incident beam. Using the value[10] $\epsilon=3.1\times10^{-15}$ ev-cm^2, we find $Y=(7.7\pm1.5)\times10^{-9}$ gamma rays (9.17 Mev)/proton.

The thick target yield of radiation was measured by use of a NaI crystal 3 in. in diameter and 4 in. long, and also one 3.5 in. in diameter and 3.5 in. long. The results for the two crystals agreed satisfactorily. Measurements were made at an angle of 0° and at distances of 6 in. and 11.5 in. from the target. The efficiencies of the crystals were obtained from the computation of Miller, Reynolds, and Snow.[11] In obtaining the total yield, the angular distribution of the radiation and the isotopic composition of the target were taken into account. The value obtained was $Y=(7.4\pm3)\times10^{-9}$ gamma ray (9.17 Mev)/proton, which may be compared with the value above and with the value of 11.5×10^{-9} obtained previously by Seagrave.[12] In the latter result, however, no correction[7] has been made for the 10% branch to the level at 6.44 Mev.

[10] W. Whaling, *Handbuch der Physik* (Springer-Verlag, Berlin, 1958), Vol. 34, p. 193.

[11] Miller, Reynolds, and Snow, Rev. Sci. Instr. **28**, 717 (1957).

[12] J. D. Seagrave, Phys. Rev. **85**, 197 (1952).

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Absolute Activation Cross Sections for Reactions of Bismuth, Copper, Titanium, and Aluminum with 14.8-Mev Neutrons*

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Absolute neutron activation cross sections at 14.8 Mev have been measured for bismuth, copper, titanium, and aluminum based on comparison with the $Cu^{63}(n,2n)Cu^{62}$ reaction (556 millibarns) which served as a standard for monitoring the flux. The reactions studied, measured half-lives, and cross sections are: $Bi^{209}(n,\alpha)Tl^{206}$, 4.29±0.05 min, 1.1±0.3 mb; $Bi^{209}(n,p)Pb^{209}$, 3.31±0.03 hours, 0.83±0.40 mb; $Bi^{209}(n,\gamma)Bi^{210}$, ≤1.7 mb; $Cu^{65}(n,2n)Cu^{64}$, 12.85±0.05 hours, 954±130 mb; $Cu^{65}(n,p)Ni^{65}$, 2.56±0.20 hours, 27±11 mb; $Ti^{50}(n,p)Sc^{50}$, 1.8±0.2 min, 27±6 mb; $Ti^{50}(n,p)Sc^{50}$, 22±3 min, 48±15 mb; $Ti^{50}(n,\gamma)Ti^{51}$, ≤9 mb; $Ti^{49}(n,p)Sc^{49}$, 58±2 min, 29±5 mb; $Ti^{48}(n,p)Sc^{48}$, 44.0±0.9 hours, 58±8 mb; $Ti^{47}(n,p)Sc^{47}$, 3.45±0.06 days, 230±40 mb; $Ti^{46}(n,2n)Ti^{45}$, 3.06±0.08 hours, 50.4±8.0 mb; $Ti^{46}(n,p)Sc^{46}$, 85±2 days, ~520 mb; $Al^{27}(n,\alpha)Na^{24}$, 15.00±0.06 hours, 114±7 mb; $Al^{27}(n,p)Mg^{27}$, 9.46±0.02 min, 53±5 mb.

Comparisons of the experimental cross sections with values estimated according to the continuum theory of the compound nucleus outlined by Blatt and Weisskopf are in agreement within an order of magnitude, except in the case of the bismuth results which exhibit large discrepancies.

From irradiations of natural titanium and highly enriched Ti^{50}, a new activity was observed having a half-life of 22±3 minutes. This activity is not produced from enriched Ti^{47} or Ti^{49} samples, and it is therefore assigned tentatively to an isomer of Sc^{50}. Further work on this activity is in progress.

INTRODUCTION

Neutron activation cross sections of bismuth, copper, titanium, and aluminum have been measured at 14.8 Mev as part of a systematic study being carried out for comparison with nuclear reaction theory. Monoenergetic neutrons of 14.8±0.9 Mev energy (taken at 0° to the beam axis) are produced by the $H^3(d,n)He^4$ reaction in thin zirconium-tritium targets giving total yields ranging from 10^9 to more than 10^{11} neutrons/second on the University of Arkansas 400-kv Cockcroft-Walton accelerator.[1]

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[1] Bronner, Ehlers, Eukel, Gordon, Marker, Voelker, and Fink, Nucleonics **17**, No. 1, 94 (1959); R. W. Fink, Atomic Energy Commission Report ORO-172, 1958 (unpublished). Detailed information on the neutrons produced in the *DD* and *DT* reactions is contained in the report by J. D. Seagrave, Atomic Energy Commission Report LAMS-2162, 1958 (unpublished), J. H. Coon, in *Fast Neutron Physics*, edited by J. B. Marion and J. L. Fowler (to be published), Chap. IV. D; and by J. Benveniste and J. Zenger, Atomic Energy Commission Report UCRL-4266, 1954 (unpublished).

EXPERIMENTAL

The samples were in the form of solid metallic foils 1.7 to 2.1 cm in diameter having surface densities in mg/cm^2 as follows: copper, 3 to 6; aluminum, 3; natural titanium, 8; and bismuth, 34 to 56. Enriched samples of titanium[2] were in the form of TiO_2 powder. All target materials were of reagent-grade purity. Irradiations were carried out for periods ranging from 10 to 60 minutes, the samples being placed in contact with the back of the target plate. In the experiments in which limits were obtained for the (n,γ) cross sections at 14.8 Mev of titanium and bismuth, the samples were wrapped in cadmium sheet in order to eliminate any possible contribution from slow neutrons. The flux passing through the samples generally was of the order

[2] The enriched samples of titanium had the following mass analyses, as supplied by Oak Ridge National Laboratory: Ti^{47}, 85.6±0.3% (with 1.7±0.1% Ti^{46}, 11.3±0.2% Ti^{48}, 0.8±0.1% Ti^{49}, and 0.6±0.1% Ti^{50} as isotopic impurities); Ti^{49}, 81.5±0.2% (with 1.3±0.1% Ti^{46}, 1.3±0.1% Ti^{47}, 14.5±0.1% Ti^{48}, and 1.4±0.1% Ti^{50} as isotopic impurities); Ti^{50}, 84.69±0.04% (with 1.25±0.01% Ti^{46}, 1.23±0.04% Ti^{47}, 10.99±0.07% Ti^{48}, and 1.84±0.03% Ti^{49} as isotopic impurities.)

of 10^7 to 10^9 neutrons/cm^2-sec, as monitored by the $Cu^{63}(n,2n)Cu^{62}$ reaction (556 ± 28 mb)[3] in thin copper foils placed before and after the sample in sandwich fashion. For cross-section measurements, radiochemical separation was not performed as it was not required. Counting was begun approximately 2 minutes after bombardment on a stable, aluminum-walled methane-flow beta-proportional counter having a 0.9-mg/cm^2 aluminized Mylar end-window. The counting rates were small enough so that dead-time losses did not exceed 0.8%. Absolute beta counting was made as precise as possible by use of the thinnest practical samples and by taking into account corrections for the following factors: counting efficiency (defined as the ratio of the events registered/total events in the sensitive volume of the detector) air and window transmission,[4] saturation backscattering,[5] self-absorption and self-scattering,[6] and background. Sample and monitor were of the same diameter and were placed in identical positions in the counter so that errors in geometry canceled. All samples were followed until decay was substantially complete.

With the exception of Cu^{64} and Ti^{45}, all of the nuclides studied emit essentially one particle per disintegration so that the counting efficiencies could be taken in the first approximation as unity. In the case of Cu^{64} we assumed that the decay proceeds 58% by emission of particulate radiation and 42% by electron capture with an intrinsic counting efficiency for the latter of 0.1 per disintegration giving an over-all counting efficiency of 0.695 ± 0.050 for Cu^{64}, assuming the K-fluorescence yield of nickel to be $\omega_K=0.366\pm0.011$.[7] In the case of Ti^{45}, we assumed 84% particulate radiation and 16% electron capture from a theoretical result given by Way, McGinnis, and van Lieshout.[8] Thus, if we accept an efficiency of 0.1 per disintegration for scandium K x-rays and a value of $\omega_K=0.20$ for scandium,[9] the counting efficiency of Ti^{45} becomes 0.92 ± 0.05.

Absolute cross sections were computed[10] from the raw counting data, after correction to infinite bombardment time from the known duration of bombardment with the assumption of constant flux during irradiation. During accelerator runs the neutron flux is monitored continuously by means of a "long counter" for neutrons and an argon-flow proportional counter which observes α particles from the $H^3(d,n)He^4$ reaction. The observed steadiness of the neutron yield ($\pm5\%$) during the bombardments justifies the constant-flux assumption. At least three runs were made for every cross section determined with the exception of the $Ti^{46}(n,p)Sc^{46}$ value, which is based on only one run.

In addition to the errors in absolute cross sections arising from the usual interpolations of the correction factors[4–8] involved in absolute beta counting, there is an error introduced in those instances where a multi-component decay must be resolved into two or more activities with similar half-lives. This difficulty appeared in the titanium bombardments in which the $Ti^{47}(n,p)$ cross section appeared to be smaller with enriched Ti^{47} than with natural titanium owing to the difficulty of resolving 3.43-day Sc^{47} [arising from the $Ti^{47}(n,p)$ reaction] and 4.8-day Ca^{47} [arising from the $Ti^{50}(n,\alpha)$ reaction] from bombardments of natural titanium, from which these activities had low counting rates after the initial decay of 44-hour Sc^{48}. By bombarding enriched Ti^{49}, the $Ti^{49}(n,p)$ cross section was established unequivocally.

The results and probable errors are listed in Tables I and II.

A new activity having a half-life of 22 ± 3 minutes was observed in bombardments of natural titanium and of enriched Ti^{50}. Radiochemical separation demonstrated that it followed scandium chemistry.[11] Since it was not observed from enriched Ti^{47} or Ti^{49} samples, we tentatively assign it to an isomer of Sc^{50}. No activity having a 22-minute half-life is produced from known impurities[12] in the natural titanium.

The 85-day half-life of Sc^{46} from the $Ti^{46}(n,p)$ reaction was followed for more than 100 days. From its counting rate extrapolated to the end of bombardment (30 counts/min above background), a rough estimate of the cross section was obtained (~520 mb).

DISCUSSION

The absolute cross sections for (n,p) reactions of Ti^{46}, Ti^{47}, Ti^{48}, and Ti^{49} show an interesting trend with mass number. The relative cross sections lie in a ratio, respectively, of 2.26:1:0.252:0.131. This agrees very well with the trend reported by Levkovskii,[13] who determined relative (n,p) yields for Ti^{47}, Ti^{48}, and Ti^{49} in the ratio, respectively, of 1:0.25:0.137. This trend, as well as a similar one for (n,α) reactions, has been interpreted by Levkovskii[13] to reflect a decreasing

[3] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[4] Gleason, Taylor, and Tabern, Nucleonics **8**, No. 5, 12 (1951).
[5] B. P. Burtt, Nucleonics **5**, No. 2, 28 (1949); J. R. Zumwalt, Atomic Energy Commission Report AECU-567, 1950 (unpublished); L. Yaffe, *Conference on Absolute Beta Counting*, Prelim. Rept. No. 8 (National Research Council, Washington, D. C., 1950).
[6] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, No. 3, 18 (1952); Walton, Thomson, and Croall, Atomic Energy Research Establishment Report AERE-c/R-1136, 1953 (unpublished); Cuninghame, Sizeland, and Willis, Atomic Energy Research Establishment Report AERE-c/R-2054 1957 (unpublished); R. G. Baker and L. Katz, Nucleonics **11**, No. 2, 14 (1958).
[7] C. E. Roos, Phys. Rev. **105**, 931 (1957).
[8] Way, King, McGinnis, and van Lieshout, *Nuclear Level Schemes, $A=40$ to $A=92$*, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).
[9] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953); P. R. Gray, Phys. Rev. **101**, 1306 (1956).
[10] D. J. Hughes, *Neutron Cross-Section* (Pergamon Press, Inc., New York, 1957).
[11] (to be published).
[12] Impurities in natural titanium foils consisted of 0.018% carbon, 0.05% iron, 0.01% nitrogen, and 0.007% hydrogen according to analyses supplied by American Silver Company, Flushing, New York.
[13] V. N. Levkovskii, J. Exptl. Theoret. Phys. U.S.S.R. **31**, 360 (1956); **33**, 1520 (1958) [translations: Soviet Phys. JETP **4**, 291 (1957); **6**, 1174 (1958)].

TABLE I. Absolute (n,p) and (n,α) cross-section measurements; comparison with previous data and with theory at 14.8 Mev.

Reaction	Product	Measured half-life	Measured cross section (millibarns) Present work	Literature	Q-value (Mev)	Calculated cross section (mb)	Ratio $\sigma_{exp}/\sigma_{calc}$
$Al^{27}(n,p)$	Mg^{27}	9.46±0.02 min	53± 5	55±15[a] 79±15[d] 52[e] 87± 7[c]	−2.59[b]	120[c]	0.44
$Al^{27}(n,\alpha)$	Na^{24}	15.00±0.06 hours	114± 7	120±15[c] 116± 8.1[g]	−3.14[f]	420[c]	0.26
$Ti^{46}(n,p)$	Sc^{46}	85±2 days	~520		−1.57[f]	610	0.85
$Ti^{47}(n,p)$	Sc^{47}	3.45±0.06 days	230±40		+2.11[f]	191	1.20
$Ti^{48}(n,p)$	Sc^{48}	44.0 ±0.9 hours	58± 8	92.7[e]	−3.21[f]	187	0.31
$Ti^{49}(n,p)$	Sc^{49}	58±2 min	29± 5		−1.27	100	0.29
$Ti^{50}(n,p)$	Sc^{50}	1.80±0.20 min	27± 6 }		−3.5[f]	280	0.26[h]
$Ti^{50}(n,p)$	Sc^{50}	22±3 min	48±15 }				
$Ti^{50}(n,\gamma)$	Ti^{51}	(5.89 min)	⩽9	3.5± 1.0[i]			
$Cu^{65}(n,p)$	Ni^{65}	2.55±0.20 hours	27±11	31 ±13[j] 19 ± 4[m]	−1.3[k]	5[l]	5.4
$Bi^{209}(n,p)$	Pb^{209}	3.31±0.03 hours	0.83± 0.40		−0.63[n]	0.043	19
$Bi^{209}(n,\alpha)$	Tl^{206}	4.29±0.05 min	1.1 ± 0.3	1.2[e]	+9.74[o]	0.063	17
$Bi^{209}(n,\gamma)$	Bi^{210}	(5.10 days)	⩽1.7	1.45± 0.15[i]			

[a] Brown, Morrison, Muirhead, and Morton, Phil. Mag. **2**, 785 (1957).
[b] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), second edition.
[c] S. Yasumi, J. Phys. Soc. (Japan) **12**, 443 (1957).
[d] Haling, Peck, and Eubank, Phys. Rev. **106**, 971 (1957).
[e] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).
[f] A. H. Wapstra, Physica **21**, 367 (1955); A. G. W. Cameron, Chalk River Laboratory Report CRP-690, 1957 (unpublished).
[g] Grundl, Henkel, and Perkins, Phys. Rev. **109**, 425 (1958).
[h] The sum of the experimental (n,p) cross sections for both isomers of Sc^{50} has been compared with the theoretical value.
[i] Perkin, O'Connor, and Coleman, Proc. Phys. Soc. (London) **72**, 505 (1958).
[j] R. S. Scalan and R. W. Fink, Nuclear Phys. (to be published).
[k] G. Brown and H. Muirhead, Phil. Mag. **2**, 473 (1957).
[l] The sum of the direct interaction cross section (30 mb) and the compound nucleus cross section (5 mb) as calculated by Brown and Muirhead gives ratio $\sigma_{exp}/\sigma_{calc}=0.74$.
[m] S. G. Forbes, Phys. Rev. **88**, 1309 (1953).
[n] L. J. Lidofsky, Revs. Modern Phys. **29**, 773 (1957).
[o] J. R. Huizenga, Physica **21**, 410 (1955).

probability for emission of a proton from an excited nucleus with decreasing concentration of protons in the nucleus. If this hypothesis be a valid one, then $(n,2n)$ cross sections should show an increase with increasing A at constant Z. Existing data on 14-Mev cross sections indicate that this trend generally is followed throughout the periodic table, but is especially marked in the low-Z and middle-Z regions.

In the case of bismuth, the (n,α) cross section is about the same in magnitude as the (n,p), and both values are a bit less than 20 times larger than predicted from the continuum theory of a compound nucleus (see Table I). However, the theoretical results were obtained from the averaged level density formula given in the next section, so that the effect of the closed shells ($Z=82$, $N=126$) which causes an anomalous decrease in the level density and hence a decrease in the calculated cross sections has not been taken into account. Thus, the deviations between theory and experiment for the (n,α) and (n,p) cross sections of bismuth in Table I really are minimal values; the true disagreements very likely are larger.

One thinks that the large (n,p) cross section of bismuth might be explained, at least qualitatively, by direct interaction of the incoming neutron with the 83rd proton, one above the closed proton shell. Brown and Muirhead[14] predict a value of about 0.5 mb in the region of bismuth on the assumption, however, that

TABLE II. Absolute $(n,2n)$ cross section measurements; comparison with previous data and with theory at 14.8 Mev.

Reaction	Measured half-life	Measured cross section (millibarns) Present work	Literature	Q-Value (Mev)	ϵ_c (Mev)	a (Mev)$^{-1}$	θ (Mev)	ϵ_c/θ	Calculated Cross section (mb)	Ratio $\sigma_{exp}/\sigma_{calc}$
$Ti^{46}(n,2n)Ti^{45}$	3.06±0.08 hours	50.4± 8.0	27.9[a]	−10.30[b]	1.2	1.0[c] 1.2[d]	3.84 3.52	0.312 0.341	54 64	0.94 0.80
$Cu^{65}(n,2n)Cu^{64}$	12.85±0.05 hours	954 ±130	935[a] 1000±100[e]	−9.79[b]	4.6	1.95[c] 1.85[d]	2.75 2.82	1.67 1.63	695 675	1.37 1.42

[a] L. A. Rayburn, Bull. Am. Phys. Soc. Ser. II, **3**, 337 (1958); Ser. II, **3**, 365 (1958).
[b] A. H. Wapstra, Physica **21**, 367 (1955); A. G. W. Cameron, Chalk River Laboratory Report CRP-690, 1957 (unpublished).
[c] Feld, Feshbach, Goldberger, Goldstein, and Weisskopf, Atomic Energy Commission Report NYO-636, 1951 (unpublished).
[d] J. Heidman and H. A. Bethe, Phys. Rev. **84**, 274 (1951).
[e] *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

[14] G. Brown and H. Muirhead, Phil. Mag. **2**, 473 (1957).

direct nucleon-nucleon interactions occur throughout the nuclear volume rather than primarily with nucleons outside of a closed shell. That the (n,α) cross section of bismuth is so large and of the same magnitude as the (n,p) is interesting in that one does not readily imagine a simple mechanism for a direct interaction process for ejection of an alpha particle from bismuth. However, Wilkinson[15] has summarized the evidence for the existence of a tendency for the region of low density in the nuclear surface to be relatively rich in nucleon clusters such as alpha particles owing to the lower average binding energies extant in the diffuse surface. The existence of such performed alpha-particle clusters could explain the large (n,α) cross sections and the evidence for direct interaction production of alpha particles in (p,α) reactions.[16] Such nucleon clusters would also explain the fact that a large measure of preformed alpha particles in the surface of heavy nuclei must be assumed in order to account for the emission rate of alpha radioactive decay.[17] Another factor which would enhance the cross sections for alpha emission in (p,α) and (n,α) reactions is the larger penetrability arising from the diffuse potential at the nuclear surface.

THEORETICAL COMPUTATIONS

Theoretical estimates of the (n,p) and (n,α) cross sections have been made using the continuum theory of the compound nucleus as outlined by Blatt and Weisskopf.[18] Level densities were computed from the formula,[18]

$$\omega_{(E)}=C\exp[2(aE)^{\frac{1}{2}}],$$

where the values of the constants C and a were taken from the work of Feld, Feshbach, Goldberger, Goldstein, and Weisskopf.[19] In determining C it was assumed[14] that

$$12C_{\text{even-even}}=2.4C_{\text{even-odd}}=2.4C_{\text{odd-even}}=C_{\text{odd-odd}}.$$

The computations have been carried out with $r_0=1.3\times10^{-13}$ cm and $a=12.2$ for bismuth[19] and $a=1$ for titanium.[19] The latter value is in agreement with that given by the semiempirical formula $a=0.035(A-12)$ Mev^{-1} of Heidman and Bethe.[20] The Q-values were obtained from total beta disintegration energies,[21] atomic mass tables based on experimental data,[22] or atomic mass tables computed from a semiempirical formula.[23]

Results of these theoretical computations are given in Table I for comparison with the experimental data.

In the case of $(n,2n)$ reactions, theoretical estimates have been computed from the formula given by Blatt and Weisskopf[18] on the assumption that neutron emission becomes predominant as soon as it becomes energetically possible. The threshold energies were taken from Segrè.[24] The results are compared with our experimental values in Table II, in which ϵ_c is the maximum emission energy of the second neutron in the $(n,2n)$ reaction and is equal to the difference between the energy of the incoming projectile and the threshold energy[20] for the reaction, a is a constant in the level density formula, and θ is the nuclear temperature. It can be seen from Table II that the theoretical estimates for the $(n,2n)$ cross sections of copper and titanium agree well with the experimental values.

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[15] D. H. Wilkinson, Phil. Mag. **4**, 215 (1959).

[16] P. E. Hodgson, Nuclear Phys. **8**, 1 (1958); C. B. Fulmer and B. L. Cohen, Phys. Rev. **112**, 1672 (1958).

[17] I. Perlman and J. O. Rasmussen, *Handbuch der Physik*, edited by S. F. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 109.

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

[19] Feld, Feshbach, Goldberger, Goldstein, and Weisskopf, Atomic Energy Commission Report NYO-636, 1951 (unpublished).

[20] J. Heidman and H. A. Bethe, Phys. Rev. **84**, 274 (1951).

[21] L. J. Lidofsky, Revs. Modern Phys. **29**, 773 (1957).

[22] A. H. Wapstra, Physica **21**, 367 (1955); A. G. W. Cameron, Chalk River Laboratory Report CRP-690, 1957 (unpublished); J. R. Huizenga, Physica **21**, 410 (1955).

[23] N. Metropolis and G. Reitwiesner, Atomic Energy Commission Report NP-1980, 1950 (unpublished).

[24] E. Segrè, *Experimental Nuclear Physics* (John Wiley & Sons, Inc., New York, 1953), Vol. 2, p. 350.