

THE RECOVERY AND STUDY OF HEAVY NUCLIDES PRODUCED IN A NUCLEAR EXPLOSION—THE HUTCH EVENT

R. W. Hoff and E. K. Hulet Lawrence Radiation Laboratory, University of California Livermore, California 94550

ABSTRACT

During the explosion of the Hutch device, the target (^{238}U) and $^{232}\text{Th})$ was subjected to a very high neutron exposure, $^{2.4}\times10^{25}$ neutrons/cm². Multiple neutron capture reactions resulted in the production of heavy nuclides, up to and including ^{257}Fm . Results of the search for species with A > 257 were negative. The recovery and chemical processing of kilograms of Hutch debris has resulted in the isolation of 1010 atoms of ^{257}Fm , which is 102 times more material than has been available for experimentation in the past. Experimentally significant amounts of other rare nuclides, e.g., ^{254}Cf , ^{251}Cf , ^{255}Es , and ^{250}Cm , have also been separated from the Hutch debris. The production of these nuclides in thermonuclear explosions is shown to be a valuable supplement to the AEC program for reactor production of transplutonium elements. The neutron flux achieved in Hutch was insufficient to even approach production of nuclides in the region of 298 114. A much more intense neutron flux is required. In future experiments, considerable attention must be given to the problem of adequate sample recovery, in order to properly use the ability to subject targets to an exceedingly intense time-integrated neutron flux.

HEAVY ELEMENT PRODUCTION—NUCLIDES WITH A ≤ 257

Explosion of the Hutch device resulted in irradiation of a mixed ²³⁸U and ²³²Th target to a total exposure of 40 moles neutrons/cm² (equivalent 20 keV neutron flux) and consequent production of appreciable amounts of certain heavy nuclides. This production was discussed by S. F. Eccles¹ and is summarized in a plot of logarithm of total atoms produced versus mass number in Fig. 1. The same data are given in Table I, where the nuclide listed for each mas number is usually the final (and longest-lived) member of each beta-decay chain. Early results and conclusions from the Hutch experiment have also been described in an earlier report by Eccles and Hulet.²

To obtain these experimental data, samples of rock containing explosion debris were recovered by drilling 2,000 ft below the surface of the ground at the Nevada Test Site. The first debris from the drilling was returned to our laboratory 7 days after the explosion. A 100-g sample of this material was processed immediately and a second 100-g sample that was especially rich in actinides was treated a day later. Hundred-gram samples were also sent to groups at Los Alamos Scientific Laboratory and Argonne National Laboratory for analysis. The rock was dissolved and the transcurium actinides were chemically isolated. Weightless samples of each element were electrodeposited and then carefully counted for alpha and spontaneous fission radioactivity. We found that the second 100 g of debris, representing 8.4×10^{-10} fraction of

^{*}Work performed under the auspices of the U.S. Atomic Energy Commission.

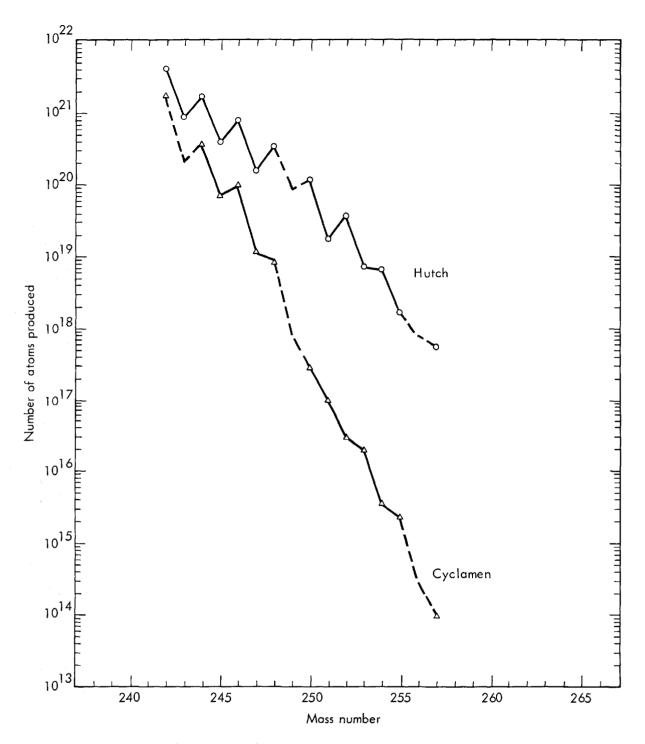


Fig. 1. Heavy element yields from the Hutch and Cyclamen experiments.

the total, contained 4×10^8 atoms of $^{257}{\rm Fm}$, or about seven times the quantity of this scarce isotope ever before isolated. The abundances of other known isotopes were equally impressive.

SEARCH FOR NUCLIDES WITH A > 257

In the analyzing of the Hutch samples, one of the most urgent problems was the search for new isotopes with mass numbers greater than 257. Extrapolating the data of Fig. 1, we estimate it should have been possible to detect nuclides with mass numbers as high as 265, provided the following conditions were satisfied: 1) the species with A > 257 do not experience a sudden increase in probability for neutron-induced fission, 2) the members of the

Table I. Total atoms for each mass number chain produced in the Hutch experiment

Nuclide	Half-life and decay mode	Total atoms (t _o , 7/16/69)	Total atoms (1/1/70)
242 _{Pu}	$3.9 \times 10^5 \text{ y } \alpha$	4.22 × 10 ²¹	Same
$243^{\mathrm{Am}}_{\mathrm{Am}}$	$8.0 \times 10^3 \text{ y } \alpha$	9.03×10^{20}	Same
244 _{Pu}	$8.3 \times 10^7 \text{ y } \alpha$	1.71×10^{21}	Same
$^{245}\mathrm{Cm}$	$8.3 \times 10^3 \text{ y } \alpha$	3.92×10^{20}	Same
$^{246}\mathrm{Cm}$	$4.7 \times 10^3 \text{ y } \alpha$	8.54×10^{20}	Same
$^{247}\mathrm{Cm}$	$1.6 \times 10^7 \text{ y} \alpha$	1.60×10^{20}	Same
248 _{Cm}	$3.8 \times 10^5 \text{ y } \alpha$	3.54×10^{20}	Same
249 _{Bk}	314 d β ⁻	(not measured)	(not measured)
249 _{Cf}	352 y α	(not measured)	(not measured)
250 _{Cm}	1.1×10^4 y SF	$\frac{-}{1.21 \times 10^{20}}$	Same
251 _{Cf}	900 y α	1.82×10^{19}	Same
252 _{Cf}	$2.7 \ \mathrm{y} \ \alpha$	3.82×10^{19}	3.4×10^{19}
253 _{Cf}	2.7 y α 18 d β -	7.20×10^{18}	1.1×10^{16}
253 _{Es}	20 d α	7.20 × 10	2.1×10^{16}
$^{254}\mathrm{Cf}$	$20~\mathrm{d}~lpha$ 60 d SF	6.82×10^{18}	9.7×10^{17}
255 _{Es}	40 d β -	1.66×10^{18}	8.9×10^{16}
$255_{\mathrm{Fm}}^{\mathrm{ES}}$		1.00 X 10	1.8×10^{15}
256_{Fm}	20 h α	(4 ala4 1:1)	1.8 X 10
257 _{Fm}	2.6 h SF	(too short-lived)	
Fm	95 d α	5.56×10^{17}	1.6 × 10 ¹⁷

beta-decay chains were not consumed rapidly by spontaneous fission decay, and 3) the final member(s) of the beta-decay chain survive(s) long enough to be detected 8 days after the Hutch explosion.

The unknown nuclides $^{259}\mathrm{Fm}$ and $^{259}\mathrm{Md}$, end products of the mass-259 beta-decay chain, offer a good chance for detection because at formation time they would presumably be only a little less abundant than $^{257}\mathrm{Fm}$. The predicted decay modes, energies, and half-lives for $^{259}\mathrm{Fm}$ and $^{259}\mathrm{Md}$ are given in Table II. Our estimates of the spontaneous fission half-lives were

Table II. Estimated decay characteristics of ²⁵⁹Fm and ²⁵⁹Md

Nuclide	SF t _{1/2}	Alpha particle energy (MeV)	Alpha t _{1/2}	Beta decay energy (MeV)	Beta t _{1/2}
259 _{Fm}	0.1 - 1 h	6.4	2 y	0.4	0.1 - 3 d
$^{259}\mathrm{Md}$	0.1 - 1 h	6.8	33 y	0	stable

made by extrapolating from data shown in Fig. 2, experimentally-measured SF half-lives plotted as a function of neutron and proton number. In the region above N = 152, SF lifetimes are decreasing rapidly, amounting to nearly 10^5 reduction upon adding two neutrons or two protons. Indeed, these short lifetimes severely restrict the chances for the formation and identification of

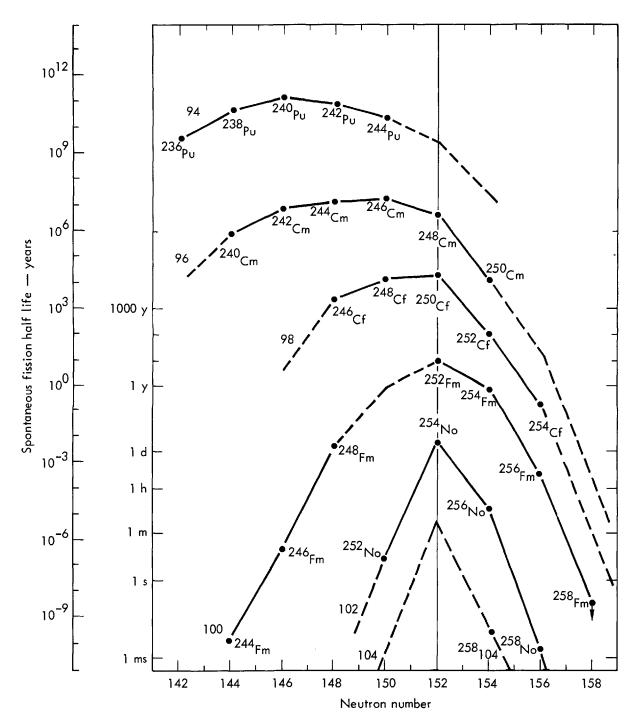


Fig. 2. Spontaneous fission half-life trends for even-even nuclei in the heavy elements.

nuclei with A > 257. One result is that the even-numbered mass chains with A > 254 are consumed by SF, leaving only the odd-numbered chains to survive because of hindrance to spontaneous fission rates associated with an odd neutron or proton. These predictions of spontaneous fission half-lives are sufficiently uncertain that there was a reasonable possibility of survival of a detectable number of atoms with A = 259 (~100 atoms are required) in our samples.

Counting of alpha particles and fission fragments from the first pure sample of fermium began ~8.5 days after the test. At that time the ratio of alpha to fission disintegration rates of $^{257}{\rm Fm}$ in the sample was 470 ± 25.

By comparing this value to an average ratio of 495 ± 15 measured later and to a ratio of 510 ± 25 for reactor-produced $^{257}{\rm Fm}$, we find no excess fissions attributable to $^{259}{\rm Fm}$. Similarly, in two samples of a combined Md-Lw fraction there was no evidence for $^{259}{\rm Md}$ in the alpha spectra, nor was there unassigned spontaneous fission activity. After counting alpha particles from the Md-Lw samples for 20 days in a Frisch-grid ionization counter, we noted only 24 events in the expected alpha-energy range for $^{259}{\rm Md}$ of 6.7 to 7.0 MeV; these were shown to be random background. In addition, we have not observed any growth of 7.03 MeV $^{255}{\rm Fm}$ alpha particles, a product of the alpha decay of either $^{259}{\rm Fm}$ or $^{259}{\rm Md}$. Thus, we have no positive evidence for the presence of $^{259}{\rm Fm}$ or $^{259}{\rm Md}$ in our samples from Hutch.

Half-life limits for $^{259}\mathrm{Fm}$ and $^{259}\mathrm{Md}$ were calculated from the observed alpha and fission rates of our samples and are listed in Table III. Essential assumptions are that neutron capture proceeded from $^{257}\mathrm{U}$ to $^{259}\mathrm{U}$ without serious fission competition and that the mass-259 beta decay chain was not terminated by SF before reaching $^{259}\mathrm{Fm}$. On that basis we expected a total of 1017 atoms of $^{259}\mathrm{Fm}$ or $^{259}\mathrm{Md}$ product.

Table III. Half-life limits for 259 Fm and 259 Md

$$^{259}{
m Fm}$$
: $T_{1/2} \le 0.5$ d (SF) or $\ge 250 {
m y}(\alpha)$ $^{259}{
m Md}$: $T_{1/2} \le 0.5$ d (SF) or $\ge 1.25 \times 10^4 {
m y}(\alpha {
m or SF})$

In the upper range, the half-life limits for either nuclide are much too long to be credible, particularly when related to systematic trends in this region of nuclei. We can only conclude that one or more members of the mass-259 chain is shorter lived than our limits. The shortest half-life limits measured in this experiment and Cyclamen ($\leq 5.5 \, \mathrm{hr}$)⁴,⁵ are still well above the predicted lifetimes for SF. Clearly, because of the long drilling time needed to recover debris samples in underground nuclear experiments, it is impossible to use this technique when trying to identify heavy species with half-lives shorter than several hours. In view of the hour or less half-lives expected, we must find new methods for the faster recovery of debris in future experiments.

We might have also expected to see alpha decay from ^{265}Lw or ^{267}Lw , providing these nuclides were produced and survived long enough to be detected. In the energy region above 7.0 MeV where one would expect to observe the alpha groups of these nuclides, we found no evidence for their existence. The intermediate odd-A mass chains probably terminate at ^{261}No and ^{263}No . The LRL analyses did not include isolation of a nobelium fraction, but studies at LASL and ANL failed to produce any evidence for the existence of these nuclides.

A search was also made for long-lived isotopes of element 104. Chemical isolation followed carrier-free Hf chemistry since the chemical properties of element 104 are expected to resemble those of Hf and Zr. There was no evidence for spontaneous fission or alpha activity in the final sample. Either element 104 was not produced in Hutch or the half-lives (spontaneous fission) are too short to allow observation in this experiment. Since the lightest isotope that might have been detected is $^{269}104$, production and detection of this nuclide was marginal at best.

One of the more intriguing postulates of recent years in the study of heavy elements has been the suggestion that nuclides in the region of ²⁹⁸114 may be long-lived enough to be detected if one can derive a means of producing them. Although the predicted half-lives and the extent of this "island of stability" are quite uncertain, it appears that one must produce nuclei with

A > 294 in order to reach a region of half-lives longer than a few hours. The neutron flux achieved in Hutch was insufficient to even approach production of atoms of mass number 294. A much more intense neutron flux is required. In addition to the question of whether there are ways to produce the necessary neutron exposure, one can ask whether the problems that prevented detection of nuclides with A = 258-265 in the Hutch debris will also prevent formation and detection of heavier nuclides. If this problem lies mainly in the destruction of heavy nuclei by neutron-induced fission during the capture phase, then one is doomed to failure in attempting to produce even higher fluxes and heavier products. If, however, the Hutch explosion resulted in the production of reasonable amounts of nuclides with A = 258-265 and these nuclides were then consumed in spontaneous fission decay of certain members of each beta-decay chain, there is still a mechanism available for producing heavier products if one finds a way to increase the total neutron exposure. Unfortunately, there are no direct clues in the data as to which mechanism is responsible for the fruitless search for heavier species.

LARGE-SCALE RECOVERY AND PROCESSING OF HUTCH DEBRIS

Perhaps the most important gain scored by the Hutch test is the opening of a new path for synthesizing useful quantities of rare nuclides. The debris offers a unique source of $^{250}\mathrm{Cm}$ and $^{257}\mathrm{Fm}$, both of which can serve as target materials for charged particle bombardments to produce hitherto unobserved isotopes and possibly new elements. The $^{250}\mathrm{Cm}$ and $^{257}\mathrm{Fm}$ are rare enough that it was considered attractive to attempt to recover 500 kg of debris for chemical processing and recovery of the heavy actinides.

Accordingly, the Phase II, large-scale sample recovery operation was carried out during the period August 22 to September 14, 1969. The recovery technique involved enlarging a drill hole, initially 10 in. in diameter, to a 15-in. diameter by reaming and collecting the loosened rock in a basket mounted below the reaming tool. A total of eleven baskets, each full of rocks and drilling mud, were recovered from three holes drilled in various positions in the debris distribution. The total weight of material recovered in this manner was approximately 1100 lb or 500 kg.

During the initial attempts to recover samples immediately following the explosion, the recovery of debris from the 2000-ft depth was accomplished by sidewall sampling. With this technique a 100- to 200-g sample is obtained by forcing a hollow sampling tool into the side of the hole and recovering a sample from the rock lodged in the tool. Early Hutch samples obtained in this manner showed relatively good concentrations, e.g., the second 100-g sample studied had a concentration of 8×10^{-12} of the device per gram of rock in the sample. During Phase I sampling, a total of approximately 10 kg of sample was recovered. A few kg were dissolved for the early studies and the remainder was saved for later dissolution.

Analysis of the 500 kg of sample recovered in Phase II showed that the concentration of device debris in this rock was much lower than expected. In fact, the average concentration in the 500 kg was $\leq 3\%$ of that in the sidewall samples recovered during Phase I. Thus, we had the remainder of the sidewall samples, 6 kg that contained 5×10^{-8} of the device, and 500 kg of sample that contained $<12\times10^{-8}$ of the device. Unless this latter batch could be enriched to some high degree through a "high-grading" technique, it was unattractive to process this material.

We have investigated the question of high-grading the 500 kg of material. After washing to remove drilling mud, the rock was screened to produce fractions classified according to particle size. The specific gamma activity of these fractions remains essentially constant. Thus, there was no gain in merely

sorting the rock according to size. In the larger-sized fractions, one can pick out pieces of fused rock which exhibit relatively high specific gamma activity. However, the yield from this operation is extremely low, e.g., 15 kg of material yields less than 100 g of fused rock.

A more promising high-grading technique has been to sort the more finely-divided material according to magnetic properties. In preliminary experiments, a factor of 4 enrichment has been obtained in a fraction that contains 87% of the spontaneous fission activity. Further study may allow us to improve this factor. Another approach which looks promising is the use of flotation processes for enriching the debris. These high-grading techniques are being further developed.

Inasmuch as we have not yet developed a high-grading technique that will give the desired degree of enrichment, we have proceeded to process a total of 10 kg of rich sample, leaving the remaining 500 kg of material for possible later treatment. Our chemical processing equipment is designed to operate on a 5-kg batch size. The process flowsheet, subject of an earlier report, involves leaching the actinide elements from finely-ground rock samples with a mixture of HNO3-HF. Following the leach step, the actinides are extracted directly into dioctylpyrophoric acid (DOPP). Following back extraction, an actinide lanthanide separation is accomplished with a LiCl-Dowex 1 anion exchange column. The final step is the production of pure elemental fractions of Fm, Es, Cf, Bk, and Cm. The processing of this 10 kg, which is nearly completed, will yield about 4×10^{-8} of the total device production. In terms of the key nuclides, this represents about 1×10^{10} atoms of 2^{57} Fm and 5×10^{12} atoms of 2^{50} Cm at this time.

RARE NUCLIDES PRODUCED IN THERMONUCLEAR EXPLOSIONS

In this section we wish to discuss some of the more rare heavy actinide isotopes produced in thermonuclear explosions and to contrast their production and availability with that for these same nuclides in reactor production. There are at least five nuclides that are most advantageously produced in thermonuclear explosions: $^{257}\mathrm{Fm}$, $^{255}\mathrm{Es}$, $^{251}\mathrm{Cf}$, $^{254}\mathrm{Cf}$, and $^{250}\mathrm{Cm}$. The first three of these nuclides exhibit large fission (and/or capture) cross sections for thermal neutrons; hence, they are rapidly destroyed in a high-flux reactor. In thermonuclear explosions, the average neutron energy during the capture process is tens of kilovolts and consequently the destruction cross sections for the entire series of multiple neutron capture products tend to be relatively uniform. The result is that certain elemental fractions, e.g., Es, Cf, and Cm, exhibit considerably less variation in isotopic composition when recovered from thermonuclear explosions as compared with reactor-produced material. Our intent is to show that production of these nuclides in thermonuclear explosions serves as a valuable supplement to the already enormously productive AEC production program centered about the High Flux Isotopes Reactor (HFIR) and the Transuranium Processing Plant (TRU) at Oak Ridge National Laboratory.

The most obvious advance in the Hutch experiment was the production of 6×10^{17} atoms of $^{257}{\rm Fm}$. This nuclide, the longest-lived isotope of fermium, has a 95-day half-life and decays predominantly by alpha emission with spontaneous fission branching ratio, ${\rm SF}/\alpha$, of 2×10^{-3} . Semiannual production of $^{257}{\rm Fm}$ in the HFIR-TRU complex at Oak Ridge has been $^{-8}\times10^{7}$ atoms per batch, recently. Future production is predicted to increase to an annual rate of approximately 2×10^{9} atoms by 1971. Thus, the amount of $^{257}{\rm Fm}$ produced in the Hutch explosion exceeded current reactor production capacity by a factor of almost 10^{10} .

Much of the interest in ²⁵⁷Fm for experimental purposes lies in the possibility of recovering enough material to serve as a target for charged-particle

and neutron irradiations. As has been already discussed by Cowan,⁵ a list of laboratory-produced nuclides one might produce with a ²⁵⁷Fm target is shown in Table IV.

Table IV. Reactions for producing new nuclides with a ²⁵⁷Fm target.

$^{257}\mathrm{Fm}$	$(n,\gamma)^{25}$	8 Fm
	$(d, p)^{25}$	$^{68}\mathrm{Fm}$
	$(t, p)^{25}$	
	$(t, n)^{25}$	
	$(\alpha, n)^{26}$	
($(\alpha, 2n)^{25}$	9 No

The goal here is to produce nuclides with neutron numbers greater than 157. To date, no heavy species has been observed that contains more than 157 neutrons in its nucleus, in spite of repeated attempts to identify $^{258}\mathrm{Fm}$ from the $^{257}\mathrm{Fm}(n,\gamma)$ reaction. It has been concluded that the half-life of $^{258}\mathrm{Fm}$ is probably shorter than 200 msec where the decay is predominantly spontaneous sion. Of importance in the experimental program for the $^{257}\mathrm{Fm}$ target outlined in Table IV is that one can repeat the experiments many times, an essential feature when one is searching for and studying short-lived nuclides. In contrast, if one is constrained to search for nuclides such as $^{259}\mathrm{Fm}$, $^{259}\mathrm{Md}$, and $^{260}\mathrm{No}$ only in the debris following the explosion of a heavy-element-producing device, the opportunities for repeated experiments are limited.

Another nuclide of interest in the Hutch debris is the 39-day alpha emitter $^{255}{\rm Es}$. In Table V we see that einsteinium produced in the High Flux Isotope Reactor contains 0.06 at.% $^{255}{\rm Es}$. To produce higher isotopic purity for experimental purposes, we have enriched these samples in the Livermore isotope separator with resultant purity as high as 90 at.% $^{255}{\rm Es}$. The $^{255}{\rm Es}$ abundance in the Hutch Es is 20 at.% (see Table V), intermediate in concentration between the raw HFIR product and the same material following isotope separation. A significant feature of the Hutch Es is that $^{254}{\rm mEs}$ is entirely absent, since the beta decay for mass 254 stops at the beta-stable $^{254}{\rm Cf}$. As an example, $^{254}{\rm mEs}$ is an objectionable impurity in samples where one is studying $^{255}{\rm Es}$ alpha decay because the lower-energy $^{254}{\rm mEs}$ alpha groups tend to overlap the alpha groups of $^{255}{\rm Es}$. Thus, Hutch Es, if further enriched in an isotope separator, offers good prospects for very pure samples of $^{255}{\rm Es}$ for experimentation.

Table V. Comparison of reactor-produced and Hutch einsteinium

	HFIR Es (a)		Hutch Es	
Half-life, type of decay	A	Atom ratio	A	Atom ratio
20.5 d α	253	1.00	253	1.00
$276~\mathrm{d}~lpha$	$254\mathrm{m}$	0.003	254m	0
39 d β ⁻ ,α	255	0.0006	255	0.231

^aThe isotope ratios listed are typical for material directly from the HFIR reactor. The composition of HFIR Es changes rapidly with time because of the short half-lives of the isotopes.

The californium fraction in the Hutch debris is significantly richer in two isotopes, $^{251}\mathrm{Cf}$ and $^{254}\mathrm{Cf}$, than is reactor-produced Cf. The comparison is made in Table VI. As feed material for isotopic enrichment, the Hutch Cf presents an opportunity to produce $^{251}\mathrm{Cf}$ samples of high purity. In addition, the Hutch Cf offers an excellent opportunity to study the spontaneous fission of $^{254}\mathrm{Cf}$. When the first samples were recovered, 98% of the spontaneous fission rate of the Cf sample was due to $^{254}\mathrm{Cf}$; even now the $^{254}\mathrm{Cf}$ is 93% of the total rate. Wolfsberg and others 12 have used the Hutch Cf to make a radiochemical study of the distribution of fission products from the spontaneous fission of $^{254}\mathrm{Cf}$.

Table VI. Comparison of reactor-produced and Hutch californium

TT 10 1:0	HFIR Cf ^a		Hutch Cf	
Half-life, type of decay	A	Atom ratio	A	Atom ratio
352 y α	249	0.0012	249	-
13 y α	250	0.0083	250	
900 yα	251		251	0.48
$2.65 \text{ y} \alpha$, SF	252	1.000	252	1.00
18 dβ ⁻	253	0.0069	253	0.19
60 d SF	254	0.0007	254	0.18

 $^{^{\}dot{a}}$ Californium of differing isotopic compositions can be produced in the HIFR, depending upon total neutron exposure, target material, etc. The composition listed here is that for $^{252}\mathrm{Cf}$, which had been irradiated for 70 days at 3 \times 10 15 n/cm 2 - sec and had served as feed material for isotopic enrichment of $^{253}\mathrm{Cf}$ and $^{254}\mathrm{Cf}$.

The Hutch curium is an invaluable source of the rare isotope, $^{250}\mathrm{Cm}.$ Reactor production of heavy curium isotopes does not produce appreciable (or even detectable) amounts of $^{250}\mathrm{Cm}$, since its production depends upon competition between beta decay of 64-min $^{249}\mathrm{Cm}$ and neutron capture by $^{249}\mathrm{Cm}.$ The 2200 m/s capture cross section for $^{249}\mathrm{Cm}$ is reported to be 2.8 barns (see p. 32 of Ref. 10). Even at the flux level of the HFIR (5 \times 10 15 n/cm² sec), a very large fraction of the $^{249}\mathrm{Cm}$ atoms beta-decay (or are destroyed by neutron-induced fission), and only the minutest amount are converted to $^{250}\mathrm{Cm}.$ The Hutch Cm contains 6.4 at.% $^{250}\mathrm{Cm}$ (Table VII). This isotope is particularly attractive for use as a target material in heavy-ion bombardments because of its high ratio of neutrons to protons. It is often desirable to use neutron-rich targets in heavy-ion bombardments, since the products tend to be on the neutron-deficient side of beta stability. As an example, recent studies of the isotopes of elements 104 and 102 involved the bombardment of a $^{248}\mathrm{Cm}$ target with oxygen ions. The half-life of $^{250}\mathrm{Cm}$, about 104 yr, is long enough that the debris can be mined and chemically processed long after the majority of fission products have decayed.

FUTURE EXPERIMENTS

From the preceeding discussion, it is clear that in future experiments considerable attention should be paid to insuring adequate sample recovery. There are two aspects of the problem that require development: rapid sample recovery and large-scale sample recovery.

In the search for short-lived species in previous experiments, a limit of ≤ 5.5 hr was set for the half-life of $^{259}{\rm Fm}$ in the Cyclamen event. In this

Table VII. Comparison of reactor-produced and Hutch curium

TT 10 1:0	HFIR Cm ^a			Hutch Cm	
Half-life, type of decay	A	Atom ratio	A	Atom ratio	
18 y α	244	5.11	244		
8300 y α	245	0.043	245	1.00	
$4700~\mathrm{y}~\alpha$	246	1.000	246	1.00	
$1.6 \times 10^7 \text{ y } \alpha$	247	0.030	247	0.41	
$3.8 \times 10^5 \text{ y} \alpha$	248	0.058	248	0.91	
1.1×10^4 y SF	250	_	250	0.31	

^aThe isotopic content of curium from HFIR will depend upon the circumstances of its irradiation. The isotopic composition listed here is for material that has received intense neutron exposure and is consdiered a useful source of isotopes with A > 244.

work a sample had been recovered, processed chemically, and was ready for counting 36 hr after the explosion. To improve upon this limit, a means of delivering and processing samples much more rapidly must be devised. Technology does exist whereby samples are brought to the surface through a fluid-filled pipe immediately following the explosion. A report by J. D. Brady¹⁴ describes the results of analyses of the contents of tanks containing the prompt sample and fluid (a dilute starch solution in water) in two separate experiments. In the Anacostia experiment, the prompt sampling system collected 10 to 40 kg of solids with a specific Pu content nearly equal to that of good puddle glass samples. These solids were dispersed throughout 6.4 m³ of starch. There remains the development of a system that will successfully retrieve a small part of the solids collected in the tank, process the material chemically, and provide a sample for counting within 1 or 2 hr after zero time. Other techniques for retrieving prompt samples that may eliminate the need for extensive chemical purification are under study at our laboratory.

Perhaps the most vital need in the entire program is the development of a reliable, economical system for large-scale recovery of device debris. While the concentration of samples recovered in the latter phases of Hutch sampling was disappointing, the reaming of the sample hole did allow the retrieval of 500 kg of material. It is likely that additional work on this technique will produce a reliable method for recovering large samples.

Another factor in this problem is the medium in which the device is exploded. A promising approach would be to detonate the next experiment in salt, as was done in the Salmon event. In Salmon, the 5-kt explosion melted 5000 tons of salt. However, the debris was efficiently scavenged from the molten salt by the oxides (CaO, SiO2, Na2O, Fe2O3) present. Hence, the radioactive products from Salmon were contained in about 50 tons of solids. Sample concentrations were at least two orders of magnitude higher than one finds in the ordinary shots in NTS alluvium. An important feature here is that the spherical cavity produced during the explosion does not collapse; the debris is contained in molten salt that runs down to form a lake in the bottom of the cavity and eventually solidifies. Since there is no collapse of the cavity, the molten puddle is not diluted by inert material that falls from the ceiling. Although an explosion in a salt medium would produce the most concentrated debris, as well as material that is particularly amenable to large-scale

chemical processing, ¹⁶ problems with doing experiments other than at NTS are so severe that other possibilities should be considered. Areas do exist within the Test Site where an experiment can be performed in dolomite rock (a mineral consisting primarily of Ca and Mg carbonate), a medium which offers some advantages in chemical processing.

In reviewing the current situation, we see that the device development phase of this program has reached a stage where it is possible to create quantities of certain nuclides that exceed reactor production by as much as ten orders of magnitude. Yet the development of a technology to recover these materials has not proceeded to where satisfactory amounts of this "ore" can be delivered to a chemical processing plant. In planning and executing future experiments, this imbalance will have to be corrected in order to obtain maximum benefits from the program.

ACKNOWLEDGMENTS

The authors wish to acknowledge the efforts of many co-workers at the Lawrence Radiation Laboratory, Livermore, Los Alamos Scientific Laboratory, and Argonne National Laboratory in analyzing samples and recovering actinides from the Hutch debris. We also wish to thank S. F. Eccles, and R. A. Heckman for their helpful discussions in preparing this manuscript.

REFERENCES

- 1. S. F. Eccles, "Production of Heavy Nuclides in Nuclear Devices," to be presented at the ANS Topical Meeting, Engineering with Nuclear Explosives, January 14-16, 1970, Las Vegas, Nevada.
- 2. S. F. Eccles and E. K. Hulet, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-50767 (October 1969), unpublished.
- 3. E. K. Hulet, R. W. Lougheed, and B. J. Qualheim, Lawrence Radiation Laboratory, Livermore, private communication.
- 4. D. C. Hoffman, Arkiv Fysik 36, 533 (1966).
- 5. G. A. Cowan, Synthesis of Neutron-Rich Nuclides, Los Alamos Scientific Laboratory Report LA-3738 (1967); presented at American Chemical Society Meeting, Miami, Florida, April 1967.
- 6. G. A. Cowan, Los Alamos National Laboratory, Los Alamos, New Mexico, and P. R. Fields, Argonne National Laboratory, Argonne, Illinois; private communication
- 7. S. G. Nilsson, et al., <u>Nucl. Phys. A131</u>, 1 (1969); H. Meldner, <u>Arkiv Fysik</u> 36, 593 (1967); and W. J. Swiatecki and W. D. Meyers, <u>Nucl. Phys.</u> 81, 1 (1966).
- 8. R. Quong and J. R. McNabb, <u>Chemical Processing Facility to Recover Actinide Isotopes Produced in Underground Nuclear Explosions</u>, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-50499 (October 1968).
- 9. E. K. Hulet, J. E. Evans, R. Quong, and B. J. Qualheim, The Chemical Recovery of Heavy Elements from Underground Nuclear Explosions, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-14838 (1968)

 Abstract; presented at American Chemical Society Meeting, San Francisco, March-April 1968.
- 10. W. D. Burch, J. E. Bigelow, and L. J. King, Oak Ridge National Laboratory Rept. ORNL-4428 (November 1969) unpublished.
- 11. E. K. Hulet, C. E. Bemis, Jr., and R. W. Lougheed, Lawrence Radiation Laboratory, Livermore, private communication
- 12. K. Wolfsberg, et al., Los Alamos Scientific Laboratory, Los Alamos, New Mexico, private communication.
- 13. A. Ghiorso, M. Nurmia, J. Harris, K. Eskola, and P. Eskola, Phys. Rev. Letters 22, 1317 (1969).

- 14. J. D. Brady, Evaluation of the Prompt Sample Systems Used for the Anacostia and Kennebec Experiments, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-50441 (March 1968).
- 15. D. Rawson, P. Randolph, C. Boardman, and V. Wheeler, J. Geophys. Res. 71, 3507 (1966).
- 16. R. Elson, et al., Coach Processing, A Study of Processes for Concentrating Actinides Formed in a Nuclear Detonation in a Salt Medium, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-14494 (May 1965).