The problem of large-scale production of plutonium-238 for autonomous energy sources

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Abstract. The article is devoted to the urgent problem of large-scale production of plutonium-238. Various starting isotopes and Pu-238 production schemes are analyzed. The isotope characteristics of the chain based on the starting Np-237 isotope are presented. It has been substantiated that the region of neutron resonant absorption for the isotope Np-237 is the preferred spectrum of its irradiation. It is proposed to form a region with a preferred neutron spectrum in the fast reactor core for irradiation of neptunium-237. The preferred spectrum is achieved by a heterogeneous target structure including neptunium-237 and a moderator with a high atomic weight (Pb, Pb-208, Bi, Pb-Bi eutectic). It is shown that when a moderator with a high atomic weight is used, it becomes possible to form an extensive region with the preferred spectrum. In conclusion, the main conditions for the large-scale production of isotopically pure Pu-238 in a fast reactor are formulated.

1. Introduction
Pu-238 is one of the most demanded radionuclides for heat generation because:

- has a high energy release (570 W/kg);
- is a long-term source of heat ($T_{1/2} = 88$ years);
- characterized by a low radiation background (there is no hard $\gamma$-radiation).

The plutonium-238 market is actively developing in connection with the large-scale plans of many countries for the development of remote regions of the Earth and space. Such a prospect will require the creation of numerous sources of autonomous power supply.

At present, Pu-238 is being produced in the United States at research reactors at the Idaho and Oak Ridge national laboratories [1]. Meanwhile, the scale of production is about 400 grams per year. However, the expected demand for plutonium-238 is at least several times higher than the current production rate.

With regard to the Russian Federation, potential consumers of Pu-238 or autonomous energy sources based on this radioisotope may be: State Corporation Rosatom, RSC Energia named after S.P. Korolev and other corporations. At the same time, the prospect of interaction with foreign consumers of Pu-238 and, in general, the development of a part of the world market for sources of long-term autonomous power supply is also relevant. Here, the Russian Federation has a successful experience of earlier work.
on the production of plutonium-238. An example is the five-year agreement signed in 1992 for the purchase of the Pu-238 isotope by the United States from Russia. As of 2005, the American side purchased approximately 16.5 kg of this isotope.

2. Isotope schemes for the production of plutonium-238

As is known, the isotope of plutonium-238 is produced in uranium fuel in nuclear reactors simultaneously with other isotopes of plutonium. However, the proportion of plutonium-238 in plutonium is usually only about 2%. Of course, it is desirable that the target plutonium-238 isotope be accompanied by a minimum of other plutonium isotopes. Therefore, plutonium-238 is produced from special starting materials. The nearest fairly stable nuclides, neutron irradiation of which leads to the formation of plutonium-238, are neptunium-237 \((T_{1/2} = 2.1 \times 10^6\) years) and americium-241 \((T_{1/2} = 433\) years). These isotopes are in significant amounts in the spent fuel of power reactors (SNF) and belong to minor actinides that must be transmuted due to their long-term radiotoxicity. Figure 1 shows a schematic diagram of the technological process for the production of the isotope Pu-238 based on these starting materials.

![Figure 1](image1.png)

**Figure 1.** Scheme of Pu-238 production in the target complex of a fast reactor.

2.1. Irradiation of americium-241

The amount of americium produced by the Russian nuclear power industry was about 15 tons in 2017. The most (11 tons) are stored in unprocessed SNF from VVER-1000 and RBMK reactors. Currently, the accumulation rate of americium is about 700 kg / year [2]. Forecast estimates say that by 2050 the total amount of americium will be about 48 tons, and will increase at a rate of about 1300 kg / year. Thus, the accumulation of americium in the spent nuclear fuel of power reactors will reach a significant amount by the middle of this century, which will require the adoption of special measures on americium utilization, for example, in fast reactors.

It should be noted that the decay of Pu-241 makes the overwhelming contribution to the accumulation of americium, as a result of which the accumulated americium is largely represented by the isotope Am-241. The share of Am-241 in a mixture of americium isotopes is over 80%. The remaining isotopes are Am-242 and Am-243.

Figure 2 shows a chain of nuclide transformations of Am-241 into Pu-238 [3].

![Figure 2](image2.png)

**Figure 2.** Scheme of obtaining Pu-238 by irradiating Am-241 in a reactor. The "X" indicates an unwanted channel in the Pu-238 production process.
It is seen that the Pu-238 production chain is short and contains unwanted irradiation channels only for the target nuclide. The latter indicates the high potential of this scheme for producing Pu-238. On the other hand, americium-241 is radioactive. The half-life of Am-241 is 433 years. Intense gamma radiation (59.5 keV) accompanies the α-decay of Am-241. The Am-241 isotope seriously complicates the radiation environment when handling materials. The addition of a noticeable amount of americium to the material significantly (by an order of magnitude or more) increases the level of its γ-radiation and imposes certain restrictions on the applied technologies for handling this material.

Unlike Am-241, the radiation characteristics of materials containing Np-237 remain close to the level of material without it. In addition, the use of Np-237 as an irradiated material serves to reduce the risk of its unauthorized use, because it has practically no spontaneous fission, and its bare critical mass (~ 60 kg) is comparable to that of uranium-235. In connection with the above mentioned, further, neptunium-237 is considered as the irradiated material for the production of Pu-238.

2.2. Scheme of the production of plutonium-238 by irradiation of neptunium in a reactor

Figure 3 shows a chain of nuclear reactions that promote or impede the conversion of neptunium-237 to plutonium-238. These are reactions of radiative capture of neutrons (n, γ), fission reactions (n, f) and β- decays [3]. Undesirable reactions that do not lead to the formation of plutonium-238 are marked with red crosses.

![Figure 3. Chain of isotopic transitions during irradiation of Np-237. The "X" indicates an unwanted channel in the Pu-238 production process.](image)

It is seen that the chain of isotopic transitions is even shorter than for the starting isotope Am-241. However, the task of producing plutonium-238 from Np-237 is being solved in highly contradictory conditions (many undesirable channels). The main thing is to find the flux density and neutron spectrum of neptunium-237 irradiation (preferred spectrum) in order to satisfy the following conditions:

- neptunium-237 should mainly capture neutrons, turning into neptunium-238, and not fission;
- neptunium-238 should mainly undergo β- decay and transform into plutonium-238, and not fission and capture neutrons. The latter imposes a limitation on the acceptable value of the neutron flux in the target material;
- plutonium-238 should fission and capture neutrons as little as possible so that its mass does not decrease and heavier isotopes of plutonium are not formed.

The search for the preferred spectrum included characterization of each isotope in the plutonium-238 production chain.

3. Analysis of the process of plutonium-238 production by irradiation of Np-237

3.1. Characteristics of Np-237 as a starting material for the production of Pu-238
Figure 4 shows the energy dependences of the microsections for the radiative capture of neutrons and fission of neptunium-237. Microsections are taken from the JENDL-4.0 Evaluated Nuclear Data Library [4].

It can be seen that in order for the initial irradiated material, neptunium-237, to mainly capture neutrons, and not uselessly fission, it is desirable that the neutron spectrum, at least, be below 0.1 MeV. And besides, with a decrease in the neutron energy, there is a favorable tendency for the ratio of the capture-to-fission cross section to increase, reaching four orders of magnitude at the thermal point.

![Image of Figure 4](image_url)

**Figure 4.** Microsections of radiation capture and fission for neptunium-237.

This means that the spectrum of neutrons, which irradiate neptunium-237, should be softer than in a fast neutron reactor. For example, in the region of resonant absorption at Np-237 (1-600 eV), the ratio of radiative capture to fission cross sections is 3-4 orders of magnitude, and the resonance integral is 700 barn. At the same time, the region of thermal and epithermal neutrons is not suitable for irradiation of the target material due to the intense consumption of the target nuclide there (see Figure 6). Thus, the region of resonant absorption of Np-237 can be considered as the preferred spectrum for its irradiation.

### 3.2. Radiation capture and fission processes on neptunium-238 and plutonium-238

Figure 5 shows the energy dependences of microsections for fission and radiative capture of neutrons for neptunium-238 [4]. It can be seen that these microsections for neptunium-238 fall with energy, approximately, according to the law $1/E^{1/2}$. At the same time, both microsections reach the highest values in the thermal region.
Figure 5. Microsections of the radiation capture and fission of neptunium-238.

The isotope neptunium-238, as an intermediate nuclide in the chain, should predominantly decay into plutonium-238 and fission and absorb neutrons as little as possible. That is, we have a restrictive condition regarding the rates of interaction processes at $^{238}$Np:

$$\lambda = \frac{\ln 2}{T_{1/2}(^{238}\text{Np})} \gg \left[ \sigma_c(^{238}\text{Np}) + \sigma_f(^{238}\text{Np}) \right] \cdot \varphi$$

(1)

Condition (1) implies two simple conclusions:
1. The region of thermal and epithermal neutrons is extremely undesirable, since here the cross sections $\sigma_c(^{238}\text{Np})$ and $\sigma_f(^{238}\text{Np})$ reach their maximum values;
2. Condition (1) essentially imposes an upper limit on the neutron flux in the target complex. At the same time, estimates show that under conditions of a preferred spectrum (1–600 eV), even for neutron flux levels of a fast reactor ($\sim 10^{15}$ n/(cm$^2 \times$ s)), the electron decay channel is dominant. This conclusion is an important argument in choosing a fast reactor for Pu-238 production.

Figure 6 shows the energy dependences of microsections responsible for the loss of the target isotope plutonium-238 (cross sections for radiative capture of neutrons and fission [4]). It is seen that for Pu-238, both types of interactions are significant over the entire energy range.
We strive for the minimum consumption of the target nuclide:

\[
\text{Min}\left\{\sigma_c^{(238Pu)} + \sigma_f^{(238Pu)}\right\} \cdot \varphi \right) \tag{2}
\]

Therefore, it is advisable to have a neutron spectrum in the region of the minimum of its cross sections, i.e. in the range from about 10 eV to 100 eV. Since it is practically impossible to keep the neutron spectrum in such a narrow energy range, the recommended range can be extended to the energy range from 1 eV to 600 eV, that is, to the resonant absorption region of Np-237. Figure 6 shows that here the minimum (2) is approximately observed and the cross sections \(\sigma_c\) and \(\sigma_f\) remain at the level of 10 barn and below.

3.3. Controlling the isotopic purity of the target nuclide

Radiative neutron capture by Np-238 and Pu-238 isotopes are channels for the appearance of non-target plutonium isotopes in the target material with Np-237. Therefore, the control of the isotopic purity of Pu-238 is to reduce the parasitic radiation capture of the isotopes Np-238 and Pu-238.

As follows from Figures 5-6, the microsections of the radiative capture of Np-238 and Pu-238 increase significantly with decreasing neutron energy, reaching large values in the thermal region (at the thermal point, almost 1000 barn). Therefore, in order to weaken the radiative capture of neutrons by these isotopes, it is proposed to screen the target material from epithermal and thermal neutrons. For this purpose, it is planned to use strong slow neutron absorbers in the target material area. These absorbers should capture only slow neutrons and should be much superior in this relation to Np-238 and Pu-238, since otherwise the isotopic purity of Pu-238 would be significantly reduced.

Figure 7 shows, for comparison, the energy dependences of microsections for radiative capture of neutrons by various absorbers of slow neutrons.
Figure 7. Microsections of the neutron radiative capture of Np-237, B-10, Cd-113, Gd-157 and Pu-238.

It can be seen that, for example, gadolinium-157 and cadmium-113 are suitable for the aim mentioned above. By adjusting the content of these absorbers around and inside the irradiated material containing neptunium-237, it will be possible to influence the degree of isotopic purity of the plutonium-238 being produced.

4. Method of plutonium-238 production and influence of the production process on the reactor safety

It is proposed to place the target complex in the fast reactor with a light / heavy liquid metal coolant [5, 6]. A region with a preferred neutron spectrum for irradiation of neptunium-237 (Np-237 resonance region) is formed in the target complex. The preferred spectrum is achieved by the heterogeneous structure of the target complex. That is, a moderator with a high atomic weight and low absorption of neutrons (Pb, Bi, Pb-Bi eutectic, radiogenic lead, Pb-208) is placed around the Np-237 target.

For a formation of the preferred spectrum, it is especially effective to use Pb-208, which is characterized by an extremely low absorption of neutrons and, therefore, makes it possible to increase the neutron flux density in the target material. Its application, thereby, accelerates the production of the target nuclide. The use of Pb-208 offers other important advantages, including an increase of the Doppler effect and the average lifetime of prompt neutrons.

Figure 8 shows an example of a annular arrangement of a target complex in a fast reactor. The central part is fuel assemblies of the low enrichment zone. Three rows of assemblies of the target complex are located inside the zone of medium enrichment. Next comes the large enrichment zone and side blanket.

Preliminary calculations have shown that the heterogeneous configuration of the target complex makes it possible to create an extensive area in a fast reactor:
- with high neutron flux;
- with a preferred neutron spectrum for Np-237 irradiation.

As a consequence, with this method there is an opportunity for efficient and large-scale production of Pu-238 of high isotopic purity.
As can be seen from Figure 8, the placement of the target complex inside the core of a fast reactor makes it possible to divide the core into two parts with neutron coupling through the target complex. At the same time, the positive void reactivity effect of the coolant is leveled. Of course, it must be said that placing a target complex in a fast reactor is a separate optimization problem determined by many specific conditions (for example, the amount of available starting material). The authors' immediate plans are to study this and other safety issues related to the operation of a fast reactor in a dual-purpose mode.

5. Conclusion

It is shown that for a large-scale production of isotopically pure Pu-238 from the starting material Np-237 in a fast reactor, the following set of favorable conditions is required:

- High neutron flux density for fast production of the target nuclide;
- Resonance spectrum of neutrons, to intensify the radiative capture of Np-237 and avoid its useless fission, as well as to reduce the radiative capture / fission of Pu-238;
- A moderator with a heavy atomic weight in the target complex (lead, lead-208, bismuth), which is characterized by a small step of neutron moderation and allows the formation of the required resonance spectrum in the target;
- The existence in the target complex of an extended region with a resonance neutron spectrum (due to the use of extremely heavy moderators);
- The presence of strong absorbers of slow neutrons (Cd-113; Gd-157) around / inside the irradiated material to increase the isotopic purity of Pu-238.

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