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(n, 2n) cross sections and the statistical model predictions

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Abstract. Published literature up to October 1973 has been scanned to obtain the experimental (n, 2n) cross sections at $E_n = 14$ to 15 MeV. Weighted average of the total (n, 2n) cross sections reported and the probable error in it are tabulated for each nuclide in the region $A = 2$ to 238. (n, 2n) cross sections at $E_n = 14.5$ MeV have been calculated using Pearlstein's formulae based on the statistical model with new constants determined by us; these theoretical cross sections are also tabulated for nuclides in the region $A = 23$ to 209.

A comparison of the theoretical values and the weighted averages of the experimental values shows that (a) the statistical model values in general agree well with the experimental values; (b) the theoretical cross sections agree well with the experimental cross sections even for target nuclides having neutron shell closures except in the case of ^{39}K and ^{54}Fe which have high (n, 2n) thresholds comparable to the incident neutron energy. It is concluded that there are no shell effects other than those manifesting in Q values and the level density parameters which have been properly taken into account in our calculations.

1. Introduction

Above its threshold, the (n, 2n) reaction is most prolific and is of practical interest to reactor physicists as it can affect the neutron distribution in reactors. For almost a decade there has been controversy regarding shell effects in the 14–15 MeV (n, 2n) cross sections; some (Strohal *et al* 1962, Bormann 1965, Manero 1966, Cuzzocrea and Notarrigo 1966, Chattarjee and Chattarjee 1969) claim to have demonstrated the existence of shell effects while others (Ruder and Sitzber 1966, Hille 1968) argue that there are no shell effects in the reported cross sections. In spite of the large number of measurements, the experimental facts in this connection have so far been unclear. The aim of this investigation is to review the experimental facts without excluding any type of measurements on subjective grounds and compare these with the theoretical values based on the statistical model using Pearlstein's formalism and thereby deduce whether any prominent shell effects occur in the (n, 2n) cross sections at 14–15 MeV incident neutron energy.

2. Evaluation of the experimental data

Whereas recent measurements using the mixed powder method (Rao and Fink 1967) and gamma detection by Ge(Li) detectors may be superior to scintillation counter detection of γ rays and GM counter detection of β rays and other neutron detection methods, the former is limited in its application to radioactive cases emitting sufficiently intense γ rays and having well determined decay schemes; this method is not so

useful in those cases where the radioactivity is mainly beta emission and cannot be used when the residual nucleus is non-radioactive or has a very long half-life. As a matter of fact, some of the beta counting methods employing thin samples when corrections for sample thickness are properly taken into account, are found to yield results just as reliable as the gamma counting methods. In view of this, we have taken into account all the measurements published in regular scientific journals; however we have not included the following types of results.

(i) Results reported without error limits.

(ii) Values reported with an error greater than 20% unless it is the only measurement available for that particular nuclide.

(iii) Partial cross sections giving σ_m or σ_g only when they are not useful to compute the experimental total (n, 2n) cross section.

(iv) All private communications and results published in internal reports or abstracts are generally neglected.

(v) When the same author or laboratory reported more than one value for the same cross section over different periods, the latest value reported is taken neglecting the others.

Most of the measurements employed a T(d, n) source with an incident neutron energy spread of about 0.2 MeV or more. Whereas the excitation function rises quickly in the neighbourhood of the (n, 2n) threshold, it does not generally rise so steeply when the incident neutron energy is more than about 2 MeV above the (n, 2n) threshold; considering the errors involved in the measurements, it is not unrealistic to group all the 14 to 15 MeV neutron cross sections reported in the literature as representative cross sections at $E_n = 14.5 \pm 0.5$ MeV. So, we have calculated the neutron cross section assuming the incident neutron energy as 14.5 MeV.

3. Computation of weighted averages and errors

The weighted average of all the total (n, 2n) cross sections reported for a nuclide is calculated giving a weight inversely proportional to the error quoted by the authors in each case:

$$\bar{\sigma} = \frac{\sum_{i=1}^N (\sigma_i / d\sigma_i)}{\sum_{i=1}^N (1/d\sigma_i)} \quad (1)$$

where $(\sigma_i \pm d\sigma_i)$ is the reported (n, 2n) cross section σ_i with the error $d\sigma_i$ for a particular nuclide.

The error in the weighted average cross section $\bar{\sigma}$ is calculated in two different ways and whichever is the larger of the two, is taken as the likely error.

$$d\bar{\sigma} = 0.6745 \left(\frac{\sum_{i=1}^N [(\sigma_i - \bar{\sigma})^2 / d\sigma_i]}{\sum_{i=1}^N (1/d\sigma_i)} \right)^{1/2} \quad (2a)$$

or

$$d\bar{\sigma} = \left(\frac{[\sum_{i=1}^N (d\sigma_i)^2]^{1/2}}{N} \right) \quad (2b)$$

where N is the number of reported values included in calculating $\bar{\sigma}$.

4. Pearlstein's method

Pearlstein (1967) reported a method of calculating (n, 2n) cross sections on the basis of the statistical model using the relation

$$\sigma_{(n,2n)} = \sigma_{ne} \left(\frac{\sigma_{n,M}}{\sigma_{ne}} \right) \left(\frac{\sigma_{n,2n}}{\sigma_{n,M}} \right) \quad (3)$$

where σ_{ne} is the non-elastic cross section, $\sigma_{n,M}$ is the sum of all neutron emission cross sections, like (n, n'), (n, 2n), (n, 3n) etc, allowed by the Q values; however, it does not include the elastic scattering (n, n). For σ_{ne} Pearlstein used an empirical relation given by Flerov and Talyzin (1957) whereas we now use the more accurate optical model values of the non-elastic cross sections given by Mani *et al* (1963). The ratio $(\sigma_{n,2n}/\sigma_{n,M})$ is calculated on the basis of the statistical model using the relation,

$$\frac{\sigma_{n,2n}}{\sigma_{n,M}} = \frac{\int_0^{E_n - S_n} \epsilon \sigma_c(\epsilon) \exp[4a(E_n - \epsilon)]^{1/2} d\epsilon}{\int_0^{E_n} \epsilon \sigma_c(\epsilon) \exp[4a(E_n - \epsilon)]^{1/2} d\epsilon} \quad (4)$$

where $\sigma_c(\epsilon)$ is the compound nucleus (CN) formation cross section, E_n is the incident neutron energy (14.5 MeV), S_n is the neutron separation energy of the target nucleus and a is the level density parameter. This equation is based on the assumption that the CN emits a second neutron after emission of the first neutron whenever it has sufficient energy to do so; the numerator is proportional to the number of emitted neutrons whose energies are such that the residual nucleus has an excitation energy greater than the neutron binding energy; the denominator is proportional to the total number of neutrons emitted. Pearlstein used the effective spin values of neutron and proton shells evaluated by Newton (1956) to find the values of the level density parameter a occurring in equation (4) whereas we have used the more recent values given by Abdelmalek and Stavinsky (1964). Pearlstein took neutron separation values, S_n , from the tables of Konig *et al* (1962) whereas we have used the latest values given by Wapstra and Gove (1971).

The ratio $(\sigma_{n,M}/\sigma_{ne})$ was evaluated by Pearlstein using an empirical formula given by Barr *et al* (1961)

$$\frac{\sigma_{n,M}}{\sigma_{ne}} = 1 - k \exp\left(-m \frac{(N - Z)}{A}\right) \quad (5)$$

where $k = 1.764$ and $m = 18.14$; Lu *et al* (1970) modified these values to $k = 1.8124$ and $m = 12.99$; Kondaiah and Athougies (1974) obtained $k = 1.365 \pm 0.04$ and $m = 10.605 \pm 0.43$ by a weighted least squares fit of $(\sigma_{n,M}/\sigma_{ne})$ values derived from experimental (n, 2n) cross sections for more than a hundred nuclides over the region $(N - Z)/A = 0.03$ to 0.22; figure 1 gives the least squares fit curve for $(\sigma_{n,M}/\sigma_{ne})$ against $(N - Z)/A$; the central one is represented by equation (5) with $k = 1.365$ and $m = 10.605$ while the curves on either side of it represent the limiting cases including errors in k and m mentioned above. We have used these values of k and m .

In equation (3) σ_{ne} is known correct to about 0.1 mb (Mani *et al* 1963); the values of S_n occurring in equation (4) are known quite accurately compared to the uncertainty of the incident neutron energy; the level density parameter a occurring in equation (4) is known to an accuracy of about 7% (Abdelmalek and Stavinsky 1964); the constants k and m occurring in equation (5) are known to an accuracy of 3 to 4% (Kondaiah and

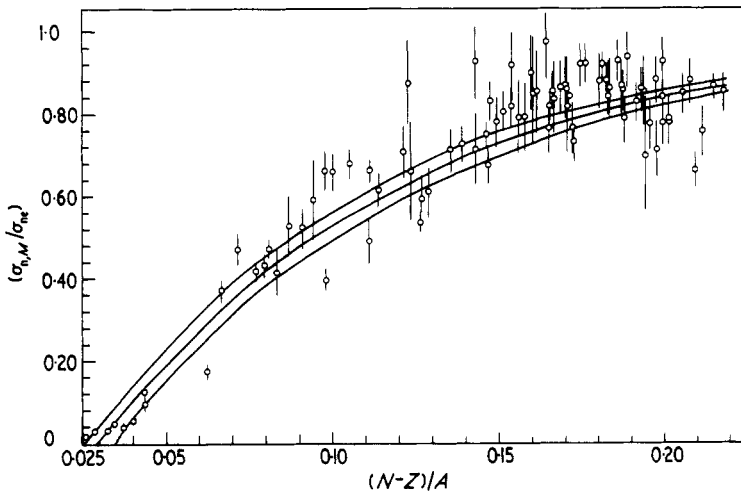


Figure 1. $(\sigma_{n,M}/\sigma_{ne})$ plotted against $(N-Z)/A$. The central curve is the least squares fitted curve; those on either side of it are the ones obtained by including the errors in k and m .

Athougies 1974); so one can expect to calculate $\sigma_{(n,2n)}$ using equations (3), (4) and (5) to an accuracy of about 10% and any large deviation from the calculated value, if confirmed by experiment, has to be attributed to the non-statistical nature of the $(n, 2n)$ reaction in that particular case.

5. Results and discussion

Table 1 gives target nuclide, weighted average of the total cross sections for each nuclide together with the error in it, and the theoretical cross section calculated using equations (3), (4) and (5) with our (Kondaiah and Athougies 1974) values for k and m ; the number in the bracket in column 2 refers to the number of experimental values included in calculating the weighted average cross section. The actual cross sections (and their references) used in arriving at the weighted average cross sections are given in an unpublished report which may be obtained from the authors by request.

An examination of table 1 shows that there is good agreement between the predicted cross sections and weighted averages of the reported experimental $(n, 2n)$ cross sections; about 60% of the cases agree to within an error of $\pm 10\%$, about 75% of cases agree to within an error of $\pm 15\%$ and about 90% of the cases agree to within an error of $\pm 25\%$. The following few cases are found to differ from the theoretical values by more than 30%.

(i) *Nuclides having high $(n, 2n)$ thresholds.* These nuclides have $(n, 2n)$ thresholds greater than or equal to 13 MeV and the residual nucleus is left with very little excitation energy ($\lesssim 1$ MeV); hence, equation (4) based on the statistical model is not generally valid; for these nuclides the $(n, 2n)$ cross section is a sensitive function of the incident neutron energy. ^{35}Cl ($E_{\text{th}} = 13.0$ MeV), ^{39}K ($E_{\text{th}} = 13.41$ MeV), ^{46}Ti ($E_{\text{th}} = 13.48$ MeV), ^{50}Cr ($E_{\text{th}} = 13.19$ MeV) and ^{54}Fe ($E_{\text{th}} = 13.87$ MeV) belong to this class.

(ii) ^{78}Kr , ^{86}Sr , ^{102}Pd and ^{174}Hf . These are measured by only one group of authors in each case and may be worth remeasuring.

Table 1. (n, 2n) cross sections at 14.0 ± 0.5 MeV.

Target	Weighted average $\sigma_{tot}(mb)$	$\sigma_{theo}(mb)$	Target	Weighted average $\sigma_{tot}(mb)$	$\sigma_{theo}(mb)$	
¹ H ₁	190 ± 14	(2)	¹⁰⁶ Cd ₅₈	995 ± 167	(2)	840
⁶ Li ₃	70 ± 6	(1)	¹⁰⁸ Cd ₆₀	865 ± 100	(1)	1023
⁷ Li ₄	56 ± 5	(1)	¹¹⁰ Cd ₈₂	1221 ± 150	(1)	1161
⁹ Be ₅	533 ± 32	(4)	¹¹⁶ Cd ₆₈	1427 ± 117	(3)	1459
¹⁴ N ₇	7.32 ± 0.72	(6)	¹¹³ In ₆₄	1624 ± 81	(4)	1212
¹⁹ F ₁₀	45.6 ± 5.1	(8)	¹¹⁵ In ₆₆	1823 ± 84	(2)	1321
²³ Na ₁₂	43.7 ± 3.6	(2)	50	1304 ± 114	(5)	971
³¹ P ₁₆	10.2 ± 1.1	(5)	10	1556 ± 188	(2)	1123
³⁵ Cl ₁₈	9.27 ± 1.36	(2)	0	1741 ± 68	(2)	1401
³⁹ K ₂₀	4.16 ± 0.62	(1)	0	1562 ± 217	(5)	1488
⁴⁸ Ca ₂₈	907 ± 107	(2)	817	1554 ± 92	(2)	1338
⁴⁵ Sc ₂₄	321.5 ± 25	(4)	287	1689 ± 127	(2)	1586
⁴⁶ Ti ₂₄	37.3 ± 7.5	(4)	56	907 ± 125	(2)	1644
⁵¹ V ₂₈	660 ± 50	(1)	518	1478 ± 117	(4)	1480
⁵⁰ Cr ₂₆	27.9 ± 1.8	(3)	58	1130 ± 110	(1)	1213
⁵² Cr ₂₈	322 ± 16	(2)	306	1355 ± 165	(1)	1333
⁵⁵ Mn ₃₀	791 ± 44	(4)	575	1530 ± 170	(1)	1432
⁵⁶ Fe ₂₈	10.7 ± 2.2	(6)	23	1698 ± 170	(1)	1644
⁵⁶ Fe ₃₀	500 ± 40	(1)	384	1700 ± 100	(1)	1699
⁵⁹ Co ₃₂	751 ± 37	(3)	526	1440 ± 88	(5)	1549
⁵⁸ Ni ₃₀	34.8 ± 1.7	(9)	42	1574 ± 100	(1)	1424
⁶³ Cu ₃₄	501 ± 36	(14)	481	1318 ± 90	(1)	1405
⁶⁵ Cu ₃₆	961 ± 34	(15)	752	1695 ± 64	(4)	1567
⁶⁴ Zn ₃₄	170 ± 19	(9)	293	1808 ± 90	(5)	1654
⁶⁶ Zn ₃₆	639 ± 60	(4)	586	1733 ± 146	(6)	1559
⁷⁰ Zn ₄₀	1307 ± 130	(1)	990	1941 ± 231	(6)	1465
⁶⁹ Ga ₃₈	1007 ± 45	(4)	716	1975 ± 160	(3)	1719
⁷¹ Ga ₄₀	834 ± 88	(2)	913	1952 ± 142	(3)	1774
⁷⁰ Ge ₃₈	614 ± 34	(4)	471	1402 ± 82	(4)	1334
⁷⁶ Ge ₄₄	1171 ± 126	(3)	1104	1500 ± 300	(1)	1779
⁷⁵ As ₄₂	1060 ± 41	(7)	1011	1739 ± 80	(1)	1626
⁷⁴ Se ₄₀	413 ± 20	(3)	373	1855 ± 140	(1)	1660
⁸² Se ₄₈	1341 ± 135	(4)	1218	1699 ± 118	(2)	1837
⁷⁹ Br ₄₄	950 ± 66	(5)	919	1982 ± 178	(1)	1583
⁸¹ Br ₄₆	1075 ± 79	(6)	1023	2115 ± 190	(1)	1659
⁷⁸ Kr ₄₂	245 ± 20	(1)	426	1911 ± 110	(4)	1814
⁸⁰ Kr ₄₄	810 ± 60	(1)	648	1870 ± 300	(1)	1654
⁸⁵ Rb ₄₈	1079 ± 112	(6)	654	1965 ± 155	(1)	1786
⁸⁷ Rb ₅₀	1199 ± 156	(5)	1109	1895 ± 133	(1)	1892
⁸⁴ Sr ₄₆	412 ± 40	(3)	591	1894 ± 101	(3)	1815
⁸⁶ Sr ₄₈	545 ± 33	(1)	811	2080 ± 110	(1)	1784
⁸⁹ Y ₅₀	819 ± 150	(4)	783	1179 ± 336	(2)	1939
⁹⁰ Zr ₅₀	747 ± 30	(4)	657	1788 ± 134	(3)	1868
⁹³ Nb ₅₂	1350 ± 250	(1)	1018	860 ± 60	(1)	1782
⁹² Mo ₅₀	199 ± 29	(10)	173	2102 ± 76	(2)	1841
¹⁰⁰ Mo ₅₈	1560 ± 161	(6)	1306	2160 ± 120	(1)	1892
⁹⁶ Ru ₅₂	650 ± 90	(4)	687	1675 ± 168	(1)	1965
⁹⁸ Ru ₅₄	1169 ± 96	(1)	883	2097 ± 132	(3)	2032
¹⁰⁴ Ru ₆₀	1443 ± 85	(2)	1297	2042 ± 125	(3)	1965
¹⁰³ Rh ₅₈	933 ± 46	(2)	1123	1876 ± 144	(2)	2012
¹⁰² Pd ₅₆	637 ± 45	(1)	834	2031 ± 113	(2)	2064
¹¹⁰ Pd ₆₄	1735 ± 116	(2)	1366	1830 ± 156	(2)	2079
¹⁰⁷ Ag ₆₀	1255 ± 114	(2)	1109	2132 ± 142	(8)	2015

Table 1—continued

Target	Weighted average $\sigma_{\text{tot}}(\text{mb})$	$\sigma_{\text{theo}}(\text{mb})$	Target	Weighted average $\sigma_{\text{tot}}(\text{mb})$	$\sigma_{\text{theo}}(\text{mb})$
$^{196}_{80}\text{Hg}_{116}$	2056 ± 180 (2)	1936	$^{204}_{82}\text{Pb}_{122}$	1901 ± 156 (5)	2044
$^{198}_{80}\text{Hg}_{118}$	2004 ± 119 (4)	1992	$^{209}_{83}\text{Bi}_{126}$	2484 ± 178 (2)	2113
$^{204}_{80}\text{Hg}_{124}$	2130 ± 89 (5)	2125	$^{232}_{90}\text{Th}_{142}$	1250 ± 57 (3)	
$^{203}_{81}\text{Tl}_{122}$	1917 ± 148 (4)	2068	$^{238}_{92}\text{U}_{146}$	690 ± 40 (1)	
$^{205}_{81}\text{Tl}_{124}$	1636 ± 111 (2)	2109			

(iii) ^{64}Zn , ^{84}Sr , ^{85}Rb , ^{130}Te and ^{176}Yb . In these cases the theoretical value agrees with one or more of the reported values even though the weighted average differs from the predicted value by more than 30%. Experimental values that agree with the theoretical values are given below for these nuclides together with the reference.

^{64}Zn	224 ± 45 mb (Paul and Clarke 1953)
^{84}Sr	482 ± 80 mb (Rao <i>et al</i> 1971)
^{85}Rb	687 ± 74 mb (Strohal <i>et al</i> 1962)
^{130}Te	1800 ± 120 mb (Bormann <i>et al</i> 1970)
	1455 ± 54 mb (Lu <i>et al</i> 1970)
^{176}Yb	1810 ± 127 mb (Spenke 1964).

6. Shell effects

It was pointed out by Qaim (1972) that the claim of Chattarjee and Chattarjee (1969) regarding the existence of shell effects in $(n, 2n)$ reactions at 14–15 MeV is based on the low experimental cross section values used in the case of ^{88}Sr , ^{93}Nb , ^{109}Ag , ^{128}Te , ^{130}Te , ^{136}Ba and ^{151}Eu ; this appears to be true as the values calculated by us agree with the observed total $(n, 2n)$ cross sections for ^{93}Nb ($\sigma_{\text{obs}} = 1350 \pm 250$ mb compared to $\sigma_{\text{cal}} = 1018$ mb), ^{128}Te ($\sigma_{\text{obs}} = 1689 \pm 127$ mb compared to $\sigma_{\text{cal}} = 1586$ mb), ^{130}Te ($\sigma_{\text{obs}} = 1800 \pm 120$ mb reported by Bormann *et al* (1970) compared to $\sigma_{\text{cal}} = 1644$ mb) and ^{151}Eu ($\sigma_{\text{obs}} = 1739 \pm 80$ mb compared to $\sigma_{\text{cal}} = 1626$ mb); total $(n, 2n)$ cross sections for ^{88}Sr , ^{109}Ag and ^{136}Ba are not yet available for comparison; for these nuclides, we calculate the total $(n, 2n)$ cross section as 912 mb, 1229 mb and 1577 mb, respectively with an error of about 10%; future measurements may test these predictions.

Furthermore, an inspection of table 1 shows that our calculated values agree well with the observed values even for closed neutron-shell target nuclei; the disagreement in the case of $^{39}_{19}\text{K}_{20}$ and $^{54}_{26}\text{Fe}_{28}$ is due to their high $(n, 2n)$ thresholds as already pointed out; the predicted value for $^{142}_{60}\text{Nd}_{82}$ agrees with the value 1500 ± 150 mb reported by Bormann *et al* (1970) though not with the weighted average value. Hence, we conclude that there are no visible consistent shell effects in 14–15 MeV $(n, 2n)$ cross sections reported to date in the literature.

7. Conclusions

Pearlstein's method of calculating $(n, 2n)$ cross sections at 14–15 MeV incident neutron energy has been improved by taking into account proper values for the constants; using

our (Kondaiah and Athougies 1974) constants k and m in equation (5), one can make reliable predictions of $(n, 2n)$ cross sections for nuclides in the region $A = 23$ to 209. There is no tangible evidence of shell effects in the $(n, 2n)$ cross sections at $E_n = 14$ to 15 MeV. The weighted averages of the experimental cross sections reported to date as well as the theoretical predictions are given in table 1.

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