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STUDY OF HIGHER ISOTOPE PRODUCTION
AS A USEFUL BY-PRODUCT IN FAST POWER REACTORS

MASTER

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by

John T. Madell

SUMMARY

A study of the possible economic value of the higher isotopes which are formed as by-products in a fast power reactor was made. A literature search revealed that it is technically feasible to produce and separate higher isotopes. The production rates of two important higher isotopes, Np^{237} and Am^{241} , were calculated as a function of the power output of the reactor. The calculations showed that a credit (sale price minus separation cost) of \$500/gm of Np^{237} and a credit of \$1000/gm of Am^{241} will each reduce the power costs by one mil/kw hr. The present sale price of Np^{237} and Am^{241} produced in thermal reactors is \$500/gm and \$1500/gm, respectively. Since the production rates are lower and the separation costs are higher for the isotopes in a fast reactor than in a thermal reactor, little or no reduction in the power costs of a fast reactor is presently expected from the sale of Np^{237} and Am^{241} . However, if the demand for Np^{237} and Am^{241} should exceed the production from thermal reactors, fast power reactors would be an attractive source of the isotopes.

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STUDY OF HIGHER ISOTOPE PRODUCTION
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Introduction

Alpha emitters are used to supply the power for some Space Nuclear Auxiliary Power (SNAP) devices. Two alpha emitters which find application in this field are Pu^{238} and Cm^{242} . These isotopes can be produced as by-products in a nuclear power reactor. Although the rates of production of Pu^{238} and Cm^{242} are of second order [successive neutron captures or (n,2n) reactions], the high value of the isotopes justifies making a cost estimate of their production. The production of the isotopes was investigated for fast reactors because fast reactors may have an advantage in producing isotopes requiring a (n,2n) reaction. At the request of the A.E.C. a study of the production of Pu^{238} and Cm^{242} as by-products in fast power reactors was undertaken. The object of the study is to determine whether the production of the isotopes is technically feasible and what economic incentive exists for the sale of these alpha-emitting isotopes. The economic incentive is expressed as a reduction in power costs.

Mode of Production

Both plutonium and uranium are considered in this study as possible fuels for fast reactors. The mode of formation of Pu^{238} from U^{235} and U^{238} is given in Figures 1 and 2, respectively. The steps in the production of Cm^{242} from the plutonium isotopes are illustrated in Figure 3.

The Pu^{238} formed in the uranium fuel elements is diluted with Pu^{239} (also a little Pu^{240} and Pu^{241}) produced from U^{238} . To obtain pure Pu^{238} from the mixture

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

an isotopic separation would be required. Since isotopic separations are extremely expensive, an intermediate product, Np^{237} , is removed by chemical means before it is transformed into Pu^{238} . The production of Pu^{238} is therefore accomplished in two irradiations. The Np^{237} is produced from U^{235} and U^{238} and then separated from the uranium and fission products. The Np^{237} is then irradiated to give Pu^{238} . Alternatively, the Np^{237} can be sold after the chemical separation from the irradiated uranium. In this study, it is assumed that the Np^{237} is sold because the price of Np^{237} is available while that of Pu^{238} is not. The production of Pu^{238} by a $(n,2n)$ reaction with Pu^{239} is not included because of the required isotopic separation.

Cm^{242} with Am^{241} as an intermediate isotope is produced from a plutonium fueled reactor. In a plutonium breeder reactor with a "dirty" plutonium feed, the plutonium isotopes, Pu^{239} , Pu^{240} , Pu^{241} , and Pu^{242} , may reach equilibrium values in the fuel cycle. This situation was assumed in the study and therefore Pu^{241} can be considered as the starting isotope for the production of Cm^{242} . As was true for the case of Pu^{238} , alternative methods are available in calculating the values of the Cm^{242} production. Am^{241} may be separated and either sold or re-irradiated separately, or it may be left in the fuel to yield Cm^{242} . Since the price of Cm^{242} is not available, it is not possible to select the most economical alternate. Therefore, each possibility is considered.

Calculation of Production

The production rate of the J^{th} isotope, N_J , in a straight chain is

$$\frac{dN_J}{dt} = P_J N_{J-1} - R_J N_J, \quad (1)$$

where P_J is the production rate of N_J ,

R_J is the removal rate of N_J .

The general solution of equation (1) is

$$N_J = \sum_{k=1}^J \left\{ \sum_{j=k}^J N_{ok} e^{-R_j t} \pi \frac{P_q}{R_q - R_j} \right\}, \quad (2)$$

where N_{ok} is the quantity of N_k at zero time. The removal term R_j consists of the product of the absorption cross section and the flux ($\sigma_a \phi$) and a decay constant (λ). The production term is either the product of the capture cross section and the flux ($\sigma_c \phi$) or the decay constant (λ). Five energy groups are used to describe the cross sections and the flux.

The production of the isotopes is calculated by a computer code. The cross sections, decay constants, flux, initial concentrations and the time are given as input data. The removal and production terms are first calculated and then the production of the isotopes is obtained from equation (2).

Nuclear Data

The data are divided into two types, nuclear data (cross sections and decay constants) and reactor operating data (flux, irradiation time and fuel cycle information).

The values of the fast absorption and the fast fission cross sections of uranium and plutonium isotopes have been measured several times and the published values are considered to be accurate enough for this work^(1, 2, 3). Data are available for the fast fission cross sections of Np^{237} and Am^{241} , but no data have been found in the literature for the fast fission cross sections of the other americium and curium isotopes nor for the fast absorption cross sections of any of the neptunium, americium, and curium isotopes. Discussions with the scientists attending the "Annual Review of AEC Transplutonium Production Program" (held at ANL on January 23, 1962) confirmed that this lack of data exists. In view of the lack of data, a method

was devised to obtain the maximum and minimum expected values of the cross section for these isotopes.

The method is based on the classical theory of neutron interaction. The classical theory states that the cross section is proportional to the nuclear radius which is in turn proportional to the atomic number to the two-thirds power. The theory is valid for neutrons having an energy greater than a few Mev. Although the average neutron energy in fast reactors is a few kev, the methods afford a means of obtaining approximate values. The maximum and minimum expected values of $\sigma(n,\gamma)$ are obtained by plotting the published values of the $\sigma(n,\gamma)$'s of the isotopes having about the same nuclear radius. The maximum and minimum cross section in each of the five energy groups are determined (see Figure 4) and assigned to all of the Np, Am, and Cm isotopes as $\sigma(n,\gamma)_{\max}$ and $\sigma(n,\gamma)_{\min}$.

The fission cross section is dependent upon the pairing energy of the nuclide. The dependency is seen in the available fission cross sections of even Z, odd N nuclides, of even Z, even N nuclides, and of odd Z, odd N nuclides in Figures 5, 6, and 7, respectively. As was done for the capture cross sections, the maximum and minimum values of the fission cross section are determined in each energy group for each type of nuclide. These values are assigned as the maximum and minimum expected values of $\sigma(n,f)$ to the Am and Cm isotopes having the same pairing energy. For example, the values of $\sigma(n,f)$ for Cm²⁴² are assumed to be equal to the maximum and minimum values of $\sigma(n,f)$ for the even Z, even N nuclides.

This method of obtaining the maximum and minimum expected values of the fast cross sections seems to be justified by the fact that all of the experimental values plotted in the four figures lie within a small range of values. With this range of cross section values, the maximum and minimum production rates are calculated. The decay constants of the nuclides of interest were taken from Sullivan's

TABLE 1. VALUES OF THE CAPTURE AND FISSION CROSS SECTIONS
AND THE DECAY CONSTANTS OF THE NUCLIDES OF INTEREST

Nuclide	$\sigma(n,\gamma)$ barns						$\sigma(n,f)$ barns						$\lambda(\text{sec}^{-1})$
	Comment	Gp I	Gp II	Gp III	Gp IV	Gp V	Comment	Gp I	Gp II	Gp III	Gp IV	Gp V	
U^{235}	exp. values	.02608	0.1438	0.2625	0.4084	0.9139	exp. values	1.280	1.240	1.331	1.694	2.750	$\sim 3 \times 10^{-17}$
U^{236}	exp. values	.03506	.2924	.2888	.3978	.8567	exp. values	0.9432	0.2620	0.0	0.0	0.0	$\sim 1 \times 10^{-15}$
U^{237}			negligible						negligible				1.188×10^{-6}
U^{238}	exp. values	.01706	.1350	.1344	.2189	.5015	exp. values	.5855	.01482	0.0	0.0	0.0	$\sim 2 \times 10^{-18}$
Np^{237}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	exp. values	1.600	1.301	0.250	0.0	0.0	$\sim 1 \times 10^{-14}$
Np^{238}			negligible						negligible				3.82×10^{-6}
Pu^{241}	exp. values	.01728	.09688	.1739	.2753	.6541			not required				1.663×10^{-9}
Pu^{242}	exp. values	.02811	.2289	.2438	.4366	.7937	exp. values	1.450	1.210	0.1322	0.0143	0.0	5.78×10^{-14}
Pu^{243}			negligible						negligible				3.865×10^{-3}
Am^{241}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	exp. values	1.40	.06745	0.0	0.0	0.0	4.75×10^{-11}
Am^{242}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	max. min.	2.00 1.20	2.00 1.20	2.075 1.256	2.80 1.70	4.20 2.50	1.203×10^{-5}
Am^{243}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	max. min.	2.0 1.00	1.50 0.329	1.60 0.0	0.0 0.0	0.0 0.0	2.75×10^{-12}
Am^{244}			negligible						negligible				4.44×10^{-4}
Cm^{242}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	max. min.	2.00 0.60	1.382 .353	0.220 0.0	0.030 0.0	0.0 0.0	4.94×10^{-8}
Cm^{243}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	max. min.	2.00 1.20	2.00 1.20	2.075 1.256	2.80 1.70	4.20 2.50	6.27×10^{-10}
Cm^{244}	max. min.	.0630 .0257	.1653 .0794	.2625 .0120	.4519 .3779	1.006 .6612	max. min.	2.00 0.60	1.382 0.353	0.220 0.0	0.030 0.0	0.0 0.0	1.19×10^{-9}
$\sigma(n, 2n)$ barns													
U^{238}	exp. values	.0250	0.0	0.0	0.0	0.0							

Trilinear Chart of Nuclides⁽⁴⁾. A list of the nuclear data for the pertinent isotopes of U, Np, Pu, Am, and Cm is given in Table 1.

Reactor Operating Data

In selecting the reactor operating data and therefore the reactor, consideration was given to picking a reactor which will produce commercial power in the near future. The only fast reactor which fits this requirement is the Enrico Fermi reactor. The EBR-II was not considered because it is an experimental reactor rather than a power reactor and, therefore, will not have a regular operational and fuel cycle schedule. Physics calculations for the Enrico Fermi reactor have been completed for the uranium core. This core is of interest in calculating the production of Np^{237} and Pu^{238} . It is conceded that other fast power reactors, for which preliminary calculation have been carried out, may be better suited to the production of Np^{237} , but the study was restricted to a fast power reactor which should be operating in the near future. The operating data of interest to the study are

- (1) power - 300 MWt, 100 MWe
- (2) average core flux - 5×10^{15} n/cm²/sec
- (3) average core burnup - 1%
- (4) irradiation time - 50 days
- (5) specific core power - 550 kw/kg of U^{235}
- (6) average core flux spectrum

Group	Energy	Flux
I	1.35 - 10.0 Mev	5.75×10^{14} n/cm ² /sec
II	0.50 - 1.35	10.00×10^{14}
III	0.18 - 0.50	14.25×10^{14}
IV	0.067 - 0.18	12.00×10^{14}
V	0 - 0.067	8.00×10^{14}

The fuel cycle for the uranium core is shown in Figure 8. After each irradiation, the plutonium, neptunium, and fission products are removed. The reprocessed fuel

contains U^{235} , U^{236} , and U^{238} . It is assumed that the U^{237} and Np^{238} have entirely decayed before the fuel is reprocessed. U^{235} and U^{238} are added to the returning fuel to compensate for the burnup of these isotopes.

Am^{241} and Cm^{242} are produced in a plutonium fueled reactor. Presently no reactor physics calculations have been published for the operation of the Enrico Fermi reactor on a plutonium cycle. A fuel cycle code, CYCLE⁽⁵⁾, was used to obtain the operating conditions of a metal plutonium fast breeder reactor with a "dirty" plutonium feed. Although the reactor for which the fuel calculations were performed is not identical to the Fermi reactor, it is felt that the production rates of Am^{241} and Cm^{242} per kilogram of fuel would be about the same in both reactors.

The pertinent operating conditions are

- (1) power - 500 MWt, 165 MWe
- (2) average core flux - 5×10^{15} n/cm²/sec
- (3) equilibrium fuel composition entering reactor

U^{238}	1852. kg
Pu^{239}	266.7
Pu^{240}	5.7
Pu^{241}	7.0
Pu^{242}	1.4

- (4) power density - 1 MWt/l of core
- (5) average core spectrum

Group	Energy	Flux
I	1.35 - 10.0 Mev	5.758×10^{14} n/cm ² /sec
II	0.5 - 1.35	11.510×10^{14}
III	0.18 - 0.50	16.468×10^{14}
IV	0.067 - 0.180	9.751×10^{14}
V	0.0 - 0.067	6.513×10^{14}

- (6) average irradiation time - 50 days

In the fuel cycle for this reactor (Figure 9) the fission products are removed at the end of each irradiation. The neptunium, americium, and curium isotopes are removed when their concentration is large enough for an economical separation.

Np²³⁷ Production

The concentration of Np²³⁷ along with that of the intermediate isotopes produced from U²³⁵ and U²³⁸ is plotted in Figures 10 and 11, respectively, as a function of the irradiation time. The minimum expected absorption cross section for Np²³⁷ was selected to give the maximum production of the isotope. In the first 100 days of irradiation the majority of the Np²³⁷ is produced from U²³⁸ but as the amount of U²³⁶ increases, more of Np²³⁷ comes from the U²³⁵, U²³⁶ chain. Since the concentration of Np²³⁷ is small for the first 250 days (5 irradiation cycles), it is probably too expensive to remove the Np²³⁷ and, therefore, no calculations of the Np²³⁷ production were made until fuel had been cycled five times. By this time an appreciable amount of U²³⁶ has built up in the system. The amount of Np²³⁷ produced per cycle in a uranium fueled Fermi reactor is tabulated in Table 2a. The total production of Np²³⁷ per cycle increases by ~30 gms for the first 10 or 20 cycles so that data in Table 2 may be easily extended. The situation of equilibrium production of Np²³⁷ (~ 2500 gm/cycle) is not realistic because the U²³⁶ concentration would prevent criticality.

Am²⁴¹ and Cm²⁴² Production

The production of Am²⁴¹ and Cm²⁴² in a plutonium fueled fast reactor is plotted as a function of the irradiation time in Figure 12. The choice of cross sections was made to give the maximum production rate of Am²⁴¹ and Cm²⁴². Two different fuel cycles are considered. In the first type of cycle, Cycle A, the Am²⁴¹ and Cm²⁴² are removed after each irradiation and in the second, Cycle B, the Am and Cm are removed after every other irradiation. In both cases, a 100 day cooling time and a 50 day reprocessing and refabrication delay time are assumed. Results of the two types of fuel cycles are given in Table 2b. Note that more Am²⁴¹ and less Cm²⁴² are produced in Cycle A than in Cycle B. Which cycle is more economical to operate depends upon the relative worth of Am²⁴¹ and Cm²⁴².

Table 2(a)

Cycle#	Production of Np^{237} /cycle			
	$\text{Np}^{237}/\text{U}^{235}$	$\text{Np}^{237}/\text{fuel}$	Total Np^{237} (grams)	$\text{Np}^{237}\text{mg/kw hr electric}$
6	0.4785×10^{-3}	0.1324×10^{-3}	232.1	2.175×10^{-3}
7	0.5406×10^{-3}	0.1493×10^{-3}	262.2	2.457×10^{-3}
8	0.5959×10^{-3}	0.1642×10^{-3}	289.0	2.709×10^{-3}
∞	5.277×10^{-3}	1.259×10^{-3}	2559.0	23.99×10^{-3}

Table 2(b)

Total Grams of Isotope										milligrams/kw hr electric	
<u>Before Irrad.</u>		<u>After Irrad.</u>		<u>After Cooling</u>							
Am		Cm		Am		Cm		Am		Cm	
Cycle "A"											
1	0.0	0.0	50.0	0.143	150.8	0.143	0.754 x 10 ⁻³	0.0715 x 10 ⁻⁵			
2	50.4	0.0	99.8	0.424	200.6	0.424	1.003 x 10 ⁻³	0.212 x 10 ⁻⁵			
3	50.4	0.0	99.8	0.424	200.6	0.424	1.003 x 10 ⁻³	0.212 x 10 ⁻⁵			
		etc.									
Cycle "B"											
1	0.0	0.0	50.0	0.143	150.8	0.143	—	—			
2	200.2	0.143	247.9	1.146	348.7	1.146	0.872 x 10 ⁻³	0.287 x 10 ⁻⁵			

3 and 4 are a repetition of cycles 1 and 2

Economic Incentive for Isotope Production

The methods of separation of Np, Am, and Cm have been developed for small-scale operations. It is, therefore, technically feasible to separate the isotopes from fuel irradiated in fast power reactors. Presently there is no information on which an accurate cost estimate of a large-scale separation can be based. It is, therefore, not possible at this time to determine the economic return that can be realized by separating and selling these isotopes.

However, the influence of the profit from selling the isotopes on the power costs can be calculated. The difference between the sale price and separation cost is called the CREDIT. The reduction in power costs is plotted against the CREDIT for the Np^{237} , Am^{241} , and Cm^{242} in Figures 13, 14, and 15, respectively. It is seen that a credit of $\sim \$500/\text{gm}$ Np^{237} , a credit of $\sim \$1000/\text{gm}$ of Am^{242} , and a credit of $\sim \$400,000/\text{gm}$ of Cm^{242} are each equivalent to a reduction of 1 mil/kw hr (electric).

The A.E.C. has recently published the prices for Np^{237} ($\$500/\text{gm}$) and Am^{241} ($\$1500/\text{gm}$). It seems likely that these figures represent the cost of separation and marketing with little or no profit included. Since the present means of producing these isotopes most likely allows a cheaper separation process than could be used for the isotopes produced in a fast reactor, the cost of separating Np^{237} and Am^{241} produced in fast reactors is equal to or greater than the sale price. The production and separation of the isotopes in a fast reactor does not, therefore, look economical.

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FIG. 1
MODE OF FORMATION OF
 Pu^{238} FROM U^{235}

FIG. 2
MODE OF FORMATION OF
 Pu^{238} FROM U^{238}

FIG. 3
MODE OF FORMATION OF Cm^{242} FROM
THE PLUTONIUM ISOTOPES

FIG. 4

THE MAXIMUM AND MINIMUM VALUES OF $\sigma(n, \gamma)$ FOR HEAVY ($A \geq 232$) NUCLIDES AS A FUNCTION OF THE NEUTRON ENERGY

FIG. 5
THE MAXIMUM AND MINIMUM VALUES OF THE FISSION CROSS SECTION OF
THE EVEN Z, ODD N NUCLIDES AS A FUNCTION OF THE NEUTRON ENERGY

FIG. 6
THE MAXIMUM AND MINIMUM VALUES OF THE FISSION CROSS SECTION
OF THE EVEN Z, EVEN N NUCLIDES AS A FUNCTION OF THE NEUTRON ENERGY

FIG. 7

THE MAXIMUM AND MINIMUM VALUES OF THE FISSION CROSS SECTION
 OF THE ODD Z, EVEN N NUCLIDES AS A FUNCTION OF THE NEUTRON ENERGY

FIG. 8
FUEL CYCLE FOR AN ENRICHED URANIUM FED
FAST POWER REACTOR

FIG. 9
FUEL CYCLE FOR A "DIRTY" PLUTONIUM
FED FAST POWER REACTOR

FIG. 10
PRODUCTION OF HIGHER ISOTOPES FROM U^{235}

FIG. 11
PRODUCTION OF HIGHER ISOTOPES FROM U²³⁸

FIG. 12
PRODUCTION OF HIGHER ISOTOPES FROM Pu^{241}

FIG. 13
REDUCTION IN POWER COSTS AS FUNCTION
OF THE CREDIT FOR Np^{237}

FIG. 14
REDUCTION IN POWER COSTS AS A FUNCTION
OF THE CREDIT FOR Am^{241}

FIG. 15
REDUCTION IN POWER COSTS AS A FUNCTION
OF THE CREDIT FOR Cm^{242}