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PREPARATION OF ADDITIONAL SUPPLIES OF $^{244}\text{Pu}^*$

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SUMMARY

In the mid-1970s, gram quantities of the man-made isotope ^{244}Pu were produced in high purity for basic research as a by-product of a program to prepare gram quantities of ^{252}Cf . Because ^{244}Pu is not produced in appreciable quantities in standard power reactor fuel, this nuclide has become extremely useful in the application of isotope dilution mass spectrometry (IDMS), a high-precision analytical technique, to problems of plutonium accountability in support of international safeguards agreements. Although only a small amount (a fraction of a microgram) is needed for each analysis, the present supply of certified nuclear reference standard SRM-996 is expected to last only an additional 3 to 5 years. Three options for producing additional supplies of this rare and valuable material were examined. The options encompassed the production of from 1 to 8.5 g during time spans of from 3 to 10 years. Unit costs ranged from \$2300 to \$7200 per mg of plutonium of $>97\%$ ^{244}Pu . While the paper concludes that the short-term option can be pursued along with further study of the longer-term options, this should not be regarded as an offer or a commitment on the part of the U.S. Department of Energy (USDOE) to furnish the described material.

INTRODUCTION

The only significant source of high-isotopic purity ^{244}Pu has been, and probably for a long time will continue to be, the so-called Cf-I housings. These assemblies initially each contained ~ 100 g of ^{242}Pu and were irradiated at the Savannah River Plant in the early 1970s with the primary objective of producing from 2 to 3 g of ^{252}Cf for the Californium Market Evaluation Program of the U.S. Department of Energy (USDOE) [then the Atomic Energy Commission]. Twenty-one of the original 86 irradiated

housings were processed at the Transuranium Processing Plant (TRU) at the Oak Ridge National Laboratory (ORNL) during 1971, 1972, and 1973, to recover the ^{252}Cf along with by-products ^{244}Cm , ^{249}Bk , and ^{254}Es . A plutonium fraction rich in ^{244}Pu was also recovered and then electromagnetically separated in the ORNL calutrons to produce ~2.25 g of 98.5% ^{244}Pu . That material was designated for use in research programs of the USDOE. A later reworking of the remaining plutonium produced another 1 g of ~97% ^{244}Pu for safeguards programs; this material ultimately became a nuclear reference material, SRM-996, available from the New Brunswick Laboratory of the USDOE.

Isotopic dilution mass spectrometry (IDMS) using ^{244}Pu has proved so useful that we have been asked to consider production of a continuing supply, or at least a replenishment when the current stock of SRM-996 is depleted in from 3 to 5 years. However, the Californium Market Evaluation Program has since been terminated and the current demand for ^{252}Cf is insufficient to justify operation of a Savannah River production reactor for this purpose. Since 1967, californium has also been produced by the USDOE Office of Basic Energy Sciences at TRU and in the High Flux Isotope Reactor (HFIR) at ORNL. The feed for this program is recycled curium and thus no ^{244}Pu is currently being generated in the United States.

In order to replenish the supply of ^{244}Pu , we must return to the initial source: the 65 remaining Cf-I housings which are stored at the Savannah River Plant and the residues from the initial processing campaigns. Approximately 14 grams of unseparated plutonium feed has been recovered at ORNL from the calutron enrichment campaigns on the material originally isolated from the first 21 housings. We have examined various

possibilities for recovering IDMS grade (>97%) ^{244}Pu from these residues and focused on three options.

Calculations of isotope enrichment by calutron separations and by irradiation in the HFIR, which were made in order to determine product yields for the different schemes, are considered to be reasonably accurate. However, the corresponding costs were only crudely estimated, based on the judgment and experience of competent engineers. Insufficient time was available to complete conceptual designs and detailed cost estimates. The results are intended to be used for planning purposes only. They are expressed on a USDOE full-cost recovery basis in FY 1986 dollars. Note that these costs do not correspond to the final price of the ^{244}Pu , but only represent the incremental costs associated with the activities described here for each option. There will certainly be costs associated with certifying the material, and there may be additional charges and credits for the feed materials and any associated coproducts.

Publication of this study does not constitute a commitment or offer by the USDOE to furnish the described material.

SUMMARY OF RESULTS

The three identified options for potential future production of >97% ^{244}Pu and the corresponding yields, schedules, and estimates of the incremental costs are as follows:

- A. Further enrichment campaigns using residues from the initial production campaigns. This option can produce ~1 g of ^{244}Pu in 3 to 4 years at a cost of ~5 M\$.
- B. Transfer of some (~18) additional Cf-I housings to TRU with processing that parallels the original 1971-73 campaigns. This option could produce ~3 g of ^{244}Pu in ~7 years at a cost of ~23 M\$.

- C. Processing of the remaining 65 housings in the Multipurpose Processing Facility (MPPF) at Savannah River, followed by isotopic enrichment of the plutonium at Oak Ridge. This option could produce ~8.5 g of ^{244}Pu in ~10 years at a cost of ~19 M\$.

The cost estimates are scoping in nature and are intended to assess, in 1986 terms, the magnitude of investment required to prepare the desired products from existing feed stocks. The following sections detail the production options considered.

DISCUSSION OF OPTIONS

OPTION A: REWORKING OF RESIDUES FROM THE INITIAL PRODUCTION CAMPAIGNS

Of the two possible sources from which enriched ^{244}Pu may be produced, the most readily accessible is the existing feed batch, FP-37, consisting of 14.2 g of plutonium having an assay of 16.92% ^{244}Pu (see Table 1 for complete isotopic analysis). Table 1 shows the calculated isotopic analysis of the ^{244}Pu produced in a single pass through the calutrons. The assay calculations are perhaps somewhat optimistic, but since assay, rather than throughput, appears to be of prime concern, the calculated values can probably be attained by careful attention to operating conditions.

The calutrons are relatively inefficient devices. Only ~15% of the feed material is collected in the designated collector pockets and the remaining 85% is distributed over the insides of the machine. A thorough washout of the machine can recover up to ~85% of the 85% uncollected feed, and after chemical purification, the plutonium may be processed through the machine again.

Table 1. Assays of calutron feed in Option A (sample FP-37)
and calculated ^{244}Pu product

Isotope	Sample FP-37 feed assay (%)	Product assay ^a (%)
^{238}Pu	0.23	0.02
^{239}Pu	2.10	0.12
^{240}Pu	31.87	1.98
^{241}Pu	4.50	0.30
^{242}Pu	44.38	3.28
^{244}Pu	16.92	94.30

^aSingle-pass calutron enrichment.

Table 2 lists the feed and product quantities computed for a multi-cycle campaign of single-pass separations resulting in a combined product of 1.1 g of plutonium at an assay of 94.30% ^{244}Pu (1.037 g of ^{244}Pu). Multi-cycle campaigns such as this are not unusual when the cost of recovering and purifying unresolved feed from the previous cycle is less than the cost of providing new feed. However, it would have to be an extremely valuable material (like ^{244}Pu) to warrant the effort and expense of a 5-cycle campaign, as illustrated in Table 2. Table 3 presents the calculated assays of the other plutonium products from the ^{244}Pu enrichment campaign and the projected weight of elemental plutonium in each sample for the assumed 5-cycle campaign. In this instance, the quality

Table 2. Quantities produced in calutron campaign using sample FP-37 as feedstock (14.2 g Pu, 16.92% ^{244}Pu)

Cycle	Feed (g Pu)	Recovered feed ^a (g Pu)	Enriched ^{244}Pu ^b (g Pu)	Cumulative ^{244}Pu product (g Pu)
1	14.2	10.25	0.380	0.380
2	10.25	7.40	0.275	0.655
3	7.40	5.35	0.200	0.855
4	5.35	3.85	0.145	1.000
5	3.85	2.75	0.100	1.100

^aAssuming 85% recovery of unresolved feed.

^bAssuming 15% process efficiency.

Table 3. Calculated assays of plutonium products coproduced with the ^{244}Pu

Isotope	Sample FP-37 feed assay (%)	Mass 239 collection (%)	Mass 240 collection (%)	Mass 241 collection (%)	Mass 242 collection (%)
^{238}Pu	0.23	0.13	0.01	0.05	0.01
^{239}Pu	2.10	56.00	0.11	0.50	0.06
^{240}Pu	31.87	17.33	97.00	9.10	1.02
^{241}Pu	4.50	2.06	0.24	74.00	0.17
^{242}Pu	44.38	18.33	2.00	12.68	98.20
^{244}Pu	16.92	6.15	0.64	3.67	0.54
Plutonium content of sample (g)		0.23	2.00	0.37	0.54

and quantity of the coproducts are insufficient to warrant recovery and purification of the materials collected; consequently, these coproducts will not share the costs of the calutron operation.

The assay of the calutron product is estimated to be 94.30% ^{244}Pu , which falls short of the specified 97% minimum. Further enrichment could be achieved by another 5-cycle pass through the calutrons, but this would produce a rather small amount (~0.45 g) of higher-than-necessary assay (>99.9% ^{244}Pu) at further substantial incremental cost. As an alternative, the calutron product may be further enhanced by two reactor cycles (~42 days) of irradiation in the HFIR to burn out lighter isotopes of plutonium and enrich the ^{244}Pu to >97%. This is possible because the ^{244}Pu has by far the lowest neutron absorption cross section of any plutonium isotope; very little ^{244}Pu burnup is expected in the proposed irradiations. Table 4 shows the calculated isotopic composition after HFIR irradiation of the materials listed in Tables 1 and 2 as product. The estimated yield of final plutonium product would be 0.96 g, assuming a loss of 7% for fabrication scrap and reprocessing losses. Of this, an estimated 5% could probably be re-covered for recycle to later campaigns.

In principal it would have been possible to reverse the order and perform an irradiation first to beneficiate the feed, and then produce >97% ^{244}Pu in a single-pass calutron enrichment. However, this arrangement is not as good a match to the relative capacities of the two systems. In particular, the HFIR has a rather poor capability to irradiate the highly fissile isotopes, ^{239}Pu and ^{241}Pu , and much better performance can be achieved after the calutrons have removed the major fractions of these

Table 4. Calculated assay of ^{244}Pu product from sample FP-37 after calutron enrichment followed by reactor irradiation^a

Isotope	Final product assay (%)
^{238}Pu	2.1×10^{-4}
^{239}Pu	2.5×10^{-4}
^{240}Pu	0.013
^{241}Pu	0.011
^{242}Pu	2.51
^{244}Pu	97.5

^aTwo cycles of irradiation in the HFIR.

isotopes. After calutron separation, all of the plutonium produced in Option A could easily be accommodated in one HFIR target instead of the six that would be required prior to calutron separation.

The schedule and cost estimates made for this option are summarized in Table 5. All of these processes are well-known and have been repeatedly demonstrated. However, the alpha-contained segment of the calutron is currently in standby and an estimated two years would be required for funding, planning, staffing, training, etc., prior to initiation of an enrichment campaign. The actual length of the calutron campaign is dominated not by the beam time, but by the length of time required to recover, purify, and recycle the unresolved feed in the 5-cycle campaign. The reactor irradiation and associated processing will consume a comparable amount of time. Thus, the total time from project authorization to delivery of product will range from 3 to 4 years.

Table 5. Schedule and cost estimate for Option A: Reworking current stock

[Feed: Sample FP-37, 14.2 g Pu, 16.92% ^{244}Pu]

Step	Elapsed time (months)	Cost \$(000,000)
1. Calutron Enrichment to ~94.3% ^{244}Pu	6-9 ^a	5.0
2. Fabrication of High Flux Isotope Reactor (HFIR) Target Assembly	3	0.015
3. Irradiation to ~97.5% ^{244}Pu	2	0.03
4. Reprocessing	<u>2</u>	<u>0.03</u>
Total	13-16 ^a	5.1
Product: 0.96 g Pu, 97.5% ^{244}Pu	~\$5,300 per mg Pu	

^aApproximately 2-year planning and preparation period prior to calutron enrichment campaign would delay availability of product until 3 to 4 years after program authorization.

Remarks: Costs are stated in FY 86 dollars on a DOE Full-Cost Recovery Basis. The identified costs are those associated with the processing campaigns and do not include the cost of the feed-stock nor any credit for unrecovered scrap or coproducts.

The costs of this option are almost entirely those of the calutron enrichment program. There are relatively few uncertainties associated with this option.

OPTION B: TRANSFER OF 18 ADDITIONAL CF-I HOUSINGS TO TRU

The other possible source of ^{244}Pu is the unprocessed irradiated housings stored at the Savannah River Plant. Table 6 shows the calculated plutonium contents of the various groups of housings adjusted to October 1, 1987 (grouped by irradiation history and by initial ^{242}Pu loading). The estimated capacity for processing these housings at TRU is ~12 per year. Thus, if the plant were operated on housings half-time (the other half on HFIR target assemblies), ~18 housings could be processed over a period of 2-1/2 years. The choice of 18 housings is somewhat arbitrary and the sub-options of 12 and 24 housings ought to be considered also. If 18 housings were to be processed, the obvious choice would be to pick the six housings of Group III plus 12 from Group I. On this basis, a combined feed of 77 g of plutonium at an assay of 10.31% ^{244}Pu would be recovered at TRU (no processing losses assumed) for use as calutron feedstock. As in Option A, the calculated isotopic compositions of the combined feed and product fractions (single-pass enrichment) are presented in Table 7, and the computed material quantities for a 5-cycle campaign are listed in Table 8. The lower feed assay relative to sample FP-37 results in a lower product assay, which in turn would require three cycles of irradiation in the HFIR to bring the ^{244}Pu to a level above 97% (Table 9). Again allowing for losses of 7%, the final estimated product would be 3.2 g of plutonium.

There are substantial problems that must be resolved in order to carry out a campaign as outlined in Option B. The housings are stored at the Savannah River Plant where there is no present capability to process them.

Table 6. Calculated plutonium assay and content of Cf-I housings irradiated at Savannah River after decay to October 1, 1987

Isotope	Group I 38 housings (wt %)	Group II 21 housings (wt %)	Group III 6 housings (wt %)	Combined total 65 housings (wt %)
^{238}Pu	0.10	0.10	0.06	0.09
$^{239}\text{Pu}^{\text{a}}$				
^{240}Pu	72.57	73.64	69.12	72.46
^{241}Pu	5.41	5.37	1.52	5.12
^{242}Pu	13.18	14.26	13.48	13.34
^{244}Pu	8.74	6.63	15.82	8.99
Pu/housing (g)	5.01	1.43	2.86	3.66
^{244}Pu /housing (g)	0.44	0.09	0.45	0.33

^aContent at discharge approximately nil. Ingrowth from ^{243}Am not calculated here.

Table 7. Calculated Option B assays of calutron feed recovered from 18 Cf-I housings and resulting ^{244}Pu product

Isotope	Feed quantity (g)	Feed assay (%)	Product assay ^a (%)
^{238}Pu	0.06	0.08	0.01
^{239}Pu	-	-	-
^{240}Pu	55.51	71.82	7.45
^{241}Pu	3.51	4.54	0.51
^{242}Pu	10.24	13.25	1.63
^{244}Pu	7.97	10.31	90.40

^aSingle-pass calutron enrichment.

Table 8. Quantities produced in calutron enrichment campaign using plutonium recovered from 18 Cf-I housings (77 g Pu, 10.5% ^{244}Pu)

Cycle	Feed (g Pu)	Recovered feed ^a (g Pu)	^{244}Pu product ^b (g Pu)	Cumulative ^{244}Pu product (g Pu)
1	77.29	55.84	1.32	1.32
2	55.84	40.34	0.95	2.27
3	40.34	29.14	0.69	2.96
4	29.14	21.05	0.50	3.46
5	21.05	15.20	0.36	3.82

^aAssuming 85% recovery of unresolved feed.

^bAssuming 15% process efficiency.

Table 9. Calculated assay of ^{244}Pu product from 18 Cf-I housings after calutron enrichment, followed by reactor irradiation^a

Isotope	Final product assay (%)
^{238}Pu	2.5×10^{-4}
^{259}Pu	1.9×10^{-4}
^{240}Pu	0.13
^{241}Pu	0.08
^{242}Pu	2.54
^{244}Pu	97.3

^aThree cycles of irradiation in the HFIR.

Such capability exists in Oak Ridge, but the housings cannot be shipped to ORNL because no suitable container exists that can meet the current requirements for transportation of radioactive materials. Even if a shipping container were available, substantial modifications to the storage facility at the Savannah River Plant would be required to permit cropping and packaging the housings and loading them into the shipping container. The first two items on the Option B schedule and cost estimate (Table 10) are concerned with moving the housings to ORNL. Once the housings have been transferred into TRU, the remainder of the campaign is straightforward. In Option B, the time required to prepare the plutonium feed sample will exceed the time required to prepare the calutrons for the enrichment campaign and these phases can run concurrently. Thus, the total

Table 10. Schedule and cost estimate for Option B:
Transfer 18 housings to ORNL

[Feed: 18 Cf-I housings, Al clad, containing 77 g of plutonium
(10.3% ^{244}Pu), and 320 g of curium.]

Step	Elapsed time (months)	Cost \$(000,000)
1. Cropping and Packaging Housings	(24)	5
2. Design, Construction and Qualification of Container for Shipment	36	1.5
3. Processing of Housings in TRU	30	11
4. Calutron Enrichment to ~90.4% ^{244}Pu	6-9	5.5
5. Fabrication of HFIR Target Assembly	3	0.015
6. Irradiation to 97.2% ^{244}Pu	3	0.043
7. Reprocessing	<u>2</u>	<u>0.036</u>
	80-83 ^a	23
Product: 3.2 g Pu, 97.2% ^{244}Pu	~\$7,200 per mg Pu	

^aApproximately 7 years after program authorization.

Remarks: Costs are stated in FY 86 dollars on a DOE Full Cost Recovery Basis. The identified costs are those associated with the processing campaigns and do not include the cost of the feedstock nor any credit for unrecovered scrap or coproducts.

time from project authorization to delivery of product is expected to be ~7 years at an overall cost of ~23 million dollars. In this option, 300 g of curium, containing an abundance of heavy isotopes, will also be recovered. This material is of value to the Transplutonium Element Production Program, so some of the costs of Option B might be allocated to the curium product. No credit for this coproduct is indicated in Table 10.

OPTION C: PROCESSING OF THE REMAINING 65 HOUSINGS IN THE MULTI-PURPOSE PROCESSING FACILITY (MPPF) AT SAVANNAH RIVER

Under this option, 238 g of plutonium would be recovered from all 65 of the remaining Ci-I housings at an average assay of 9.0% (see Table 6). The recovered plutonium can be easily shipped to Oak Ridge in conventional shipping packages. The somewhat lower average feed assay will result in a slightly lower single-pass product assay (estimated to be 88%). This in turn will require additional irradiation (estimated 4 HFIR cycles) to achieve >97% assay on the final product. The details were not worked out in Option C, but will clearly parallel those of Options A and B. The key to Option C is the availability of MPPF for processing the housings. This facility is capable of a high throughput of actinides, but at present is in standby. There are plans under consideration to refurbish and reactivate the facility for another program, but even after that, additional modifications would be required to enable MPPF to dissolve the housings. The schedule and cost figures in line 1 of Table 11 are our estimates which include the cost of the modifications plus the operating cost based on some verbal input from SRP personnel relating to the scope of the project. One difficulty is that plans for the MPPF indicate that it will be committed to the other program until 1991, so that ^{244}Pu cannot be made available before 1995. The result of this study is that under Option C, ~8.5 g of plutonium at an assay >97% might be made available sometime around 1995 at a total cost of ~19 million dollars, although some of this cost might be allocated to recovery of the curium in the housings.

Table 11. Schedule and cost estimate for Option C:
Reprocessing of 65 housings at MPPF

[Feed: 65 Cf-I housings, Al-clad, containing 238 g of plutonium
(9.0% ^{244}Pu), and 900 g of curium.]

	Elapsed time (months)	Cost \$(000,000)
1. Upgrade MPPF and Process 65 housings for Plutonium Recovery	18 ^a	12
2. Calutron Enrichment to ~88% ^{244}Pu	9-10	6.5
3. Fabrication of HFIR Target Assemblies (4)	4	0.06
4. Irradiation to >97% ^{244}Pu	4	0.23
5. Reprocessing	<u>4</u>	<u>0.07</u>
Total	39-40 ^a	19
Product: 8.5 g of Pu, >97% ^{244}Pu		~\$2300 per mg Pu

^aMPPF will not be available to start this program until 1991, making material available no sooner than 1995.

Remarks: Costs are stated in FY 86 dollars on a DOE Full-Cost Recovery Basis. The identified costs are those associated with the processing campaigns and do not include the cost of the feedstock nor any credit for unrecovered scrap or coproducts.

CONCLUSIONS

The analyses of the three options have been carried out in sufficient detail to serve as a basis for planning.

It would appear that Option C could provide the greatest quantity of ^{244}Pu product at the lowest unit cost. However, the exclusive reliance on Option C could entail some planning risks. Given that ^{244}Pu is extremely valuable for precise IDMS analysis of plutonium, and hence an integral part of International Safeguards, there should be a continuing supply available to maintain this vital role. The present stock (SRM-996) is probably not sufficient to last until 1995 or 1996. Also, when the time comes, the MPPF may be needed for other high-priority missions, thus delaying further the time when the 8.5 g supply would become available.

Option A has relatively few risks and its schedule appears to be compatible with the expected lifetime of the present supply of SRM-996. The unit cost falls in the mid-range of the three options, although the investment is the least. Further, this option is completely independent of Option C and thus both could be pursued; A for the near term and C for the longer range future (10-15 y hence) when the additional gram produced in Option A would also be consumed.

Option B is intermediate in terms of schedule and quantity made available but it appears to have the highest unit cost. However, this option has the greatest advantage to the Transplutonium Element Production Program and it is possible that a portion of the expense could be allocated to that program. However, this option seriously suffers from the unresolved technical problems associated with the shipment of the housings from Savannah River to Oak Ridge.

In summary, we conclude that the best strategy for extending the supply of ^{244}Pu would be an early exercise of Option A with concurrent studies of costs and schedules associated with moving housings from Savannah River to Oak Ridge. By approximately 1989, sufficient new information should be available (including a better understanding of long term ^{244}Pu needs) to permit a rational decision on Options B and C.

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