Nordheim (7). The flux ϕ (u) is normalized to 1 at 5 eV. γ varies only very little with rod diameter and, considering the uncertainty in ϕ (u) ($\pm$ 1 %), the variation can be neglected. The same value of γ (1.04 $\pm$ 0.015) has been used for all dimensions.

For energies above 10 keV we have no experimental information about the energy dependence of ϕ(u). The decrease in ϕ (u) with increasing energy that starts already at 1 keV most certainly continues. Bigham and Pearce's (3) calculations for the NRX lattice give a ratio of ϕ (u) at 100 keV and 5 eV of about 0.93. Similar conditions might be expected for the RI lattice. The effect on RI is, however, quite small and is neglected.

This might be permissible for an undisturbed cell boundary flux. The large piece of cadmium around the samples in the central channel could, however, introduce some additional effects. The nearest fission sources are depressed, which will enhance the deviation from 1/E at high energies.

To check the magnitude of such an effect threshold detectors were irradiated both inside the normal cadmium tube (l = 220 mm) and in a small one (l = 12 mm). As threshold detectors sulfur (S^{32} (n, p) P^{32}) phosphorus (P^{31} (n, p) Si^{31}) and aluminium (Al^{27} (n, α) Na^{24}) were used. We found that the normalized activities from the small tube were for all three detectors only about 1 % higher than those from the large box. Although the effective threshold energies for the detectors used lie between 3 and 5 MeV the results should be approximately representative for the flux conditions in the energy region around 100 keV.

4. Results

As may be easily verified, the resonance integral RI is obtained from an expression of the form

$$RI = \frac{1}{\gamma} \left[\frac{A^{U}_{epi} \; A^{Au}_{th} \; g^{U} \sigma^{U}_{o} \, (RI^{Au} + \sigma^{Au}_{1/v})}{A^{Au}_{epi} \; A^{U}_{th} \; g^{Au} \; \sigma^{Au}_{o}} - \sigma^{U}_{1/v} \right]$$

A^{U}_{epi} and A^{Au}_{epi} are the monitored neptunium and gold activities from the central channel runs

A^{U}_{th} and A^{Au}_{th} are the corresponding activities from the thermal column runs

$g^{U} \sigma^{U}_{o}$ and $g^{Au} \sigma^{Au}_{o}$ are the 2200 m/sec cross sections corrected for deviations from the 1/v dependence

$(RI^{Au} + \sigma^{Au}_{1/v})$ is the effective epicadmium absorption cross section for the lead-gold foil used

$\sigma^{U}_{1/v}$ is the epicadmium 1/v cross section for uranium.

When evaluating RI from the expression given above the following cross sections have been used. $\sigma^{U}_{o} = 2.71 \pm 0.02$ b (8), $\sigma^{Au}_{o} = 98.8 \pm 0.3$ b (8), $g^{U} = 1.002$ (9), $g^{Au} = 1.006$ (9), $\sigma^{U}_{1/v} = 1.20 \pm 0.05$ b and, after correction for self shielding, $RI^{Au} + \sigma^{Au}_{1/v} = 1525 \pm 35$ b (10, 11).

The correction factor γ only takes into account the influence of the non-1/E dependence of the neutron flux on the resonance absorption in uranium. It is true that the experimental values of ϕ (u) show a tendency to increase with decreasing energy below the lowest resonance in uranium. However, a neglect of as large an increase as 10 % at the cadmium cut-off energy affects the resonance integral for the thickest uranium rod with less than 1 % and still less for thinner rods. The effect has therefore been neglected.

The values of RI for metal and oxide obtained from different runs are given in Table I.

Table I.

Sample	U - metal $\rho = 18.80$		UO_2 $\rho = 10.60$	
Diam. in mm	10.0	28.0	12.5	17.0
RI	14.55 14.92 15.00 15.00 14.90 14.96	10.22 10.44 9.90 9.91 10.19 9.89 9.88	18.37 18.98 18.83 18.74	16.47 16.65 16.69 16.72 16.70 16.97
Mean value	14.89	10.06	18.73	16.70
From ref.	14.23	9.64	18.74	16.69

Assuming a linear dependence of RI as a function of $\sqrt{\frac{S}{M}}$ the following expressions are obtained:

$$\text{Metal: } RI = 2.95 + 25.8 \sqrt{\frac{S}{M}}$$

$$\text{Oxide: } RI = 4.15 + 26.6 \sqrt{\frac{S}{M}}$$

Taking into account the experimental uncertainties as well as those for the cross sections used, the limits of error for the resonance integrals calculated from the two formulas are estimated as $\pm$ 4 %.

The expression for uranium oxide is the same as that given in (2). For the metal the new expression gives resonance integrals which are 4.5 % larger than those obtained from the old formula. The reason for this discrepancy is not clear. The old measurements were no doubt less accurate than the new ones and in addition, a minor systematic error has been discovered for the metal measurements in (2). During the metal irradiations one of the uranium rods in the inner ring of the reactor lattice had been removed by the reactor operators. This causes a slight decrease of the high energy tail of the neutron spectrum, which decreases the resonance absorption for the lumped uranium relative to that for the "infinitely" thin uranium standard. This effect could be of the order of 1 % but hardly larger than that.

There are some other remarks which should be made concerning the old measurements. The deviation from 1/E of the neutron flux was not known at that time and no correction factor γ was applied. γ for those measurements was only about 1.03 as the cross section calibration occurred at a somewhat higher energy than for the present work. On the other hand, for the estimate of the self screening effect in the thin uranium samples the Doppler broadening of the 6.7 eV resonance was disregarded. The self screening factor thereby came out about 1.5 % too high, yielding too low values of the effective resonance integrals by the same amount. The two effects counteract each other. A third point of interest is the placing of gold foils for monitoring purposes on the cylindrical surface of the cadmium tube containing the uranium rods. It is no doubt desirable to have the monitors close to the measuring samples, but the small difference between the

energy of the main resonance in gold (4.9 eV) and that of the largest resonance in uranium (6.7 eV) can introduce complications. A special arrangement was used to monitor, in principle, only the neutron current entering the uranium rods (compare (2) for details). For the thickest uranium rods, however, the larger depression of the neutron flux around the 6.7 eV resonance will be shifted into the region of the gold resonance by oxygen scattering. A minor local depression could therefore appear also in the neutron current registered by the gold monitors. The same effect arises if the shielding of the monitor foils from the beam transmitted through the uranium rods is not complete. For our present work we have therefore chosen to place the gold monitors farther away from the uranium samples. Some additional comments concerning a similar problem are discussed in the next section.

A direct comparison with the results obtained by Smith et al. (1) is hampered by the large difference in the energy distribution of the neutron fluxes for the two measurements. Their measurements were made in a fairly close-packed light water system. The neutron spectrum for such a system shows a peak for high energy neutrons. Klein has estimated the excess absorption in this peak to about 1.5 b (private communication) compared to that for a 1/E spectrum. Considering this, their ratio of the resonance absorption for metal and oxide agrees with ours within the limits of error.

III. Measurement of the resonance capture in the presence of fission sources

1. Method

A comparison has been made between the epicadmium absorption in a fuel rod of the R1 lattice and that of a cadmium-covered rod placed in an approximate cell boundary flux. For convenience the ratio of the two quantities will be called δ. The reactor lattice was completed at the center of the reactor by

inserting an 80 cm long fuel rod surrounded by heavy water in the central channel. The rod was divided into two equal pieces. Between these pieces a small cadmium box (height 12 mm) could be placed. The box contained three disks of uranium, the middle one of which served as a measuring sample. The sample was irradiated in the same way as earlier described and its activity measured. This activity was compared to the activity from samples irradiated in the absence of fission sources and D_2O in the channel. All samples from these runs had the same diameter as the reactor fuel (29 mm). Gold and molybdenum were used as monitors. The gold foils were placed inside the cadmium tubes on the cylindrical surface of the sample and the molybdenum foils were placed horizontally between two of the uranium disks.

2. Studies of various effects

a) Monitoring

As mentioned above gold and molybdenum were originally used for monitoring purposes. The main resonances for the two substances lie around 5 eV and 500 eV respectively.

The value of δ with gold as a monitor was found to be 1.070 ± 0.015. When molybdenum was used the difference should be smaller as the flux increases more strongly with E close to the fuel rod than at the cell boundary. δ then came out as 1.020 ± 0.015.

The closeness of the main resonance in gold and the lowest resonance in uranium (6.7 eV), however, introduces a systematic error which must be accounted for. In the neighbourhood of the gold resonance the small local depression in the neutron current entering the rods (discussed earlier in section II) is different for the rod surrounded by air than for that directly surrounded by heavy water. In addition, fairly thick gold foils were used for activity reasons. Self screening was therefore considerable and could have been

different for the two runs because of the somewhat different neutron angular distribution. The two effects mentioned were studied by comparing the activities from gold and indium monitors. For indium with the main resonance at 1.46 eV the depression effect is negligible as the neutrons which pass through the uranium rod and return to the indium foil must make a head-on or nearly head-on collision with oxygen or deuterium nuclei. For oxygen the energy loss is too small to move the dip into the indium resonance and for deuterium it is too large. Furthermore the short half life of In^{116} (54^{m}) allows the use of very thin detectors, actually a lead indium alloy was used in foils 0.1 mm thick and containing 0.1 weight per cent of indium. For foils of such a thickness the small difference in neutron angular distribution is of negligible importance.

The correction factor for δ obtained from the comparison between the gold and indium activities was 0.98 ± 0.01. The value 1.070 ± 0.015 for δ has therefore to be reduced to 1.050 ± 0.018.

b) Effect of the cadmium

The depression of the fission sources inside and near to the cadmium box is believed to have very little effect for the absorption in the resonance region. The absorption of very high energy neutrons on the other hand will be considerably reduced. As this absorption is generally included in ϵ, the reduction mentioned is unimportant for our measurement. To illustrate the influence of the size of the cadmium a 20 mm high cadmium box was substituted for the smaller one. A value of 1.060 ± 0.015 was obtained for δ instead of the original 1.070 ± 0.015 (gold monitored values). Although the limits of error overlap the decrease in δ is probably real and is caused by the still more pronounced depression of the fission sources.

c) Capture above 100 keV

The capture above 100 keV of neutrons which originates in the rod and which have made no collision outside the rod is usually included in the fast fission factor ϵ. The contribu-

tion to the measured activity from such capture must therefore be subtracted before a proper value can be obtained for δ. For the rod completely covered with cadmium the high energy contribution is small and has been neglected. For the sample inside the fuel rod it was estimated in the following way. Catcher foils of aluminium were placed between the uranium disks in the small cadmium box as well as between similar disks in the rod completely covered with cadmium. The assemblies were irradiated in the usual way and the catcher foil activities were measured. Suitably placed dummy foils of aluminium made a subtraction of induced activities in the aluminium possible. With no fission sources in the sample rod the fission products on the catcher foils arise almost exclusively from U^{235} fissions. With fission sources the U^{235} fission contribution is almost the same, but in addition there is a certain contribution of U^{238} fissions. The activities of the catcher foils were measured in β-counters earlier used by Nylund for ϵ measurements (12). Nylund has made a relative calibration of the efficiency of the equipment in detecting U^{235} and U^{238} fissions. By using this calibration we found that the ratio of U^{238} to U^{235} fissions was 0.47 ± 0.05 for the disk in the small box. With the aid of this value, together with the effective fission and capture cross sections for U^{238} in a fission spectrum, and the epicadmium fission and capture cross sections in a 1/E flux for U^{235} and U^{238} respectively, the capture in U^{238} above 100 keV can be estimated relative to that below that energy. We find a value of (2 ± 0.5) %. Evidently, the capture of neutrons slowed down below the U^{238} fission threshold by inelastic scattering but with their energies still above 100 keV is not included in the figure given. We cannot measure this contribution separately but an estimate of the effect is obtained from the calculations of ϵ by Carlvik and Pershagen (13). For a 29 mm diam. rod the increase in neutron capture above 100 keV is about 30 % because of inelastically scattered neutrons. For our samples the effect must be smaller because of the cadmium. The ac-

tual high energy contribution should therefore be somewhere between 2 and 2.5 % of the total epicadmium absorption.

d) 1/v part

The form of the neutron spectrum for the two cases is very similar in the energy region of importance for the 1/v absorption. For both runs the 1/v part constitutes about the same fraction of the total epicadmium absorption and is approximately obtained from the values of the 1/v cross section and the resonance integral.

3. Results

After subtration of the 1/v and the high energy capture our final value for the ratio of the resonance absorption of a fuel rod in a lattice position in R1 and that in a cadmium-covered rod at an approximate cell boundary becomes 1.030 ± 0.020. If one refers to a 1/E flux instead of the cell boundary flux the corresponding ratio approximately becomes $\gamma\ 1.030 = 1.070 \pm 0.025$. The values given may serve as examples, and it should be pointed out they depend on the reactor lattice as well as on the neutron energy at which the normalization has been made.

Acknowledgement

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