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November 19, 1952

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ABSTRACT

Making use of the predictions of "alpha systematics" on the extent of alpha branching of the isotope AcK and of the properties of the resultant At^{219} daughter, the branching was looked for and found in the francium fraction isolated from a 20 mc source of Ac^{227} . The observed branching ratio is ca 4×10^{-5} , and the At^{219} daughter decays predominately by the emission of 6.27 Mev alpha particles with a half-life of 0.9 minutes to the new isotope Bi^{215} , which in turn emits β^- particles with a half-life of 8 minutes. At^{219} also decays by β^- emission to An^{219} about once in 30 disintegrations. It is pointed out that this is the first reported instance of a chemical identification of an isotope of element 85 from a natural source, although other shorter-lived astatine isotopes are known to exist.

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I. INTRODUCTION

The only isotope of element 87 occurring in nature is the 21 minute beta emitting isotope, AcK , which arises from a 1.2 percent alpha branching of Ac^{227} and was discovered by M. Perey in 1939.¹ An extensive series of researches have been carried out by Mlle. Perey and her collaborators¹⁻⁹ to study the chemical properties of francium and to determine the radiation characteristics of AcK .

Recently, considerable effort in this laboratory¹⁰, as well as in many laboratories throughout the world, has been devoted to the

¹M. Perey, J. Phys. et Radium 10, 435 (1939).

²M. Perey, J. Chim. Phys. 43, 155, 269 (1946).

³M. Perey, Theses, University de Paris (1946).

⁴M. Perey, J. Phys. et Radium [8] 8, 179 (1947).

⁵M. Perey and M. Lecoïn, J. Phys. et Radium 10, 439 (1939).

⁶M. Perey and M. Lecoïn, Compt. Rend. 212, 893 (1941).

⁷M. Guillot and M. Perey, Compt. Rend. 225, 330 (1947).

⁸Lecoïn, Perey, Riou, and Teillac, J. Phys. et Radium 11, 227 (1950).

⁹Lecoïn, Perey, and Tsien, Cahiers Phys. 26, 10 (1944).

¹⁰Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).

correlation of the wealth of new data collected on the decay characteristics of the heavy element nuclides and to the development of an alpha decay systematics which would facilitate the prediction of the alpha decay characteristics of those isotopes for which this information has not yet been obtained.

It was of interest to apply these methods to the prediction of the alpha decay characteristics of Fr^{223} and its unknown daughter, At^{219} , to establish the feasibility of a search for this decay transition.

It was estimated that the alpha particle energy of Fr^{223} must lie in the range 5.2-5.4 Mev and its half-life in the range of several months to tens of years. The daughter isotope, At^{219} , must have an alpha particle energy of 6.1 to 6.4 Mev and an alpha decay half-life of 15 seconds to 15 minutes with the best estimate of the order of one minute. The alpha decay half-lives in both cases are uncertain because of the limits on the alpha particle energy and because of the general phenomenon of hindrance in the alpha decay of odd nucleon nuclei, the nature and magnitude of which is only crudely understood. Since the beta decay half-life of Fr^{223} is only 21 minutes, the predicated alpha branching ratio was quite low, 10^{-4} to 5×10^{-7} . The energy available to At^{219} for beta decay to Em^{219} is approximately 1.5 Mev from which one can estimate a beta decay half-life of the order of minutes. Hence, it was not certain whether alpha or beta decay would be more prominent for this nuclide. Alpha decay of At^{219} produces the unknown nuclide Bi^{215} which is certain to decay predominantly by beta emission. The beta decay half-life should fall in the range of a few seconds to a few minutes.

II. EXPERIMENTAL

These preliminary considerations and in particular the low value of the branching ratio of AcK pointed up the difficulties in the search for this alpha branching but suggested that the experiment was feasible given a sufficiently active source of Ac²²⁷. We had available to us a 20 mc source and using this as a starting material, the experiments discussed below were carried out with the results summarized in Figure 1. Certain data of the 4n + 3 decay series pertinent to this investigation are included in this figure in addition to those facts reported here for the first time.

Starting with the 20 mc source of Ac²²⁷, our procedure was to isolate and purify the AcK daughter, to isolate an astatine fraction from this francium fraction by chemical or physical methods, and to examine it for the expected radioactivity. This examination was done in our alpha ray differential pulse analyzer, an instrument with 48 electronic channels to analyze the pulses developed in an ionization chamber by the alpha particles emitted by the sample. The instrument used is a modification of the original design of Ghiorso et al.¹¹

The francium was isolated by the improved method developed by Hyde and described in detail elsewhere.¹² Briefly the method consists of the precipitation of free silicotungstic acid from a solution of Ac²²⁷ in saturated hydrochloric acid followed by the separation of the francium from the carrier on a cation exchange resin (Dowex 50). By

¹¹Ghiorso, Jaffey, Robinson, and Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, The Transuranium Elements: Research Papers, Paper No. 16.8 (McGraw-Hill Book Company, Inc., New York, 1949).

¹²E. K. Hyde, J. Am. Chem. Soc. 74, 4181 (1952).

this method we could isolate francium carrier-free and radioactively pure in 40 minutes or less. The disintegration rates of the samples at this time were $1-2 \times 10^8$ disintegrations per minute. The Ac^{227} solution was set aside until a new francium sample was required, at which time this simple silicotungstic acid separation procedure was repeated.

In the first successful experiment astatine was extracted from a 3 M hydrochloric acid solution of the AcK into an equal volume of a 10 percent solution of tributyl phosphate in butyl ether. Preliminary test runs with appropriate tracers had established that extraction of astatine is nearly complete and extraction of radium lead and bismuth is very slight under these conditions. This later consideration is quite important since a few disintegrations per minute of At^{219} activity were to be extracted from a solution containing 10^4 to 10^5 disintegrations per minute of Ra^{223} , Pb^{211} and Bi^{211} . After a quick washing of the tributyl phosphate with 3 M hydrochloric acid, the organic layer was evaporated to dryness on a platinum plate and examined in the alpha pulse analyzer. The observed alpha spectrum is shown in Figure 2.

No At^{219} alpha peak was observed because several minutes had elapsed from the time of isolation of the astatine until the above data were taken. An alpha peak corresponding to Po^{215} was observed. This Po^{215} was not formed by the decay of An^{219} as shown by the absence of the An^{219} peak. The area under the Po^{215} peak decayed with a half-life of 8 minutes as shown in Figure 3. An examination of Figure 1 will show that this can be so only if the Po^{215} is being maintained by the beta decay of an 8 minute Bi^{215} . The bismuth isotope in turn must be the product of the decay of At^{219} . The

greater part of the activity in the Bi^{211} peaks is present because of a slight extraction of Pb^{211} , but some of it represents $\text{Pb}^{211} - \text{Bi}^{211}$ formed from the decay of the Bi^{215} prior to the pulse analyzer measurements.

To speed up the separation so that the alpha group of At^{219} could be observed, a volatilization separation requiring only a few seconds was used. The purified carrier-free francium fraction was evaporated on a platinum filament. A platinum disc was mounted 1-2 mm above this filament and an electric current was passed through the filament to warm it and volatilize the astatine to the collector disc. Since francium, lead and bismuth will also volatilize if the temperature is sufficiently high, a number of test runs were made to determine at what current settings for our particular apparatus volatilization of astatine would occur without concomitant volatilization of the others. Using this method, astatine was collected on the collector disc during a 15 second milking period and then quickly inserted in the differential pulse analyzer. The first analysis of the alpha spectrum began within 1/2 minute of the end of the milking period and was continued for one minute. The data obtained are shown in Figure 4. The analysis was repeated seven times during the first 12 minutes so that the behavior of the alpha groups with time could be checked. A new alpha group was observed at an energy identical with the 16 percent alpha particle group of Bi^{211} , i.e., at 6.27 Mev. This group decayed with a half-life of 0.9 minutes and almost certainly is the alpha particle of At^{219} . The Po^{215} peak was observed to grow attaining a maximum at 4-6 minutes and then to decay. This is an important point since this behavior should be observed if the following chain is correct:

Quantitative agreement of the observed growth and decay of the Po^{215} alpha group with a theoretical curve constructed for the chain as given was not obtained because of statistical error of the limited number of events recorded.

The upper curve of Figure 4 is a cumulative curve integrating the data collected from the 7 one-minute runs on the astatine fraction. One important piece of data obtained from this curve is the alpha/beta branching ratio of At^{219} . The An^{219} peak arises from the beta decay of At^{219} since there is no Ra^{223} in the sample. The $\text{At}^{219}/\text{An}^{219}$ peak ratio gives the alpha/beta branching ratio directly. This ratio is approximately 30 from this and other runs giving an At^{219} beta half-life of roughly 1/2 hour.

The astatine collection was repeated using the same francium source and identical collection conditions. Pulse analyzer data were repeated as in the case of the first sample with corroboration of all observations.

This entire procedure was repeated seven times. The amount of At^{219} found in each milking was plotted on semi-log paper as a function of the time of volatilization and shown to decay with the half-life of AcK within the experimental error (± 3 minutes).

These experiments were repeated several times with fresh samples of AcK isolated from the 20 mc Ac^{227} source. Half-life determinations on At^{219} ranged from 0.8-1.0 minute, the best value being 0.9 minute. A typical decay curve is shown in Figure 5. The lower limit of the AcK alpha branching was calculated from the amount of At^{219} actually

observed in the pulse analyzer runs making appropriate corrections for pulse analyzer geometry, decay of the AcK source to time of volatilization, etc. The amount of AcK used as a source was determined by counting the beta activity of a measured aliquot of the sample and cross-checked by counting the alpha activity of the Ra²²³ and its daughters several hours after preparation of the sample. Analysis of the data from several runs set $3-5 \times 10^{-5}$ as the lower limit for the AcK alpha branching.

An attempt was made to get a more accurate value of this ratio by isolating Bi²¹⁵ chemically from a measured quantity of AcK making the initial separation after a timed growth period and using RaE tracer to determine the chemical yield. Several procedures were developed to isolate bismuth quickly in a carrier-free state sufficiently decontaminated from AcK, Ra²²³, and Pb²¹¹. The shortest procedure took about 25 minutes. No details of these methods are given here since the results were negative, but from our experiments we can set an upper limit of 10^{-4} to the branching ratio. The assistance of Mr. Bruce Raby in this series of experiments is gratefully acknowledged.

This low branching ratio makes it impossible to detect the AcK alpha particle by our present instrumental techniques because of the rapid growth of interfering Ra²²³ daughter activity even in freshly purified samples.

III. DISCUSSION

This research is of interest not only because of its demonstration of the usefulness of "alpha systematics" in planning experiments but also because of its bearing on the question of the existence of astatine in nature.

The Viennese workers Karlik and Bernert¹³⁻¹⁶ have carried out an extensive series of experiments in which previously unreported alpha particle groups were observed in samples isolated from the naturally occurring radioactive series and ascribed to astatine isotopes produced by the rare beta branching of the A-products in the chains. The very short-lived isotope At^{215} , arising from a 5×10^{-4} percent beta branching of AcA , was postulated to explain the observation^{15,16} of 8.4 Mev alpha particles in a sample of actinon. The isotope At^{216} , with a half-life short compared to the 54 second half-life of thoron and arising from a 1.35×10^{-2} percent beta branching of the A, was postulated to explain the observation^{14,16} of 7.64 Mev alpha particles in a fresh sample of thoron. In the radium series, alpha particles of 6.72 Mev energy, with a half-life of a few seconds, were observed^{13,16} to grow into a freshly deposited sample of RaA and these were ascribed to At^{218} , resulting from a beta branching of 5×10^{-2} percent in RaA .

The not inconsiderable experimental difficulties in these investigations were overcome with ingenuity by Karlik and Bernert, but unfortunately the interpretations in the first two cases are open to some question from energetic considerations as recognized by the authors¹⁶ themselves and as discussed by Feather¹⁷ and by Perlman

¹³B. Karlik and T. Bernert, Naturwiss. 31, 298 (1943).

¹⁴B. Karlik and T. Bernert, Naturwiss. 31, 492 (1943).

¹⁵B. Karlik and T. Bernert, Naturwiss. 32, 44 (1944).

¹⁶B. Karlik and T. Bernert, Zeit f. Physik. 123, 51 (1944.).

¹⁷N. Feather, Nucleonics 5, 22 (1949). See also, British Atomic Energy report, BR 640 (1945).

et al.¹⁰ In the third case the conclusions followed more directly from the experimental data, and in particular the observation of the growth of the new activity strengthens the interpretation. The experiments in this case have been confirmed by Walen¹⁸, and since the properties ascribed to At^{218} do not conflict with "systematics," it seems likely that the interpretation of Karlik and Bernert is correct.

At^{218} may then be taken as the first isotope of astatine to be found in a natural source. At^{215} and At^{216} may also exist but confirmatory evidence would be desirable. Recently, Peppard et al.¹⁹ have reported the occurrence of minute amounts of the $4n + 1$ series in uranium and thorium ores. These experiments imply the existence of the 0.02 second At^{217} in these ores since it is a member of the $4n + 1$ decay chain. The results reported in the present paper add another isotope of astatine to this list and supply the first case in which the half-life of the astatine is sufficiently long that a chemical identification (as for example, by extraction into tributyl phosphate or volatilization at low temperature) of the activity was possible.

It is worthy of note that other authors have published predictions similar to those discussed above on the alpha branching of AcK and that these predictions are in substantial agreement with the ex-

¹⁸R. Walen, Compt. Rend. 227, 1090 (1948).

¹⁹Peppard, Mason, Gray, and Mech, U. S. Atomic Energy Commission
Declassified Document AECD-3378, April 10, 1952; submitted for
publication in J. Am. Chem. Soc.

perimental results reported here. Vigneron²⁰ predicted an alpha particle energy of 5.45 ± 0.15 Mev for AcK and an α/β^- branching ratio in the range of 1/300 to 1/2700. He also estimated an alpha energy of 6.20 ± 0.15 Mev for At²¹⁹ and an alpha half-life of 15 seconds to 15 minutes. Karlik²¹ predicted an alpha particle energy of 5.55 Mev and an α/β^- branching ratio of 2×10^{-3} for AcK. Feather²² predicted a branching ratio of a few per thousand. Jentschke²³ predicted an alpha branching ratio of 4×10^{-5} to 6×10^{-4} for AcK and indicated α/β^- branching at At²¹⁹ and beta emission at Bi²¹⁵.

ACKNOWLEDGMENTS

We wish to thank Dr. F. Hagemann of the Argonne National Laboratory for making the Ac²²⁷ source available to us and Dr. J. Hollander of this laboratory for helpful discussions on this problem.

²⁰L. Vigneron, Compt. Rend. 225, 1067 (1947).

²¹B. Karlik, Acta Phys. Austria 2, 182 (1948).

²²N. Feather, Reports Prog. Phys. 11, 19 (1948).

²³W. Jentschke, Phys. Rev. 77, 99 (1950).

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Fig. 1. The $4n+1$ series below actinium including new data on the alpha branch products of AcK.

Fig. 2. Alpha spectrum of radioactivity extracted into tributyl phosphate from hydrochloric acid solution of AcK. Upper curve represents calibration run with Ra²²³ standard.

Fig. 3. Determination of half-life of Bi^{215}
from decay of Po^{215} peak in tributyl phosphate fraction. |

MU-4484

Fig. 4. Alpha spectrum of astatine fraction separated from AcK by volatilization. Lower curve represents first one-minute run. Upper curve shows cumulative data on first 7 one-minute runs. Discriminator settings on pulse analyzer instrument not identical with those for figure 2.

Fig. 5. Determination of half-life of At²¹⁹ from decay of 6.27 Mev alpha particle peak.