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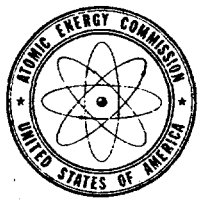
RICHLAND FIVE-YEAR 02 R&D PROGRAM

TRANSPLUTONIUM



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BATTELLE-NORTHWEST
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RICHLAND FIVE-YEAR

O2 R&D PROGRAM

TRANSPLUTONIUM PROGRAM-MISSION 3

June 30, 1968

RICHLAND OPERATIONS OFFICE

ATLANTIC RICHFIELD HANFORD COMPANY

BATTELLE - NORTHWEST

DOUGLAS UNITED NUCLEAR, INC.

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RICHLAND FIVE-YEAR

O2 R&D PROGRAM

TRANSPLUTONIUM PROGRAM-MISSION 3

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R I C H L A N D F I V E - Y E A R

0 2 R & D P R O G R A M

TABLE OF CONTENTS

TRANSPLUTONIUM PROGRAM - MISSION 3

Introduction

This program is directed to the production of higher weight plutonium isotopes and transplutonium isotopes from the irradiation of transuranic elements, particularly plutonium. Since production of Pu-238 from the irradiation of Np-237 and Am-241 is the subject of Mission 4, Pu-238 Program, it is excluded from this Mission. Specific products which are of prime interest are Cm-244 and plutonium containing a high concentration of either the Pu-240 isotope or the Pu-242 isotope. Because of the potential interest in Cm-244 as a heat source in the 1970's, the main emphasis has been to provide the capability for producing this isotope. In support of this program, irradiations have been performed to obtain isotopic buildup rates, and a production and economic calculational model has been prepared for determining production methods and costs of producing Cm-244 in the Richland complex.

The Richland production reactors have particular advantages in producing higher weight plutonium isotopes from the irradiation of plutonium because of a desirable neutron flux spectrum and a high heat removal capability. Specifically, the high temperature thermal neutron flux maximizes the capture-to-fission ratio of the Pu-239 and Pu-241 isotopes; the high heat removal capability provides for the large decrease in heat generation in the plutonium elements with minimum effect on reactor power levels and efficiency.

Based on the developed technology, a program for irradiating 10 kg of plutonium to produce higher weight plutonium isotopes, Am-243, and Cm-244, was recently authorized; preparation for this irradiation and fabrication of the elements is proceeding. The Cm-244 production program and possibly the fast reactor program may require substantial plutonium irradiation by 1970. The authorized irradiation will serve to demonstrate the capability of the Richland production complex to perform such large scale plutonium irradiations.

This program also includes investigation of the attractiveness of producing Cf-252 in the reactor from the irradiation of Cm-244 and Am-243.

Scope and Objectives

The principal objective of this program is to identify and to provide the capability for producing large quantities of Cm-244 and the intermediate plutonium and americium isotopes from the irradiation of plutonium in the Richland reactors.

Reactor operation with an inventory of up to 150 kg to plutonium in C Reactor and up to 300 kg in the K and N Reactors is being examined. Safety and operability problems and the economics of such irradiations are being investigated. The cost and best methods will be determined for meeting potential customer requirements, whether they be for a plutonium fuel containing the heavier plutonium isotopes, for a satisfactory feed material for the Cm-244 program, or to obtain substantial quantities of Cm-244 directly. This also includes integration of the plutonium with other loadings which may be in the reactor to achieve

maximum utilization of the neutrons and provide smooth operational transition as the fissionable isotopes are consumed. Also, concentrated effort is required to predict isotopic buildup in the reactors. In-reactor irradiations have been completed which support the calculational model development. The scheduled irradiation will provide more precise data for establishing effective neutron cross sections and will aid in developing fabrication and reprocessing technology. In the fuels area, efficient manufacturing processes will be developed with primary emphasis, at least initially, on PuAl elements, but investigations will be made of oxide fuels and other carrier materials. Investigation of the feasibility and production capability of producing Cf-252 from the irradiation of Cm-244 will be investigated. It is planned that this study will include test scale irradiations.

The plutonium recycle program in Pacific Northwest Laboratories has provided much of the needed separations technology. However, handling large quantities of plutonium efficiently requires the development of flow sheets to utilize existing facilities. The technical bases will be developed for processing irradiated plutonium fuels of high burnout in Atlantic Richfield Hanford Company-operated facilities, recovering not only the plutonium, but also the trans-plutonium elements, americium and curium. Also, the technology for processing americium targets for curium recovery and for purification of americium and curium products will be provided.

Incentives

The incentives for irradiating plutonium in the reactor are directly dependent upon the market for the ashes of the irradiation. However, since the plutonium would be utilized as an enrichment for other irradiations, the reactor processing costs will be relatively small. At present three potential markets are foreseen: Cm-244, plutonium fuel of specific isotopic content for the fast reactor development program, (fast breeder plutonium fuel is included in mission 1, Basic Production) and high Pu-240 plutonium for exploiting the Phoenix fuel concept. Since the most promising market foreseen is for Cm-244 (either as a heat producing isotope or as feed for producing Cf-252), this program is receiving prime emphasis. However, the developed technology is fully applicable to plutonium processing for other purposes.

In the Cm-244 program, the Richland reactors are ideally suited for producing the Pu-242, which is an intermediate isotope in the production of Cm-244. These advantages are as follows: The higher temperature thermal neutron flux of the graphite moderated reactors maximizes the quantity of feed Pu-239 converted to Pu-242; second, the high heat removal capability of the Richland reactors permits burning of the fissionable isotopes, Pu-239 and Pu-241, in a single irradiation without requiring major expense to maintain full operating efficiency; third, the Richland reactors have a versatile capability for utilizing the neutrons produced in the plutonium fuel at the start of the irradiation for producing a variety of products, and for adjusting the loading as the plutonium burns out without compromising reactor operating economics or neutron utilization efficiency.

Also, a substantial portion of the Pu-242 can be converted to Am-243 and Cm-244 directly without an intermediate separation and fabrication step.

The Richland reactors can also be utilized to obtain a wide range of assays by utilizing the wide ranges of plutonium assays available for initial fuel material, controlling the residence time, and tailoring the neutron energy spectrum by such means as varying the graphite moderator temperature and the makeup of the plutonium element.

The Phoenix fuel program would require plutonium containing a large concentration of Pu-240. Also, productive investigations in the fast reactor program could utilize plutonium with a very high Pu-240 concentration. The neutron energy spectrum in the Richland reactors can be made particularly efficient for converting Pu-239 to Pu-240 with a minimum amount of fissions and for concentrating the Pu-240 with a minimum conversion to Pu-241. Experimental results have confirmed that Pu-240 concentrations in plutonium in excess of 60 percent could be attained. A plutonium irradiation program also complements the plan for considering Richland as a plutonium storage center. Plutonium having a wide range of isotopic assays would be available and could be obtained quickly for fabricating and irradiating in the reactors to meet customer requirements.

The incentives for irradiating Cm-244 to produce Cf-252 in the Hanford reactors are uncertain. The flux level in the Hanford reactors may not be sufficiently high to make such irradiations attractive; however, the effective cross sections for these higher weight isotopes in the Hanford flux spectrum are unresolved. Consequently, a small scale Cm-244 irradiation from which the cross sections and hence the Cf-252 capability can be assessed is considered a desirable extension of the technology.

Progress During the Report Period

The results of the small scale irradiation completed in Fy-1967 were subjected to rigorous analysis to assess the pertinent cross sections and to determine the confidence level to assign to the buildup predictions. The following relative neutron cross-section values were derived from the analysis:

$$\sigma_a^{40}/\sigma_a^{49} = .166 + .483 \times 10^{-5}/N^{40}$$

$$\sigma_a^{41}/\sigma_a^{49} = .965$$

$$\alpha^{49} = .503$$

$$\alpha^{41} = .398$$

$\sigma^i \equiv$ effective neutron microscopic cross section

$\alpha^i \equiv$ capture-to-fission ratio

$N^{40} \equiv$ the concentration of the Pu-240 isotope in units of atom/barn-Cm.

Based on these measured α's, 9.5% of the original Pu-239 would escape fission and thus be converted to Pu-242, a value which matches calculations. Evaluation of the Pu-242 absorption cross section was not possible because of uncertainties and apparent discrepancies in the measured Am-243 concentrations in the irradiated plutonium elements; re-measurements of the Am-243 concentrations are scheduled. However, measured concentrations of Cm-244 were consistent with the predicted values.

The fabrication, loading, and operating plans for irradiating the recently authorized 10 kg of plutonium have been developed. The operating mode is designed to maximize the plutonium conversion rate to the higher weight isotopes in an efficient manner. Also, plutonium of a high Pu-240 content ($\sim$ 20 percent) was obtained and fabrication of the plutonium fuel elements has been initiated. Supporting studies and tests were progressing satisfactorily to meet a charge date in September, 1968. A major modification was made to the Cm-244 production code (CUPID) to more closely match the experimental data. The Cm-244 model is fully operational for planning and optimizing a generalized Cm-244 production program at Richland.

The processes required to decontaminate americium and curium from fission products and to separate the americium from the curium were developed and used to recover an americium and curium product of high purity from the waste which resulted from processing the Shippingport blanket. The processes used at Hanford were a di (2-ethylhexyl) phosphoric acid (D2EHPA) solvent extraction purification of the Am-Cm-Rare Earth crude which had been collected by 50% TBP extraction of the Shippingport waste; followed by chromatographic ion exchange using DTPA (diethylenetriaminepentaacetic acid) and NTA (nitrilotriacetic acid) as complexants for successive elutions. The function of the first elution was to separate the Am-Cm from the lathanide rare earths and the second to separate the Am from the Cm. Subsequent to the recovery campaign, the D2EHPA solvent extraction process was refined, SAMREX vice PAMEX processes, such that separation of the Am-Cm from most of the lanthanides is possible in this step. Further work on separation of Am from Cm by simple solvent extraction technology appears less favorable at this time. In the course of the recovery campaign, considerable technology applicable to the recovery and purification of americium and curium was developed. The technologies used are listed in Table I. Typical concentration curves during the DTPA and NTA elutions are given in Figures I and II. The developed techniques are powerful tools that can be useful in rare earth-actinide processing, with the major unresolved problem being the path of the transcuriums, berklium and californium, and the operating parameters which could optimize the productive capacity of the process. It is likely also that the process could be adapted to the recovery of products from irradiated plutonium, and it is proposed that the plutonium currently planned for irradiation at Hanford be processed in this manner. Although the separation of Cm-242 from Am-241 by the chromatographic process does not appear too favorable due to the high energy output of the Cm-242, it appears that yttrium will follow curium and that yttrium-curium-242 mixtures of reasonable heat output per gram of mixture could be processed successfully. These processes need further development.

Evaluation of Work to Date

The experimental results have confirmed, with minor modifications to neutron cross sections, the validity of the production model in predicting isotope

TABLE I

AMERICIUM-CURIUM TECHNOLOGY

	BNW		ARHCO
	LAB SCALE	PILOT SCALE	PLANT SCALE
CRUDE RECOVERY			
POWER REACTOR WASTE			
SULFATE PPT	X		
TBP EXT.	X	X	(X)
HANFORD PUREX WASTE			
SULFATE PPT	X	X	X
D2EHPA EXT.	X	X	X
LANTHANIDE-ACTINIDE REFINING			
D2EHPA	X	X	(X)
LANTHANIDE-ACTINIDE SPLIT			
CHROMATOGRAPHIC ION EXCHANGE	X	(X)	
ACTINIDE SEPARATION AND PURIFICATION			
CHROMATOGRAPHIC ION EXCHANGE	X	(X)	

(X) TECHNOLOGY USED FOR SHIPPINGPORT Am-Cm RECOVERY

FIGURE I

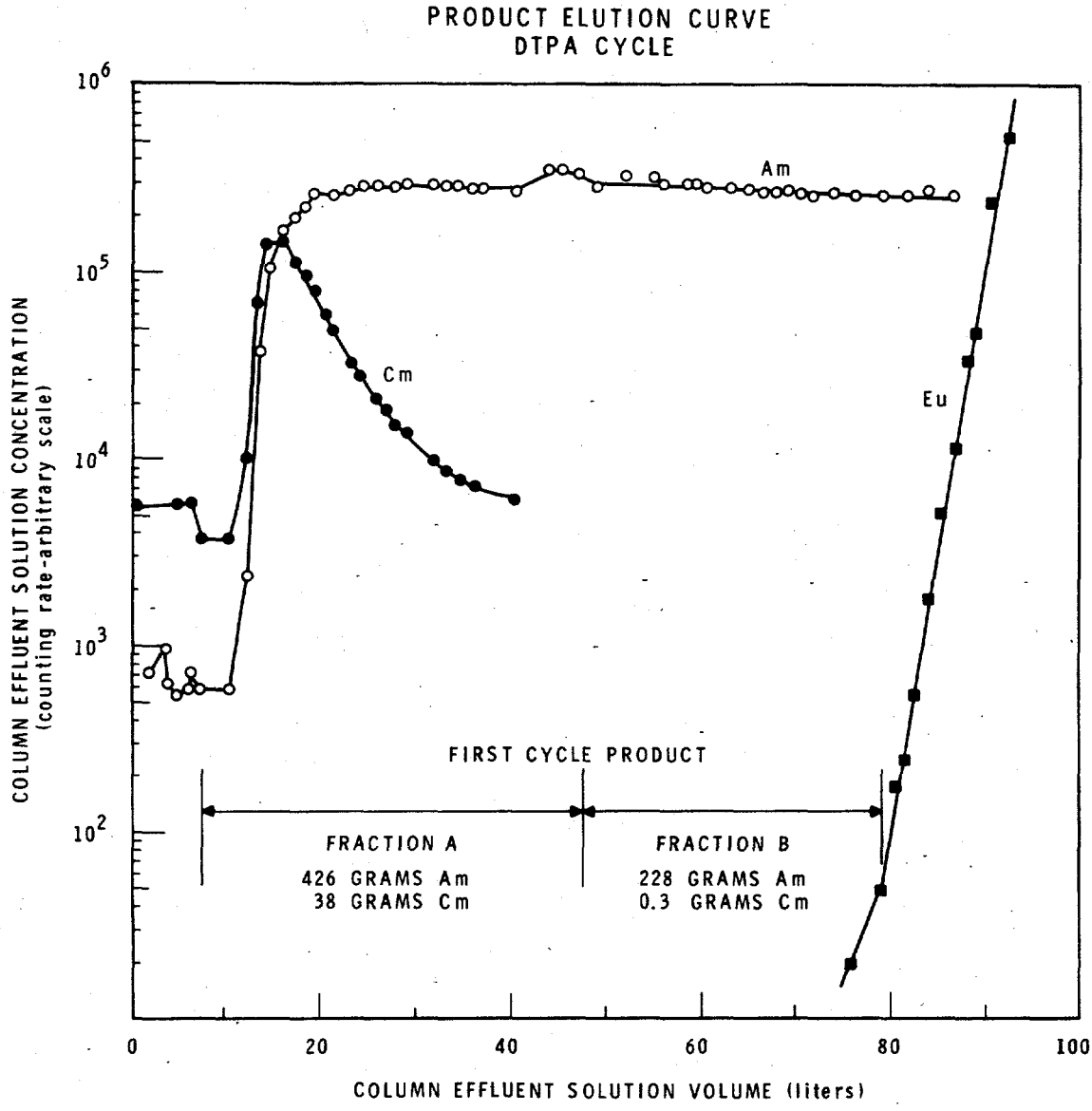
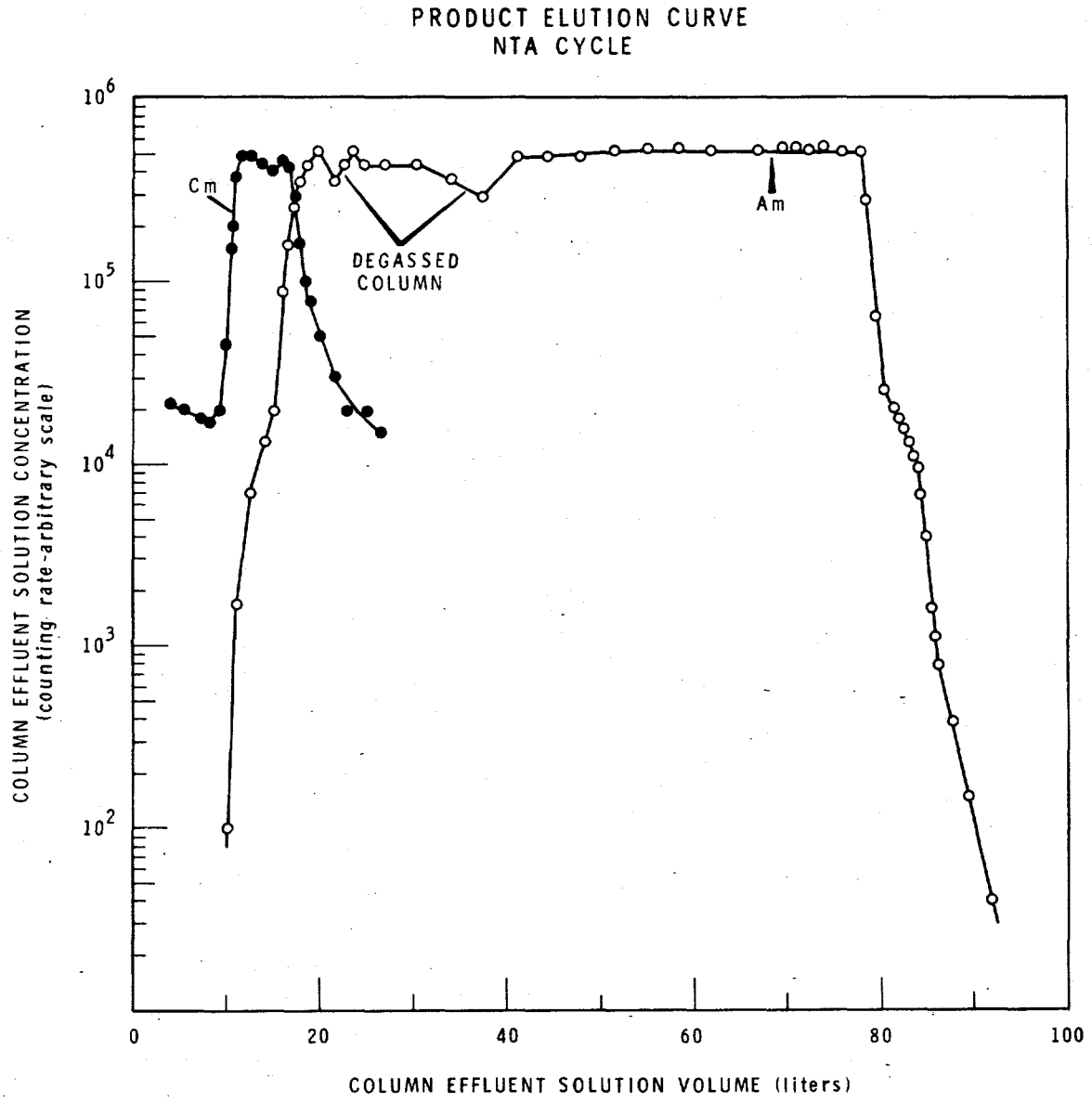


FIGURE II



formation rates up to Cm-244. As noted above, the experimental results confirm the advantages of the Hanford reactors in maximizing the conversion of Pu-239. Development of irradiation techniques, including flux tailoring, has proceeded to the point that a pilot scale plutonium loading and operating modes have been designed which should maximize the transmutation rate of the plutonium and overall reactor production efficiency.

Processes were devised and used to recover and purify americium and curium from waste generated by recovering the plutonium and neptunium from the Shippingport reactor blanket. The significant achievement is that these processes are in a nitric acid system which is compatible with stainless steels normally used in separations plants. Processes under development for use at other sites utilize solvent extraction in a chloride system which requires exotic materials of construction, and a precipitation process for separation of Am and Cm, which is difficult to operate with a high degree of separation. Subsequent to the recovery campaign, the solvent extraction processes were improved (SAMREX vice PAMEX) to the extent that capacity and performance of the final ion exchange process is further enhanced. Incidental to the technology developed, the isotopic composition of the curium appears to be very useful to the atomic energy program, since the Cm-245 content is substantially higher than that of material which has been available. The isotopic composition is as follows:

Cm-243 - 1.49%
-244 - 93.91%
-245 - 3.98%
-246 - 0.62%

Budget Period Plans and Expected Results

The irradiation of 10 kg of plutonium in the K Reactors was recently authorized. Fabrication has been initiated and reactor charging will take place early in FY-1969. Evaluation and optimization of the target fabrication process will be undertaken during the fabrication of the test material. Capability for fabricating Pu-Al target elements on a sustained program will be studied during the present campaign.

Although the irradiation time for this loading has not been defined, it is anticipated that an 18-month to five-year period will be selected, depending on the final use and scheduled need for the material. The maximum Pu-240 content ($\sim$ 60%) will occur with less than six-months irradiation while the maximum Pu-242 content will occur after about two years of irradiation. If it is desired to obtain curium in a single irradiation step, an irradiation time of three to five years is more optimum. Based on previously estimated Pu-239 burnout required prior to transferring the material to a Savannah River high flux charge, and irradiation time of nine months to one year is minimum. Monitor columns of the plutonium elements will be discharged at intermediate exposures to assure satisfactory element performance and accurate prediction of isotopic formation rates.

The detailed evaluation of the Pu-242 absorption cross section from re-measurement of the Am-243 content is scheduled to be completed early in Fy-1969. Also, a comprehensive production and economic study of an optimized curium production

mode at Richland with current technology will be performed in response to indicated needs for the information.

Fabrication of capsules containing small, test quantities of Cm-244 will be completed for charging in K Reactor late in FY-1969. The purpose of this irradiation, which will probably require two to four years' reactor residence time, will be to assess the buildup characteristics of trans-curium isotopes in the Hanford reactors.

No work in the separations area is planned during FY-1969 due to the lack of funds. In FY-1970, plans to process the irradiated plutonium and Cm-244 will be implemented. The irradiated plutonium will be processed by the combination of solvent extraction and ion exchange processes indicated, and if possible, the path of the transcuriums will be determined. The processes will be optimized and other complexing agents tested. Engineering studies on the application of these processes will be made.

Reports Issued

E. J. Wheelwright, F. P. Roberts, L. A. Bray, G. L. Ritter, A. L. Boldt, "Simultaneous Recovery and Purification of Pm, Am and Cm by the Use of Alternating DTPA and NTA Cation Exchange Processes, BNWL-SA-1492, March 6, 1968.

L. A. Bray and G. L. Ritter, "Solvent Extraction Purification of Americium with Di(2-ethylhexyl) Phosphoric Acid," Invention Report, CPDIR-142, May 8, 1967.

L. A. Bray, "Solvent Extraction Purification of Americium with Di(2-ethylhexyl) Phosphoric Acid," BNWL-SA-1446A, October 2, 1967.

L. A. Bray, "Solvent Extraction of the Shippingport Transplutonium and Rare Earth Elements using Di(2-ethylhexyl) Phosphoric Acid," BNWL-CC-1433, December 1, 1967.

G. L. Richardson, "Pilot Plant Demonstration of the SAMREX Process," BNWL-CC-1455, January 18, 1968.

Schedule of Activities

Complete fabrication and initiate irradiation of 10 kg of plutonium - September, 1968.

Complete capsule fabrication and initiate irradiation of small quantity of Cm-244 - FY-1969.

Discharge and analyze first plutonium irradiation monitor column - complete analysis September 1970.

Discharge 10 kg loading - FY-1970-72.

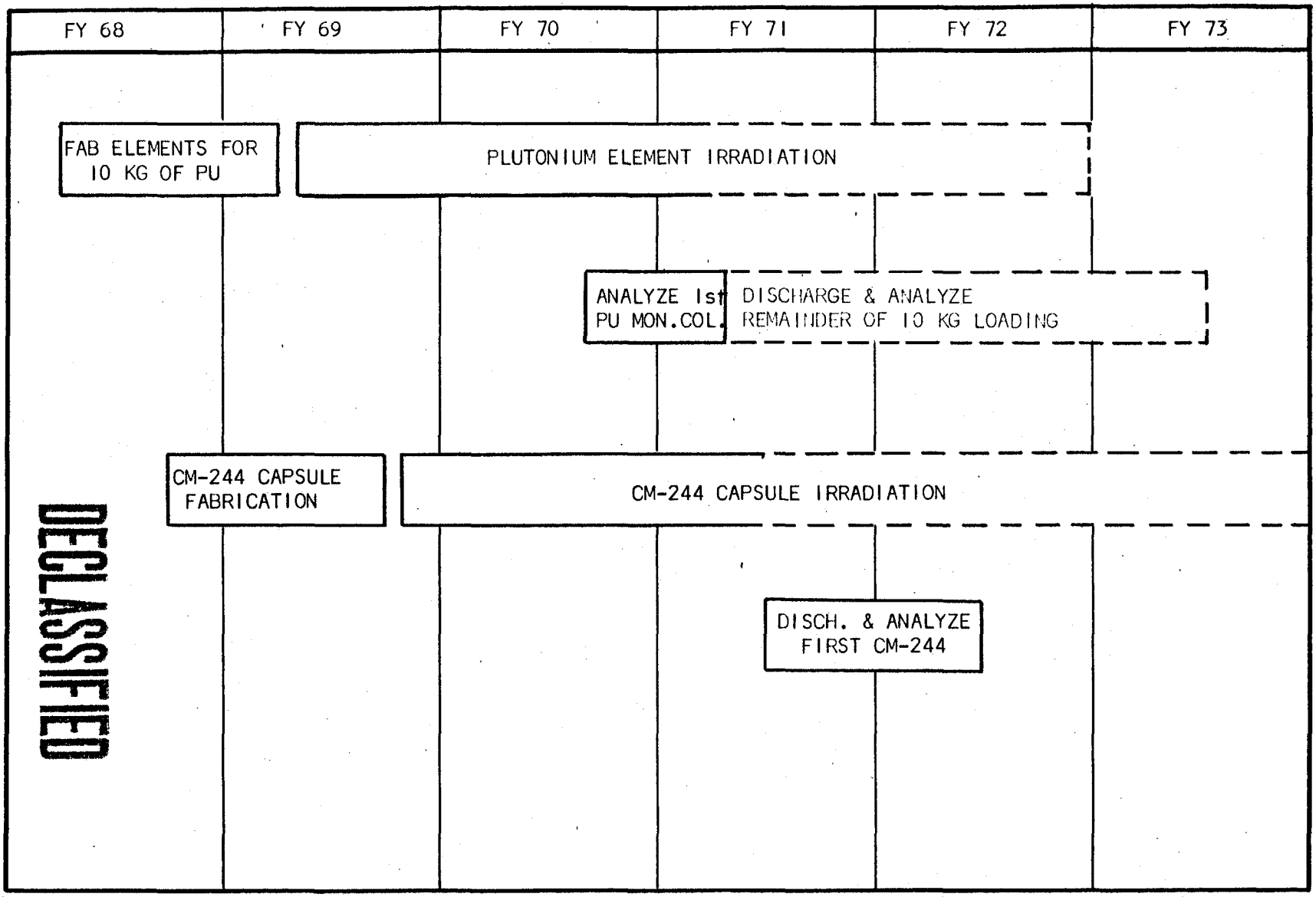
Discharge and analyze Cm-244 irradiation - FY-1971 or FY-1972.

[redacted]

STATISTICAL SUMMARY SCHEDULE

	Fiscal Year				
	1969	1970	1971	1972	1973
Dollars (in thousands)					
DUN	0	0			
ARHCO	0	100			
Total	0	100	130	210	210
Man Years					
DUN	0	0			
ARHCO	0	0.25			
Total	0	0.25			
Equipment					
DUN	0	0			
ARHCO	0	0			
Total	0	0			

[redacted]



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