INVESTIGATION OF CHARACTERISTICS OF FAST POWER REACTOR WITH NATURAL AND RADIOGENIC LEAD AS COOLANT FOR LARGE-SCALE PRODUCTION OF PLUTONIUM-238

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Abstract

A dual-purpose fast reactor with heavy liquid metal coolant and a large-scale production of plutonium-238 is considered. A universal target complex for large-scale production of Pu-238 is located inside the reactor. Np-237 is considered as a starting material for the production of Pu-238. Np-237 is in a significant amount in the spent fuel of light water reactors and does not exhibit significant gamma radiation, unlike americium-241. The target complex has a heterogeneous structure, including neptunium-237 and a moderator with a high atomic weight. In the target complex, an area is formed with favorable conditions for the production of plutonium-238 of the required isotopic purity:
- High neutron flux density for intensive irradiation of the starting material (typical for fast reactors);
- Resonance spectrum of neutrons, to intensify the radiative capture on Np-237 and avoid its useless fission, as well as reduce the radiative capture / fission of plutonium-238;
Use of a moderator with a high atomic weight and low absorption of neutrons around the Np-237 target (Pb, Bi, Pb-Bi eutectic, radiogenic lead, Pb-208) allows to form a vast region with neutron spectrum concentrated near resonance neutron spectrum. This circumstance determines the scale of the production of plutonium-238. It is especially useful for the formation of the resonance spectrum to use Pb-208, which is characterized by extremely low absorption of neutrons.
The scale of Pu-238 production is, of course, determined by the resource of Np-237 separated from SNF. Nevertheless, the proposed method for the production of Pu-238 will make it possible to obtain in the target complex of a fast reactor up to several kilograms of Pu-238 per year compared to ~500 g/year currently being produced in research reactors. The use of Np-237 for the production of Pu-238 serves also to reduce the risk of its unauthorized use, because it is a well-fissionable isotope in the fast neutron spectrum (bare critical mass - 57 kg) with practically no spontaneous fission.
It is shown that the use of radiogenic lead/Pb-208 opens up the possibility of improving a number of neutronic parameters of a fast reactor, including safety parameters:
- To reduce significantly, by about 20%, the critical loading of plutonium;
- To increase considerably (about three orders of magnitude) the mean lifetime of prompt neutrons, which can significantly improve the nuclear reactor safety in the case of reactivity accidents;
- To increase the Doppler effect by about 1.5 times.

1. INTRODUCTION

The report examines the problem of large-scale production of plutonium-238 in a fast reactor, taking into account the improvement of its safety characteristics.

1.1. The problem of large-scale production of plutonium-238

Plutonium-238 is a unique and irreplaceable material for autonomous power supply of devices operating in remote regions of the Earth and in space. The uniqueness of plutonium-238 is determined by its properties:

- 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 γ-radiation).

The existing scale and, especially, the near future of space research [1] and the development of remote regions of the Earth, will obviously require the creation of numerous sources of long-term autonomous power supply based on the Pu-238 isotope.

At present, Pu-238 is being produced at research reactors in the Russian Federation and the United States. However, the scale of its production (0.5 ~ 1.0 kg / year) cannot satisfy the growing demand for it due to the noted trends in the active development of remote regions of the Earth and outer space. New methods are required for the large-scale production of high quality Pu-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 and americium-241. 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. Fig. 1 shows a schematic diagram of the technological process for the production of the isotope Pu-238 based on these starting materials.

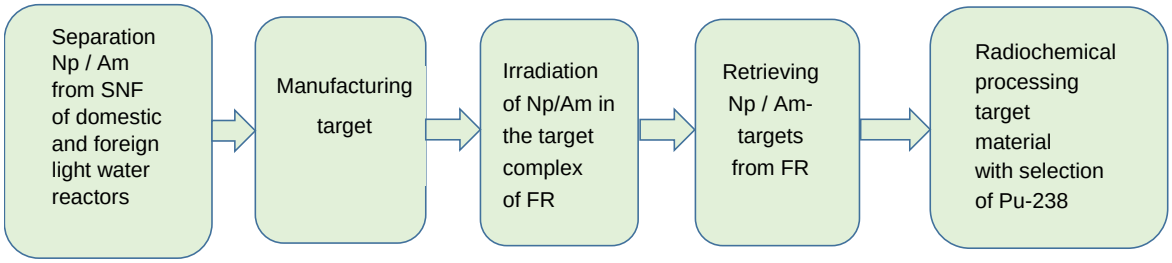

FIG. 1. Scheme of Pu-238 production in the target complex of a fast reactor.

It should be noted that the chains for obtaining Pu-238 from these starting isotopes in the neutron flux of a nuclear reactor are relatively short. However, they contain nuclear reactions that prevent the conversion of starting isotopes into plutonium-238. These are undesirable reactions of radiation capture of neutrons (n, γ), fission reactions (n, f), as well as alpha and electronic decay of americium isotopes. Thus, the problem of producing plutonium-238 from starting isotopes is being solved in very contradictory conditions, and the main thing here is to find the spectrum of irradiation neutrons (preferred spectrum) in order to minimize the contribution of unwanted channels in the chains of Pu-238 production. Naturally, the search for the preferred spectrum includes the characterization of each of the isotopes in the plutonium-238 production chains.

A method for forming the preferred neutron spectrum for the production of plutonium-238 is proposed in the report. The method consists in the use of moderators with a small step of deceleration (and, therefore, high atomic weight) and weak absorption of neutrons. An example of such moderator is radiogenic lead /lead-208. The use of these materials makes it possible to create closed spatially extended regions with a neutron spectrum preferable for the production of Pu-238 from starting materials (Np-237, Am-241). The report makes an impact assessment of radiogenic lead /lead-208 as a neutron moderator on the safety characteristics of a fast reactor.

1.2. Some features of radiogenic lead

Radiogenic lead is a product of radioactive decay of uranium and thorium isotopes. Resulting from several α and β decays, ^{232}Th transforms into a stable ^{208}Pb isotope, ^{238}U transforms into ^{206}Pb isotope, and ^{235}U transforms into ^{207}Pb isotope. Therefore, the isotopic composition of radiogenic lead extracted from thorium and thorium–uranium ores is determined by their composition and age. Hereinafter we will consider radiogenic lead with a high lead-208 content.

The lead-208 isotope is characterized by unique neutron-physical properties: an extremely small radiative neutron absorption cross section and location of inelastic scattering cross section threshold at a significantly higher neutron energy level than in other lead isotopes [2–4]. The unusual properties of radiogenic lead rich in lead-208 can manifest in a fast reactor with lead coolant as follows:

- The weak neutron absorption by lead-208 results in longer neutron migration lengths, making it possible for neutrons to penetrate deeply into the neutron reflector with low losses as they penetrate. Consequently, neutrons generated in the core and leaving the core for a physically thick reflector can stay there for quite a long time, then return to the core, and take part in a fission chain reaction, but with a time delay. This time delay due to the neutron staying in a weakly absorbing reflector can significantly increase the mean lifetime of prompt neutrons and thus slow down the development of the fission chain reaction in the case of a hypothetical reactivity accident;
- The microscopic cross section of neutron absorption for lead-208 in a wide range of energies (from thermal energy to few tens of keV) is significantly smaller than other lead isotopes (Fig. 2) and natural lead, whose replacement with lead-208 decreases the useless neutron absorption by the coolant and helps reduce plutonium critical loading;
- The high threshold energy of inelastic neutron scattering by lead-208 isotope hardens the neutron spectrum and, consequently, improves the plutonium breeding properties, and reduces the critical loading of plutonium.

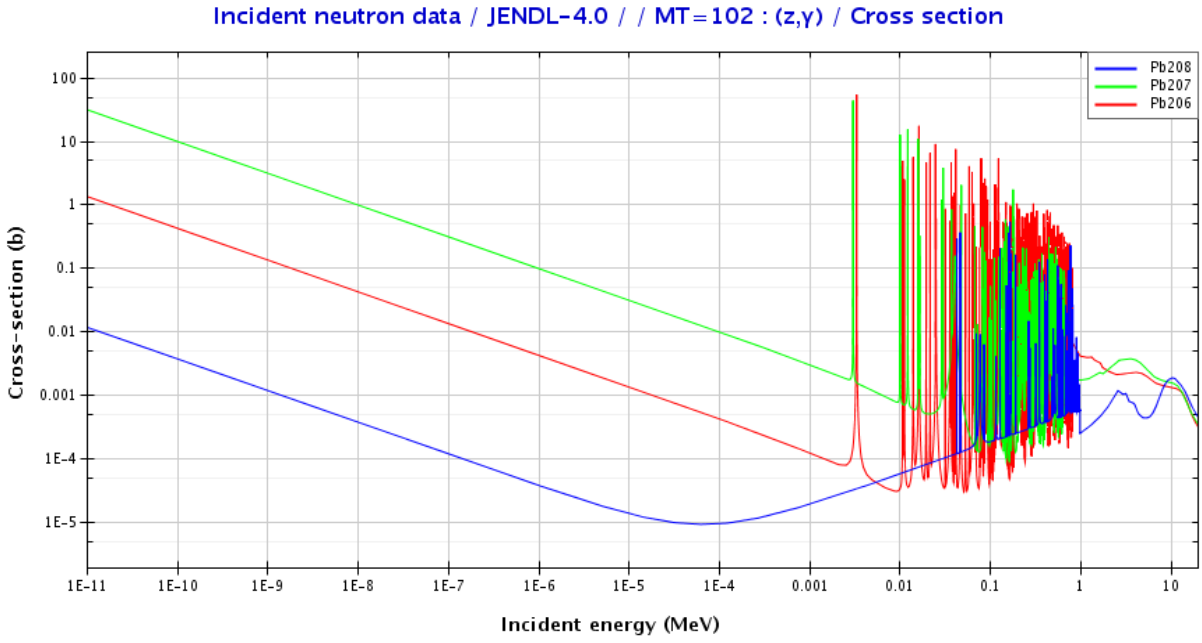

FIG. 2. Comparison of radiation capture microscopic cross sections of Pb-206, Pb-207 and Pb-208 isotopes.

2. PHYSICAL FOUNDATIONS OF LARGE-SCALE PRODUCTION OF PLUTONIUM-238 IN A FAST REACTOR

Below we will consider the process of plutonium-238 production in a fast reactor using the example of Np-237 irradiation. Fig. 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 [5]. Undesirable reactions that do not lead to the formation of plutonium-238 are marked with red crosses.

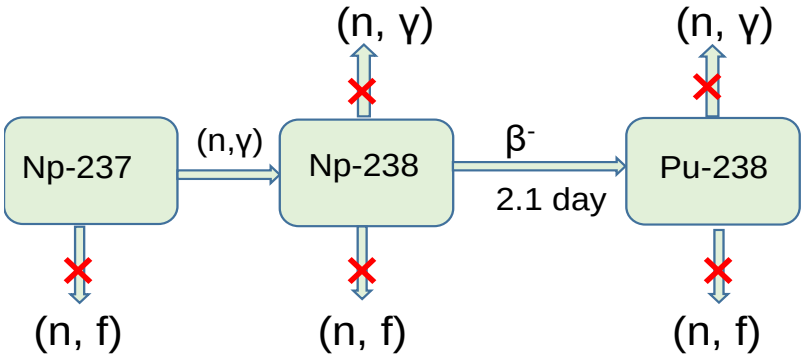

FIG. 3. Chain of isotopic transitions during irradiation of Np-237 in a fast reactor. The "X" indicates an unwanted channel in the Pu-238 production process.

It is seen that the chain of isotopic transitions is short. However, the task of producing plutonium-238 from Np-237 is being solved in highly contradictory conditions (many undesirable channels). And 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.

2.1. Analysis of the process of plutonium-238 production by irradiation of Np-237 in a fast reactor

2.1.1. Characteristics of Np-237 as a starting material for the production of Pu-238

Fig. 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 [6].

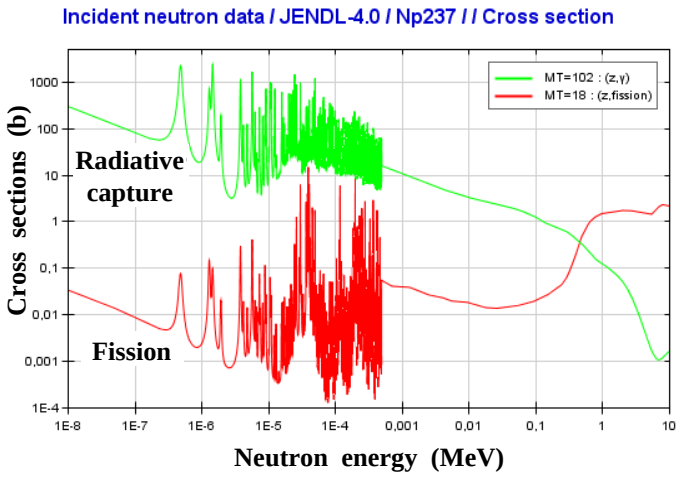

FIG. 4. Microsections of radiation capture and fission for neptunium-237.

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.

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 Fig. 6). Thus, the region of resonant absorption of Np-237 can be considered as the preferred spectrum for its irradiation.

2.1.2. Radiation capture and fission processes on neptunium-238 and plutonium-238

Fig. 5 shows the energy dependences of microsections for fission and radiative capture of neutrons for neptunium-238 [6]. 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.

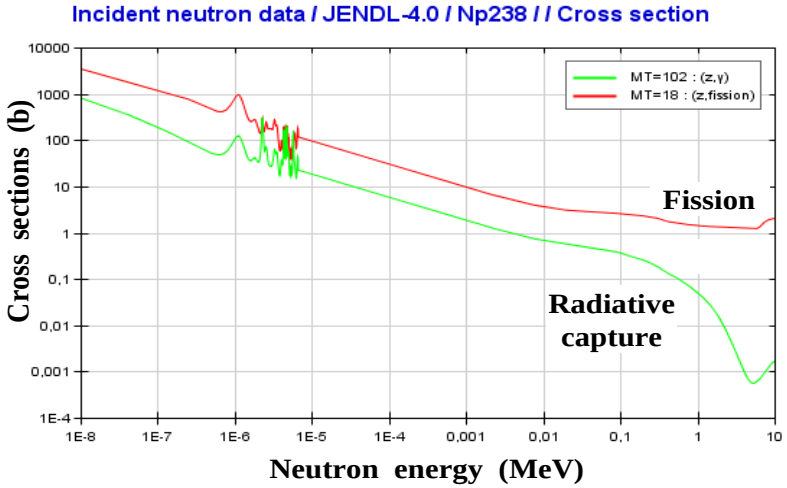

FIG. 5. Microsections of the radiation capture and fission of neptunium-238.

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:

$$\lambda = \frac{ln2}{T_{1/2}(^{238}Np)} \gg \left[\sigma_c(^{238}Np) + \sigma_f(^{238}Np)\right] \cdot \varphi \qquad (1)$$

where φ is neutron flux [n/(cm^2×s].

Condition (1) implies two simple conclusions:

1) The region of thermal and epithermal neutrons is extremely undesirable, since here the cross sections σ_c (^{238}Np) and σ_f (^{238}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 (~ 10^{15} n/(cm^2×s)), the electron decay channel is dominant. This conclusion is an important argument in choosing a fast reactor for Pu-238 production.

Fig. 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 [6]). It is seen that for Pu-238, both types of interactions are significant over the entire energy range.

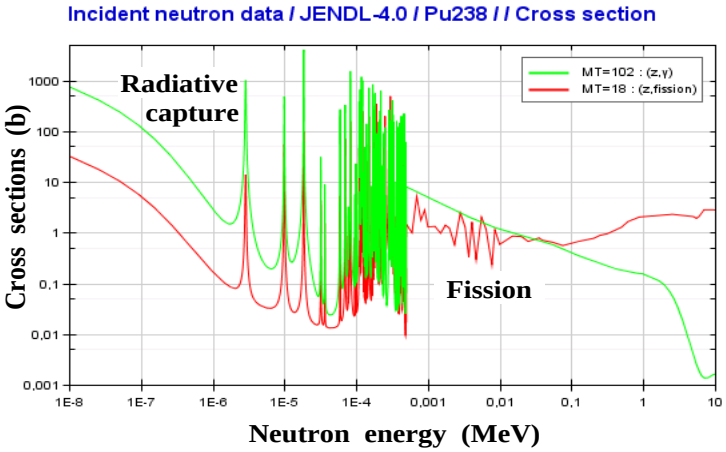

FIG. 6. Microsections of radiation capture and fission of plutonium-238.

We strive for the minimum consumption of the target nuclide:

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

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 the minimum (2) is approximately observed here and the cross sections σ_c and σ_f remain at the level of 10 barn and below.

2.2. Method for producing plutonium-238

It is proposed to place the target complex in the end part (reflector) of a fast reactor with a heavy liquid-metal coolant [7]. 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, the Np-237 target is surrounded by a moderator with a high atomic weight and low neutron absorption (radiogenic lead /Pb-208).

To form a preferred spectrum, it is especially effective to use radiogenic lead or Pb-208, which are characterized by an extremely low absorption of neutrons and, therefore, make it possible to increase the neutron flux density and soften the spectrum in the target material. Its application, thereby, accelerates the production of the target nuclide. As will be shown below, the use of Pb-208 offers other important advantages associated with increasing the safety of a fast reactor (including an increase in the Doppler effect and the average lifetime of prompt neutrons).

3. ONE-DIMENSIONAL MODEL OF A FAST REACTOR WITH LEAD COOLANT AND REFLECTOR

It is known [7, 8] that greatly flattened cores are typical of modern fast reactor projects. Therefore, in order to study the special aspects of neutron-physical characteristics, which are determined by the axial distribution of the neutron field, it is quite possible to use in the calculations the one-dimensional plane (axial) model of the reactor along its height (Fig. 7).

Neutron-physical calculations have been carried out to assess the logic of the above assumptions about the advantages of using lead-208 as a coolant and reflector. In these calculations, TIME26 software was used [9], which considers one-dimensional models of fast reactors in a 26-group diffusion approximation. The constant provision of TIME26 is based on the BNAB-78 evaluated nuclear data library that is processed by the ARAMACO-C1 auxiliary program to prepare blocked microscopic cross sections in each zone of the reactor [10]. The computer code ARAMACO-S1 prepares 26-group micro cross-sections with accounting for self-

shielding effect and Doppler-effect within the energy range of resolved and non-resolved resonances with application of self-shielding Bondarenko factors and the conception of diluted micro cross-sections.

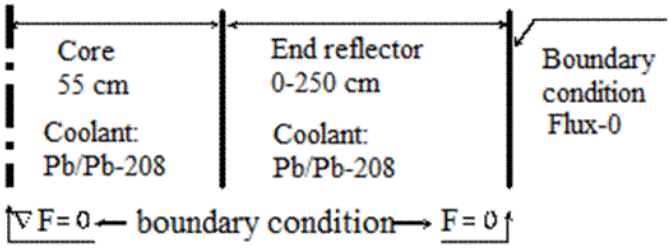

FIG. 7. Geometric model of a fast reactor with the neutron reflector characterized by the absolute absorption at the outer boundary.

4. NEUTRON SPECTRUM AND FLUX IN A LEAD REFLECTOR OF A FAST REACTOR

Three-group spectra and fluxes of neutrons in 250- cm thick end reflectors of natural lead (Fig. 8) and lead-208 (Fig. 9) are shown. The spectrum consists of fast neutrons (with energies above uranium-238 resonances), resonance neutrons (with energies within the resonance range of uranium-238 from 4.65 eV to 21.5 keV), and epithermal and thermal neutrons (with energies below the uranium-238 resonances).

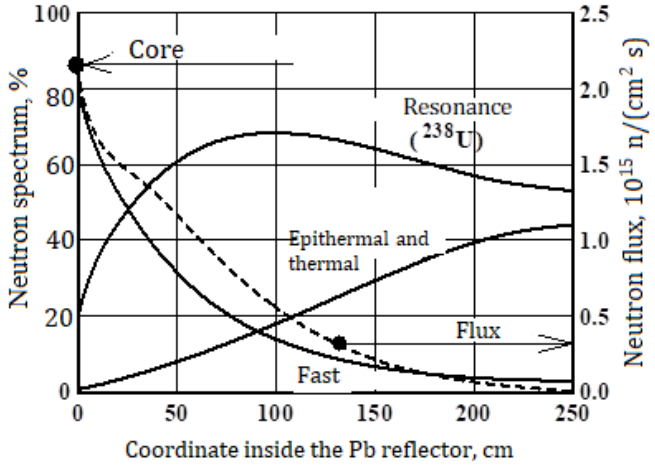

FIG. 8. Neutron spectrum and flux in the Pb reflector.

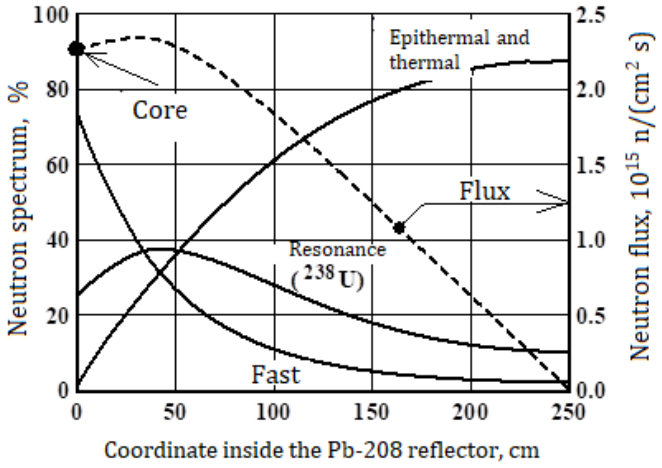

FIG. 9. Neutron spectrum and flux in the ^{208}Pb reflector.

Naturally, the general behavior of the dependencies for both types of lead is the same: as the penetration into the reflector increases (with the growth of coordinate inside the reflector), the neutrons slow down and the neutron flux weakens. The share of fast neutrons continuously decreases, the share of resonance neutrons first grows and then decreases, and the share of epithermal and thermal neutrons continuously grows. This is explained by the fact that fast neutrons are the source of resonance neutrons, which, in turn, are the source of epithermal and thermal neutrons. At large coordinates of the reflector (close to 250 cm), the neutron spectra almost reach plateau and change weakly.

However, there are also significant differences between these dependences in natural lead and lead-208. For a lead-208 reflector, a significantly higher neutron flux and a slight shift of the neutron spectrum towards epithermal energies are observed (since lead-208 softens the neutron spectrum). These circumstances serve to intensify the production of plutonium-238 in the region of resonant absorption of neutrons on neptunium-237. Moreover, if the irradiation of starting material is carried out at a sufficiently large distance from the core, then it does not significantly disturb the core itself.

5. LEAD REFLECTOR INFLUENCE ON FAST REACTOR PARAMETERS

Calculations using the TIME26 program were carried out in the following order.

1. The share of the plutonium fraction in mixed uranium–plutonium nitride fuel corresponding to the reactor's critical state was found at the maximum reflector thickness of 250 cm. Expectedly, the critical share of plutonium was noticeably higher (~11.3%) in the natural lead option than in the lead-208 option (~9.1%).

2. Then, the reflector thickness was reduced and the following neutron-physical parameters of the reactor were calculated: the effective multiplication factor (K_{eff}) and the mean lifetime of prompt neutrons (l_{prm}). The results of the calculations are shown in Fig. 10.

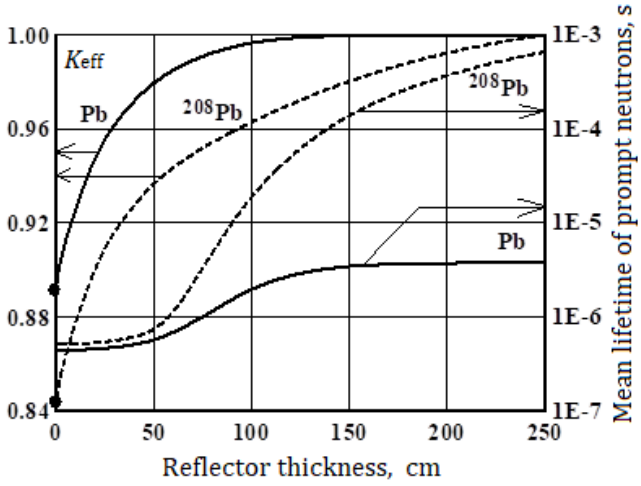

FIG. 10. Influence of the reflector thickness on the effective multiplication factor and the mean lifetime of prompt neutrons.

It is shown that the advantage of a neutron reflector (change of K_{eff} from the bare core to the maximum thickness of the reflector) is much greater when lead-208 is used instead of natural lead. Hence, with a 250-cm thickness of the reflector, this advantage is $\Delta K_{eff} = 0.047$, i.e., about $13 \cdot \beta$ at the fraction of delayed neutrons $\beta = 0.37\%$. This is mainly explained by the better reflecting properties of lead- 208 due to its low neutron absorption as compared to natural lead.

It is also shown, that the coolant and reflector made of lead-208 can provide three orders of magnitude longer lifetimes of prompt neutrons. When natural lead is used as the coolant and reflector, the average lifetime of prompt neutrons remains around a few microseconds even at the maximum thickness of the reflector. Whereas using lead-208 for coolant and reflector we can shift the lifetime range of prompt neutrons to milliseconds.

This is due to the radically low neutron absorption by lead-208 compared to natural lead, which allows leakage neutrons escaping from the core to penetrate deeply into the reflector, slow down there, and yet have a

significant chance to return to the core and influence the fission chain reaction. In natural lead, this is impossible, because the natural lead absorbs moderated neutrons, so they cannot return from the great depth of the reflector. While these are exactly the neutrons that can increase the mean lifetime of prompt neutrons.

The computations showed that absolute value of negative Doppler constant K_D for the reactivity Doppler-effect caused by the reactor heating-up was larger by a factor of 1.5 in the case of ^{208}Pb-coolant / reflector than that in the case of natural lead (- 0.0108 versus – 0.0072). The reason consists in a weak neutron absorption by ^{208}Pb. That is why ^{208}Pb-reflector is able to provide intense return of slow neutrons to the reactor core where they are absorbed by expanded resonances of ^{238}U. At the same time, natural lead as a neutron reflector provides intense absorption of slow neutrons without their coming back to the reactor core.

6. CONCLUSIONS

1. It has been shown that for a large-scale production in a fast reactor Pu-238 from starting material Np-237, the following favorable conditions are required:
   - High neutron flux density for fast production of the target nuclide;
   - Resonance spectrum of neutrons (1 ~ 600 eV) to intensify the radiative capture on Np-237 and avoid its useless fission, as well as to reduce the radiative capture / fission on Pu-238;
2. The use of radiogenic lead as a moderator makes it possible to form a spectrum in the target with a maximum concentration of neutrons in the resonance region (1 ~ 600 eV);
3. Due to the use of radiogenic lead, the resonance region in the target complex has an extended character, which is important for the large-scale production of Pu-238;
4. It is shown that the transition to radiogenic lead as not only a coolant, but also a reflector opens up the possibility of improving a number of neutronic parameters of a fast reactor, including safety parameters:
   - To achieve in the reflector a higher flux of resonance neutrons, which is retained at a considerable distance from the core. The latter opens up wider possibilities for the placement of the target complex for the production of Pu-238;
   - To reduce significantly, by about 20%, the critical loading of plutonium;
   - To increase considerably (about three orders of magnitude) the mean lifetime of prompt neutrons, which can significantly improve the nuclear reactor safety in the case of reactivity accidents;
   - To increase the Doppler effect by about 1.5 times.

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