

3 3679 00062 6012

BNWL-1950 PT4

UC-48

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1974
TO THE
USAEC DIVISION OF BIOMEDICAL AND ENVIRONMENTAL RESEARCH
PART 4 PHYSICAL AND ANALYTICAL SCIENCES

By

J. M. Nielsen
with Major Contributions from
Staff Members of
Radiological Sciences Department
Physics and Instrumentation Department

February 1975

On January 19, 1975, research and development programs of the U.S. Atomic Energy Commission (AEC) became part of the newly formed Energy Research and Development Administration (ERDA). In this report, since it refers to work done in 1974, most references are to AEC programs.

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

• MEDICAL GRADE PLUTONIUM-238 FROM AMERICIUM-241

The objective of this program is to compare and evaluate the economics and the technical practicability of producing medical grade ^{238}Pu by irradiating ^{241}Am recovered from spent power reactor fuel. The ^{241}Am target irradiation could be performed in power reactors.

During 1974 the ALTHAEA computer code was calibrated to match experimental data for americium and curium compositions; irradiation calculations were carried out with the ALTHAEA code to define compositions of spent nuclear fuel and americium target elements; work progressed on development of a computer program for the ^{241}Am irradiation cycle analysis and cost optimization calculations; and, preliminary estimates of ^{241}Am recovery costs from spent power reactor fuel were developed.

Medical Grade ^{238}Pu from ^{241}Am R. W. McKee

For applications in medical devices such as pacemakers and artificial hearts, a supply of ^{238}Pu with less than 0.3 ppm of ^{236}Pu is needed because of the disproportionately high radiation emitted by the ^{236}Pu daughters. Up to the present time, the principal source of ^{238}Pu has been through neutron irradiation of ^{237}Np in AEC reactors and subsequent chemical separation of the ^{238}Pu that was formed by neutron capture. However, a small amount of ^{236}Pu is also formed. Power reactors will be discharging spent fuel containing significant quantities of ^{241}Am which, if separated, could be irradiated to produce a ^{238}Pu product free of ^{236}Pu . The objective of this program is to evaluate the economics and technical practicability of producing medical grade ^{238}Pu by irradiating ^{241}Am recovered from spent power reactor fuel. The ^{241}Am target irradiation could be performed in power reactors.

The scope of the program includes: 1) projecting the availability of ^{241}Am , 2) identifying the facilities and cost of americium recovery, and 3) identifying the optimum target irradiation and processing cycle and cost of producing ^{238}Pu .

During 1974 work on this project consisted of: a) calibrating the physical constants in the ALTHAEA computer code to produce irradiation calculations that match experimental data for americium and curium compositions, b) irradiation calculations for spent fuel and americium target compositions, c) development of the AMRAD computer program for analyses of the ^{241}Am target irradiation cycle, and d) analyses and preliminary estimates of the cost for recovering ^{241}Am from spent power-reactor fuel.

Analysis of the feasibility of producing ^{238}Pu from americium recovered from spent power reactor fuel is dependent on irradiation burnup calculations at two points: 1) defining the

quantities of the americium isotopes in the spent fuel and 2) defining the composition of the americium target elements after target irradiation. The ALTHAEA¹ computer code has been used at PNL to carry out these types of calculations. The physical constants in the code have been calibrated to match analytical data for spent fuel compositions of power reactor fuels and results are quite reliable for the isotopic compositions up through the plutonium isotopes. However, the physical constants related to the production of the higher actinides including americium and curium have not been as well calibrated. For this reason an effort was made to improve the calibrations related to the americium and curium calculations and to establish the magnitude of the uncertainty in these calculations.

The resulting differences between calculated and experimental compositions were still relatively large for these isotopes and point out the desirability of further improving the calibration. Results indicate an uncertainty of 25 to 35% in the calculated americium and curium compositions. Calculated americium and curium compositions for PWR fuel are shown in Table 5.1.

A series of ALTHAEA cases were run to develop target irradiation data over the range of conditions believed to be practical. The three primary variables are irradiation time, americium isotopic composition and ameri-

cium density in the targets. Results for one case that is believed to be well within feasible limits are shown in Table 5.2.

In addition to the transmutation reactions, there will also be some fission reactions taking place in the target elements. To evaluate the feasibility of early reprocessing of the target elements, knowledge of the fission product content of discharged targets is also essential. The ORIGEN computer code² is being used for this purpose. This code provides a complete list of all of the fission products as well as the transuranium isotopes present at selected times following discharge. Physical constants in this code were adjusted to produce results that coincide with the ALTHAEA results.

The principal nuclear reactions involved in americium target irradiations are illustrated in Figure 5.1. The entire americium cycle for production of ²³⁸Pu is illustrated in Figure 5.2. A computer program based on this cycle and called AMRAD is being developed to calculate production rates, product compositions, production costs, etc. It will be used

TABLE 5.1. Americium and Curium Content of PWR Fuel at 32,000 MWd/MT Exposure.

Isotope	U Enriched Fuel g/MT		1st Pu Recycle g/MT	
	150-Day Cooling	365-Day Cooling	150-Day Cooling	365-Day Cooling
²⁴¹ Am	64	97	484	626
^{242m} Am	0.6	0.6	8	8
²⁴³ Am	79	79	872	872
²⁴² Cm	8.2	3.3	71	29
²⁴³ Cm	0.1	0.1	1.3	1.3
²⁴⁴ Cm	17.1	17.3	391	382

TABLE 5.2. Americium Irradiation Products.

Irradiation Conditions:

11-Month PWR Irradiation

Target is 33% ²⁴¹Am/67% ²⁴³Am

Target Density is 0.3 g Am/cm³

Product from First Reprocessing After 3 Months Decay:

²³⁸Pu Yield = 9 g/100 g Americium

²³⁸Pu Purity = 72%

²³⁶Pu Content = 4 Parts Per Billion*

Product from Second Reprocessing 18 Months After First Reprocessing:

²³⁸Pu Yield = 5 g/100 g Americium

²³⁸Pu Purity = 90%

²³⁶Pu Content = 0

*From neutron capture by ²³⁷Np from alpha decay of 433-year ²⁴¹Am.

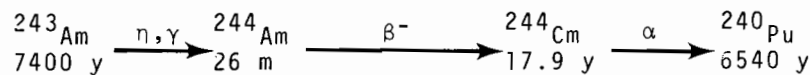
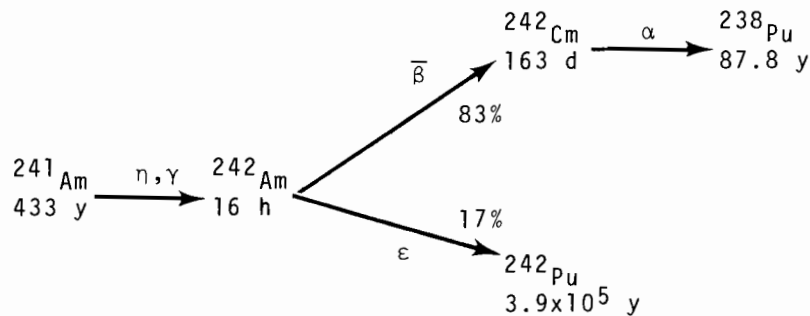


FIGURE 5.1. Americium Irradiation Reactions.

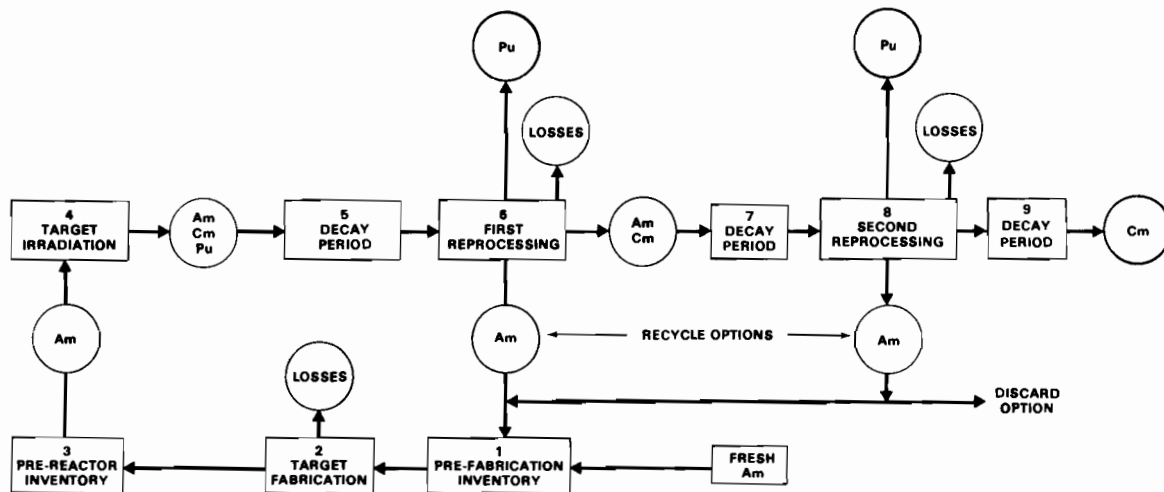


FIGURE 5.2. Americium Irradiation Cycle.

for optimizing the cycle and evaluating the various alternatives illustrated in Figure 5.2. One option not illustrated in Figure 5.2 is the number of times the americium is to be recycled. One recycle appears almost certainly desirable. At some point, however, the ${}^{241}\text{Am}$ will be too depleted to be useful for recycle.

A recent study³ by the General Atomic Company of ${}^{244}\text{Cm}$ and ${}^{237}\text{Np}$ recovery from spent-power-reactor fuels for AEC's Space Nuclear Systems Division appears to provide most of

the information needed to develop costs for recovery of americium from reactor fuel reprocessing waste. The process flow sheet developed for this study provides for processing all of the high-level liquid waste from the Barnwell Nuclear Fuel Plant, after a 3-year storage period. The reference flowsheet for curium recovery consists of a solvent extraction step and an ion exchange step. The americium follows the curium in the process and is not separated until the final elution step. A pure americium fraction

could be collected in place of or together with the curium at no additional cost. Thus the costs developed for curium recovery also apply to americium recovery. If both elements were recovered, the cost could be shared.

Preliminary price estimates for ^{241}Am recovery were developed based on limited details from the above study and will be revised when additional details are obtained. The principal conclusion that can be drawn is that when artificial heart

use is sufficient to require near capacity recovery of ^{241}Am (around 1990), it should be available at \$50 to \$100/g (1974 dollar values with actual escalated cost possibly several times larger). The low end of this price range would require co-product recovery and sale of ^{244}Cm . These ^{241}Am prices should result in acceptable ^{238}Pu production costs. The ^{241}Am may cost substantially more during the introduction period of artificial hearts when production rates are low.