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MODELING OF THE PU-238 PRODUCTION PROCESS IN A VVER-440 REACTOR USING DIFFERENT STARTING MATERIALS AND THEIR ARRANGEMENT STRUCTURES

Annotation

The paper investigates the feasibility of producing high-purity Pu-238 in a VVER-440 thermal reactor by irradiating the actinide Neptunium-237 (Np-237) from spent nuclear fuel. The study uses Serpent2 Monte Carlo simulations to model neutron interactions and optimize key configurations, including lattice height and moderation environment. The goal is to produce approximately 3 kilograms of high-purity plutonium annually, with an isotopic specification of no less than 80% ^{238}Pu and not exceeding 2 ppm of ^{236}Pu.

Keywords: ^{237}Np, ^{238}Pu, irradiation device, lattice pitch, Serpent2, ppm (parts per million), radioisotope thermoelectric generators (RTGs), VVER-440 reactor.

МОДЕЛИРОВАНИЕ ПРОЦЕССА ПРОИЗВОДСТВА PU-238 В РЕАКТОРЕ ВВЭР-440 С ИСПОЛЬЗОВАНИЕМ РАЗЛИЧНЫХ СТАРТОВЫХ МАТЕРИАЛОВ И СТРУКТУРЫ ИХ РАЗМЕЩЕНИЯ

Аннотация

В работе исследуется возможность получения высокочистого Pu-238 в тепловом реакторе ВВЭР-440 путем облучения актинида нептуния-237 (Np-237), извлеченного из отработавшего ядерного топлива. В исследовании используются методы Монте-Карло (код Serpent2) для моделирования нейтронных взаимодействий и оптимизации ключевых конфигураций, включая высоту решетки и условия замедления нейтронов. Целью является ежегодное производство приблизительно 3 килограммов высокочистого плутония с изотопным составом, содержащим не менее 80% ^{238}Pu и не более 2 ppm (частей на миллион) ^{236}Pu.

Ключевые слова: ^{237}Np, ^{238}Pu, облучательное устройство, шаг решетки, Serpent2, ppm (частей на миллион), радиоизотопные термоэлектрические генераторы (РИТЭГ), реактор ВВЭР-440.

VVER-440 REAKTORIDA PU-238 ISHLAB CHIQARISH JARAYONINI TURLI XIL BOSHLANG'ICH MATERIALLAR VA ULARNI JOYLASHTIRISH TUZILMALARIDAN FOYDALANGAN HOLDA MODELLASHTIRISH

Annotatsiya

Maqolada ishlatilgan yadro yoqilg‘isidan aktinid Neptunium-237 (Np-237) nurlanishi orqali VVER-440 termal reaktorida yuqori toza Pu-238 ishlab chiqarishning maqsadga muvofiqligi o‘rganiladi. Tadqiqot neytronlarning o‘zaro ta’sirini modellashtirish va asosiy konfiguratsiyalarni, shu jumladan panjara balandligi va moderatsiya muhitini optimallashtirish uchun Serpent2 Monte Carlo simulyatsiyalaridan foydalanadi. Maqsad har yili taxminan 3 kilogramm yuqori toza plutoniy ishlab chiqarish, izotopik spetsifikatsiyasi kamida 80% ^{238}Pu va 2 ppm ^{236}Pu dan oshmaydi.

Kalit so‘zlar: ^{237}Np, ^{238}Pu, nurlanish moslamasi, panjara qadami, Serpent2, ppm (millionga qism), radioizotopli termoelektr generatorlari (RTG), VVER-440 reaktor.

Problem statement. Plutonium-238 (Pu-238) is an essential isotope for powering radioisotope thermoelectric generators (RTGs), which are critical for long-duration space missions. However, global production has been insufficient to meet growing demand. It should be noted that the task of producing ^{238}Pu is associated with the need to meet several requirements for its suitability for space RTGs-the ^{238}Pu content must be at least 80%, and the ^{236}Pu content must be no more than 2 ppm-complicating the initial problem. And here are the specific objectives for the research:

- to review and analyze existing methods of Pu-238 production in thermal reactors;
- to investigate the application of the Serpent2 Monte Carlo simulation code for modeling Pu-238 accumulation;
- to prepare and assess various starting material compositions based on Neptunium isotope;
- to develop homogeneous and heterogeneous models of the irradiation assembly;
- to simulate neutron-physical parameters under different moderation and structural scenarios;
- to examine the influence of moderators such as H_2O, ZrH_2, natural Pb, and ^{208}Pb on Pu-238 production;
- to compare the effectiveness of different configurations in terms of yield, isotopic purity, and compliance with production standards.

Research Methodology. The irradiation device was placed in the center of the VVER-440 reactor core (Figure 1). The main parameters of the VVER-440 reactor and fuel assemblies are as follows:

- thermal power of the reactor- 1375 MW;

• number of FAs in the reactor core- 349;
• fuel- enriched uranium dioxide (3.6% ^{235}U), density: 10.4 g/cm^3;
• fuel rod cladding- alloy of 99% Zr and 1% Nb, density: 6.5g/cm^3;
• coolant- light water, density: 0.7g/cm^3;
• size of the hexagonal FA "turnkey"- 14.4 cm;
• diameter of the fuel pellet- 9.1 mm;
• diameter of the central hole- 3 mm;
• gap thickness between fuel and cladding- 0.075 mm;
• cladding thickness- 0.6 mm;
• triangular lattice pitch of fuel rods- 12.2 mm;
• height of the fuel column- 250 cm.

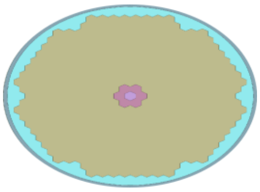

Figure 1. Core model of the VVER-440.

This basic model consists of a central NpO_2 FA, surrounded by a Pb-208 moderator, UO_2, water, and a core box.

Analysis and results

Here, the main limitation on the development process:

• Pu-238 content in plutonium ≥ 80%;
• Pu-236 content ≤ 2*ppm*.

The main difficulties in achieving the criterion for Pu-236, which is formed by the $^{237}Np(n,2n)\ ^{236}Pu$ reaction and to solve the problem of slowing down fast neutrons, we surrounded Np-237 with various moderators.

To achieve the primary research objective, six different core configurations were investigated, each employing neptunium dioxide (NpO_2) at the center and varying surrounding moderator environments. The examined cases are as follows:

1. NpO_2 located at the core center, surrounded by a layer of standard UO_2-based fuel assemblies;
2. NpO_2 at the center, with a surrounding shell of enriched ^{208}Pb encapsulated in Zr-Nb alloy cladding;
3. NpO_2 at the center, surrounded by two concentric layers of lead, comprising 18 Pb-based assemblies.
4. NpO_2 at the center, surrounded by natural lead assemblies also clad in Zr-Nb alloy;
5. NpO_2 at the center, surrounded by light water (H_2O) as a moderator;
6. NpO_2 at the center, surrounded by ZrH_2 as a moderator.

In variant 2, the use of ^{208}Pb rods was motivated by two main considerations. First, the ^{208}Pb isotope possesses a very low neutron absorption cross-section. Second, it is capable of shifting the spectrum of slowing-down neutrons into the resonance energy range, which may enhance the production rate of ^{238}Pu in the central NpO_2-based fuel assembly.

For variants 1 through 6, calculations were performed to evaluate the isotopic composition and production rate of plutonium generated within the central NpO_2 assemblies. All simulations were conducted under the condition of a constant triangular lattice pitch of 12.2 mm for the fuel rods. The results are presented in Table 1.

№	Moderator	Pu-238 production rate [kg Pu-238/year]	Pu-238/Pu, %	Pu-236/Pu, ppm
1	UO_2	3.70	99.1	56.7
2	^{208}Pb	4.73	98.5	20.3
3	2 layers ^{208}Pb	2.70	98.2	13.3
4	Natural Pb	4.27	98.7	24.7
5	H_2O	2.56	99.3	37.5
6	ZrH_2	1.50	98.9	30.4

Table 1 - Plutonium production parameters in the irradiation device of the VVER-440 reactor.

Without a lead-containing environment, the central NpO_2 assembly produces plutonium with an unacceptably high fraction of ^{236}Pu. Using water as a surrounding medium (variant 5) significantly reduces ^{238}Pu yield. In contrast, lead moderators improve both the production rate and isotopic composition. Yet, all variants exceed the 2 ppm limit for ^{236}Pu. While a single layer of ^{208}Pb provided high ^{238}Pu yield, increasing it to a double layer in order to reduce ^{236}Pu resulted in a drop in ^{238}Pu production. Therefore, this approach is not effective, and adjusting the lattice pitch is required in future studies. To mitigate this, future variants will explore increasing the lattice pitch to soften the neutron spectrum in the NpO_2 assembly.

Within the Serpent framework, options for heterogeneous placement of Np fuel elements with different lattice pitches were studied. The considered options are presented in Figure 2.

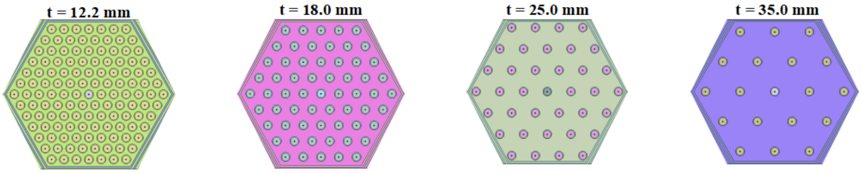

Figure 2. Design of NpO_2-FA under variable lattice pitch.

Lattice pitch of NpO_2 pins, mm	12.2	18	25	35
Initial amount of *Np*, kg	215	102.3	61.5	30.7
^{238}Pu mass, kg	4.60	4.11	3.70	3.07
^{238}Pu fraction, %	98.4	96.8	94.7	89.3
^{236}Pu fraction, ppm	19	7.5	2.84	1.3

Table 2 - Production rate and isotopic composition of *Pu* with increasing lattice pitch of NpO_2 fuel elements. Surrounded by six natural Pb FAs.

Table 2 shows how increasing the lattice pitch of NpO_2 fuel assemblies, surrounded by Pb-based assemblies, affects the plutonium yield and isotopic composition; as the pitch increases, less ^{237}Np is loaded per assembly, leading to a reduced total Pu yield.

However, a key observation is the significant reduction in the fraction of ^{236}Pu, decreasing from approximately 19 ppm to 1.3 ppm. This reduction brings the ^{236}Pu level well within the required limits for high-purity isotopic fuel applications (typically ≤ 2 ppm). Meanwhile, the ^{238}Pu fraction decreases slightly (from 98.4% to 89.3%) yet remains well above the standard threshold (85%) required for radioisotope thermoelectric generator (RTG) applications in space systems.

Considering the particularly attractive nuclear characteristics of ^{208}Pb, a set of core design configurations was analyzed in which a central NpO_2 fuel assembly (FA) was surrounded by six ^{208}Pb-based assemblies. In the simulation, the lattice pitch between the NpO_2 fuel rods was varied (12.2 mm, 18 mm, 25 mm, and 35 mm) to assess its effect on isotopic composition and plutonium yield.

Due to the minimal influence of changing the lattice pitch of the ^{208}Pb rods, that parameter was excluded from the scope of the analysis. The results of the calculations are summarized in Table 3.

Lattice pitch of NpO_2 pins, mm	**12.2**	**18**	**25**	**35**
Initial amount of *Np*, kg	215	102.3	61.5	30.7
^{238}Pu mass, kg	4.86	4.55	4.31	3.45
^{238}Pu fraction, %	98.2	96.4	93.6	88.1
^{236}Pu fraction, ppm	19.2	5.23	4.6	1.4

Table 3 - Production rate and isotopic composition of *Pu* with increasing lattice pitch of NpO_2 fuel elements. Surrounded by six Pb-208 FAs.

It is evident that switching to ^{208}Pb in configurations with a wider NpO_2 fuel rod lattice results in an 11% increase in the total mass of produced plutonium and a 10% improvement in specific production. At the same time, the quality of the resulting ^{238}Pu meets the required standard in both natural lead and ^{208}Pb environments, as the ^{236}Pu fraction remains at or below 2 ppm.

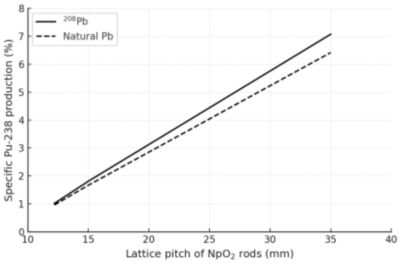

Figure 3. Specific plutonium production with increasing pitch of Np fuel elements in variants using natural lead and ^{208}Pb in the FA environment.

Figure 3 shows the dependence of specific plutonium production (per unit mass of loaded neptunium) on the increasing lattice pitch of neptunium fuel rods in configurations using natural lead and ^{208}Pb in the surrounding fuel assemblies. A significant—eightfold or greater—increase in specific plutonium production is evident with increasing Np lattice pitch.

Conclusions

As a result of the work performed, the following conclusions can be made:

1. Using the Serpent program, a model of the VVER-440 reactor was built, within the framework of which the problem of Pu-238 production was considered.

2. During this scientific work, Np-237 was considered as the starting material. The specificity of this starting material was to satisfy two conflicting criteria for Pu-238 and Pu-236. Calculations showed that the main difficulty was the criterion for Pu-236 $\leq 2ppm$.

3. Different protective layers of moderators around the starting material have been investigated. For a standard/close lattice of Np- rods pitch, even the best of them (Pb-208) do not allow the 2 ppm criterion to be achieved.

4. The study of the effect of Np-rod pitch on achieving the Pu-238 and Pu-236 criteria has been carried out; it was found that both criteria were achieved for the Neptunium environment with a wide grid of Np-rods (t=35 mm) and 6 FAs Pb-208.

5. If both criteria are met, the Pu-238 production parameters are as follows:

- ^{238}Pu mass - 3.5 kg;
- ^{236}Pu fraction- 1.4 ppm;
- ^{238}Pu fraction- 88%;
- $^{238}Pu/^{237}Np$ − 11.2%.

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