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NEUTRON CROSS-SECTIONS FOR NATURAL ZIRCONIUM IN THE ENERGY RANGE 0.0001 eV to 6 keV

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Abstract

The currently available resolved resonance data for the isotopes of zirconium are evaluated, with the view to compile a file of basic tabulated neutron cross-section data. (Data file number 9 now available in UKAEA Nuclear Data Library).

These data are also compared with integrally measured data, and from the discrepancies it is concluded that:

(i) there is a need for better measurements of the radiation widths of the resonances for all zirconium isotopes, and especially for the s-wave resonances of Zr91,

(ii) the measured values of the thermal neutron capture cross-sections of the isotopes are not consistent with the data for natural zirconium,

(iii) there are still considerable uncertainties in the resonance absorption integrals both for natural zirconium and the separated isotopes.

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1. INTRODUCTION

This report presents an evaluation of the data currently available on neutron induced reactions in zirconium, which have been used in the current data file for this material (Data File No.9) in the UKAEA Nuclear Data Library. A general account of the UKAEA Nuclear Data Library and an extensive bibliography may be found in the Geneva conference paper of Story et al. (1). The format used to represent the data on computer cards and magnetic tape is described by Parker (1a).

The neutron cross-sections of zirconium were recently evaluated by Hemmings (2) in the energy range 0.025 eV to 17 MeV. Since then much more information has become available, and the low energy cross-sections for the range 0.0001 eV to 6 keV have been re-evaluated by the author and the cross-sections have been calculated from the preferred set of resonance parameter values. These cross-sections have been combined with Hemmings' compilation for the energy range above 6 keV, to form the new data file, Date File No.9. The present report examines the experimental data available in the resolved resonance region and lists preferred values for the resonance parameters (see Section 3 below) and other pertinent information.

1.1 Natural zirconium

Zirconium occurs in nature as a mixture of five stable isotopes. Their relative atomic abundances (ref. 41) and assumed nuclear radii are quoted in Table 1.

Table 1

Stable Isotopes of Zirconium

Isotope	Abundance (%)	Nuclear radius (r) (cms^{-1})
Zr 90	51.46	7.40
Zr 91	11.23	7.43
Zr 92	17.11	7.45
Zr 94	17.40	7.50
Zr 96	2.80	7.56

The ground state spin and parity, I^{π}, are 0^{+} for each of the even-even isotopes, and $5/2^{+}$ for the odd one, Zr 91. Most of the observed resonance structure is due to Zr 91; the relatively close resonance spacing being a consequence of both the large spin value and large neutron binding energy. In contrast the abundant Zr 90 is a closed shell nucleus and the associated resonances are very widely spaced.

2. NEUTRON REACTIONS IN THE ENERGY RANGE 0.0001 eV - 6 keV

Table 2 shows that below 6 keV only the following neutron induced reactions are possible:-

(a) Elastic scattering (n,n),

(b) Radiative capture (n,γ), and

(c) Neutron induced α-emission (n,α).

The threshold energies of all other reactions are considerably greater than 6 keV.

Table 2

Q values in keV for neutron reactions with Zr isotopes (42)

Isotope	Reaction (n,γ)	(n,α)	(n,p)
90	7194 ±5	1741 ±5	-1512 ±7
91	8640 ±5	5647 ±7	- 758 ±10
92	6750 ±6	3400 ±7	-2846 ± 20
94	6468 ±6	2062 ±15	-4220 ± 200
96	5575 ± 22	-332 ± 100	-6100 ± 1000

2.1 Elastic scattering

In calculating the elastic scattering cross-section, the nuclear radii were taken as

$$r = 1.35\ (A^{\frac{1}{3}} + 1)\ \text{Fermis}$$

which gives the values listed in Table 1 for the different isotopes. The resonances are widely spaced and between resonances the scattering cross-section should be very nearly the same as the value for the potential scattering. On examining the calculated curve below 6 keV this is seen to be the case.

At 0.0253 eV, the scattering cross-section for natural zirconium is calculated as 5.7 barns, which agrees well with the value of 6.0 ± 0.5 barns recommended by Sjöstrand & Story (21), the difference is equivalent to an increase of only 2½% in the nuclear radii, and it was considered not worth while repeating the calculations with such a small change.

No account was taken of coherence effects in the slow neutron scattering even though the tabular cross-sections extend down to 0.10 meV.

2.1.1 Angular distribution

The angular distribution of elastically scattered neutrons of energy less than 6 keV is treated as isotropic in the centre of mass frame of reference. Deviations from isotropy may be expected at the p-wave resonances, but the effect should be very small in the average and has been ignored.

2.2 Neutron induced α-emission

Hemmings (2) did not include specifically the (n,α) reaction amongst his tabulated cross-sections. However, Table 2 shows that this reaction is energetically possible even with thermal neutrons for four of the stable isotopes of zirconium. However at the low neutron energies discussed in this report the reaction is strongly inhibited by the Coulomb barrier.

The energy release is greatest for the reaction Zr 91(n,α)Sr88, so the penetration factor was calculated for this reaction with 6 keV neutrons by using the computer programme CWF (see ref. 3). It was assumed that $\ell = 0$ for the neutron and $\ell = 2$ for the alpha particle. Using the penetration factor calculated in this way it was estimated that the (n,α) cross-section for natural zirconium is substantially less than 0.01 microbarns below 6 keV. This reaction was therefore ignored in the resonance region.

3. RESONANCE PARAMETERS

An interim set of cross-section data below 4 keV was derived from the set of resonance parameters listed in Table 3. This interim set formed part of Data File No.38 which has since been withdrawn. At about the same time a similar evaluation was carried out independently by Breton & Vidal (4). As one might expect, the selected parameter values were almost identical in these two evaluations (compare Table 3 with Table 2 of reference 4), since they were based on the same measurements; mainly those of Ohlsen et al. (5) and Morgenstern et al. (6). However more data have subsequently become available (see refs. 7-17) and with their aid the cross-sections in the resonance region have been computed, and are tabulated up to 6 keV in DFN9.

Table 3

Set of resonance parameter values used for an earlier tabulation below 4 keV

Energy keV	Target Nucleus	$2g\Gamma_n$ eV	Γ_n eV	Γ eV	Γ_γ eV	J	g
0.181	91	0.009			0.282		$(\frac{1}{2})$
0.2395	91	0.004			0.290		$(\frac{1}{2})$
0.291	91		0.600		(0.290)	3	
0.300	96		0.270	0.640	0.370	$\frac{1}{2}$	
0.675	91		0.690		(0.290)	3	
0.878	91	0.046			"		$(\frac{1}{2})$
1.110	91				"		
1.518	91		7.370		"	3	
1.933	91	0.260			"		$(\frac{1}{2})$
1.992	91	0.232			"		$(\frac{1}{2})$
2.218	91	1.340			"		$(\frac{1}{2})$
2.448	91	3.800			"		$(\frac{1}{2})$
2.661	92 or 94		26.500		"	$\frac{1}{2}$	
3.812	90		13.200		"	$\frac{1}{2}$	

The 0.24 keV resonance only is assumed to be excited by p-wave neutrons, all others assumed to be s-wave resonances. No data were available on the widths of the 1.11 keV resonance. It was assumed to be very small and was ignored.

All numbers in parentheses are assumed, somewhat arbitrarily.

Table 4 lists the resolved resonances and the preferred values of the resonance parameters.

Table 4

Preferred resonance parameter values used as a basis for DFN9 below 6 keV

Energy keV	Target Nucleus	$2g\Gamma_n$ eV	Γ_n eV	Γ_γ eV	J	ℓ	g
0.1593	91*				(3)	1	
0.1817	91	0.0105		0.315	(4)	1	
0.2411	91	0.0046		0.286	(2)	1	
0.2934	91	0.604	0.518	0.190	3	0	7/12
0.3014	96		0.240	0.370	$\frac{1}{2}$	0	1
0.442	91	0.0034		(0.302)	(3)	1	
0.682	91	0.793	0.680	0.491	3	0	7/12
0.898	91	0.037		(0.302)	(4)	1	
1.110	91*						
1.538	91	8.825	7.564	"	3	0	7/12
1.860	92		0.350	"	$\frac{1}{2}$	0	1
1.948	91	0.380		"	(2)	1	
2.018	91	0.250		"	(4)	1	
2.218	91	1.34		"	(2)	0	
2.258	94		1.40	"	$\frac{1}{2}$	(0)	
2.380	96		1.50	"	$\frac{1}{2}$	(0)	
2.478	91	5.2		"	(3)	0	
2.655	96		8.0	"	$\frac{1}{2}$	(0)	
2.693	92		21.85	"	$\frac{1}{2}$	(0)	
2.730	91	14.5		"	(2)	0	
2.770	91*			"			
3.173	91	2.6		"	(3)	0	
3.200	92≠		2.0	"	$\frac{1}{2}$	(0)	
3.300	92		1.9	"	$\frac{1}{2}$	(0)	
3.500	92		1.9	"	$\frac{1}{2}$	(0)	
3.630	91*						
3.813	96		7.063	"	$\frac{1}{2}$	(0)	
3.862	90		15.04	"	$\frac{1}{2}$	(0)	
3.870	91*						
4.010	91*						
4.160	92		3.8	"	$\frac{1}{2}$	(0)	
4.325	91*						
4.600	96		8.1	"	$\frac{1}{2}$	(0)	
4.683	92		14.03	"	$\frac{1}{2}$	(0)	
4.694	90		8.0	"	$\frac{1}{2}$	(0)	
4.750	91*						
4.940	94*						
4.970	91*						
5.100	92		19.97	"	$\frac{1}{2}$	(0)	
5.447	96	2.8		"	$\frac{1}{2}$	(0)	
5.600	91		24.27	"	(2)	0	
5.813	94		24.27	"	$\frac{1}{2}$	(0)	
5.920	96		11.3	"	$\frac{1}{2}$	(0)	
6.47	91*						
6.66	92*			"	$\frac{1}{2}$		
6.70	90		10.20	"	$\frac{1}{2}$	(0)	
6.85	94		31.5	"	$\frac{1}{2}$	(0)	
6.888	92		73.15	"	$\frac{1}{2}$	(0)	

Table 4 (continued)

Energy keV	Target Nucleus	$2g\Gamma_n$ eV	Γ_n eV	Γ_γ eV	J	ℓ	g
7.06	91_*			(0.302)			
7.16	94		47.0	"	$\frac{1}{2}$	(0)	
7.185	90		10.2	"	$\frac{1}{2}$	(0)	
7.72	94_*			"			
7.78	91_*			"			
8.51	91_*			"			
8.84	90_*			"			
9.07	91_*			"			
9.26	91_*			"			
9.83	92_*			"			
9.87	91_*			"			
10.40	94_*			"			
12.73	94		21.5	"	$\frac{1}{2}$	(0)	
13.626	90		49.7	"	$\frac{1}{2}$	(0)	
15.40	96		120.0	"	$\frac{1}{2}$	(0)	
15.40	$92_{\neq}$			"	$\frac{1}{2}$	(0)	
17.716	90		229.0	"	$\frac{1}{2}$	(0)	
17.80	94		59.1	"	$\frac{1}{2}$	(0)	
19.62	94		203.0	"	$\frac{1}{2}$	(0)	
22.7	92_*			"			
24.0	94		90	"	$\frac{1}{2}$	(0)	

*No parameter values available; it is assumed that this resonance may be neglected.

≠It seems likely that the effect of this resonance has already been accounted for by the parameters of one or more adjacent resonances, see Appendix A, accordingly this resonance has been ignored in the calculations.

All numbers in parentheses are assumed, somewhat arbitrarily.

These resonance parameters were derived from single level Breit-Wigner analyses of transmission data. In order to make use of these parameters for a multi-level synthesis of the neutron cross-section, it is necessary to identify or assign values of the orbital angular momentum quantum number ℓ and compound spin state J, as well as the neutron width n and radiation width Γ_γ.

As Table 4 shows, there appear to be 69 resolved resonances, but the isotopic assignment of some of these is still somewhat uncertain; see Appendix A. Should a resonance be wrongly assigned it is virtually impossible to re-assign it and revise the partial widths without specific knowledge of the relative abundances of the isotopes in the particular samples used in the experiments. However any errors arising from faulty assignments are likely to be relatively insignificant.

3.1 Orbital angular momentum, ℓ

Seven of the low energy resonances of Zr91 have been attributed to p-wave excitations because of the very small values of their reduced neutron widths; see Appendix A. There is some disagreement over the level at 182 eV; the small neutron width is suggestive of p-wave excitation. All the other resonances listed in Table 4 are taken as due to s-wave excitations.

3.2 The compound nucleus spin, J

One requires the J-value for two reasons in the multi-level calculations; (i) nearby resonances of the same ℓ and J interfere with one another (see ref. 18); (ii) the statistical factor g depends on J, where as usual

$$g = \frac{2J + 1}{2(2I + 1)} \qquad (3)$$

and I is the target nucleus spin; g is needed to calculate Γ_n from the measured $2g\Gamma_n$.

For s-wave neutron excitation of the even isotopes, I = 0 (see Section 1), therefore $J = \frac{1}{2}$ and hence $g = 1$. For Zr91, I = 5/2 (see Section 1); so that J = 2 or 3 for s-wave neutrons, and J = 1,2,3, or 4 for $\ell = 1$. Values of J have been reported in the literature for only three of these resonances, so the remainder have been nominated arbitrarily subject to the assumption that for a given ℓ-value the frequency of resonances of a particular J-value is approximately proportional to 2J+1.

3.3 The neutron width, Γ_n

Usually it is the parameter "$g\Gamma_n$" which is deduced from the measurements so that Γ_n depends to some extent on the chosen value of J.

Of the 69 resolved resonances listed in Table 4 the neutron widths have only been reported for 44. It has been assumed that the other 25 resonances are so weak that they have negligible effect on the neutron cross-sections below 6 keV, and they were ignored when generating tabular cross-sections for the data file. Although resonances have been resolved up to 24 keV, the range over which the detailed resonance structure is known is limited by the information available on the isotope Zr91 (see Section 1).

3.4 The radiation width, Γ_γ

The radiation width has only been determined for five of the resolved resonances, and the weighted value of the mean radiation width $\langle \Gamma_\gamma \rangle$ is

$$\langle \Gamma_\gamma \rangle = 302 \quad 40 \text{ meV}.$$

For want of better information this value has been used for all other resonances in calculating the cross-sections for the data file.

There is a serious lack of experimental information on Γ_γ ; and if we accept that Γ_γ may vary markedly from one isotope to another, and even with the parity of the state, the quoted uncertainty of ± 40 meV is decidedly optimistic. Further discussion of this question is given in Appendix B.

4. DERIVED CROSS-SECTIONS

Using the parameter values given in Section 2.1 and Table 4, and the computer programmes MLBW (ref. 18) and TEMPO (ref. 19), the Doppler-broadened cross-sections for elastic scattering and radiative capture were calculated and tabulated together with the total cross-section in the energy range 0.0001 eV to 5.9759 keV. The curve for the total cross-section was compared with the curve of experimental values, particularly at the resonance peaks and in the troughs between resonances. Exact agreement cannot be expected since the effects of instrumental resolutions have been ignored, however the general agreement seemed reasonably satisfactory.

4.1 Radiative capture

Table 5 shows the most reliable of the measured values of the capture cross-section of natural zirconium at 0.0253 eV.

Table 5

Measured thermal capture cross-section data for zirconium

σ_γ°[Zr] (millibarns)	Reference	Remarks
186 ± 18	25	Revised assuming σ_γ°[Au] = 98.4 barns
186 ± 7	26	Corrected for H_f impurity.
182 ± 2	27	
183 ± 4	31	

The value adopted in DFN9 was 183 ± 4 millibarns

The value at 0.0253 eV calculated from the resonance parameters was only 90 millibarns. The difference must be attributed to unresolved resonances and especially to the nearest negative energy resonances.

In view of the limitations of the data available it was felt sufficiently accurate to represent the unresolved contribution by a 1/v term

$$14.79/\sqrt{E} \text{ millibarns}$$

where E is in eV.

This addition was adopted in DFN9 and is sufficient to correct the low energy cross-section without introducing a significant error into the cross-section at higher energies. This 1/v term is of course not affected by Doppler broadening.

5. DISCUSSION OF THE DATA

Results calculated from the resolved resonance parameters have been compared with directly measured quantities. Particular interest attaches to the thermal capture cross-sections of the separate isotopes, and to the resonance integrals.

5.1 Thermal cross-sections

5.1.1 Isotopic thermal capture cross-sections

Values of thermal cross-sections have been measured for the separate isotopes as well as for the natural element, and are shown here in Table 6, while the corresponding results calculated from the chosen values of the resonance parameters are given in Table 7.

Table 6

Measured thermal capture cross-section data for the zirconium isotopes

Target nucleus	σ_γ^o	Contrib. to Nat. Zr. (millibarns)	Reference
90	104 ± 73	53.5	22
91	1580 ± 126	177.4	22
92	260 ± 78	44.5	22
94	75 ± 8	13.2	23
96	54 ± 5	1.5	23
	σ_γ^o[Nat.Zr]	290.1 ± 42 mb.	

N.B. The data of ref. (22) have been increased by 4%. assuming that the capture cross-section of gold, which was used as reference standard, should be taken as 98.8 barns at 0.0253 eV.

Table 7

Thermal capture cross-section data for the Zr. isotopes calculated from resonance parameters

Target nucleus	$\sigma_\gamma^o \pm$ Error	Contrib. to Nat. Zr. (millibarns)
90	42.5 ± 5.5	21.9
91	323.7 ± 15	36.4
92	124.4 ± 16	21.3
94	51.3 ± 7	8.9
96	301.5 ± 12	8.4
	σ_γ^o[Zr] =	96.9 ± 4.3 mb.

N.B. These results were computed with the programme RESIN (ref. 24) which makes an estimate for contributions from unresolved positive resonances. This is the reason why the value obtained for natural Zr is somewhat higher than that reported in Section 4.1 which was calculated with MLBW (ref. 18). However neither programme makes any allowance for unresolved negative energy resonances.

Attention is drawn to two serious discrepancies among the data listed in Tables 6 and 7:

(i) The calculated value of the thermal capture cross-section of Zr96 is much greater than the measured value (23). The value obtained from the resonance parameters may perhaps be too great by a factor 2 because of the uncertainty in the radiation widths, but this is not enough to resolve the discrepancy. The major contribution to the calculated value comes from the resonance at 301 eV.

(ii) The measured values of thermal capture cross-sections for the separate isotopes yield much too large a value for the thermal capture cross-section of natural zirconium. Although one might suspect some hafnium contamination of the enriched samples used, it is not very profitable to speculate on the possible distribution of such a contaminant between the different isotopes.

Column 4 of Table 6 shows the contribution to σ_γ^o[Zr(Nat)] from the measured values of each isotope; however the sum of these contributions, 290 millibarns greatly exceeds the recommended value of 183 millibarns, given in Section 4.1. The major contribution comes from Zr91 (177 millibarns), while the contribution from Zr96 is relatively unimportant even though it may be wrong by an order of magnitude. Adopting reasonable values for the contributions of the four even isotopes we are able to derive a more realistic value for σ_γ^o[Zr91] from the more accurate value for natural zirconium.

It is proposed then that σ_γ^o[Zr96] is put equal to 180 ± 130 millibarns; this contributes only 5 millibarns to the thermal cross-section of natural zirconium. The other three even isotopes contribute a further 111 millibarns (from column 3 of Table 6), leaving only 67 millibarns as the contribution from Zr91 to complete the 183 millibarns for the natural element. This corresponds to a thermal cross-section for Zr91 of 0.60 barns. If the smaller values for σ_γ^o quoted in Table 7 are taken for the isotopes Zr90, Zr92, and and Zr94 an overestimate is obtained for σ_γ^o[Zr91] of 1.2 barns. It is concluded that σ_γ^o[Zr91] = 0.9 ± 0.3 barns, instead of the value of 1.58 ± 0.12 barns obtained from the measurements of ref. (22).

5.2 Resonance integrals

We may also compare the integrally measured and the calculated values of the resonance absorption integrals at infinite dilution.

Measured values of the excess resonance integral for natural zirconium are listed in Table 8, and for the separate isotopes in Table 9. The values for the separate isotopes calculated from resonance parameters are listed in Table 10 and from them one may derive the excess resonance absorption integral for natural zirconium.

$$I'[Zr] = 1.56 \pm 0.06 \text{ barns}$$

Table 8

Measured values of resonance absorption integral for natural zirconium

Excess resonance integral (barns)	Reference	Remarks
2.9 ± 2	28)	Ref. (4) suggests Hf impurities may be present.
3.6 ± 0.5	29)	
2.2 ± 0.5	30)	
0.85 ± 0.15	31)	Pile oscillator measurements.
1.06 ± 0.14	33)	
1.0 ± 0.2	43	

The quoted errors indicate a preferred weighted mean = 0.97 ± 0.09 barns.

Measurements by Tattersall et al. (32) and by Hone et al. (34) are also available but they require corrections for self-screening; see ref. (4).

Table 9

Measured values of resonance absorption integral for separate isotopes of zirconium

Target nucleus	Excess resonance integral (barns)	Reference	Remarks
Zr90	0.16 ± 0.02	43	
Zr91	6.7 ± 0.8	43	
Zr91	5.4 ± 1.6	35	Self-screening correction yet to be applied.
Zr94	0.2 ± 0.03	43	

Table 10

Calculated values of excess resonance absorption integral, using the resonance parameter values listed in Table 4

Target nucleus	Excess resonance integral (I') (barns)	No. of resonances	Contribution to I'(nat.Zr) (barns)
Zr90	0.224 ± 0.017	7	0.115 ± 0.011
Zr91	9.17 ± 0.46	30	1.030 ± 0.052
Zr92	0.926 ± 0.086	11	0.158 ± 0.015
Zr94	0.360 ± 0.053	11	0.062 ± 0.009
Zr96	7.112 ± 0.327	8	0.199 ± 0.009
	Total for natural zirconium		1.564 ± 0.057

N.B. The calculated values include allowances for unresolved resonances. The total contribution to I' for natural zirconium from unresolved resonances is about 0.07 barns.

A slightly smaller value of the excess resonance absorption integral, 1.55 barns, was calculated from Data File No.9 by using GALAXY (ref. 20); this trifling discrepancy results from the discrete number of data points used and from rounding errors.

The measurements of refs. 28, 29, 31, 32, 33, and 34 are pile oscillator measurements; those in refs. 31, 32, and 34 being measured in D_2O moderated reactors and in ref. 33 in a H_2O moderator. The possible sources of error in this type of measurement have been discussed before, but it is worth emphasising some of these in order to appreciate how difficult it is to obtain a reliable measurement:

(i) Very pure samples are needed; if the cross-section is small, even a small amount of impurity may seriously affect the results. For example Breton & Vidal (4) have suggested that the large values of I reported by refs. 28 and 29 may be due to Hf-impurities of maybe 1000 ppm. Dobrynin et al. (30) however have corrected their results for Hf impurities.

(ii) The minimum size of a sample is dictated by the reactivity response of the equipment. If the capture cross-section is small one must use a thick sample in order to obtain a measurable signal. Hence the measurements can only yield an effective resonance integral in a neutron flux which is depressed at the energies of high cross-section, and a self-screening correction is necessary. The thicker the sample, the greater the self-screening correction. For materials of small capture cross-section the effects combine to inhibit accurate measurements. It may be added that the problem of resonance self-screening corrections has not been adequately resolved for the case of strong resonance scattering.

(iii) The resonance integral measurement requires a 1/E flux spectrum. Bigham et al. (36) have shown how difficult this is to achieve and the error becomes more serious when the resonance capture occurs at high energies (~ 1 keV or above) while the reference standard samples the epithermal flux in the low eV region. For example when Au is used as the standard, it effectively measures the value of the neutron flux at about 5 eV; deviations from the 1/E law may be relatively unimportant for materials with strong resonances near 5 eV, but are not trivial if the resonance integral comes mainly from energies of 100 eV or more.

For the most recent measurements of zirconium (31) and (33) care was taken over sample purity and self-screening, but it is less clear that adequate attention has been given to spectrum measurement. A crude estimate of the possible magnitude of this effect is given in Appendix C and it is suggested that the integral data may not be inconsistent with a value for I'(nat. Zr) of 1.45 $\pm$ 0.17 barns. The calculated value obtained from resonance parameters is based upon a rather sweeping assumption about the radiation widths, and Appendix B considers the effect of uncertainties in this parameter. It is concluded that even adopting the lowest reasonable value for the radiation widths (viz. Γ_γ = 190 meV) the excess resonance absorption integral for natural zirconium

$$I'(\text{nat.Zr}) = 1.35 \pm 0.05 \text{ barns}$$

still exhibits a marked discrepancy with the weighted mean value of 0.97 $\pm$ 0.08 barns obtained from the measurements in Table 8.

6. CONCLUSIONS

The evaluation reported in this document shows that there is a need for better measurements of the radiation widths of the zirconium resonances for all isotopes, and especially for the s-wave resonances of Zr91.

The measured values of the thermal neutron capture cross-sections of the zirconium isotopes are not consistent with the data for natural zirconium.

There are still considerable uncertainties in the resonance absorption integrals both for natural zirconium and the separated isotopes.

7. RECOMMENDATIONS FOR FUTURE EVALUATIONS

In future evaluations of the zirconium data the work of Biggerstaff et al. (37), Bergman et al. (38), Dovbenko et al. (39) and Seth (40) should be taken into account.

8. ACKNOWLEDGEMENTS

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REFERENCES

1. J.S. Story, M.F. James, W.M.M. Kerr, K. Parker, I.C. Pull & P. Schofield. 3rd Geneva Conf. on Peaceful Uses of Atomic Energy P/168 (1964).

1a. K. Parker, AWRE 0-70/63 (1963).

2. P.J. Hemmings. "A compilation of nuclear data for natural zirconium". AHSB(S)R62 (1963).

3. D.S. Norton & M.F. James. "CWF and TABLE - Two Fortran programmes for the calculation of Coulomb penetration and shift factors". AEEW-M611 (1965).

4. D. Breton & R. Vidal. "Analysis of the resonance integral of zirconium". EANDC(E)42'U' (1963).

5. W.C. Ohlsen & P.P. Singh. BAPS 6 (No. 1), 69 (1961). [See also BNL 666 (1961)].

6. J. Morgenstern, C. Corge, V-D. Huynh, J. Julien & F. Netter. "Resonance parameters of zirconium and molybdenum". BAPS 7, 288 (1962). [See also Comptes rendus 254, 4009 & 4162 (1962)].

7. S.S. Moskalev, G.V. Muradyan & Yu. V. Adamchuk. Nuc. Phys. 53, 667 (1964).

8. G.V. Muradyan. Phys. Letters 14, 123 (1965).

9. H.J. Kim, J.A. Biggerstaff & W.M. Good. ORNL 3425, 32 (1963).

10. J.A. Biggerstaff, W.M. Good & H.J. Kim. ORNL 3582, 82 (1964).

11. R.C. Block. ORNL 3425, 32 (1963).

12. R.C. Block. ORNL 3582, 61 (1964). (Also published in Phys. Rev.).

13. A. Stolovy, J.A. Harvey. Phys. Rev. 108, 353 (1957).

14. F. Netter. Unpublished. (1962)

15. H.E. Jackson. EANDC(US) 61 'U' (1964). [See also Phys. Rev. 131, 2153 (1963)].

16. J.A. Moore, A.P. Jain, W. Lee, R.E. Chrien & H. Palevsky. WASH 1041, 9 (1962).

17. R.C. Block, R.W. Hockenbury & J.E. Russell. ORNL-TM-874, 6 (1964).

18. K. Gregson, M.F. James & D.S. Norton. "MLBW - A Multi-level Breit-Wigner Computer Programme". AEEW-M517 (1965).

19. K. Gregson & M.F. James. "TEMPO - A General Doppler Broadening Programme for Neutron Cross-Sections." AEEW-M517 (1965).

20. V.J. Bell, L.W. Blott, W.M.M. Kerr, K. Parker, I.C. Pull, R.D. Wade & D.V.J. Williams. "A Users Guide to GALAXY 3". AEEW-R379 (1964).

REFERENCES (contd.)

21. N.G. Sjöstrand & J.S. Story. "Thermal Neutron Absorption and Scattering Cross-Sections of All Elements". AEEW-M197 (1962).

22. H. Pomerance. Phys. Rev. 88, 412 (1952).

23. W.S. Lyon. Nucl. Sci. Eng. 8, 378 (1960).

24. A.L. Pope. "RESIN - A Computer Programme for Calculating Resonance Integrals and Thermal Cross-Sections from Resonance Parameters". To be published.

25. H. Pomerance. Phys. Rev. 83, 643 (1953).

26. D. Jowitt, S.K. Pattenden, H. Rose, V.G. Small & R.B. Tattersall. AERE R/R2516 (1959).

27. Progress Report on Nuclear Data Research in the EURATOM-Community - Service des Expérience Neutroniques, CEA (France) - 25.3.3 - Mesure des sections efficaces thermiques sur ZOE (1966).

28. R.L. Macklin & H.S. Pomerance. P/833 Geneva Conference (1955).

29. V.B. Klimentov & V.N. Griazev. Journal of Nuclear Energy, Vol. 9, p.20 (1959).

30. P. Dobrynin, G.A. Dorofeyev & I.E. Kutikov. Atomnaya Energiya 3, 323 (1957).

31. E. Hellstrand, G. Lindahl & G. Lundgren. AE 59 (1961) and Arkiv För Fysik 20 (6), 559 (1962).

32. R.B. Tattersall, H. Rose, S.K. Pattenden & D. Jowitt. AERE-R2887 (1959).

33. R. Vidal & E. Cardoso-Martinho. "Mesure des Integrales de Resonance d'Absorption (Mn, Fe, Co, Ni, Cu, Zr, Mo)". CEA-R2840 (1965).

34. D.W. Hone, K. Serdula & E. Critoph. RPI 59 (1959).

35. F. Feiner. "A Measurement of the Resonance Integral of Zr91". KAPL 2000-8 (Dec. 1959).

36. C.B. Bigham & R.M. Pearce. AECL-1228 (1961).

37. J.A. Biggerstaff, W.M. Good & H.J. Kim. ORNL-3778 p.76. See also "International conference on the study of nuclear structure with neutrons" P.67 Antwerp (July 19th-23rd, 1965.)

38. A.A. Bergman, S.P. Kapchigashev, Ju. P. Popov & S.A. Romanov. "International conference on the study of nuclear structure with neutrons" P.182 Antwerp (July 19th 23rd, 1965).

39. A.G. Dovbenko, S.M. Zakharova, V.E. Kolesov & A.V. Malyshev. Soviet Atomic Energy 18 (No.2), 140 (1965).

40. K.K. Seth. Physics Letters 16 (No.3), 306 (1965).

REFERENCES (contd.)

41. W. Seelmann-Eggebert, G. Pfennig, H. Münzel & G. Zundel. Second edition of "Chart of the Nuclides" (1961).

42. J.H.E. Mattauch, W. Thiele & A.H. Wapstra. Nuclear Physics 67, 1 (1965).

43. S.P. Kapchigashev. Atomnia Energia 19, 294 (1965).

Appendix A

Literature values of the resolved resonance parameters of the zirconium isotopes (from which the preferred set given in Table 3 was derived)

Table A1

Zr 90

E_r (eV)	Γ_n (eV)	ℓ	Ref.	Remarks
3800	32.7	0	10	
3812	13.2 ± 2		6	
3860			17	
3880	8.2 ± 1.3		11	
3905	7.5 ± 1.5		5	
3912	13.6 ± 0.5		7	
4690	8.0 ± 3.0		7	
4697			5	
6700	10.2	0	10	[See Zr92, E_r = 6660 eV, ref.17]
7100	10.2	0	10	
7270			17	
8840			17	
13400	60.8	0	10	
13500	37 ± 19		11	
13670	65 ± 15		7	
13934	66 ± 15		5	
17200	213	0	10	
17800	240 ± 44		11	Ref.11 finds a doublet at this energy.
17850	200 ± 70		7	
18007	263 ± 20		5	
19400		0	10	

Table A1 continued

Zr 91

E_r (eV)	$2g\Gamma_n$ (meV)	ℓ	Ref.	Remarks
158.5		1	14	
160.0			17	
180	10 ± 2		6	J = 2 or 3
182		0	8	ℓ = 0 assignments seem quite
182	8 ± 1		5	doubtful in view of
182.2	11 ± 2	1	11	small Γ_n.
182.2			17	
182.5	13 ± 2			
238	4 ± 1	1	6	Γ = 290±50meV, Γ_γ = 286±50meV.
239		1	14	
240 ± 1	6 ± 3		7	ℓ = 1 is inferred from small Γ_n^o
240	3.8 ± 0.4	1	11	
240.6			17	
242		1	8	
247		1	15	
291	700 ± 70		6	Γ = 800±100meV, Γ_γ = 200±120meV J = 3.
293			8	
293	680 ± 50	0	11	ℓ = 0 is inferred from large Γ_n^o.
293.2			17	
293.3±.5	570 ± 100		7	Γ = 750±100meV, Γ_γ = 180±140 meV.
294	475 ± 55		5	
296 ± 9			13	Γ = 1400 meV.
420		1	16	
445	3.4 ± 1.0	1	11	ℓ is inferred from small Γ_n^o.
447		1	14	
448		1	15	
450			17	
675	800 ± 100		6	Γ = 1100±150meV, Γ_γ = 414 ± 190 meV. J = 3
683	880 ± 90	0	11	ℓ is inferred from large Γ_n^o.
683			17	
685	761 ± 65		5	Γ = 1610±400meV, Γ_γ = 850 ± 410 meV. J = 3.
687	730 ± 120		7	
700		0	15	
(878	0.046 ± .010		6	
(890	0.028 ± .010	1	11	
(893			17	
(930		1	15	
1110			6	

Table A1 continued

Zr 91 continued

E_r (eV)	$2g_n$ (meV)	ℓ	Ref.	Remarks
1518	8.6 ± 1.0		6	Γ = 8.0±1.0, J=3 [Γ_γ=0.74 ± 1.4]
1535			17	
1538±6	10.0 ± 1.0		7	
1540	8.4 ± 0.8	0	11	ℓ inferred from large Γ_n^o.
1542	8.3 ± 1.1		5	
1550		0	15	
1993	0.26± 0.08	1	6	
1950	0.05± 0.30		7	
1960			17	
1992	0.232±0.060	1	6	
2010			17	
2020	2.3 ±0.7		7	
2050		1	15	
2218	1.34 ±0.16		6	[This res. may be Zr92, see refs. 7,11,17].
2390			17	[See Zr96, E_r = 2380eV, ref. 7]
2448	3.8 ±1.0		6	
2480	3.3 ±1.0		5	
2480			17	
2490±12	6.5 ±1.5		7	
2490	7.2 ±1.1	0	11	ℓ inferred from large Γ_n^o.
2720	18.0 ±7.0		7	
2730			17	
2740	11.0 ±1.5	0	11	ℓ inferred from large Γ_n^o.
2770			17	
3170		0	17	[See Zr92, E_r = 3200eV, ref.10]
3175±30	2.6 ±0.8		7	
3630			17	
3870			17	
4010			17	
4320			6	
4330			17	
4750			17	
4970			17	
5600±50	2.5 ±1.0		7	
5600	7.0 ±4.0	0	11	
6470			17	
6750			9	[See Zr92, E_r = 6660eV, ref.17]

Table A1 continued

Zr 91 continued

E_r (eV)	$2g_n$ (meV)	ℓ	Ref.	Remarks
7060			17	
7780			17	
8510			17	
9070			17	
9260			17	
9870			17	
			Zr 92	
1860	0.35 ± 0.10		7	
2010			17	
2661	26.5 ± 3.0		6	
2680			17	
2699			5	Assigned in ref. 5 to Zr90 or Zr96.
2700	23.5	0	10	
2700	23 ± 2		11	
2720 ± 15	14.4 ± 2.0		7	
3200	2.0	0	10	[See Zr91, E_r = 3170eV, ref. 17 and 7.]
3300	1.9	0	10	
3500	1.9	0	10	
4140	4.1 ± 1.5		11	
4150 ± 30	3.5 ± 0.5		7	
4150			17	
4200	3.8	0	10	
4650			17	
4660	10 ± 4		7	
4670	12.5 ± 3.5		11	
4750	19.6	0	10	
5050			17	
5150	6.0	0	10	
6660			17	[See Zr90, E_r = 6700eV, ref. 10].
6830			17	
6880	80 ± 10		7	
6890	72 ± 9		11	
6900	70.6	0	10	
6942	70 ± 10		5	
9830			17	

Table 1 continued

Zr 92 continued

E_r (eV)	Γ_n (eV)	ℓ	Ref.	Remarks
15400		0	10	
17100		0	10	
22700		0	10	

Zr 94

2250	2.0 ± 0.6		11	[See also Zr91, E_r = 2218eV, ref. 6].
2250			17	
2273±10	0.8 ± 0.1		7	
2750			17	
4940			17	
5700			9	
5700	23.8	0	10	
5840	22 ± 5		11	
5840			17	
5870	27 ± 7		7	
6800			9	
6850	31.5	0	10	
7100	22 ± 7		11	
7220	72 ± 8		7	
7720			17	
10100			17	
10700			9	
12600	23 ± 18		11	
12800±160	20 ± 7		7	
12800			9	
15800			9	
17800			9	
17800	59.1	0	10	
19000	264.0	0	10	
19500			9	
19860	125 ± 40		7	
20000	220 ± 50		11	
24000	90 ± 50		11	

Table 1 continued

Zr 96

E_r (eV)	Γ_n (eV)	ℓ	Ref.	Remarks
300	0.270 ± 0.025		6	Γ = 0.640±0.080eV, Γ_γ = 0.370±0.090eV
301	0.22 ± 0.03		11	
302	0.300 ± 0.040		5	
302.7±0.5	0.17 ± 0.02		7	
2380	1.5 ± 0.3		7	[See Zr9, E_r=2390eV, ref.17]
2620			9	
2690	8 ± 3		11	
2820			9	
3800	5.69	0	10	
3830	11 ± 4		11	
3840±40	4.5 ± 1.0		7	
4600	8.1	0	10	
5400	18.9	0	10	
5430	25 ± 4		11	
5510	16.0 ± 8.0		7	
5900	13.6	0	10	
5940	9 ± 5		11	
6700			9	
15400	120 ± 40		11	
29300			9	

When examining resonance parameters measured by different experimenters one may discern some systematic trends in resonance energies. For example refs. 5 and 15 quote resonance energies systematically higher than others, while ref. 6 quotes lower energies.

Appendix B

Radiation widths of zirconium resonances

One may see from Table A1 that very few determinations of the radiation widths of the resonances are available; and many of these are quite inaccurate. The most accurate values are those for p-wave resonances which may, after all, be quite different from those for s-wave resonances which have opposite parity.

For Zr 90, 92 and 94 no radiation widths have been measured.

For Zr 96 only one, and that rather larger than the adopted value for the mean.

For Zr 91, the radiation width for s-wave excited resonances may be determined from the neutron widths and total widths of two resonances at 291 eV and 680 eV. The results for the 291 eV resonance seem in little doubt since only the spin value J = 3 seems acceptable; the measurements for this resonance give $\Gamma_\gamma \approx 190 \pm 120$ meV. However the measurements at 680 eV are much less satisfactory. The spin J could be 2 or 3, and the following table shows the effect on Γ_γ of varying J.

Table B1

E_r (eV)	$2g\Gamma_n$ (meV)	Γ (meV)	Ref.	J	Γ_γ (meV)
675	800 ± 100	1100 ± 150	6	3	414 ± 190
				2	240 ± 190
685	761 ± 65	1610 ± 400	5	3	850 ± 410
				2	697 ± 410

These values for Γ_γ have such large uncertainties that with J = 2, the results are by no means inconsistent with the value of 190 meV obtained for the 291 eV resonance.

Table B2 shows the calculated values of the resonance integrals of the zirconium isotopes and natural zirconium under the assumption that Γ_γ = 190 meV for all s-wave resonances. The values may be compared with those given in Table 10 in the body of the report.

Appendix B continued

Table B2

Material	I' (barns)	No. of res. resonances	Contrib. to I'(nat.) (barns)
Zr 90	0.143 ± 0.011	7	0.074 ± 0.006
Zr 91	8.29 ± 0.38	30	0.931 ± 0.043
Zr 92	0.620 ± 0.056	11	0.106 ± 0.010
Zr 94	0.235 ± 0.035	11	0.041 ± 0.006
Zr 96	6.906 ± 0.327	8	0.193 ± 0.009
	Natural zirconium	67	1.345 ± 0.05

N.B. The calculated values include allowances for unresolved resonances. The total contribution to I' for natural zirconium from unresolved resonances is about 0.05 barns.

Appendix C

Effect of spectrum uncertainties on measured resonance integrals

The mechanism for an oscillator requires considerable space; this space may be obtained by removing a fuel element from the lattice. The conditions are then similar to those examined by Bigham et al. (36) in which the spectrum is changed, and using their figure 13 one would expect the measured value of I'(nat. Zr) to be too small by approximately 0.4/0.6. Under these conditions the weighted mean value, I' = 0.97 barns, obtained from the measurements in Table 8 should be increased to

$$I'(\text{nat. Zr}) \approx 0.97 \times \frac{0.6}{0.4} = 1.45\ (\pm\ 0.17)\ \text{barns}$$

The example is crude, but it serves to illustrate the size of the correction which may be needed.