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REPORT

FOR

GENERAL RESEARCH

JUNE 26 TO SEPTEMBER 18, 1950

(Actinium Volume)

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(Actinium Volume)

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Date: October 23, 1950

Approved By: [Signature]

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Problem Title - Preparation of Actinium-228

Report By - J. B. Otto

Work Done By - J. B. Otto

INTRODUCTION

In studying the tracer chemistry of actinium, one is faced with the problem of obtaining an isotope of this element which can be easily counted and is at the same time fairly readily available. Actinium-228, which has a beta energy of 1.55 million electron volts and a half life of 6.13 hours, meets these requirements but its half life is so short that it can only be used for short-term experiments. Despite this disadvantage, it is presently the most desirable isotope with which to study the tracer chemistry of actinium. Actinium-228 has been employed for this purpose by McLane and Peterson.¹ These authors² describe a method for separating actinium-228 from its parent radium-228 with a procedure originally worked out by Haissinsky.³ McLane and Peterson's modification of the method allows one to prepare either radioactively pure or carrier free actinium-228. The present investigation involves the working out of a satisfactory procedure for obtaining actinium-228 by McLane and Peterson's method or a modification thereof.

DETAILED REPORT

The Haissinsky-McLane and Peterson method of separating actinium-228-lanthanum from radium-228-barium mixture depends on the solubility of the nitrates of the former pair of elements in absolute ethanol. Radium-228 is

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the daughter of thorium-232, the major isotope of thorium in commercial thorium salts. The relationship of radium-228 and actinium-228 to thorium-232 and the remainder of its decay products is seen below:^{4,5,6}

Aged thorium-232 serves as a source of radium-228 from which actinium-228 can be separated by the Haissinsky-McLane and Peterson method. Rough calculations show that one gram of thorium produces about 4.7×10^{-11} gram of radium-228 in one year which is about 10 per cent of the equilibrium value. This quantity of radium-228 gives about 23,600 beta disintegrations per minute. Radium-228 and actinium-228 are nearly in equilibrium in one day.

Radium-228

The radium isotopes were precipitated from samples of thorium nitrate thought to be at least six years old with barium as a carrier. The barium was precipitated either as the chloride or nitrate. Nothing could be found in the literature concerning procedures for separation of radium from thorium.

Chloride Procedure

1. One gram of the thorium nitrate was weighed out and dissolved in about 1.5 milliliters of water in a 10-milliliter centrifuge tube. (The per cent thorium in this material was not known because it was associated with an unknown amount of water of hydration. Therefore it is proposed to analyze a sample of this thorium nitrate for the per cent thorium at a later date.)
2. Ten milligrams of barium carrier was added as the chloride in 50 lambda of solution. A small amount of precipitate was formed at this point; probably barium nitrate.
3. Barium chloride was precipitated in the cold with seven milliliters of a 6:1 by volume solution of concentrated hydrochloric acid and ether in the manner described by Ames et al.⁷ The tube was centrifuged, and the supernatant liquid containing the thorium and nearly all its daughter activity except radium-224 and radium-228 was discarded. The barium chloride precipitate was dissolved in about one milliliter of water and reprecipitated twice in the same manner.

4. The last barium chloride precipitate was dissolved in about five milliliters of water, and barium sulfate was precipitated by the addition of about 10 drops of one normal sulfuric acid. This precipitate was washed twice with water and transferred to a small Bradley slide for counting. Only one transfer of the slurried barium sulfate could be made to the slide because of the limited capacity of the slide, so only an estimated 80 to 90 per cent was transferred and counted.

Nitrate Procedure

1. and 2. Same as chloride procedure (see above).

3. Barium nitrate was precipitated by the addition of seven milliliters of cold concentrated nitric acid. The precipitate was white and appeared to be dense and granular. The tube was centrifuged, and the supernatant liquid containing the thorium was discarded. The barium nitrate was dissolved in about one milliliter of water and reprecipitated twice as the nitrate in the manner described above.

4. Same as chloride procedure (see above).

The radium-barium Bradley slides prepared by both the chloride and nitrate procedures described above were allowed to decay 22 days before counting. This allowed time for nearly all the radium-224 initially separated and its daughters to die out leaving what should be only radium-228 and actinium-228 activity in equilibrium. The daughter of actinium-228, thorium-228, is comparatively inactive after only 22 days of growth because of its relatively long half life. Counting data from these slides is given in Table I.

What fraction of the 20,000 beta counts is due to radium-228 has not been determined as yet. This fraction may be small since radium-228 has a very weak beta.

Actinium-228

Samples of barium nitrate containing radium-228 activity were prepared by both the chloride and nitrate procedures as described above. The barium chloride obtained by the chloride procedure was converted to the nitrate because this form is required for the Haissinsky-McLane and Peterson method.

Actinium-228 was separated from the above samples of barium-radium-228 nitrates by the procedure described by McLane and Peterson,² an outline of which is shown in Figure 1. The separated lanthanum-actinium-228 was prepared for counting in the following manner. The lanthanum-actinium-228 oxide was dissolved in one normal nitric acid, transferred to a one-inch watch glass, and the nitrate solution was evaporated to dryness. The resulting lanthanum-actinium nitrate was too hygroscopic to be suitable for counting, so it was converted to the oxalate by adding 10 per cent excess oxalic acid to a solution of the nitrate on the watch glass. The solution was evaporated, and the resulting dry residue of lanthanum-actinium-228 oxalate was counted in a Bradley counter. The foregoing procedure was carried out on a sample of barium-radium-228-actinium-228 separated from thorium by chloride procedure. The lead-bismuth sulfide scavenger precipitation step was omitted in this particular experiment. The results of this separation are given in Table II.

This result compares favorably with the results shown in Table I for samples of radium-228 and actinium-228 in equilibrium. In fact, the 14,000 beta counts obtained corrected for decay to separation time is slightly higher than the 20,000 beta counts given in Table I, but it should be recalled that only an estimated 80 to 90 per cent of the radium-radium sulfate could be transferred to the slides used for the data in Table I. The half life of the separated material compares favorably with that of actinium-228.

It should be pointed out that the entire separation should be carried out as soon as possible after the completion of the first step, i.e., the solution and evaporation to dryness of the barium, radium-228, actinium-228, thorium, and lanthanum nitrate mixture. If one or two days elapse between the times this step and remainder of the procedure are carried out, a very small fraction of the actinium-228 will be extracted. This phenomenon indicates the separation of radium-228 and actinium-228 must take place when the barium-radium-228-lanthanum-actinium-228 solution is evaporated to dryness and crystallized. If the lanthanum nitrate is extracted one or two days after crystallization, the actinium-228 associated with it at the time of crystallization would have nearly all decayed away. This point is illustrated by an extraction carried out on two barium-radium-228-actinium-228 samples separated by the chloride method. In this experiment, two days elapsed between the times the first step and the remainder of the procedure was carried out. The results of this experiment are given in Table III.

Comparison of the results shown in this table with those in Tables I and II show what a small fraction of the actinium-228 present was separated. The half life of the activity compares well with that of actinium-228, 6.13 hours.

Conclusions

Procedures have been worked out for separating radium activity from a sample of aged thorium by coprecipitation with inactive barium. The chloride and nitrate procedures were found to be equally effective. It is not clear as to what fraction of the 20,000 beta counts given in Table I is due to radium-228 but quite probably it will prove to be small because of the low energy of the beta particles from this isotope. Work is in progress on this point.

Actinium-228 has been separated in good yield by the procedure described by McLane and Peterson from samples of radium-228 which had been separated from a sample of aged thorium nitrate.

REFERENCES

1. McLane, C. K. and Peterson, S., National Nuclear Energy Series IV-14B, p. 1371 (1949).
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6. Seaborg, G. T. and Perlman, I., Table of Isotopes, Reprinted from Rev. Mod. Phys. 20, 585 (1948).

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Table I

DATA OBTAINED BY COUNTING SLIDES OF RADIUM ISOTOPES 22 DAYS AFTER SEPARATION FROM A THORIUM SOLUTION BY COPRECIPITATION WITH BARIUM

<u>Sample Number</u>	<u>Counts Per Minute at 50 Per Cent Geometry</u>		
	<u>Alpha and Beta</u>	<u>Alpha</u>	<u>Beta</u>
Chloride Procedure			
1	22,982	816	22,166
2	22,113	830	21,283
Nitrate Procedure			
1	20,881	809	20,072
2	20,978	794	20,184

Table II

THE SEPARATION OF ACTINIUM-228 FROM RADIUM-228 BY THE HAISSINSKY - McLANE AND PETERSON METHOD

Step 1 finished: 10:00 A.M. 9-13-50

Separation time: 10:30 A.M. 9-13-50

<u>Sample Number</u>	<u>Time Counted</u>	<u>Counts Per Minute at 50 Per Cent Geometry</u>		
		<u>Alpha and Beta</u>	<u>Alpha</u>	<u>Beta</u>
1	3:00 P.M. 9-13-50	14,047	14	14,033
1	11:30 A.M. 9-14-50	1,441	23	1,418

Table III

THE SEPARATION OF ACTINIUM-228 FROM RADIUM-228
BY THE HAISSINSKY - McLANE AND PETERSON METHOD

Two Days Elapsed Between the Time the First Step
and the Remainder of the Procedure was Carried Out

Step 1 finished: 8-28-50

Separation time: 10:00 A.M. 8-30-50

<u>Sample Number</u>	<u>Time Counted</u>	<u>Counts Per Minute at 50 Per Cent Geometry</u>		
		<u>Alpha and Beta</u>	<u>Alpha</u>	<u>Beta</u>
1	3:30 P.M. 8-30-50	84	2	82
2	3:30 P.M. 8-30-50	80	0	80
1	2:08 P.M. 8-31-50	6	1	5
2	1:44 P.M. 8-31-50	2	0	2

FIGURE 1

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OUTLINE OF THE PROCEDURE USED FOR
THE SEPARATION OF ACTINIUM 228 FROM
RADIUM 228

Problem Title - The Separation of Actinium from Radium
Report By - P. M. Engle
Work Done By - W. T. Brown and P. M. Engle

INTRODUCTION

Design work on all phases of the "cave" installation to be built for the actinium project has continued. Some equipment construction has begun.

DETAILED REPORT

The design of the "cave" structure is now practically complete. Specifications for the various types of construction are being written. Progress is also being made on the layout of the interior partitions, services, and other features required in the section of the Semi-Works Building to be used by this program. The above design work is being accomplished in cooperation with the Engineering Division.

The length of the "cave" to be built has been increased approximately 50 per cent to a total of 29 feet. This additional length will provide space for a second unit of processing apparatus which will be of a development nature.

A surface protective system for the interior of the "cave" that will permit decontamination by stripping one or more layers and will also prevent corrosion and contamination of the basic structure is needed. A series of combinations of various paint and tape samples are being tested for this purpose.

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Work has begun on the construction of various items of processing apparatus. A quantity of glassware has been completed and some of the remote controlled mechanical devices have been started. As this apparatus is finished, it will be assembled and tested as far as possible with inactive solutions.

A detailed preliminary cost estimate for the construction and materials required by this project was prepared at the request of the Atomic Energy Commission. This estimate in its final form did not, however, include the cost of the development equipment mentioned above or the cost of certain items of a semioperational nature such as shielded carriers, crucibles, etc.

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