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Radioactive Argon

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When bombarded with high speed deuterons, argon gas is found to yield a radioactive product which emits negative electrons, and decays with a period of 110 ± 1 minutes. Chemical tests show that the activity is due to an isotope of argon, and the reaction involved is doubtless $A^{40}+H^2=A^{41}+H^1$. Absorption measurements of the β-particles indicate that their maximum energy is about 1.1 MV. This is in satisfactory agreement with the results of a cloud chamber study of their distribution curve made by Kurie, Richardson and Paxton, which shows a strong group with an upper limit at 1.5 MV, and apparently also a weak group with an upper limit at about 5 MV. The radioactivity is accompanied by the emission of a γ-ray, the energy of which is 1.39 MV, as measured by the energies of its Compton recoils in a cloud chamber. The excitation function of the radioactivity favors the Oppenheimer-Phillips theory for this type of reaction, rather than the Gamow theory for the penetration of the potential barrier by a charged particle. The same radioactive substance has been made by subjecting argon to an intense bombardment with neutrons.

SO many new radioactive isotopes have recently been created by the action of neutrons and high speed charged particles upon stable substances that an element for which no induced radioactivity has been reported is almost a rarity. Argon has been one of the few elements in this category. It may be of interest therefore to report the activation of argon by bombardment with deuterons and neutrons, and to describe the identification and some of the properties of the radioactive substance thereby produced.

EXPERIMENTAL

The magnetic resonance accelerator of Lawrence and Livingston[1] served as a source for the high speed deuterons and (in later tests) for the neutrons used in activating the argon. In the present form of this apparatus, it is possible to allow the deuteron beam to emerge from the vacuum through a thin platinum window so that it passes across the closed inner end of a brass tube recessed in the wall of the accelerating chamber (Fig. 1). In order to bombard argon with deuterons it was therefore necessary merely to close off the region traversed by the deuteron beam with a vacuum-tight seal, penetrated only by a slender tube to be used for introducing and withdrawing the sample of gas. This tube led either (a) to a Hyvac pump, or (b) to a mercury transfer pump and argon reservoir. The transfer pump was used to fill the bombarding chamber with argon at any desired pressure, and (after bombardment) to remove the activated gas and convey it to a thin-walled vessel. The latter was then sealed off, and the sample was ready for examination. This technique was used whenever a fairly strong sample of active gas was required; it has an advantage in that it avoids possibilities

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[1] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608 (1934).

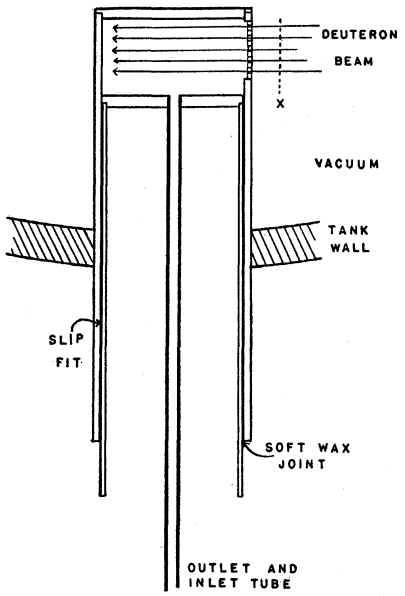

FIG. 1. Experimental arrangement used for activating argon with deuterons. For activation with neutrons, a beryllium target is introduced at ×.

of contamination such as would arise, for example, in the collection of active recoils propelled by the deuterons onto a piece of metal placed in the path of the beam in an atmosphere of argon.

THE DECAY PERIOD OF THE RADIOACTIVITY

The first measurements of the decay period were made with samples of commercial argon which was roughly 90 percent pure. Samples were activated on three occasions with about 1 microampere of 3 MV deuterons, the activation lasting about one hour. The radioactivity induced was moderate in intensity, and its decay was followed for three half-lives or more with a quartz-fiber electroscope of the Lauritsen type.[2] Readings were started 20 minutes after cessation of bombardment. Each sample gave evidence of only one decay period, the mean value of which was 108±3 minutes. Later an additional determination was made with a sample of 99 percent pure argon, more strongly activated by bombardment for 1½ hours with 3 microamperes of 5-MV deuterons. The decay of this sample was followed over a factor of 1000 with a pressure ionization chamber connected to an F.P. 54 electrometer tube and a d.c. amplifier. (I am indebted to Dr. S. N. Van Voorhis for the use of this apparatus.) The decay curve is reproduced in Fig. 2; it will be seen that the semilogarithmic plot is quite accurately linear. and indicates a half-life of 110±1 minutes.*

The sign of the emitted particles was observed by collecting some of the active recoils upon a piece of platinum, and placing the platinum in a cloud chamber in a magnetic field. The particles had a negative charge; only a very few positives were observed, which could easily be attributed to a slight carbon contamination of the platinum.

IDENTIFICATION OF THE RADIOACTIVE ISOTOPE

According to Aston,[3] argon possesses two isotopes; their masses are, respectively, 36 and 40, and A^{36} exists in less than one percent of the abundance of A^{40}. This fact alone makes it probable that the heavier isotope is involved in making the new radioactive substance; however, it is well for the sake of completeness to consider

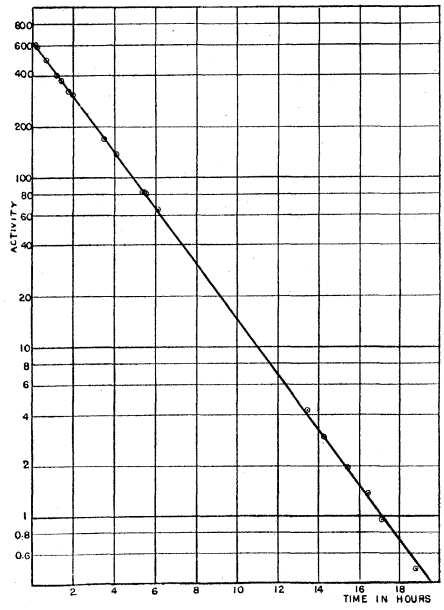

FIG. 2. Decay curve for radioactive argon, obtained with high pressure ionization chamber. Half-life: 110±1 minutes.

[2] J. Read and C. C. Lauritsen, Phys. Rev. **45**, 433 (1934).

* Because of a misprint, this period was erroneously quoted as 10±1 minutes in an earlier report (Phys. Rev. **49**, 207 (1936)).

[3] F. W. Aston, *Mass Spectra and Isotopes* (Longmans, Green and Co., 1933).

the six possible reactions which might be expected to take place. They are

$$_{18}A^{36,\,40}+{}_1H^2\rightarrow{}_{18}A^{37,\,41}+{}_1H^1, \quad (1)$$

$$_{18}A^{36,\,40}+{}_1H^2\rightarrow{}_{17}Cl^{34,\,38}+{}_2He^4, \quad (2)$$

$$_{18}A^{36,\,40}+{}_1H^2\rightarrow{}_{19}K^{37,\,41}+{}_0n^1. \quad (3)$$

Of these the third is unlikely because K^{41} is a common isotope which certainly does not have the radioactive properties which are here described, while K^{37} would be expected to emit positive electrons, and not the negatives which are found to decay with the 110-minute period. As regards the second reaction, a chemical test was carried out which showed that the radioactivity was not due to an isotope of chlorine. A small amount of chlorine gas was added to the activated argon, and the mixture was slowly drawn through a copper trap containing powdered antimony spread upon small pieces of filter paper. The trap was provided with a thin wall made of 0.001-inch copper, through which radioactive β-particles could escape. The experiment was done at room temperature, and also with the trap immersed in liquid air. In both cases no chlorine passed the trap, and the trap showed no activity after the residual active gas was flushed out: all of the activity followed the argon which had passed through. The reaction therefore is undoubtedly of type (1), and the active isotope is probably A^{41}. This identification is supported not only by the relative abundance of A^{36} and A^{40}, but by the fact that A^{41} would then decay to a known and (presumably) stable potassium isotope:

$$A^{41}\rightarrow K^{41}+e^-,$$

whereas if A^{37} were the active body the radioactive change

$$A^{37}\rightarrow K^{37}+e^-$$

would lead to potassium isotope which is unknown, and the existence of which in a stable form would constitute a violation of the "odd number rule."

The identification of A^{41} as the carrier of the 110-minute activity is in accord with the form of the excitation function for the radioactivity, and with the results of the bombardment of argon by neutrons. These matters are dealt with later in the paper.

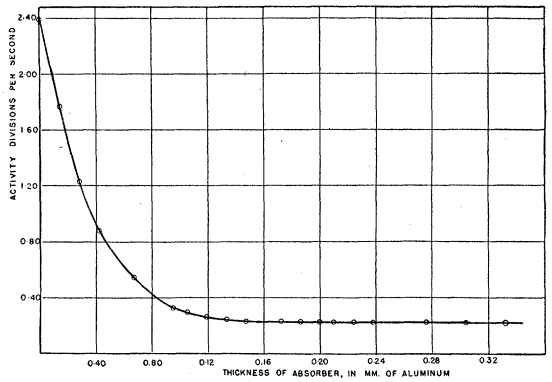

FIG. 3. Absorption curve for the β-rays of radio-argon in aluminum. Erratum: In the scale of abscissae, the figures to the right of 0.80 should all be multiplied by 10.

THE RADIOACTIVE β-PARTICLES

The absorption of the β-particles was studied by fixing a sample of the active gas two inches from a quartz fiber electroscope and ionization chamber, and placing between them various thicknesses of aluminum absorber. Three different tests yielded similar curves, one of which is reproduced in Fig. 3. It will be seen that when the absorber thickness reaches 1.6 mm of aluminum the β-rays are apparently entirely absorbed, and there remains a well-marked γ-ray background. Applying Feather's[4] empirical rule relating the maximum range of β-rays with their energy, one obtains the value 1.1 MV for the maximum energy of the particles emitted by radioactive argon.

A more detailed study of the argon β-ray spectrum has been carried out by Kurie, Richardson and Paxton,[5] who have recently examined the β-ray spectra of several artificially prepared radioactive light elements by measuring the curvature of the tracks in a hydrogen-filled cloud chamber traversed by a magnetic field. They have found that in general the shapes of their distribution curves are in very good agreement with the Konopinski-Uhlenbeck[6] modifications of the Fermi theory of continuous β-ray emission, and have accordingly adopted the practice of obtaining the upper limits of the spectra by fitting

[4] N. Feather, Phys. Rev. **35**, 1559 (1930).
[5] F. N. D. Kurie, J. R. Richardson and H. C. Paxton, Phys. Rev. **49**, 368 (1936).
[6] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 7, 107 (1935).

the shapes of the curves with theory, and extrapolating to an upper limit. The extrapolation usually gives a value for the upper limit which is somewhat greater than the energy of the fastest particle observed, and is rendered necessary by the fact that the number of particles having energies which approach the upper limit is, according to the Konopinski-Uhlenbeck theory, so small that there is little chance of observing them in a cloud chamber without measuring so many tracks as to make the problem wholly impracticable. In the case of argon, Kurie, Richardson and Paxton have found evidence for two groups of β-particles. The lower energy group has an upper limit of 1.5 MV, and the higher (and relatively much less abundant) group has an upper limit which is tentatively set at about 5 MV. The tracks were obtained by collecting active recoils upon a piece of clean platinum placed in the deuteron beam as it traversed an atmosphere of argon, and then placing the sample in a recess in the cloud chamber. The sample was allowed to "age" before photographs were taken, in order to allow activities due to short period contaminants to subside. To ensure that the high energy component of the spectrum was not due to a contaminant on the platinum, nor to the platinum itself, a trace of the activated gas was later fed into the cloud chamber. Examination of some thirty photographs taken under these conditions showed that the high-energy tracks were still present in about the same relative number as before.

The results of this cloud chamber work are not in disaccord with the absorption measurements. The thinly populated high energy group could not be expected to be separable with the electroscope from the γ-ray background, and the upper limit of 1.5 MV found by the method of Kurie, Richardson and Paxton for the lower energy group is in good enough agreement with the value 1.1 MV given by Feather's rule.

The Radioactive γ-Rays

The energy of the γ-rays emitted from radio-argon has been studied by Mr. J. R. Richardson,[7] who has measured the curvature of the tracks of the Compton recoils in a cloud chamber in a magnetic field. The recoil electrons were driven from a lamina of mica placed inside the chamber. Mr. Richardson finds that the γ-ray spectrum of radio-argon consists of a single line, which has an energy of 1.39 MV.

The Excitation Function for the Radioactivity

To investigate the nature of the transmutation function for the radioactivity, use was made of a method which has become standard in this laboratory. A stack of ten platinum foils, each having a stopping power of about 1 cm of air, was arranged in a holder such that they were equally spaced 3.2 mm apart. The stack was immersed in argon at atmospheric pressure, and the deuteron beam was allowed to pass through the alternate layers of platinum and argon so formed. The platinum foils served both to retard the beam and to collect the activated argon recoils, so that the strength of the argon activities on the foils was a measure of the yield of radioactive atoms at the deuteron energies concerned. Variations in the deuteron current during the course of bombardment caused no trouble, for all foils automatically received the same total bombardment. The progressive decrease in the intensity of the beam due to scattering and to nuclear reactions was entirely negligible.

The stopping powers of the foils were measured independently with polonium α-particles. However, since the deuterons passing through the foils had velocities which for the most part were considerably greater than the velocity of the α-particles, it was necessary to correct the measured thicknesses of the foils to suit the faster particles. Mano[8] has given data relating the atomic stopping powers of a number of common substances with the velocities of charged particles passing through them, and his results were used in making the necessary corrections. As applied here, however, the corrections may be in error, because in using Mano's formula to calculate the stopping power of the platinum for the polonium α-particles (and likewise the slower deuterons), it was necessary to extrapolate to velocities lower than those for which the formula is valid. The

[7] J. R. Richardson, Phys. Rev. **49**, 203 (1936), and later work, as yet unpublished.

[8] G. Mano, J. de physique **5**, 628 (1934).

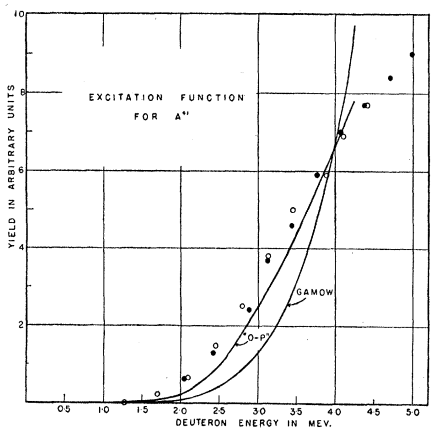

FIG. 4. The excitation function for radioactive argon—experimental points and two theoretical curves.

stopping powers of the foils were calculated for each foil individually and in turn. In the extreme case of the fastest deuterons the correction amounted to an increase in the measured stopping power of the first foil by about 40 percent. The similar correction for variation of the stopping power of the argon with deuteron velocity was negligible, because of its lower atomic number and the smaller total retardation for which it was responsible.

The experimental points of Fig. 4 represent the results of two measurements of the excitation function for the argon radioactivity, obtained and corrected in this manner. In both experiments the initial deuteron energy was measured by stopping the deuteron beam in air and aluminum foil of known thickness; in the first test (circles) the initial deuteron energy so measured was 4.5 MV (17.5 cm range), and in the second test (dots) it amounted to 5.1 MV (21 cm range). In either case this measurement may be in error by as much as ± 0.3 MV. The arrangement of the foils in their holder was reversed between the two experiments. The activities of the individual foils were measured with the electroscope, and the decay of the radio-argon on each foil was followed for several hours. The activities obtained in the two tests have been adjusted in Fig. 4 to fit at the deuteron energy of 4.0 MV.

For comparison with the experimental points, the figure also shows the theoretical excitation functions as given, respectively, by the theory of Gamow[9] for the penetration of a potential barrier by a charged particle, and by the theory of Oppenheimer and Phillips[10] for the "acceptance" by the bombarded nucleus of the neutron belonging to the incident deuteron, and the "rejection" of its constituent proton. The curve for the Gamow theory has been calculated from the formula

$$\sigma = K(1/v^2)e^{-4\pi^2 Ze^2/hv},$$

in which σ represents the effective cross section for the nuclear reaction, K is a proportionality constant, v is the velocity of the incident deuteron, and Z is the charge of the bombarded nucleus. The "O−P" curve has been calculated from the expression given by Oppenheimer and Phillips in their paper, using for the binding energy of the deuteron the value 2.1 MEV. Adjustment has been made in each case to fit the experimental data at the abscissa of 4.0 MV. The Oppenheimer-Phillips theory applies only to reactions of the type ${}_zX^n + {}_1H^2 = {}_zX^{n+1} + {}_1H^1$, and has been experimentally established by Lawrence McMillan and Thornton[11] for sodium, aluminum, silicon and copper. In the case of magnesium also, a very clear-cut experimental distinction between these two types of excitation function has recently been given by Henderson.[12] For argon, it is apparent that the experimental points give better agreement with the theory of Oppenheimer and Phillips than they do with the theory of Gamow; this is to be expected, for it is in agreement with the identification of A^{41} as the radioactive isotope.

A second correction might be applied to the experimental points in Fig. 4 to allow for the variation of recoil range with deuteron velocity, for the foils traversed by the faster deuterons collect recoils from a greater depth of argon than do those traversed by the slower deuterons. Lacking a knowledge of the momenta of the protons emitted during the reaction, this correction has not been made. Its application would decrease the slope of the experimental curve, and make the agreement with either theoretical curve less good. If one neglects the effect of the protons,

[9] G. Gamow, *Atomic Nuclei and Radioactivity* (Oxford University Press, 1931).
[10] J. R. Oppenheimer and M. Phillips, Phys. Rev. **48**, 500 (1935).
[11] E. O. Lawrence, E. McMillan and R. L. Thornton, Phys. Rev. **48**, 493 (1935).
[12] M. C. Henderson, Phys. Rev. **48**, 855 (1935).

and assumes that the whole of the momentum of the incident deuterons is transferred to the argon recoils (certainly a gross over-correction), one finds that the slope of the experimental curve is decreased by roughly 20 percent. However, the uncorrected points, as reproduced in Fig. 4, are probably nearer to the true excitation function.

In the two tests, the argon activities on the foils struck by deuterons with an energy of 4 MEV were equivalent, respectively, to about 1.7 and 0.8 microcuries, at the time of cessation of activation. In the first case, the total deuteron bombardment amounted to 90 microampere-minutes, and in the second case it amounted to 48 microampere-minutes. Assuming that the A^{41} recoils had a range of 1 mm, the effective cross section (σ) for the making of radio-argon is found from both tests to have the value of roughly 8×10^{-26} cm^2 for deuterons with an energy of 4 MEV.

Activation of Argon by Neutrons

It would be expected that radio-argon could be made by subjecting argon to the action of neutrons; in particular, slow neutrons should be especially effective in inducing the 110-minute activity. To see if this is so, about 150 cc of argon at slightly above atmospheric pressure was placed in a cavity at the center of a large paraffin block. The block was placed upon the cyclotron in such a position that the argon sample was about 20 cm from the beryllium target which, when struck by 5.5 microamperes of 5 MV deuterons, constituted the neutron source. Activation was continued for 80 minutes. The argon was then transferred to a thin-walled vessel, and tested for radioactivity with the pressure ionization chamber previously mentioned. The activity was exceedingly weak. Its decay was followed for an hour, but the background fluctuations were large enough to render the readings very erratic; however, they were not inconsistent with a decay period of 110 minutes. To make the test more conclusive, it was repeated with stronger neutron activation. In the second test the possible gain to be obtained by using slow instead of fast neutrons was sacrificed in favor of better geometrical conditions. The paraffin block was dispensed with, and the argon (at 1.2 atmospheres) was contained in a cylindrical volume with its axis two inches from the beryllium target. A deuteron current of 8 microamperes at 5.7 MV striking the beryllium provided an exceedingly powerful neutron source. Activation lasted 60 minutes. The induced activity was again found to be very weak when examined in the same thin-walled vessel with a quartz-fiber electroscope. However, it was possible at the time of this test to introduce the gas directly into the ionization chamber of the electroscope. When this was done the rate of discharge was increased to about ten times the background, and by following the decay for $3\frac{1}{2}$ hours, the presence of the 110-minute period was definitely established.

Although these two tests were not designed to be in any way quantitative, it is possible to deduce from them roughly the relative efficiencies of fast and slow neutrons in activating argon. The two activated samples were exposed, respectively, to the pressure ionization chamber and to the electroscope under geometrical conditions which were nearly alike. By knowing approximately the relative sensitivities of the pressure chamber and of the electroscope, and that the same quantity of gas was present in each case, it is estimated that the two samples had about the same absolute activity. During bombardment, on the other hand, the geometrical conditions and the strength of the neutron source together favored the second sample by a factor of about 30; it can accordingly be concluded that the slow neutrons were very roughly 30 times more effective in producing radio-argon than were the fast neutrons.

Acknowledgments

The writer is greatly indebted to Professor E. O. Lawrence for many suggestions made during the course of this work, and also he wishes to express his thanks to his colleagues in the Radiation Laboratory for helpful discussions. The generous personal support of the Royal Commissioners for the Exhibition of 1851 is gratefully acknowledged. The experimental work has been supported by the Chemical Foundation, the Research Corporation, and the Josiah Macy Junior Foundation.