Neutron activation resonance integrals
of ^{64}Zn, ^{68}Zn, ^{85}Rb, ^{100}Mo, ^{102}Ru,
^{113}In, ^{123}Sb, and ^{180}Hf

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The ratio of resonance integral to thermal activation cross sections of the isotopes ^{64}Zn, ^{68}Zn, ^{85}Rb, ^{100}Mo, ^{102}Ru, ^{113}In, ^{123}Sb, and ^{180}Hf relative to gold has been determined by measuring cadmium ratios.

The standard gold or a secondary standard was mixed uniformly in the samples and the activities resolved with a lithium-drift germanium γ-ray spectrometer. Expressed as Westcott S_0 values the results were: ^{64}Zn $= 2.06 \pm 0.03$, ^{68}Zn $= 3.72 \pm 0.14$, ^{85}Rb $= 18.4 \pm 0.6$, ^{100}Mo $= 21.7 \pm 0.8$, ^{102}Ru $= 3.76 \pm 0.03$, ^{113}In $= 24.7 \pm 0.5$, ^{123}Sb $= 28.3 \pm 2.1$, ^{180}Hf $= 2.17 \pm 0.09$. All measurements are relative to $S_0 = 17.7$ for gold.

Introduction

This work is a continuation of that already published (Ricabarra *et al.* 1968) in which accurate intercomparison of activation resonance integral to thermal activation cross sections are measured by means of a lithium-drift germanium gamma-ray spectrometer.

The high resolution attainable with a Li-drifted Ge gamma-ray detector makes it possible, by activation analysis of the samples, to select an energy region which is free from contaminants and, because the samples were prepared with a solution including the standard, weight calibration and corrections due to irradiation position and counting geometry were eliminated.

The flux in the RA-1 reactor where the irradiations were made was large enough (2×10^{12} n/cm^2 s) to allow the use of very thin samples, with negligible self-shielding correction.

For short half-lives the measurement of the ratio of cadmium ratios was made over a period of time comparable to the half-lives involved, and the periods agreed with those currently found in the literature.

For half-lives longer than 10 days the measurement was made for a period of time usually less than the relevant half-life, but activities were checked to prove that they were decaying with the expected half-life.

All S_0 values were determined relative to gold, but for activities with half-lives of about an hour or less ^{115}In was used as a secondary standard.

For activities with half-lives longer than 10

days, the isotopes selected as secondary standards were ^{64}Zn and ^{74}Se, which by neutron capture produces ^{65}Zn (245 d) and ^{75}Se (120 d).

The cadmium ratios of ^{74}Se and ^{115}In relative to ^{197}Au were measured in our previous work, and that of ^{64}Zn was measured relative to gold in this work.

The experimental method and analysis have been described in detail previously (Ricabarra *et al.* 1968).

Briefly the method consists of preparing samples containing the same proportion of unknown element and standard from a solution.

The samples were prepared by soaking small disks of filter paper in the solution and drying them slowly on a watch glass. Pairs of samples were irradiated, uncovered and cadmium covered, in the internal graphite reflector of the RA-1 by a pneumatic rabbit system.

A gold–cadmium ratio measurement at the irradiation site, $R_{Cd}(\text{Au}) = 1.674 \pm 0.003$, gives an epithermal index $r\sqrt{(T/T_0)} = 0.0805 \pm 0.0002$.

The gamma-ray spectrum of the two samples was then measured in the spectrometer and the relative number of counts in the photopeak calculated.

The final experimental result was expressed in the Westcott formalism (Westcott *et al.* 1958):

$$S_0 = \frac{2}{\sqrt{\pi}} \frac{I'}{\sigma_0}$$

Where I' is the reduced resonance integral and σ_0 is the absorption cross section at energy $E_0 = 0.025$ eV.

In order to obtain S_0 the experimental results were corrected by G_r which is a factor for neutron epithermal self-shielding, F, which takes into account the transmission of cadmium for resonance neutrons; g is the ratio of the activation of the detector in a maxwellian flux to that of a $1/v$ detector having the same σ_0 and W, which is the difference of epithermal neutron activation and that of a $1/v$ detector, normalized to $\sigma_0 = 1$, below the cadmium cutoff.

The neutron spectrum at the irradiation site was calculated with multigroup diffusion theory (Boix and Solanilla 1967) and a correction that takes into account the deviation from the $1/E$ spectrum was applied to the resonance integral.

The errors assigned to experimental S_0 take into account the standard error of the average value of the ratio of cadmium ratios of the standard and unknown obtained from several irradiations; the systematic error introduced by resonance parameters of the standard is not included.

Many previous measurements of activation resonance integrals have been handicapped by systematic errors due to poor contaminant discrimination, self-shielding of epithermal neutrons in the samples, and uncertain description of epithermal neutron spectrum in the site of irradiation. These systematic errors have been carefully avoided by the present technique, as has been remarked above and described in our previous work.

Zinc

Cadmium ratios of ^{64}Zn and ^{68}Zn were both measured relative to gold.

^{64}Zn

Mixed samples containing 10 mg/cm^2 of zinc and 0.0003 mg/cm^2 of gold were irradiated, uncovered and cadmium covered, for 6 h. Six days later the 1.114 MeV gamma ray of ^{65}Zn was measured relative to the 411.8 keV gold gamma ray.

The averaged result from six irradiations gives

$$R_{Cd}(^{64}Zn)/R_{Cd}(^{197}Au) = 3.21 \pm 0.03$$

or

$$R_{Cd}(^{197}Au) = 1.674 \pm 0.003$$
$$R_{Cd}(^{64}Zn) = 5.37 \pm 0.05$$

Due to the importance of ^{64}Zn as a secondary standard, a measurement of the cadmium ratio for two different thicknesses of zinc foils was made

TABLE I

Neutron resonance self-shielding in ^{64}Zn

Thickness (mm)	$R_{Cd} - 1$	G_r
0.25	4.50	0.961
0.08	4.40	0.986
Extrapolated	4.35	1.000

in a conventional well-type crystal gamma-ray spectrometer. The results are listed in Table I.

The extrapolated cadmium ratio obtained with this method agrees with the value obtained by the first method.

Since resonance neutrons shielded by the cadmium were negligible, $F = 1$. The spectrum correction was calculated to be -12%. We then obtain

$$S_0 = 2.06 \pm 0.03$$

The reduced resonance integral calculated using the Breit–Wigner equation with the resonance parameters for neutrons with orbital angular momentum $l = 0$ (s capture) (Goldberg et al. 1966) gives $I' = 0.59$ b, but if p capture in the keV neutron resonances of ^{64}Zn is taken into account by means of the statistical model, $I' = 0.8 \pm 0.2$ is obtained.

The measured thermal cross section (Beckurts and Wirtz 1964), $\sigma_0 = 0.47 \pm 0.05$ b and $I' = 0.8 \pm 0.2$ b, gives $S_0 = 2.0 \pm 0.5$ in agreement with our experimental result. Another evidence of the importance of p capture in the activation resonance integral of ^{64}Zn is that experimental self-shielding, as is shown in Table I, is much less than would be estimated applying multiple scattering self-shielding correction in the s-neutron resonances of ^{64}Zn.

A previous experimental result by Brune and Jirlow (1963) gives $S_0 = 1.72$, in reasonable agreement with our value.

^{68}Zn

Samples containing 8.21 mg/cm^2 of zinc and 0.002 mg/cm^2 of gold were irradiated, uncovered and cadmium covered, for 1 h. Twenty hours later the 438 keV gamma ray of ^{69}Zn^m was measured, over two periods, relative to the 411.8 keV gold gamma ray.

The averaged result from six irradiations gives

$$R_{Cd}(^{68}Zn)/R_{Cd}(^{197}Au) = 2.184 \pm 0.054$$

or

TABLE II

Summary of resonance integral values for ^{100}Mo

Integral measurements	σ_0 (b)	I' (b)	S_0
Cabell (1960)	0.199 ± 0.003	3.71 ± 0.20*	21.03
Baumann (1963)	0.199 ± 0.003†	4.06‡	23.01
This work	0.199 ± 0.003†	3.82 ± 0.15‡	21.65 ± 0.75

*Obtained from the Cabell value, subtracting the $1/v$ part, and corrected for a small (2%) epithermal self-shielding effect in the gold monitor.
†Assumed (Cabell 1960).
‡Derived from S_0 determination.

$$R_{Cd}(^{68}Zn) = 3.68 \pm 0.09$$

Since self-shielding and cadmium shielding of the main resonance were negligible, $F = 1$ and $G_r = 1$ and with a spectrum correction of -10% we get

$$S_0 = 3.72 \pm 0.14$$

The resonance integral calculated from the parameters given by Goldberg et al. (1966), including a small contribution from p captures is $I' = 3.01$ b and this, with the recommended thermal cross section for total capture $\sigma_0 = 1.095$ b, gives $S_0 = 3.1$, in reasonable agreement.

The experimental results of Brune and Jirlow (1963) give $S_0 = 1.97$ which is around 90% lower than our experimental value.

This discrepancy may be explained by the difficulty of discriminating in a low resolution experiment against the 0.511 MeV annihilation gamma-ray ^{64}Cu (12.8 h) produced by neutron capture in the copper impurities of zinc, because half-lives for ^{64}Cu and ^{65}Zn are similar. As the S_0 for ^{63}Cu is less than one (Dahlberg et al. 1961), the contribution of ^{64}Cu activity in the 440 keV gamma ray of ^{69}Zn will give an apparently lower S_0.

Rubidium (^{85}Rb)

Samples containing 3.17 mg/cm^2 of rubidium and 7.12 mg/cm^2 of zinc were irradiated uncovered and cadmium covered. Six days later the 1080 keV gamma ray of ^{86}Rb (18.7 d) was measured relative to the 1114 keV gamma ray of ^{65}Zn.

The averaged result from five irradiations gives

$$R_{Cd}(^{85}Rb)/R_{Cd}(^{64}Zn) = 0.295 \pm 0.002$$

or

$$R_{Cd}(^{64}Zn) = 5.37 \pm 0.05$$
$$R_{Cd}(^{85}Rb) = 1.58 \pm 0.02$$

With $F = 1$ and $G_r = 1$ and including a -9.8% spectrum correction, we get

$$S_0 = 18.4 \pm 0.6$$

The resonance integral calculated from the parameters recommended by Goldberg et al. (1966), $I' = 2.77$ b, and the activation thermal cross section measured in a thermal spectrum by $4\pi\beta-\gamma$ coincidence technique of Keish (1963), $\sigma_0 = 0.45 \pm 0.05$ b, give $S_0 = 6.9$, in disagreement with our result.

Early measurements of the activation thermal cross section of ^{85}Rb by Seren et al. (1947) and Lyon (1960) give a σ_0 value which is a factor of two higher than that of Keish, but these measurements were both made in a reactor spectrum. Since S_0 for ^{85}Rb is 18.4, the contribution of epithermal neutron activation in Seren and Lyon's σ_0 values will be quite high and will depend on the epithermal index in the site of irradiation.

An approximate value of the epithermal index can be obtained from the cadmium ratio of ^{55}Mn and ^{60}Co quoted by Lyon, and using our experimental S_0, it is possible to correct Lyon's value to obtain $\sigma_0 = 0.5$ b, in agreement with Keish's value. The recent determination of Para and Bettoni (1967) also agrees with the Keish σ_0 value.

Molybdenum (^{100}Mo)

Samples containing 0.675 mg/cm^2 of molybdenum and 0.012 µg/cm^2 of indium were irradiated for 5 min uncovered and cadmium covered. The relative intensities of the 191 keV of ^{101}Mo and 307 keV of ^{101}Te were measured relative to the 416 keV of ^{116}In over four ^{101}Mo half-lives.

The averaged result from five irradiations was

$$R_{Cd}(^{100}Mo)/R_{Cd}(^{115}In) = 0.858 \pm 0.010$$

or

$$R_{Cd}(^{100}Mo) = 1.49 \pm 0.02$$

Here $G_r = 1$ and $F = 1$, and with a spectrum correction of -10% we get

$$S_0(^{100}Mo) = 21.7 \pm 0.8$$

The resonance integral calculated with the recommended resonance parameters $I' = 8.04$ b and $\sigma_0 = 0.199$ b (Cabell 1960) gives $S_0 = 45.6$, in disagreement with our result, but if the calculated $\sigma_0 = 0.36$ b is used, $S_0 = 25.0$ is obtained, in better agreement.

In Table II our result is compared with previous experimental results. Good agreement is obtained with previous experimental work, so that a serious discrepancy still remains between the calculated and experimental resonance integral for ^{100}Mo (Schmidt 1966).

Ruthenium (^{102}Ru)

Samples containing 1.8 mg/cm^2 of ruthenium and 0.97 mg/cm^2 of selenium were irradiated, uncovered and cadmium covered, for 6 h. Three months later the 500 keV gamma ray of ^{103}Ru (140 d) was measured relative to the 265, 280, and 405 keV gamma ray of ^{75}Se.

The averaged result of six irradiations was

$$R_{Cd}(^{102}Ru)/R_{Cd}(^{74}Se) = 1.655 \pm 0.005$$

or with

$$R_{Cd}(^{74}Se)/R_{Cd}(^{197}Au) = 1.322 \pm 0.006$$

and

$$R_{Cd}(^{197}Au) = 1.674 \pm 0.003$$

we have

$$R_{Cd}(^{102}Ru) = 3.66 \pm 0.02$$

With $F = 1$ and $G_r = 0.996$ and correcting by -9% for the spectrum deviation of the $1/E$ spectrum, we calculated

$$S_0 = 3.76 \pm 0.03$$

Only one resonance at 200 eV (Bolotin and Chrien 1963) is known for this nuclide which has a contribution of $I' = 5.4$ b to the total resonance integral. With our measured S_0 and the experimental $\sigma_0 = 1.44 \pm 0.16$ b (Hughes et al. 1958), one obtains $I' = 4.80$ b.

A measurement of 4.14 b for the total activation resonance integral of ^{102}Ru was made by Lantz (1965). Subtracting the $1/v$ contribution and normalizing to the thermal cross section

quoted by the author, $I' = 3.65$ b and $S_0 = 3.5 \pm 0.3$ can be deduced. This result agrees with ours within the error and may be lower because of the probable self-shielding effect for 10 mg samples used by Lantz (4% epithermal self-shielding effect for 10 mg/cm^2 natural ruthenium samples).

Indium (^{113}In)

Samples with 0.187 mg/cm^2 of indium and 8.8 mg/cm^2 of selenium were irradiated uncovered and cadmium covered. About a month later the 190 keV of ^{114}In (50 d) was measured relative to the 265, 280, and 405 keV gamma ray of ^{75}Se.

The averaged result of six independent irradiations is

$$R_{Cd}(^{113}In)/R_{Cd}(^{74}Se) = 0.675 \pm 0.003$$

or

$$R_{Cd}(^{113}In) = 1.49 \pm 0.01$$

Values of $F = 0.984$, $W = 0.069$, and $g = 1.000$ were calculated because ^{113}In has a low energy resonance at 1.8 eV, and with a -1.8% spectrum correction we obtain

$$S_0 = 24.7 \pm 0.5$$

The calculated resonance integral $I' = 258.9$ b (Hughes and Schwartz 1958) and the recommended thermal cross section $\sigma_0 = 11.1$ b (Goldberg et al. 1966) give $S_0 = 26.3$, in good agreement with our experimental value.

Antimony (^{123}Sb)

Samples with 0.029 mg/cm^2 of antimony and 8.58 mg/cm^2 of zinc were irradiated, uncovered and cadmium covered, for 6 h. Four months later, the 600 keV gamma ray of ^{124}Sb (60 d) was measured relative to the 1114 keV gamma ray of ^{65}Zn.

The averaged result from three irradiations is

$$R_{Cd}(^{123}Sb)/R_{Cd}(^{64}Zn) = 0.262 \pm 0.005$$

or

$$R_{Cd}(^{123}Sb) = 1.407 \pm 0.030$$

with $F = 1$ and $G_r = 1$ and with a -3.5% spectral correction, we get

$$S_0 = 28.3 \pm 2.1$$

The parameters recommended by Goldberg et al. (1966), corrected for the error mentioned in the "note added in proof", were used in the calculation of $I' = 114.3$ b. The assumed thermal

TABLE III

Summary of resonance integral values for ^{180}Hf

Integral measurements	σ_0 (b)	I' (b)	S_0	Method
Scoville et al. (1964)	11 ± 2	$11\pm6\ddagger$	1.128	Absorption
Fulmer (1968)	—	$43\pm8\ddagger$	—	Oscillation
Macklin and				
Pomerance (1956)	—	15.5§	—	Activation
This work	$12.6\pm0.7*$	$24.3\pm1.6\dagger$	2.17 ± 0.09	Activation
From resonance parameters				
Parameters listed by				
Goldberg et al. (1966)	12.6 ± 0.7	29.6	2.65	—

*Assumed (Goldberg et al. 1966).
†Derived from S_0 determination.
‡References do not specify if the resonance integral quoted includes the $1/v$ contribution.
§The $1/v$ contribution calculated with $\sigma_0 = 12.6$ b and $E_{Cd} = 0.4$ was subtracted from $I_{Cd} = 21.8$ b (Macklin and Pomerance, 1956).

TABLE IV

Summary of results

Nuclide	Measured S_0	σ_0 (b) (assumed)	This experiment (derived)	Resonance parameters	Other measurements
			I' (b)		
^{64}Zn	2.06 ± 0.03	0.47 ± 0.05	0.86 ± 0.09	0.8	0.67^a
^{68}Zn	3.72 ± 0.14^b	1.095 ± 0.15	3.61 ± 0.51	3.01	—
^{85}Rb	18.4 ± 0.6	0.45 ± 0.04	7.34 ± 0.68	2.77	—
^{100}Mo	21.7 ± 0.8	0.199 ± 0.003	3.82 ± 0.15	8.04	3.71 ± 0.20^c
^{102}Ru	3.76 ± 0.03	1.44 ± 0.16	4.80 ± 0.52	d	4.4 ± 0.4^g
^{113}In	24.7 ± 0.5	11.1 ± 1.3	243.1 ± 29.0	258.9	—
^{123}Sb	28.3 ± 2.1	4.50 ± 0.14	112.9 ± 8.7	114.3	138^e
^{180}Hf	2.17 ± 0.09	12.6 ± 0.7	24.3 ± 1.6	29.6	f
Standard					
^{197}Au	17.7	98.8 ± 0.3	—	1551	—

^{a}Brune and Jirlow (1963). $\sigma_0 = 0.44$ b is reported by the authors.
$^b R_{Cd}$ for ^{69}Zn^m was measured, supposing equal isomeric yield ratio for thermal and epithermal neutrons.
^{c}See Table II.
^{d}There is only one assigned resonance for ^{102}Ru.
^{e}Macklin and Pomerance (1956).
^{f}See Table III.
^{g}Lantz (1965). Subtracting $1/v$ contribution and normalizing to $\sigma_0 = 1.44$ b.

cross section was the average of the experimental values of Courtemanche et al. (1966) and Sims and Juhnke (1968), which is $\sigma_0 = 4.5$ b. Using these values an $S_0 = 28.7$ is obtained, in excellent agreement with our experimental result.

Previous experimental results from Macklin and Pomerance (1956) and from Sims and Juhnke (1968) give a reduced resonance integral around 138 b which is 20% higher than the calculated value.

Hafnium (^{180}Hf)

Samples with 0.0069 mg/cm^2 of hafnium and 3.53 mg/cm^2 of zinc were irradiated uncovered and cadmium covered. About a month later the 482 keV gamma ray of ^{181}Hf was measured relative to the 1114 keV gamma ray of ^{65}Zn.

The average of three independent irradiations is

$$R_{Cd}(^{180}\text{Hf})/R_{Cd}(^{64}\text{Zn}) = 0.994 \pm 0.030$$

or

$$R_{Cd}(^{180}\text{Hf}) = 5.34 \pm 0.16$$

Here $F = 1$ and $G_r = 1$ and with a spectrum correction of -7%, we obtain

$$S_0 = 2.17 \pm 0.09$$

The resonance integral calculated from the resonance parameters $I' = 29.6$ b and $\sigma_0 = 12.6 \pm 0.7$ b (Goldberg et al. 1966) gives $S_0 = 2.65$, in reasonable agreement with our experimental value.

Our result is compared with previous experimental results in Table III.

Discussion and Conclusions

In Table IV the final results are compared with the calculated values and with results from previous experimental determinations.

A better agreement is observed between our experimental resonance integrals and the calculated values than previous experimental determinations.

There have been no previously published values for ^{85}Rb.

^{85}Rb and ^{100}Mo experimental resonance integrals are in serious disagreement with calculated values.

Previous calculations made by Schmidt (1966) for ^{100}Mo with different resonance parameters show the same discrepancy, and the excellent agreement obtained with the results of Cabell (1960) and Baumann (1963) proves that there is no serious systematic error in the experimental S_0 value of ^{100}Mo.

Our value of the resonance integral and that of Baumann have been normalized to the thermal cross-section measurement of Cabell, which is a factor of two lower than the calculated σ_0 value.

The early activation cross-section determination of Seren et al. (1947) would apparently support the higher calculated value, but this determination has been made in a pile spectrum, and since the S_0 for ^{100}Mo is around 22, it is reasonable to expect Seren's value to be considerably higher than that of Cabell.

Since there is no serious systematic error in the experimental resonance integral of ^{100}Mo, the discrepancy may be attributed to the uncertainty in the resonance parameters used in the calculation of the resonance integral.

For the important resonances of ^{100}Mo, $\Gamma_n > \Gamma_\gamma$, then I' will be quite sensitive to the assumed value of Γ_γ. The Γ_γ value of 0.260 eV assumed in our calculation has not been directly measured but is based on measured Γ_γ values of other isotopes (Harvey et al. 1955) and may be lower by a factor of two or greater if the results of Huynh et al. (1967) are correct.

On the other hand, Baumann's boron filter measurements shows that a significant portion of the ^{100}Mo resonance integral comes from resonances at energies much higher than 364 eV. This is in disagreement with the resonance integral calculation that takes into account the s-wave neutron resonances above 364 eV (Gold-

berg et al. 1966), which indicates that only 30% of the neutron captures comes from higher energies.

These additional neutron captures may be due to unassigned p- or s-wave neutron resonances in the keV energy region of ^{100}Mo.

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