to oxygen molecules gives support to the view[18] that in E region a twofold reaction occurs, namely, the ionizing radiation first produces positive ions and free electrons and the free electrons become rapidly attached to form negative oxygen ions. If the ratio of the rate of electron attachment to the rate of ionic recombination were sufficiently great the ionization of E region would be predominately ionic, rather than electronic as far as the refraction of radio waves is concerned, the disappearance of ions would be by the recombination formula (15.1), and y of E would agree with (1) and hence with observation. Numerically, however, the ratio of b/α should be about 3 times greater than the value given by (15.1) and (16.1). This would occur either if α of (15.1) were a little less, or if Bradbury's[16] curve of h against electron energy continued to increase with decreasing energy in the domain below 0.2 electron volts. Further experimental knowledge of attachment and recombination coefficients might contribute to the question in an important manner.

[18] Hulburt, Phys. Rev. **34**, 1167 (1929); **35**, 240 (1930).

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Emission of Neutrons from Argon, Chlorine, Aluminum and Some Heavier Elements Under Alpha-Particle Bombardment

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The emission of neutrons from chlorine, argon, scandium, titanium, manganese and iron under alpha-particle bombardment was established. The yield from argon is considerable and enabled a measurement of the energy of the neutrons to be made: the majority are associated with a group of energy change -5.6 ± 1.0 Mev, but two groups must be present. The excitation curve for these neutrons was plotted for alpha-particle energies between 3.5 and 9.0 Mev and varies smoothly in agreement with penetration through a barrier of radius 7.6×10^{-13} cm. This smooth variation means that the total neutron yield does not change rapidly as a new group is excited, from which it is deduced that observations on single groups would show apparent resonance effects. The excitation curve for chlorine fits a theoretically derived function for a nuclear radius of 6.0×10^{-13} cm and a similarly plotted curve for aluminum agrees with a radius of 5.8×10^{-13} cm. These last elements have radii approximately fitting the formula $R=R_0A^{\frac{1}{3}}$ with $R_0=1.94\times10^{-13}$, while argon has a radius which is definitely too large to fit the above relation. This abnormally large radius is linked with the large neutron content of the argon nucleus.

INTRODUCTION

THE use of a boron trifluoride filled ionization chamber surrounded by paraffin is so sensitive a neutron detector that neutron emission can be detected even with weak alpha-particle sources. Using an arrangement of this kind we have subjected a number of elements to bombardment by Th C′ alpha-particles and looked for the emission of neutrons. It was found that a slight yield could be detected from scandium, titanium, manganese and iron, indicating that the absorption of an alpha-particle with emission of a neutron is a general occurrence in this section of the periodic table. The yields from these elements were, however, roughly equal to the background of the detecting apparatus and therefore not suitable for detailed study. On the other hand argon and chlorine were found to give considerable yields of neutrons (in the case of argon the yield is second only to that from beryllium if Th C′ alpha-particles are used) and the nature of the neutron yield from these two elements was therefore further studied. An estimate of the energy of a neutron group was made for argon and excitation curves for both elements plotted. As a check an excitation curve for the neutrons from aluminum was also determined: from these three excitation curves values for the nuclear radii of the three elements can be found. A preliminary report of the experiments on argon and chlorine appeared early in 1937.[1]

[1] E. Pollard, H. L. Schultz and G. Brubaker, Phys. Rev. **51**, 140 (1937).

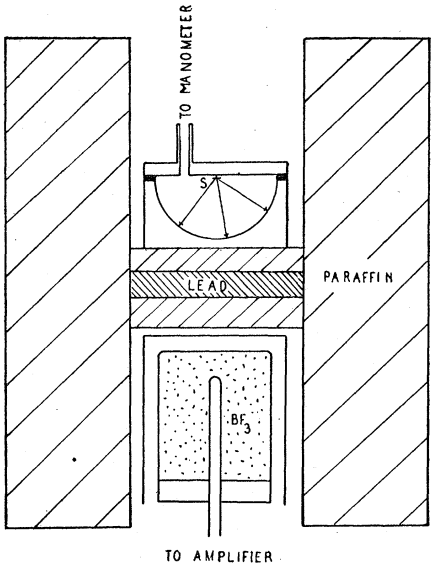

FIG. 1. The arrangement of source, ionization chamber and surrounding paraffin.

EXPERIMENTAL ARRANGEMENT

The essentials of the arrangement for bombardment and detection are shown in Fig. 1. The radioactive source *S* was placed at the center of a hemisphere of four centimeters radius cut in a brass block. Targets of various materials mounted on hemispherical bases could be placed inside the block, or gases could be bombarded with a silver hemisphere as backing. For preliminary experiments a smaller bombardment arrangement was used in which the material bombarded was placed in a brass tray and exposed to, or shielded from, the source, by rotating a small copper plate over the source button. The ionization chamber was of the shape drawn: it was filled with boron trifluoride at atmospheric pressure and a potential of a thousand volts applied to the outer electrode; the collection electrode was connected to the input of a conventional linear amplifier. The source box and ionization chamber were placed in a cylinder of paraffin of outside diameter 30 cm and inside diameter 12 cm; varying thicknesses of paraffin could be placed between box and chamber. A block of lead was interposed to diminish the gamma-ray background from the source. No difficulty was experienced in separating the kicks produced by the neutrons from the background due to gamma-rays except when sources larger than fifty millicuries were employed.

For general trial experiments different targets were used and the yield measured with target alternately exposed to, or shielded from, the alpha-particles: for gaseous targets the yield was compared with the yield when the box was evacuated. Excitation curves for the two solid targets were plotted by varying the pressure of oxygen in the hemispherical container and measuring the neutron yield; and for argon by varying the pressure of argon itself. Sources of thorium and radium active deposits and polonium were used, deposited on buttons of either nine or ten millimeters in diameter: both silver and monel metal buttons were used for Th C′ and Ra C, while the polonium source was deposited on palladium.[2]

EXPERIMENTAL RESULTS

General survey

The first result of general survey experiments was an apparent yield of neutrons from the source itself. This phenomenon has been mentioned by Szilard.[3] It appears to be greater for Th C′ sources than for Ra C sources of equal strength and is not observed for polonium: the probable explanation is that it is partly due to the transmutation of the material of the source button and partly to the nuclear photoeffect on the deuterium present in the paraffin. Whatever the cause, the presence of a yield of roughly two counts per minute above the background (also roughly two per minute) rendered it difficult to study the properties of neutron yields from elements except where this yield is large. The data in Table I are the results of experiments intended to test the presence or absence of neutron emission when an element is bombarded by Th C′ alpha-particles. Only relative yields should be considered as the counting level was not always the same. The limits of error given are not to be regarded as rigorous: they merely serve to give some indication of the extent of the counting in each determination. Following a somewhat arbitrary convention, we have divided

[2] The polonium source was kindly supplied by Professor Bearden and Dr. Kanne of Johns Hopkins University.
[3] L. Szilard, Nature **136**, 950 (1935).

the value by the square root of the number of particles counted to give the limits above. We consider that where a yield is claimed to exist no impurity in the material could account for the observed emission. This general survey shows clearly that elements of moderately high atomic number will emit neutrons when bombarded by alpha particles of roughly 9 Mev energy. The experiments of Ridenour and Henderson[4] in which induced radioactivity has been observed in a wide variety of elements also indicate this. Since the yields we observe with natural sources are so small it is clearly preferable to use artificial alpha-particles for further study in this field and we therefore contented ourselves with the above data except for the two elements argon and chlorine where the yields are large enough to enable the plotting of excitation curves, and in the case of argon to determine the energy of the emitted neutrons.

Argon

Energy of neutrons.—In order to obtain a large yield of neutrons a brass cylinder was filled with a mixture of argon and fifty millicuries of radon and recoil protons from paraffin looked for with a proportional counter as used in experiments on proton emission. A lead block was interposed between the neutron "bomb" and the counter to diminish the effect of the gamma-radiation and a paraffin layer was placed on the counter side of the block. A source of Th C′+Be giving roughly one third as many neutrons as the argon-radon bomb gave a yield of recoil protons equal to three times the counter background. The argon radon mixture gave no detectable effect (one-third the background) with counters having foils of greater than 2.0 cm air equivalent over their opening so that very few neutrons of energies greater than one Mev can be emitted from this mixture. The counter was then filled with hydrogen at 47 cm pressure which would be capable of detecting recoil protons of range less than six millimeters air equivalent. A rise in counts from twelve per hour to seventy per hour was observed when the argon-radon mixture was brought near, indicating the presence of a yield of relatively low energy neutrons. From estimates of the size of oscillograph deflections compared with protons of known energy, we estimated that the majority of the neutrons gave recoil protons of ranges between four and six millimeters air equivalent. The neutrons from the $A+\alpha$ reaction, using radon and its products for bombarding are therefore *mainly confined to less than 500,000 electron volts energy.* This conclusion has been verified by the more satisfactory work of McCarthy of this laboratory who measured the lengths of sixty recoil tracks in a cloud chamber filled with hydrogen and found the majority of tracks to lie between 6 and 8 millimeters air equivalent with no tracks greater than eighteen millimeters air equivalent. If we suppose, as a simplifying assumption that the neutrons are due to bombardment by 6.5 Mev alpha-particles and that they are emitted with 0.5 Mev energy, then the nuclear energy in the reaction:

$$A^{40}+He^{4}\rightarrow Ca^{43}+n^{1}+Q$$

is

$$Q=-5.6\pm1.0 \text{ Mev.}$$

[4] L. N. Ridenour and W. J. Henderson, Phys. Rev. **52**, 889 (1937).

TABLE I. *Results of alpha-particle bombardment.*

ELEMENT	TARGET	YIELD PER MINUTE	BACKGROUND PER MINUTE	CONCLUSION	REACTION
Titanium	Metal	4.4±0.4	3.5±0.4	True yield[a]	Ti[α, n]Cr
Chlorine	"arochlor" gas	6.3±0.6	3.9±0.4	True yield	Cl[α, n]K
		5.0±0.5	1.6±0.3	" "	
Scandium	Sc_2O_3	5.4±0.6	4.4±0.5	Probable yield[b]	Sc^{45}[α, n]V^{48}
Iron	Filings	3.1±0.3	2.4±0.2	True yield	Fe^{56}[α, n]Ni^{59}
Manganese	MnO_2	3.8±0.5	2.3±0.3	True yield	Mn^{55}[α, n]Co^{58}
Carbon	Lamp black	3.8±0.2	3.5±0.2	No yield	
Oxygen	Tank gas	3.7±0.2	3.5±0.2	No yield	
Nitrogen	Air	6.0±0.5	2.2±0.3	Yield	N^{14}[α, n]F^{17}
Argon	Gas	7.0±0.9	1.9±0.3	Large yield	A^{40}[α, n]Ca^{43}

[a] Walke (Phys. Rev. **52**, 784 (1937)) obtained two radioactive products from the activation of Ti with alpha-particles which he suggests may be due to this type of reaction.
[b] This reaction has been definitely established by Walke (Phys. Rev. **52**, 671 (1937)) who observed the artificial radioactivity of V^{48}.

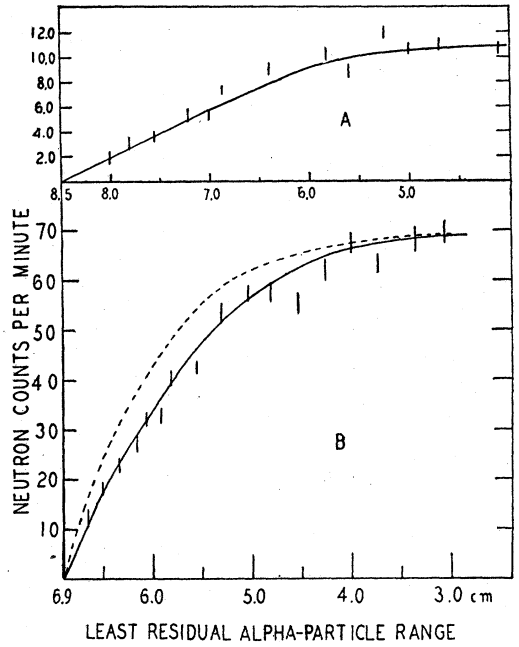

FIG. 2. Graph showing the neutron yield from argon bombarded by alpha-particles from Th C′ (A) and Ra C (B) as the gas pressure in the hemispherical chamber is varied. The line drawn through the points in A is the theoretically expected yield if the nuclear radius is 7.3×10^{-13} cm; that in B for a radius of 7.6×10^{-13} cm. The dotted line in B is the theoretical curve for a radius of 6.8×10^{-13} cm which is close to the value expected from the relation $R=R_0A^{\frac{1}{3}}$.

Excitation curves.—A gaseous target is not suitable for plotting an excitation curve directly as it must be confined between walls of a material which is both thin and vacuum tight. If, however, the stopping power of the gas is known it is quite possible to derive an excitation curve from a set of data giving the variation of yield with pressure in the hemispherical bombardment chamber as described already. Thus, when the hemisphere is evacuated, only alpha-particles of full residual range traverse the space. As gas is admitted a band of energies from maximum down to a value governed by the gas pressure is effective in the space until at high pressures the alpha-particles are fully stopped in the gas and do not reach the limits of the container. The increase in yield corresponding to an increase in pressure is therefore the yield due to the increased absorption and if desired the excitation curve for a thin layer of gas may be derived from the yield-pressure curve by differentiation. Or preferably, a theoretical excitation function may be integrated for progressively widening limits and compared with the experimental results. This procedure was followed in a number of runs made with Th C′, Ra C and polonium sources with the results given in Fig. 2. The yield is plotted against the lower limit of alpha-particle range corresponding to the pressure. It can be seen that for Th C′ particles (curve A) the yield rises linearly until alpha-particles of less than 6.5 cm range are present after which it falls away indicating that alpha-particles with residual ranges between 6.5 and 8.6 cm are equally effective in causing transmutation while those below 6.5 cm are progressively less effective. The Ra C curve B shows the lower energy portion in more detail and indicates that alpha-particles of as low as 3.0 cm continue to cause transmutation. The results with polonium verify this last deduction since a definite yield was observed for alpha-particles of less than 3.0 cm range. In curve A 500 particles were counted at each point; in curve B, 1000.

The lines drawn in Fig. 2 are derived from the Gamow penetration formula ($l=0$) assuming radii of 7.3 and 7.6×10^{-13} for the argon nucleus. The theoretical curve for a thin layer has been integrated to compare with the experimental results. It can be seen that the fit is quite good and we suggest the value $7.6\pm0.3\times10^{-13}$ for the nuclear radius of argon. We give greater weight to the results with Ra C on account of the larger number counted and the fact that a single alpha-particle group is present.

Presence of two groups of neutrons.—A point of some interest arises from the fact that alpha-particles of relatively low energy can cause the emission of neutrons. It was definitely established that alpha-particles of three centimeters range can set free neutrons and if it be assumed as a limiting case that at this range the emitted neutrons have zero energy the nuclear energy change cannot be less than -4.0 Mev, which is 1.6 Mev greater than found from the measured neutron energies.[5] This discrepancy means that some explanation other than the simple reaction with a single nuclear energy change must be sought and two reasonable possibilities present

[5] With this nuclear energy change the average range of projected protons due to neutrons from Rn+A as in our experiments would be expected to be 7 cm air equivalent as against 0.7 cm found.

themselves. The first is that the emission of two neutrons may occur according to $A^{40}[\alpha, 2n]Ca^{42}$; the second that at high alpha-particle energies an excited state of Ca^{43} is formed which reverts to the ground state with emission of gamma-radiation. The first process requires a considerable energy change in going from A^{40} to Ca^{42} and for that reason, we think, is improbable. The second process, the formation of Ca^{43} in an excited state is normally to be expected and is much more probably the true explanation. On this view the neutrons emitted would have energies corresponding to two "Q" values, the one the value -5.6 Mev already found and a more energetic group of relatively low yield of Q value certainly exceeding -4.0 Mev (which just permits the release of neutrons by 3.0 cm alpha-particles). The presence of this second group could not be established with certainty by direct experiments as the available yields were not large enough. It is, of course, possible that several groups are present. If this view is accepted there is an interesting deduction to be made from the excitation curve. We find a smooth fit to the Gamow formula for the *total* neutron emission, whereas it would be expected that a sharp drop in yield would occur at the critical alpha-particle energy below which the -5.6 Mev group could not be emitted. Since this group is the more prominent this drop in yield should be very definite, whereas Fig. 2 shows no sign of a clear discontinuity. If, then, the *total* emission is governed only by the penetrability of the barrier, the yield of any one group must be affected by

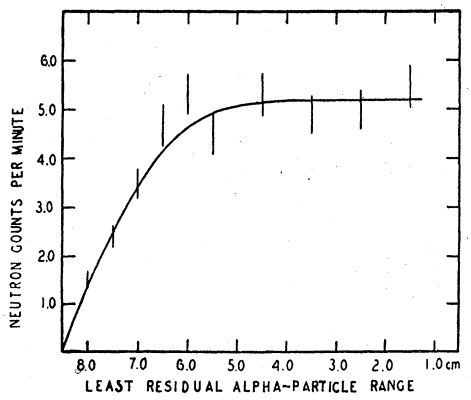

FIG. 3. Neutron yield from chlorine gas at varying pressures in a hemispherical chamber. The line is the theoretical curve for a nuclear radius of 6.0×10^{-13} cm.

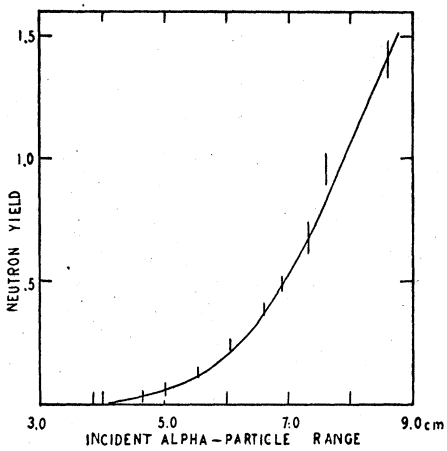

FIG. 4. Neutron yield from a thick layer of "Arochlor" (containing chlorine, carbon and hydrogen) as the range of the bombarding alpha-particles is varied by introducing oxygen at different pressures between source and target. The line is the theoretical curve for a nuclear radius of 6.0×10^{-13} cm.

the presence or absence of other groups and in particular the yield of the "Q" greater than 4.0 Mev group must fall when the 5.6 Mev group appears, thus giving the appearance of resonance. This explanation of "resonance" effects has been proposed by Waring and Chang[6] and Chang and Szalay[7] to explain the variations in the yield of induced radioactivity from the reactions $Al^{27}[\alpha, n]P^{30}$ and $Mg^{24}[\alpha, n]Si^{27}$. Our experiments lend confirmation to their views.

Chlorine

The majority of the experiments were conducted with "Arochlor" (a chlorinated diphenyl) as a target and our work was confined to plotting an excitation curve. Two runs were made with chlorine gas following the procedure used for argon, with the results shown in Fig. 3. Only the general behavior should be inferred from this curve as the number of particles counted to a point was between one hundred and two hundred: the great chemical activity of chlorine gas renders this type of experiment unpleasant. The curve through the points is the thoretical function for penetration through a barrier of radius 6.0×10^{-13} cm integrated as for argon; it can be seen that although the points are rather scattered,

[6] J. R. S. Waring and W. Y. Chang, Proc. Roy. Soc. **157**, 652 (1936).

[7] W. Y. Chang and A. Szalay, Proc. Roy. Soc. **159**, 72 (1937).

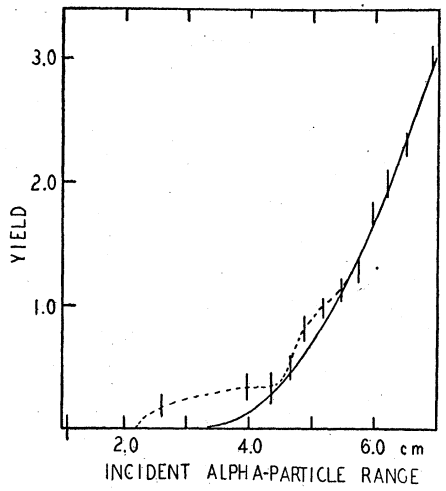

FIG. 5. Excitation curve for a thick layer of aluminum. The full line is the theoretical relation for a nuclear radius of 5.8×10^{-13} cm. The dotted line indicates possible resonance effects at 4.8 cm and 2.3 cm alpha-particle range.

the fit is good. In Fig. 4 is shown the results of bombarding a thick layer of "Arochlor" with both Th C′ and Ra C sources. The curve below 6.9 cm alpha-particle range is more accurately known although the certainty is considerably less than that for argon. A net count of between three and five hundred particles was made at each point. The curve through the points is the theoretical curve for a nuclear radius of 6.0×10^{-13} cm: the agreement is good and the fact that this is the same radius as found in Fig. 3 justifies the method of plotting excitation curves for gaseous targets. We suggest the value $6.0\pm0.5\times10^{-13}$ cm for the nuclear radius of chlorine.

The interpretation we originally gave for this process was the reaction:

TABLE II. *Nuclear radii.*

ELEMENT	NUMBER OF NUCLEAR PARTICLES (A)	$A^{\frac{1}{3}}\times1.94\times10^{-13}$	FOUND RADII (cm)
Al	27	5.80×10^{-13}	$5.80\pm0.3\times10^{-13}$
Cl	35	6.30×10^{-13}	$6.00\pm0.5\times10^{-13}$
A	40	6.63×10^{-13}	$7.60\pm0.3\times10^{-13}$

$$Cl^{37}+He^{4}\rightarrow K^{40}+n^{1}.$$

In view of the experiments of Hurst and Walke[8] and Ridenour and Henderson[4] it is certain that part of the neutrons are due to the reaction:

$$Cl^{35}+He^{4}\rightarrow K^{38}+n^{1}.$$

It is most probable that both processes occur.

Aluminum

A thick hemispherical target was bombarded at various alpha-particle ranges with the results shown in Fig. 5. Roughly four hundred particles were counted at each point. It can be seen that while no striking resonance effects are present, yet there may be small irregularities present as suggested by Waring and Chang.[6] The smooth curve drawn through the points is derived from the Gamow penetration formula for a nuclear radius of 5.8×10^{-13} cm. The agreement is satisfactory. We suggest limits of error of 0.3×10^{-13} cm.

Nuclear radii of light elements

Our results give us the values of the nuclear radii for aluminum, chlorine and argon. These should vary proportionally to the cube root of the number of particles (A) in the nucleus. The values found, are given in Table II together with the values of the expression $A^{\frac{1}{3}}\times1.94\times10^{-13}$ which fits the value for aluminum. It can be seen that the values found vary with respect to the calculated value; this is in part to be expected by reason of the possible errors: we feel, however, that the large value for argon lies beyond the limits of error and represents a real anomaly. This can be linked with the fact that argon has an unusually large excess of neutrons over protons.

It is a pleasure to thank Professor A. F. Kovarik for his interest and encouragement, and Mr. R. M. Ryder for assisting with the calculations of barrier penetration.

[8] D. G. Hurst and H. Walke, Phys. Rev. **51**, 1033 (1937).