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PLUTONIUM-238 - PROGRAM ANALYSIS

R. G. Geier

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PLUTONIUM-238 - PROGRAM ANALYSIS

I. PREFACE

The program is designed to use the Richland reactors to increase the national production of Pu-238 in as efficient and economical a manner as practicable while giving attention to any short-term market requirements for Pu-238.

II. INTRODUCTION

The Pu-238 program contains four thrusts:

- A. To enhance neptunium production, it is important that uranium rich in U-236 be put back into the reactors on an expedited basis. It is also important that the materials with the highest U-236 concentrations be placed in the N Reactor because of its more advantageous effective U-236 cross section. However, substantial gains can also be realized by increasing the U-236 concentrations at the other three reactors. Under the present economic ground rules, enhancing U-236 concentration in natural and slightly enriched fuels by re-enriching with uranium of higher enrichment and recycling increases the ultimate cost of the Pu-238. Consequently, the degree to which the U-236 inventories can be maintained in the reactor system requires detailed consideration of the incremental effect on the feed-cycle cost. However, a further source of U-236 is available from slightly enriched uranium irradiated in other reactors, such as that available from NFS. Thus, substantial increases in neptunium production within economic limits can be realized through the prudent management of U-236 supplies which are becoming available.
- B. An efficient process for the irradiation of neptunium to produce Pu-238 has been worked out and demonstrated. It has been determined that maximum production levels are attained when the exposure time is adjusted to yield a plutonium product containing approximately 90 percent Pu-238. The relatively short (50-60 days) out-of-reactor time, demonstrated at Richland, is an equally important factor in obtaining maximum Pu-238 production levels. As a result of achieving the programmatic goals to date, the technology is now established for full-scale production of Pu-238 using

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Richland neptunium with an optimum relationship between the fabrication, irradiation, and reprocessing steps.

- C. Of particular importance is the production of Pu-238 containing minimal Pu-236. Experiments have shown that the substitution of graphite for aluminum as a matrix in neptunium target elements reduces the Pu-236 formation from 0.5 to 0.32 ppm Pu-238 at comparable Pu-238 quality (Pu-238/total Pu). As will be discussed later, additional experiments are being planned to determine the feasibility of producing Pu-238 with an even lower Pu-236 content. It is hoped that the Pu-236 level may be lowered to 0.2 ppm Pu-238.
- D. The technical development of producing Pu-238 from the irradiation of Am-241 will be initiated in FY 1969. The immediate need for this capability is small as the result of the rather minor quantities of Am-241 which will be available in the very near future. However, this production route is particularly attractive because the Pu-238 should contain negligible Pu-236 contaminant and can potentially be processed in a manner to yield very pure Pu-238.

III. INCENTIVES

The operation of the Richland reactors results in the production of substantial quantities of Np-237 as a byproduct. Since the production of Pu-238 is limited by the national availability of Np-237, there is considerable incentive for adapting feasible and economically attractive methods for enhancing the production of Np-237.

The short-term incentive for moving part of the neptunium irradiation to Richland stems primarily from the increased production capabilities. Richland's long-term incentive for participating in the conversion of Np-237 to Pu-238 stems primarily from characteristics of the Richland complex which probably make Richland the optimum site for the ultimate production of Pu-238. Advantages at Richland include a favorable thermal neutron energy spectrum (which results in a significantly lower ratio of Pu-238 destruction neutron cross section to

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Pu-238 formation neutron cross section compared to thermal reactors with lower moderator temperature); the availability of existing facilities for processing the neptunium and Pu-238 with only very small capital modifications for low throughput and only moderate capital modifications for high throughput capability, the capability for a closely integrated fabrication, irradiation, and separation process with relatively low operating cost (which permits minimum out-of-reactor time for recycled neptunium); and the reactor capability to vary the flux level to optimum values. A further important consideration is that due to the type of charge-discharge scheduling and the potential for charging and discharging neptunium elements during operation, the flexibility of the Richland reactors permits nearly continuous flow of the neptunium at a precise exposure.

In addition, there is an advantage to the AEC for capability to process neptunium at a second site. This will provide the flexibility to mesh Pu-238 production with other programs in an optimum manner as the Pu-238 program increases in the future. Also, second site participation can be expected to result in efficiency and cost gains as a result of the competition between the two sites.

Production of Pu-238 from Am-241 provides a potential for substantially increased Pu-238 production capability which will be significant in the middle and late 1970s when quantities of Am-241 become significant from spent power reactor fuels. Being free of the Pu-236 contaminant, Pu-238 produced by this route may be particularly attractive for medical purposes. It is expedient that the technology be developed now to provide well-integrated expansion into this production area.

The most recent market studies indicate the need to produce Pu-238 from Hanford-generated and central station power reactor neptunium by 1971 at maximum levels of 50 kg in 1971, 120 kg in 1975, and 600 kg in 1980. A need for producing clean Pu-238 by FY 1972 has been identified.

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IV. PROGRAM TASKSA. Engineering Section

Although operational charge-discharge of neptunium targets was demonstrated during the previous Np-237 test irradiation, there are undoubtedly areas where improvements and/or optimization could be effected. It is expected that this would be particularly true as the number of tubes requiring operational charge-discharge increased.

B. Process and Programs Section

The Pu-238 program is expected to occupy the time of 0.1 man year per year of program management. In addition, process engineering assistance will be necessary to provide and administer limits as the tests progress. Process physics personnel will be required to monitor and perhaps tailor the flux as the irradiation proceeds. The engineering and physics support are estimated at 0.1 man year/year total.

C. Technology Section

Several Technology Subsections are involved in the tasks described below. These include Reactor Physics Technology Subsection, Fuel and Target Technology Subsection, Reactor Technology Subsection, Nuclear Safety Technology Subsection, and possibly Reactor Materials Technology Subsection.

1. Neptunium Irradiations to Produce Clean Pu-238Graphite and Aluminum Matrix Configuration in a Block Configuration--

It is proposed to irradiate four neptunium target columns in a 2 x 2 array in a K Reactor. Two of the central zone columns would contain Np-Al targets and the other two would contain sintered Np-graphite targets. The target elements would occupy 5 feet of the tube length and contain 40 grams of neptunium per foot.

The block loading would result in a reduction in the fast flux from surrounding fuel with only a slight reduction in the thermal flux level. Thus, a substantial reduction in the amount of Pu-236 produced could be achieved.

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2. Containment of Np-237 in Thin Annulus

A second potential method for reducing the level of Pu-236 is to confine the Np-237 in an annulus between two thin aluminum cylinders. As an example, the Np, NpO_2 , or a mixture of NpO_2 and an inert oxide might be plated on an aluminum tube having an outside diameter somewhat less than required for a given tube. A second aluminum tube having an O.D. appropriate for the tube would then be slipped over the plated section and bonded by a Magneform process. The ends would be welded shut. A graphite rod or tube would be inserted into the central core to provide proper flow balance. There are also mechanical methods of producing the annular sections. However, in either case, providing the technology to produce targets in this concept will require substantial development work.

3. Enhanced U-236 Concentration

In order to verify the gains in Np-237 production by the irradiation of uranium containing an increased concentration of U-236, three tests are in progress:

a. B Reactor

Twelve columns of natural uranium containing 400 ppm U-236 (virgin natural uranium contains less than 10 ppm U-236) were irradiated and discharged. The analytical data have been completed and are being correlated.

b. K Reactor

Six columns of 95-Metal containing 1000 ppm U-236 are being irradiated in a K Reactor. The tubes are scheduled for discharge in March 1969.

c. N Reactor

Twenty-two columns of Mark IC fuel (95-Metal) containing 1145 ppm U-236 are being irradiated in N Reactor. Discharge of the tubes is scheduled for mid-1969, and the tubes will be sent in batches to ARHCO beginning in December 1969.

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4. Americium Irradiation

As pointed out earlier, the irradiation of Am-241 is an attractive supplemental route for producing Pu-238, and the Pu-238 produced from Am-241 should be free of Pu-236 contamination. Following the reprocessing of the Shippingport Reactor blanket, ARHCO separated an Am-Cm fraction from the waste. BNW then separated the americium from the curium. Of the available Am-241-Am-243 fraction, 180 grams were assigned to Hanford, 130 grams for DUN and 50 grams for ARHCO. Because of the tight budget situation in ARHCO, it has been agreed that DUN can count on 150 grams. Such an amount will permit the fabrication of 18 six-inch-long targets--two containing 30 grams Am per foot, two containing 15 grams Am per foot, and two containing 5 grams Am per foot. The targets will be irradiated in the central zone tubes of a K Reactor for periods of 3, 6, and 9 months.

V. CURRENTLY SCHEDULED PROGRAM BUDGET - FY 1969 (Dollars in Thousands)

Engineering	0
Process and Programs	20
Technology	<u>136</u>
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VI. SCHEDULE

Initiate Np Irradiations	FY 1969
Complete Np Irradiations	FY 1969
Initiate Am-241 Irradiation	FY 1969
Complete Am-241 Irradiation	FY 1970
Complete Analysis of Np Irradiations	FY 1970
Document Clean Pu-238 Study	FY 1970
Complete Irradiations for U-236 Study	FY 1970
Document U-236 Study	FY 1971
Have Capability for 50 kg Production Level	FY 1971
Insure Capability for Clean Pu-238	FY 1972
Document Am-241 Study	FY 1972
Have Capability for 120 kg Production Level	1975
Have Capability for 600 kg Production Level	1980

NOTE: None of these tests will be influenced by the power level restrictions at the K Reactors.

VII. COMMENT

From a budgetary standpoint, it appears there is no question that the aluminum matrix-graphite matrix neptunium irradiations and the U-236 study will proceed as planned. Preliminary negotiations with Livermore indicate that they have the capability to encapsulate the americium and would do so for a modest cost. If their estimates hold up after a forthcoming visit to Livermore by our personnel to discuss our requirements in detail, the americium irradiation can also proceed as scheduled. Some funds probably can support the development of the annular concept. However, if the encapsulation cost increases drastically, it may be that either the americium portion of the program or the annular concept development may have to be delayed.

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DUN is presently authorized to divert 500 grams of Np-237 from the stream. A letter requesting the diversion of an additional 700 grams has been prepared for transmission to the AEC to provide for the initial irradiations. If the diversion is approved, there is no problem. However, if it is not, the only irradiations which can be carried out would be a central zone-fringe comparison using graphite-matrix targets.

VIII. REFERENCES

DUN-AOP-44, "Pu-238 Production Program at Hanford," W.K. Woods, et al, 7-28-67 (Secret)

DUN-2913, "Final Report, PT-68 Results of N-Reactor Neptunium Irradiation," D.W. Constable, 7-31-67 (Secret)

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DUN-3305, "PT-NR-95, Irradiation and Analysis of High U-236 Mark IC Fuel," H.E. Jackel, 1-26-68 (Secret)

DUN-4349, "Effective Pu-238 and Np-237 Neutron Absorption Cross Section in N and K Reactors," S.P. Roblyer, 6-10-68 (Secret)

DUN-4354, "K Reactor Pu-238 Demonstration Results, Interim Report PTA-063," J.C. Nosler, 6-13-68 (Secret)

RL-4-4, "Richland Five-Year 02 R&D Program - Plutonium-238," Richland Operations Office, AEC, et al, 6-20-68 (Secret).

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