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Research Article

Possibility for large-scale production of ^{238}Pu for radioisotope thermoelectrical generators in a supercritical light-water reactor*

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Abstract

The paper analyzes the possibility for large-scale production of highly pure plutonium suitable for use in radioisotope thermoelectrical generators in a VVER-SKD light-water power reactor with supercritical coolant parameters. Neptunium dioxide (NpO_2) was used as the starting material. Neptunium can be extracted from minor actinides, i.e. from the main components of radioactive transuranium waste. Large-scale production of useful high-purity plutonium via neutron irradiation of useless radioactive waste can be estimated as one of the possible ways to convert harmful waste to a harmless form. The assembly with the starting material was placed in the central part of the reactor core with the maximum neutron flux to increase the plutonium accumulation. Numerical studies were based on a multi-physical approach with coupled analyses of neutronic and thermal-hydraulic processes in the downward and upward reactor zones. The results obtained in numerical studies have demonstrated that large quantities of high-purity plutonium with a low content of ^{236}Pu and a high content of ^{238}Pu could be accumulated in the central NpO_2 assembly if the following conditions are satisfied. Firstly, it is desirable that the lattice of NpO_2-containing rods would be sufficiently wide with a high volume fraction of light-water coolant. Secondly, the central NpO_2 assembly needs to be surrounded by protective assemblies to form a barrier against high-energy fission neutrons which are capable to intensify the threshold ^{237}Np(n,2n)^{236}Pu reaction. The protective function can be performed by the assemblies containing rods with natural lead or radiogenic lead. The paper estimates the production scales of high-purity plutonium in the central assembly with a wide lattice of NpO_2-containing rods and surrounded by a layer of protective Pb-containing assemblies.

Keywords

^{236}Pu, ^{238}Pu, radioisotope thermoelectrical generators, light-water power reactors with supercritical coolant parameters

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Introduction

The value of low-background plutonium with a high content of ^{238}Pu (according to the NASA specification (Wong, 2001), there should be not more than 80 % ^{238}Pu in plutonium) is explained by unique properties of this isotope. Intensive heat emission by ^{238}Pu (570 W/kg) can be converted to electricity with an efficiency of 10 to 15 % in radioisotope thermoelectric generators (RTG), which provide energy for space vehicles. The half-life of ^{238}Pu (87.7 years) allows one to expect their energy supply will be long-term and stable.

The capacity currently available worldwide for producing this isotope is insufficient and does not cover the growing annual demand (Start-up Plan for Plutonium-238 Production for Radioisotope Power Systems, 2015). Normally, it is produced in research reactors. As of 2019, the output of ^{238}Pu at two research reactors at the Idaho and Oak Ridge National Laboratories was estimated at 800 to 900 g per year. The NASA program provides for the scale of the isotope production to be increased to 1.5 kg/year by 2025 (Sutliff et al. 2019). In this case, the composition of produced plutonium is expected to be as required by energy sources for space exploration (Wong, 2001). In the Russian Federation, one of the leading manufacturers of ^{238}Pu is PO Mayak. Currently, PO Mayak has two Generation III research reactors, *Ruslan* and *Lyudmila*. Both feature unique neutronic performance and allow obtaining a broad range of radioactive isotopes for civilian applications. In September 2015, PO Mayak began to develop a new reactor designed as a multipurpose facility with the ability to generate electricity and produce a broad variety of radioisotopes, including ^{238}Pu (The Mayak plant has begun designing a new research reactor, 2015).

Basically, plutonium can be used in charges of nuclear explosive devices (NED). However, it is exactly plutonium with a major fraction of ^{238}Pu that is unfit for being used in NEDs due to its high heat liberation value. According to the IAEA's INFCIRC/153 (INFCIRC/153, 2008), plutonium with a ^{238}Pu fraction of over 80 % cannot be used in NEDs and is exempted from the IAEA safeguards. It is exactly such plutonium that is required for space-borne RTGs.

Currently, the key starting materials for production of low-background plutonium in nuclear reactors are different ^{237}Np compounds. Neutrons that bombard this isotope are capable to initiate a useful ^{237}Np(n,γ)^{238}Pu reaction leading to the accumulation of the desired ^{238}Pu isotope and a needless ^{237}Np(n,2n)^{236}Pu reaction leading to the accumulation of the unwanted ^{236}Pu isotope (Shmelev et al. 2020). The fact that the presence of ^{236}Pu is not desired in plutonium for RTGs is explained by the activity of its decay products, which include high-power sources of hard gamma radiation (e.g. ^{208}TI). Plutonium is believed to be suitable for spacecraft RTGs if it contains not more than 2 ppm of ^{236}Pu (Wong, 2001).

It seems reasonable to produce low-background plutonium in Np-containing FAs irradiated by thermal neutrons in light-water power reactors. A soft neutron spectrum of light-water reactors can enhance the useful ^{237}Np(n,γ)^{238}Pu reaction and suppress the needless ^{237}Np(n,2n)^{236}Pu reaction, which only fast neutrons are capable to initiate. Estimates are provided in Shmelev et al. 2022 for the possibility of low-background plutonium production in the VVER-1000 light-water power reactor. These estimates showed that the reactor was capable to produce low-background plutonium at a rate of up to 3 kg/year in a central FA with neptunium dioxide used as the starting material. The produced plutonium contains ^{238}Pu at a level of not less than 90 % and ^{236}Pu at a level of not more than 2 ppm.

VVER-SKD reactors are promising largely due to being more efficient than traditional VVER-type reactors. In the event of a nuclear power plant with a reactor operating at a water pressure of 25 MPa and at an outlet temperature of about 600 °C, the efficiency value can reach 48 % against 30 % to 32 % in the event of a nuclear power plant with VVER-type reactors. Higher efficiency means less waste heat and a smaller environmental impact from VVER-SKD reactors. In addition, VVER-SKD reactor designs feature both acceptable safety characteristics and specific capital expenditures (Lapin et al. 2020).

It is exactly for this reason that light-water reactors with supercritical coolant parameters are currently viewed as one of the most promising options for the evolution of reactor technologies. Notably, international experts list light-water SKD reactors among six promising Generation IV reactor systems (Generation IV International Forum, 2001). Also noteworthy is that specifically important to the development of reactors with supercritical water parameters is that the existing industrial and infrastructural capabilities can be used to the maximum extent for the light-water reactors in operation and under construction.

The study assesses the ability of light-water reactors with supercritical coolant parameters to produce large quantities of low-background plutonium for space-borne RTGs.

Multi-physical model of a VVER-SKD reactor

The coolant used in the VVER-SKD reactor is water with supercritical parameters heated to 260 °C, and the core axial water density and enthalpy change several-fold (Deev et al. 2015). This leads to the neutron spectrum changing heavily from resonant at the core coolant inlet to fast at the outlet. All these circumstances indicate the need for using a multi-physical model of coupled neutron and thermal-hydraulic processes to describe correctly the reactor characteristics.

This study uses a multi-physical approach of coupled neutron and thermal-hydraulic processes as a converging sequence of axial thermal-physical and neutronic calculations of the SKD reactor's downward and upward zones (Fig. 1).

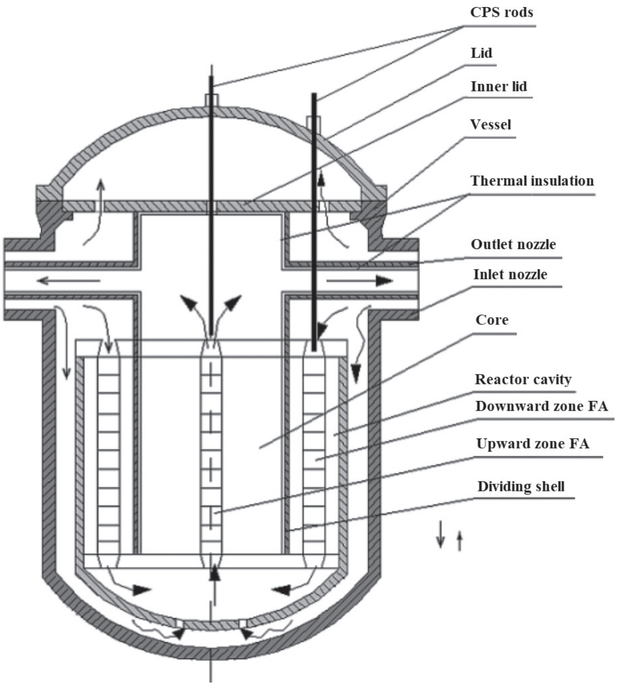

Figure 1. Two-way reactor cooling flow diagram (Glebov and Klushin 2014).

The calculations were based on the design of the VVER-SKD-1600 reactor facility (Deev et al. 2015; Kirillov and Bogoslovskaya 2019) with a two-way flow diagram of coolant movement in the core (Fig. 2). Key parameters of the VVER-SKD-1600 core:

- thermal capacity – 3682 MW;
- water pressure – 24.5 MPa;
- downward zone water temperature – 280 to 382.9 °C;
- upward zone water temperature – 382.9 to 540 °C;
- downward zone FA number – 109;
- upward zone FA number – 132;
- FA shape – hexagonal;
- FA flat-to-flat dimension – 20.5 cm;
- fuel lattice pitch – 12 mm;
- kernel diameter – 9.6 mm;
- fuel core material – uranium dioxide;
- fuel rod active part length – 400 cm;
- fuel cladding thickness – 0.55 mm;
- fuel cladding material – EP-172 stainless steel;
- mass velocity of water, ρ_W, for downward zone – 164.3 g/(cm^2·s);
- mass velocity of water, ρ_W, for upward zone – 181.5 g/(cm^2·s).

Estimates for the possibility of large-scale low-background plutonium production in a VVER-SKD-1600 light-water reactor were obtained using the TIME26 code (Kuzmin et al. 2015) and the WaterSteamProCalculator code (WaterSteamProCalculator Online Software). Using these codes made it possible to analyze comprehensively, as was required, the neutronic and thermal-physical processes in the reactor.

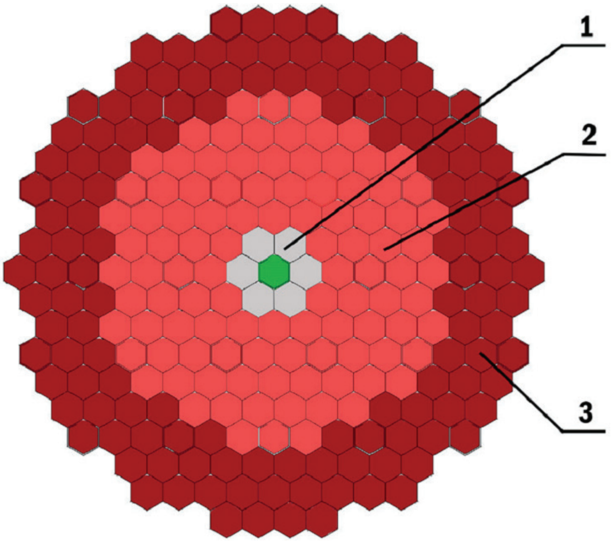

Figure 2. VVER-SKD reactor core layout taken as a basis for coupled thermal-hydraulic and neutronic calculations (Deev et al. 2015): 1 – irradiation device; 2 – upward zone; 3 – downward zone.

The TIME26 code is designed to determine the spatial energy distribution of the neutron flux and analyze the time-dependent behavior of the fuel isotope composition as part of a 26-group diffusion approximation for axial and radial models of nuclear reactors. The code uses group microscopic cross-sections of neutron reactions from the BNAB-78 library of evaluated nuclear data (Abagyan et al. 1981).

The WaterSteamProCalculator code allows one to obtain temperature dependencies of the water density and enthalpy as water moves along the FA in the reactor core downward and upward zones. Temperature dependencies of the water enthalpy and density were added to the TIME26 code in the form of tabulated arrays.

Algorithm for coupled thermal-physical and neutronic calculations

The axial water enthalpy, temperature and density distribution in the FA in the downward and upward zones were calculated in the following sequence. At the initial stage, neutronic calculations are performed for one-dimensional axial models of FAs in the reactor core downward and upward zones. These calculations made it possible to obtain an axial dependence of the FA linear power density, $q_{L\text{-}FA}(z)$, and to use it to determine the FA axial distributions of the water enthalpy, temperature and density.

The axial dependences of the water enthalpy, temperature and water density were calculated using the following iterative sequence.

1. The initial simplified distribution of the water density along the FA height was determined. For example, the water density was assumed to be the same

throughout the FA height and equal to the water density at the inlet temperature.

2. Neutronic calculation was undertaken, as a result of which the axial dependence of the FA linear power density, $q_{l,\mathrm{FA}}(z)$, was determined. This dependence was used to calculate the water enthalpy distribution along the FA height:

$$J(z) = J(0) + \int_0^z \frac{q_{l,FA}(\dot{z})d\dot{z}}{\rho_W \cdot \varepsilon(H_2O) \cdot S_{FA}}$$

where $J(z)$ is the water enthalpy, J/g; $q_{l,\mathrm{FA}}(z)$ is the linear power density, W/cm; ρ_W is the mass velocity of water, g/(cm^2·s); $\varepsilon(H_2O)$ is the volume fraction of water in the FA; and S_{FA} is the FA cross-section area, cm^2.

Calculating the water enthalpy distribution allows one to determine the water temperature distribution using the tabulated arrays introduced into the code.

The FA axial linear power density was normalized such that the water temperature at the FA outlet corresponds to the specified values (382.9 °C at the downward zone outlet and 540 °C at the upward zone outlet).

3. Knowing the axial water temperature distribution makes it possible to find the new distribution of water density along the FA height using the above tabulated dependencies of water density and enthalpy on its temperature.

After the updated distribution of water density along the FA height is found, the sequence of calculations went back to the neutronic calculation of the new axial distribution of the FA linear power density, $q_{l,\mathrm{FA}}(z)$. Axial dependencies of the water enthalpy and temperature were calculated again. Such iterative process continued until the axial distribution of water temperature stabilized.

The obtained axial dependencies of water density were used to determine the average values of water density in the downward and upward zone FAs.

Calculations of plutonium accumulation in the central fa of the reactor core cylindrical model

The average water densities in the downward and upward zone FAs obtained at the previous stage were used for the computational analysis of the time-dependent behavior of the space-energy distribution of the neutron flux density and the fuel isotope composition in the cylindrical model of the reactor core.

Neptunium dioxide was considered as the starting material for the accumulation of low-background plutonium, NpO_2. Steel-clad fuel elements with neptunium dioxide were installed in the central FA. Variable parameters, changed to create the best spectral conditions for production of low-background plutonium, were the NpO_2 fuel lattice pitch, $t(NpO_2)$, and the composition of FAs in the immediate vicinity of the central FA.

Light water, natural lead and radiogenic lead with a high content of the ^{208}Pb isotope were used as materials for the central FA surrounding.

Increasing the NpO_2 fuel lattice pitch will increase the volume fraction of water in the central FA, soften the neutron spectrum, increase the accumulation rate of the valuable ^{238}Pu isotope through the ^{237}Np(n,γ)^{238}Pu reaction, and slow down the accumulation rate of the unwanted ^{236}Pu isotope through the ^{237}Np(n,2n)^{236}Pu reaction.

Surrounding the central NpO_2 FA with a layer of six FAs containing light water or lead (natural or radiogenic) creates a barrier against the penetration of fast fission neutrons from the reactor's main UO_2 FAs capable of intensifying the unwanted ^{237}Np(n,2n)^{236}Pu reaction. In all versions of NpO_2 fuel rods differing in the fuel lattice pitch and the central NpO_2 FA differing in the nearest surrounding, the fraction of ^{235}U in the reactor's UO_2 FA was selected such that the effective neutron multiplication factor at the beginning of the NpO_2 FA irradiation cycle was equal to $k_{\mathrm{eff}} = 1.10$.

The results of calculating the plutonium accumulation rate and the isotope composition are presented in Tables 1 through 3.

The data in Tables 1–3 allows making the following conclusions.

Table 1. Central FA surrounded by six water-filled FAs

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	9.27	99.7	387.0	1.13
20	7.01	99.5	118.7	2.37
40	6.20	97.3	35.5	8.36
60	5.64	92.2	21.6	17.1
80	4.98	**85.8**	**17.0**	26.9

Table 2. Central FA surrounded by six FAs with natural lead

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	7.90	99.7	158.5	0.96
20	7.22	99.4	23.5	2.43
40	6.49	96.9	4.24	8.76
60	5.54	90.8	2.29	16.9
80	4.49	**84.0**	**1.82**	24.3

Table 3. Central FA surrounded by six FAs with radiogenic lead (100 % ^{208}Pb)

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	8.72	99.6	208.2	1.06
20	7.37	99.4	28.7	2.49
40	6.55	96.8	4.61	8.84
60	5.67	90.4	2.40	17.2
80	4.66	**83.1**	**1.91**	25.2

1. The results of calculating the ^{238}Pu accumulation in the VVER-SKD reactor has shown that the isotope accumulation rate was ~ 4.7 kg/year, which is much higher than in the classic VVER-1000 reactor (~ 3.0 kg/year, (Shmelev et al. 2023)).

2. The major difficulties in the accumulation of low-background plutonium are caused not by the failure to observe the ^{238}Pu fraction limit (not less than 80 %) but rather by observing the ^{236}Pu fraction limit (not more than 2 ppm).
3. Light water is a weak barrier against fast fission neutrons produced by the reactor's main UO_2 FAs and penetrating into the central NpO_2 FA. As a result, plutonium is inadmissibly heavily contaminated by the ^{236}Pu isotope.
4. Natural lead has proved to be a stronger barrier against fast fission neutrons as compared to water and radiogenic lead. A record-breaking weak neutron absorption by the ^{208}Pb isotope, which plays a positive role when using radiogenic lead as coolant instead of natural lead, turned a drawback in this case. Radiogenic lead is too transparent for fast fission neutrons, which have increased the ^{236}Pu accumulation rate in the central NpO_2 FA.

In addition, radiogenic lead is transparent for resonant and thermal neutrons capable of intensifying the accumulation of the fissile ^{239}Pu isotope in the central assembly. Fast fission neutrons of this isotope intensify the ^{237}Np(n,2n)^{236}Pu reaction and increase the fraction of ^{236}Pu in produced plutonium.

The possibility of increasing the accumulation rate of low-background plutonium was also considered, making the barrier of six FAs more transparent by introducing a minor fraction of water (10%) that flows about lead rods. The results of calculations with such central NpO_2 FA surrounding are presented in Table 4.

Table 4. Six FAs around the NpO_2 FA contain 90 % lead and 10 % water

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	9.89	99.6	159.1	1.20
20	8.44	99.1	26.8	2.85
40	7.63	95.3	5.20	10.3
60	6.56	87.6	2.96	19.9
80	5.33	**79.2**	**2.44**	28.8

It can be seen that even such a small fraction of water in the six FAs that surround the NpO_2 FA weakens noticeably the protective capabilities of these FAs. Naturally, the plutonium accumulation rate increased by about 13 % compared to the case of no water (5.3 kg/year against 4.7 kg/year), but the plutonium isotope composition does not satisfy the requirements for either the ^{236}Pu content or the ^{238}Pu content.

The protective barrier against the fast fission neutrons produced by the reactor's main fuel assemblies can be reinforced by adding another layer of 12 assemblies around the central NpO_2 FA. This FA will be then surrounded by 18 assemblies containing natural lead and water. The results of calculations with the thicker central NpO_2 FA surrounding are presented in Tables 5, 6.

Table 5. 18 assemblies around the NpO_2 FA contain 90 % lead and 10 % water

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	9.44	99.3	43.7	1.15
20	8.76	98.1	5.72	2.96
25	8.49	97.2	2.99	4.47
29.8	8.22	**96.0**	**2.00**	6.16

Table 6. 18 assemblies around NpO_2 -TVS contain 78 % lead and 22 % water

$t(NpO_2)$, mm	Pu, kg/year	^{238}Pu/Pu, %	^{236}Pu/Pu, ppm	Pu/Np, %
12	11.27	98.6	34.2	1.37
20	10.31	96.6	6.27	3.48
30	9.47	93.0	2.83	7.19
40	8.58	88.8	2.16	11.6
50	7.64	**84.4**	**2.00**	16.1

It can be seen that the barrier of 18 assemblies with natural lead and water makes it possible to increase greatly the accumulation rate of low-background plutonium in the central NpO_2 FA. The accumulation rate is expected to be 7.6 to 8.2 kg/year depending on the volume fraction of water. At the same time, the plutonium produced satisfies the declared requirements for the content of ^{236}Pu and ^{238}Pu. With a volume fraction of water in excess of 22 %, it becomes however impossible to satisfy both requirements at a time. If the ^{238}Pu content is below 80 %, the ^{236}Pu content remains over 2 ppm.

Conclusion

1. A multi-physical model of coupled neutronic and thermal-hydraulic calculations is proposed, taking into account major nonlinear changes in the thermal-physical properties of water along the VVER-SKD-1600 reactor core height. Using this model made it possible to estimate the output of low-background plutonium more closely to the actual situation in the reactor core.
2. The estimates for production of low-background plutonium in a VVER-SKD-1600-type reactor have allowed making the following conclusions:

   - it has been shown to be conceptually possible to produce large quantities of low-background plutonium (at a level of 7–8 kg/year) that satisfies the requirements with respect to plutonium for space-borne RTGs in a light-water reactor with supercritical coolant parameters;
   - it is possible to produce large quantities of low-background plutonium in the central FA containing neptunium dioxide only with this assembly being surrounded by a sufficiently dense protective barrier against fast fission neutrons. Such barrier can be formed by assemblies containing natural lead rods.

3. It is important to note that the accumulation of plutonium with a high content of ^{238}Pu (over 80 %) will be accompanied by the formation of a large fraction of the ^{239}Pu isotope with perfect fissionable properties (up to 20 %). Fast fission neutrons of this isotope are capable to intensify the threshold ^{237}Np(n,2n)^{236}Pu reaction and increase the content of the unwanted ^{236}Pu isotope. Assessing such “self-contamination” effect of low-background plutonium may require further investigations.

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