Rapid Diagnosis Method for Transplutonium Isotopes Production in High Flux Reactor

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Abstract

This paper proposes a rapid diagnostic method for the irradiation production process of transplutonium isotopes, achieving yield enhancement of Pu-238, Cm-242, Cm-244 and Cf-252.

Full Text

Preamble

Rapid Diagnosis Method for Transplutonium Isotopes Production in High Flux Reactor

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ABSTRACT

Transplutonium isotopes are scarce and must be produced through irradiation in high-flux reactors, yet their production remains inefficient, necessitating optimization studies. This paper analyzes the physical nature of transplutonium isotope production using Cf-252, Cm-244, Cm-242, and Pu-238 as examples. Traditional methods based on Monte Carlo burnup calculations suffer from excessive computational demands and cannot analyze individual energy intervals in detail, thus failing to support refined evaluation, screening, and optimization of irradiation schemes. By grasping the underlying physics and simplifying the production process complexity, we propose a rapid diagnosis method for evaluating irradiation schemes based on the concepts of "Single Energy Interval Value (SEIV)" and "Energy Spectrum Total Value (ESTV)." This method not only avoids tedious burnup calculations but also provides direction for optimization. Optimal irradiation schemes for producing Cf-252, Cm-244, Cm-242, and Pu-238 are determined using this rapid diagnosis method.

The optimal schemes substantially improve production efficiency. Compared with the initial scheme, the optimal scheme enhances production efficiency by 7.41 times for Pu-238, 11.98 times for Cm-242, 65.20 times for Cm-244, and 15.08 times for Cf-252. This work achieves refined analysis of transplutonium isotope production and provides a theoretical basis for improving production efficiency.

Keywords: Transplutonium isotopes, Rapid Diagnosis Method, Production optimization, Single Energy Interval Value, Energy Spectrum Total Value.

1. INTRODUCTION

Transplutonium isotopes [1] refer to nuclides with atomic numbers greater than or equal to 94 (plutonium). Most are radioactive isotopes with unstable nuclei that can spontaneously emit radiation (α, β, γ) or neutrons through spontaneous fission. Transplutonium isotopes primarily include the 94th element plutonium (Pu), the 95th element americium (Am), the 96th element curium (Cm), the 97th element berkelium (Bk), the 98th element californium (Cf), and others. All transplutonium isotopes are metals, as shown in Figure 1 [Figure 1: see original paper]. Due to their research and application value, transplutonium isotopes have attracted extensive scientific attention [2-4].

Figure 1. Photographs of plutonium, americium, berkelium, and californium.

Transplutonium isotopes are scarcer and more expensive than medical isotopes [5-8]. For example, Cf-252 is the most expensive element, costing $27 million per gram—650,000 times more than gold. Cf-252 is a strong neutron source, emitting 2.35×10^{6} neutrons per microgram per second with a half-life of 2.65 years.

Its neutron spectrum resembles that of a fission reactor, making it ideal for reactor startup [9]. Cf-252 is also effective in cervical cancer treatment [10], oil exploration [11], and prompt gamma neutron activation analysis (PGNAA) [12]. Pu-238 is widely used in radioisotope thermoelectric generators due to its moderate half-life, high thermal power density, and easy -decay radiation shielding [13]. Cm-242 and Cm-244 serve as portable -ray sources for powering satellites and spacecraft and are also used as target materials to produce Cf-252 [14].

Most transplutonium isotopes do not occur naturally and must be artificially produced [15-18]. Three production methods exist: (1) accelerator production [19], (2) thermonuclear explosion production [20], and (3) high-flux reactor irradiation [21]. Among these, high-flux reactor irradiation is the most stable and efficient method and the only commercially viable approach for Cf-252 production. Only two laboratories worldwide can stably produce Cf-252 using high-flux irradiated curium targets [22]: Oak Ridge National Laboratory (ORNL) in the United States and the Research Institute of Atomic Reactors (RIAR) in Russia. However, global production remains low. Over 78 production runs in the past 60 years [23], only 1.2 grams of Bk-249, 10.2 grams of Cf-252, 39 milligrams of Es-253, and 15 picograms of Fm-257 were produced through the High Flux Isotope Reactor (HFIR) [24] and the Radiochemical Engineering Development Center (REDC) [25] in the United States.

Neutron capture and decay reactions occur during reactor irradiation production [26-27]. Each neutron absorbed by a nuclide increases its atomic number by one, transforming it into a new nuclide. Thus, target materials evolve from lower-mass to higher-mass nuclides through successive nuclear reactions (radiative capture, -decay). Figure 2 [Figure 2: see original paper] illustrates this nuclide conversion process for transplutonium isotope production in a high-flux reactor [28].

Figure 2. Nuclide conversion process for producing transplutonium isotopes.

In addition to absorption reactions, fission reactions also occur. Once fission occurs, transplutonium isotope production ceases, causing fission losses and low conversion rates. Therefore, reducing fission reactions while enhancing absorption reactions improves production efficiency. Since cross sections depend on the neutron spectrum [29-30], the neutron spectrum around the target must be studied, as it significantly influences production efficiency [31]. Based on literature review, two key questions must be answered to improve transplutonium isotope production efficiency: (1) how to find the optimal neutron spectrum, and (2) how to achieve it. The optimization background is illustrated in Figure 3 [Figure 3: see original paper].

Figure 3. Background of transplutonium isotopes production optimization.

Addressing the first question is challenging due to the high complexity of the production process, and no existing neutronics model can analyze individual energy intervals. Traditional methods search for optimal neutron spectra through

numerous Monte Carlo burnup calculations. Since burnup equations are energy-independent, multi-group burnup calculations are impossible. Consequently, traditional methods require extensive computations and can only macroscopically analyze the entire energy spectrum, not specific intervals in detail, thus failing to support refined evaluation, screening, and optimization of irradiation schemes. A rapid diagnosis method for transplutonium production is needed—one that grasps the production process essence, simplifies complexity, achieves refined neutronics analysis, and supports optimization to improve efficiency and reduce costs.

For the second question, the neutron spectrum has a complex relationship with design parameters, necessitating reactor physics calculations [32-34] and sensitivity analysis. By analyzing how design parameters affect production efficiency, the optimal neutron spectrum can be achieved by optimizing these parameters: (1) moderator selection, (2) moderator layout, (3) target position, (4) target size, and (5) target shape. Adjusting these parameters yields a high-efficiency production scheme—specifically, an optimal neutron spectrum for transplutonium isotope production.

This paper analyzes transplutonium isotope production optimization using Cf-252, Cm-244, Cm-242, and Pu-238 as examples. By proposing the concepts of "Single Energy Interval Value (SEIV)" and "Energy Spectrum Total Value (ESTV)," we establish a rapid diagnosis method enabling detailed analysis of the neutron spectrum around the target. This method avoids tedious burnup calculations while providing direction for irradiation scheme optimization. An optimal irradiation scheme is then proposed based on sensitivity analysis using the rapid diagnosis method. The paper is organized as follows: Chapter 2 introduces the rapid diagnosis method, Chapter 3 provides numerical and experimental verification, Chapter 4 presents the optimization process based on sensitivity analysis, and Chapter 5 concludes.

2.1 The High Flux Reactor

All analyses are performed for a high-flux lead-bismuth reactor under design. It features a 90-day refueling cycle with a maximum flux of 6.7×10^{15}. The inlet temperature is 170°C, outlet temperature is 536.5°C, and coolant velocity is 4.0 m/s. ^{208}Pb-Bi serves as coolant, and ^{208}Pb as reflector. Figure 4 [Figure 4: see original paper] shows the X-Y and X-Z cross-sections of the reactor.

Figure 4. X-Y and X-Z sections of the reactor.

The fuel rod is a simplified model comprising fuel pellets, an air gap, and cladding, with a total length of 50 cm. The fuel assembly has a hexagonal design containing 91 fuel rods. The fuel rod and assembly geometries are shown in Figure 5 [Figure 5: see original paper]. Detailed reactor parameters are provided in Table 1 .

Figure 5. Geometry of the fuel rod and fuel assembly.

Table 1. Detailed parameters of the high-flux reactor.

Parameters	Value
Thermal power	150 MW
Fuel loading capacity / U-235	779 kg / 175.3 kg
Equivalent diameter of active zone	58.14 cm
Height of active zone	50 cm
Average line power density	1130 W/cm^{3}
Maximum line power density	U-10%Zr alloy
Average volume power density	4 / 4.6 mm
Fuel material	Helium
Enrichment of ^{235}U	0.1 mm
Inner / outer diameter of fuel rod	0.2 mm
Filled gas in gaps	80 / 120 cm
Fuel rod clearance width	
Cladding material	
Thickness of cladding	
Ratio of pitch to diameter (P/D)	
Number of assemblies	
Number of fuel rods per assembly	
Axial / radial thickness of reflector layers	

The Cf-252 production target is a mixture of plutonium, americium, and curium [9], with nuclide composition shown in Table 2 . The target undergoes five irradiation cycles, each consisting of 25 days, representing typical californium production campaign operations.

Table 2. Nuclide composition of the Cf-252 production target.

Nuclide	Weight
Pu-238	0.002 g
Pu-239	0.001 g
Pu-240	0.565 g
Pu-242	0.016 g
Am-243	3.903 g
Cm-244	8.734 g
Cm-245	0.271 g
Cm-246	22.349 g
Cm-247	0.647 g
Cm-248	4.743 g

2.2 Production Evaluation by Monte Carlo Burnup Calculation

Transplutonium isotope yields are obtained through Monte Carlo burnup calculations, which couple Monte Carlo criticality calculations with burnup calculations. Figure 6 [Figure 6: see original paper] shows the flowchart for evaluating transplutonium isotope production via this method.

Figure 6. Flowchart for production evaluation by Monte Carlo burnup calculation.

The point burnup equation describes nuclide transmutation over time during high-flux reactor irradiation. For each nuclide in the burnup chain, its time-dependent equation is:

$$\frac{dn_i}{dt} = -\lambda_i^{\text{eff}} n_i + \sum_{j \neq i} b_{j,i}^{\text{eff}} \lambda_j^{\text{eff}} n_j$$

where n_i is the density of the ith nuclide, λ_i^{eff} is the effective decay constant, and $b_{i,j}^{\text{eff}}$ is the branching ratio for transmutation from the ith to jth nuclide. These are calculated as:

$$\lambda_i^{\text{eff}} = \lambda_i + \sum_j \sigma_{i,j} \phi$$

$$b_{i,j}^{\text{eff}} = \frac{\sigma_{i,j} \phi}{\lambda_i + \sum_k \sigma_{i,k} \phi}$$

where λ_i is the decay constant, ϕ is neutron flux, and $\sigma_{i,j}$ is the one-group cross section for reactions converting the ith nuclide to the jth.

During burnup calculations, one-group cross sections and neutron flux are assumed constant within each step, making the point burnup equation a first-order linear ODE. In matrix form:

$$\frac{d\mathbf{n}}{dt} = \mathbf{A}\mathbf{n}$$

where $\mathbf{A}$ is the coefficient matrix of the N-order system, treated as constant within a single burnup step.

Given initial conditions, the solution is:

$$\mathbf{n}(t) = e^{\mathbf{A}t}\mathbf{n}(0)$$

where $\mathbf{A}t$ is the burnup matrix.

Due to extremely low transplutonium isotope yields and complex target composition, the point burnup model typically employs a complex nuclide system covering over a thousand isotopes, including many short-lived ones. For example, the ORIGEN-S database [35] contains ~1,500 nuclides, some like Po-212 with half-lives of 10^{-7} s. Thus, transplutonium production burnup equations are numerous and stiff, complicating their solution.

As Equation (2) shows, target cross sections are required for Monte Carlo burnup calculations, necessitating criticality calculations. The k-eigenvalue neutron transport equation is [36]:

$$\mathbf{L}\phi + \mathbf{C}\phi + \mathbf{S}\phi = \frac{1}{k_{\text{eff}}}\mathbf{M}\phi$$

where **L**, **C**, **S**, and **M** are leakage, collision, scattering, and fission operators, k_{eff} is the effective multiplication factor, and ϕ is neutron flux.

Physical parameters around the target (neutron flux, fission/absorption rates) are obtained by solving Equation (5). Monte Carlo reaction rates are calculated as:

$$R = \int \Sigma(\mathbf{r}, E)\phi(\mathbf{r}, E)d\mathbf{r}dE$$

Thus, the cross section for burnup calculations is:

$$\sigma = \frac{R}{\int \phi(\mathbf{r}, E)d\mathbf{r}dE}$$

These formulas demonstrate that Monte Carlo burnup calculations are complex and computationally expensive [37-38]. Testing on the high-flux reactor in Figure 4 with 1,000,000 neutrons/cycle, 100 inactive cycles, 300 active cycles, and 10 burnup steps requires 150 minutes per calculation using 64 threads on a 2nd Gen AMD EPYC™ 7742. Even considering only Cf-252, Cm-244, Cm-242, and Pu-238 production and only five design parameters (Figure 9 [Figure 9: see original paper]), 9,720 calculations are needed—totaling 1,458,000 minutes (1,012.5 days), which is unacceptable.

Moreover, since Monte Carlo burnup calculations only require one-group cross sections, they can only assess whole-spectrum effects, not analyze individual energy intervals. During neutron spectrum optimization, we can only work from macroscopic design parameters without knowing which energy intervals favor or hinder production, preventing detailed optimization guidance.

Therefore, we must understand the production process physics and develop a new rapid diagnosis method that simplifies calculations while enabling detailed production process analysis. Such a method would facilitate irradiation scheme evaluation, screening, and optimization.

2.3 Rapid Diagnosis Method Based on SEIV and ESTV

The root cause of low transplutonium isotope production is fission loss [14]. During high-flux reactor irradiation, target materials evolve from lower-mass to higher-mass nuclides via absorption reactions. However, fission reactions also occur, terminating transplutonium production and causing fission losses that lead to low conversion rates. Therefore, the absorption-to-fission rate ratio (A/F) is a crucial physical quantity affecting production efficiency. Efficiency can be assessed by analyzing A/F values throughout the nuclide chain, and A/F can be calculated for individual energy intervals, enabling refined neutronics analysis. Using Cf-252 production from the Table 2 target as an example, Figure 7 [Figure 7: see original paper] shows A/F values for intermediate nuclides: Cm-244, Cm-245, Cm-246, Cm-247, Cm-248, Bk-249, Cf-250, and Cf-251, with nuclear data from JANIS [39].

Figure 7. A/F values of intermediate nuclides for Cf-252 production.

As Figure 7 shows, A/F values vary significantly among nuclides and energy intervals, meaning different nuclides and energy intervals have markedly different effects on production efficiency. Whole-spectrum analysis alone is insufficient; detailed energy interval analysis is necessary.

We propose a rapid diagnosis method based on the neutron spectrum around the target to enable fast, refined evaluation of production schemes. The model only requires the neutron spectrum from Monte Carlo criticality calculations [40-41] to evaluate scheme efficiency, avoiding burnup calculations and drastically reducing computational resources. Thus, the rapid diagnosis method can quickly screen optimal transplutonium isotope production schemes and guide irradiation scheme optimization.

The method comprises four steps: (1) obtain the neutron spectrum around the target via Monte Carlo criticality calculation, described by 46-group neutron flux, fission rate, and absorption rate; (2) define "Single Energy Interval Value (SEIV)" and calculate it for all 46 energy intervals; (3) define "Energy Spectrum Total Value (ESTV)" and calculate it using 46-group neutron flux and SEIV; and (4) evaluate scheme efficiency by comparing ESTV values—higher ESTV indicates higher production efficiency and yield. The schematic is shown in Figure 8 [Figure 8: see original paper].

Figure 8. Schematic diagram of the rapid diagnosis method.

SEIV is calculated as:

$$\mathrm{SEIV}_i = \frac{R_{a,i}}{R_{f,i}} \cdot \frac{R_{a,i}}{\phi_i}$$

where subscript i denotes the energy interval number, $R_{a,i}$ is the absorption rate, $R_{f,i}$ is the fission rate, and ϕ_i is neutron flux in the ith interval.

SEIV is the product of two terms: the A/F ratio, indicating absorption versus fission tendency, and the absorption rate-to-flux ratio, indicating absorption probability. A higher SEIV means absorption is favored over fission and occurs more frequently in that interval. High-SEIV intervals are crucial for production, and neutron flux in these intervals should be maximized.

ESTV is calculated as:

$$\mathrm{ESTV} = \sum_{i=1}^{46} \mathrm{SEIV}_i \cdot \phi_i$$

ESTV integrates SEIV across the neutron spectrum, indicating the target nuclide' s overall neutron absorption tendency without fission. Higher ESTV corresponds to more efficient transplutonium production. Since ESTV depends only on the neutron spectrum, burnup calculations are unnecessary, enabling rapid scheme evaluation.

3.1 Numerical Verification

We verify the rapid diagnosis method by comparing burnup calculations with ESTV. Production efficiency is optimized by adjusting five design parameters: (1) moderator selection, (2) moderator layout, (3) target position, (4) target size, and (5) target shape. Optional values are shown in Figure 9 [Figure 9: see original paper].

Figure 9. Optional values for the five design parameters.

We use a large hexagonal prism target at Position #1 with zirconium hydride moderator as the initial scheme. Different irradiation schemes are generated by modifying moderator layout. Criticality calculations provide the neutron spectrum around the target, from which SEIV and ESTV are determined to evaluate scheme efficiency.

Pu-238, Cm-242, Cm-244, and Cf-252 serve as examples. The Pu-238 production target is Np-237; Cm-242 uses Am-241; Cm-244 uses Pu-239; and Cf-252 uses the composition in Table 2. SEIV and ESTV are calculated for the bare target and six moderator layouts, yielding seven production schemes per isotope. SEIV values are given in Table 3 and ESTV values in Table 4 .

Table 3. SEIV for producing Pu-238, Cm-242, Cm-244, and Cf-252.

Energy (MeV)	Pu-238	Cm-242	Cm-244	Cf-252
[1.00E-11, 1.00E-07]	4.55E+04	3.06E+03	4.96E+00	2.42E+00
[1.00E-07, 4.14E-07]	2.78E+04	2.10E+03	1.10E+01	2.53E+00
[4.14E-07, 8.76E-07]	6.99E+04	1.88E+03	9.02E-01	1.75E+01
[8.76E-07, 1.86E-06]	1.93E+04	3.89E+03	4.87E+02	1.16E+02
[1.86E-06, 5.04E-06]	3.17E+02	7.34E-02	1.89E-02	1.48E+00

Energy (MeV)	Pu-238	Cm-242	Cm-244	Cf-252
...	...	...	...	...

Table 4. ESTV for producing Pu-238, Cm-242, Cm-244, and Cf-252.

Scheme	Pu-238	Cm-242	Cm-244	Cf-252
Bare target	4.50E-03	5.64E-03	3.92E-06	9.22E-05
Layout #1	1.36E-01	1.92E-02	2.55E-05	4.41E-04
Layout #2	6.78E-01	4.05E-02	5.00E-05	1.08E-03
Layout #3	1.60E+00	7.14E-02	7.59E-05	1.72E-03
Layout #4	5.46E+00	1.74E-01	1.77E-04	4.39E-03
Layout #5	6.26E+00	1.99E-01	2.01E-04	4.94E-03
Layout #6	1.11E+01	2.94E-01	2.42E-04	4.59E-03

As Table 3 shows, the optimal neutron spectrum is not always softer. The interval [4.14E-07 MeV, 8.76E-06 MeV] has the highest SEIV for Pu-238; [1E-11 MeV, 1E-07 MeV] for Cm-242; [1E-07 MeV, 4.14E-07 MeV] for Cm-244; and [8.76E-07, 1.86E-06] for Cf-252. SEIV variation across intervals is substantial—for Cf-252, the maximum deviation reaches 1.37×10^{9} times. Thus, transplutonium isotope efficiency can be improved by modifying the neutron spectrum around the target [42-43].

Table 4 shows ESTV varies significantly across schemes. The bare target always has the lowest ESTV, confirming that moderator placement is essential. Layout #6 yields the highest ESTV for Pu-238, Cm-242, and Cm-244, while Layout #5 is optimal for Cf-252. Since SEIV and ESTV enable detailed energy interval analysis, they guide irradiation scheme optimization.

Comparing schemes solely by SEIV and ESTV is not rigorous, so verification is needed. We perform burnup calculations using the RMC code [44] to calculate yields for Pu-238, Cm-242, Cm-244, and Cf-252. Since the neutron spectrum changes during burnup, we first simulate a one-second burnup to ensure consistent spectra for ESTV and yield calculations. This establishes the ESTV-yield relationship under a fixed spectrum. One-second yields are given in Table 5 .

Table 5. One-second yields for Pu-238, Cm-242, Cm-244, and Cf-252 (g^{-1}).

Scheme	Pu-238	Cm-242	Cm-244	Cf-252
Bare target	5.03E-13	1.53E-12	1.68E-45	1.94E-37
Layout #1	1.07E-12	2.29E-12	4.96E-45	6.90E-37
Layout #2	1.86E-12	4.20E-12	1.07E-44	1.32E-36
Layout #3	2.86E-12	6.75E-12	1.92E-44	1.79E-36
Layout #4	6.78E-12	1.62E-11	2.44E-43	1.38E-35

Scheme	Pu-238	Cm-242	Cm-244	Cf-252
Layout #5	7.62E-12	1.80E-11	2.85E-43	1.46E-35
Layout #6	1.05E-11	2.64E-11	1.60E-43	5.79E-36

As Table 5 shows, one-second yields increase with ESTV for all isotopes, confirming that ESTV evaluates yield for a given spectrum. However, one-second burnup only describes early irradiation, not the full cycle. To verify ESTV' s applicability to the entire cycle, we simulate 90-day target irradiation. The 90-day yields are given in Table 6 .

Table 6. 90-day yields for Pu-238, Cm-242, Cm-244, and Cf-252 (g^{-1}).

Scheme	Pu-238	Cm-242	Cm-244	Cf-252
Bare target	1.65E+00	1.28E+00	1.96E-02	7.95E-03
Layout #1	3.03E+00	1.74E+00	4.51E-02	2.07E-02
Layout #2	4.64E+00	2.68E+00	7.91E-02	3.42E-02
Layout #3	6.07E+00	3.58E+00	2.98E-01	4.20E-02
Layout #4	9.44E+00	6.57E+00	1.00E+00	8.23E-02
Layout #5	1.01E+01	7.08E+00	7.08E+00	9.80E-02
Layout #6	1.16E+01	1.05E+01	1.05E+01	8.30E-02

Table 6 shows 90-day yields also increase with ESTV for all isotopes, demonstrating that ESTV indicates both early-stage and full-cycle efficiency. Thus, the SEIV- and ESTV-based rapid diagnosis method effectively evaluates transplutonium isotope production efficiency.

3.2 Experimental Verification

Since the Figure 4 reactor is not yet built, experimental verification is conducted on the High Flux Isotope Reactor (HFIR) [45]. HFIR has produced Cf-252 since 1966, currently supplying 70% of global Cf-252, and possesses extensive measured transplutonium production data.

Rated at 100 MW (currently operating at 85 MW), HFIR has the world's highest steady-state neutron flux (2.6×10^{15} cm$^{-2}\cdot$s^{-1}). It is a light-water-cooled, moderated flux-trap reactor using highly enriched ^{235}U. The core comprises concentric annular zones ~61 cm high (fuel height 51 cm). At the center is a 12.70 cm cylindrical "neutron flux trap" containing 37 vertical experimental holes, surrounded by two concentric fuel assemblies. The core and flux trap are shown in Figure 10 [Figure 10: see original paper] [46].

Figure 10. HFIR reactor core and neutron flux trap.

Since the rapid diagnosis method uses ESTV to evaluate scheme efficiency, Monte Carlo criticality calculations are performed to compute ESTV. The RMC

code models HFIR, with input cards provided in the attachment. Core and flux trap modeling are shown in Figure 11 [Figure 11: see original paper].

Figure 11. HFIR core and flux trap modeling in RMC.

To verify the model, radial thermal and total neutron flux distributions are calculated and compared with reference solutions in Figure 12 [Figure 12: see original paper]. The RMC flux distributions agree well with references, confirming correct HFIR modeling.

Figure 12. Comparison of radial flux distributions.

Experimental verification proceeds using the target design in Figure 13 [Figure 13: see original paper], irradiated in HFIR for five days. Four schemes were formed with two target compositions and two positions: 245Cm_A, 245Cm_C, Cf_A, and Cf_C. Heavy nuclei yields (all nuclides heavier than target nuclides) were measured [9], and corresponding ESTV values calculated. Results are in Table 7 .

Figure 13. Experimental target design.

Table 7. Experimental verification results.

Irradiation Scheme	Nuclide Composition	Irradiation Duration	Yields of Heavy Nuclei (Experiment)	ESTV (Simulation)
245Cm_A	245Cm: 45.9±0.3 ng	5 days	249Cf: 0.099±0.001 ng	7.861 ng
245Cm_C	249Cf: 42.4±0.3 ng	5 days	250Cf: 11.3±0.2 ng	9.308 ng
Cf_A	251Cf: 30.2±0.5 ng	5 days	7.861 ng	1.00E-12
Cf_C	-	5 days	9.308 ng	1.09E-12

Since target composition must be identical for ESTV comparison, we compare 245Cm_A vs. 245Cm_C and Cf_A vs. Cf_C. Table 7 shows 245Cm_A' s measured heavy nuclei yield and ESTV are lower than 245Cm_C' s, and Cf_A' s yield and ESTV are lower than Cf_C' s. These results confirm the rapid diagnosis method' s principle: higher ESTV yields higher production efficiency.

Thus, experimental verification validates the rapid diagnosis method. The next chapter applies it to rapidly screen and determine optimal irradiation schemes.

4.1 The Logic of Optimization

Five design parameters require optimization: (1) moderator selection, (2) moderator layout, (3) target position, (4) target size, and (5) target shape.

As Table 3 shows, the highest-SEIV energy interval is always in the thermal range, regardless of isotope, so moderators must soften the neutron spectrum around the target. We seek the material with strongest moderating power to achieve the best spectrum with minimal moderator. Moderating power is an intrinsic material property unaffected by other parameters, so moderator material can be determined first. Using a large hexagonal prism target at Position #1 with Layout #1 as the initial scheme, nine moderator materials are tested, with resulting neutron spectra shown in Figure 14 [Figure 14: see original paper].

Figure 14. Neutron spectra with different moderators.

Zirconium hydride exhibits the best moderating power and is selected as the moderator for all subsequent analyses.

To determine the optimal production scheme, we must optimize moderator layout and target position, size, and shape. Starting from a large hexagonal prism bare target at Position #1, we optimize the four remaining parameters. Optimization order is critical and determined by sensitivity analysis. The sensitivity coefficient of design parameter d to production efficiency is:

$$\eta = \frac{\Delta P/P}{\Delta d/d}$$

where P is production efficiency and d is the design parameter.

The sensitivity coefficient correlates with the maximum ESTV increase achievable by adjusting a parameter. Larger potential increases indicate greater importance, so such parameters are optimized first. After optimizing one parameter, the improved scheme becomes the baseline for optimizing remaining parameters, continuing until all are optimized. The optimization logic is diagrammed in Figure 15 [Figure 15: see original paper]. This paper details Cf-252 scheme optimization; Pu-238, Cm-242, and Cm-244 optimal schemes are given in Section 4.6.

Figure 15. Optimization logic diagram.

4.2 First Round Sensitivity Analysis and Optimization

The first round considers four parameters: moderator layout, target position, size, and shape. Optional values are in Figure 9. New schemes adjust one parameter from the initial scheme. ESTV values are in Table 8 , where η is the sensitivity coefficient.

Table 8. ESTV for first-round optimization.

Parameter	Variation	ESTV
Layout	Bare target	9.22E-05
	Layout #1	4.41E-04

Parameter	Variation	ESTV
	Layout #2	1.08E-03
	Layout #3	1.72E-03
	Layout #4	4.39E-03
	Layout #5	4.94E-03
	Layout #6	4.59E-03
Shape	Hexagonal prism	9.22E-05
	Sphere	9.78E-05
	Cylinder	9.22E-05
Position	Position #1	9.22E-05
	Position #2	1.07E-04
	Position #3	5.43E-04
	Position #4	6.97E-04
Size	Large	9.22E-05
	Medium	9.35E-05
	Small	9.22E-05

Moderator layout provides the greatest ESTV improvement and is optimized first. Layout #5 yields the highest ESTV, so it is selected. After the first round, the scheme becomes: large hexagonal prism target at Position #1 with Layout #5.

4.3 Second Round Sensitivity Analysis and Optimization

The second round considers three parameters: target position, size, and shape. All schemes build on the first-round result. ESTV values are in Table 9 .

Table 9. ESTV for second-round optimization.

Parameter	Variation	ESTV
Position	Position #1	4.94E-03
	Position #2	3.45E-03
	Position #3	2.23E-03
	Position #4	1.04E-03
Shape	Hexagonal prism	4.94E-03
	Sphere	4.03E-03
	Cylinder	4.75E-03
Size	Large	4.94E-03
	Medium	5.42E-03
	Small	6.28E-03

Target position provides the greatest ESTV improvement. Position #1 yields the highest ESTV and is selected. After the second round, the scheme remains: large hexagonal prism target at Position #1 with Layout #5.

4.4 Third Round Sensitivity Analysis and Optimization

The third round considers two parameters: target size and shape. All schemes build on the second-round result. ESTV values are in Table 10 .

Table 10. ESTV for third-round optimization.

Parameter	Variation	ESTV
Shape	Hexagonal prism	4.94E-03
	Sphere	4.03E-03
	Cylinder	4.75E-03
Size	Large	4.94E-03
	Medium	5.42E-03
	Small	6.28E-03

Target size and shape have similar effects, with size slightly more influential. Size is optimized in this round. The small target yields the highest ESTV and is selected. After the third round, the scheme becomes: small hexagonal prism target at Position #1 with Layout #5.

4.5 Fourth Round Sensitivity Analysis and Optimization

The fourth round considers one parameter: target shape. All schemes build on the third-round result. ESTV values are in Table 11 .

Table 11. ESTV for fourth-round optimization.

Parameter	Variation	ESTV
Shape	Hexagonal prism	6.28E-03
	Sphere	5.19E-03
	Cylinder	6.10E-03

The hexagonal prism yields the highest ESTV and is selected. After the fourth round, the final scheme is: small hexagonal prism target at Position #1 with Layout #5.

4.6 Final Irradiation Scheme

The optimized Cf-252 production scheme is a small hexagonal prism target at Position #1 with Layout #5. Pu-238, Cm-242, and Cm-244 schemes were similarly optimized. The final schemes are summarized in Table 12 , with efficiency comparisons shown in Figure 16 [Figure 16: see original paper].

Table 12. Final optimized irradiation schemes.

Design Parameter	Pu-238	Cm-242	Cm-244	Cf-252
Moderator selection	Zirconium hydride	Zirconium hydride	Zirconium hydride	Zirconium hydride
Moderator layout	Layout #6	Layout #6	Layout #6	Layout #5
Target position	Position #1	Position #1	Position #1	Position #1
Target shape	Hexagonal	Hexagonal	Cylinder	Hexagonal
Target size	Small	Small	Small	Small

Figure 16. Efficiency comparison before and after optimization.

After four optimization rounds, optimal transplutonium isotope production schemes are rapidly obtained. As Figure 16 shows, the optimal schemes substantially improve efficiency over the initial scheme: Pu-238 by 7.41×, Cm-242 by 11.98×, Cm-244 by 65.20×, and Cf-252 by 15.08×. Thus, the rapid diagnosis method-based optimization strategy effectively enhances transplutonium isotope production.

5. CONCLUSION

Reactor irradiation for transplutonium isotope production is inefficient, requiring optimization studies. The production physics is complex to quantify. By grasping the physical nature and simplifying process complexity, we propose SEIV and ESTV concepts to quantitatively analyze production efficiency. A rapid diagnosis method based on SEIV and ESTV is established, and optimization designs for Cf-252, Cm-244, Cm-242, and Pu-238 are implemented. The optimal scheme improves efficiency by 7.41×for Pu-238, 11.98×for Cm-242, 65.20×for Cm-244, and 15.08×for Cf-252 compared to the initial scheme.

This paper addresses two key questions in transplutonium isotope production: (1) how to find the optimal neutron spectrum, and (2) how to achieve it. For the first, SEIV and ESTV enable detailed neutron spectrum analysis around the target. For the second, we optimize target design parameters to achieve the optimal spectrum, improving computational efficiency.

This work realizes refined analysis of transplutonium isotope production and provides a theoretical basis for efficiency improvement. Future research will focus on energy spectrum conversion technology for finer optimal spectrum achievement.

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