Pramāṇa, Vol. 17, No. 1, July 1981, pp. 99–104. © Printed in India.

Pre-equilibrium effects in $(n, 2n)$ reactions at 14·2 MeV+

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MS received 12 January 1981; revised 9 June 1981

Abstract. With a view to study the pre-equilibrium effects in neutron-induced reactions, the activation cross-sections for $(n, 2n)$ reactions at 14·2 ± 0·2 MeV in the heavy mass region have been measured using the versatile mixed powder technique and high resolution Ge(Li) detection. The experimental cross-sections are found to be consistently smaller than the predictions based on the statistical theory and this is attributed to the effect of pre-equilibrium decay in these reactions. The cross-sections due to pre-equilibrium decay were estimated using exciton, hybrid and unified models. When this cross-section was included in comparing the experimental cross-sections with theory, we obtained better agreement, within the limitations of the present-day preequilibrium theories.

Keywords. Activation cross-sections; mixed powder technique; statistical theory estimates; pre-equilibrium decay; neutron-induced reactions.

1. Introduction

The advent of isochronous cyclotrons in the last few decades made possible the study of excitation functions and particle spectra in the energy range 20-200 MeV in a variety of charged particle-induced reactions. Certain new experimental features which emerged from these studies defied explanation in terms of well-known direct interaction (Austern 1963) or compound nucleus (Böhr 1936) models. Consequently a new reaction model (Griffin 1966) known as the ' pre-equilibrium model ' (hereafter referred to as PE model) has been proposed to explain some of these experimental features. The basic concept of the new model is the finite probability for particle emission during the pre-equilibrium phase of a nuclear reaction and hence the name.

Whereas this model has been thoroughly tested in charged particle-induced reactions (Blann 1975), only a few attempts (Braga-Marcazzan *et al* 1972; Augustyniak *et al* 1977) have been made in the area of neutron-induced reactions. Typical calculations (Holub and Cindro 1975; Seidel *et al* 1976) have shown that appreciable precompound effects can be anticipated even in 14 MeV neutron-induced reactions in as much as the compound system is raised to excitation energies of the order of 20-25 MeV. A characteristic feature of precompound emission is that the emitted

+Preliminary results of this work were reported at the Nuclear Physics and Solid State Physics Symposium, Pune (1978).

nucleons carry away relatively large amounts of excitation energy. In fast neutron-induced reactions, for example, it may happen that after the first neutron emission in the PE phase, the residual nucleus may be left at such low excitation that a second neutron emission is energetically forbidden. This would detract from the $(n, 2n)$ cross-section. In an activation measurement of $(n, 2n)$ cross-sections where the product nuclei are identified, the evident result is a considerable decrease in the experimentally measured cross-section as compared to what is expected on the basis of pure evaporation from an equilibrium compound nucleus. This decrease, as pointed out earlier, can be due to the PE contribution. Using the PE models (Blann 1975; Ernst and Rama Rao 1977), the extent of this contribution can be evaluated. When this is included in the cross-section estimated on the basis of pure evaporation and compared with the experimental cross sections, one has to observe a better agreement.

With this end in view, a systematic investigation of $(n, 2n)$ reaction cross-sections in the heavy mass region, where PE contribution is appreciable, has been undertaken in the present work using the activation method. The versatile mixed powder technique (Venugopala Rao and Fink 1967) and high resolution Ge(Li) detection were used to make precision measurements of the cross-sections.

2. Experimental procedure and results

2.1 *Irradiation and monitor reactions*

The neutron irradiations were carried out at the 600 keV Cockcroft-Walton accelerator of the Andhra University. Thick tritium targets, supplied by Bhabha Atomic Research Centre, Bombay (India), were used to produce 14 MeV neutrons through $T(d, n)\,\alpha$ reaction. All the irradiations were performed at 90° to the incident deuteron beam such that the neutrons irradiating the samples had their energy, $14{\cdot}2 \pm 0{\cdot}2$ MeV. The neutron flux was of the order of $10^8 n/\mathrm{cm}^2/\mathrm{sec}$. The constancy of the flux was monitored by an auxiliary BF_3 counter embedded in a paraffin block. The samples for irradiation were prepared by mixing metallic powder or oxide of the target material of natural isotopic composition and $> 99{\cdot}9\%$ pure, (supplied by Koch-Light Labs., E. Merck Co.) with monitor substance in 'specpure' form. The mixed powder was encapsulated in thin-walled ($< 0{\cdot}5$ mm thick) perspex discs of 3·3 cm in diameter. The thickness of the wafer was 1 or 2 mm in all cases. The following reactions served as monitors:

1. $^{27}\mathrm{Al}\,(n, \alpha)\,^{24}\mathrm{Na}$, $T_{1/2} = 15\mathrm{h}$, $\sigma = 115{\cdot}5 \pm 3$ mb (Robertson *et al* 1973)
2. $^{27}\mathrm{Al}\,(n, p)\,^{27}\mathrm{Mg}$, $T_{1/2} = 9{\cdot}5\mathrm{m}$, $\sigma = 72 \pm 5$ mb (our value)
3. $^{63}\mathrm{Cu}(n, 2n)\,^{62}\mathrm{Cu}$, $T_{1/2} = 9{\cdot}7\mathrm{m}$, $\sigma = 593 \pm 36$ mb (Qaim 1972)
4. $^{65}\mathrm{Cu}\,(n, 2n)\,^{64}\mathrm{Cu}$, $T_{1/2} = 12{\cdot}7\mathrm{h}$, $\sigma = 926 \pm 60$ mb (Qaim 1972)

2.2 *Counting and errors*

The gamma activity of the reaction products was measured using a 35 cc coaxial Ge(Li) detector (FWHM: 4·6 at 1332 keV) in conjunction with a ND 512 channel analyser system. Areas of the photopeaks of interest were determined with the help of a computer program which corrects for a linear background and makes a Gaussian fitting of the photopeaks. The relative photopeak efficiency of the detector, corrected for self-absorption and scattering of gamma rays within the sample, was calibrated by the simulation technique, using standard radioactive sources, ^{75}Se, ^{133}Ba and ^{152}Eu. The activation cross-sections for the reaction was evaluated using the standard formula.

The errors in the present cross-section measurements are the root-mean-square errors and are composed of the following: (i) the relative photopeak efficiency of the detector, the error in which was $\sim 3\%$, (ii) the error in the determination of the photopeak areas which was the standard deviation in counting statistics, (iii) the errors in weighing and mixing of the samples, duration of bombardment and transport of irradiated samples were negligible. The errors in the monitor cross-sections, half-lives, and absolute gamma ray abundances were not included but the spectroscopic data used are shown in table 1. This is because any revision in the decay schemes and conversion coefficient values permits easy recalculation of the cross-section in future.

2.3 *Experimental results*

The cross-sections measured in the present work are presented in table 1 together with the spectroscopic data used in the evaluation of the cross-section. 'θ' is the absolute gamma ray abundance in photons per disintegration.

A comparison of the present measurements with those reported in the literature (CINDA 78/79) showed a general agreement with those who employed similar technique, the main emphasis in the present work being on precision and uniformity in measurement.

Table 1. Experimental $(n, 2n)$ reaction cross-sections measured in the present work at 14·2 ± 0·2 MeV

Reaction	Decay data of the product nuclide[a]			Measured cross-section (mb)
	$T_{1/2}$	E_r (keV)	θ	
^{142}Nd$(n, 2n)^{141m}$Nd	62 sec	756	0·92[b]	515 ± 57
^{142}Nd$(n, 2n)^{141g}$Nd	2·5 h	511	0·06[c]	1220 ± 73
^{160}Gd$(n, 2n)^{159}$Gd	18 h	364	0·1	1868 ± 95
^{162}Er$(n, 2n)^{161}$Er	3·2 h	827	0·61	1886 ± 240
^{168}Yb$(n, 2n)^{167}$Yb	17·7 m	113	0·90	1948 ± 195
^{170}Yb$(n, 2n)^{169}$Yb	32 d	198	0·36	2037 ± 122
^{176}Hf$(n, 2n)^{175}$Hf	70 d	343	0·85	2124 ± 128

[a] Taken from Bowman and Mac Murdo (1974), unless otherwise stated.
[b] Lederer and Shirley (1978)
[c] Grissom *et al* (1966).

3. Comparison of experimental results with theory

The experimental total $(n, 2n)$ cross-sections measured in the present work are compared with the theoretical estimates based on statistical theory (Blatt and Weiskopf 1952) and are shown in column 3 of table 3. The statistical model cross-sections are calculated using the inverse cross-sections evaluated using Blann's code ' Overlaid Alice ' (Blann 1976). The computations were performed on IRIS 55 computer at ECIL, Hyderabad. The Q-values were taken from Gove and Wapstra (1972). It should be mentioned that the statistical model estimates are sensitive to the choice of the level density parameter. Wide ranging values from A/8 to A/20 have been used by different authors on an empirical basis. To obviate this uncertainty, in the present work, experimentally deduced values of the level density parameter, compiled by Holmes *et al* (1976) have been used.

It can be seen from table 3 that invariably in all cases the experimental cross-sections are lower than the theoretical estimates. As mentioned earlier, this diminution can be attributed to the PE effects in $(n, 2n)$ reactions at 14 MeV. The PE contribution as such, *i.e.*, both the neutrons being emitted in pre-equilibrium phase is also quite unlikely since the available excitation energy is limited and each PE neutron tends to take away a large chunk of the energy. The observed cross-section mainly comes from the compound nucleus evaporation, which, however, is reduced due to competition from the non-compound effects.

Ever since Griffin (1966) propounded the exciton model, several groups (Harp *et al* 1968, Harp and Miller 1971, Blann 1971, 1972) have concentrated on PE calculations. Blann proposed the hybrid model and more recently Ernst and Rama Rao (1977) proposed the unified model which bridges the gap of ideological differences between exciton and hybrid models. They have also developed a comprehensive computer code which calculates the PE share of the total reaction cross-section based on the above three models. Using this code, the calculations were performed on an IBM 1130 computer of the Andhra University and the PE contributions in all the reactions were estimated. A two particle—one hole configuration was assumed, thus giving a value of two for the initial exciton number (n_0). Table 2 gives the cross-

Table 2. Cross-sections due to PE decay in $(n, 2n)$ reactions at 14·2 MeV.

Reaction	Excitation energy of the compound system (MeV)	Cross-section due to PE decay based on			
		Exciton model (mb)	Hybrid model (mb)	Unified exciton model (mb)	Unified hybrid model (mb)
$^{121}Sb(n, 2n)^{120}Sb$	20·89	99	115	104	105
$^{123}Sb(n, 2n)^{122}Sb$	20·55	100	117	107	108
$^{142}Nd(n, 2n)^{141}Nd$	20·25	102	121	111	112
$^{160}Gd(n, 2n)^{159}Gd$	19·74	108	129	117	118
$^{162}Er(n, 2n)^{161}Er$	21·02	93	109	101	102
$^{168}Yb(n, 2n)^{167}Yb$	20·98	92	109	100	101
$^{170}Yb(n, 2n)^{169}Yb$	20·74	94	111	102	103
$^{176}Hf(n, 2n)^{175}Hf$	20·50	105	120	110	111

section values due to PE decay together with the excitation energies of the compound systems. These cross-sections are obtained by compounding the differential cross-sections over and above the threshold energy of the reaction (in the present set of reactions, it is the binding energy of the neutron).

It can be noticed from column 2 of table 2 that the excitation energy of the compound system in all the reactions is more or less the same, being around 20 MeV. As pointed out by Blann (1975), the PE characteristics mainly depend on the nature of the incident particle and the excitation energy of the compound system, irrespective of the details of the target nucleus. Taking the unified exciton model as a typical example, it is clearly observed that the cross-section due to PE decay is more or less constant in this mass region.

The total $(n, 2n)$ cross-sections measured in the present work are compared with the statistical theory estimates in which the contribution due to PE decay is subtracted, and are given in column 4 of table 3. The PE cross-sections based on the unified exciton model are taken into consideration. It is observed that the agreement between theory and experiment is improved, though a slight deviation still persists. This slight deviation is due to some inherent deficiencies in the PE models: (i) the choice of the initial exciton number is left arbitrary, although the calculated result depends very strongly on it. The selection of a particular value for the initial exciton number is usually guided by the goodness of the fit between theory and experiment in the case of other reactions induced by the same particle. In the case of neutron-induced reactions at 14 MeV such guidance is scarcely available. The value of $n_0=2$ used in the present calculations is inspired by the proton-induced reactions. (ii) In the present theories, the internal decay rates are derived from free nucleon-nucleon scattering data corrected for the Pauli effect. Also, the Fermi gas or the equidistant spacing models are used in evaluating state densities. Hence the theoretical computations represent only ' nuclear matter calculations ' which do not take into account the nuclear structure effects.

Judging from the state-of-art of the present day PE theories, the above agreement between theory and experiment is considered very reasonable.

Table 3. Comparison of experimental $(n, 2n)$ cross-sections with the theoretical estimates.

Reaction	σ expt (mb)	σ St. theory (mb)	σ theory (mb) (PE corrected)
^{121}Sb$(n, 2n)$ ^{120}Sb	1599 ± 75[a]	1840	1736
^{123}Sb$(n, 2n)$ ^{122}Sb	1692 ± 85[a]	1861	1754
^{142}Nd$(n, 2n)$ ^{141}Nd	1735 ± 93	1943	1832
^{160}Gd$(n, 2n)$ ^{159}Gd	1868 ± 95	2193	2076
^{162}Er$(n, 2n)$ ^{161}Er	1886 ± 240	2167	2066
^{168}Yb$(n, 2n)$ ^{167}Yb	1948 ± 195	2214	2114
^{170}Yb$(n, 2n)$ ^{169}Yb	2037 ± 122	2246	2144
^{176}Hf$(n, 2n)$ ^{175}Hf	2124 ± 128	2286	2176

[a]Lakshmana Das *et al* (1978).

Acknowledgements

The authors are thankful to Professor V Lakshminarayana and Dr K Parthasaradhi for their interest in the present work. One of the authors (NLD) acknowledges the receipt of a fellowship from the University Grants Commission, New Delhi.

References

Augustyniak W, Herman M, Marcinkowski A and Zwieglinski B 1977 *Nucl. Phys.* **A285** 145

Austern N 1963 in *Fast neutron physics* (eds.) J B Marion and J L Fowler, Part 2 (New York: John Wiley), p. 1113, and related references quoted in it.

Blann M 1971 *Phys. Rev. Lett.* **27** 337

Blann M 1972 *Phys. Rev. Lett.* **28** 757

Blann M 1975 *Ann. Rev. Nucl. Sci.* **25** 123

Blann M 1976 *Overlaid Alice*, Report (00-3494-72) with several additional corrections

Blatt J M and Weiskopf V F 1952 *Theoretical nuclear physics*, (New York: John Wiley), p. 484

Böhr N 1936 *Nature (London)* **137** 344

Bowman W W and Mac Murdo K W 1974 *Atomic and Nuclear Data Tables* **13** 89

Braga-Marcazzan G M, Gadioli-Erba E, Millazzo-Colli L and Sona P G 1972 *Phys. Rev.* **C6** 1398

CINDA 78/79 1978 *An index to the literature on microscopic neutron data*, (IAEA, Vienna)

Ernst J and Rama Rao J 1977 *Z. Phys.* **A281** 129

Gove N B and Wapstra A H 1972 *Nucl. Data Tables* **11** 127

Griffin J J 1966 *Phys. Rev. Lett.* **17** 478

Grissom J T, Koehler D R and Alford W L 1966 *Phys. Rev.* **142** 725

Harp G D, Miller J M and Berne B J 1968 *Phys. Rev.* **165** 1166

Harp G D and Miller J M 1971 *Phys. Rev.* **C3** 1847

Holmes J A, Woosley S E, Fowler A William and Zimmerman B A 1976 *Atomic and Nuclear Data Tables* **18** 305

Holub E and Cindro N 1975 *Phys. Lett.* **B56** 143

Lakshmana Das N, Srinivasa Rao C V, Thirumala Rao B V and Rama Rao J 1978 *Pramana* **11** 595

Lederer C M and Shirley V S (ed.) 1978 *Table of Isotopes* (New York: John Wiley)

Qaim S M 1972 *Nucl. Phys.* **A185** 614

Robertson J C, Audric B and Kolkowskii P 1973 *J. Nucl. Energy* **27** 531

Seidel K, Seelinger D and Meister A 1976 *Sov. J. Nucl. Phys.* **23** 39

Venugopala Rao P and Fink R W 1967 *Phys. Rev.* **154** 1023