1.42, 1.42, and 1.41 as calculated from Rose's data.[1] The ratios of the total number of transitions to the number of gamma rays and the number of K-converted transitions are then 6.6 and 5.5, respectively. Assuming internal conversion of the high-energy transitions of Lu^{168} to be negligible, the ratios of the relative numbers of transitions in Yb^{168} are 49:10:13, respectively. The relative number of K x rays can be corrected for fluorescence by dividing the 100 K x rays observed by the K fluorescence yield in ytterbium which is 0.937.[2] The result is 107. Applying the ratio 5.5 to the 87-kev transitions implies that approximately 9 of the 107 K x rays result from internal conversion of the 87-kev transition and approximately 98 result from K capture to the levels of Yb^{168}.

Figure 1 shows a proposed energy level scheme for the decay of Lu^{168}. The approximate branching ratios of electron capture to the levels of Yb^{168} were obtained by determining the difference between the number of transitions from each level and the number of transitions into the same level. The number of K x rays remaining after correcting for internal conversion and fluorescence was used as the relative number of electron capture transitions to the ground state of Yb^{168}.

There is no information currently available in the literature concerning the radioactive decay of Lu^{168} nor have any energy levels been established in Yb^{168} by Coulomb excitation. The natural abundance of the 168 mass number in ytterbium is only 0.14%.

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Cross Sections for the $(n,2n)$ Reaction in N^{14}, P^{31}, Cu^{63}, and Pr^{141}

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The $(n,2n)$ cross sections have been measured for N^{14}, P^{31}, Cu^{63}, and Pr^{141}, for neutron energies from 12.5 to 18 Mev. The annihilation radiation emitted from the product nuclides was counted with two NaI(Tl) crystals in coincidence. In the energy range measured, the cross sections were found to vary, as follows: N^{14}, 3.03 to 11.67 mb; P^{31}, 0 to 74 mb; Cu^{63}, 186 to 836 mb; Pr^{141}, 1231 to 1737 mb. The results are generally in agreement with those of others. The data are compared with curves plotted from Weisskopf's theoretical expression for $(n,2n)$ cross sections.

INTRODUCTION

ALTHOUGH many $(n,2n)$ cross-section measurements have been made for 14-Mev neutrons,[1–7] relatively few measurements have been made over a range of neutron energies.[8–11] Weisskopf and Ewing presented an approximate theoretical equation for the variation of the $(n,2n)$ cross section for nuclides with $A>50$ as a function of neutron energy as long ago as 1940.[12] However, this equation possesses two parameters (the cross section for the emission of one neutron from the compound nucleus and the nuclear temperature) which in general are not known, and hence it is difficult to compare the theory with experimental data at only one energy.

It was, therefore, decided to take advantage of the unique energy-angle relationship of the neutrons produced by the $T(d,n)He^4$ reaction to measure some $(n,2n)$ reactions in the range from 12 to 18 Mev. Four nuclides were studied: N^{14}, P^{31}, Cu^{63}, and Pr^{141}. These were chosen to give a wide range in atomic weight and for experimental convenience (each of the product nuclides, N^{13}, P^{30}, Cu^{62}, and Pr^{140}, is a positron emitter). The yield of the reaction for a given neutron

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bombardment was obtained by counting the annihilation radiation from the product nuclide.

EXPERIMENTAL APPARATUS AND TECHNIQUES

Neutrons were produced by the $H^3(d,n)He^4$ reaction, with the NRDL Van de Graaff accelerator providing a beam of 1.80-Mev deuterons. The target consisted of a 0.025-inch thick platinum foil onto which had been evaporated a 1 mg/cm^2 layer of zirconium which was later converted to ZrH^3, with the H^3 content being approximately 0.4 curie.[13]

The target materials were melamine ($N_6C_3H_6$) for nitrogen, elemental phosphorus and copper, and a mixture of oxides with the effective formula Pr_6O_{11} for praseodymium. All of the samples were in a powdered form, and were placed in $\frac{7}{16}$-inch diameter test tubes, filled to a depth of one inch.

On a typical run, the sample was held in a thin aluminum holder at the appropriate angle and at a distance of 9 cm from the tritium target. The sample was irradiated for about two half-periods of the product nuclide, and then was placed in position in the annihilation radiation counter within 60 seconds. The sample was counted for about two half-periods.

The time-integrated neutron flux for each run was computed using as data the counts from a 1.5-inch diameter, 0.5-inch height $Li^6I(Eu)$ scintillation counter. The technique for using this crystal as a neutron counter has been previously discussed.[14,15] The $Li^6I(Eu)$ crystal was placed at an angle of 110 degrees to the deuteron beam, and the neutron flux at the sample position was computed from the differential relative cross-section values of Bame and Perry.[16]

Since the bombarding time was two half-periods, it was necessary to correct the time-integrated neutron flux for the number of product nuclei which decayed during the bombardment. An expression for the ratio of nuclei surviving to a time T to the total number of nuclei produced during a bombardment may be obtained in the following manner. The number of nuclei produced between times t and $t+dt$ is proportional to φdt where φ is the neutron flux per unit time. Of these nuclei, the number left at a later time T is proportional to $e^{-\lambda(T-t)}\varphi dt$, where λ is the decay constant of the radionuclide being produced. If we integrate φdt over the neutron bombarding time we obtain the total number of nuclei produced, while if we integrate $e^{-\lambda(T-t)}\varphi dt$ we obtain the total number surviving to a time T. Hence,

$$s=\frac{\text{number of atoms left at end of bombardment}}{\text{number of atoms produced during bombardment}}$$

$$=\int_0^T e^{-\lambda(T-t)}\varphi dt \Big/ \int_0^T \varphi dt. \quad (1)$$

This quantity (called the "survival factor") was determined by feeding the output of the Li^6I neutron monitor into an electronic device similar to a count-rate meter. The RC time constant of this "count-rate meter" was set equal to the mean life of the product nuclide under study. The output current of the count-rate meter is proportional to the numerator of Eq. (1),[17] while the denominator is proportional to the total number of input pulses to the count-rate meter. Thus s was determined from the readings of the count-rate meter and a scaler monitoring the output of the Li^6I crystal.

The annihilation-radiation counter consists of two 4-inch diameter 4-inch height NaI(Tl) crystals placed 6 cm apart. After bombardment the test tube containing the sample was placed in an aluminum container thick enough to stop the positrons between the NaI crystals. After amplification, the output of each crystal was fed into a differential discriminator. The differential discriminator outputs were run in coincidence and the coincidence counts were recorded with a scaler. The windows of the differential discriminators were set to accept pulses from the full-energy peak of the 0.511-Mev annihilation radiation. The counter was calibrated with a standard Na^{22} source after each run to correct for any drift in the electronic equipment.

The efficiency of the counter was obtained with a Na^{22} source prepared by the Chemical Technology Division of NRDL. Small correction factors had to be applied to the efficiency to correct for the different ranges of the positrons, and for the effect of the 1.27-Mev gamma ray in Na^{22}. The correction for the gamma ray was obtained by studying the Na^{22} singles and coincidence spectra with a 100-channel analyzer. The correction for the different ranges of the positrons was obtained by moving a small Na^{22} source around to determine the effects of small spatial displacements of the source. These corrections were never more than 10%.

Also, high-activity samples of N^{13}, P^{30}, Cu^{62}, and Pr^{140} were produced by taping the small test tubes directly to the tritium target and irradiating. These samples were counted both in coincidence and singly for comparison with the Na^{22} standard. Checks were made on the half-life of the activity produced. The gamma-ray pulse-height spectra were examined for interfering activity or unreported gamma rays. All

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TABLE I. $(n,2n)$ cross sections in millibarns.

Neutron energy (Mev)	Energy spread (Mev)	N^{14} $\sigma_{n,2n}$ (mb)	N^{14} Standard deviation (mb)	P^{31} $\sigma_{n,2n}$ (mb)	P^{31} Standard deviation (mb)	Cu^{63} $\sigma_{n,2n}$ (mb)	Cu^{63} Standard deviation (mb)	Pr^{141} $\sigma_{n,2n}$ (mb)	Pr^{141} Standard deviation (mb)
12.41	±0.12	3.03	±0.75			186	±19	1231	±111
12.81	±0.15					233	±21		
13.77	±0.20	5.18	±0.6			378	±34	1386	±125
14.74	±0.27	8.69	±0.9	8.7	±2.7	507	±45	1591	±143
15.78	±0.32	9.25	±1.0			649	±58	1737	±156
16.96	±0.34	10.49	±1.0	43.6	±5.2	758	±68	1606	±145
17.98	±0.24	11.57	±1.2	74.0	±7.4	836	±75	1667	±150

the data obtained agreed with the literature,[18] except for the half period of Pr^{140}, which was found to be 200±5 seconds.

RESULTS

The number n of radioactive nuclei which are present at the start of the positron counting depends on the one hand on the cross section σ; on the other hand, n may be expressed in terms of the number of positron coincidence counts A:

$$\varphi\sigma LWsa/G=n=A(1-e^{-\lambda t})^{-1}/p\eta. \quad (2)$$

The remaining symbols in the equation are φ, the time-integrated neutron flux density at the midplane of the sample; L, Avogadro's number; W, the weight of the sample; s, the survival factor; a, the relative isotopic abundance of the target isotope; G, the target gram atomic weight; λ, the radioactive decay constant; p, the number of positrons per disintegration; η, the efficiency of the annihilation-radiation counter; and t, the length of time that the annihilation radiation is counted. The cross sections have been evaluated from Eq. (2). The number of positrons per disintegration is taken to be 1.00 for N^{13} and P^{30}, 0.98 for Cu^{62}, and 0.54 for Pr^{140}.

The results are tabulated in Table I and plotted in Fig. 1. Each point on the experimental curves is based on from two to six runs, with the number of annihilation-radiation counts varying from 20 to 1000. The results of other experimenters[3–9] are shown in the figures for comparison.

The significant sources of error and an estimate of their magnitudes are: (1) uncertainty in the $Li^6(n,t)He^4$ cross section, 5%[14]; (2) uncertainty in the relative differential cross section for the $H^3(d,n)He^4$ reaction, 4%; (3) statistical error in the number of neutron monitor counts, 2 to 3%; (4) uncertainty in the survival factor, 2%; (5) uncertainty in the annihilation-radiation counter efficiency, 5%; (6) uncertainty in the sample position with respect to the target, 2%; (7) statistical error in the number of annihilation-radiation counts, 1 to 15%. The first, second, and fifth sources of error introduce systematic errors; the others are expected to vary from run to run.

DISCUSSION

The following approximate equation for the $(n,2n)$ cross section, based on the compound nucleus model of nuclear reactions, is given by Weisskopf and Ewing[11]:

$$\sigma_{n,2n}=\sigma(n,n_1)[1-(1+\epsilon/\Theta)e^{-\epsilon/\Theta}], \quad (3)$$

where Θ is the temperature in Mev of the target nucleus at the bombarding energy, and ϵ is the difference between the incident neutron energy and the $(n,2n)$ threshold energy E_{th}. The quantity $\sigma(n,n_1)$ represents the cross section for the emission of at least one neutron from the compound nucleus, and ideally would be equal to the sum of the cross sections of all the reactions which involve emission of at least one neutron from the compound nucleus. In this equation, however, the competing effect of other secondary reactions, such as (n,np), are ignored. Hence the equation would be expected to overestimate the $(n,2n)$ cross section as the energy is increased to the point where secondary and tertiary reactions become important.

To compare the equation with the experimental data, one must obtain values for E_{th}, $\sigma(n,n_1)$, and Θ. The threshold energies were computed from data given by Ajzenberg-Selove and Lauritsen,[19] Sullivan,[20] and Way et al.[21] For the four nuclides considered, the neutron inelastic scattering cross section has been measured at 14 Mev only for nitrogen.[22] However, accurate values for the nonelastic cross section are available.[23] For the purposes of this paper it is assumed that

$$\sigma(n,n_1)=\sigma_{ne}-(\sigma_{np}+\sigma_{n\alpha}), \quad (4)$$

where $\sigma(n,n_1)$ is the cross section for the emission of the first neutron from the compound nucleus, as in

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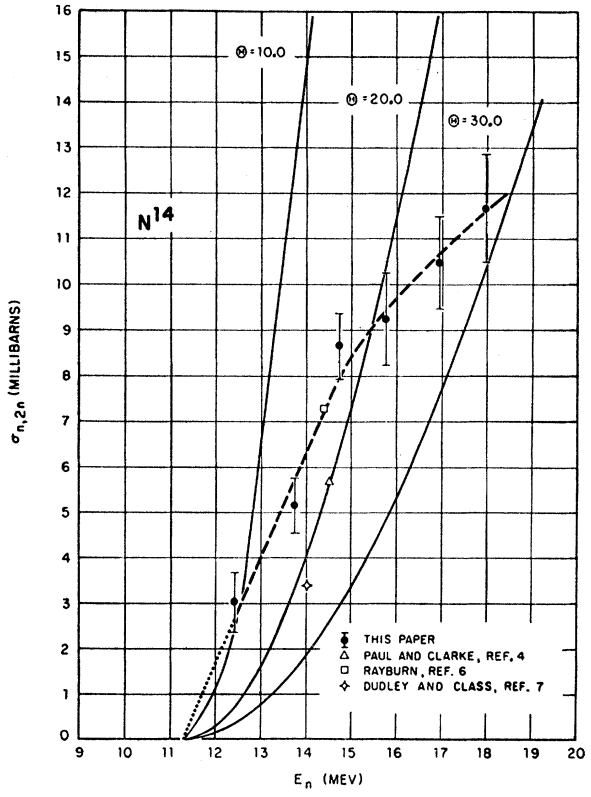
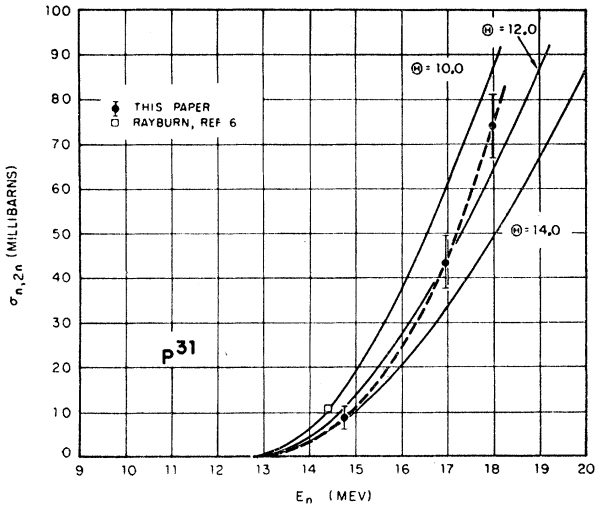


(a)



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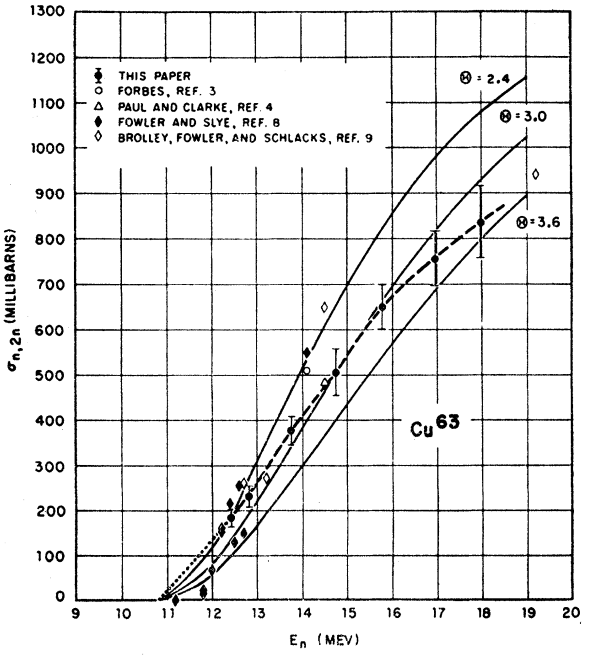

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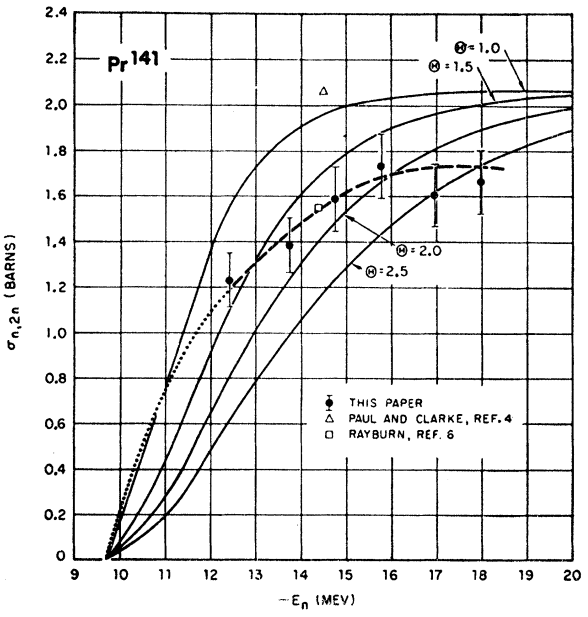

(d)

FIG. 1. The solid lines are theoretical estimates of the $(n,2n)$ cross sections using Eq. (3), where the values of $\sigma(n,n_1)$ are given in Table II, and the values of Θ are indicated on the figure. The points are the experimental cross sections given in this paper and by other investigators. The symbols representing each of the investigators are shown in the relevant figure.

TABLE II. Data used for theoretical estimates.

	σ_{ne} (barns)	σ_{np} (barns)	$\sigma_{n\alpha}$ (barns)	$\sigma(n,n_1)$ (barns)	E_{th} (Mev)
N^{14}				0.48[e]	11.30[g]
P^{31}	1.13[a]	0.077[b]	0.146[b]	0.91[f]	12.78[h]
Cu^{63}	1.49[a]	0.120[c]	0.030[d]	1.35[f]	10.82[i]
Pr^{141}	2.11[a]	0.050[d]	0.01[d]	2.06[f]	9.70[h]

[a] See reference 23.
[b] See reference 4.
[c] See reference 25.
[d] Estimated from cross sections of neighboring nuclides.
[e] See reference 22.
[f] Calculated from Eq. (4).
[g] See reference 19.
[h] Calculated from nuclear masses from reference 20.
[i] See reference 21.

Eq. (3), σ_{ne} is the nonelastic cross section, and σ_{np} and $\sigma_{n\alpha}$ are the cross sections for proton and α-particle emission. The values used for estimating $\sigma(n,n_1)$ are given in Table II. Where there were no measured values available for σ_{np} or $\sigma_{n\alpha}$, the cross section was estimated from those of neighboring nuclides.[4,24,25]

In using Eqs. (3) and (4), we have ignored the effect of other competing reactions, such as $\sigma(n,np)$ and $\sigma(n,d)$. Also, since Eq. (3) is based on the compound nucleus model of nuclear reactions, we have ignored the effect of direct interactions. Solutions of Eq. (3) for the $\sigma(n,n_1)$ values given in Table II, and several different values of Θ are plotted with the experimental results in Fig. 1.

Values of Θ were chosen so that the theoretical $\sigma_{n,2n}$ curves bracket the experimental ones. For the isotopes with higher mass numbers (Pr^{141} and Cu^{63}), these values for Θ are reasonable when compared with those estimated on p. 372 of Blatt and Weisskopf.[26] For the isotopes with lower mass numbers (P^{31} and N^{14}) much larger values of Θ than those estimated in reference 26 have to be used to bracket the experimental curves. These considerations suggest that compound nucleus formation is a relatively less important mechanism for the $(n,2n)$ reaction in isotopes with low mass numbers. In particular, Hassler, Zatzick, and Eubank[27] have obtained angular distributions of emitted particles for the $N^{14}(n,p)$, $P^{31}(n,p)$, $N^{14}(n,d)$, and $P^{31}(n,d)$ reactions for 14-Mev neutrons, and all their data show angular distributions characteristic of the direct interaction process.

In the direct interaction process, the average energy of the "first neutron" given off in an interaction is expected to be larger than for a compound nucleus reaction. Hence there are fewer cases when there is enough energy available for a second neutron to escape and the $(n,2n)$ cross section is expected to be smaller.

The Cu^{63} cross sections show the best agreement with Eq. (3). The data of Rosen et al.[28] indicate that the compound nucleus process accounts for 85 to 90% of the inelastic interactions for medium and heavy weight nuclides, so one would expect reasonable agreement for Cu^{63}. The experimental cross section for Cu^{63} appears to increase less rapidly than the theory predicts at the higher energies. This can be attributed to the effect of secondary nuclear reactions which were neglected in Eqs. (3) and (4).

The Pr^{141} cross section appears to be decidedly lower than theory predicts, especially at higher energies. As in the case of Cu^{63}, this discrepancy is attributed to the effect of secondary reactions. The threshold for the $Pr^{141}(n,np)$ reaction is about 5.5 Mev, and the threshold for the $Pr^{141}(n,d)$ reaction is about 3.3 Mev. It is, therefore, not unreasonable to assume that in the high-energy region these reactions are important enough to account for the difference between the theroretical curves for $\Theta=1$ or 1.5 Mev and the observed cross sections.

In conclusion, it appears that Eq. (3) overestimates the $(n,2n)$ cross section for nuclides with low mass numbers for reasonable values of the nuclear temperature. This lack of agreement is consistent with the view that the direct interaction process predominates for nuclides with low mass numbers. For nuclides with medium and high mass numbers, the equation agrees reasonably well near the threshold, but overestimates the cross section at higher energies where other secondary reactions not included in Eq. (3) begin competing effectively with the $(n,2n)$ process.

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