

fission fragments relative to that for  $\alpha$ -rays should, in fact, be nearly the same over the whole part of the range where the stopping is mainly caused by electronic encounters. There it is also pointed out that the straggling must be ascribed practically entirely to the end part of the range, where the effect of nuclear collisions is predominant. It is just because of the relatively greater length of this part of the range in light gases that the resultant straggling in the total range is approximately the same in helium and argon.

Corresponding to the statistical analysis of the branch distribution on the argon tracks, an examination of the branch distribution was also made in helium. We have here not much material from cloud-chamber pictures at low pressure but, fortunately, since in helium the branches are relatively longer in comparison with the length of the whole track, it was possible to perform the analysis at the relatively high pressures used in the range determination of the tracks from thin uranium targets. This circumstance gives the advantage that the separation into groups need not be made by indirect

statistical methods but can be based directly on the difference in range. The photograph (Fig. 2) shows a typical example of the paired fragment tracks in helium, with several branches on each member.

The results of the counts indicated a preponderance in branches of the shorter track, with a factor of about 1.5 near the end of the range, and about 1.8 over the remainder of the range. While the number of branches—about 120 in all—is too small to justify definite conclusions, the agreement with the previously estimated values is satisfactory and may be considered to support the general character of the range-velocity curves of the two groups of fragments. A comparison of the absolute numbers of branches within definite energy limits near the end of the range in argon and in helium also gave good agreement with the theoretical expectations.

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## Protons from the Deuteron Bombardment of the Separated Isotopes of Chlorine

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The bombardment of chlorine by 3.2-Mev deuterons is found to give rise to several groups of protons and possibly to one group of alpha-particles. By bombarding targets in which the proportion of heavy chlorine was more than doubled an assignment of some of the groups has been possible. The assignment of  $Q$  values is as follows.  $\text{Cl}^{35}(dp)\text{Cl}^{36}$ : 6.31; 5.35; and 1.50 Mev.  $\text{Cl}^{37}(dp)\text{Cl}^{38}$ : 4.02; 3.02; 2.10. These figures lead to masses of 35.9808 and 37.9806 for the isotopic masses of  $\text{Cl}^{36}$  and  $\text{Cl}^{38}$ , respectively. A group which appears at 12 cm range we assign to the light isotope according to the reaction  $\text{Cl}^{35}(d\alpha)\text{S}^{33}$ . The  $Q$  value is 9.1 Mev which gives a mass of 32.9828 for  $\text{S}^{33}$  in reasonable agreement with the mass derived from the  $\text{S}^{32}(dp)\text{S}^{33}$  reaction.

### INTRODUCTION

THE fact that the bombardment of chlorine by deuterons would give rise to protons has been known, since both the isotopes  $\text{Cl}^{36}$

and  $\text{Cl}^{38}$  have been prepared by this reaction.<sup>1</sup> The direct detection of the protons and the measurement of the ranges of the resulting groups is of interest since it permits information

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<sup>1</sup> D. C. Grahame and H. J. Walke, see Livingood and Seaborg, *Rev. Mod. Phys.* **12**, 35 (1940); F. N. D. Kurie, J. R. Richardson and H. C. Paxton, *Phys. Rev.* **49**, 368 (1936).

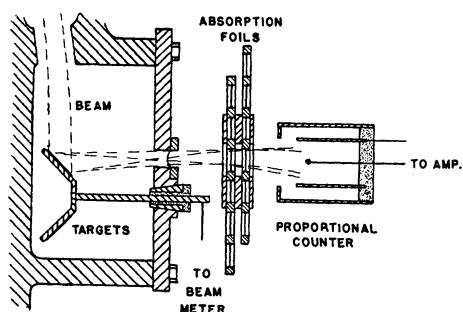


FIG. 1. Bombardment chamber, and detecting arrangements. The beam strikes the target as indicated, causing protons to be emitted in all directions. Those emitted at right angles to the beam emerge through an aluminum foil, pass through various absorbing foils and are detected in a proportional counter.

regarding masses of several nuclear species to be obtained. This information cannot, however, be obtained unless the assignment of the appropriate groups to the isotope responsible can be made, an assignment which is impossible unless the proportion of the two isotopes in targets can be altered. By means of a thermal diffusion isotope separation apparatus it has been possible to obtain targets with definitely different proportions of the two isotopes and an assignment made of all the groups definitely resolved.

#### EXPERIMENTAL ARRANGEMENTS

The apparatus for the separation of the isotopes has been described elsewhere.<sup>2</sup> The ratio of the heavy to the light isotope was determined by bombarding samples of HCl gas containing the normal proportions and separated proportions by the cyclotron beam for the same time and same beam current. The relative amounts of the 37-minute  $\text{Cl}^{38}$  were then used to measure the amounts of  $\text{Cl}^{37}$  present in the two samples. This is a simple method of making such a determination and is particularly to be recommended in this case as HCl is rather unpleasant to introduce into a mass spectrometer.

The chlorine was bombarded as silver chloride, previous experiments having shown that the silver gave a negligible yield in comparison with the yield of protons from the chlorine. The general technique has been described in a previous paper<sup>3</sup> and needs no remark except as

<sup>2</sup> E. F. Shrader, in course of publication.

<sup>3</sup> E. Pollard, W. L. Davidson, Jr. and H. L. Schultz, *Phys. Rev.* **57**, 1117 (1940); E. Pollard and W. W. Watson, *ibid.* **58**, 12 (1940).

concerns the preparation of the targets. We found that a target prepared by precipitation of AgCl with silver nitrate gave uncertain results at long ranges, and decided that some nitrogen had been entrained with the precipitate of silver chloride. Moreover, the layer of AgCl was non-uniform and unsuitable for the detection of proton groups. We therefore prepared targets by electrolysis as follows. The chlorine to be bombarded was obtained first in the form of approximately 0.02N HCl. A thoroughly clean piece of silver about 3 cm<sup>2</sup> in area forming the anode, a thin and very uniform layer of AgCl was electroplated from the solution on to the silver. A current of 0.075 ampere flowing for about two minutes is sufficient to coat the surface of the silver with one to two milligrams of chlorine per square centimeter. After allowing to dry in darkness the target was fastened on to an all-silver mounting by means of small silver bolts. In this way contamination may be kept to a minimum. We tested for the prevalent contamination by carbon from the pump oil by bombarding a target for about ten minutes and then looking for  $\text{N}^{13}$  due to  $\text{C}^{12}(dn)\text{N}^{13}$ . We found no activity except the 37-minute activity of  $\text{Cl}^{38}$ , which means that no proton groups could be caused by carbon contamination. We used thicker targets to study the longer range, low yield groups and targets of about one centimeter air equivalent (about 0.3 Mev) for the more prolific shorter range groups.

#### EXPERIMENTAL RESULTS

Figure 1 shows the arrangements for bombarding. Absorption curves from targets of

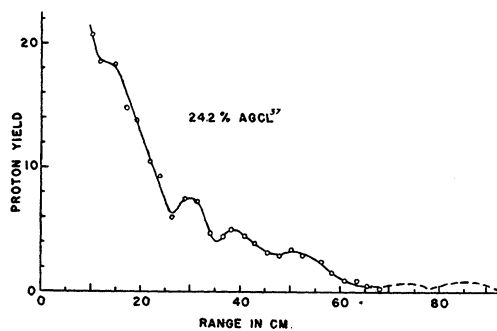


FIG. 2. Absorption curve for protons from a normal target of silver chloride. Six groups appear, with a seventh at very small absorption which is ascribed to alpha-particles.

normal and heavy chlorine (24.2 percent and 53 percent  $\text{Cl}^{37}$  bombarded by deuterons) were plotted and are shown in Figs. 2, 3 and 4. It is seen from these curves that there are at least six definite proton groups having ranges of 27, 33, 44, 54, 77, and 93 cm (uncorrected for variation of range in Al with velocity) with a possible error of  $\pm 2$  cm. If we call the group of 27 cm range group *A*, that of 33 cm range group *B*, and so on, it can be seen that the relative yield of the three groups *B*, *C*, and *D* to each other has remained very nearly the same in going from normal chlorine to heavy chlorine. This fact is evidence that all three of these groups arise from the same isotope of chlorine. At the same time the yield of group *A* has decreased relative to groups *B*, *C*, and *D* in Fig. 3 as compared to Fig. 2. Further the apparent range of the group has decreased as would be expected if the yield of group *B* had been built up relative to *A* thus changing the shape of the composite curve. This establishes that group *A* is due to  $\text{Cl}^{35}$ , while groups *B*, *C*,

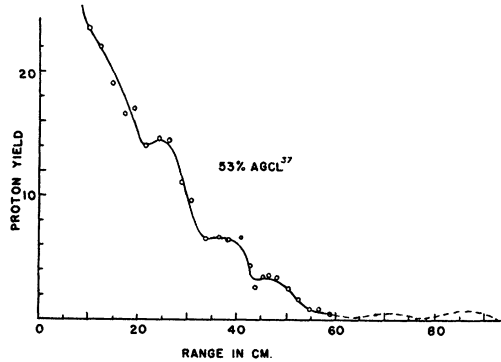


FIG. 3. Absorption curve for protons from a target of silver chloride enriched in the heavy isotope. The six groups are present, but the abundance of the three groups ending at 33, 44 and 54 cm is increased relative to the groups at 27, 77, and 93 cm. These three groups are therefore caused by the bombardment of  $\text{Cl}^{37}$ .

and *D* must be assigned to  $\text{Cl}^{37}$ . The identification of the long range groups *E* and *F* was made by comparing them with the yield of group *D*. Using the yield at 50 cm as a standard it was found that the yield of both the groups, while remaining the same relative to each other, changed by a factor of approximately three in going from one target to the other, the normal sample giving the larger yield. This is the ratio one would expect using the isotope concentra-

tions given. This fact again points to the identification of group *D* as due to  $\text{Cl}^{37}$  while at the same time it establishes the fact that the two long range groups *E* and *F* are caused by  $\text{Cl}^{35}$ . The yield of these two long range groups was so small that the background caused by neutron recoils rendered us a little unhappy about our measurement of their ranges although we made something like ten runs on this part of the curve alone. It was therefore decided to use the

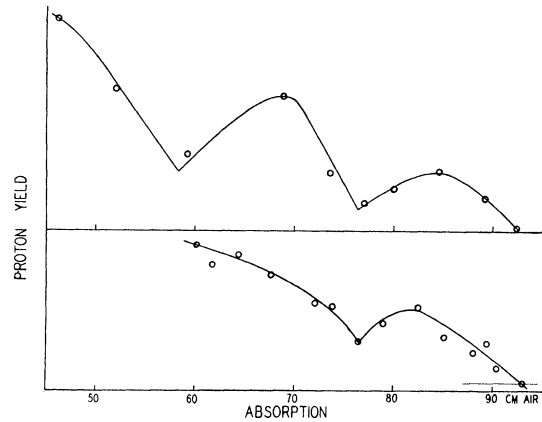


FIG. 4. Absorption curve showing the detail of the long groups. The upper curve was taken with single counting while the lower was taken by counting double coincidences, which reduces the random background caused by neutron recoils. Three groups are seen, of which the two of longest range are caused by  $\text{Cl}^{35}$ .

method of coincidence counting to study this part of the curve<sup>4</sup> with the results shown in Fig. 4 where the upper curve is the average of runs taken by direct counting and the lower is the curve found by coincidence counting. In the second case the yield of coincidences when a thick "infinity" absorber was interposed was about two per minute (these are probably all caused by spurious pulses originating in electrical disturbances) while the yield of coincidences from the chlorine target at 82 cm range was thirty per minute. It can be seen that the coincidence method confirms the previous findings.

There is evidence that the group at 12 cm range is an alpha-particle group caused by the reaction  $\text{Cl}^{35}(d\alpha)\text{S}^{33}$ . Its observation is greatly complicated by the very large yield of protons and the fact that the scattered beam has a range only a little less. The fact that it appears

<sup>4</sup> E. Pollard and R. F. Humphreys, Bull. Am. Phys. Soc. 15, 7 (1940), Abstract No. 4.

most prominently on the light target is evidence in favor of this assignment.

The groups found here by no means exhaust the number which may actually be present, but rather represent the most favorable transitions in the nuclei of  $\text{Cl}^{36}$  and  $\text{Cl}^{38}$ .

#### DISCUSSION

In Table I are given the energy change values for the reactions  $\text{Cl}^{35}(dp)\text{Cl}^{36}$ ,  $\text{Cl}^{37}(dp)\text{Cl}^{38}$ , and  $\text{Cl}^{35}(d\alpha)\text{S}^{33}$ .

The limiting energy change value for the three reactions can be used to derive masses if the masses of the target elements are known. These are not very satisfactory. If a chain of nuclear reactions is employed the best values obtainable for the two stable chlorine isotopes are 34.98107 and 36.97829,<sup>5</sup> while mass-spectroscopic values are 34.97903 and 36.97786.<sup>6</sup> The transmutation values are derived from a series of good determinations of energy change values which agree with the mass-spectroscopic value for  $\text{P}^{31}$ ,<sup>7</sup> but not with the value for  $\text{S}^{32}$ .<sup>7</sup> The mass-spectroscopic value for  $\text{Cl}^{35}$  given above does not agree with the  $Q$  value for the  $\text{S}^{32}(\alpha p)\text{Cl}^{35}$  reaction. There is need for much further work in this region of the elements both by transmutation and the mass spectrometer. If we take the transmutation values, the derived masses for  $\text{Cl}^{36}$  and  $\text{Cl}^{38}$  are 35.9808 and 37.9806, with errors as far as the energy changes in this work are concerned of 0.0003.

A check on the consistency of the energy measurements can be made by using the fact

TABLE I. Energy change values.

PROTON ENERGY (MEV)	Q VALUE (MEV)
<i>Reaction <math>\text{Cl}^{36}(dp)\text{Cl}^{36}</math></i>	
9.10	6.31
8.15	5.35
4.40	1.50
<i>Reaction <math>\text{Cl}^{37}(dp)\text{Cl}^{38}</math></i>	
6.85	4.02
5.90	3.02
5.00	2.10
<i>Reaction <math>\text{Cl}^{35}(d\alpha)\text{S}^{33}</math></i>	
	9.1

<sup>5</sup> E. Pollard, Phys. Rev. **57**, 1186 (1940).

<sup>6</sup> T. Okuda, K. Ogata, K. Aoki and Y. Sugawara, Phys. Rev. **58**, 578 (1940).

<sup>7</sup> F. W. Aston, Nature **138**, 1094 (1936).

that the energy of the beta-ray emitted by  $\text{Cl}^{38}$  is 4.8 Mev.<sup>8</sup> This gives us the mass of  $\text{A}^{38}$ , which is linked with that of  $\text{Cl}^{35}$  by the reaction  $\text{Cl}^{35}(\alpha p)\text{A}^{38}$ .<sup>9</sup> The mass of  $\text{A}^{38}$  which we derive from the work here described is 37.9754, while if the data of Pollard and Brasefield are corrected for the air equivalent of aluminum foils according to Livingston and Bethe<sup>10</sup> the value derived from the alpha-particle bombardment of chlorine is also 37.9754. With the two mass-spectroscopic values given above the values for  $\text{A}^{38}$  by the two approaches are 37.9734 and 37.9740. Both figures agree within the experimental errors given, but the agreement is more satisfactory for the masses derived from transmutation.

The fact that  $\text{Cl}^{36}$  has a maximum beta-ray energy of 0.7 Mev<sup>11</sup> enables the mass of  $\text{A}^{36}$  to be calculated, giving 35.9800. Since  $\text{Cl}^{36}$  can also emit a positron the mass of  $\text{S}^{36}$  must also be very close to this value.

The group at 12 cm range, if correctly assigned, leads to the value 32.9828 for the mass of  $\text{S}^{33}$ , which is in fairly good agreement with the value 32.9826 derived from the  $\text{S}^{32}(dp)\text{S}^{33}$  reaction.<sup>5</sup>

The energy levels obtained for the product nucleus  $\text{Cl}^{38}$  are consecutive and so can be discussed. It will be noticed that their spacing is roughly one Mev. This can be compared with the spacing of 2 Mev found in the stable nucleus of  $\text{A}^{38}$  in the reaction  $\text{Cl}^{35}(\alpha p)\text{A}^{38}$ . There may be some significance in the fact that  $\text{Cl}^{38}$  is unstable while  $\text{A}^{38}$  is stable, but we prefer to think that the manner of production of the final nucleus is responsible for exciting different energy levels and that the spacing observed depends more on selection rules than on the nature of the nucleus itself.

In conclusion we wish to thank Professor W. W. Watson for discussion and help with the isotope separation, and Mr. Harry Schultz for assistance in running the cyclotron. We are also grateful for a grant from the George Sheffield Fund which made possible the construction of the cyclotron.

<sup>8</sup> F. N. D. Kurie, J. R. Richardson and H. C. Paxton, reference 1.

<sup>9</sup> E. Pollard and C. J. Brasefield, Phys. Rev. **50**, 890 (1936).

<sup>10</sup> M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 272 (1937).

<sup>11</sup> D. C. Grahame and H. J. Walke, reference 1.