DECLASSIFIED

[redacted]

AEC RESEARCH AND DEVELOPMENT REPORT

RECORD CENTER FILE

HW-25205
TECHNOLOGY - HANFORD PROCESSES

COPY NO. ~~16~~

[redacted]

BEST AVAILABLE COPY

THE DETERMINATION OF AMERICIUM IN PLUTONIUM PRODUCT SOLUTIONS

BY
A. CHETHAM-STRODE, JR.
TECHNICAL SECTION
ENGINEERING DEPARTMENT

AUGUST 15, 1952

THIS DOCUMENT IS
PUBLICLY AVAILABLE

~~RESTRICTED DATA~~

~~THIS [illegible] CONTAINS RESTRICTED DATA AS DEFINED IN THE [illegible] ENERGY ACT OF 1946. ITS TRANSMITTAL OR THE [illegible] OF ITS CONTENTS IN ANY MANNER TO [illegible] UTHORIZED PERSON IS PROHIBITED.~~

RECEIVED
JUN 20 1956
300 AREA
CLASSIFIED FILES

NUCLEONICS DIVISION
RICHLAND, WASHINGTON

GENERAL ELECTRIC

[redacted]

DECLASSIFIED

DECLASSIFIED

[redacted]

HW-25205 Copy-1

Technology - Hanford Processes

This document consists
of 6 pages. Copy No. ~~16~~
of ~~18~~ copies. Series ~~A~~

THE DETERMINATION OF AMERICIUM IN PLUTONIUM PRODUCT SOLUTIONS

By

A. Chetham-Strode, Jr.
Analytical Research
Applied Research Unit

August 15, 1952

HANFORD WORKS
RICHLAND, WASHINGTON

Classification Change to DECLASSIFIED
)
By Authority of 3-31-56
TID-1113
By
au Townsend 4/8/99
PD Aman 4-20-99

Operated for the Atomic Energy Commission by the
General Electric Company under Contract # W-31-109-Eng-52

[redacted]

This document [redacted] as defined in the Atomic Energy Act of [redacted] the disclosure of its contents in any [redacted] an unauthorized p[redacted]rohibited.

Route To	Read By	Date	Route To	Read By	Date
Yellow Copy RECORD CENTER FILE					

[redacted]

DECLASSIFIED

DECLASSIFIED

[redacted]

Technology - Hanford Processes

COPY NUMBER	INTERNAL DISTRIBUTION
1	O. H. Greager - W. K. Woods
2	F. W. Albaugh
3	W. J. Ozeroff
4	A. H. Bushey - H. R. Schmidt
5	John Hall
6	O. F. Hill
7	J. W. Healy
8	O. P. Amacker
9	L. M. Knights
10	Extra Copy
11	G. W. Watt, University of Texas
12	A. Chetham-Strode, Jr.
13	300 File
14	700 File
15	Pink Copy
16	Yellow Copy

	EXTERNAL DISTRIBUTION
17 - 19	Atomic Energy Commission, Washington
20	Chicago Patent Group
21 - 25	duPont Company
26	Hanford Operations Office
27 - 30	Knolls Atomic Power Laboratory
31	Patent Branch, Washington
32	Savannah River Operations Office (Augusta)
33	Savannah River Operations Office (Wilmington)
34 - 48	Technical Information Service, Oak Ridge

[redacted]

DECLASSIFIED

THE DETERMINATION OF AMERICIUM IN PLUTONIUM PRODUCT SOLUTIONS

INTRODUCTION

The routine method for the separation of americium and curium from plutonium and uranium is that due to H. W. Miller(1). In this scheme the plutonium is oxidized to the fluoride-soluble hexavalent state, and the combined americium and curium is carried on cerium fluoride. Miller has shown(1), however, that a small fraction of the plutonium present in solution is carried on the fluoride precipitates, presumably by mechanisms of adsorption and occlusion, and has estimated the extent of this carrying to be $\simeq$0.02% of the total plutonium present per 5 mg. of Ce^{+4} under the conditions of the analysis. This value has been confirmed by M. M. Jones(2) and A. Chetham-Strode, Jr.(3). It is readily seen that in concentrated plutonium solutions containing only small amounts of americium, coprecipitated plutonium can easily obscure the relatively small americium activity and lead to very large errors in the analysis.

SUMMARY

The cerium fluoride method for the determination of americium in the presence of plutonium and uranium has been extended with modifications to application to plant streams of very high plutonium to americium ratios. The precision of the modified method has been determined to be ± 2.24% at the 99% limits.

DISCUSSION

Some experiments have been made to determine the error in Miller's method resulting from coprecipitated plutonium. Table I indicates the magnitude of this error. The data are taken from the americium recovery run of December, 1951(3). The amount of plutonium expected to coprecipitate on cerium fluoride has been calculated(1, 2, 3). Alpha energy analysis

[redacted]

of the actual sample discs showed them to be highly contamined with plutonium. The extent of this cross-contamination agreed with the estimated values as to order of magnitude.

TABLE I

THE CONTAMINATION OF SOME AMERICIUM DISCS WITH COPRECIPITATED PLUTONIUM

Plutonium Concentration of sample c/m/m.	% Total Activity Due to coprecipitated Plutonium - measured	% Total Activity Due to coprecipitated Plutonium - caluclated
1×10^7	5	2
2.9×10^9	70	90
2.7×10^{10}	100	100

A method originally used to separate plutonium from uranium in dissolver solution was applied to a similar problem involving the determination of americium in plutonium solutions (4). This procedure involves the dissolution of the first cerium fluoride precipitate with zirconium, following removal of the bulk of the supernate. The cerium fluoride is then reprecipitated from the solution of greatly reduced plutonium concentration. This method has now been successfully extended to product solutions with good precision and accuracy.

EXPERIMENTAL

Americium determinations have been made on nine different product samples drawn from routine process runs. Four determinations have been made on each sample. The procedure given at the end of this discussion was rigorously adhered to in every case. The precision of the method was found to be 2.24% at the 99% confidence level.

The percentage of americium with respect to alpha activity in these samples varies from 0.1% to 1%. This composition corresponds to that encountered in process streams in the course of an americium recovery run.

The accuracy of the analysis can not be determined directly due to the lack of suitable standards. The separation has been repeatedly checked with the alpha energy analyzer, however, and no indication of plutonium contamination of the americium fraction has been detected. The carrying of americium on cerium fluoride has been shown to be quantitative with and without the zirconium dissolution step$^{(4)}$.

PROCEDURE

An aliquot containing 1×10^6 total alpha d/m is pipeted into a 3 ml. centrifuge cone and diluted to 2 ml. with 2 N HNO_3. The cone is transferred to a water bath and heated for 15 min. at 85-95°C.

Four drops (250 ul.) of 0.1 M Ce^{+4} are added, and the sample is digested for 15 min. at 85-95°C, then cooled to room temperature.

Cerous fluoride is precipitated by the addition of 10 drops (500 ul.) of 27 N HF, which has been previously treated with $Cr_2O_7^=$ and stored in a platinum container. The precipitate is digested for 5 min., and centrifuged. The supernate is discarded.

One drop of 0.05 M $Cr_2O_7^=$, 3 drops of 0.1 M $ZrO(NO_3)_2$, and 2 ml. 2 N HNO_3 are added and the sample stirred until the cerous fluoride precipitate is completely dissolved. Cerous fluoride is reprecipitated as before by the addition of 10 drops of the dichromate-treated HF.

The precipitate is washed three times with a 1 N HNO_3-1 N HF solution, slurried with a few drops of 2 N HNO_3, and transferred to a platinum disc. The sample is dried, ignited, and counted for alpha activity.

A. Chetham-Strode, Jr.

AC-S:dt

[redacted]

[redacted]

REFERENCES

(1) Miller, H. W., "The Quantitative Separation of Americium from Plutonium and Uranium", HW-22267.

(2) Bushey, A. H., Quarterly Report, Oct. - Dec., 1951, HW-23508.

(3) Chetham-Strode, Jr., A., Unpublished data, Secret Notebook HW-4606-T.

(4) Bushey, A. H., loc. cit.

[redacted]