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HEAVY ISOTOPES PRODUCTION CAVE RUN**

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INTRODUCTION

A capsule, containing Cm and a mixture of Cm and Pu which was irradiated at the Materials Testing Reactor (MTR) for two years and finally at the Savannah River Reactor (SRR) for two months, was processed for transcurium isotopes by the Berkeley Heavy Element Production Group. Because of the increased flux available at the SRR, maximum effort was placed in processing the slug as rapidly as possible after the end of the irradiations.

IRRADIATIONS

In February 1963, a Cm capsule, the descendent of the Berkeley "Napkin Rings", was processed.¹ The Cm fraction, 34 mg of Cm (see Table I) was returned to the MTR as one end of a capsule, UCRL-1b-150. The other end of the capsule contained a mixture of 25 mg ²⁴⁴Cm and 200 (?) mg ²⁴⁰Pu.

From March 1963 until January 1965, UCRL-1b-150 received 2.21×10^{22} neutrons/cm² at the MTR. The slug was then transferred to the higher flux (2.5×10^{15} n/cm²-sec) at SRR where it received 8.7×10^{21} neutrons/cm² during the two months ending April 27, 1965.

PROCESSING

General interest in the curium fraction, from the Cm end of the capsule, required that the two halves of the slug not be mixed, at least until the curium fractions had been separated. A further requirement² that the capsule be dissolved in acid, made it necessary to remove as much aluminum as possible in order to minimize the evolution of H₂. A small lathe was installed in the 4 ft cave³ and after neutron counting the whole capsule, the capsule was machined into two pill shaped portions, each weighing about 3 gms. Because the Cm end of the slug, as expected, emitted about 7 times as many neutrons as the Cm-Pu end, it was transferred to the 6 ft cave to be processed first. (See Simplified Flow Sheet.)

The 3 gm Al pill, containing the Cm, was dissolved in 50 cc of 7.5 N HCl with 0.1 gm of HgCl₂ added as a catalyst. The dissolution was done under a negative 5 lbs pressure in a sealed system. The H₂ and rare gasses were collected in a previously evacuated cylinder outside of the cave box. After dissolution, the Al was removed by precipitating the actinides with 10 M KOH, and trace amounts of Cu were removed by digesting the precipitate with 6 M NH₄OH. Following a water wash, the precipitated actinides and fission products were dissolved in 15 cc of 12 M LiCl - 0.1 N HCl.

The fission products and the bulk of the Cm were separated from the transcurium elements by elution from Dowex-1 using 10 M LiCl. The column size was 5 mm x 12 cm and the characteristic glows of the rare earths and the Cm indicated that approximately 5% of the resin bed was used during loading. The glass wool plug above the resin bed served as a filter for the black insoluble residue² which would not dissolve in the load solution. The transcurium fraction was stripped from the resin with

6 N HCl after the bulk of the curium had been eluted. In order to reduce bubbling⁴ on the column 4% CH₃-OH was added to all of the LiCl solutions and the temperature lowered from 90°C to 60°C.

The intra-actinide separations were carried out by elution from the NH₄ form of Dowex 50X12 resin with α -hydroxy-isobutyric acid. To clean up the transcurium fraction after the LiCl column, and, prior to the "But" column, the actinides were precipitated as hydroxides using Fe⁺⁺⁺ as a carrier, redissolved in 9 M HCl and passed through an anion column (Dowex-1, 5 mm \times 9 cm) to remove the Fe and trace amounts of Zr. At this point the transcurium fraction was evaporated to dryness and transferred to a clean chemistry box in the 4 ft cave to insure that no recontamination with the rare earths or curium occurred. Final purification was performed by eluting the actinides with 1.5 N HCl from a 3 mm \times 5 cm Dowex 50X4 cation column. The purified actinides were evaporated to dryness, redissolved with 0.05 M HCl, loaded on a 3 mm \times 5 cm "But" column, and eluted with 0.175 M α -hydroxy isobutyric acid at a pH of 4.9. The α and $\beta\gamma$ activity per drop was recorded⁵ as each drop formed, giving an immediate plot of the elution position of each actinide (curve No. 1).

The Cm-Pu portion of the capsule was processed in almost the same manner as the Cm portion. The two exceptions were: an anion column before the LiCl column to remove the Pu fraction, and the addition of all supernate scrounges from the Cm end of the capsule. (See Simplified Flow Sheet).

RESULTS

Amounts at the time of separation

Cm end		Cm-Pu end	
92 hrs after irradiation		Pu ²⁴²	31 mg
Cm	26.8 mg (See Table II)	Cm	17.4 mg (See Table IV)
Bk ²⁴⁹	7.85 µg	Bk ²⁴⁹	1.75 µg
Cf	60.3 µg (See Table III)	Cf	13.8 µg (See Table V)
Es ²⁵³	415 ng	Es ²⁵⁴	3.4 ng
Es ²⁵⁴	(.13 ng*)	Es ²⁵⁵	0.13 ng
Fm ²⁵⁴	0.26 ng	Fm ²⁵⁵	0.12 ng,
Fm ²⁵⁶	0.2 pg,	Fm ²⁵⁷	4.0 d/m
Md	0.0	Fm ²⁵⁷	1.0 d/m

* Estimated

DISCUSSION OF RESULTS

A. Chemical Separation

The chemical separations were entirely satisfactory during the cave run. Although the actual yields of several isotopes were different than the anticipated yields, the discrepancy appears to belong to low irradiation production (or over optimistic calculations) rather than to low chemical yields. The separations of the various actinide elements eluted from the "But" columns was exceptionally good.

A study⁶ of "But" columns after the January '64 cave run showed that Dowex 50X12 resin appears to deteriorate with time in its ability to reach equilibrium quickly. Thus, if the eluting solution is forced through the resin too rapidly, poor separations are effected. But on the

other hand, if the eluting solution is passed through too slowly, peroxide formation and thermal heating, dry out the resin producing channeling and poor separations. Although the addition of 4% CH_3OH to the α -hydroxyisobutyric acid inhibits peroxide formation it doesn't stop it completely. Each batch of resin must be periodically checked to determine the maximum elution rate.

In an attempt to observe the predicted short lived isotopes⁷ of Fm^{258} , Fm^{259} and/or Md^{259} the Cm end of the capsule was processed as rapidly as possible. The separated actinides were removed from the caves 92 hrs after the end of the irradiation. (40 hrs were spent in transportation from SRR to Berkeley.) About 1% of the activity was lost per hydroxide carry during the rapid processing of the Cm end. These losses, which were predicted, were recovered and added to the Cm-Pu end. No appreciable losses (less than 0.01%) were observed when the hydroxide carries were allowed to digest overnight in the Cm-Pu processing.

B. Berkelium

Previous cave runs have left confusion concerning the chemical yield vs irradiation yield of Bk^{249} . Although the "Recommended Flow Sheet" of J. L. Green et al.² was not followed in regards to extracting the Bk after the LiCl column, the slug was dissolved in acid and assayed for the Bk present in the dissolver solution. The assay consisted of a LiCl column to remove rare earths and H.D.E.H.P. extraction. The results (see Table VI) indicate that all of the Bk present in the dissolver solution was separated on the final "But" column and recovered.

In order to compare the results of this cave run to past cave runs, the Bk²⁴⁹ recovered is shown as a function of the Cm²⁴⁸ and Cf²⁵⁰ recovered (Table VII). Changes in the total neutron flux between various capsules processed in past cave runs makes the comparison of Bk²⁴⁹ to Cf²⁵² unreliable.

ACKNOWLEDGMENTS

A cave run requires the cooperation and help of a large number of people. Thanks are extended to all members of Health Chemistry, under the direction of Pat Howe, who provided the cave room facilities. Thanks are also extended to Maynard C. Michel for the mass analysis and to Sherman Fried for his general help and interest. The cave run would not have been possible without the Health Chemistry Personnel, who did the actual manipulations. Special thanks, therefore, to Dick Bolton, John Depew, Bruce Duncan, Bob Gribben, F. T. "Mac" McCarthy, and Jean Rees.

Simplified Flow Sheet

(Yields for Cf²⁵² based on neutron cts shown in μg)
not corrected for Cf²⁵⁴

UCRL-16-150

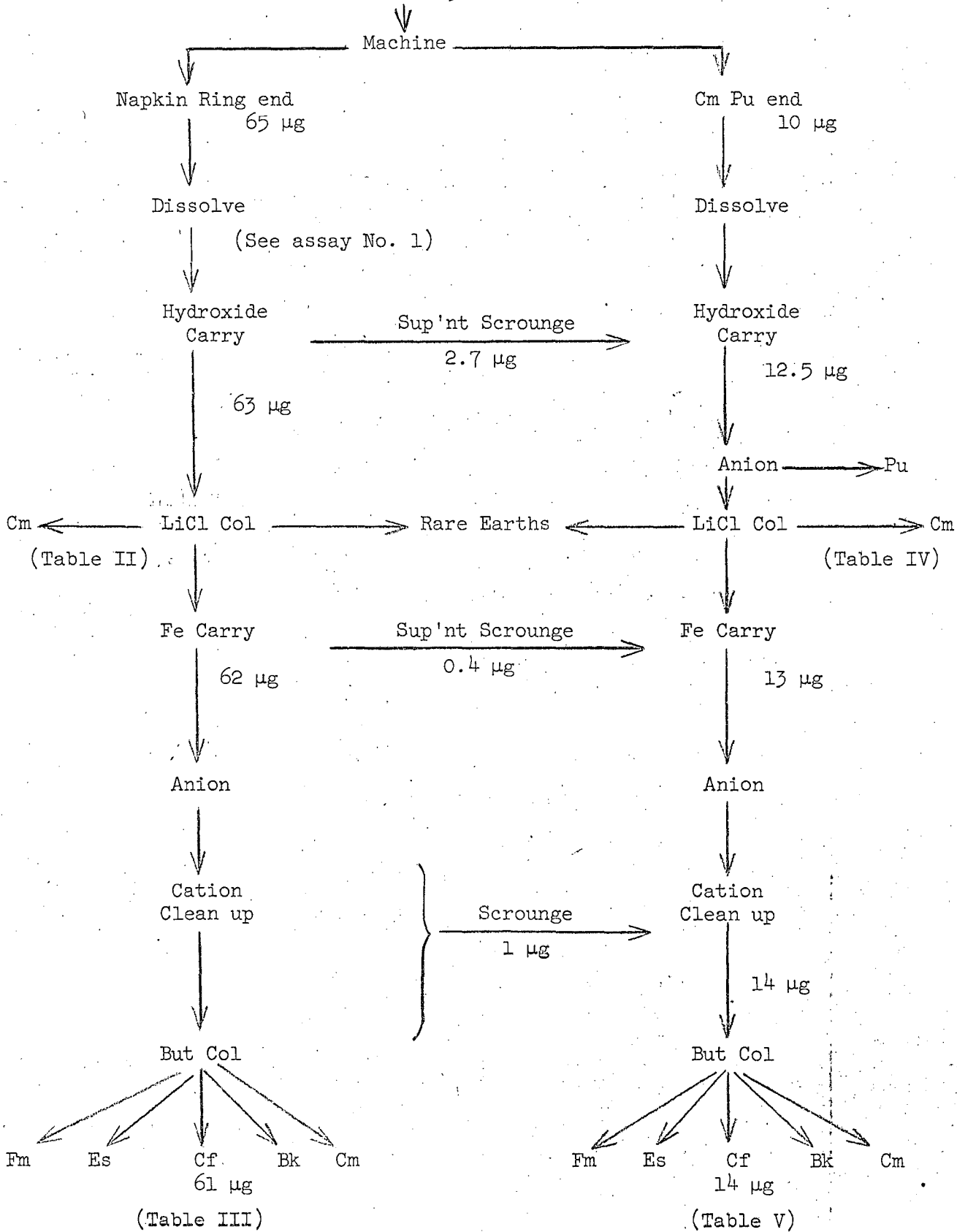


Table I. Mass analysis of the Cm prior to irradiation.

Cm ²⁴⁴	24.9 mg	74.25%
Cm ²⁴⁵	0.25	0.73
Cm ²⁴⁶	7.7	22.65
Cm ²⁴⁷	0.25	0.74
Cm ²⁴⁸	0.55	1.63

Table II. Mass analysis of the Cm after irradiation.

Cm ²⁴⁴	15.4 mg	57.5%
Cm ²⁴⁵	0.12	0.46
Cm ²⁴⁶	9.95	37.2
Cm ²⁴⁷	0.27	1.0
Cm ²⁴⁸	1.04	3.9

Table III. Mass analysis of the Cf fraction from Cm end.

Cf ²⁴⁹	0.12 μ g	0.20% (Bk ²⁴⁹ present)
Cf ²⁵⁰	6.28 μ g	10.42%
Cf ²⁵¹	1.45 μ g	2.41%
Cf ²⁵²	51.64 μ g	85.62%
Cf ²⁵³	0.83 μ g	1.37% (Es ²⁵³ present)
Cf ²⁵⁴	22.3 ng	0.037%

Table IV. Mass analysis of Cm from Cm-Pu after irradiation.

Cm ²⁴⁴	13.2 mg	75.2%
Cm ²⁴⁵	0.12	0.67
Cm ²⁴⁶	3.8	21.6
Cm ²⁴⁷	0.1	0.58
Cm ²⁴⁸	≤0.24	≤1.35
(Th ²³² O ⁺ contamination)		

Table V. Mass analysis of Cf from Cm-Pu end.

Cf ²⁴⁹	0.026 μg	0.19% (Bk ²⁴⁹ present)
Cf ²⁵⁰	1.46	10.61
Cf ²⁵¹	0.38	2.77
Cf ²⁵²	11.85	85.89
Cf ²⁵³	0.07	0.52
Cf ²⁵⁴	0.002	0.02

Table VI. Assay No: 1.

Cm ²⁴⁴	16 mg
Bk ²⁴⁹	7.1 μg
Cf ²⁵²	59 μg
(includes S.F. from Cf ²⁵⁴)	
Es	2.2×10^{10} d/m
Fm ²⁵⁴	5×10^9 d/m

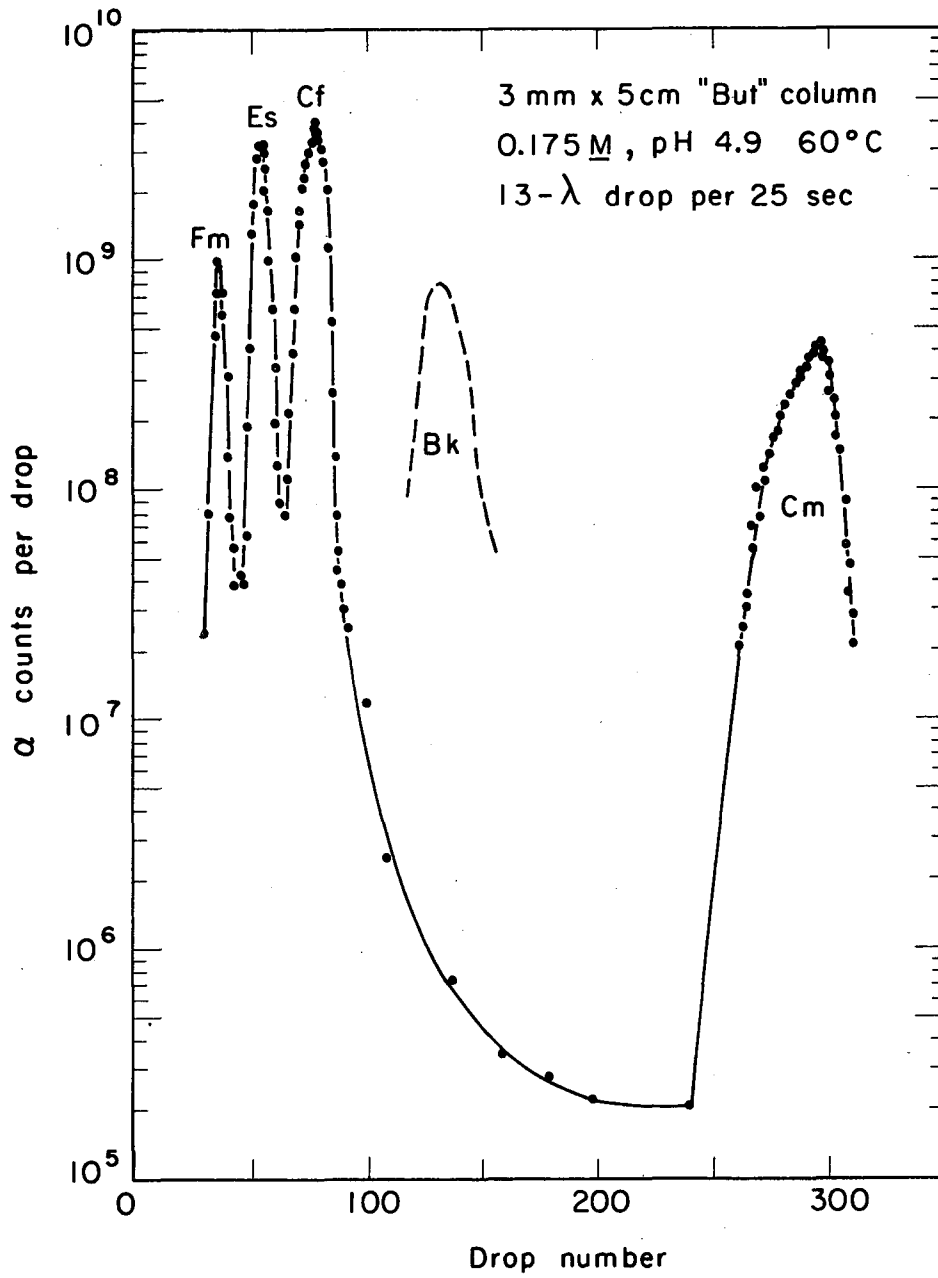
Table VII. Mass yield ratios of Cm²⁴⁸:Bk²⁴⁹:Cf²⁵⁰
from this and previous cave runs.

	$\frac{\text{Bk}^{249}}{\text{Cm}^{248}}$	$\frac{\text{Bk}^{249}}{\text{Cf}^{250}}$
Predicted Ratio ⁷	8×10^{-3}	1.6
May 65 (1b-150)*		1.24
Cm end	7.5×10^{-3}	1.24
Pu end	7.3×10^{-3}	1.20
Jan 64 Am 3 and 4	3.8×10^{-3}	0.82
Feb 63 (1b-145)*	8.3×10^{-3}	1.28
July 61 (1b-19)*	6.3×10^{-3}	1.17

* Note slug No. 1b-19 was feed for No. 1b-145 which in turn was feed for No. 1b-150.

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Fig. 1

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