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Studies of the Effective Total and Resonance Absorption Cross Sections for Zircaloy 2 and Zirconium

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STUDIES OF THE EFFECTIVE TOTAL AND RESONANCE ABSORPTION CROSS SECTIONS FOR ZIRCALOY 2 AND ZIRCONIUM.

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Summary.

Using pile oscillator technique, the total absorption cross section for zircaloy 2 plates has been determined in the neutron spectrum of the reactor R1. The plate thickness was varied in six steps from 0.2 mm to 6.4 mm. The thermal cross section for the alloy was calculated from cross section data and the known composition of the alloy. By subtracting this value from the measured cross sections and dividing by the factor $\alpha = \frac{2}{\sqrt{\pi}} r \sqrt{\frac{T}{T_0}}$ the effective resonance integrals were obtained. After subtraction of a constant amount for resonance contributions from hafnium, tin etc., effective resonance integrals for zirconium could be evaluated. An extrapolated value of 0.85 ± 0.15 b was obtained for the infinitely dilute integral (1/v part excluded). The ratio of the resonance integral at plate thicknesses 0.2 and 6.4 mm came out as 1.65 ± 0.25.

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1. Introduction.

Zirconium and zircaloy, i.e. zirconium alloys containing small amounts of other metals (Sn, Fe, Cr etc.), have found wide applications in reactor technology. With a few per cent zirconium added to the uranium metal in fuel elements the stability of the elements is much improved. Higher burn up of the fuel may be attained in such a way. In addition, thin layers of zirconium may be used as bonding material between the fuel and the canning to improve the heat transfer. Zircaloy on the other hand is widely used as canning material, especially in reactors fuelled with natural or slightly enriched uranium, because of its high resistance against corrosion and low neutron absorption cross section etc. On account of its good mechanical properties, zircaloy is also used for pressure tubes in reactors using such tubes around the fuel and cooling medium.

The thickness of the zirconium or zircaloy for these different applications varies from a few tenths of a millimeter in bonds to 5-10 mm in pressure tubes. For reactor calculations it is therefore desirable to know effective neutron absorption cross sections for the range of thicknesses mentioned above. The thermal neutron cross section for these materials is low (0.18 - 0.21 b). No appreciable thermal neutron flux depression will therefore occur even for the largest thicknesses. The resonance absorption, on the other hand, may be expected to vary with the thickness of the material. Due to the lack of detailed knowledge of the zirconium resonances, calculations of the effective resonance integrals cannot be performed. A series of experiments have therefore been undertaken to study the dependence of the zirconium resonance integral on the thickness of the material. Earlier measurements will be discussed at the end of the paper.

2. Method of measurement.

Of the five zirconium isotopes, Zr^{91} has the largest resonance absorption. However, neutron capture in Zr^{91} leads to the stable Zr^{92} isotope. It is therefore not possible to measure the resonance integral of natural zirconium by an activation technique. The pile oscillator method was chosen for the measurement.

The oscillations were made in the central channel of the heavy water - natural uranium reactor R1. The experimental arrangements are illustrated in Figs. 1 a and b. The samples were oscillated between the center of the reactor and a point about 80 cm above the D_2O level. To minimize the effect of the neutrons scattered out of the channel a large piece of graphite was placed below the sample "in" position. Above the center, the effective width of the channel (diam. 155 mm) was reduced with the aid of a graphite tube having an inner diameter of 60 mm.

The time function for the oscillation was an approximate square wave with a 40 sec period. The time for the samples to move from the "in" position to the "out" position or vice versa was about 1.8 sec.

The measurements could in principle be made in two ways. By performing the oscillations completely inside a long cadmium cylinder the epithermal neutron absorption could be studied apart from the thermal absorption. The great advantage of this method is that one is independent of the thermal cross section of the samples and does not require to know the ratio of the epithermal flux to the thermal flux at the "in" position. On the other hand there are several severe disadvantages with performing the oscillations inside a cadmium tube. The large amount of cadmium may seriously disturb the neutron spectrum. The absorption signal becomes quite small. Corrections for slowing down of neutrons by the samples below the cadmium cut-off are important and difficult to evaluate. The use of a 1/v absorber (boron) as a cross section standard is unsatisfactory because of the uncertainty of the true cadmium cut-off energy. A resonance absorber (gold) may of course be used as a standard. However, to obtain large enough signals, "infinitely" thin samples cannot be used. The effective resonance integral for the sample used must therefore be determined by a special measurement.

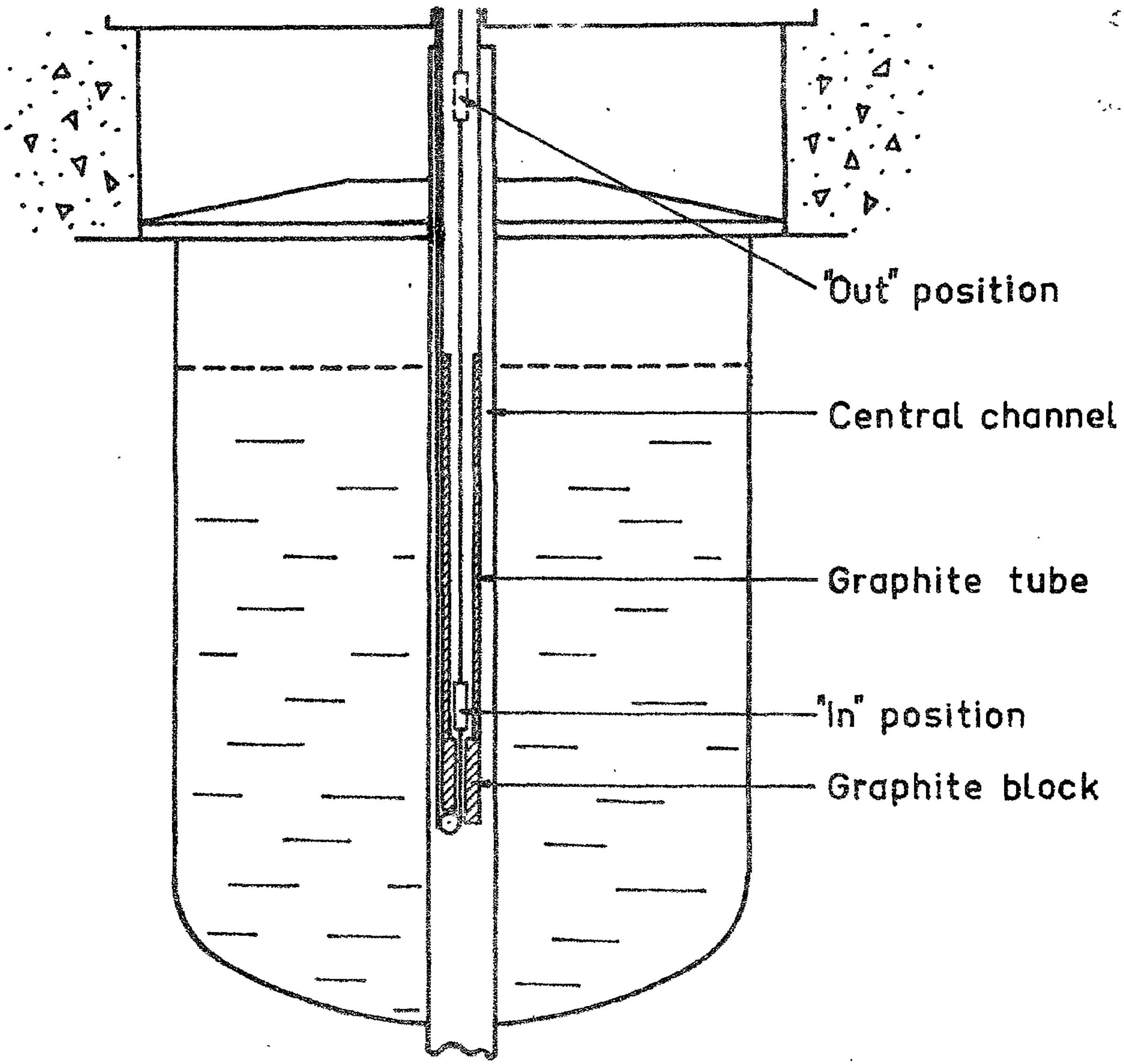
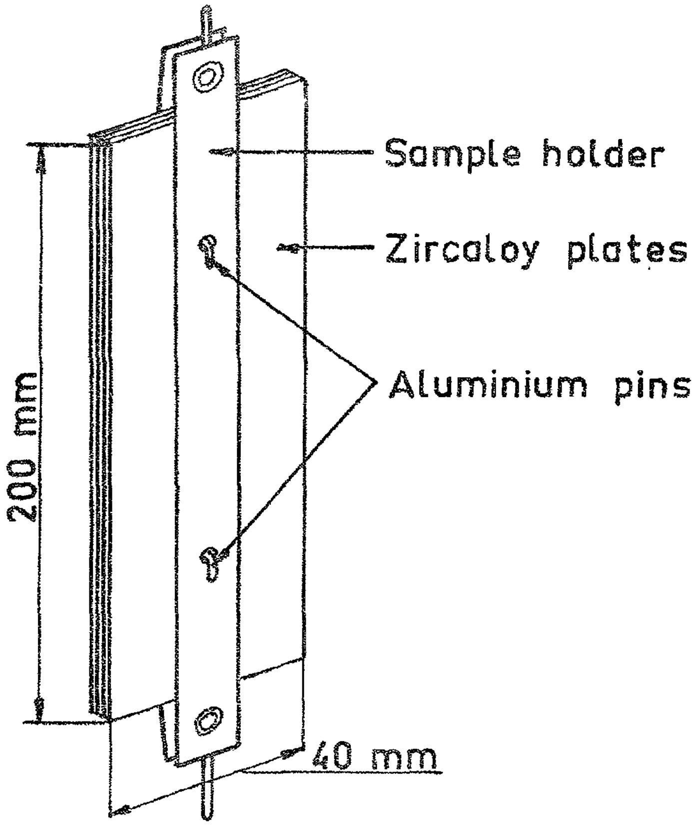


Fig. 1 a) Arrangement for the oscillation in the central channel.



b) Sample holder

Because of the disadvantages connected with the use of cadmium we chose to measure the absorption cross section of our samples in the total reactor spectrum. The resonance integrals were then determined by subtracting the thermal absorption cross section from the measured cross section. Boron was used as the cross section standard.

3. Apparatus.

The block diagram of the electronic parts of the pile oscillator equipment is shown in Fig. 2. The current from the ionization chamber consists of a d-c component, i_o, corresponding to the mean reactor power and a small alternating current, i, caused by the oscillations. The current passes through a resistance, R, in series with a backing off voltage. This latter voltage is adjusted to cancel the d-c voltage Ri_o. Thus only the a-c component is amplified in the succeeding amplifier and fed to a sine-cosine potentiometer. The output from the potentiometer gives the product of the amplified voltage F(t) and sin ωt or cos ωt. This product is then integrated in the velodyne integrator. By an appropriate mechanical system the periods of the oscillations and of the resistance variation of the potentiometer are made identical. Integration over a whole number of periods therefore yields the coefficients for the fundamental mode in a Fourier expansion of F(t). For small perturbations these coefficients are proportional to the effective absorption cross section of the samples and may serve as a relative measure of this quantity.

The principle of the measuring system was first used at the critical facility Zeep in Canada (1) and at the graphite reactor Gleep at Harwell (2). Although our system is closely similar to the English unit it has been described in some detail to facilitate the description of some special studies discussed later on.

For an investigation of the present kind good linearity of the equipment is required. The linearity was tested with the aid of a square wave generator as well as with signals from an RC circuit, both types of signals being synchronized with the frequency of the sine-cosine potentiometer. When plotted against the test signal amplitude, the integrated signals for moderate and large input signals fell on a straight line. But extrapolated towards zero input

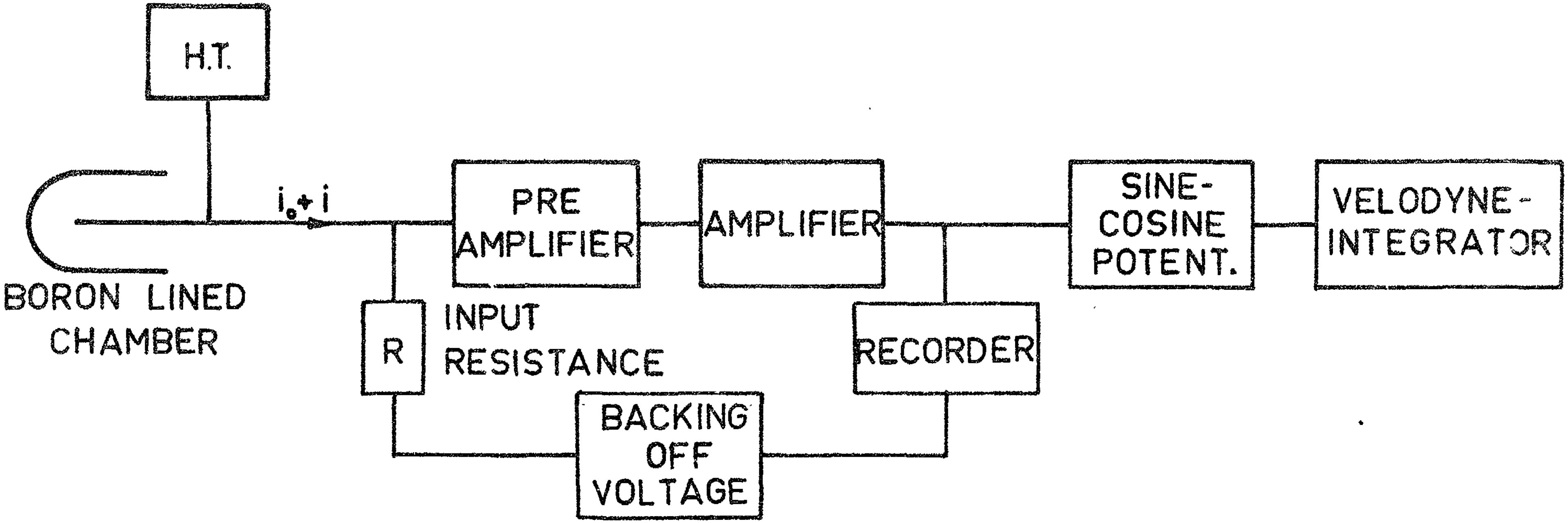


Fig. 2 Block diagram of the electronic parts.

signal the line did not pass through the origin. The deficiency of the apparatus for small signals may arise from friction in the velodyne motor. It is believed that most of the consequences of the lack of linearity were avoided by following a particular procedure of measurement. A thorough additional test of.the whole equipment is described under 5 e.

4. Experimental procedure.

No zirconium with well enough known purity was available and therefore all measurements were performed with zircaloy-2. The samples were in the form of plates, 200 mm long and 40 mm wide. The following plate thicknesses were used 0.2, 0.4, 0.8, 1.6, 3.2 and 6.4 mm. Originally measurements were also made on plates 0.1 mm thick, but the signals obtained were too small to give reasonable limits of error. The plates were fixed to the sample holder in the way shown in Fig. 1 b.

Boron was used as the cross section standard. A water solution of boric acid was pipetted on to a filter paper of about the same length as the zircaloy plates and with a width of about 22 mm. The borated filter paper was wrapped in a thin aluminium foil. An identical dummy element containing no boron was also made.

The concentration of boron in the solution was determined in two ways. Firstly the solution was prepared by dissolving a weighed amount of boric acid in water to a known volume. Secondly, the concentration of the solution was checked by titration with sodium hydroxide in the presence of mannitol and with bromothymol blue as indicator.

The boric acid was taken from a stock of known B^{10} content. The absorption cross section for this boron was 758 ± 4 b (3). The amount of boron in each standard was 3.33 ± 0.02 mg or about 0.08 mg/cm^2. For a homogeneous distribution this corresponds to a macroscopic cross section of 0.003 $cm^2/_{cm}2$. To check that the boron was not lumped together in certain spots the filter paper was closely examined with the aid of a microscope. Despite the use of a magnification of 100 times, no boric acid crystals could be seen. The inhomogeneous structure of the filter paper apparently impedes the growth of crystals.

The distribution of boron along the paper was checked by cutting a sample into eight equal pieces. The boron content of each was determined by titration. The maximum deviation from the mean value was 20 %. Considering the small variation of the neutron flux along the samples (compare below) such a variation of the boron distribution was tolerable.

A correction factor for self screening in the samples was obtained from Bothe´s formula for monoenergetic neutrons (4) (compare also (5)), which, in an approximate form, was weighted over a maxwellian spectrum. The factor used was 0.993 ± 0.005.

The sample holder was at first made as light as possible to minimize the signal from the empty holder. Because of the lack of linearity of the apparatus for small signals, the holder was later loaded with a small piece of iron to increase the signal from the holder. The sample signals were thereby obtained as the difference between two reasonably large signals. This procedure made the resulting sample signals slightly more uncertain but systematic errors due to lack of linearity were decreased.

The pile power during oscillations was 1 kW. To decrease the effect of power drifts the signal from the amplifier was multiplied by weighting factors in the way described by Schmid (6) and Persson (7). The number of periods for the integration was three, and thus drifts up to the fourth power in t (t = time) were eliminated.

5. Special studies.

5 a. Neutron flux distribution.

The neutron flux distribution at the sample position was studied by activating bare and cadmium covered gold foils. The result of the activations is given in Fig. 3. As may be seen, the epithermal and thermal fluxes are proportional and vary only slightly along the length of the samples.

5 b. Scattering and moderation.

Scattering events in the samples influence the oscillator signal in two ways. Firstly, despite the fact that the "in" position is at zero flux gradient, some neutrons are lost through scattering out of the channel. This effect concerns neutrons of all energies although the largest contributions comes from scattered thermal neutrons. Secondly, epithermal neutrons scattered in the samples lose energy. The

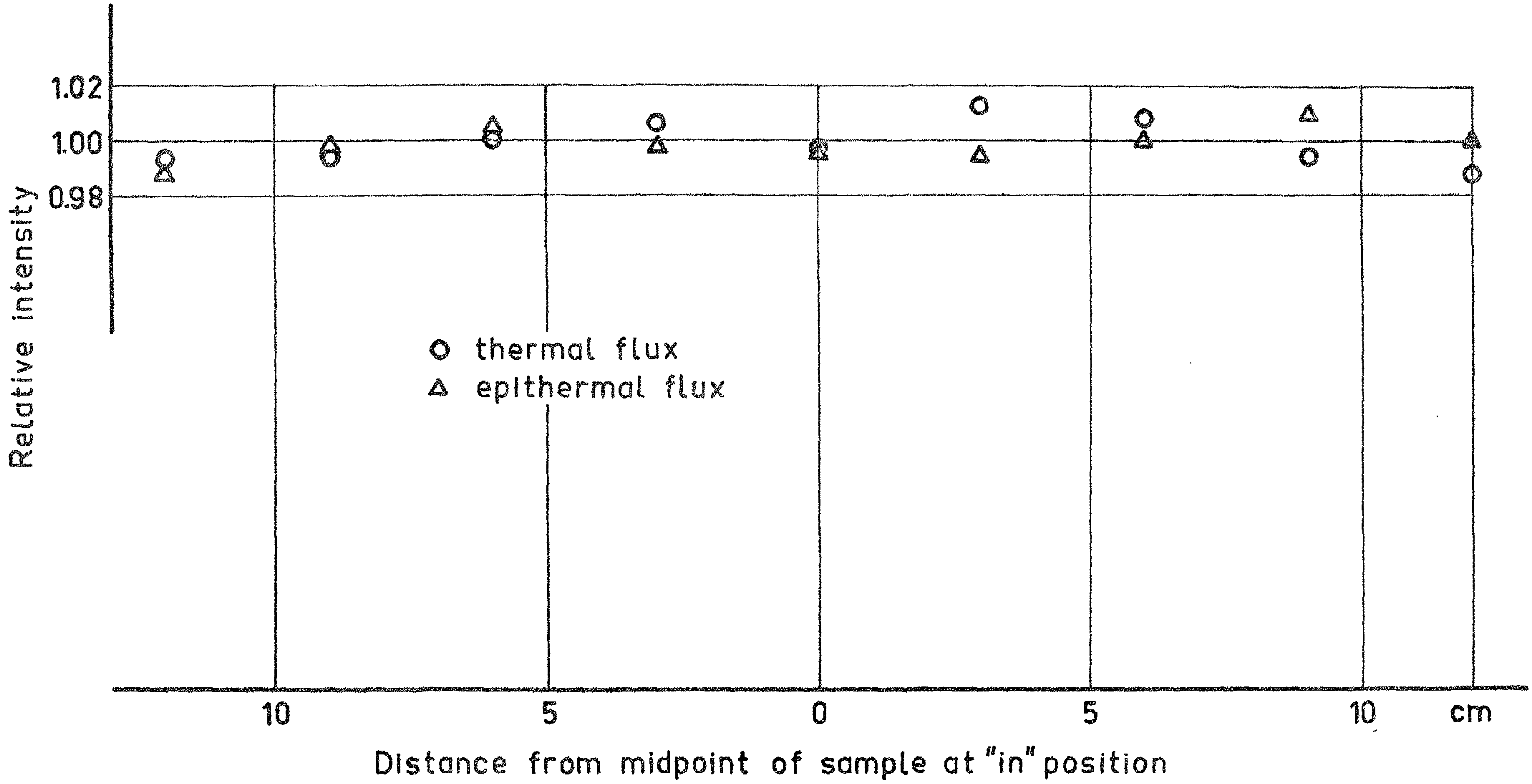


Fig. 3 The thermal and epithermal neutron flux distributions at the sample "in" position.

resonance escape for these neutrons is enhanced and the effect acts as a negative absorption.

The magnitude of the two effects was studied with the aid of several materials with low absorption cross sections. As can easily be verified, the oscillator signal may be written in the following way (the non 1/v correction factor g (8) for the sample is assumed to be equal to 1)

$$\frac{1}{k_a} S = \sigma_o + \alpha RI + \frac{k_s}{k_a} \sigma_{st} - \frac{k_m}{k_a} \xi \sigma_{se} - \sigma_N \quad \ldots\ldots\ldots\ldots\ldots \quad (1)$$

S	is the oscillator signal per atom of sample.
$\frac{1}{k_a}$	is a proportionality factor transforming the oscillator signal into barns.
σ_o	is the 2200 m/s absorption cross section.
α	is a measure of the ratio of the epithermal to total neutron densities. With Westcott's nomenclature it is equal to $\frac{2}{\sqrt{\pi}} \, r \sqrt{\frac{T}{T_o}}$ (8).
RI	is the effective resonance integral (1/v part excluded).
$\frac{k_s}{k_a}$ and $\frac{k_m}{k_a}$	are proportionality constants transforming σ_{st} and $\xi \, \sigma_{se}$ into effective absorption cross sections.
ξ	is the average logarithmic energy decrement per scattering collision.
σ_{st} and σ_{se}	are the scattering cross section for thermal and epithermal neutrons respectively.
σ_N	is a fictitious cross section per sample atom, representing the effect of nitrogen-displacement by the samples.

$1/k_a$ was determined from the boron runs for which Eq. 1 may with good accuracy be written $\frac{1}{k_a} S = \sigma_{o\ eff}$. $\sigma_{o\ eff}$ is the 2200 m/s absorption cross section corrected for self screening. Thus $\frac{k_s}{k_a}$ and $\frac{k_m}{k_a}$ could be obtained by oscillating samples with known absorption and scattering properties and, preferably, with large differences in the ξ-values.

Lead, bismuth, heavy water and graphite were chosen for this investigation. The lead and bismuth samples were of "guaranteed reagent" quality. The D_2O was of reactor grade quality containing 99.5 weight per cent of D_2O[1]. The graphite came from a stock intended for use as reflector material for the R0 reactor. The diffusion length for this graphite had earlier been determined as 52.8 cm (C. Wikdahl, private communication). A separate boron analysis[1] of an oscillated sample showed a boron content of 1.5 ppm. An effective value of the 2200 m/s graphite absorption cross section of 4.5 ± 0.5 mb was derived from these quantities. Cross section data for lead and bismuth were obtained from (9) and (10). The following values of k_s/k_a and k_m/k_a were obtained

$$k_s/k_a = (0.50 \pm 0.17)\ 10^{-3}$$

$$k_m/k_a = (5.0 \pm 0.8) \times 10^{-3}$$

Applied to zirconium, the net effect of scattering and moderation is a slight increase of 3.3 ± 1.5 mb in the absorption cross section. It is true that the effect varies somewhat with plate thickness owing to multiple scattering, but in view of the large uncertainty and the small size of the total effect the same correction has been applied for all sample thicknesses.

The effect of the displacement of nitrogen by the samples (σ_N in Eq. 1) has been calculated using a macroscopic absorption cross section of $7.5 \cdot 10^{-5}$ cm^{-1} for nitrogen (9) (air temperature 25°C and pressure 760 mm Hg).

5 c. Determination of α.

The evaluation of the effective resonance integrals requires a knowledge of the factor α in Eq. 1. α has been determined from the cadmium ratio, R_{Cd}, of gold, assuming the thermal and resonance absorption cross sections for gold to be known. The following relation is valid

$$\alpha = \frac{g}{g\, R_{Cd} \sqrt{\frac{4\, E_o}{E_{Cd}}} + \left(R_{Cd} - 1\right) \frac{RI}{\sigma_o}} \quad \ldots\ldots\ (2)$$

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[1] The authors are indebted to C. Evertson for help with the D_2O analysis and to G. Tydén for the determination of the content of boron and other impurities.

g is the non $1/v$ correction factor (8).
E_o is the energy corresponding to a neutron velocity of 2200 m/s.
E_{Cd} is the cadmium cut-off energy.

R_{Cd} was determined from a lead-gold alloy containing about 100 µg of gold per cm^2. Measurements were made with two different foil diameters, 25 mm and 10 mm, with a minimum of cadmium around the foils for the cadmium runs. Cadmium thickness was 0.5 mm. The R_{Cd} values obtained with the two foil sizes agreed within the limits of error (less than 1 %). It was therefore concluded that the disturbance of the epithermal flux by the cadmium was negligibly small.

The cadmium ratio was determined as 2.290 ± 0.020. With a cadmium cut-off energy of 0.45 ± 0.05 eV and the following constants for gold $g = 1.005$ (8), $\sigma_o = 98.8 \pm 0.3$ b (9) and RI = 1480 ± 35 b (obtained from a dilute integral value of 1500 ± 35 b (11, 12) and a self screening correction of 20 b) one obtains

$$\alpha = 0.0492 \pm 0.015.$$

5 d. Scattering effects during sample movement.

During the transition from the "in" position to the "out" position or vice versa tha samples pass through a region of neutron flux distribution with non zero gradient. Neutron scattering in the samples therefore causes a positive reactivity signal for each transition. This signal has a frequency twice that of the oscillator signal. Under ideal conditions it should therefore not contribute to the output signal from the pile oscillator. However, the output from the sine-cosine potentiometer may contain a small first harmonic besides the fundamental. The integral of the product of this first harmonic and the scattering signal would not be zero. In addition, although the time for the transition of the samples in the two directions is the same, their speed is not exactly the same close to a rest position. Thus the reactivity effect may be slightly different for samples moving out of the reactor and for samples moving into the reactor.

To get an idea of the magnitude of these possible effects the following studies were made. A boron and a zircaloy sample were both oscillated at two different periods, 40 sec and 60 sec, but with the same transition time for all runs. For boron the absorption cross section is much larger than the scattering cross section, for zircaloy the opposite is true. Thus if the scattering effect during the sample movement causes an effect outside the estimated limits of error, the ratio of the boron signals for the two periods would differ from the same ratio for the zircaloy sample. This was not the case.

As an additional check, the phase angle between the mechanical oscillations and the reactor response as seen at the amplifier output was determined by alternatively integrating the product $F(t) \sin \omega t$ and $F(t) \cos \omega t$ and taking the ratio between the two integrals. This was done for several samples with different values of σ_s/σ_a . No significant difference in phase angle was noted.

5 e. Test of apparatus and method with gold foils.

The preliminary results of the zircaloy measurements showed a considerably smaller variation of the resonance integral with plate thickness than had earlier been estimated by approximate calculations. (R.M. Pearce, private communication.) A need for a thorough test of the measuring equipment was therefore felt. Such a test was made by comparing the effective resonance integrals for gold foils measured with two different methods in the following way.

Firstly, gold foils of four different thicknesses, about 0.005, 0.009, 0.027 and 0.10 mm, were oscillated in the reactor and their effective absorption cross sections were determined relative to boron. All foils were of the same size and so chosen that the signal from the thinnest foil was of the same order as the smallest signal during the zircaloy measurements.

For the used foil thicknesses, self screening effects for thermal neutrons were not negligible. The necessary correction factors were obtained in the same way as for the boron samples. With the aid of these correction factors and the known g factor and 2200 m/s absorption cross section for gold, the effective resonance integrals for the four thicknesses could be evaluated from Eq. 1.

Secondly, the cadmium ratios for the four thicknesses were determined at the oscillator sample "in" position in the reactor. Before counting, the foils were dissolved in nitromuriatic acid. The reason for this step was twofold. The thinnest sample consisted of several small pieces, which were used both for the oscillations and the succeeding activation. The pieces did not have a uniform size and could not easily be counted with the same counter efficiency. For the thickest foils, the different distribution of the induced activity in the bare and cadmium covered foils could have caused a small systematic error if the measurements had been made directly on the foils. By dissolving them, this problem was avoided. The intensity of the 411 keV γ line was measured in a γ spectrometer.

From the measured cadmium ratios, R_{Cd}, the effective resonance integrals were determined from the expression

$$RI_{eff} = \frac{\sigma_{o\,eff}}{R_{Cd}-1} \left[\frac{g}{\alpha} - \sqrt{\frac{4\,E_o}{E_{Cd}}}\; R_{Cd} \right] \ldots\ldots\ldots\ldots\ldots \quad (2)$$

where $\sigma_{o\,eff}$ is the thermal cross section corrected for self screening. The other constants are defined in connection with Eqs. 1 and 2.

The result of the measurement is summarized in Table I and in Fig. 4.

Table I.

Comparison between results from pile oscillator and cadmium ratio measurements on gold.

Approximate tickness, mm	RI_{eff}	
	Pile osc.	Cd-ratio
0.005	900 ± 65	915 ± 40
0.009	820 ± 45	790 ± 35
0.027	540 ± 40	510 ± 20
0.10	310 ± 40	295 ± 12

The values of the resonance integral from the oscillator measurement are, especially for the thick foils, very sensitive to the correction factors applied to the thermal cross section. As these factors are not too accurately known the errors in RI become large.

To facilitate a comparison between the two results, each of the two sets of points in Fig. 4 have been joined with a curve. As may be seen from the figure, the agreement between the resonance integrals determined in the two ways is good. Especially, it is unlikely that any large systematic difference exists between the way in which the two sets of resonance integrals vary with foil thickness. Thus, the result of the test raises our confidence in the zircaloy results. However, it should be pointed out that for zircaloy the large ratio of scattering to absorption (about 30 compared to about 0.1 for gold) may cause un-

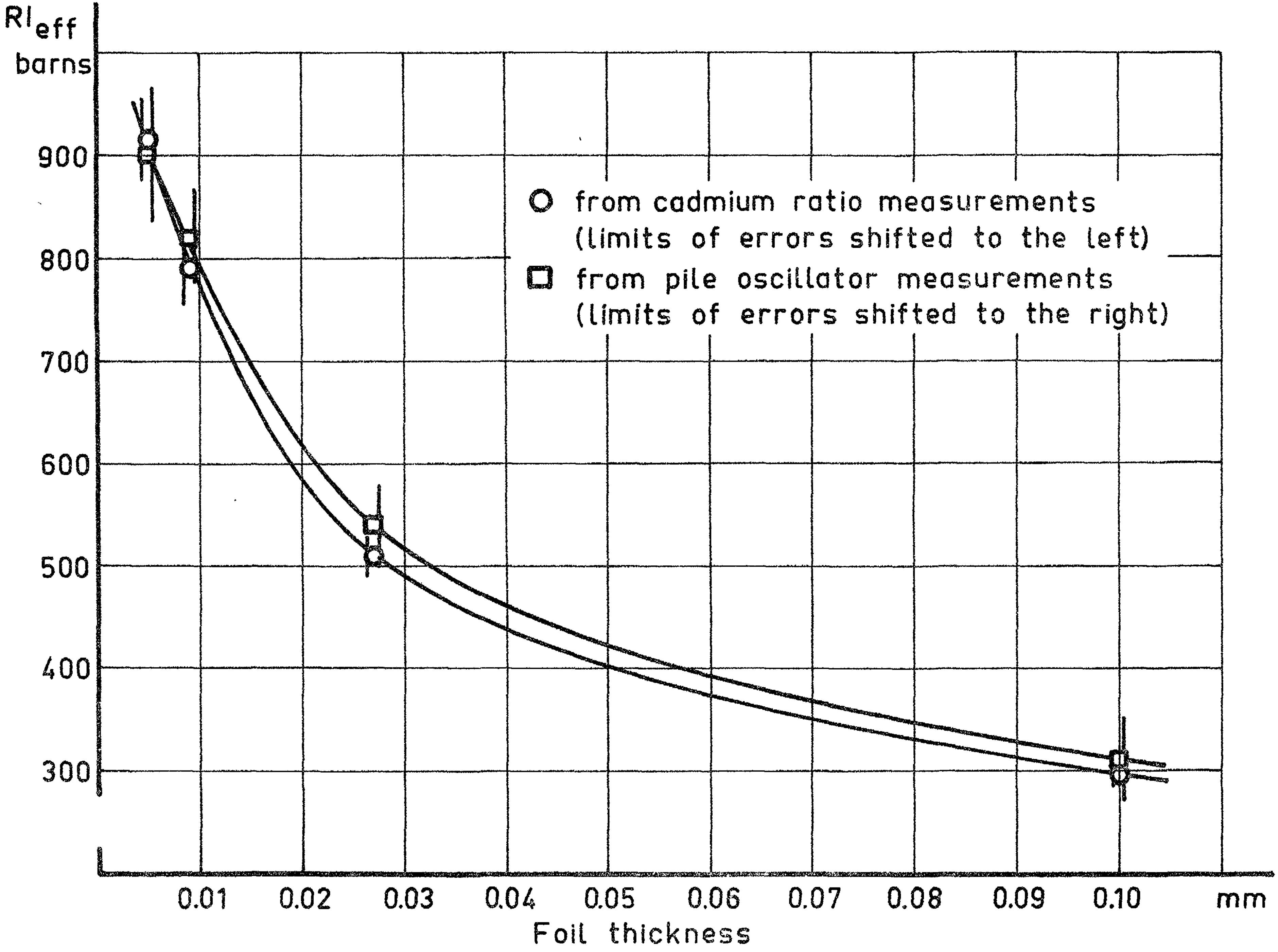

Fig. 4 The effective resonance integral for gold determined with pile oscillator and cadmium ratio methods.

known complications making the gold test less significant.

5 f. Energy dependence of the neutron flux.

According to the measurements by E. Johansson et al (13 and private communication) the epithermal neutron flux follows the 1/E distribution rather well to at least 10 keV. It is true that resonance capture in the uranium fuel causes a small gradual decrease in the neutron flux per unit lethargy with decreasing energy. However, as the reactor response to a perturbation is proportional to the product of the real and adjoint fluxes and these deviate from 1/E in opposite directions, the effect of the small deviation from 1/E is negligible.

Above 10 keV no experimental information exists about the spectrum. Approximate age theory calculations show that there should be no serious deviations from 1/E below at least 100 keV.

6. Results and discussion.

6 a. Evaluation.

With the aid of the results from the boron runs and with due consideration to the effects discussed in section 5, Eq. 1 has been used to obtain the effective cross sections, $\sigma_{eff} = \sigma_o + \alpha \mathrm{RI}$, for the different zircaloy 2 plates. From these cross sections, the resonance integrals may be obtained, if σ_o is known. This quantity was calculated from the known composition of the zircaloy samples. For the melt from which our plates had been made, the manufacturer of the zircaloy (Imperical Chemical Industries, Ltd, England) furnished the analysis given in Tabel II.

Table II.

Chemical composition of the zircaloy 2.

Element	Weight percent	Δ %[2]	Element	Ppm[1]	Δ ppm[2]
Zr			Al	60	
Sn	1.42	± 0.03	Cd	< 0.2	
Fe	0.11	± 0.005	C	100	
Cr	0.09	± 0.005	Hf	65	± 15
Ni	0.04	± 0.002	Pb	< 20	
			Mn	< 15	
			N_2	70	
			Si	< 25	
			Ti	35	
			B	0.3	± 0.05
			Co	< 10	
			Cu	60	
			H_2	80	
			Mg	< 10	
			Mo	< 30	
			0_2	1700	
			W	< 20	
			V	< 30	

1) Parts per million (weight)

2) Uncertainty as given by the manufacturer

The predominant contribution to the resulting thermal cross section comes from zirconium. Unfortunately, there is some uncertainty about what value to use for this material. The old value of 180 ± 4 m b given in BNL-325 Sec. ed. has in Supplement No. 1 been raised to 185 ± 4 m b (9). The new reference is to pile oscillator work at Harwell yielding a value of σ_o = 191 ± 4 m b (14). However, this value may be somewhat high for two reasons. Firstly, scattering effects have been neglected, although measurements on graphite reported in the relevant paper indicate that the effect is not quite negligible. Secondly, the hafnium content was not known to better than "less than 100 ppm". 100 ppm of hafnium corresponds to a cross section of about 5 m b and the result could therefore be too high by that amount. Instead of the value 185 ± 4 m b recommended in the BNL-325 Supplement we have therefore chosen a value of 183 ± 4 m b for the Zr 2200 m/s absorption cross section. The resulting cross section for zircaloy 2 thereby becomes 209 ± 5 m b. This cross section has been used for plate thicknesses less than 0.8 mm. For thicker samples a minor correction for self screening must be applied. The correction factor, η , was measured with the aid of very thin copper foils for the 6.4 mm thick plate (actually 8 plates 0.8 mm thick) and found to be 0.993, i.e. the effective 2200 m/s cross section was for this thickness reduced to 0.993 σ_o .

As η was close to unity even for the thickest sample, the correction for other thicknesses was obtained by assuming a linear variation of $1-\eta$ with thickness.

The results of the measurements are summarized in Table III. To obtain the values of the zirconium resonance integrals a correction for resonance absorption in hafnium, tin etc. must be performed. With the aid of the chemical analysis of the alloy and cross sectional data from (10) and (11) a value of 0.165 b has been estimated for the non-zirconium contribution to the measured resonance integrals.

The effective corss section in the reactor spectrum has been plotted as a function of plate thickness, d , in Fig. 5. Fig. 6 gives the zirconium resonance integral as a function of effective plate thickness, d_{eff} . d_{eff} is the thickness of an infinite plate giving the same surface to mass value, S/M, as the finite plates. This representation is preferred when comparing the measured values with calculations.

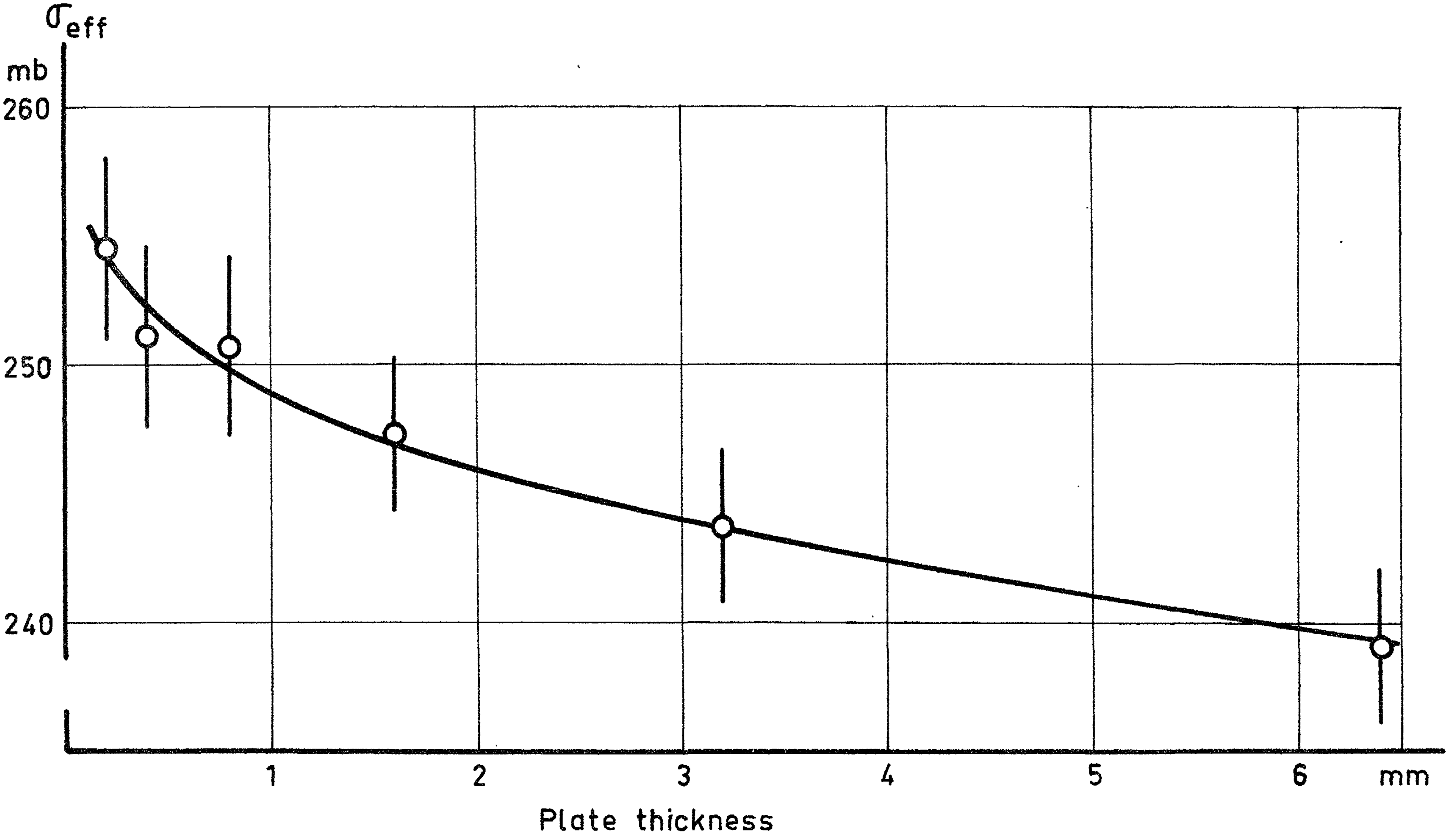

Fig. 5 The effective absorption cross section of zircaloy 2 in the R1 central flux.

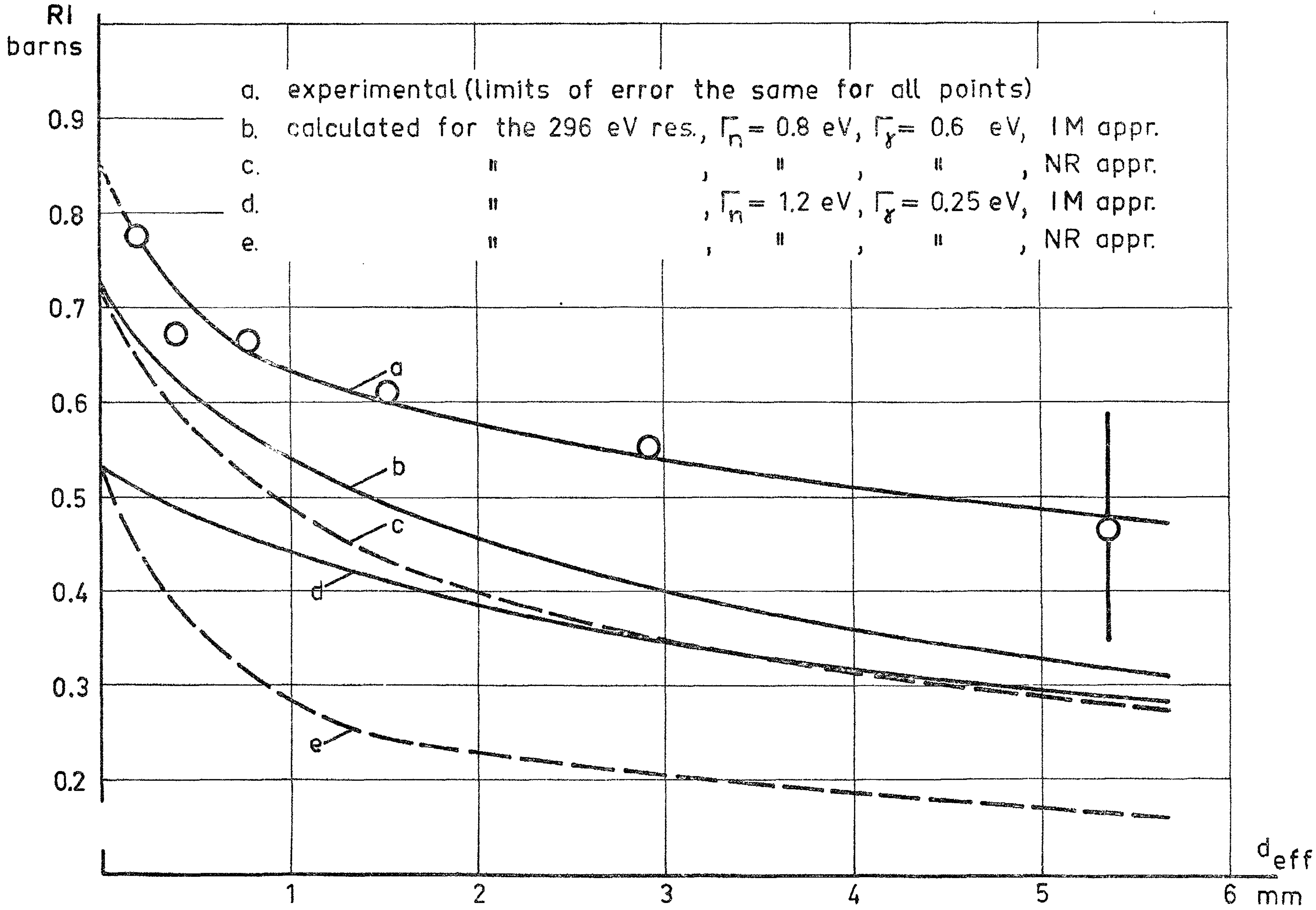

Fig. 6. The effective resonance integral for zirconium,
a. experimental, b.-e. calculated for the 296 eV resonance.

Table III.

Effective cross section for zircaloy-2 and
resonance integrals for zirconium for
different plate thicknesses.

Plate thicknesses mm	Effective thickness	σ_{eff}, mb	RI b
0.2	0.2	256 ± 3.5	0.78 ± 0.12
0.4	0.4	251 ± 3.5	0.67 ± 0.12
0.8	0.8	250 ± 3.5	0.67 ± 0.12
1.6	1.5	247 ± 3	0.61 ± 0.12
3.2	2.9	240 ± 3	0.55 ± 0.12
6.4	5.3	239 ± 3	0.47 ± 0.12

6 b. Errors.

The standard deviation of the effective cross sections for the different thicknesses is a combination of uncertainties in the mean value of the pile oscillator signal (0.5 - 1 %) and in the conversion of the oscillator signal into barns via the boron standard (about ± 1 %).

The resonance integral values are obtained from the effective absorption cross section by first subtracting the thermal cross section and contributions from scattering, moderation and nitrogen as described above and then dividing by the factor α. The combined error of the subtracted cross sections is about 5 mb. α is known to within ± 3 %. Combining all estimated errors results in a standard deviation of about ± 0.12 b for the zirconium resonance integrals. The absolute limits of error become almost equal for all dimensions.

6 c. Comparison with calculations.

A comparison between measured and calculated values is greatly hindered by the lack of knowledge of detailed data of the zirconium resonances. The only one, that has been at least partially resolved is the 296 eV resonance in Zr^{91}. For this resonance the values $\Gamma_n = 1.2$ eV and $\Gamma_\gamma = 0.25$ eV are given in (9). However, a recent measurement by P.P. Singh at Chalk River (private communication through J. Harvey) has yielded the values $\Gamma_n \approx 0.8$ eV and $\Gamma_\gamma \approx 0.6$ eV. The total width of the resonance therefore seems to be about 1.4 eV but the division into Γ_n and Γ_γ is uncertain.

The effective resonance integral for the 296 eV resonance has been calculated by the method used for the calculation of effective resonance integrals for uranium and thorium rods. For either set of Γ values the practical width of the resonance is about the same as the maximum energy loss of a neutron scattered by the absorber atom at resonance energy. The practical width is the energy interval in which the resonance cross section is larger than the potential scattering cross section. Thus, neither the narrow resonance (NR) approximation nor the infinite mass (IM) approximation (for the definition and meaning of these expression , compare for instance (15))can be expected to be a good approximation for the present case. One may, however, assume that calculations with the two different approximations will yield extreme values covering the true value. In Fig. 6, the results of the calculations for the 296 eV resonance are given together with the experimental points. The IM curves contain only the so called surface term, i.e. the absorption of neutrons which enter the plates with resonance energy. In reality, a small amount should be added for the contribution from neutrons slowed down into the resonance through scattering with zirconium atoms. It has been omitted because it is small. It may still be remarked that its inclusion in the calculations would flatten the IM curves somewhat as it is relatively more important for thick plates than for thin ones.

The effect of interference between resonance and potential scattering has been omitted in the calculations. For the IM approximation this is unimportant. For the NR approximations on the other hand, the interference will increase the absorption and somewhat decrease the variation of RI as a function of d. No effect has been made to estimate the magnitude of this latter effect as inclusion of interference in the calculations leads to quite complicated expressions.

Finally, it is evident from Fig. 6 that the 296 eV resonance alone cannot be responsible for the total resonance absorption in zirconium. A contribution from resonances at higher energies has to be added to the calculated curves. As generally self screening effects become smaller with increasing energy, the high energy contribution is less sensitive to the plate thickness d . With these different effects taken into account the apparent large difference in the relative variation of RI with d between, at least, the curves c and e and the experimental curve becomes smaller. The latter curve has been drawn by eye through the experimental points. As most of the indicated limits of error arises from uncertainties which affect all points in the same way, the relative variation of RI with d is better known than what could be assumed from the given errors. The ratio of RI for the thinnest and thickest plates (0.2 and 6.4 mm) is 1.65 ± 0.25.

From the form of the calculated curves for small d values a crude extrapolation of the experimental curve forwards zero plate thickness may be attempted. A value of 0.85 ± 0.15 b is in this way obtained for the infinitely dilute zirconium resonance integral.

6 d. Comparison with earlier measurements.

Table IV contains a summary of earlier measurements, which are known to the authors.

Table IV.

Results from earlier measurements.

Reference	Method	Sample	RI
1. Macklin & Pomerance (11)	?	?	3
2. Klimentov et al (16)	Pile period	?	3.6 ± 0.5
3. Dobrynin et al (17)	Beam experiment	Disc, 22 mm diam, thickness 10 mm	2.3 ± 0.5
4. Tattersall et al (10)	Pile oscillator	Plate 1.8 mm thick	0.60 ± 0.09
5. Feiner (18)	Pile oscillator	4 g ZrO_2, enriched to 88 % in Zr^{91}	> 0.5

In addition to the results in the table an unpublished measurement at Chalk River has yielded a value of 0.59 b for a tube, outer diam. 83 mm, thickness 4 mm (R.M. Pearce, private communication). Our results agree well with this value and that of Tattersall et al., but disagree far outside the limits of error with the first three in the table. The source of the value quoted by Macklin and Pomerance is not known and the value is probably quite inaccurate. The Russian measurements, numbers 2 and 3 in the table, are well described in the articles referred to and there are no obvious reason to question the results. A minor decrease from 3.6 to 3.3 b may be obtained for number 2 by using the more up-to-date value of 2800 b for the hafnium resonance integral instead of 1500 b used by Klimentov et al. However, for such a low absorbing material as zirconium it is much easier to construct an explanation for too high a value than for one too low. Many impurities (B, Cd, Hf etc.), even in small concentrations, raise the absorption appreciably, and if unknown cause erraneous integral values. Too low values may be obtained if moderation effects are underestimated, but for a lattice with a reasonably high resonance escape probability and low fast leakage factor such an error can never be large.

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