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The Radioactivity Produced in Nickel by Deuteron Bombardment[1]

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Nickel bombarded with 5 MV deuterons exhibits an intense positron activity of half-life 3.4±0.1 hours. Chemical analysis shows this activity to be isotopic with copper. The energy excitation curve for this reaction (proton capture) between 3 and 5 MV has been obtained, and is found to be very different from that found for copper (neutron capture). The variation with energy of the disintegration cross section agrees satisfactorily with theoretical formulae for the penetration of a deuteron into a nucleus of radius 4.5×10^{-13} cm. This result does not support the larger nuclear radii recently suggested by Bethe. The upper limit of the positron spectrum is approximately at $H\rho$ 4400 gauss-cm.

EARLY attempts to induce artificial radioactivity in nickel through neutron bombardment by Fermi and his collaborators[3] led to negative results. More recently Rotblat[4] has obtained evidence for two active isotopes by this method—a water-sensitive activity of several hours half-life which he attributes to an isotope of nickel, and a shorter period of twenty minutes which could be associated with cobalt. In view of the extreme weakness of the activity produced, however, these disintegrations cannot be regarded as being definitely established.

In the experiments to be described, nickel has been subjected to bombardment with several microamperes of deuterons accelerated to 5 MV by means of a magnetic resonance accelerator or cyclotron. The results obtained show the production of a single positron radioactive isotope of copper; no evidence being found for the nickel activity reported by Rotblat. The dependence of the induced activity upon the deuteron energy, and the energy distribution of the positrons have also been ascertained.

The nickel was bombarded in the form of a pure metal. For the determination of the period and of the excitation curve, thin foils (0.0001″ in thickness) were used; while for the later experiments on the β-ray spectrum a somewhat thicker target was employed. In each case the purity of the nickel was attested by the observation that only a single period that could be ascribed to an impurity was obtained—the ten-minute period of radionitrogen.

The energy excitation curve was determined through the simultaneous bombardment of a number of thin foils. The foils were mounted in suitable holders which were protected from bombardment, and the decay curve of each was established. A typical group of such curves is shown in Fig. 1. From these plots the relative activities of the several foils could be readily and accurately

[1] A preliminary report of these experiments has been made to the American Physical Society, Phys. Rev. **49**, 207 (1936).
[2] The greater part of the experiments described were carried out while the writer was at the Radiation Laboratory of the University of California. The cloud chamber experiments were performed at the University of Michigan.
[3] Amaldi *et al.*, Proc. Roy. Soc. **A149**, 522 (1935).
[4] Rotblat, Nature **136**, 515 (1935).

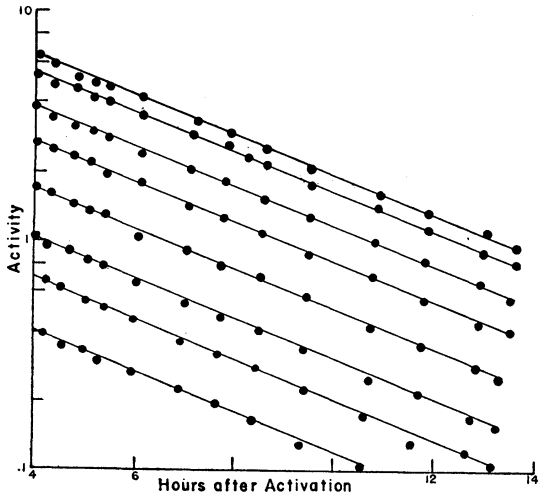

FIG. 1. Semi-logarithmic plot of the decay curves for activated nickel foils.

ascertained, and from the known stopping power of the foils the differential excitation curve of the reaction determined. But before this can be done a number of corrections must be applied to the observations.

The stopping power of the individual foils was measured relative to air by means of polonium α-particles. It was found that the metal was not completely uniform, and a correction to reduce all foils to a common thickness was necessary. This amounted to as much as 10 percent—the mean thickness being 8.6 mm air equivalent. In addition it is necessary to correct for the variation of stopping power with the velocity of the incident deuterons. This was done using the theory developed by Mano.[5] The activation of each foil except the first is enhanced through the recoil of activated atoms from the preceding foil; it is thus to be expected that the measured intensity of the first foil will be somewhat too low. Since the observed points lie along a smooth curve, it was assumed that this correction was small and no allowance was made for it.

The energy of the incident deuterons was determined from range measurements in aluminum. Errors arising from this determination may be as high as 6 percent; relative energy measurements between the several foils are certainly considerably more accurate than this.

The decay curves for the foils were followed with a Lauritsen type electroscope, and with a pressure ionization chamber vacuum tube electrometer combination constructed by Dr. S. N. Van Voorhis.

The complete decay curves for the activated nickel may be satisfactorily explained on the basis of two periods—an intense ten-minute activity which in all probability is due to carbon contamination, and a 3.4 ± 0.1 hour activity which chemical analysis established as being isotopic with copper. The chemical procedure followed that outlined by Bray.[6] Copper and cobalt salts were added to the dissolved nickel, and the three elements precipitated separately. It was found that the relative activities of the three precipitates were 10.0, 0.1 and 0.05 for the copper, nickel and cobalt, respectively.

Investigation of the active copper in a Wilson cloud chamber showed that positive electrons were emitted, and thus the resulting element must be an isotope of nickel. The isotopic constitution of nickel has been investigated by Dempster[7] who found isotopes of mass numbers 58, 60, 61, 62 and 64. The reaction may thus be most probably written

$$_{28}\mathrm{Ni}^{N}+{}_{1}\mathrm{H}^{2}\rightarrow{}_{29}\mathrm{Cu}^{N+1}+{}_{0}n^{1}$$

$$_{29}\mathrm{Cu}^{N+1}\rightarrow{}_{28}\mathrm{Ni}^{N+1}+e^{+}$$

where the possible values of N are 58, 60 and 61. Of these the first possibility results finally in an unknown isotope of nickel, and since no other activity than the single period was observed, it is more probable that the disintegration involves either Ni^{60} or Ni^{61}. The possibility of a radiative capture of the deuteron, although unlikely, is of course not excluded.

Since the formation of this active copper isotope results from the capture of a proton accompanied by neutron emission, considerable interest attaches to the shape of the energy excitation curve in comparison with those obtained for cases of neutron capture and proton emission. In order to account for the shape of the latter curves, and for the large disintegration cross sections observed for neutron capture reactions for elements of high

[5] Mano, J. de phys. et rad. **5**, 628 (1934).

[6] Noyes and Bray, *Qualitative Analysis for the Rare Elements* (Macmillan, 1927). I am indebted to Dr. H. W. Newson for his help with the chemical analysis.

[7] Dempster, Phys. Rev. **50**, 98 (1936).

atomic number, Oppenheimer and Phillips[8] have suggested that such disintegrations do not require the penetration of the deuteron as a whole into the nucleus. On this basis a formula giving the variation of yield with deuteron energy was obtained, which has been shown by Lawrence, McMillan and Thornton[9] and by Henderson[10] to be in excellent agreement with experiment for such reactions in the cases of sodium, aluminum, silicon, copper and magnesium.

More recently, however, Bethe[11] has suggested that such considerations are not necessary to explain the observations, and that they may be interpreted as evidence for a larger nuclear radius than has hitherto been accepted. In addition to the arguments advanced by Bethe, further support for this view is given by Pollard[12] through the observation of low energy protons emitted from certain light elements under α-particle bombardment, and from estimates of the yields obtained.

In a number of papers Sexl[13] has investigated the penetration of charged particles through nuclear potential barriers, and has developed a formula showing the dependence of the disintegration cross section σ upon the energy of the

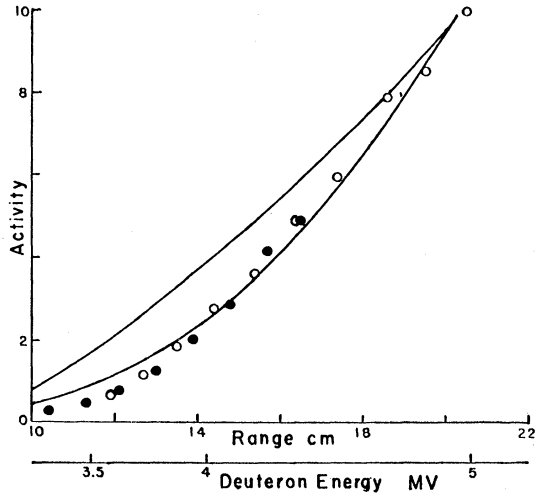

FIG. 2. Energy excitation curves for nickel (lower curve) and for copper (upper curve). The curve through the experimental points for nickel corresponds to a nuclear radius of 4.5×10^{-13} cm.

[8] Oppenheimer and Phillips, Phys. Rev. **48**, 500 (1935).
[9] Lawrence, McMillan and Thornton, Phys. Rev. **48**, 493 (1935).
[10] M. C. Henderson, Phys. Rev. **48**, 855 (1935).
[11] Bethe, Phys. Rev. **50**, 977 (1936).
[12] Pollard, Phys. Rev. **51**, 290 (1937).
[13] Sexl, Zeits. f. Physik **87**, 105 (1933).

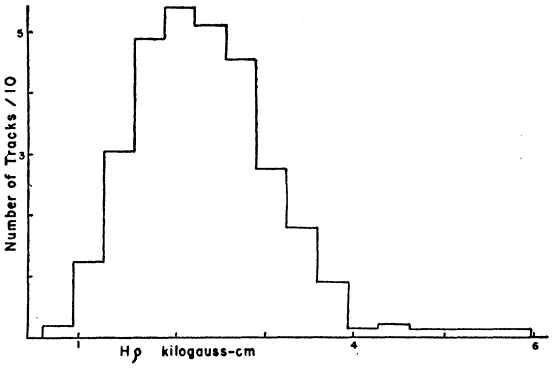

FIG. 3. Momentum distribution of 300 positrons from activated nickel target.

projectile and upon the nuclear radius. This formula may be written

$$\sigma(E)=(\text{const.}/E)e^{-\chi(2\alpha_0-\sin 2\alpha_0)},$$

where E is the energy of the bombarding deuteron of velocity v, $\chi=2e^2Z/\hbar v$, $\cos^2\alpha_0=Er_0/e^2Z$ and r_0 is the nuclear radius. In deriving this expression, only the probability of penetration of the deuteron has been taken into account. To completely describe the cross section, the probability of the reaction proceding after the penetration of the deuteron should also be considered. This, however, cannot be done as yet, but we may reasonably assume that the emission of a charged particle will correspond to a somewhat steeper excitation curve than that for the emission of a neutron.

Thus the comparision of the excitation curves for proton capture in nickel ($Z=28$) with those for neutron capture in copper ($Z=29$) provides a convenient test for these points of view. Since the nuclear radius is approximately proportional to the cube root of the total number of nuclear particles, it should not be very different for the two cases. Hence if in each case the deuteron as a whole penetrates the nucleus, we may expect the copper curve to be slightly steeper than that for nickel (Z is larger and a charged particle must escape). On the other hand if the views of Oppenheimer and Phillips are correct, and also if we assume a radius for the natural radioactive atoms of 9×10^{-13} cm (about 5×10^{-13} for nickel), then the nickel curve should be the steeper.

The experimental data presented in Fig. 2, corrected as outlined previously, are the result of two independent runs taken at different initial

deuteron energies. The two curves were adjusted to fit at 16.5 cm deuteron range. In comparison with the observations for nickel are shown the data obtained by Van Voorhis[14] for copper (the upper curve in Fig. 2). The different character of the two curves is immediately evident. The second curve in Fig. 2 through the experimental points is a plot of Sexl's formula for a nuclear radius $r_0=4.5\times10^{-13}$ cm, adjusted to fit the experimental points at 20 cm range. The agreement would appear to be satisfactory. It was found that an increase of the radius to 5.0×10^{-13} cm resulted in a curve lying completely above the experimental points, while a value of 8×10^{-13}, which Bethe's value of 13×10^{-13} for the natural radioactive elements would suppose, is very decidedly too large. Thus from the data presented here it would seem that it is not possible to account for the "flat" excitation curves observed for neutron capture reactions by solely supposing an increase in the nuclear radius.

Measurements of the radii of curvature of 300 positron tracks in a Wilson cloud chamber in a plane perpendicular to a magnetic field of 340 gauss resulted in the histogram presented in Fig. 3. Only tracks were measured which clearly originated in the target and had a length greater than 7 cm. The presence of the five tracks observed beyond the "inspection" upper limit at $H\rho$ 4400 gauss-cm (0.9 MV) may be explained either as being due to γ-rays which are known to be present but have not as yet been measured, or to the presence of a second and higher energy positron group as has been observed in other cases.[15] Decision between these alternatives must await further experiments.

It is a pleasure for the writer to acknowledge his very great indebtedness to Professor E. O. Lawrence, and to Professor J. R. Oppenheimer for his friendly advice upon the theoretical questions involved. The support of the Research Corporation, the Chemical Foundation and the Josiah Macy, Jr. Foundation at the University of California, and of the Horace H. and Mary A. Rackham Fund at the University of Michigan is also gratefully acknowledged.

[14] Van Voorhis, Phys. Rev. **50**, 895 (1936). I am much indebted to Dr. Van Voorhis for his kindness in sending me a copy of his experimental curve.

[15] E.g., Kurie, Richardson and Paxton, Phys. Rev. **49**, 368 (1936); M. V. Brown and Mitchell, Phys. Rev. **50**, 593 (1936).

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Neutron Yields from Artificial Sources

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Measurements of the total yields of neutrons from the reactions D+D, D+Li, D+Be and D+C have been carried out for the voltage range between 300 kv and 1000 kv using the observing technique developed by Amaldi and Fermi, in which the neutron source is surrounded by a large tank of water. Neutrons of all energies are thus reduced to thermal energies, and the total yield of neutrons is obtained by integration from measurements of the density of slow neutrons at various distances from the source. Similar measurements were made on the neutrons from Rn+Be, with the same geometrical conditions. The neutron yields per microampere of pure D^2 ions from the artificial sources are shown in the table below for various voltages; each entry gives the number of millicuries of Rn+Be required to give the same total yield of neutrons per second. A total yield of 25,000 neutrons per second from one millicurie of Rn+Be is calculated from the measurements of this source. Absolute yields for the various sources computed on the basis of this figure should not be considered as reliable within 20 percent, perhaps the chief error arising from the fact that the average ionization per beta-ray is assumed to be the same for the beta-rays from the rhodium detector and for those from

REACTION	300 kv	400 kv	600 kv	800 kv	1000 kv
D+$D_2O(P_2O_5)$	48	140	250	550	860
D+Li	(40)	(160)	(800)	(4200)	...
D+Be	9	100	700	2400	6800
D+C	..	...	10	140	680