Rare Nuclear Reactions Induced by 14.7-MeV Neutrons*

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Activation cross sections for 14.7±0.2-MeV neutrons were measured for $[(n,n\alpha)+(n,\alpha n)]$ reactions with Cu^{65}, Zn^{70}, Ga^{71}, and Nb^{93}, while upper limits were set for this reaction for V^{51}, Ge^{76}, Br^{81}, Rb^{87}, Ag^{107}, Ag^{109}, In^{115}, Au^{197}, and Tl^{203}. Cross-section limits also were set for $(n,2p)$ reactions on Si^{29}, K^{41}, Sc^{45}, Ti^{50}, V^{51}, Mn^{55}, As^{75}, Y^{89}, Nb^{93}, Cs^{133}, La^{139}, Pr^{141}, and Tb^{159}; and for (n,He^3) reactions on Sc^{45}, Nb^{93}, Au^{197}, and Tl^{205}. Cross sections were determined for (n,γ) reactions on Y^{89}, Nb^{93}, and Pr^{141}, and upper limits were set for $(n,3n)$ reactions with Pr^{141}, Au^{197}, and Tl^{203}. The $[(n,np)+(n,pn)]$ reaction was detected with Ni^{58}, but not with Mo^{92}. A value of 520±120 mb was established for the former, and an upper limit set for the latter. Absolute disintegration rates were obtained by both beta and gamma spectroscopy counting methods. Extensive use of radiochemical separation was made in order to isolate the low-yield rare reaction products from large target samples. Statistical theory was employed (using parameters selected from a critical analysis of the literature) to make theoretical cross section estimates for many of the reactions studied. The theoretical cross sections for $(n,\alpha n)$ reactions agree remarkably well with the experimentally determined $[(n,n\alpha)+(n,\alpha n)]$ cross section sums, thereby suggesting that the path for this reaction may be predominantly the $(n,\alpha n)$ process. For certain cases, statistical theory predicts appreciable $(n,2p)$ cross sections at 14.7 MeV; e.g., for Cr^{50}, Ni^{58}, Kr^{78}, and Mo^{92}. It is suggested that in studies of emitted proton spectra which show an "excess" of low-energy protons, a contribution may be present from the $(n,2p)$ as well as the (n,np) reaction. A new gamma at 176±4 keV in about 5% of 1.8-day Sc^{48} decays is confirmed.

1. INTRODUCTION

THERE exist for most nuclei, in addition to $(n,2n)$, (n,p), and (n,α) reactions, others which are energetically possible with 14–15 MeV neutrons, but which usually are more improbable, and as a consequence they have not been investigated as thoroughly. These "rare" reactions include (n,d), (n,np), (n,pn), (n,t), (n,dn), $(n,2p)$, (n,He^3), (n,γ), $(n,n\alpha)$, $(n,\alpha n)$, and $(n,3n)$ processes. There are several reasons why these are interesting. The cross sections are useful in testing nuclear reaction theories in this energy region, and they are important in interpreting energy and angular distributions obtained in the study of emitted particles, since in many cases the spectra of emitted protons or alphas have a somewhat distorted Maxwellian shape, and occasionally exhibit an "excess" of low-energy particles, due to contributions from such competing reactions as $[(n,np)+(n,pn)+(n,d)]$, $(n,2p)$, and $[(n,n\alpha)+(n,\alpha n)]$. In connection with an understanding of reaction mechanisms in this energy region, it is generally thought that "clustering" should occur in the diffuse nuclear surface. Thus, it is of interest to examine the relative emission of such clusters as H^2, H^3, He^3, He^4, He^6 in nuclear reactions; for example, with 14.7-MeV neutrons.

Also, due to the increasing use of fast neutron activation analysis for determining trace impurities (with sensitivities often down to parts per billion), it is important to know what interferences might be present from possible contributions from rare reactions. Finally, knowledge of cross sections for these rare reactions is an aid in the proper mass assignment of new activities found in fast neutron studies; especially as the cross sections on occasion are quite large, as for certain of the $[(n,np)+(n,pn)+(n,d)]$ group at 14–15 MeV.

Thus, in the present work we have searched for examples of rare reactions with 14.7-MeV neutrons by means of very sensitive activation techniques. The large output of 14.7-MeV neutrons from the Arkansas 400-kV Cockcroft-Walton accelerator, (presently as high as 3×10^{11} DT neutrons/sec from a new TiT target), and the versatile two-crystal NaI(Tl) scintillation spectrometer with 200-channel analyzer provided the means for a very sensitive search for low-yield reactions. Extensive use of radiochemical separations from large target samples was made to identify and count the reaction products.

In Table I we have summarized cross-section values from the literature[1–36] for (n,d), (n,np), and $[(n,p\gamma)$

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[1] F. L. Ribe, Phys. Rev. **87**, 205 (1952).
[2] G. M. Frye, Jr., Phys. Rev. **93**, 1086 (1954).
[3] D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (2nd ed.) (1958); D. J. Hughes, B. A. Magurno, and M. K. Brussel, BNL-325 (2nd ed., Suppl. 1) (1960).
[4] R. J. Howerton, University of California Radiation Laboratory Report UCRL-5420, 1958, (unpublished).
[5] M. E. Battat and F. L. Ribe, Phys. Rev. **89**, 80 (1953).
[6] G. M. Frye and J. H. Gammel, Phys. Rev. **103**, 328 (1956).
[7] F. L. Ribe and J. D. Seagrave, Phys. Rev. **94**, 934 (1954).
[8] A. B. Lillie, Phys. Rev. **87**, 716 (1952).
[9] G. E. Velyukhov, A. N. Prokofev, and S. V. Starodubtsev, Dokl. Akad. Nauk SSSR **4**, 837 (1960) [translation: Soviet Phys.—Doklady **4**, 781 (1960)].
[10] G. E. Velyukhov, A. N. Prokofev, and S. V. Starodubtsev, Zk. Eksperim. i Teor. Fiz. **39**, 563 (1960) [translation: Soviet Phys.—JETP **12**, 395 (1961)].
[11] D. L. Allan, Proc. Phys. Soc. (London) **A70**, 195 (1957).
[12] L. Colli, M. G. Marcazzan, F. Merzari, P. G. Sona, and F. Tonolini, Nuovo Cimento **16**, 991 (1960).

TABLE I. Cross sections from the literature for (n,d), (n,np), (n,pn), and $(n,p\gamma)$ reactions at 14–15 MeV.

Target nuclide	Method of measurement	Neutron energy (MeV)	Cross sections[a] (n,d)	(n,np)	$[(n,p\gamma)+(n,pn)]$ Compound nucleus	Direct interaction	Reference
Li^6	telescope	14.2	~140				1
Li^6	emulsion	14	166±15				2
Li^6	not reported	14	89±10				3
Li^6	prop. counter	14.1	~200				4
Li^7	emulsion	14	(observed no value)				2
Li^7	not reported	14	9.8±1.1				3
Li^7	activation of 0.83-sec He^6	14.2	~200				5
B^{10}	emulsion	14	128±19				6
B^{10}	telescope	14	21±3				7
N^{14}	cloud chamber	14.1	<100				8
O^{16}	cloud chamber	14.1	15±5				8
F^{19}	telescope	14.1	21.4±1.1*				9
Ne^{20}	telescope	14.1	5*				9
Al^{27}	emulsion	14		70	70		11
Al^{27}	telescope	14.8	30±4	157	89	27	36
Al^{27}	emulsion	13.2			70	80	13
Al^{27}	emulsion	14		53±11			14
Si^{28}	emulsion	14		27±22	243±22		14
P^{31}	telescope	14.1	21.8±1.2*				9
P^{31}	telescope	14.1	14.5±3				12
P^{31}	emulsion	14		163±14	184±14		14
S^{32}	telescope	14.1	20.4±1.5*				10
S^{32}	emulsion	14		105±25	365±25		14
S^{32}	emulsion	14.4		$0.33\sigma_{(n,p)}$			15
S^{32}	telescope	14.1	14±4				12
K^{39}	scintillation crystal	14		186±28	354±54		16
Ca^{40}	emulsion	14		205±38			14

+ (n,pn)] reactions at 14–15 MeV. Other rare reactions from the literature are discussed in the text. From the present investigation, cross sections and upper limits at 14.7 MeV are tabulated (Tables II, III, IV, and V). Finally, in the course of these studies, a number of (n,p), (n,α), and $(n,2n)$ cross sections were measured; these are summarized (Table VI) together with literature values. It needs to be pointed out, in connection with Table I, that in the analysis of emitted proton spectra from fast neutron reactions, the (n,p) cross section cannot be determined directly, for the residual nucleus may have sufficient excitation energy to evaporate a neutron to give the (n,pn) reaction. Instead, one determines the $[(n,p\gamma)+(n,pn)]$ sum, and it is these sums which are given in Table I (Column 6).

2. EXPERIMENTAL

Monoenergetic neutrons were produced by the $H^3(d,n)He^4$ reaction on the Arkansas 400-kV Cockcroft-Walton accelerator. With a maximum subtended angle of 40° from the beam direction, it is estimated that more than 80% of the neutrons passing through the sample have energies of 14.7±0.2 MeV. Monitor disks of 0.00025-in. copper foil or 0.001-in. aluminum foil were cut into 0.5-in. diam and placed in front and in back of the sample. Measurements of very short-lived activities could also be carried out with the aid of a pneumatic transport system, consisting of 0.5-in.-diam polyethylene tubing[37] with polyethylene capsules. The

[13] G. Brown, G. C. Morrison, H. Muirhead, and W. T. Morton, Phil. Mag. **2**, 785 (1957).
[14] D. L. Allan, Nucl. Phys. **24**, 274 (1961).
[15] B. Antolkovic, Nuovo Cimento **22**, 853 (1961).
[16] M. Bormann, H. Jeremie, G. Andersson-Lindström, H. Neuert, and H. Pollehn, Z. Naturforsch. **15a**, 200 (1960).
[17] I. Slaus, P. Tomas, and N. Stipcic, Nucl. Phys. **22**, 692 (1961).
[18] P. Avignon and L. Rosier, Compt. Rend. **247**, 1849 (1958).
[19] P. V. March and W. T. Morton, Phil. Mag. **3**, 143 (1958).
[20] D. L. Allan, Nucl. Phys. **10**, 348 (1959).
[21] D. M. Chittenden, D. G. Gardner, and R. W. Fink, Phys. Rev. **122**, 860 (1961).
[22] K. H. Purser and E. W. Titterton, Australian J. Phys. **12**, 103 (1959).
[23] I. Kumabe and R. W. Fink, Nucl. Phys. **15**, 316 (1960).
[24] R. N. Glover and E. Weigold, Australian National University Report, ANU/P-236, 1961, (unpublished).
[25] R. N. Glover and K. H. Purser, Nucl. Phys. **24**, 431 (1961).
[26] R. S. Storey, W. Jack, and A. Ward, Proc. Phys. Soc. (London) **75**, 526 (1960).
[27] W. G. Cross, R. L. Clarke, K. Morin, G. Slinn, N. M. Ahmed, and K. Beg, Bull. Am. Phys. Soc. **7**, 335 (1962).
[28] P. V. March and W. T. Morton, Phil Mag. **3**, 577 (1958).
[29] I. L. Preiss and R. W. Fink, Nucl. Phys. **15**, 326 (1960).
[30] L. Colli, U. Facchini, I. Iori, M. G. Marcazzan, and A. M. Sona, Nuovo Cimento **13**, 730 (1959).
[31] A. V. Cohen, S. B. Hyder, and P. H. White, Nucl. Phys. **1**, 278 (1956).
[32] C. H. Reed, Ph.D thesis, University of Utah, 1960 (unpublished).
[33] J. F. Barry, R. F. Coleman, B. E. Hawker, and J. L. Perkin, Proc. Phys. Soc. (London) **74**, 632 (1959).
[34] A. D. Poularikas, University of Arkansas Annual AEC Report, 1960, (unpublished).
[35] F. L. Ribe, Phys. Rev. **106**, 767 (1957).
[36] R. N. Glover and F. Weigold, Nucl. Phys. **24**, 630 (1961).

[37] Kindly supplied by Phillips Petroleum Company, Bartlesville, Oklahoma.

TABLE I (*Continued*).

Target nuclide	Method of measurement	Neutron energy (MeV)	Cross Sections[a] (n,d)	(n,np)	[(n,pγ)+(n,pn)] Compound nucleus	Direct interaction	Reference
V^{51}	scintillation crystal	14.4			27		17
Cr^{50}	emulsion	14		153±21	265±21		14
Mn^{55}	emulsion	14			110±15		18
Fe^{54}	emulsion	14		218±13	382±13		14
Fe^{54}	emulsion	13.5		224	306	70	19
Fe^{54}	emulsion	14		220	460		11
Fe^{54}	emulsion	14		195	395	20	20
Fe^{56}	emulsion	14		0	100	90	11
Fe^{56}	emulsion	13.2			70	25	13
Fe^{56}	emulsion	14		35±7	82±7		14
Fe^{56}	emulsion	13.5		0	75	20	19
Fe^{57}	activation of 1.7-min Mn^{56}	14.8		6.1±2.6			21
Ni^{58}	emulsion	14		220	310		11
Ni^{58}	activation of 270-day Co^{57}	14.1		160±40			22
Ni^{58}	emulsion	14.8		150	270	>17	23
Ni^{58}	activation of 270-day Co^{57}	14.8		570±55			24
Ni^{58}	telescope	14.8	25±6	340	430±27	60	25
Ni^{58}	emulsion	15.7			754±15		26
Ni^{58}	emulsion	14		343±27	440±27		14
Ni^{58}	activation of 270-day Co^{57}	14.5		680±80			27
Ni^{60}	emulsion	13.5		68	87	68	28
Ni^{60}	emulsion	14		60	200	40	11
Ni^{60}	emulsion	14		51±9	124±9		14
Ni^{61}	activation of 10.5-min Co^{60m}	14.8		3.8±1.0			29
Ni^{62}	activation of 1.7-h Co^{61}	14.8		0.65±0.15			29
Ni^{64}	activation of 1.4-h Co^{63}	14.8		0.93±0.04			29
Cu	emulsion	14		128	98	20	20
Cu^{63}	emulsion	14		130	100	20	11
Cu^{63}	telescope	14		181±18	46±5		30
Cu^{63}	emulsion	14		152±9	118±9		17
Cu^{65}	emulsion	14		<40	<40		11
Zn^{64}	emulsion	14		281±18	179±18		14
Zn^{66}	emulsion	14		50±4	34±4		14
As^{75}	emulsion	14			115±15		18
Se^{80}	activation of 9-min As^{79}	14.5		<0.8			31
Zr^{90}	telescope	14		55	30 to 50		30
Zr^{94}	activation of 10-h Y^{93}	14.1		<0.8±0.1			32
Mo^{92}	telescope	14		112	31		30
Rh^{103}	emulsion	13.2			5	20	13
Pd^{108}	activation of 23-min Rh^{107}	14.5			4.0±0.8		33
Pd^{108}	activation of 23-min Rh^{107}	14.5		<65			31
Ag	telescope	14		0	28		31
Te^{128}	activation of 90-h Sb^{127}	14.5			0.33±0.08		34
Te^{130}	activation of 90-h Sb^{129}	14.5			0.17±0.02		34
CsI[b]	scintillation crystal			6±1	17±3		16
Ce^{142}	activation of 3.8-h La^{141}	14.5		<18			32
Ce^{142}	activation of 3.8-h La^{141}	14.5			1.0±0.2		34
W^{186}	activation of 49-min Ta^{185}	14.5		<1.6			32
W^{186}	activation of 49-min Ta^{185}	14.5			0.11±0.05		34
W^{186}	activation of 49-min Ta^{185}	14.8		1.5±0.8			35

[a] Cross sections are in millibarns, except where indicated by asterisks in which case they are in units of mb/sr at 0°.
[b] Total (n,np) cross section for both Cs^{133} and I^{127}.

transfer time from neutron target to detector is typically 420 msec for a distance of 39 m.

Experience with TiT targets under high-current bombardment with deuterium ion beams in this laboratory shows that the neutron output during constant beam bombardment drops off exponentially with a "half-life" of about 2.5 h, with a typical beam current of about 700 μA (consisting of approximately 40–50% D_3^+ at 125 keV, 40% D_2^+ at 188 keV, 15–20% D^+ at 380 keV, and 5–10% of neutrals which produce about 8% of the neutrons). This half-life is considerably longer at much lower beam currents.

Since cross sections are computed generally on the assumption that the neutron flux is constant during irradiations, it is necessary in some experiments to correct for the change in flux during bombardment. It is readily shown that the ratio of the cross section σ_s', obtained by assuming that the neutron flux decreases exponentially during a bombardment of duration t, to the cross section σ_s, obtained on the assumption of constant neutron flux during bombardment, is

$$\frac{\sigma_s'}{\sigma_s}=\frac{(\lambda_m-\Delta)(e^{-\Delta t}-e^{-\lambda_s t})(1-e^{-\lambda_m t})\lambda_s}{(\lambda_s-\Delta)(e^{-\Delta t}-e^{-\lambda_m t})(1-e^{-\lambda_s t})\lambda_m}, \quad (1)$$

where subscript s refers to the sought nuclide in the sample, and subscript m refers to the nuclide used as a monitor. Δ is defined by

$$f/f^0=e^{-\Delta t}, \quad (2)$$

where f is the flux at time t, and f^0 is the flux at the start of the irradiation. Δ is a constant ($=0.693/2.5$ h^{-1} in the present case).

From Eq. (1), it is obvious that if both λ_s and λ_m are greater than Δ, and if the irradiation time t is short (so that $e^{-\Delta t}\approx 1$), then $\sigma_s'=\sigma_s$, and the flux decrease causes no error in the cross section measurement. Under any other conditions, however, there will be a difference between σ_s' and σ_s. Furthermore, when Δ is greater than λ (of either sample or monitor), no equilibrium is attained (i.e., *the activity does not reach a saturation value*), the activity merely increasing to a maximum and then decreasing with time. The time to reach the maximum, $t_{\max}$, is readily found to be

$$t_{\max}=\frac{\log_e(\lambda/\Delta)}{(\lambda-\Delta)}. \quad (3)$$

Under the usual conditions prevailing in cross section work in this laboratory, where previously the flux average was taken as a measure of the flux during bombardment, this effect might account for variations in cross section values not exceeding some 10 to 20% (well within the experimental error limits) in most cases, nor about 50% in the most extreme cases.

In most of the samples the (n,α), (n,p), or $(n,2n)$ reaction with an isotope of the element under study served as an internal monitor for the flux. This eliminates flux errors arising from geometry differences. In one case, copper powder to serve as a monitor was mixed intimately with a sample powder to obtain similar sample-monitor geometry. Where no internal monitor was suitable, copper and aluminum monitor foils were used.

Due to "masking" of the low-yield rare reaction products by the more probably (n,p), (n,α), and $(n,2n)$ reactions and also to the necessity often of irradiating large, bulky samples (up to 100 g) in order to get a significant yield of the rare reaction products, absolute counting is exceedingly difficult, because of uncertainties in such counting factors as the self-scattering–self-absorption corrections. For these reasons, radiochemical separation and identification of the reaction products was performed whenever possible.

Counting[38] was done in the following ways. Samples decaying mostly by energetic beta emission were counted with an aluminum-walled methane-flow proportional counter with 1.0 mg/cm^2 aluminized Mylar end-window. For counting of very low-energy beta and electron emitters, a windowless, stainless steel, flow-proportional counter using argon-methane gas was used. This counter also is sensitive to soft x rays. For identifying beta end-point energies or to count betas within a given energy interval, a scintillation beta spectrometer was employed, consisting of a 0.5-in.$\times$1.5-in. cylindrical plastic scintillator and 200-channel RIDL transistorized analyzer. Calibration was achieved with conversion electrons from In^{114}(162 keV), Sn^{113}(364 keV), and Cs^{137}(624 keV). For identification of beta end-point energies, samples were evaporated onto thin Mylar film from liquid solution. Gamma spectra were studied by means of a two-crystal 3-in.$\times$3-in. NaI(Tl) scintillation spectrometer[39] and 200-channel analyzer. Counting of low-energy gammas and x rays was also done with a 25-mm$\times$2-mm NaI(Tl) crystal.

When the product of a desired rare reaction could not be observed, an upper limit for its cross section was obtained as follows: For beta-decay analysis, a line was added to the decay curve with a slope corresponding to the half-life of the unobserved product. This added line represented the amount of product activity which would have had to be present in order that a change in the slope of the decay curve would have been evident. Similarly, cross-section limits from gamma spectra were obtained by drawing in photopeaks of gammas which

[38] Counting techniques, in general, used in this laboratory for cross-section measurements by activation methods have been discussed by A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959) and R. G. Wille and R. W. Fink, *ibid.* **118**, 242 (1960); **112**, 1950 (1958). The improvement in the precision of activation cross-section measurements that is possible by the combined use of the technique of cross comparisons between beta and gamma scintillation counting and gamma scintillation coincidence spectrometry has been pointed out by J. Kantele and D. G. Gardner, Nucl. Phys. **35**, 353 (1962), based on work in this laboratory.

[39] J. Kantele and R. W. Fink, Nucl. Instr. Methods **15**, 69 (1962).

TABLE II. Upper limits for $(n,2p)$ cross sections at 14.7 MeV.

Reaction	Q-value (MeV)	Product half-life[a]	Monitor reaction and cross section	(mb)	Reference	Upper limit to $(n,2p)$ cross section (mb)
$Si^{29}(n,2p)Mg^{28}$	−13.39[b]	21.3 h	$Al^{27}(n,\alpha)Na^{24}$	(114)	c	0.50
$K^{41}(n,2p)Cl^{40}$	−14.52[b]	1.4 min	$K^{41}(n,\alpha)Cl^{38}$	(30)	d	0.13
$Sc^{45}(n,2p)K^{44}$	−12.21[e]	22 min	$Sc^{45}(n,\alpha)K^{42}$	(63)	d	0.21
$Ti^{50}(n,2p)Ca^{49}$	−16.63[b]	8.8 min	$Ti^{50}(n,p)Sc^{50}$	(28)	d	0.28
$V^{51}(n,2p)Sc^{50}$	−13.56[b]	1.7 min	$V^{51}(n,p)Ti^{51}$	(55)	d	0.030
$Mn^{55}(n,2p)V^{54}$	−15.7[f]	55 sec	$Mn^{55}(n,\alpha)V^{52}$	(33)	g	0.30
$As^{75}(n,2p)Ga^{74}$	−11.76[e]	7.8 min	$As^{75}(n,\alpha)Ga^{72}$	(93)	d	0.50
$Y^{89}(n,2p)Rb^{88}$	−11.81[b]	18 min	$Al^{27}(n,p)Mg^{27}$	(82)	h	0.030
$Nb^{93}(n,2p)Y^{92}$	−8.81[b]	3.7 h	$Nb^{93}(n,\alpha)Y^{90m}$	(5.9)	d	0.50
$Cs^{133}(n,2p)I^{132}$	−9.15[b]	2.3 h	$Cs^{133}(n,\alpha)I^{130}$	(1.0)	d	0.005
$La^{139}(n,2p)Cs^{138}$	−10.26[b]	32 min	$La^{139}(n,\alpha)Cs^{136}$	(1.87)	i	0.046
$Pr^{141}(n,2p)La^{140}$	−8.21[b]	40.2 h	$Pr^{141}(n,\gamma)Pr^{142}$	(2.3)	d	0.84
$Tb^{159}(n,2p)Eu^{158}$	−10.0[f]	60 min	$Tb^{159}(n,p)Gd^{159}$	(2.2)	d	0.080

[a] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[b] See Ref. 49.
[c] See Ref. 79.
[d] Monitor cross section was determined in the present work (see Table VI).
[e] See Ref. 80.
[f] Value calculated using semi-empirical masses from A. G. W. Cameron, Can. J. Phys. **37**, 44 (1959).
[g] I. Kumabe, E. Takekoshi, H. Ogata, Y. Tsuneoka, and S. Oki, J. Phys. Soc. (Japan) **13**, 325 (1958) and E. Weigold, Australian J. Phys. **13**, 186 (1960).
[h] See Ref. 44.
[i] R. F. Coleman, B. E. Hawker, L. P. O'Connor, and J. L. Perkin, Proc. Phys. Soc. (London) **73**, 215 (1959).

should have been observed from the rare product activity and comparing these with a photopeak from the monitor, after making the usual corrections, e.g., for crystal efficiency and peak-to-total ratio.

Error limits affixed to cross sections in the present work are the probable errors based on a propagation of the estimated error in each term used in computing the cross section; e.g., duration of bombardment, flux decreases during bombardment, activities at end of bombardment, half-lives, sample and monitor weights, *and* monitor cross sections. The estimated error in the activities at the end of bombardment include the propagated errors arising from such correction factors as self-scattering–self-absorption,[38] chemical yields, peak-to-total ratios, conversion coefficients, and counting efficiencies. The errors in weights, half-lives, and duration of bombardment usually were negligible. Absolute counting correction factors were estimated to within 5%, photopeak areas and chemical yields generally to within 10%.

3. EXPERIMENTAL RESULTS

Some of the cases reported here are discussed individually below for special reasons; however, the details of all cases are given in full elsewhere.[40]

3.1 $(n,2p)$ Reactions

No case of a detectable $(n,2p)$ reaction at 14.7 MeV could be observed. A list of the cases studied together with the upper limits of the cross sections determined from a consideration of the sensitivity of each experiment is given in Table II.

The $C^{12}(n,2p)Be^{11}$ (14 sec) reaction also was investigated with targets of lampblack, graphite, redistilled benzene, redistilled cyclohexane, naphthalene, and *n*-heptane, but in each case severe masking from activities of 7.4 sec N^{16} and 2.3 min Al^{28} from (n,p) reactions on traces of oxygen and silicon impurities prevented the detection of 14 sec Be^{11}, so that no conclusion could be reached regarding the $(n,2p)$ reaction with carbon.

In studying the $Tb^{159}(n,2p)Eu^{158}$ (60 min) reaction, the existence of 60 min Eu^{158} rests on only one preliminary report.[41] To check on this, enriched (92.87%) Gd^{158} oxide was irradiated and counted without chemical separation. Activities of 11 min (also found from irradiated Tb_2O_3) and about 51 min (not found in irradiated Tb_2O_3) were resolved in beta decay. Although an assignment of the 11-min activity is not made, presumably the 51-min species belongs to Eu^{158}. Thus, a limit on the $Tb^{159}(n,2p)$ reaction could be set (Table II).

3.2 (n,He^3) Reactions

No case of a detectable (n,He^3) reaction at 14.7 MeV could be observed. Previously[42] upper limits were established for this reaction with Mg^{26}, Al^{27}, P^{31}, K^{41}, V^{51}, Mn^{55}, Co^{59}, Cu^{63}, As^{75}, Zr^{94}, Rh^{103}, and Cs^{133}. In the present work, we have investigated the additional cases Sc^{45}, Nb^{93}, Au^{197}, and Tl^{205} with radiochemical separation of the products. The new results are summarized in Table III.

[40] E. T. Bramlitt, Ph.D thesis, University of Arkansas, 1962, issued as AEC report, TID-16949, 1963, available from Office of Technical Services, U. S. Department of Commerce, Washington 25, D. C.

[41] L. Winsberg, Natl. Nucl. Energy Ser. Div. IV **9**, 1292 (1951).
[42] E. T. Bramlitt, R. W. Fink, D. G. Gardner, and A. Poularikas, Phys. Rev. **125**, 297 (1962).

TABLE III. Upper limits on (n,He^3) reaction cross sections at 14.7 MeV.

Reaction	Q-value (MeV)	Product half-life[a]	Monitor reaction and cross section (mb)		Upper limit (n,He^3) cross section (mb)
$Sc^{45}(n,He^3)K^{43}$	−11.36[b]	22 h	$Sc^{45}(n,\alpha)K^{42}$	(53)[c]	0.30
$Nb^{93}(n,He^3)Y^{91m}$	−7.68[b]	51 min	$Nb^{93}(n,\alpha)Y^{90m}$	(5.3)[c]	0.060
$Au^{197}(n,He^3)Ir^{195}$	−7.40[b]	2.3 h	$Au^{197}(n,\alpha)Ir^{194}$	(0.43)[d]	0.020
$Tl^{205}(n,He^3)Au^{203}$	−8.02[e]	55 sec	$Tl^{205}(n,\alpha)Au^{202}$	(0.75)[c]	0.070

[a] See Ref. 49.
[b] See Ref. 79.
[c] Monitor cross section was determined in the present work (see Table VI and Ref. 40).
[d] R. F. Coleman, B. E. Hawker, L. P. O'Connor, and J. L. Perkin, Proc. Phys. Soc. (London) **73**, 215 (1959).
[e] Value computed from semi-empirical masses from A. G. W. Cameron, Can. J. Phys. **37**, 44 (1959).

3.3 $[(n,n\alpha)+(n,\alpha n)]$ Reactions[43]

The $Cu^{65}(n,n\alpha)Co^{61}$ reaction was detected and reported earlier[42] to have a cross section of 2.3±1.3 mb. Kantele and Gardner[44] have confirmed this with a precision measurement of 2.8±0.3 mb at 14.7 MeV. The excitation function for this reaction also has been studied[45] up to 19.6 MeV.

Table IV lists the targets irradiated to study the $[(n,n\alpha)+(n,\alpha n)]$ reaction. The cross section determined is given for cases in which the reaction was clearly detected; otherwise, the upper limit for the cross section is given.[40]

During the investigation of the $V^{51}(n,n\alpha)Sc^{47}$ reaction, a new gamma was found in the decay of 1.8-day Sc^{48} from the competing (n,α) reaction, having an energy of 176±4 keV and amounting to about 5% intensity.[40] Kantele[46] has confirmed this new gamma at 180±5 keV with intensity 6 to 8% of Sc^{48} decays and has shown that it is the first member of a (180)(1040)(1314)(986) keV quadruple gamma cascade fed in the decay of Sc^{48}. Hillman[47] also has found the 175-keV gamma in 4% of Sc^{48} decays.

The $V^{51}(n,n\alpha)$ reaction also has been studied by Vonach and Münzer[48] (<0.1 mb at 14.1 MeV), and the excitation function has been studied up to 19.6 MeV.[45]

The $Zn^{70}(n,n\alpha)$ Ni^{66} (55 h) reaction was detected with samples as large as 100 g, since Zn^{70} is only 0.63% abundant in nature. Although the activity of radiochemically separated Ni^{66} was quite low, it was possible to follow its gross beta decay for some three half-lives of 55±5 h each, no longer lived activities being detected.

The $Ga^{71}(n,n\alpha)Cu^{67}$ (58 h) reaction was detected from targets of gallium metal and gallium nitrate after radiochemical separation of copper. Gross beta counting of the copper fraction exhibited a 58±2-h half-life. The Cu^{67} was further identified by its two strong gammas at 92 and 182 keV[49,50] and by its beta end point in the region of 0.4–0.6 MeV.

The possibility of producing Cu^{67} from the (n,p) reaction on impurity Zn^{67}, which might have been present, was carefully examined. In such a case, 12.8-h Cu^{64} also would have been produced from the $Zn^{64}(n,p)$ reaction, since Zn^{64} is some 12 times more abundant than Zn^{67} in natural zinc, and its (n,p) cross section is some 8 times greater[51] than that of Zn^{67}. The 12.8-h Cu^{64}, if it had been present, would have been observed readily in gross beta counting. Further proof of its absence was obtained from gamma spectra which showed an absence of 1.34-MeV gammas (present to the extent of 1% in Cu^{64} decay) and of annihilation radiation (from the 19.6% of positrons in Cu^{64} decay). The $Ga^{71}(n,n\alpha)Cu^{67}$ cross section was determined relative to that of $Ga^{69}(n,p)Zn^{69m}$ (14 h). From gross beta counting on nonradiochemically separated gallium, 14-h Zn^{69m} and 58-h Cu^{67} were resolved. The former decays by a 0.44-MeV isomeric transition to the 59-min ground state, which decays by emission of a 0.92-MeV beta transition.[49] The absolute disintegration rate of Zn^{69m} was obtained by employing a total conversion coefficient of 0.06[49] and by taking into account the fact that its equilibrium daughter activity also is counted. For Cu^{67} beta branching ratios of 20, 35, 45, and 0.6% were used for the 577-, 484-, 393-, and 189-keV beta transitions, respectively, in obtaining the true disintegration rate of Cu^{67}.[49,50] Thus, the $Ga^{71}(n,n\alpha)/Ga^{69}(n,p)$ cross-section ratio was determined to be 0.038±0.025. Similarly, by comparing the 0.18-MeV photopeak ($\alpha_{total}=0.06$) from Cu^{67} with the 0.44-MeV

[43] For brevity, we designate this reaction sum as the $(n,n\alpha)$ reaction, the understanding being that in activation work it is always the sum which is determined.
[44] J. Kantele and D. G. Gardner, Nucl. Phys. **35**, 353 (1962).
[45] M. Bormann, S. Cierjacks, R. Langkau, and H. Neuert, Z. Physik **166**, 477 (1962).
[46] J. Kantele, Nucl. Instr. Methods **17**, 33 (1962).
[47] M. Hillman, Bull. Am. Phys. Soc. **7**, 462 (1962).
[48] H. Vonach and H. Münzer, Oesterr. Akad. Wiss. Math. Naturw. Kl. Sitzber. Abt. II. **169**, No. 13, 199 (1960); and private communication.
[49] *Nuclear Data Cards*, compiled by K. Way *et al.* (Printing and Publishing Office, National Research Council–National Academy of Science, Washington 25, D. C.).
[50] G. Blosser, C. D. Goodman, and T. H. Handley, Phys. Rev. **110**, 531 (1958).
[51] See D. G. Gardner, Nucl. Phys. **29**, 373 (1962).

TABLE IV. Cross sections and upper limits for $[(n,n\alpha)+(n,\alpha n)]$ reaction sums at 14.7 MeV.

Reaction[a]	Q-value (MeV)	Product Half-life[b]	Monitor reaction and cross section	(mb)	Reference	$(n,n\alpha)$ cross section (mb)[a]
$V^{51}(n,n\alpha)Sc^{47}$	−10.27[c]	3.4 days	$V^{51}(n,\alpha)Sc^{48}$	(23)	d	<5
$Cu^{65}(n,n\alpha)Co^{61}$	−6.79[c]	1.6 h	$Cu^{65}(n,2n)Cu^{64}$	(954)	e	2.9 ±0.8
$Zn^{70}(n,n\alpha)Ni^{66}$	−5.92[c]	55 h	$Zn^{68}(n,\alpha)Ni^{65}$	(18)	d	0.89±0.40
$Ga^{71}(n,n\alpha)Cu^{67}$	−5.16[c]	58 h	$Ga^{69}(n,p)Zn^{69m}$	(24)	f	2.1 ±1.8
$Ge^{76}(n,n\alpha)Zn^{72}$	−8.37[g]	49 h	$Al^{27}(n,\alpha)Na^{24}$	(114)	h	<1.0
$Br^{81}(n,n\alpha)As^{77}$	−6.45[c]	39 h	$Br^{79}(n,\alpha)As^{76}$	(9.2)	d	<6.5
$Rb^{87}(n,n\alpha)Br^{83}$	−7.85[c]	2.3 h	$Rb^{87}(n,\alpha)Br^{84}$	(39)	f	<1.5
$Nb^{93}(n,n\alpha)Y^{89m}$	−1.64[c]	16 sec	$Cu^{63}(n,2n)Cu^{62}$	(507)	i	2.5 ±1.1
$Ag^{107}(n,n\alpha)Rh^{103m}$	−2.19[c]	54 min	$Ag^{109}(n,\alpha)Rh^{106}$	(10.5)	j	<2.0
$Ag^{109}(n,n\alpha)Rh^{105m}$	−3.03[c]	36 h	$Ag^{109}(n,\alpha)Rh^{106}$	(10.5)	j	<0.60
$In^{115}(n,n\alpha)Ag^{111g}$	−3.87[c]	7.5 days	$In^{115}(n,\alpha)Ag^{112}$	(2.7)	k	<0.055
$Au^{197}(n,n\alpha)Ir^{193}$	+1.50[g]	12 days	$Au^{197}(n,\alpha)Ir^{194}$	(0.43)	k	<0.040
$Tl^{203}(n,n\alpha)Au^{199}$	+0.031[c]	3.15 days	$Tl^{203}(n,\alpha)Au^{200}$	(0.37)	k	<0.012

[a] For brevity, the $[(n,n\alpha)+(n,\alpha n)]$ reaction sum is designated as $(n,n\alpha)$.
[b] See Ref. 49.
[c] See Ref. 79.
[d] Monitor cross section was determined in the present work (see Table VI).
[e] See Ref. 63.
[f] See Ref. 52.
[g] See Ref. 80.
[h] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[i] See Ref. 53.
[j] B. G. Dzantiev, V. N. Levkovskii, and A. D. Malievskii, Dokl. Akad. Nauk SSSR **2**, 135 (1957) [translation: Soviet Phys.—Doklady **3**, 537 (1957)].
[k] R. F. Coleman, B. E. Hawker, L. P. O'Connor, and J. L. Perkin, Proc. Phys. Soc. (London) **73**, 215 (1959).

photopeak from Zn^{69m}, and assuming that the 0.18-MeV gamma transition occurs in 45% of Cu^{67} decays,[49,50] this cross-section ratio is found to be 0.089±0.009. Using a value of 24±19 mb for the $Ga^{69}(n,p)$ cross section,[52] a value of 2.1±1.8 mb (Table IV) is obtained for the $Ga^{71}(n,n\alpha)$ cross section.

A check on this value was made using the $Al^{27}(n,\alpha)Na^{24}$ reaction as a flux monitor with gamma counting of the 0.18- and 1.37-MeV gammas from Cu^{67} and Na^{24}, respectively. Good agreement was obtained, which also supports the value of the $Ga^{69}(n,p)$ cross section used. Bormann, Cierjacks, Langkau, and Neuert[45] have studied the excitation function of the $Ga^{71}(n,n\alpha)$ reaction from 15.4 to 19.6 MeV, finding a cross section of 6±3 mb at the lowest energy (15.4 MeV). Extrapolation of their excitation curve down to 14.7 MeV gives a value comparable with our result.

Finally, it is possible that Cu^{67} could be produced via the (n,He^3) reaction on Ga^{69}, but in view of the generally negative results for this reaction (Sec. 3.2 and Ref. 42), this is considered unlikely.

In the case of $Br^{81}(n,n\alpha)As^{77}$, which was not detected, the $(n,n\alpha)$ product, As^{77}, is masked by the much more abundant $Br^{79}(n,\alpha)$ product, As^{76}. It was impossible in our case to distinguish small amounts of As^{77} from relatively large amounts of As^{76} by beta or gamma spectroscopy.

Bormann *et al.*[45] produced a 39-h beta activity in a KBr crystal irradiated with neutrons from 14 to 19.6 MeV, but here also the difficulty of distinguishing a small activity of As^{77} in the presence of As^{76} rendered the experiment insensitive to the $Br^{81}(n,n\alpha)$ reaction.

The $Nb^{93}(n,n\alpha)Y^{89m}$ (16 sec) reaction was detected, an activity of 16.3±1.3 sec being observed from irradiated spectroscopically pure niobium metal. The gamma spectra exhibited a single photopeak at 0.91 MeV, decaying with this half-life straight for over five half-lives.

The cross section for this reaction was measured by irradiating an intimate mixture of metallic niobium and copper powders, in a 10.1 ratio by weight, to insure identical sample and monitor geometry. The irradiated mixture was analyzed by comparing the decay of the 0.91-MeV gamma photopeak with the annihilation peak arising from positron decay of 10-min Cu^{62} from the $Cu^{63}(n,2n)$ reaction, for which a cross section of 507±4 mb was used.[53] Using a total conversion coefficient of 0.01 for the 0.91-MeV transition in Y^{89m} decay,[49] the $Nb^{93}(n,n\alpha)Y^{89m}$ cross section was determined to be 2.5±1.1 mb (Table IV). No chemical separation was possible due to the short half-life of 16 sec, but there is little doubt of the identification of the $(n,n\alpha)$ reaction from niobium, especially as these same niobium samples, when irradiated for much longer times, showed no activities which might arise from yttrium or strontium impurities.

3.4 $[(n,np)+(n,pn)+(n,d)]$ Reactions[54]

This reaction has been the one most extensively studied, and since cross sections mostly have been estimated from emitted-particle spectra, it is of interest to check these results with the activation technique. However, in only two cases was it possible to compare cross sections for the (n,np) reaction[54] obtained from

[52] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).

[53] J. M. Ferguson and W. E. Thompson, Phys. Rev. **118**, 228 (1960).

[54] For brevity, this reaction is designated (n,np), although it is the sum that is determined in activation methods.

TABLE V. Cross sections for $[(n,np)+(n,pn)+(n,d)]$, $(n,3n)$, and (n,γ) reactions at 14.7 MeV.

Reaction[a]	Q-value (MeV)	Product half-life[b]	Monitor reaction and cross section	(mb)	Reference	Cross section (mb)
$Ni^{58}(n,np)Co^{57}$	−7.91[c]	270 days	$Al^{27}(n,\alpha)Na^{24}$	(114)	d	520±120
$Mo^{92}(n,np)Nb^{91m}$	−7.89[c]	62 days	$Mo^{92}(n,p)Nb^{92g}$	(60)	e	<50
$Pr^{141}(n,3n)Pr^{139}$	−17.2[f]	4.5 h	$Pr^{141}(n,\gamma)Pr^{142}$	(2.3)	e	<10
$Au^{197}(n,3n)Au^{196}$	−14.5[f]	180 days	$Al^{27}(n,\alpha)Na^{24}$	(114)	d	<0.1
$Tl^{203}(n,3n)Tl^{201g}$	−15.9[g]	72 h	$Tl^{203}(n,2n)Tl^{202}$	(1300)	h	<10
$Y^{89}(n,\gamma)Y^{90m}$	+6.62[c]	3.1 h	$Al^{27}(n,\alpha)Na^{24}$	(114)	d	1.1 ±0.6
$Y^{89}(n,\gamma)Y^{90g}$	+6.62[c]	64 h	$Y^{89}(n,\gamma)(Y^{90m+g})$	(2.9)	i	1.8 ±0.6
$Nb^{93}(n,\gamma)Nb^{94m}$	+7.19[c]	6.6 min	$Al^{27}(n,\alpha)Na^{24}$	(114)	d	0.44±0.26
$Pr^{141}(n,\gamma)Pr^{142}$	+5.90[c]	19.1 h	$Al^{27}(n,\alpha)Na^{24}$	(114)	d	2.3 ±1.1

[a] For brevity, the $[(n,np)+(n,pn)+(n,d)]$ reaction sum is designated as (n,np).
[b] See Ref. 49.
[c] See Ref. 79.
[d] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[e] Monitor cross section was determined in the present work.
[f] See Ref. 80.
[g] Value calculated using semi-empirical masses from A. G. W. Cameron, Can. J. Phys. **37**, 44 (1959).
[h] R. J. Prestwood and B. P. Bayhurst, Phys. Rev. **121**, 1438 (1961).
[i] See Ref. 56.

emitted-particle spectra with those determined by activation. These two cases are discussed below and are listed in Table V.

The $Ni^{58}(n,np)Co^{57}$ (270 days) reaction was studied by irradiating nickel foils and nickel nitrate for periods up to 15 h, followed by radiochemical separation of cobalt from the nickel nitrate targets, the metallic foils being counted directly. Gamma spectra of the irradiated foils and cobalt fraction revealed the 122-keV peak which decayed with a 270-day half-life (followed for 0.5 year).

In order to measure the $Ni^{58}(n,np)Co^{57}$ cross section, the activity of 36 h Ni^{57}, from the competing $(n,2n)$ reaction, must be subtracted. By following the decay of the 1.37-MeV gamma peak in Ni^{57} decay, a cross section of 31±4 mb was determined (without chemistry) for the $(n,2n)$ reaction (Table VI), on the assumption that the 1.37-MeV gamma occurs in 75±6% of Ni^{57} decays.[55] A gamma of identical energy was counted from the decay (100%) of the Na^{24} monitor product, and a very low solid angle (~3%) was used to minimize summing.

Thus, assuming a total conversion coefficient[49] of 0.011 for the 122-keV gamma in Co^{57} decay, a cross section of 520±120 mb at 14.7 MeV was obtained (Table V) for the $Ni^{58}(n,np)Co^{57}$ reaction, taking the $Al^{27}(n,\alpha)Na^{24}$ reaction as the monitor.

The literature gives values for the $Ni^{58}(n,np)$ cross section that are summarized in Table I. Values obtained from emitted-particle spectra vary widely from 220 to 754 mb, while those based on activation methods are 160 mb,[22] 570±55 mb,[24] and 680±80 mb.[27] The present result agrees best with the 570-mb value of Glover and Weigold.[24]

The $Mo^{92}(n,np)$ reaction gives Nb^{91}, for which two isomers exist; the upper one of half-life 62 days and a lower one, which has yet to be observed, of estimated half-life 10^4–10^5 years.[49] The 62-day isomer decays by isomeric transition (97.5%) and by weak electron capture (2.5%) followed by a 1.208-MeV gamma.[49] The 104 keV isomeric gamma transition has a very large conversion coefficient (~50),[49] so that its intensity is only about 0.2 that of the 1.208-MeV gamma. Nothing is known of the long-lived ground state.

Irradiations of molybdenum foil and oxide powder, followed by radiochemical separation of niobium, were performed in attempts to detect 62-day Nb^{91m} from the $Mo^{92}(n,np)$ reaction. Gamma spectra revealed a very weak peak at about 1.2 MeV from both the niobium fraction and from molybdenum foils, but this is inconclusive due to the extremely low intensity. Likewise, attempts to detect the conversion electrons (about 82 keV) from the Nb^{91m} isomeric transition were inconclusive. A limit on the $Mo^{92}(n,np)$ cross section was set at <50 mb by using the $Mo^{92}(n,p)$ reaction as a monitor with cross section 60±15 mb (Table VI), this value being determined relative to the $Al^{27}(n,\alpha)Na^{24}$ monitor by following the decays of the 0.92-MeV gamma from 10-day Nb^{92} and the 1.37-MeV gamma from Na^{24}.

Although the 62-day Nb^{91m} isomer is not produced in detectable yield at 14.7 MeV, the possibility that the $Mo^{92}(n,np)$ reaction gives the long-lived Nb^{91} ground state cannot be checked by activation methods. In view of the results of Colli *et al.*[30] (Table I), who determined a 112-mb cross section for this reaction with a counter telescope, it would appear that the major path for this reaction leads directly to the long-lived ground state of Nb^{91}.

3.5 $(n,3n)$, (n,γ), and (n,t) Reactions

The $(n,3n)$ reaction is more difficult to detect than any of the other rare reactions at 14.7 MeV because of the generally large $(n,2n)$ cross sections and the in-

[55] G. Chilosi, S. Monaro, and R. A. Ricci, Nuovo Cimento **26**, 440 (1962).

ability to separate $(n,3n)$ and $(n,2n)$ products by radiochemistry. Moreover, Q values for $(n,3n)$ reactions seldom are more positive than -14 MeV, except for certain of the heavy nuclei. Since large samples are required to obtain a noticeable yield, gamma counting is the only satisfactory method, if suitable gammas are emitted. As a consequence, only a very few cases are amenable to study at 14.7 MeV. No example of an $(n,3n)$ reaction could be detected at 14.7 MeV.

A fairly extensive report of (n,γ) cross sections at 14–15 MeV has been given by Perkin, O'Connor, and Coleman[56] and by Wille and Fink.[57] In the course of the present work, a few additional (n,γ) cross sections were determined at 14.7 MeV by using cadmium-wrapped samples. These cases are given in Table V (see also Ref. 40).

The $Y^{89}(n,\gamma)Y^{90m}$ (3.1 h) reaction was detected with cadmium-wrapped Y_2O_3 samples, the decay of the 0.48-MeV gamma being followed. A cross section, based on the $Al^{27}(n,\alpha)Na^{24}$ monitor reaction, of 1.1 ± 0.6 mb was determined. Perkin, *et al.*[56] obtained a value of 2.9 ± 0.3 mb for the $Y^{89}(n,\gamma)Y^{90g}$ (64 h) reaction. Since the 3.1-h isomer was unknown at that time, this actually represents a total cross section for formation of both Y^{90m} and Y^{90g}. Subtracting our value of 1.1 mb for the production of the 3.1-h isomer, a value is obtained of 1.8 ± 0.6 mb for the reaction to the 64-h ground state (Table V).

The $Nb^{93}(n,\gamma)Nb^{94m}$ (6.6 min) reaction was detected by counting irradiated 0.001-in. thick niobium foils in a windowless flow proportional counter. By accepting a 100% counting efficiency for conversion electrons from the 41.4-keV isomeric transition, which is essentially completely converted, and for x rays, a cross-section value, relative to the $Al^{27}(n,\alpha)Na^{24}$ monitor, of 0.44 ± 0.26 mb was determined (Table V).

Irradiation of $Pr(NO_3)_3$ cadmium wrapped samples gave 19.1-h Pr^{142} from the (n,γ) reaction. Beta counting revealed the 19.1-h decay, while gamma spectra showed the 1.57-MeV gamma, which occurs in 100% of Pr^{142} decays.[49] A cross-section value of 2.3 ± 1.1 mb, relative to $Al^{27}(n,\alpha)Na^{24}$, was found, which compares favorably with reported values of 3.33 ± 0.33 mb[56] and 2.1 ± 1.0 mb.[57] The present value was used to set the limit on the Pr^{141} $(n,2p)$ reaction (Table II and Ref. 40).

The (n,t) reaction at 14.7 MeV has been studied in this laboratory by direct counting of tritium in the work of Poularikas and Gardner.[58] Their results, together with a summary of the literature, are being published. Consequently, we have not studied the (n,t) reaction specifically in the present work.

3.6 (n,p), (n,α), and $(n,2n)$ Reactions

Along with the study of rare reactions, a number of (n,p), (n,α), and $(n,2n)$ reactions were measured at 14.7 MeV. These are gathered into Table VI, together with previous values from the literature.

Generally good agreement exists between the present values and the literature. Noted exceptions occur for the results of Khurana and Hans[59] and of Strohal, Cindro, and Eman.[60] The former authors report values for the $Sc^{45}(n,\alpha)$ and $Ti^{50}(n,p)$ cross sections which are an order of magnitude larger than the present values, while their $Sc^{45}(n,2n)$ value is lower by some 40%; similarly, they report a $Ti^{49}(n,p)$ cross section of 97 mb, although the literature values agree well at 33, 29 ± 5, and 29 ± 8 mb (see Ref. 51). (Their neutron source gave only 10^8 DT neutrons/sec, suggesting poor statistics in the counting experiments.) Strohal *et al.*[60] give cross sections for the $Zr^{92}(n,p)$, $Zr^{94}(n,p)$, and $Br^{81}(n,\alpha)$ reactions which are markedly larger than the present results, whereas their $Mo^{92}(n,p)$ value is much smaller. Moreover, other cross sections reported in their paper show drastic disagreements with the results of others (see discussion in Ref. 40).

Some remarks on (n,p), (n,α), and $(n,2n)$ cross sections which are appropriate to the measurements of the present values (Table VI) are given below, but full details of all determinations can be found in Ref. 40.

The $Sc^{45}(n,\alpha)K^{42}$ cross section, 63 ± 12 mb from present measurements, agrees well with the value of 53.5 ± 3.0 mb of Bayhurst and Prestwood.[61] The $Sc^{45}(n,2n)Sc^{44g}$ cross section from beta counting is 205 ± 6 mb[61] and from gamma counting[62] is 198 ± 15 mb. The values of Khurana and Hans[59] are, for both reactions, out of line with these results. It is possible that their low neutron flux (10^8 DT neutrons/sec total) with consequent poor statistics, and the fact that only one count was usually made on the iron foil monitors, may account for the disagreement. Furthermore, the neglect of the large contribution[61–63] from 2.4-day Sc^{44m}, from the $(n,2n)$ reaction, could explain the large (n,α) and low $(n,2n)$ values of Khurana and Hans.

Since Poularikas and Fink[63] report a 27 ± 6 mb cross section for the $Ti^{50}(n,p)Sc^{50}$ (1.8) min) reaction and Khurana and Hans[59] give 147 ± 13 mb, both groups employing beta counting of irradiated titanium, this reaction was remeasured in the present investigation. The former workers used isotopically enriched Ti^{50} oxide as well as natural titanium metal foil. In the present study, a gamma counting comparison of the 1.56-MeV gamma from 1.8-min Sc^{50} decay with the 1.02-MeV gamma from Mg^{27} decay, from the $Al^{27}(n,p)Mg^{27}$

[56] J. Perkin, L. P. O'Connor, and R. F. Coleman, Proc. Phys. Soc. (London) **72**, 505 (1958).
[57] R. G. Wille and R. W. Fink, Phys. Rev. **118**, 242 (1960).
[58] A. Poularikas and D. G. Gardner, University of Arkansas Annual AEC Report, 1963 (to be published); and A. Poularikas, M.S. thesis, University of Arkansas, 1962.
[59] C. S. Khurana and H. S. Hans, Nucl. Phys. **13**, 88 (1959).
[60] P. Strohal, N. Cindro, and B. Eman, Nucl. Phys. **30**, 49 (1962).
[61] B. P. Bayhurst and R. J. Prestwood, AEC report, LA-2439, 1960 (unpublished).
[62] L. A. Rayburn, Phys. Rev. **122**, 168 (1961).
[63] A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959).

TABLE VI. Summary of cross sections of observed (n,p), (n,α), and $(n,2n)$ reactions at 14.7 MeV.

Reaction	Product half-life[a]	Cross sections (mb) This work[b]	Literature values	Monitor reaction reference
$K^{41}(n,p)Ar^{41}$	1.8 h	69±17	81±32[c]	d
$Ti^{50}(n,p)Sc^{50}$	1.7 min	28±12	27±6,[e] 147±13[f]	g
$V^{51}(n,p)Ti^{51}$	5.8 min	55±12	53±5,[h] 23±7,[c] 27±4[i]	j
$Cu^{65}(n,p)Ni^{65}$	2.6 h	29.3±3.2	<40, 31±13, 27±11, 27±5, ~20, 19±4, 17±4, 11±1[e]	k
$Zn^{64}(n,p)Cu^{64}$	12.8 h	230±30	386±60, 295, 284±20, 216, 171±18, 18±4, ~17[e]	d
$Zr^{92}(n,p)Y^{92}$	3.7 h	22±4	20.7±0.9,[l] 76±16,[m] ~21[k]	d
$Zr^{94}(n,p)Y^{94}$	17 min	7±4	~11,[n] 10.8±0.6,[l] 48±12,[m] 11±4[c]	f
$Mo^{92}(n,p)Nb^{92g}$	10 days	60±15	108±55,[c] ~31,[o] ~14.5[p]	d
$Mo^{94}(n,p)Nb^{94}$	6.6 min	6.0±1.5		f
$Mo^{96}(n,p)Nb^{96}$	26 h	37±9	21±7[p]	d
$Mo^{98}(n,p)Nb^{98}$	51 min	9±2		d
$Tb^{159}(n,p)Gd^{159}$	18 h	2.2±1.3		d
$K^{41}(n,\alpha)Cl^{38}$	37 min	30±12	31.4±11.0,[c] 50±24,[q] 12±5,[r]	f
$Sc^{45}(n,\alpha)K^{42}$	12.5 h	63±12	53.5±3.0,[s] 132±8[t]	d
$V^{51}(n,\alpha)Sc^{48}$	1.8 days	23±4	28.6±5.7,[c] 30±10,[q] 43.7±8.6,[u] 13.5±1.4,[v] 30.5±0.4,[i] 18±3[w]	d
$Cu^{65}(n,\alpha)Co^{62m}$	1.6 min	1.9±0.6		k
$Cu^{65}(n,\alpha)Co^{62g}$	13.9 min	14±10	7.5±2.0[x]	k
$Zn^{68}(n,\alpha)Ni^{66}$	2.6 h	18±5	7.6±0.8,[y] 51±10[z]	d
$As^{75}(n,\alpha)Ga^{72}$	14 h	9.3±3.1	12.3,[c] 10.2±0.7[aa]	d
$Br^{79}(n,\alpha)As^{76}$	27 h	9.2±2.0	9±3,[q] 10.0±1.8[y]	d
$Br^{81}(n,\alpha)As^{78}$	90 min	6.6±1.4	103±26,[c] 107±20[m]	d
$Y^{89}(n,\alpha)Rb^{86m}$	1 min	0.91±0.45		
$Zr^{90}(n,\alpha)Sr^{87m}$	2.8 h	2.8±1.3	3.34±0.16,[l] 3.3±0.6[y]	d
$Zr^{94}(n,\alpha)Sr^{91}$	9.7 h	4.3±1.1	6.0±0.4,[aa] 4.9±0.6,[n] 3.6±0.5,[y] 3.99±0.16,[l] 4.1±0.6[q]	d
$Zr^{96}(n,\alpha)Sr^{93}$	7 min	5±4	4.8±0.7[l]	f
$Nb^{93}(n,\alpha)Y^{90m}$	3.1 h	5.9±2.0	5±2[ab]	d
$Nb^{93}(n,\alpha)Y^{90g}$	64 h	8.6±2.5[ac]	9±3,[q] 9.0±2.2,[ad] 9.4±0.4[aa]	d
$Mo^{92}(n,\alpha)Zr^{89g}$	79 h	20±8		d
$Cs^{133}(n,\alpha)I^{130}$	12.6 h	1.0±0.9	1.1±0.5,[q] 1.0±0.3,[ad] 1.9±0.2[ae]	d
$Sc^{45}(n,2n)Sc^{44}$	4.0 h	204±25	198±15,[t] 205±6,[aa] 129±9[s]	d
$Zn^{64}(n,2n)Zn^{63}$	38 min	153±36	167±13,[t] 119±13,[af] 224±45,[c] 254±20,[z] 150±30[ag]	d
$Ni^{58}(n,2n)Ni^{57}$	36 h	31±4	52±5,[z] 40.6±12,[c] 33.4±2.7,[ah] 40±5[ai]	d
$Zr^{90}(n,2n)Zr^{89m}$	4.4 min	84±12	74±3,[m] 79.8±16[c]	f
$Nb^{93}(n,2n)Nb^{92g}$	10 days	499±91	430±70,[aj] 530±60[ak]	d
$Nb^{93}(n,2n)Nb^{92m}$	13 h	≤1.2[al]		d
$Mo^{92}(n,2n)Mo^{91g}$	16 min	198±40	211±16,[u] 132±20,[af] 190±30,[c] 320±90[n]	d

[a] See Ref. 49.
[b] Values which disagree with the literature are discussed in the text and in Ref. 40.
[c] See Ref. 52.
[d] $Al^{27}(n,\alpha)Na^{24}$ (114 mb) from S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[e] See summary in Ref. 51.
[f] See Ref. 63.
[g] $Cu^{63}(n,2n)Cu^{62}$ (507 mb) from Ref. 53.
[h] See Ref. 64.
[i] See Ref. 14.
[j] $Al^{27}(n,p)Mg^{27}$ (82 mb) from Ref. 44.
[k] $Cu^{65}(n,2n)Cu^{64}$ (954 mb) from Ref. 63.
[l] C. H. Reed, Ph.D. thesis, University of Utah, 1960.
[m] See Ref. 60.
[n] J. E. Brolley, M. E. Bunker, D. R. F. Cochran, R. L. Henkel, J. P. Mize, and J. W. Starner, Phys. Rev. **99**, 330 (1955).
[o] See Ref. 30.
[p] J. E. Brolley, J. L. Fowler, and L. K. Schlacks, Phys. Rev. **88**, 618 (1952).
[q] See Ref. 3.
[r] See Ref. 16.
[s] See Ref. 59 and Nuovo Cimento **26**, 1328 (1962).
[t] See Ref. 62.
[u] I. Kumabe, E. Takekoshi, H. Ogata, Y. Tsuneoka, and S. Oki, J. Phys. Soc. (Japan) **13**, 325 (1958).
[v] B. Karlik (private communication).
[w] M. Hillman, Nucl. Phys. **37**, 78 (1962).
[x] See Ref. 44.
[y] See Ref. 51.
[z] See Ref. 29.
[aa] See Ref. 61.
[ab] W. L. Alford, D. R. Koehler, and C. E. Mandeville, Phys. Rev. **123**, 1365 (1961).
[ac] Includes contribution from the (n,α) reaction leading to Y^{90m}.
[ad] H. G. Blosser, C. D. Goodman, T. H. Handley, and M. L. Randolph, Phys. Rev. **100**, 429 (1955).
[ae] R. F. Coleman, B. E. Hawker, L. P. O'Connor, J. L. Perkin, Proc. Phys. Soc. (London) **73**, 215 (1959).
[af] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[ag] A. V. Cohen and P. H. White, Nucl. Phys. **1**, 73 (1956).
[ah] L. A. Rayburn, Bull. Am. Phys. Soc. **4**, 288 (1959).
[ai] See Ref. 27.
[aj] See Ref. 48.
[ak] V. L. Glagolev and P. A. Yampolskii, Sov. Phys. JETP **14**, 1220 (1961).
[al] E. T. Bramlitt and R. W. Fink, J. Inorg. Nucl. Chem. **24**, 1317 (1963), showed that no isomeric state of Nb^{92} exists with a half-life of 13 h.

monitor was employed in the irradiations of spectroscopically pure titanium metal. Although a wide range of values are reported for the $Al^{27}(n,p)$ cross section,[51] a recent careful investigation and critical analysis of this reaction by Kantele and Gardner[44] has established a precision value of the cross section of 82±10 mb at 14.7 MeV. The gamma counting method used in this study thus gives a value for the $Ti^{50}(n,p)Sc^{50}$ (1.8 min) cross section of 28±12 mb (Table VI), thereby confirming the beta counting value of 27±6 mb of Poularikas and Fink.[63] This value was used to set the limit on the $Ti^{50}(n,2p)Sc^{50}$ reaction[40] (Table II).

Our $V^{51}(n,p)Ti^{57}$ cross section of 55±12 mb agrees very well with the value of 53±5 mb of Poularikas,[64] but it is about twice the 27-mb value of Paul and Clarke[52] or the 23-mb value of Allan,[14] based on analysis of proton spectra in nuclear emulsions with consequent poor statistics. The values near 55 mb probably represent the correct cross section.

The $V^{51}(n,\alpha)Sc^{48}$ cross section, relative to the $Al^{27}(n,\alpha)Na^{24}$ monitor, was found to be 25±7 mb by beta counting and 23±4 mb (Table VI) by relative gamma counting of the 1.31- and 1.37-MeV gammas from Sc^{48} and Na^{24}, respectively. The latter cross section is the more accurate because of the close similarity of sample and monitor gamma counting.

The $Zn^{68}(n,\alpha)Ni^{65}$ (2.56 h) cross section was determined both by beta and by gamma counting. The 1.49-MeV gamma peak was followed, occurring in 18% of Ni^{65} decays,[49] and relative to $Al^{27}(n,\alpha)Na^{24}$, a cross section was found to be 18±5 mb (Table VI). The reasons for the wide disagreement in the literature are not understood.

In establishing the upper limit for the $Br^{81}(n,n\alpha)$ reaction cross section, beta decay of the arsenic fraction permitted a measurement of the $Br^{79}(n,\alpha)/Br^{81}(n,\alpha)$ cross section ratio to be made, 1.40±0.10. Thus, using the present value for the $Br^{79}(n,\alpha)$ reaction (9.20±2.0 mb), a value of 6.6±1.4 mb is obtained for the $Br^{81}(n,\alpha)$ reaction (Table VI). The former agrees well with the literature values of 9±3 and 10.0±1.8 mb (Table VI). However, the latter value falls into sharp disagreement with the 103-mb value of Paul and Clarke[52] and the 107-mb value of Strohal, Cindro, and Eman,[60] whose results are discussed above. It is not clear why such a discrepancy exists with the $Br^{81}(n,\alpha)$ cross section. However, chemical separation was not done by Paul and Clarke.[52] With radiochemical separation, it is possible to get a very accurate value of the $Br^{79}/Br^{81}(n,\alpha)$ cross section ratio, as in the present study, since only two activities are present in the arsenic fraction. The complex decay curve resulting from irradiated, nonchemically separated bromine is, on the other hand, very difficult to resolve. That the present results are in accord with the Levkovskii trend[65] of decreasing (n,α) cross sections with increasing mass number for a given element also supports the present values.

[64] A. D. Poularikas, University of Arkansas Annual AEC Report, 1960 (unpublished).

4. CALCULATION OF CROSS SECTIONS FROM STATISTICAL THEORY

Theoretical estimation of cross sections based on statistical theory of compound nuclear reactions[66–69] has been employed for 14.7-MeV neutron reactions in the present work. The cross section $\sigma_{(a,b)}$ from statistical theory can be written as

$$\sigma_{(a,b)}=\sigma_a(F_b/\sum F_i)\,, \tag{4}$$

where σ_a is the cross section for formation of the compound nucleus, F_i is a function corresponding to the relative emission probability of a particle i, and F_b is the emission probability for particle b. The F-functions can be written as

$$F_b=Kg_b\mu_b\int_0^{E_{b(\max)}} E_b\sigma_{b(E_x)}\omega_{(E_x)}dE_b\,, \tag{5}$$

where K is a constant, μ_b is the reduced mass of the emitted particle, g_b is a statistical weighting factor given by $2S_b+1$, where S_b is the spin of the emitted particle; E_b is the kinetic energy of the emitted particle; $\omega_{(Ex)}$ is the level density of the residual nucleus which retains excitation energy E_x after emission of particle b; and $\sigma_{b(E_b)}$ is the cross section for compound nucleus formation by the inverse reaction (in which particle b is absorbed by an excited product nucleus having excitation energy E_x). The residual nucleus excitation energy E_x is given by $(E_a+Q_{(a,b)}-E_b)$, where $Q_{(a,b)}$ is the Q value for reaction (a,b), and the term $[E_a+Q_{(a,b)}]$ gives the value of the maximum kinetic energy available to particle b, $E_b(\max)$.

Since it has been shown[70,71] that some 10–15% of the measured nonelastic cross section at 14–15 MeV arises from direct interactions, we arbitrarily take the values for σ_a in Eq. (4) to be given by

$$\sigma_a=0.85\sigma_{\text{nonelastic}}\,. \tag{6}$$

[65] V. N. Levkovskii, Zh. Eksperim. i. Teor. Fiz. **33**, 1526 (1957); **31**, 360 (1956) [translations: Soviet Phys.—JETP **6**, 1174 (1958); **4**, 291 (1957)].

[66] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), pp. 311–564.

[67] R. G. Moore, Jr., Rev. Mod. Phys. **32**, 101 (1960).

[68] K. J. LeCouteur, in *Nuclear Reactions*, edited by P. M. Endt and M. Demeur (North-Holland Publishing Company, Amsterdam, 1959), Vol. I, pp. 318–355.

[69] T. Ericson, Phil. Mag. **9**, 425 (1960).

[70] J. Benveniste, Report UCRL-5220 (1958), in *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, R. 2494.

[71] M. H. MacGregor, Report UCRL-5229 (1958), in *Comptes Rendus du Congrès International de Physique Nucléaire; Interactions Nucléaires aux Basses Energies et Structure des Noyaux, Paris, July, 1958*, edited by P. Guggenberger (Dunod, Paris, 1959), pp. 609–611.

TABLE VII. Effects on theoretical cross sections due to variation in the nuclear temperature parameter of the level density expression ν.

Target nuclide	ν	Calculated reaction cross section (mb) $(n,2n)$	(n,pn)	$(n,2p)$	$(n,p\gamma)$	$(n,\alpha n)$	$(n,\alpha\gamma)$
Ni^{58}	10	324	435	82	47	164	202
Ni^{58}	13	252	420	109	65	163	328
Ni^{58}	20	146	362	120	114	158	388
V^{51}	10	963	67	0	13	0	27
V^{51}	13	860	84	0	22	0	42
V^{51}	20	680	106	0	51	0	106
Cu^{65}	10	1220	17	0	1.5	1.0	4.1
Cu^{65}	13	1180	24	0	4.0	1.8	9.5
Cu^{65}	20	1060	34	0	11	4.5	29
Mo^{92}	10	673	174	62	8.4	8.1	14.8
Mo^{92}	13	542	196	102	54	12	33
Mo^{92}	20	365	274	200	172	31	146
Nb^{93}	10	1650	19	0	1.24	30	5.6
Nb^{93}	13	1530	29	0	3.0	52	14
Nb^{93}	20	1290	40	0	9.0	176	88

Values of $\sigma_{\text{nonelastic}}$ were taken from the compilation of Howerton.[72] Cross sections for reactions in which alphas are emitted were computed using an optical model with a complex nuclear potential given by Huizenga and Igo.[73] For reactions involving emission of protons and neutrons, cross sections were obtained from Blatt and Weisskopf,[66] based on a sharp cutoff black-body model having a square well potential. Radius constants of 1.5 and 1.3 F were used for protons and neutrons, respectively.

The form of the level density term was taken as

$$\omega_{(E_x)}=C\exp[2(aE_x)^{1/2}], \tag{7}$$

where the coefficient C was assumed to be energy-independent and to show a dependence on mass number A given by El-Nadi and Wafik[74] to be

$$C=0.82\exp(0.071A-0.00026A^2), \tag{8}$$

and odd-even effects were taken into account through the expression given by Varshni[75]:

$$C_{\text{odd-odd}}=2.43C_{\text{odd-}A}=15.05C_{\text{even-even}}. \tag{9}$$

The nuclear temperature coefficient a in Eq. (7) is given by $a=A/\nu$, where A is the mass number and ν is a constant in units of MeV. The value $\nu=13$ was used[76–78] in most of the calculations; however, the effect on the theoretical cross sections caused by using other values of ν was studied.[40] Since the nuclear temperature coefficient a is in the exponent of the level density expression, Eq. (7), the calculated cross sections depend rather critically on the choice of its value.[14,59,75–77] To explore the effect of the choice of ν (in $a=A/\nu$) on the theoretical cross sections, calculations were made using values of ν of 10, 13, and 20 MeV (using Method A described in Sec. 4.2). The results are shown in Table VII, from which it is seen that with increasing ν the $(n,2n)$ values decrease slightly, but for all other reactions the cross sections increase considerably. Since most of the $(n,2n)$ cross sections calculated with $\nu=13$ lie slightly higher than experiment, a somewhat larger value of ν would be suggested, but in that case, the calculated cross sections for such rare reactions as (n,pn) and $(n,\alpha n)$ become much too large. Thus, a choice of $\nu=13$ appears to be the best.

The solution of Eq. (5) for the F_i values was carried out with an IBM-650 computer, assuming $i=\alpha$, n, or p only (i.e., that alphas, neutrons, or protons only are emitted). A program written in FORTRAN gave values of the integrand of Eq. (5) in steps of 0.5 MeV in E_i and integrated to give F_i values. The individual values for the integrand at various E_i were plotted as a function of E_i to give the theoretical energy distribution of the emitted particles i, and the integration of portions of these spectra, as required, was done by use of a compensating polar planimeter. Q values mostly were obtained from the tables of Ashby and Catron[79] or from the tables of König, Mattauch, and Wapstra.[80]

4.1 Theoretical Cross-Section Calculations of $(n,\alpha n)$ Reactions and Discussion of the $[(n,n\alpha)+(n,\alpha n)]$ Measurements

One feature of the observed $(n,n\alpha)$ studies is that the reactions that could be detected resulted from the highest-A isotope of a given element. Since activation methods are used, it was possible to look for this reaction only with these higher-A isotopes. Preiss and Fink[29] observed (n,np) reactions with the higher-A nickel isotopes, where the cross sections amounted only to a few millibarns, in contrast to the lower-A nickel isotopes, where the (n,np) cross sections become very large (Table I). If a similar Levkovskii trend[65] exists for $(n,n\alpha)$ reactions, it would suggest that very much larger $(n,n\alpha)$ cross sections might be found for lower-A isotopes of a given element; for example, with Cu^{63}, Zn^{64}, Ga^{69}. Unfortunately, activation is not a suitable technique to check this question, so that it remains to be done by emitted particle methods.

The $(n,n\alpha)$ cross sections apparently decrease with increasing Z, so that the reaction is not detectable at $Z=32$, 36, or 37. Probably the relatively high cross section for Nb^{93} reflects a closed shell effect, since an (n,α) reaction yields Y^{90} (8.6 mb, 51 neutrons), while

[72] R. J. Howerton, University of California Radiation Laboratory Report UCRL-5351, 1958, (unpublished).
[73] J. R. Huizenga and G. Igo, Nucl. Phys. **29**, 462 (1962).
[74] M. El-Nadi and M. Wafik, Nucl. Phys. **9**, 22 (1958).
[75] Y. P. Varshni, Nuovo Cimento **22**, 145 (1961).
[76] R. D. Albert, J. D. Anderson, and C. Wong, Phys. Rev. **120**, 2149 (1960).
[77] D. W. Barr, D. I. Browne, and J. S. Gilmore, Phys. Rev. **123**, 859 (1961).
[78] D. C. Diven, J. Terrell, and A. Hemmendinger, Phys. Rev. **120**, 556 (1960).
[79] V. J. Ashby and H. C. Catron, University of California Radiation Laboratory Report UCRL-5419, 1959 (unpublished).
[80] L. A. König, J. H. E. Mattauch, and A. H. Wapstra, *Nuclear Data Tables, Consistent Set of Q-Values*, (U. S. Government Printing Office, Washington 25, D. C., 1960), Parts 1 and 2.

the $(n,n\alpha)$ reaction gives Y^{89} (2.5 mb, 50 neutrons).[80a]

It is of interest to ask whether the $(n,n\alpha)$ or the $(n,\alpha n)$ process predominates in these reactions, the possibility of a pickup reaction to form He^5 being ruled out since no bound states exist for this nuclide. Since neutron emission is more probable than alpha emission, due to the Coulomb barrier, one might, at first, expect that the reaction would occur predominantly following the (n,n') process; i.e., an $(n,n\alpha)$ mechanism. For such a process to compete effectively with the $(n,2n)$ reaction, it is necessary that S_α (from the target nuclide) be much lower than S_n, a condition which practically is always true. There is an additional requirement for the $(n,n\alpha)$ mechanism, that S_α must be lower than the proton separation energy S_p, otherwise proton emission will predominate [an (n,np) process], as the Coulomb barrier is about half as great for protons as for alphas. In Table VIII are listed S_n, S_p, and S_α values for nuclides studied. It is clear that in general $S_\alpha<S_p<S_n$. Although the (S_p-S_α) difference becomes much greater with increasing Z, suggesting that the $(n,n\alpha)$ process should compete more effectively with (n,np) reactions at increasing Z, the increasing Coulomb barrier inhibits alpha emission and reduces the $(n,n\alpha)$ cross section. The Coulomb barrier inhibition should be more effective for alphas from the $(n,n\alpha)$ process, since they necessarily have lower energies than alphas from the $(n,\alpha n)$ process. This may account for the fact that the reaction is not detectable with higher-Z nuclides. On the other hand, the negative (S_p-S_α) value for V^{51} suggests that an $[(n,n\alpha)+(n,\alpha n)]$ reaction with this nucleus is not likely, although it was detected. The slight (S_p-S_α) difference (0.7 MeV) for Cu^{65} also would not seem sufficient to account for the observed reaction with this nucleus.

TABLE VIII. Separation energies for neutrons (S_n), protons (S_p) and alphas (S_α) for nuclides investigated for the $[(n,n\alpha)+(n,\alpha n)]$ reaction.[a]

Target nuclide	S_n (MeV)	S_p (MeV)	S_α (MeV)	(S_p-S_α) (MeV)
V^{51}	11.0	8.0	0.3	−2.3
Cu^{65}	9.9	7.5	6.8	0.7
Zn^{70}	9.2	11.7*	5.9	5.8*
Ga^{71}	9.2	7.8	5.1	2.7
Ge^{76}	9.6	11.7*	6.5*	5.2*
Br^{81}	10.0	7.4	6.4	1.0
Rb^{87}	10.0	8.7	7.8	0.9
Nb^{93}	8.7	6.0	1.6	4.4
Ag^{107}	9.4	5.6	2.2	3.4
Ag^{109}	9.1	6.4	3.0	3.4
In^{115}	9.1	6.9	3.9	3.0
Au^{197}	8.0	5.8	−2.0*	7.8*
Tl^{203}	8.8	6.1	−0.03	6.1

[a] Values were obtained from Ref. 79 unless indicated by asterisk in which case they were calculated using empirical mass data from A. G. W. Cameron, Can. J. Phys. **37**, 44 (1959).

To further these implications that the reaction proceeds by an $(n,\alpha n)$ process, predominantly, theoretical values for the cross section of the $(n,\alpha n)$ process *alone* have been computed from the statistical theory of compound nuclear reactions. (No satisfactory direct-interaction theory exists which permits calculation of cross sections for alpha-emitting reactions).

The $(n,\alpha n)$ cross section is considered to be the product of the cross section for emission of an alpha and the probability that a neutron is subsequently emitted from the residual excited nucleus:

$$\sigma_{(n,\alpha n)}=\sigma_{(n,\alpha)}P_n, \tag{10}$$

where P_n denotes the probability of subsequent neutron emission. To find P_n it is assumed that if the residual nucleus after alpha emission has excitation energy at least as high as S_n, then a neutron is emitted. In other words, the probability that the alpha has energy between zero and $(E_n+Q_{(n,\alpha)}-S_n)$ is equivalent to P_n. Hence, P_n is of the form

$$P_n=\frac{\int_0^{[E_n+Q_{(n,\alpha)}-S_n]} E_\alpha\sigma_{\alpha(E_\alpha)}\omega_{(E_x)}dE_\alpha}{\int_0^{[E_n+Q_{(n,\alpha)}]} E_\alpha\sigma_{\alpha(E_\alpha)}\omega_{(E_x)}dE_\alpha}. \tag{11}$$

A somewhat similar expression is used for determining the $(n,\alpha\gamma)$ cross section with the exception that the limits on the integral in the numerator go from $[E_n+Q_{(n,\alpha)}-S_n]$ to $[E_n+Q_{(n,\alpha)}]$.

In Table IX are listed the nuclei irradiated in attempts to detect the $[(n,n\alpha)+(n,\alpha n)]$ reaction, together with the theoretical values for the $(n,\alpha n)$ and $(n,\alpha\gamma)$ cross sections. The experimental cross sections have been listed for comparison. It is seen that excellent agreement exists between theory and experiment up to niobium, the experimental cross sections agreeing within a factor of two with theoretical values for both $(n,\alpha n)$ and $(n,\alpha\gamma)$ reactions. For niobium, however, the theoretical $(n,\alpha n)$ cross section is some 20 times larger than the experimental one, while the $(n,\alpha\gamma)$ values are in excellent agreement. It is to be recalled, however, that the $(n,\alpha n)$ reaction on Nb^{93} was detected by counting the 16 sec isomeric state Y^{89m} only. The large discrepancy might thus be accounted for if the $(n,\alpha n)$ reaction proceeds predominantly to the stable Y^{89} ground state. The measurement of this cross section by emitted-particle techniques would be extremely valuable in confirming this hypothesis.

For the remaining cases, the theoretical $(n,\alpha n)$ cross sections are of the same order as the limits established, whereas the theoretical $(n,\alpha\gamma)$ values are considerably smaller than the experimental ones. Differences of this latter type usually are attributed to direct interactions.

[80a] *Note added in proof.* T. Ebrey and P. C. Gray (private communication) have observed the $Rh^{103}(n,n\alpha)Tc^{99m}$ (6.0 h) reaction by activation and radiochemical separation and find a cross section of somewhat less than 100 μb. In the same experiments, an upper limit well below 0.1 μb was set for the $Rh^{103}(n,He^3)Tc^{101}$ (14 min) reaction.

TABLE IX. $(n,\alpha n)$ and $(n,\alpha\gamma)$ theoretical cross sections for nuclides in Table VIII and comparison with experimental values at 14.7 MeV.

Target nuclide	$(n,\alpha n)$ cross sections Theoretical (mb)	Experimental[a,b] (mb)	$(n,\alpha\gamma)$ cross sections Theoretical (mb)	Experimental[a] (mb)	
V^{51}	<1.2	<5	38	23 ± 4	
				28.6 ± 6.0	c
				30 ±10	d
				43.7 ± 8.6	e
				13.5 ± 1.4	f
				30.5 ± 0.4	g
Cu^{65}	1.6	2.9 ±0.8	8.5	7.5 ± 2.0	h
				14 ±10	
Zn^{70}	0.70	0.89 ±0.40	9.9		
Ga^{71}	1.6	2.1 ±1.8	6.1		
Ge^{76}	0.48	<1.0	1.5		
Br^{81}	0.67	<6.5	6.7	6.6 ± 1.4	
				103 ±26	c
				107 20	i
Rb^{87}	0.004	<1.5	0.27		
Nb^{93}	47	2.5 ±1.1	12	8.6 ± 2.5	
				9 ± 3	d
				9.4 ± 0.4	j
				9.0 ± 2.2	k
Ag^{107}	3.8	<2.0	3.8		
Ag^{109}	0.87	<0.60	1.3	10.5 ± 2.0	l
In^{115}	0.086	<0.055	0.32	2.9 ± 0.3	m
				2.5 ± 0.4	n
Au^{197}	~0	<0.040	0.012	0.43 ± 0.04	m
Tl^{203}	0.0006	<0.012	0.0005	0.37 ± 0.04	m

[a] Cross sections were determined in the present work unless indicated by a reference in parentheses.
[b] These values include any contribution arising from $(n,n\alpha)$ reactions.
[c] See Ref. 52.
[d] See Ref. 3.
[e] I. Kumabe, E. Takekoshi, H. Ogata, Y. Tsuneoka, and S. Oki, J. Phys. Soc. (Japan) **13**, 325 (1958).
[f] B. Karlik (private communication).
[g] See Ref. 64.
[h] See Ref. 44.
[i] See Ref. 60.
[j] See Ref. 61.
[k] H. G. Blosser, C. D. Goodman, T. H. Handley, and M. L. Randolph, Phys. Rev. **100**, 429 (1955).
[l] B. G. Dzantiev, V. N. Levkovskii, and A. D. Malievskii, Dokl. Akad. Nauk SSSR **2**, 135 (1957) [translation: Soviet Phys.—Doklady **3**, 537 (1957)].
[m] R. F. Coleman, B. E. Hawker, L. P. O'Connor, and J. L. Perkin, Proc. Phys. Soc. (London) **73**, 215 (1959).
[n] See Ref. 51.

Since direct reactions arise mostly from surface phenomena, it is reasonable that their relative contributions would be more evident at higher A. In view of the additional (n,α) contribution from direct interaction, one might suppose that there also should be an increase in the $(n,\alpha n)$ contribution from direct reaction, particularly since S_n becomes smaller at higher A. According to Butler,[81] however, it appears that direct interaction processes are significant only for those cases where the reaction proceeds to a low-lying level of the residual nucleus. Thus, the residual nucleus from a direct (n,α) reaction is not expected to allow further nucleon evaporation. This picture is consistent with the nonobservation of $(n,\alpha n)$ reactions at high A.

4.2 Theoretical $(n,2p)$ Cross Sections and Comparison with Competing Reactions

Calculations of theoretical values for $(n,2p)$ cross sections were made[40] for nuclides having rather large (S_n-S_p) differences. Two statistical theory methods were employed. In Method A, it was assumed that particle emission from a residual nucleus occurs whenever the excitation energy is greater than the particle separation energy. In Method B, however, we have taken into account the following considerations.

Allan[14] has pointed out that with (n,np) reactions a significant yield of protons with energies below about 2 MeV is not found. Ericson[62] has suggested that angular momentum effects hindering particle emission may be considerable, particularly in cases involving multiple particle emission. Since emitted neutrons must penetrate an angular momentum barrier, assumed by Allan[14] to be about 1 MeV, and since emitted protons below 2 MeV are not found, we have assumed in Method B that (a) proton emission can occur from a residual nucleus whenever the excitation energy exceeds (S_p+2) MeV, (b) neutron emission can occur whenever the residual nucleus has excitation energy larger than (S_n+1), and (c) neutron emission always occurs in preference to proton emission, gamma decay being preferred to alpha emission.

Results of the $(n,2p)$ calculations using both methods are given in Table X. The experimental data so far do not permit a choice to be made between the two methods. Method B predicts smaller cross sections for $(n,2p)$ reactions than Method A, except for Mo^{92}, for which both methods predict the $(n,2p)$ cross section to be larger than that for the (n,p) reaction.

The competing $(n,2n)$, (n,pn), $(n,p\gamma)$, $(n,\alpha n)$, and $(n,\alpha\gamma)$ cross sections also have been computed from statistical theory for comparison with $(n,2p)$ values and are given in Table X. Here the $(n,2n)$ and (n,pn) values were calculated in a manner analogous to that described above for $(n,\alpha n)$ reactions, whereas the $(n,p\gamma)$ values

TABLE X. Theoretical cross sections for nuclides most likely to give the $(n,2p)$ reaction at 14.7 MeV.

Target nuclide	Method[a]	Cross sections in millibarns for reactions $(n,2n)$	(n,np)	(n,pn)	$(n,2p)$	$(n,p\gamma)$	$(n,\alpha n)$	$(n,\alpha\gamma)$
S^{33}	A	530	0	51	14	70	58	35
S^{33}	B	482	0	34	0	102	42	47
Ti^{47}	A	795	0	39	48	39	10.5	15
Ti^{47}	B	722	0	16	27	83	5.5	20
Cr^{50}	A	121	224	252	133	157	3.5	77
Cr^{50}	B	33	209	142	44	355	0.08	74
Fe^{54}	A	126	298	390	139	139	2.9	33
Fe^{54}	B	18	309	250	4.2	370	0.19	36
Ni^{58}	A	207	137	365	84	54	148	212
Ni^{58}	B	127	178	288	38	180	70	290
Se^{74}	A	586	342	164	54	56	64	64
Se^{74}	B	358	451	104	0	171	34	95
Se^{77}	A	1300	0	13	18	6.6	2.4	12
Se^{77}	B	1270	0	4.6	7.5	25	5.9	8.2
Kr^{78}	A	600	255	188	102	27	22	35
Kr^{78}	B	352	500	119	88	113	10	47
Mo^{92}	A	460	742	166	87	46	10	28
Mo^{92}	B	127	956	101	103	65	3.8	35

[a] For reactions of the type (n,ji), method A assumes $P_i=0$ if $E_j>(E_{\max}-P_i)$, whereas method B assumes $P_i=0$ if $E_j>(E_{j\,\max}-B_i-K)$, where K is 2 for i=protons and 1 for i=neutrons. Both Method A and Method B assume $P_n\gg P_p\gg P_\gamma\gg P_\alpha$ (see discussion in text).

[81] S. T. Butler, Proc. Roy. Soc. (London) **A208**, 559 (1951); and *Nuclear Stripping Reactions* (John Wiley & Sons, Inc., New York, 1957); Phys. Rev. **106**, 272 (1957); and S. T. Butler and N. Austern, *ibid.* **92**, 350 (1953).

were computed in the same manner as for the $(n,\alpha\gamma)$ cross sections.[40]

The most probable cases for occurrence of the $(n,2p)$ reaction are those for which the (S_n-S_p) difference is the largest. Q-value tables[79,80] were searched for the most favored cases, which are S^{33}, Ar^{36}, Ti^{47}, Ti^{49}, Cr^{50}, Fe^{54}, Ni^{58}, Zn^{64}, Zn^{67}, Se^{74}, Se^{77}, Kr^{78}, Kr^{80}, Kr^{83}, Mo^{92}, Mo^{94}, Mo^{95}, Mo^{97}, Rh^{96}, and Rh^{98}, among nuclei up to $A=100$. Unfortunately, none of these yield $(n,2p)$ products which are radioactive, except the krypton isotopes, which were not studied because of experimental problems with gaseous targets.

As an example that these nuclides should likely give appreciable $(n,2p)$ reactions, we discuss the competition of 14.7-MeV neutron reactions with Ni^{58}, which gives an excited compound nucleus Ni^{59*}, which may decay by proton emission to excited Co^{58*} or by neutron emission to excited Ni^{58*}. Since the level density of odd-odd Co^{58} is some 15 times higher than that of even-even Ni^{58} [Eq. (9[75])], de-excitation of Ni^{59*} to Co^{58*} by proton emission is significant in spite of the fact that the Coulomb barrier for protons is greater than any angular momentum barrier for neutrons. Thus, we find at 14.7-MeV bombarding energy that the $Ni^{58}(n,p)$ cross section is about 300–400 mb,[51] whereas the (n,n') cross section (of natural nickel) is only about 900 mb,[72] although generally at 14–15 MeV, $\sigma_{(n,p)} \approx 0.1\sigma_{(n,n')}$. Now, when we consider the decay of excited Ni^{58*} (resulting from the (n,n') reaction), we find that the $Ni^{58}(n,np)/Ni^{58}(n,2n)$ cross section ratio at 14.7 MeV is about 260/40 (taking the (n,np) contribution alone, rather than the sum of such reactions) experimentally, so that proton emission obviously is greatly favored. In this case, since both residual nuclei, Co^{57} and Ni^{57}, respectively, are odd-A and have comparable level densities, the predominance of the (n,np) over the $(n,2n)$ reaction must be ascribed to the difference in the neutron and proton separation energies, $(S_n-S_p)=4$ MeV.

Similarly, decay of excited Co^{58*} (from the $Ni^{58}(n,p)$ reaction) by emission either of a proton or a neutron leads to odd-A products, so that the level density difference is not important, and since the separation energy difference (S_n-S_p) is about 1.5 MeV, we may still expect considerable proton emission; that is, the $Ni^{58}(n,2p)$ reaction should be appreciable (Table X). In principle, the contribution of the $(n,2p)$ reaction with Ni^{58} could be estimated by subtracting the (n,np) cross section, as measured by activation, from the cross section arising from the "excess" of low-energy protons reported in emitted particle studies. Unfortunately, the discordant experimental results of the latter (Table I) do not permit such an estimate to be made at present. These considerations do suggest, however, that emitted proton spectra which show an "excess" of low-energy protons may contain protons from the $(n,2p)$ as well as from the (n,np) reaction.

One of the best ways to detect the $(n,2p)$ reaction would be to observe the emitted proton pairs in coincidence using $(dE/dx)-E$ detector telescopes, such as solid state detectors, in a scattering chamber experiment. There are to date no experiments of this type reported for $(n,2p)$ reactions.

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