Concentration of A^{36} and A^{38} and Study of the $A^{36}(d, p)A^{37}$ Reactions and the Beta-Activity of A^{39}*

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The concentration of A^{36} up to 95.9 percent by the thermal diffusion method is described. Observation of the proton groups resulting from the deuteron bombardment of this gas gives the following Q-values for the $A^{36}(d,p)A^{37}$ reaction: 6.49, 5.05, 3.93, 2.95, 2.09, 1.86, 1.42 and 0.64 Mev. Comparison with the proton distribution from $A^{40}(d,p)A^{41}$ shows the importance of having well-separated isotopes for the resolution of proton groups of nearly the same energy from each isotope. The enrichment of A^{38} to 0.75 percent in this same gas makes measurement possible on the betas from the $A^{38}(d,p)A^{39}$ reaction. These A^{39} beta particles have a half-life of 160 ± 5 seconds and a maximum energy of about 2.1 Mev.

I. INTRODUCTION

IN a recent article,[1] Davison, Buchanan, and Pollard reported their observations on the proton groups from the deuteron bombardment both of ordinary argon and of argon with a 26.2 percent content of A^{36}. This A^{36} concentration was produced by Buchanan[2] in a multi-stage thermal diffusion apparatus. Despite this very considerable enrichment in A^{36} from the normal 0.31 percent, there was evident masking of most of the short range A^{37} proton groups by overlapping A^{41} groups. To display clearly the proton groups from the $A^{36}(d,p)A^{37}$ reactions, we have repeated these measurements, using argon gas in which the A^{36} isotope was concentrated to 95.9 percent. The concentration of the rare A^{38} to 0.75 percent in the same gas has also made possible the determination of the half-life of A^{39} and roughly the maximum energy of the β-particles which it emits.

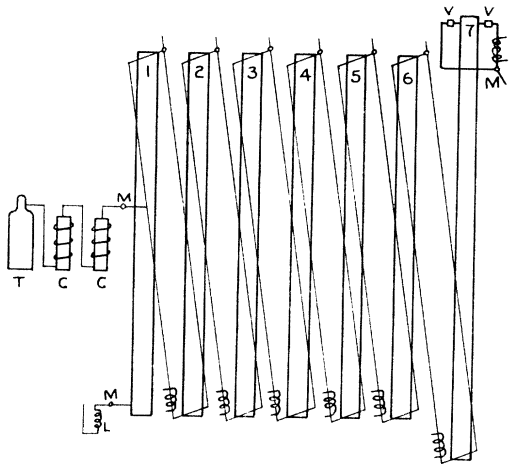

FIG. 1. Schematic diagram of all-metal multi-stage thermal diffusion apparatus for concentration of A^{36}. T—argon cylinder, C—hot calcium traps, M—McKay valves, L—capillary leak, V—sylphon bellows, wide-opening valves; 1–6 are concentric cylinder columns, 7 is a hot-wire column.

II. A^{36} AND A^{38} CONCENTRATION

The major part of our thermal diffusion apparatus consisted of the same six all-metal columns, each based on a copper-clad Calrod heater of 200 cm effective heating length, used by Buchanan and described recently[3] as a portion of some equipment for the concentration of Ne^{22}. Since for neon the heavy isotope is to be concentrated, whereas for argon one wishes to concentrate the light isotope A^{36}, it was necessary to reverse the convective coupling pipes connecting the columns. The temperature of the hot walls was 450°C, as determined by measuring the extension of the Calrods.

As a seventh unit in this chain, we connected convectively, just as Buchanan did, a 3-meter hot-wire column. This column we constructed entirely of metal, however, so that we could operate the apparatus at elevated pressures. The details of this hot-wire column are similar to those of a column we had operated for some time[4] in a high pressure thermal diffusion apparatus for the concentration of He^3. An innovation was to make the hot wire of 20-mil nichrome which operated successfully for considerable periods of time at 1000°K.

Figure 1 gives the schematic arrangement of this apparatus. It was necessary to remove the nitrogen impurity (about 0.4 percent) in the tank argon, for otherwise the "light" end volume at the top of column 7 would soon become choked with nitrogen. The gas was therefore first purified by passing it through two stainless-steel traps containing calcium metal shavings at about 400°C.

Since our aim was to attain a maximum separation factor, we calculated the gas pressure which would give the desirable value[5] of about two for the ratio K_c/K_d of the two remixing coefficients. This calculation showed that, if $p=1.06$ atmos. this condition would prevail in

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[1] Davison, Buchanan, and Pollard, Phys. Rev. **76**, 890 (1949).

[2] J. O. Buchanan, Phys. Rev. **75**, 1332A (1949).

[3] Watson, Onsager, and Zucker, Rev. Sci. Inst. **20**, 924 (1949).

[4] Schuette, Zucker, and Watson, Rev. Sci. Inst. (to be published).

[5] R. C. Jones and W. H. Furry, Rev. Mod. Phys. **18**, 151 (1946).

columns 5 and 6, with the transport $H=3.48\times10^{-5}$ g sec.$^{-1}$, $K_c=2.59\times10^{-3}$ g cm sec.$^{-1}$, $K_d=1.37\times10^{-3}$ g cm sec.$^{-1}$, and the separation factor $q=33.4$. For columns 1 and 2 at this same pressure $K_c/K_d=10.4$ which is a value well below the critical ratio of ~25 at which turbulence might set in.[6] A preliminary calculation had shown that $\frac{3}{8}''$ i.d. copper pipe was the best available size for the cold wall of the hot-wire column. For this column 7 at this optimum pressure $H=1.69\times10^{-5}$ g sec.$^{-1}$, $K_c=5.72\times10^{-4}$ g cm sec.$^{-1}$, $K_d=2.60\times10^{-4}$ g cm sec.$^{-1}$, and $q=440$.

Column 1 was operated as a scrubber, and the gas was fed into the convective coupler between columns 1 and 2. The argon input was at the rate of about 250 cm^3/hr. At this rate the argon bled into the atmosphere through a capillary leak at the bottom of the scrubber was depleted of about half of its A^{36} content.

The product of the calculated separation factors for the last three columns alone has the impressive value of 14,700, sufficient to give a concentration of 98 percent A^{36} at the top of column 7. In actual performance, however, the A^{36} content in the 200 cm^3 end volume after 200 hours was but 20 percent, while after an additional 200 hours the enrichment had reached only 22.5 percent. In steady operation we found the apparatus capable of producing 200 cm^3 of 20 percent A^{36} every 70 hours. This relatively low experimental separation factor we attribute as usual to a fairly large coefficient of parasitic remixing. The total power consumption of this apparatus was about 7 kw.

To increase the concentration of A^{36} to the desired 90 percent we proceeded to recycle this 20 percent product in a hot-wire column. This was a glass column 3 meters long, with cold wall of 1.018 cm i.d., and with a 20-mil tungsten wire operated at 1500°K. Convective circulation was maintained through an end volume both at the top and the bottom as shown in Fig. 2. The "heavy" end volume had a capacity of 200 cm^3, that at the "light" end 50 cm^3, while the column itself had a volume of 250 cm^3. At 1500°K and 1 atmos. pressure calculation gave for this column $H=1.085\times10^{-5}$ g sec.$^{-1}$, $K_c=3.48\times10^{-4}$ g cm sec.$^{-1}$, $K_d=5.23\times10^{-4}$ g cm sec.$^{-1}$ and $q=42$.

The column was charged with 500 cm^3 of the 20 percent A^{36} gas, operated for 12 hours, and then the lower end volume was isolated and its gas content removed. A fresh 200 cm^3 batch of 20 percent A^{36} was then pumped into this reservoir and connection to the column was re-established for 12 more hours. Then this process was repeated once again. With this 36 hours of operation the separation was completed, the "light" end volume was isolated, and its contents were withdrawn into a breakseal tube.

In order to conserve the concentrated A^{36} material, the increase in concentration was followed by an analysis of the gas in the "heavy" end each time a new batch was about to be added. These mass spectrometer analyses are given in Table I. Knowing the total amount of A^{36} in the column and assuming a uniform concentration gradient up the column, we could then calculate that the light end volume should contain 91.9 percent A^{36}. A mass spectrometer analysis of this product showed its A^{36} content to be 92.6 percent. From another similar set of operations 50 cm^3 of 95.9 percent A^{36} were obtained. The A^{38} content of these samples averaged 0.75 percent.

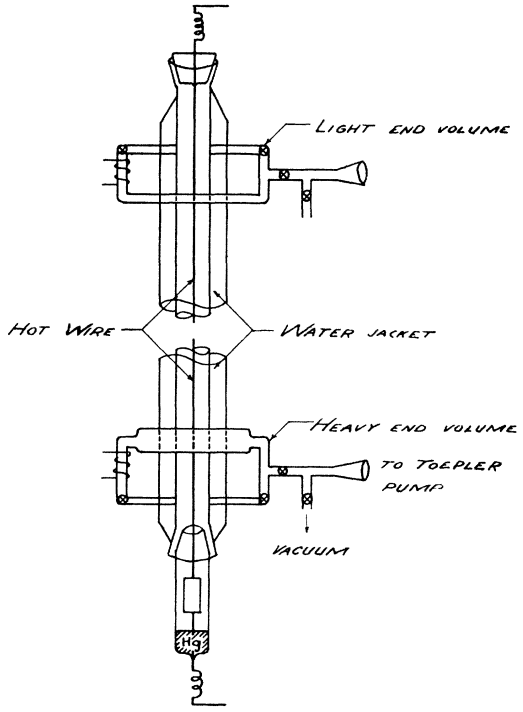

FIG. 2. Details of glass hot-wire column used for recycling the argon partially concentrated in A^{36}.

We conclude that this recycling of the partially concentrated light argon is an effective procedure. The over-all efficiency could be increased by feeding back into the first stage of the all-metal apparatus the product withdrawn from the lower end-volume of the glass hot-wire column before injection of each new batch of 20 percent A^{36} gas. Also it seems obvious that the isotope of intermediate weight, A^{38}, might have its greatest concentration at that point in the glass column where the A^{36}/A^{40} ratio is 50/50, for the thermal diffusion plus thermal syphoning effects would tend to send the A^{38} towards this intermediate point from either end. We

TABLE I. Concentration of A^{36} with time.

Running time (hours)	Concentration A^{36}
0	~20.0 percent
12	3.35
24	5.0
36	7.95

[6] L. Onsager and W. W. Watson, Phys. Rev. **56**, 474 (1939).

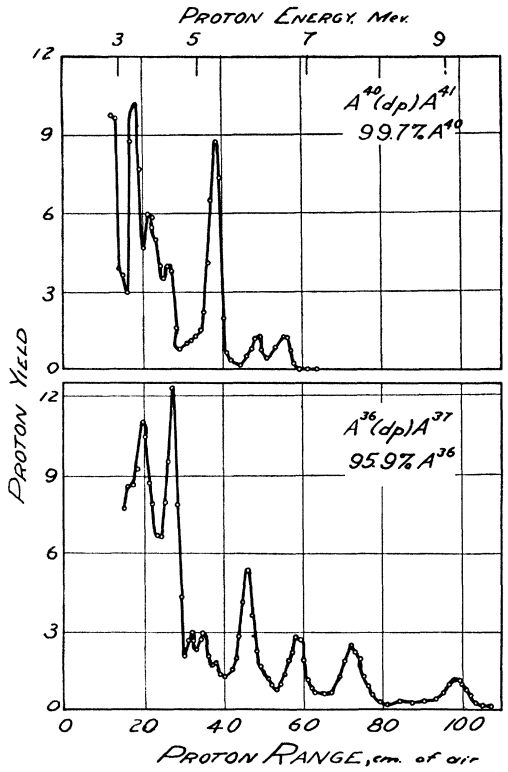

FIG. 3. Yield *vs.* energy plots of the protons from the deuteron bombardment of argon 36 and argon 40.

intend to investigate the distribution of A^{38} along this column both to check this reasoning and to obtain argon with a higher A^{38} content for further research on the β activity of A^{39} (see below).

III. THE $A^{36}(d,p)A^{37}$ REACTIONS

We have investigated the proton groups from the bombardment of this argon highly enriched in A^{36} by 3.9 Mev (extrapolated) deuterons. Before the argon was introduced into the target chamber it was freed of any nitrogen impurity by several passages through a trap containing hot calcium metal. The argon pressure in the chamber was 6.3 cm of Hg. The emitted protons were counted at an angle of 90° with the incident deuteron beam, using a proportional counter so adjusted as to count only those protons whose paths terminated in the counter. The proton energy was measured by interposing varying amounts of absorber between the bombarded gas and the counter.

TABLE II. Proton range and Q-values for the $A^{36}(d,p)A^{37}$ reactions and the energy levels of A^{37}.

Proton range cm	Q Mev	Relative yield	A^{37} energy levels Mev
106.26	6.49±0.08	1.0	ground
78.46	5.05±0.05	2.3	1.44
62.73	3.93±0.05	2.6	2.56
48.51	2.95±0.05	5.0	3.54
37.37	2.09±0.07	2.8	4.40
34.69	1.86±0.07	2.8	4.63
29.62	1.42±0.03	11.5	5.07
21.80	0.64±0.07	10.2	5.85

Figure 3 (lower half) is a plot of the yield of protons against proton energy. For comparison the corresponding measurements for $A^{40}(d,p)A^{41}$ made by Davison, Buchanan, and Pollard[1] are shown in the upper half of the figure. Close inspection of the two plots shows how important it is to have a high degree of isotope separation. The two unambiguous A^{37} proton groups with extrapolated ranges of 106 cm and 78 cm were discovered by Davison, Buchanan, and Pollard. We find no evidence confirming their suspicion that the group at range 78 cm is double. The group at 62-cm range, which is just beyond the longest range A^{41} group, was also accurately measured by Davison *et al.*, but their judgment that this group also is double is not substantiated by our observations. The A^{37} group at 48-cm range was detected and correctly identified by Davison *et al.*, but they could not determine its range accurately because of partial masking by the A^{41} group of 51 cm range.

The sharp dip occurring at 38 cm range in the lower plot in Fig. 3 we believe to come from the 3.5 percent A^{40} present in this enriched gas. The peak for this dip comes just where the prominent peak of the 40-cm-range A^{41} group is observed. We deduce from this that no other A^{37} group shown in our plot could arise from A^{40} contamination. For if the high yield 40-cm-range A^{41} group produces only such a small effect in the A^{37} proton distribution curve, none of the A^{37} peaks higher than the small one extrapolating to 38-cm range could be assigned to A^{41}.

The two A^{37} proton groups at 37-cm and 34-cm range were resolved by means of a range cell which enabled us to vary the proton absorption path by steps of a fraction of a centimeter. The most intense group at 29-cm range is so nearly at the same energy as an A^{41} group that only the argument given above assures us that this is a genuine A^{37} proton group. Finally, there is a proton group at 22 cm air equivalent, partially fused with another group at shorter range.

Table II gives all of these proton ranges and the corresponding Q-values and energy levels of the A^{37} nucleus. The proton groups at ranges 22 cm, 29 cm, 34 cm, and 37 cm are all new, while the group at 48-cm range has been measured more accurately than was possible with argon less well enriched in A^{36}.

The energy levels for A^{37} and A^{41} are plotted in Fig. 4. Most striking is the greater density of excited states in A^{41} as compared to A^{37}. Up to 4 Mev of excitation, the A^{41} nucleus has 9 levels while A^{37} has but 3. Both this difference and the appearance of levels in A^{37} at higher energies are probably related to the lesser stability of A^{41} with its four extra neutrons.

IV. BETA-ACTIVITY OF A^{39}

The A^{39} nucleus should by β-emission change to stable K^{39}. To date the only reference in the literature

to this β-activity is the assignment to A^{39} by Seaborg and Perlman in their table of isotopes[7] of a 4-minute half-life found by Pool, Cork, and Thornton[8] in the neutron bombardment of potassium. The A^{39} might have been produced in their experiment by a $K^{39}(n,p)A^{39}$ reaction. In our deuteron-bombarded argon containing 0.75 percent A^{38}, the A^{39} should be produced in detectable amounts by a (d,p) reaction. We do indeed find a β-activity of half-life 160±5 sec. which from all indications is assignable to A^{39}. There is definitely no 4-min. activity in this enriched gas. Argon 37 decays by K-capture, while A^{41} is a β-emitter of half-life 110 min. The latter is, of course, always present because of the 3.3 percent A^{40} content, but our success in measuring the A^{39} β-activity has been aided as much by the markedly lowered background of A^{41} activity as by the increased percentage of A^{38}.

This A^{38}-enriched argon at a pressure of 15.6 cm of Hg was bombarded for 15 minutes by the deuteron beam in the same target chamber used for the proton-counting experiments. The cyclotron beam was then stopped and the gas was transferred by means of a Toepler pump to a 200 cm^3 counting chamber closed at one end with a 1-mil Al foil and shielded by lead on the side facing the cyclotron. The gas pressure in the counting chamber was 7.9 cm of Hg. Counting of the β's was done with a thin-mica, end-window Geiger counter. The decay curve after correction for background counts, including those from the 110-min. A^{41} β's, clearly indicates just one half-life of 160±5 seconds. Repetition of the experiment using ordinary argon gave an activity of 110 min. within our limits of error, with a slight indication of a short-lived activity at the start of the counting.

To obtain the energy spectrum of these betas with half-life 160 sec., we have done some of this counting with Al absorbers between the counting chamber and the Geiger counter. A Fermi plot made from these data indicates that their maximum energy is about 2.1 Mev. We hope to increase the accuracy and completeness of these energy measurements when argon better enriched in A^{38}, becomes available. It can be taken as definite, however, that the maximum energy of the 160-sec. betas is considerably less than the 3.01 Mev value for Al^{28}, a possible contaminant emitter having about this half-life. The maximum energy of the A^{41} betas is 1.18 Mev.

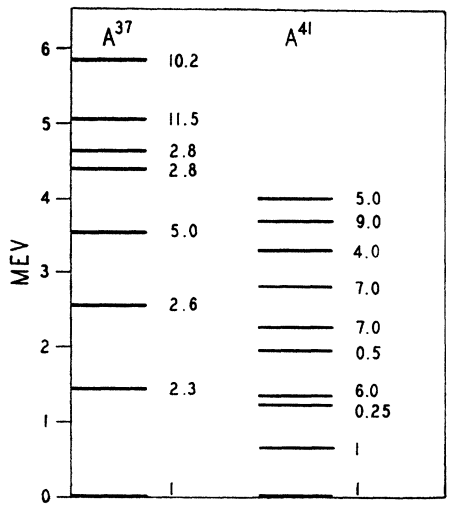

FIG. 4. Energy levels with relative abundances for A^{37} and A^{41}. Note the greater density of levels (3 to 1) for A^{41} as compared to A^{37} in the range up to 4 Mev.

From all the evidence, then, we conclude that these betas of half-life 160±5 sec. and maximum energy roughly 2.1 Mev come from A^{39}.* This energy value should be measured with greater accuracy with a beta-ray spectrometer, using material with a higher concentration of A^{38}.

We wish to thank Professor E. C. Pollard for helpful discussions.

[7] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).
[8] Pool, Cork, and Thornton, Phys. Rev. **52**, 239 (1937).

* *Note added in proof:* A beta-activity with half-life longer than 15 years and assigned to A^{39} has recently been reported by by Brosi, Zeldes, and Ketelle (Phys. Rev. **79**, 902 (1950)). They do not have positive isotope assignment, and the time necessary for the processing of their irradiated material probably would preclude the detection of an activity with a period as short as that we observe. We of course would have missed a very long lived activity. Also Haslam, Katz, Moody, and Skarsgard (Phys. Rev. **80**, 318 (1950)) suggest that, since in their study of Cl^{39} no activity that could be attributed to A^{39} was observed, the half-life of A^{39} must be longer than 5 years. Admittedly it is improbable that isomerism exists in so light a nucleus even though it immediately follows the magic number $n=20$. Further research on A^{39} produced by d, p, and n, α reactions in argon better enriched in A^{38} is in progress.