

## Some Cross-Section Limits for the Occurrence of the $(n, \text{He}^3)$ Reaction at 14.5 Mev\*

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The  $(n, \text{He}^3)$  reaction at  $14.5 \pm 0.9$  Mev has been investigated carefully with scintillation spectroscopy and radiochemical techniques. No case of an  $(n, \text{He}^3)$  reaction could be detected at this energy. Upper limits were set on the  $(n, \text{He}^3)$  cross section for the following target nuclides:  $\text{Mg}^{26}$ (3.0),  $\text{Al}^{27}$ (0.70),  $\text{P}^{31}$ (0.25),  $\text{K}^{41}$ (2.5),  $\text{V}^{51}$ (0.33),  $\text{Mn}^{55}$ (0.42),  $\text{Co}^{59}$ (0.10),  $\text{Cu}^{63}$ (0.08),  $\text{As}^{75}$ (0.51),  $\text{Zr}^{94}$ (0.4),  $\text{Rh}^{103}$ (0.09), and  $\text{Cs}^{133}$ (0.15), where the value in parentheses is the upper limit in millibarns. One case of the  $(n, n'\alpha)$  reaction was detected at this energy, on  $\text{Cu}^{65}$  with a cross section of a few millibarns.

### INTRODUCTION

RECENTLY, four examples ( $\text{Mn}^{55}$ ,  $\text{Co}^{59}$ ,  $\text{As}^{75}$ , and  $\text{Rh}^{103}$ ) of the  $(n, \text{He}^3)$  reaction at 14.5 Mev were reported to occur with cross sections of the order of a few millibarns.<sup>1</sup> During a study in this laboratory of neutron reactions of arsenic, however, radiochemical separation revealed an activity in the gallium fraction which could be ascribed only to the  $(n, \alpha)$  product, no activity arising from a possible  $(n, \text{He}^3)$  reaction being detectable. Therefore, it became necessary to reinvestigate the four cases reported previously,<sup>1</sup> especially as a similar investigation<sup>2,3</sup> of the  $(n, t)$  reaction at this energy indicated that cross sections in the region of microbarns, rather than millibarns, were to be expected, as it would be rather a surprise if the  $(n, \text{He}^3)$  reaction were to predominate over the  $(n, t)$  reaction in this energy region.

### EXPERIMENTAL

Measurements of activation cross sections were made with  $14.5 \pm 0.9$ -Mev neutrons (at  $0^\circ$  to the beam) produced by the  $\text{H}^3(d, n)\text{He}^4$  reaction in the Arkansas 400-kv Cockcroft-Walton accelerator. Total neutron yields between  $10^9$  and  $10^{11}$  neutrons/sec were obtained from ZrT or TiT targets. Target samples consisted of metallic foils or powders of the best commercial purity. In the case of copper, isotopically enriched oxide samples were used. During irradiation, samples were covered with cadmium foil which effectively removes thermalized neutrons.

In most of the samples, the  $(n, p)$ ,  $(n, \alpha)$ , or  $(n, 2n)$  reaction on the target nuclide served as an internal flux monitor for determining cross-section limits. Thus, errors arising from differences in geometry are eliminated. Moreover, when the  $(n, \alpha)$  product serves as the internal monitor, any errors which might result from

uncertainties in the yield of radiochemical separation are avoided. In bombardments of magnesium and rhodium, thin foils of copper (0.00025 in.) and of aluminum (0.001 in.), respectively, were placed in front and in back of the sample to serve as monitors by means of the  $\text{Cu}^{63}(n, 2n)\text{Cu}^{62}$  (556 mb) and  $\text{Al}^{27}(n, p)\text{Mg}^{27}$  (87 mb) reactions. For each case, the monitoring reaction used is listed in Table I, together with its appropriate cross section. For those  $(n, p)$  reactions used as monitors, the systematics compiled by Gardner<sup>4</sup> was used to select a "best" value of the  $(n, p)$  cross section; for other monitoring reactions, an average of literature values was taken (see Table I).

Gross beta counting and scintillation spectroscopy were carried out on various samples both with and without radiochemical separation. For beta counting, the decay was followed with an aluminum-walled methane-flow proportional counter fitted with a 0.9-mg/cm<sup>2</sup> aluminized Mylar end window. Gamma spectroscopy was performed with a  $3 \times 3$ -in. NaI(Tl) scintillation spectrometer and RIDL 200-channel analyzer.

Absolute  $2\pi$  beta counting for cross-section determinations was made as precise as possible by the use of thin samples mounted on aluminum saturation backscattering shelves and by taking into account corrections<sup>5</sup> for counting efficiency, air-window absorption, saturation backscattering, self-absorption—self-scattering, and background.

When beta decay was followed, the method employed for placing an upper limit on the  $(n, \text{He}^3)$  reaction was to add a line with a slope corresponding to the activity of  $(n, \text{He}^3)$  product to the decay line of the longer lived activities present. A sufficient amount was added so that if such a decaying nuclide had been present, then a change in the slope of the decay curve would have been evident.

Similarly, limits were obtained from the gamma spectra by drawing in photopeaks corresponding to gammas which would result from the products of an  $(n, \text{He}^3)$  reaction and comparing with a photopeak from

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<sup>2</sup> A. P. Baerg and G. C. Bowes, Can. J. Chem. **39**, 684 (1961).

<sup>3</sup> D. M. Chittenden, D. G. Gardner, and R. W. Fink, Phys. Rev. **122**, 860 (1961).

<sup>4</sup> D. G. Gardner, Nuclear Phys. (to be published).

<sup>5</sup> A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959); R. G. Wille and R. W. Fink, *ibid.* **118**, 242 (1960); **112**, 1950 (1958).

TABLE I. Upper limits to  $(n, \text{He}^3)$  reaction cross sections at 14.5 Mev.

Reaction	Q value (Mev)	Product half-life <sup>a</sup>	Monitor reaction and cross section	$(n, \text{He}^3)$ cross-section limit (mb)
$\text{Mg}^{26}(n, \text{He}^3)\text{Ne}^{24}$	-17.125 <sup>a</sup>	3.38 min	$\text{Cu}^{63}(n, 2n)\text{Cu}^{62}$ (556 mb) <sup>d</sup>	<3.0
$\text{Al}^{27}(n, \text{He}^3)\text{Na}^{25}$	-14.654 <sup>a</sup>	60 sec	$\text{Al}^{27}(n, p)\text{Mg}^{27}$ (87 mb) <sup>e</sup>	<0.70
$\text{P}^{31}(n, \text{He}^3)\text{Al}^{29}$	-13.229 <sup>a</sup>	5.56 min	$\text{P}^{31}(n, 2n)\text{P}^{30}$ (11.9 mb) <sup>f</sup>	<0.25
$\text{K}^{41}(n, \text{He}^3)\text{Cl}^{39}$	-12.599 <sup>a</sup>	55.5 min	$\text{K}^{41}(n, \alpha)\text{Cl}^{38}$ (40 mb) <sup>g</sup>	<2.5
$\text{V}^{51}(n, \text{He}^3)\text{Sc}^{49}$	-12.600 <sup>a</sup>	57 min	$\text{V}^{51}(n, p)\text{Ti}^{51}$ (53 mb) <sup>e</sup>	<0.33
$\text{Mn}^{55}(n, \text{He}^3)\text{V}^{53}$	-13.180 <sup>b</sup>	1.7 min	$\text{Mn}^{55}(n, \alpha)\text{V}^{52}$ (40 mb) <sup>h</sup>	<0.42
$\text{Co}^{59}(n, \text{He}^3)\text{Mn}^{57}$	-11.625 <sup>a</sup>	1.7 min	$\text{Co}^{59}(n, \alpha)\text{Mn}^{56}$ (40 mb) <sup>g</sup>	<0.10
$\text{Cu}^{63}(n, \text{He}^3)\text{Co}^{61}$	-9.528 <sup>a</sup>	1.65 hours	$\text{Cu}^{63}(n, 2n)\text{Cu}^{64}$ (954 mb) <sup>i</sup>	<0.08
$\text{As}^{75}(n, \text{He}^3)\text{Ga}^{73}$	-9.894 <sup>a</sup>	5 hours	$\text{As}^{75}(n, \alpha)\text{Ga}^{72}$ (12 mb) <sup>g</sup>	<0.51
$\text{Zr}^{94}(n, \text{He}^3)\text{Sr}^{92}$	-12.9	2.71 hours	$\text{Zr}^{94}(n, \alpha)\text{Sr}^{91}$ (4.1 mb) <sup>j</sup>	<0.4
$\text{Rh}^{103}(n, \text{He}^3)\text{Tc}^{101}$	-8.188 <sup>b</sup>	14 min	$\text{Al}^{27}(n, p)\text{Mg}^{27}$ (87 mb) <sup>e</sup>	<0.09
$\text{Cs}^{133}(n, \text{He}^3)\text{I}^{131}$	-7.782 <sup>a</sup>	8.05 days	$\text{Cs}^{133}(n, \alpha)\text{I}^{130}$ (1.0 mb) <sup>k</sup>	<0.15

<sup>a</sup> V. J. Ashby and H. C. Catron, University of California Radiation Laboratory Report UCRL-5419, TID-4500 (14th ed.) UC-34, 1959 (unpublished).

<sup>b</sup> A. G. W. Cameron, Atomic Energy of Canada, Limited Report CRP-690 (AECL-433), 1958 (unpublished).

<sup>c</sup> *Nuclear Data Sheets* (National Academy of Science, Washington, D. C., 1960).

<sup>d</sup> S. Yasumi, J. Phys. Soc. (Japan) 12, 443 (1957).

<sup>e</sup> See reference 4.

<sup>f</sup> L. A. Rayburn, Bull. Am. Phys. Soc. 3, (7), 365 (1958).

<sup>g</sup> E. B. Paul and R. L. Clarke, Can. J. Phys. 31, 267 (1953); *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (U. S. Government Printing Office, Washington, D. C., 1955).

<sup>h</sup> See reference 7.

<sup>i</sup> See reference 10.

<sup>j</sup> See reference 15.

<sup>k</sup> H. G. Blosser, C. D. Goodman, and T. H. Handley, Phys. Rev. 110, 531 (1958).

the monitor, making the usual corrections for crystal efficiency and peak-to-total ratio. Individual cases are discussed below.

### Magnesium

Magnesium foil, 4.0 mg/cm<sup>2</sup>, monitored by 3-mg/cm<sup>2</sup> copper foils fore and aft, was irradiated. No  $(n, \text{He}^3)$  product activity (3.38-min Ne<sup>24</sup>) was detectable, by beta proportional counting, the cross section being, therefore, less than 3.0 mb (Table I). Owing to the difficulty of resolving 38.0-sec Ne<sup>23</sup>, formed by the  $\text{Mg}^{26}(n, \alpha)$  reaction, from 60-sec Na<sup>25</sup>, formed by the  $\text{Mg}^{25}(n, p)$  reaction,<sup>6</sup> the total cross section of these two reactions together is estimated to be 160 mb, in good agreement with the literature value of 146 mb.<sup>6</sup>

### Aluminum

Aluminum foil, 2.3 mg/cm<sup>2</sup>, was irradiated, but no activity arising from an  $(n, \text{He}^3)$  product (60-sec Na<sup>25</sup>) was detectable in the gross beta decay. Na<sup>25</sup> emits betas having endpoint energy above 2 Mev. A limit on the cross section was set, assuming a value of 87 mb for the  $\text{Al}^{27}(n, p)\text{Mg}^{27}$  reaction<sup>4</sup> (Table I).

### Phosphorus

P<sub>2</sub>O<sub>5</sub> was irradiated, and no 6.5-min Al<sup>29</sup> activity (decaying with 1.5- and 2.7-Mev betas) from an  $(n, \text{He}^3)$  reaction was detectable in the gross beta decay. An

upper limit of 250  $\mu\text{b}$  was set, based on the  $\text{P}^{31}(n, 2n)\text{P}^{30}$  reaction (11.9 mb) (Table I).

### Vanadium

Vanadium foil, 9 mg/cm<sup>2</sup>, was irradiated, and two activities could be resolved in the gross beta decay, 6.0 $\pm$ 0.5-min Ti<sup>51</sup> and 44 $\pm$ 2-hr Sc<sup>48</sup>, from the  $(n, p)$  and  $(n, \alpha)$  reactions on V<sup>51</sup>, respectively. An upper limit of 330  $\mu\text{b}$  on the  $\text{V}^{51}(n, \text{He}^3)$  reaction was set, based on the  $\text{V}^{51}(n, p)$  cross section of 53 mb (Table I), since the product, Sc<sup>49</sup>, of the  $(n, \text{He}^3)$  reaction decays with the emission of 2.9-Mev betas.

### Manganese

Spectroscopically pure Mn<sub>2</sub>O<sub>4</sub> was irradiated and counted in an attempt to find the  $(n, \text{He}^3)$  product, 1.7-min V<sup>53</sup>, which emits a 2.5-Mev beta in coincidence with a 1.0-Mev gamma. The gross beta decay exhibited a 3.7-min activity only, attributed to an admixture of 3.75-min V<sup>52</sup>, from the  $(n, \alpha)$  reaction, and 3.6-min Cr<sup>55</sup>, from the  $(n, p)$  reaction. On the assumption that  $\sigma_{(n, p)} \simeq \sigma_{(n, \alpha)} \simeq 40$  mb,<sup>7</sup> an upper limit for the  $\text{Mn}^{55}(n, \text{He}^3)\text{V}^{53}$  reaction could be set at 1.5 mb. A further attempt to detect 1.7-min V<sup>53</sup> was made by means of gamma scintillation spectrometry, in order to find the strong 1.0-Mev gamma<sup>8</sup> in V<sup>53</sup> decay. In order to confirm the fact that V<sup>53</sup> exhibits a strong 1.0-Mev gamma decay, enriched Cr<sup>53</sup> was irradiated to produce the  $(n, p)$  reaction, and a strong gamma at 1.0 Mev and a much

<sup>6</sup> E. B. Paul and R. L. Clarke, Can. J. Phys. 31, 267 (1953); M. J. Nurmi and R. W. Fink, Nuclear Phys. 8, 139 (1958); and D. L. Allen, *ibid.* 24, 274 (1961).

<sup>7</sup> I. Kumabe, J. Phys. Soc. Japan 13, 325 (1958); D. L. Allen, Nuclear Phys. 24, 274 (1961).

<sup>8</sup> A. W. Schardt and B. J. Dropesky, Bull. Am. Phys. Soc. 1, 162 (1956).

weaker 1.3-Mev gamma were observed, the relative intensities being about 12:1, respectively. In the irradiated samples of manganese, no 1.0-Mev gamma was present after subtraction of the 1.4-Mev gamma from  $\text{V}^{52}$ , present from the  $\text{Mn}^{55}(n, \alpha)$  reaction; thereby, an upper limit on the  $\text{Mn}^{55}(n, \text{He}^3)$  reaction could be set by extrapolating the 1.4-Mev photopeak area to the end-of-bombardment, correcting for the peak-to-total ratio and the efficiencies of the 1.4-Mev and 1.0-Mev gammas, and using a value of 40 mb for the  $\text{Mn}^{55}(n, \alpha)$  cross section. Table I gives the most sensitive limit for the ( $n, \text{He}^3$ ) reaction of manganese.

In the earlier report<sup>1</sup> of the  $\text{Mn}^{55}(n, \text{He}^3)$  reaction, there is considerable doubt as to whether the gross beta decay curve was correctly resolved to give two components, since it was also possible to draw a single straight line corresponding to a half-life of 3.5 min through the points. Even if a 2-min activity were present, however, it undoubtedly arose from impurities, since no 2-min activity was detectable when spectroscopically pure manganese oxide was irradiated. Recently,<sup>9</sup> moreover, the "2-min isomer" in  $\text{Mn}^{54}$  has again been shown conclusively not to exist.

### Cobalt

Spectroscopically pure  $\text{Co}_3\text{O}_4$  was irradiated in an attempt to detect 1.7-min  $\text{Mn}^{57}$  from the  $\text{Co}^{59}(n, \text{He}^3)$  reaction, but no activity other than 2.6-hour  $\text{Mn}^{56}$  from the ( $n, \alpha$ ) reaction was detectable in the gross beta decay. By comparison with the ( $n, \alpha$ ) reaction cross section of 40 mb, an upper limit of 0.10 mb could be set for the  $\text{Co}^{59}(n, \text{He}^3)$  reaction (Table I). In the present investigation, the same cobalt foils which were used in the previous work<sup>1</sup> were irradiated and found to give rise to an activity of about 2-min half-life, exhibiting a gamma at 1.8 Mev and a weak peak at about 80 kev. Gammas of these energies are not reported in  $\text{Mn}^{57}$  decay. Since this 2-min activity was absent when specpure  $\text{Co}_3\text{O}_4$  was used, it seems likely that it must arise from impurities, e.g., traces of silicon in the cobalt foils, giving the  $\text{Si}^{28}(n, p)$  reaction (292 mb) and 2.3-min  $\text{Al}^{28}$  (1.8-Mev gamma). Gamma spectrometry on irradiated  $\text{Co}_3\text{O}_4$  did not exhibit the 117- or 134-kev strong gammas<sup>10</sup> associated with  $\text{Mn}^{57}$ .

### Copper

$\text{CuO}$  and  $\text{CuCl}_2$  were bombarded and subjected to radiochemical separation of cobalt activities. Yellow potassium hexacobaltinitrite was precipitated as the cobalt fraction, which revealed half-lives of 1.6 hr and 10–14 min in gross beta decay. Scintillation spectrometry of the cobalt fraction, taken after a cooling time of 80 min after irradiation showed a strong 72-kev, a weak

40-kev, and a very weak 59-kev peak, no other gammas up to 440 kev being evident. The 72-kev gamma followed a 1.6-hr half-life, which indicates its assignment to  $\text{Co}^{61}$ , the 59-kev gamma most likely arises from 10.5-min  $\text{Co}^{60m}$ , from the  $\text{Cu}^{63}(n, \alpha)$  reaction.

Since 1.6-hr  $\text{Co}^{61}$  may be formed either by the  $\text{Cu}^{63}(n, \text{He}^3)$  or the  $\text{Cu}^{65}(n, n'\alpha)$  reaction, enriched samples of  $\text{CuO}$  (i.e., 99.8%  $\text{Cu}^{63}$  and 99.64%  $\text{Cu}^{65}$ ) were irradiated in order to determine which reaction prevailed. The irradiated  $\text{Cu}^{63}\text{O}$  sample exhibited only two activities; namely, 10-min  $\text{Cu}^{62}$  and 12.8-hr  $\text{Cu}^{64}$  [from the ( $n, 2n$ ) reaction on the 0.2%  $\text{Cu}^{65}$  present]. A radiochemical separation of cobalt from irradiated enriched  $\text{Cu}^{63}\text{O}$  was carried out, the chemical yield being determined by addition of a known quantity of 5.2-yr  $\text{Co}^{60}$  tracer before the separation. No activity other than the  $\text{Co}^{60}$  tracer was detectable in the cobalt fraction. The copper fraction was recovered as the sulfide so that both the  $\text{Cu}^{63}(n, 2n)\text{Cu}^{62}$  and the  $\text{Cu}^{65}(n, 2n)\text{Cu}^{64}$  reactions could be used as monitors, the chemical recovery being determined gravimetrically. From the estimated sensitivity of these experiments, it is possible to set an upper limit for the cross section of the  $\text{Cu}^{63}(n, \text{He}^3)$  reaction at 80  $\mu\text{b}$  (Table I).

Irradiation of enriched  $\text{Cu}^{65}\text{O}$  and analysis of the gross beta decay revealed half-lives of 1.6 min [ $\text{Co}^{62m}$ , from the  $\text{Cu}^{65}(n, \alpha)$  reaction,  $\sigma = 1.9 \pm 0.6$  mb to isomeric state only from the present work], 2.6 hr [due to  $\text{Ni}^{65}$ , from the  $\text{Cu}^{65}(n, p)$  reaction,  $\sigma = 29.3 \pm 3.2$  mb determined in the present work], and 12.8 hr, [from the  $\text{Cu}^{65}(n, 2n)\text{Cu}^{64}$  monitoring reaction for which a value<sup>11</sup> of 954 mb and a beta counting efficiency of 62% for  $\text{Cu}^{64}$  were used].

The  $\text{Cu}^{65}(n, \text{He}^3)$  reaction leads to  $\text{Co}^{63}$  which has been reported to decay with a 1.41-hr half-life<sup>12</sup> which cannot be distinguished by beta counting from 1.65-hr  $\text{Co}^{61}$  from the  $\text{Cu}^{65}(n, n'\alpha)$  reaction. However, the  $\text{Cu}^{65}(n, \text{He}^3)$  reaction would seem to be ruled out perhaps by the following argument. Since the  $\text{Cu}^{63}(n, \text{He}^3)\text{Co}^{61}$  reaction has a cross section below 80  $\mu\text{b}$ , it is very unlikely that the  $\text{Cu}^{65}(n, \text{He}^3)\text{Co}^{63}$  reaction, having a  $Q$ -value 3.05 Mev less favorable, would have a cross section much larger than this. Moreover, the 1.65-hr  $\text{Co}^{61}$  product was clearly identified in the scintillation spectrum by its 72-kev gamma,<sup>13</sup> the decay of which was followed. It is concluded that the 1.6-hr activity observed belongs to  $\text{Co}^{61}$  from the  $\text{Cu}^{65}(n, n'\alpha)$  reaction, which is favored energetically because of its low threshold energy of 6.3 Mev.<sup>14</sup> Recently, Neuert and co-workers<sup>15</sup> have measured the excitation function of the  $\text{V}^{51}(n, n'\alpha)\text{Sc}^{47}$  re-

<sup>11</sup> A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959).

<sup>12</sup> I. L. Preiss and R. W. Fink, Nuclear Phys. **15**, 326 (1960).

<sup>13</sup> P. Erdős, P. Jordan, D. Maeder, and P. Stoll, Helv. Phys. Acta **28**, 323A (1955).

<sup>14</sup> R. J. Howerton, University of California Radiative Laboratory Report UCRL-5351, TID-4500, 1958 (unpublished).

<sup>15</sup> M. Bormann, S. Cierjacks, L. Langkau, H. Neuert, and H. Pollehn, J. phys. radium (to be published); and H. Neuert (private communication).

<sup>9</sup> O. Dietzsch, R. A. Douglas, and V. Gomes, Nuclear Phys. **26**, 113 (1961).

<sup>10</sup> B. L. Cohen, R. A. Charpie, T. H. Handley, and E. L. Olson, Phys. Rev. **94**, 953 (1954).

action from 12 to 19.6 Mev. Although the threshold energy for this reaction is about 11.6 Mev,<sup>13</sup> the reaction was not observed until an energy of about 16 Mev was reached. Even if the  $\text{Cu}^{65}(n,n'\alpha)$  reaction also is not detectable until the energy is some 4 to 5 Mev greater than threshold, the reaction should be observed at about 9 Mev or higher. Thus, it is concluded that the  $\text{Cu}^{65}(n,n'\alpha)$  reaction is entirely feasible at 14.5 Mev, in agreement with our observation of it with a  $2.3 \pm 1.3$  mb cross section at this energy.

Radiochemical separation of cobalt from irradiated enriched  $\text{Cu}^{65}\text{O}$  revealed in the cobalt fraction activities having 14-min and 1.6-hr half-lives, the latter followed straight for more than six half-lives. This proves clearly that 1.6-hr  $\text{Co}^{61}$  is produced by the  $\text{Cu}^{65}(n,n'\alpha)$  reaction, with a measured cross section of approximately  $2.3 \pm 1.3$  mb. The 14-min activity arises from the  $\text{Cu}^{65}(n,\alpha)\text{Co}^{62g}$  reaction, with a measured cross section of approximately  $20 \pm 10$  mb to the  $\text{Co}^{62}$  ground-state isomer.

### Arsenic

Gallium was separated radiochemically from irradiated arsenic in a search for 5-hr  $\text{Ga}^{73}$  from the  $\text{As}^{75}(n,\text{He}^3)$  reaction. A solvent extraction using HCl and isopropyl ether was performed, and the gallium fraction counted in a beta proportional counter as gallium oxinate. The 15-hr  $\text{Ga}^{72}$  produced by the  $\text{As}^{75}(n,\alpha)$  reaction (12 mb) served as an internal monitor. No detectable amount of a 5-hr activity appeared in the gallium fraction from beta decay, which sets a limit on the  $\text{As}^{75}(n,\text{He}^3)$  cross section of 510  $\mu\text{b}$  (Table I). In the previous report<sup>1</sup> on this reaction, it should be noted that radiochemical separation had not been done on irradiated arsenic. Furthermore, re-investigation of the data in the earlier report revealed the absence of a 5-hr activity when the 18-day  $\text{As}^{74}$  and 15-hr  $\text{Ga}^{72}$  activities are subtracted successively rather than subtracting just one "long-lived" activity as was done previously.

In the gallium fraction, an unidentified 15-min period was observed, which could be an impurity or possibly an isomeric state in gallium.

### Zirconium

One-gram samples of natural zirconium nitrate were irradiated, and the strontium fraction was radiochemically separated, specifically, from yttrium and zirconium. Following irradiation, the sample was dissolved in a hot solution containing strontium and yttrium carriers, and  $\text{SrSO}_4$  was precipitated with conc.  $\text{H}_2\text{SO}_4$ , which was added in excess to redissolve an ephemeral precipitate of zirconyl sulfate. A second cycle of purification of the strontium fraction was performed by dissolving the  $\text{SrSO}_4$  precipitate in dilute HCl with heat, adding hold-back carriers of yttrium and zirconium, and reprecipitating  $\text{SrSO}_4$  with excess  $\text{H}_2\text{SO}_4$ . The precipitate was washed, dried with ethanol, and heated to 110°C. The

separation requires about one hour and gives essentially a quantitative yield of strontium.

In order to distinguish  $\text{Sr}^{91}$  (9.7 hr) from  $\text{Sr}^{87m}$  (2.8 hr), the decay of the well-known gamma spectrum of each was followed and cross sections established from the gamma spectrometry. Absolute  $2\pi$  beta counting<sup>5</sup> also was performed as an additional search for the  $\text{Zr}^{94}(n,\text{He}^3)$  reaction.

The  $\text{Zr}^{94}(n,\text{He}^3)\text{Sr}^{91}$  (9.7 hr) cross section<sup>16</sup> (4.1 mb) was used as an internal monitor. Upper limits could be set at 0.4 mb (Table I) for the  $\text{Zr}^{94}(n,\text{He}^3)\text{Sr}^{92}$  (2.71 hr) reaction and at 2.6 mb for the  $\text{Zr}^{96}(n,n'\alpha)\text{Sr}^{92}$  (2.71 hr) reaction. A measurement of the  $\text{Zr}^{90}(n,\alpha)\text{Sr}^{87m}$  (2.8 hr) cross section from gamma spectrometry gave a value of  $2.8 \pm 0.4$  mb.

### Rhodium

Beta decay of the irradiated rhodium foil previously used<sup>1</sup> revealed an activity closer to 5–7 min rather than 14 min as previously reported. As a further check to show that 14-min  $\text{Tc}^{101}$  was not present after bombardment of rhodium, attempts were made to detect the 307-keV gamma which occurs in essentially 100% of the  $\text{Tc}^{101}$  decays. Scintillation spectrometry exhibited no such gamma and by using aluminum foils as flux monitors, an upper limit of 90  $\mu\text{b}$  could be set (Table I). Presumably, the short-lived activity is either an impurity or 4.4-min  $\text{Rh}^{104}$  produced from the  $(n,\gamma)$  reaction.

### Cesium

Following bombardment of  $\text{Cs}_2\text{CO}_3$ , the iodine fraction was radiochemically separated as silver iodide for beta counting. No activity other than 1.26-hr  $\text{I}^{130}$  from the  $\text{Cs}^{133}(n,\alpha)$  reaction was detectable, and an estimate of 150  $\mu\text{b}$  could be made for the  $\text{Cs}^{133}(n,\text{He}^3)$  reaction (Table I) to give 8-day  $\text{I}^{131}$ .

### DISCUSSION

Having observed no detectable case of an  $(n,\text{He}^3)$  reaction at  $14.5 \pm 0.9$  Mev (Table I), it is clear that, despite the fact that certain  $(n,\text{He}^3)$  reactions are energetically possible at this energy, the cross sections for this reaction are at least one or two orders of magnitude smaller than reported previously.<sup>1</sup>

The recent report by Baerg and Bowes<sup>2</sup> on  $(n,t)$  reactions at 14 Mev contained only one case of a detectable reaction at this energy, namely, the  $\text{S}^{32}(n,t)\text{P}^{30}$  reaction with a cross section of only  $4 \pm 1$   $\mu\text{b}$ . Upper limits of 20, 200, 75, and 100  $\mu\text{b}$ , respectively, were set for  $(n,t)$  reactions on  $\text{Ca}^{40}$ ,  $\text{Cr}^{50}$ ,  $\text{Fe}^{54}$  to  $\text{Mn}^{52g}$  (5.6 days), and  $\text{Zn}^{64}$ . One case of a fairly large  $(n,t)$  reaction was found recently<sup>3</sup> where a cross section of  $600 \pm 100$   $\mu\text{b}$

<sup>16</sup> C. H. Reed, thesis, University of Utah, February, 1960 (unpublished); H. G. Blosser, C. D. Goodman, T. H. Handley, and M. L. Randolph, Phys. Rev. **100**, 429 (1955); H. G. Blosser, C. D. Goodman, and T. H. Handley, *ibid.* **110**, 531 (1958).

was reported for the  $\text{Fe}^{54}(n, t)\text{Mn}^{52m}$  (21 min) reaction.

One would expect *a priori* that ( $n, \text{He}^3$ ) reactions near threshold should have cross sections somewhat smaller than similar ( $n, t$ ) reactions owing to the larger Coulomb barrier against  $\text{He}^3$  particle emission. Even with a diffuse nuclear surface and consequent lower barrier, one would not expect  $\text{He}^3$  emission to predominate over  $\text{H}^3$  emission near threshold. If the nuclear surface exhibited a tendency for clustering into  $\text{H}^2$ ,  $\text{H}^3$ ,  $\text{He}^3$ ,  $\text{He}^4$ , etc. groupings,<sup>17</sup> then the probability for emission of tritons and  $\text{He}^3$  particles should be about equal<sup>18</sup> if one neglects the Coulomb barrier effect.

<sup>17</sup> D. H. Wilkinson, Phil. Mag. 4, 215 (1959).

<sup>18</sup> B. L. Cohen and A. G. Rubin, Phys. Rev. 114, 1143 (1959).

Thus, the fact that we have been able to show conclusively that ( $n, \text{He}^3$ ) cross sections at this energy are not larger than the limits found for ( $n, t$ ) reactions at this energy is in agreement with *a priori* expectations.

Work in this laboratory is continuing in an attempt to study rare reactions at 14.5 Mev; e.g., ( $n, t$ ), ( $n, \text{He}^3$ ), ( $n, n'\alpha$ ), and ( $n, 2p$ ).

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### Radiative Proton Capture in $\text{O}^{18}\dagger$

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The excitation function for radiative proton capture in  $\text{O}^{18}$  has been observed at zero degrees with a 3-in.  $\times$  3-in. NaI crystal. Bias adjustments were selected in order to establish the resonances for proton capture which lead to the emission of high-energy radiation. About twenty resonances were found for protons ranging in energy from 1.6 to 3.0 Mev, and there were indications for a number of weaker additional resonances. A slow-coincidence technique was used to investigate the gamma decay schemes for  $\text{F}^{19}$  for six levels of excitation from 8.56 to 10.10 Mev. Quantitative conclusions are drawn regarding the decay of these levels to the ground "triplet" of  $\text{F}^{19}$ .

#### I. INTRODUCTION

**P**RELIMINARY results on the reaction  $\text{O}^{18}(p, \gamma)$  were previously reported from this laboratory.<sup>1</sup> The excitation function for this reaction has been measured by Butler and Holmgren<sup>2</sup> for protons up to 2 Mev, and the gamma-ray spectra at seven resonances have been reported.<sup>3</sup> The present experiment overlaps this work and extends it to 3.0 Mev. Hill and Blair<sup>4</sup> have studied energy levels in the same region through the  $\text{O}^{18}(p, \alpha)\text{N}^{15}$  and  $\text{O}^{18}(p, n)\text{F}^{18}$  reactions, and Carlson *et al.*<sup>5</sup> have observed elastic scattering and the yield of alpha particles in the bombardment of  $\text{O}^{18}$  by protons. Resonances established in these reactions are compared with those which we have observed through proton capture.

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<sup>1</sup> E. L. Hudspeth, I. L. Morgan, and J. T. Peoples, Phys. Rev. 99, 643(A) (1955); J. W. Nelson and E. L. Hudspeth, Bull. Am. Phys. Soc. 5, 109 (1960).

<sup>2</sup> J. W. Butler and H. D. Holmgren, Phys. Rev. 99, 1649(A) (1955).

<sup>3</sup> J. W. Butler and H. D. Holmgren, Phys. Rev. 116, 1485 (1959).

<sup>4</sup> H. A. Hill and J. M. Blair, Phys. Rev. 104, 198 (1956).

<sup>5</sup> R. R. Carlson, C. C. Kim, J. A. Jacobs and A. C. L. Barnard, Phys. Rev. 122, 607 (1961).

At six of the more prominent resonances for proton capture, we have observed the decay scheme of  $\text{F}^{19}$  to the "ground triplet," i.e., to the ground state or to one of the low-lying excited levels at 110 or 198 keV.

#### II. EXPERIMENT AND ANALYSIS OF DATA

##### A. Generator and Targets

This work was performed with the University of Texas Van de Graaff generator. The proton beam was analyzed by a 90° magnet. The magnetic field was measured and monitored with a proton resonance gaussmeter, and gaussmeter frequency was read directly on a Beckman EPUT meter. Absolute calibration of the gaussmeter-magnet system was accomplished on the  $\text{Li}^7(p, n)$  threshold, which was taken to be 1.8811 Mev. In order to check this calibration, three  $\text{F}^{19}(p, \alpha\gamma)$  resonances were measured at 872.7, 1346, and 1372 keV with a thin  $\text{MnF}_2$  target.

Targets used for the  $\text{O}^{18}(p, \gamma)$  studies were  $\text{ZrO}_2$ . They were prepared by heating 0.025-in. zirconium in the presence of water vapor<sup>6</sup> which was enriched to 85%

<sup>6</sup> Enriched water produced by Weizmann Institute, Rehovoth, Israel.