NUCET NUCLEAR ENERGY AND TECHNOLOGY

Nuclear Energy and Technology 9(4): 297–301
DOI 10.3897/nucet.9.117199

Research Article

Assessment of the possibility for large-scale ^{238}Pu production in a VVER-1000 power reactor*

Anatoly N. Shmelev[1], Nikolay I. Geraskin[1], Vladimir A. Apse[1], Gennady G. Kulikov[2], Evgeny G. Kulikov[2], Vasily B. Glebov[1]

1 *MEPhI, 31 Kashirskoe shosse, 115409 Moscow, Russia*

2 *State Atomic Energy Corporation ROSATOM, 24 Bolshaya Ordynka str., 119017 Moscow, Russia*

Corresponding author: Evgeny G. Kulikov (egkulikov@mephi.ru)

Academic editor: Snejana Grozev ♦ **Received** 14 November 2022 ♦ **Accepted** 9 September 2023 ♦ **Published** 21 December 2023

Citation: Shmelev AN, Geraskin NI, Apse VA, Kulikov GG, Kulikov EG, Glebov VB (2023) Assessment of the possibility for large-scale ^{238}Pu production in a VVER-1000 power reactor. Nuclear Energy and Technology 9(4): 297–301. https://doi.org/10.3897/nucet.9.117199

Abstract

The paper presents the estimates for the possibility for large-scale production of ^{238}Pu in the core of a VVER-1000 power reactor. The Np-fraction of minor actinides extracted from transuranic radioactive waste is proposed to be used as the starting material. The irradiation device with NpO_2 fuel elements is installed at the reactor core center. The NpO_2 fuel lattice pitch is varied and the irradiation device is surrounded by a heavy moderator layer to create the best possible spectral conditions for large-scale production (~ 3 kg/year) of conditioned plutonium with the required isotopic composition (not less than 85% of ^{238}Pu and not more than 2 ppm of ^{236}Pu). Plutonium with such isotopic composition can be used as the thermal source in thermoelectric radioisotope generators and in cardiac pacemakers. It has been demonstrated that the estimated scale of the ^{238}Pu production in a VVER-type power reactor exceeds considerably the existing scale of its production in research reactors.

Keywords

^{238}Pu, thermoelectric radioisotope generators, irradiation device, VVER-1000 reactors

Problem statement

The ^{238}Pu plutonium isotope has a half-life of $T_{1/2}$ = 87.7 years. This value, on the one hand, is comparatively small for one to be able to say that the specific heat generation is intense (~ 570 W/kg), and, on the other hand, is high enough to be able to state that heat will be generated for a long time.

These properties make the ^{238}Pu isotope a valuable source of thermal and electric energy for use in thermoelectric radioisotope generators (TRG) of spacecraft and in cardiac pacemakers (Pustovalov et al. 2005).

At the same time, the capacities currently available in Russia and worldwide for its production are insufficient and fail to cover the growing annual demand (Start-up Plan for Plutonium-238 Production for Radioisotope Power Systems 2010, Austin et al. 2020). This isotope is produced largely in research reactors.

It should be noted that production of ^{238}Pu involves the need for satisfying a number of requirements (e.g., the NASA's) as regards its suitability for space-borne TRGs (the content of ^{238}Pu is not less than 85%, and the fraction of ^{236}Pu is not more than 2 ppm (Daily, McDuffee 2020)

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2023, n. 3, pp. 118–126.

which makes it more difficult to solve the original problem. The purpose of this study is to find out if it is possible to create the optimal spectral conditions in the irradiation device (ID) for large-scale production of plutonium suitable for being used in TRGs by selecting the NpO_2 fuel lattice pitch and surrounding the ID with a heavy moderator layer.

As preliminarily estimated, the replacement of seven uranium FAs in the VVER-1000 reactor core center for seven ID assemblies will lead to the reactor power drop at a level of 5%. This loss is expected to be however made up in part at least via production of ^{238}Pu in kg quantities, the cost of which is estimated at $4000 per gram (The VVER Today: Evolution, Design, Safety). Nevertheless, the authors apprehend the need for justifying further the reactor safety during operation in a dual-purpose mode.

Methods

^{238}Pu generation method

It is proposed that the irradiation device be installed in the VVER-1000 core center (Fig. 1). The key parameters of the VVER-1000 reactor and the FAs are as follows (Shmelev et al. 2020):

- reactor thermal power – 3200 MW;
- number of FAs in the reactor core – 163;
- fuel – enriched uranium dioxide (4.4% of ^{235}U), density – 10.7 g/cm^3;
- fuel cladding – 99%Zr-1%Nb alloy, density – 6.5 g/cm^3;
- coolant – light water, density – 0.73 g/cm^3;
- hexagonal FA flat-to-flat dimension – 23.4 cm;
- fuel pellet diameter – 7.57 mm;
- central hole diameter – 1.40 mm;
- fuel-cladding gap thickness – 0.075 mm;
- cladding thickness – 0.65 mm;
- triangular fuel lattice pitch – 12.75 mm;
- fuel column height – 353 cm.

The irradiation device represents a configuration of seven VVER-1000 FAs. A standard VVER-1000 FA, in which enriched uranium dioxide has been replaced for neptunium dioxide (NpO_2), is accommodated in the ID center (Fig. 2). Neptunium exactly is the most suitable starting material for the ^{238}Pu production (Kuzmin et al. 2015). And the configuration and composition of the six FAs that surround it, have been selected such that to accelerate the production of ^{238}Pu in the NpO_2 FA.

The preferred spectrum (the ^{237}Np resonance region) is formed by way of the ID heterogeneous structure, that is, the FAs that contain moderator with a high atomic weight and a small neutron absorption (Pb, Bi, Pb-Bi eutectics, radiogenic lead, ^{208}Pb) surround the ^{237}Np FA (Fig. 3).

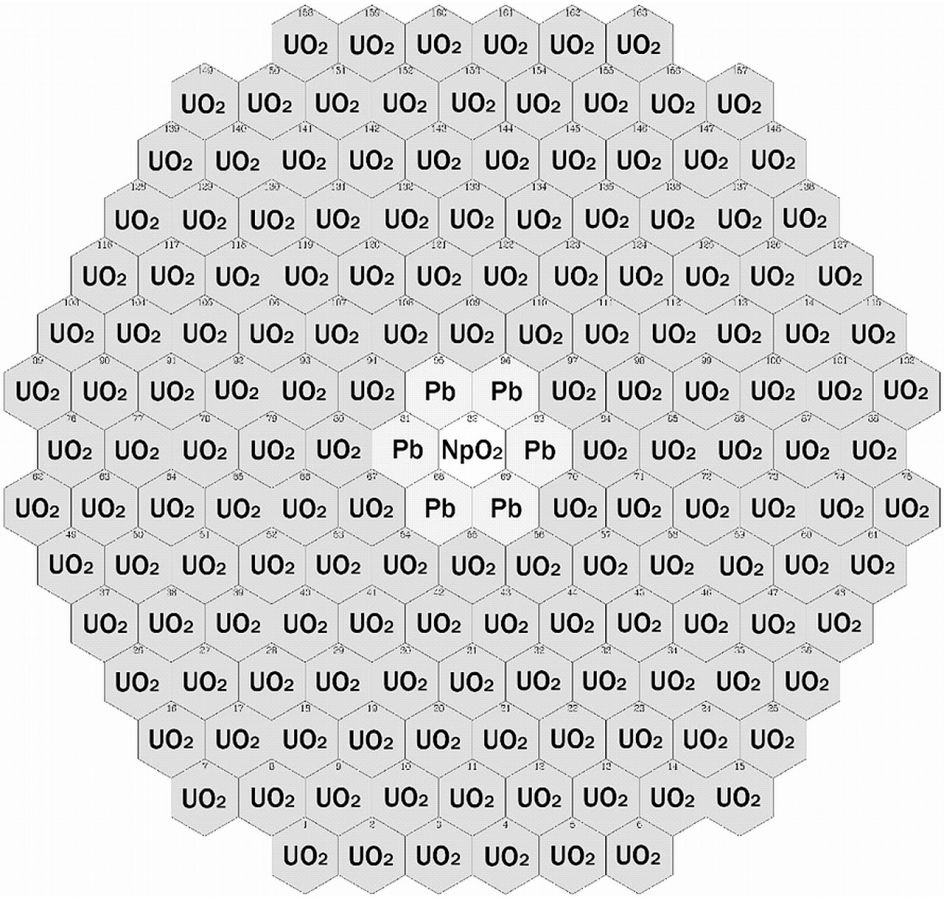

Figure 1. ID position in the VVER-1000 reactor core.

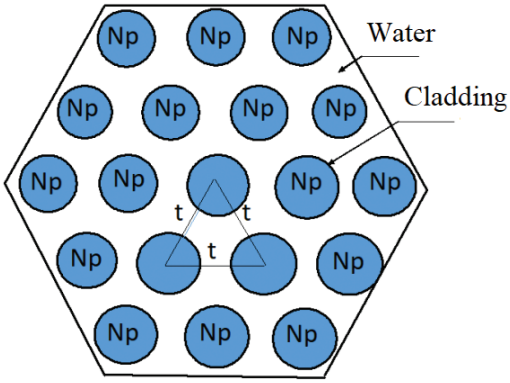

Figure 2. FA with the starting material in the form of NpO_2 (t is the Np rod triangular lattice pitch).

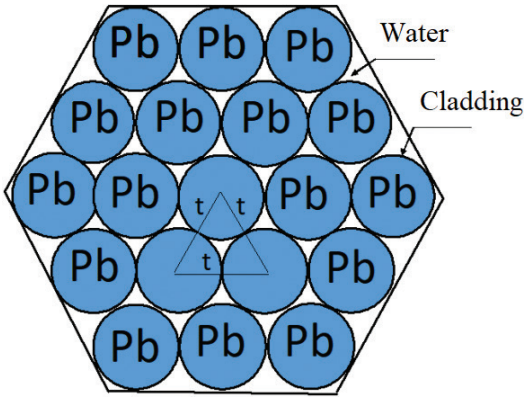

Figure 3. FA with heavy lead moderator (t is the triangular fuel lattice pitch).

The efficient way to form the preferred spectrum is to use ^{208}Pb that is characterized by an extremely small absorption of neutrons and allows, therefore, increasing the neutron flux in the target material. Using it leads thus to accelerated production of the desired nuclide. Using ^{208}Pb also offers other important advantages, including an enhanced Doppler effect and a longer average prompt neutron lifetime.

It has been shown by preliminary estimates that the heterogeneous configuration of the ID makes it possible to create an extensive in-core area with a high neutron flux and with the neutron spectrum preferred for the ^{237}Np irradiation. As a consequence, such method allows efficient and large-scale production of highly conditioned ^{238}Pu.

Mathematical model

The neutronic calculations were based on the TIME26 computer code (Janis 2016) that considers radial models of nuclear facilities in a 26-group diffusion approximation. The constants used in the code have been borrowed from the BNAB evaluated nuclear data library, which is processed by the ARAMAKO-S1 auxiliary program for preparing blocked microscopic cross-sections in each nuclear facility zone.

The geometrical model of the VVER-1000 reactor core represented a system of 8 circular layers of hexagonal FAs isolated from the reactor vessel with a water layer (Fig. 4).

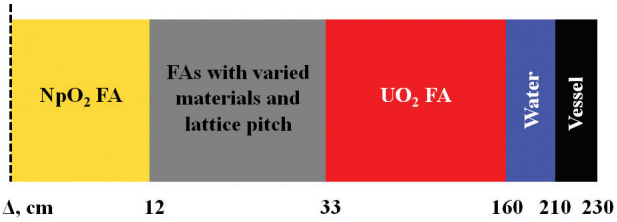

Figure 4. A radial circular model of the VVER-1000 reactor.

Results

In the initial option (No. 1), all 163 FAs were standard VVER-1000 UO_2 FAs (no ID). Further options took into account the availability of the ID, that is, the central NpO_2 FA was surrounded by a layer of six neighboring FAs, which included the following materials:

- Option No. 2 – a layer of standard UO_2 FAs.
- Option No. 3 – a layer of light water.
- Option No. 4 – natural lead rods in Zr-Nb cladding.
- Option No. 5 – ^{208}Pb rods in Zr-Nb cladding.

Two important circumstances were the cause for ^{208}Pb rods having been introduced into Option No. 5. The ^{208}Pb lead isotope is characterized by a very small neutron absorption and by the capability for shifting the delayed neutron spectrum into the resonance region, this expected to accelerate the ^{238}Pu production in the central NpO_2 FA.

For options Nos. 2 through 5, the parameters were estimated which characterize the plutonium production rate and isotopic composition in the central NpO_2 FA. The calculations were conducted in conditions of an invariable FA triangular lattice pitch (12.75 mm). The results are presented in Table 1.

Table 1. Parameters of plutonium production in the VVER-1000 ID

Option No.	Plutonium production rate, kgPu/yr	Fraction of ^{238}Pu/Pu, %	Fraction of ^{236}Pu/Pu, ppm
1	3.50	1.5	0
2	3.92	99.5	158
3	1.11	99.6	51
4	3.76	99.5	21.4
5	4.24	99.3	21.3

It can be seen that introducing NpO_2 FAs into the reactor core with no lead surrounding leads to plutonium production with a substantial fraction of unwanted ^{236}Pu. Introducing water into the surrounding FAs (Option 3) leads to an abrupt decrease in the intensity of ^{238}Pu production. At the same time, introducing lead moderator into the surrounding FAs has improved greatly the plutonium production rate and isotopic composition. However, none of the options considered satisfied the limit for the fraction of ^{236}Pu in plutonium since their respective fractions proved to be much higher than the 2 ppm permitted.

The fraction of ^{236}Pu in plutonium can be apparently reduced through the neutron spectrum mitigation in the central NpO_2 FA, that is, at the expense of reducing the ^{237}Np (n, 2n) ^{236}Pu reaction rate. To this end, the central NpO_2 FA options were calculated, in which the triangular NpO_2 rod lattice pitch was increased with increasing, respectively, the FA water fraction. This is expected to lead to a mitigated spectrum of the starting material irradiation.

Consideration was also given to the produced plutonium parameters as a function of the lead rod dense lattice pitch in the Pb FAs. Table 2 shows the change in the plutonium production rate and isotopic composition with the Pb rod pitch growth for the NpO_2 rod lattice pitch equal to 12.75 mm. The results obtained have shown the lead rod lattice pitch to have minor effect on the produced plutonium parameters. A decision was made therefore for further calculations to be undertaken with an invariable Pb rod lattice pitch (40 mm) to reduce the excessive calculated data.

Table 2. Plutonium production rate and isotopic composition with the Pb rod pitch growth

	Pb rod lattice pitch, mm			
	12.75	20	30	40
Pu mass, kg	3.76	3.83	3.87	3.89
Fraction of ^{238}Pu, %	99.5	99.2	99.2	99.2
Fraction of ^{236}Pu, ppm	21.4	21.1	20.9	20.8
Pu / Np, %	0.78	0.79	0.80	0.80

Table 3 shows the change in the plutonium production rate and isotopic composition with the NpO_2 fuel rod pitch growth with six surrounding Pb FAs.

Table 3. Plutonium production rate and isotopic composition with the NpO_2 fuel lattice pitch growth (six surrounding Pb FAs)

	NpO_2 fuel lattice pitch, mm			
	12.75	20	30	47
Quantity of starting material (Np), kg	484	197	87.5	35.6
Pu mass, kg	3.89	3.59	3.15	2.45
Fraction of ^{238}Pu, %	99.2	98.2	96.0	91.6
Fraction of ^{236}Pu, ppm	20.8	7.5	3.6	1.9

It can be seen that an increase in the neptunium fuel rod spacing (that is, an increased water fraction) leads to the ^{236}Pu content decreasing to the required standard value (2 ppm). The fraction of ^{238}Pu decreases slightly as well while remaining, however, in excess of the produced plutonium quality standard value (^{238}Pu fraction of over 85%).

Due to the specifically attractive properties of ^{208}Pb, calculations were conducted for the ID design options, in which the central NpO_2 FA was surrounded by six ^{208}Pb FAs. The NpO_2 fuel rod spacing (12.75 mm / 20 mm / 30 mm / 44 mm) was varied in the course of the calculations. No ^{208}Pb rod spacing change was considered because of its insignificant effect. The results obtained are presented in Table 4.

For the NpO_2 fuel wide lattice option, as it can be seen in the event of the transition to ^{208}Pb, the produced plutonium mass increases by 25% and the specific production

Table 4. Plutonium production rate and isotopic composition as a function of the NpO_2 fuel lattice pitch growth (six surrounding ^{208}Pb FAs)

	NpO_2 fuel lattice pitch, mm			
	12.75	20	30	44
Quantity of starting material (Np), kg	484	197	87.5	40.7
Pu mass, kg	4.47	4.16	3.70	3.06
Fraction of ^{238}Pu, %	99.1	97.9	95.2	91.0
Fraction of ^{236}Pu, ppm	20.8	7.0	3.3	1.9

grows by 9%. The quality of ^{238}Pu, both for natural lead and ^{208}Pb, meets the criterion that the fraction of ^{236}Pu ≤ 2 ppm.

Fig. 5 shows specific plutonium production (per unit of the loaded neptunium mass) as a function of the increase in the neptunium fuel rod spacing for options with natural lead and ^{208}Pb used in the surrounding FAs. There is an evident growth in specific production of plutonium (by a factor of eight and more) with the Np lattice pitch growth.

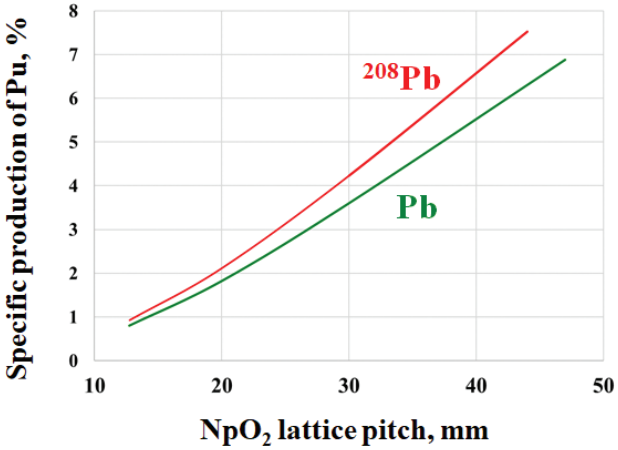

Figure 5. Specific production of plutonium with increased neptunium rod sparing in options with natural uranium and ^{208}Pb used in the surrounding FAs.

Conclusions

An analysis of the above results from the ^{238}Pu production calculation allows for the following conclusions.

1. No moderator around the Np target leads to plutonium being produced with a very large fraction of the unwanted ^{236}Pu isotope (its value is 60 times higher than permitted).
2. Introducing water instead of the surrounding FAs (ID design option 3) leads to the ^{238}Pu production intensity decreasing by four times while reducing greatly, though, the fraction of ^{236}Pu.
3. A layer of ^{208}Pb transmits much more soft neutrons promoting so the accumulation of plutonium in the ID. As the result, the substitution of Pb for ^{208}Pb leads to the plutonium accumulation in the ID increasing to 4.24 kg/yr, the fraction of ^{236}Pu being the same.

4. Introducing lead surrounding for the NpO_2 FA maintains intensive production of plutonium while reducing greatly, at the same time, the fraction of the unwanted ^{236}Pu. The fraction of this isotope remains however ~ 10 times higher than permitted.

5. An increase in the lead rod dense lattice pitch (layer 2 of the radial model), as compared with the standard pitch (h = 12.75 mm), has minor effect on the performance of produced plutonium. This circumstance allowed further calculations to be conducted with an invariable lead rod spacing.

6. A fundamentally essential role is played by the increase in the fraction of water in the NpO_2 FA. The content of ^{236}Pu drops to the required standard value (2 ppm), and the fraction of ^{238}Pu remains in the limits of the standard value in terms of the produced plutonium quality (the ^{238}Pu fraction is over 85%).

7. A major growth in the specific production of plutonium (by a factor of eight and more) is observed as the fraction of water increases in the NpO_2 FA. This leads to the plutonium production in the ID decreasing by a factor of just 1.6 even if the starting material quantity is reduced from 484 kg to 35.6 kg. The plutonium becomes so conditioned (the ^{236}Pu fraction is less than 2 ppm). Substituting natural uranium for ^{208}Pb leads to a further 9% increase in its specific production.

8. In the event of a natural lead layer, the best plutonium parameters are achieved with the NpO_2 fuel lattice pitch being 47 mm. Plutonium is accumulated in the amount of 2.35 to 2.45 kg and contains 91.6 to 91.9% of ^{238}Pu and 1.9 to 2 ppm ^{236}Pu.

9. In the event of a ^{208}Pb layer, the best plutonium parameters are achieved with the NpO_2 fuel lattice pitch being 44 mm. Plutonium is accumulated in the amount of 2.88 to 3.06 kg and contains 91.0 to 91.3% of ^{238}Pu and 1.9 to 2 ppm of ^{236}Pu.

Acknowledgements

The research was supported by a grant from the Russian Science Foundation, project No. 22-22-00287.

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