

**One-group fission cross-sections for plutonium and minor actinides
inserted in calculated neutron spectra of fast reactor cooled
with ^{208}Pb or lead-bismuth eutectic**

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Abstract

This paper presents the one-group fission cross-sections of Pu and MA in LFR and ADS spectra with the goal of increasing these values by choosing a coolant which hardens neutron spectra. It is shown that the replacement of coolant from Pb-Bi or Pb-nat with ^{208}Pb in the fast reactor RBEC-M, designed in the Russian Federation, and small ADS installation, proposed by authors, leads to hardening the core and blankets mean neutron energy approximately by 6% and increasing one-group fission cross-sections of MA up to 8-11%.

Introduction

Since 1999 authors have proposed a new coolant for nuclear power installations with intermediate and fast neutron spectra [1]. This coolant is based on lead-enriched with the lead isotope, lead-208, which is characterised by very low neutron capturing and slow neutron moderation. These features of lead-208 allow having some gain in the quantity of plutonium [2] to reach criticality of fast reactor (FR) or in hardening the neutron spectra of the core and its lateral and topical blankets. Natural lead consisting of 52.3% of ^{208}Pb , 22.6% of ^{207}Pb , 23.6% of ^{206}Pb , 1.5% of ^{204}Pb and ^{208}Pb -enriched up to 100% is assumed. This paper provides the one-group fission cross-sections for plutonium and minor actinides inserted in the calculated neutron spectra of the FR RBEC-M [3] and small power ADS [4].

Methods of spectra calculations and results

The 900 MW_{thermal} reactor core of the RBEC-M lead fast reactor (LFR) design [3] is shown in Figure 1. Mixed uranium-plutonium nitride fuel ($\text{U}_{0.863}+\text{Pu}_{0.137}\text{N}$) is used, which is composed of power grade plutonium recovered from typical light water reactor spent fuel and depleted uranium with 0.1 wt. % of ^{235}U . The core zones are surrounded by lateral (radial) and topical blankets; the structural material is steel; and the standard coolant material is lead-bismuth eutectic. The geometry of the calculation model of the RBEC-M reactor is shown in Figures 1 and 2. The model includes 12 physical zones, differing from each other by volume fractions and temperatures of materials.

Figure 1: Actual core configuration of the 900 MW(th) RBEC-M reactor

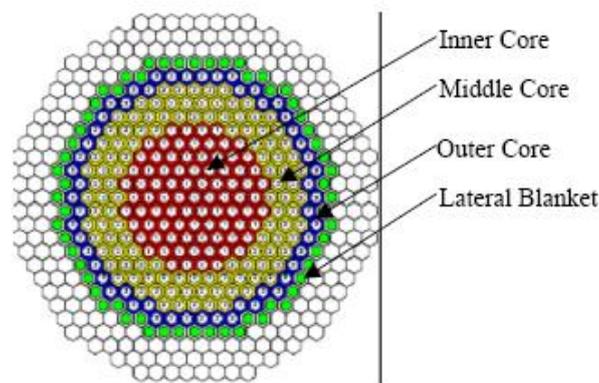
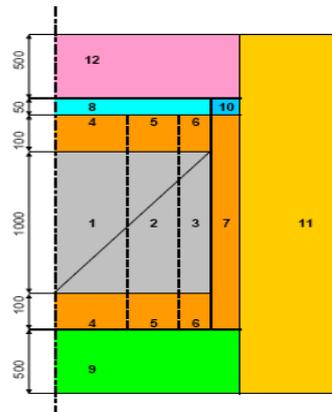


Figure 2: Geometry of calculation model of the RBEC-M reactor

In LFR RBEC-M in replacement its standard lead-bismuth coolant with lead-208 the neutron multiplication factor K_{eff} increases from its standard value, $K_{eff}=1.00957$, to the value $K_{eff}=1.0246$, i.e. on 1.5%. It was calculated that, to leave this coefficient at the standard level, i.e. to ensure the criticality, $K_{eff}=1.00957$, the quantity of power grade plutonium in uranium-plutonium nitride fuel must be decreased from 3 595 kg to 3 380 kg i.e. Pu enrichment in the fuel will be decreased from its initial value of 13.7% down to 13.0%. With respect to the coolant temperature, it can be assumed unchanged in coolant replacement due to its relatively high standard values for Pb-Bi coolant, $T_{inlet}/T_{outlet}=380/520^{\circ}\text{C}$.

For core and blankets cooled with $^{208}\text{Pb}/^{nat}\text{Pb}$ -Bi the neutron fluxes were calculated, using MCNP5 code [5] and nuclear data library ENDF/B-VII.0 generated for different temperatures by NJOY code.

Figures 3 and 4 present the calculated results of neutron fluxes for the cell numbers 1, 2, 3 and 7. Neutron fluxes are presented in the ABBN- energy-group structure system having 28 neutron energy groups [6].

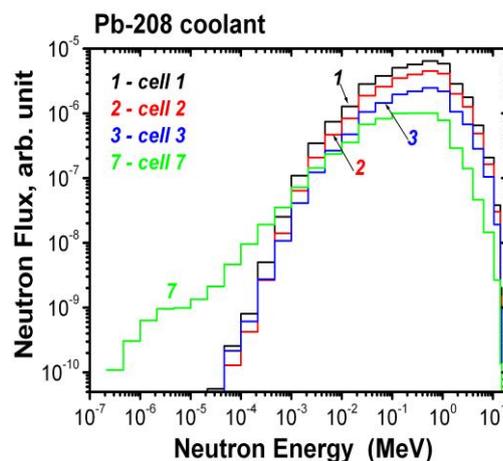
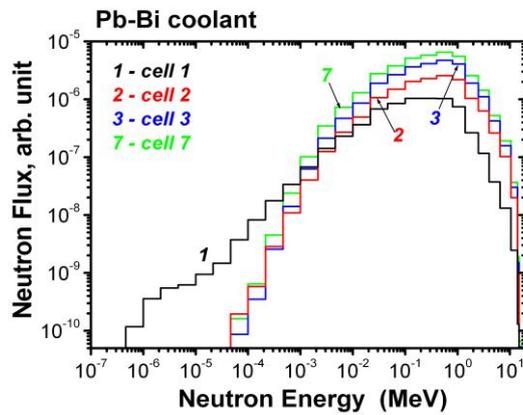
Figure 3: Neutron fluxes for the cell numbers 1, 2, 3 and 7 cooled with ^{208}Pb 

Figure 4: Neutron fluxes for the cell numbers 1, 2, 3 and 7 cooled with $^{nat}\text{Pb-Bi}$ 

In Figures 5-8 the calculated results of the neutron flux ratio for a case of the LFR core cooling with molten ^{208}Pb and $^{nat}\text{Pb-Bi}$ are presented for the cell numbers 1, 2, 3 and 7, respectively.

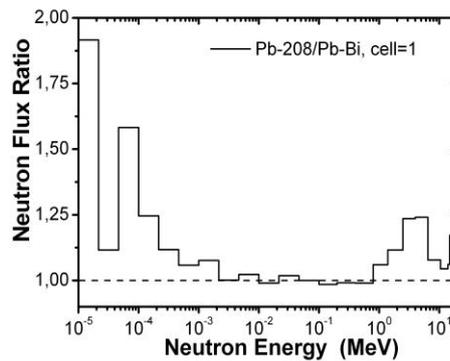
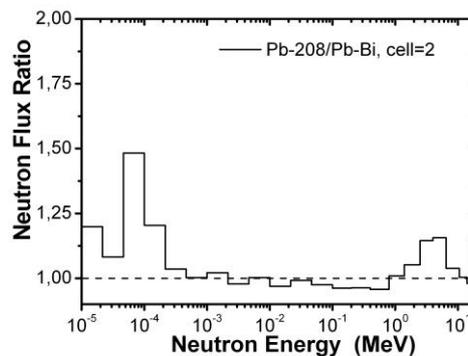
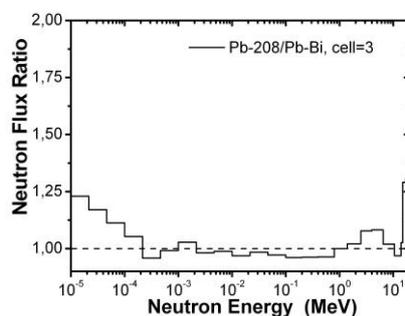
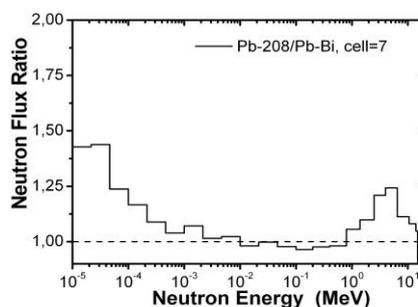
Figure 5: Neutron flux ratio for cell = 1 cooled with ^{208}Pb and $^{nat}\text{Pb-Bi}$ **Figure 6: Neutron flux ratio for cell = 2 cooled with ^{208}Pb and $^{nat}\text{Pb-Bi}$** 

Figure 7: Neutron flux ratio for cell = 3-cooled with ^{208}Pb and natPb-Bi **Figure 8: Neutron flux ratio for cell=7 cooled with ^{208}Pb and natPb-Bi** 

On the base of calculated neutron spectra for several zones of the LFR core the mean energies of neutrons ($\langle E_n \rangle$) for corresponding neutron spectra of the core zones were performed. Table 1 shows neutron energies calculated for the RBEC-M-cooled with ^{208}Pb and natPb-Bi .

Table 1: Neutron energies averaged over the neutron spectrum of FR RBEC-M core and blankets cooled with ^{208}Pb (bold) or natPb-Bi eutectic

	Cell 4	Cell 5	Cell 4 6	Cell 1	Cell 2	Cell 3	Cell 7
	Topical blanket	Topical blanket	Topical blanket	Inner core	Middle core	Outer core	Lateral blanket
$\langle E_n \rangle$, MeV	0.3010/ /0.2746	0.3051/ /0.2850	0.2869/ /0.2757	0.4246/ /0.3992	0.4408/ /0.4209	0.4431/ /0.4307	0.2662/ /0.2509
$\Delta \langle E_n \rangle$, $E_{n,208} - E_{n,Pb-Bi} / E_{n,Pb-Bi}$, %	9.6140	7.0526	4.0624	6.3627	4.7280	2.8790	6.0980

As a result of coolant replacement, the core neutron energy increases by 3-6% and the blanket neutron energy increases up to 4-10%. Table 2 presents one-group fission cross sections $\langle\sigma(n, f)\rangle = \frac{\sum \sigma_{fiss} \cdot \varphi_n}{\sum \varphi_n}$ of fuel nuclides: ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu and ^{241}Am averaged over the neutron spectrum of FR RBEC-M inner core cooled with ^{208}Pb or $^{\text{nat}}\text{Pb-Bi}$.

As follows from Table 1, the replacement of coolant from Pb-Bi with ^{208}Pb in RBEC-M leads to increasing the inner core mean neutron energy from its standard value 0.3992 MeV to 0.4246 MeV, i.e. on 6.4%. As regards Pu isotopes, their one-group fission cross-sections become slightly changed. More dramatically ^{241}Am one-group fission cross-section is changed, by 9.6%, as a result of neutron spectrum hardening. It should be noted that 5-6 % of ^{241}Am is usually presented in the power grade plutonium obtained from spent fuel of PWR or VVER. Such mix of power grade plutonium and 5-6 % of ^{241}Am is usually proposed as a fuel for FR's core.

Table 2: Neutron energy and one-group fission cross-sections of fuel nuclides: ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu and ^{241}Am averaged over the neutron spectrum of FR RBEC-M inner core cooled with ^{208}Pb or $^{\text{nat}}\text{Pb-Bi}$

Reactor and its coolant	Neutron energy averaged over inner core spectrum, MeV	Pu-238 one-group fission cross section, barns	Pu-239 one-group fission cross section, barns	Pu-240 one-group fission cross section, barns	Pu-241 one-group fission cross section, barns	Pu-242 one-group fission cross section, barns	Am-241 one-group fission cross section, barns
LFR cooled with ^{208}Pb	0.4246	1.1223	1.7091	0.4101	2.3079	1.6371	0.2882
LFR cooled with Pb-Bi	0.3992	1.0997	1.7030	0.3871	2.3122	1.6387	0.2629
Difference between data, %	+6.3627	+2.0551	+0.3582	+5.9416	-0.1860	-0.0010	+9.6234

Table 3 shows the neutron energy and one-group fission cross-sections of long-lived minor actinides in the Cell 7 – lateral blanket.

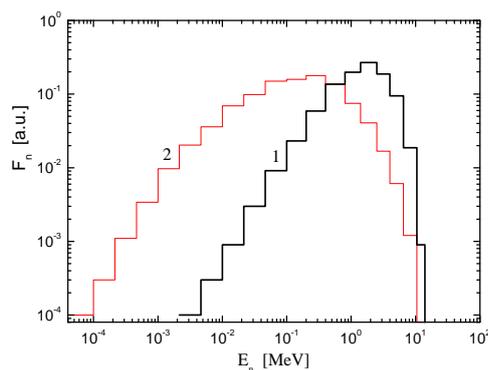
Table 3: Neutron energy and one-group fission cross-sections of long-lived minor actinides averaged over the neutron spectrum of FR RBEC-M lateral blanket cooled with lead-208 or lead-bismuth eutectic

	Np-237	Am-241	Am-243	Cm-242	Cm-244	Cm-246	Cm-248
LFR cooled with ^{208}Pb	0.2307	0.1671	0.1167	0.0847	0.3158	0.1942	0.2085
LFR cooled with Pb-Bi	0.2140	0.1521	0.1050	0.0752	0.2956	0.1861	0.1871
Difference between data, %	7.8505	9.8619	11.1429	12.6330	6.8336	4.3525	11.4378

As a result of switching coolants from Pb-Bi to ^{208}Pb lateral blanket mean neutron energy increases from 0.2509 to 0.2662 MeV, i.e. by 6.1%. As a result of the neutron spectrum hardening, the one-group fission cross-sections of ^{237}Np , ^{241}Am , ^{243}Am , ^{242}Cm , ^{244}Cm , ^{246}Cm , ^{248}Cm increase up to 8-11%. As is known these minor actinides need higher neutron energy, more than 0.1-0.2 MeV, to be fissile. The result of increasing MA one-group cross-sections is very important as it allows reducing the time of minor actinides burning in the FR lateral blanket for their exclusion from nuclear wastes.

In nuclear power installations with fast neutrons, ADSs and FRs, the mean energy of core neutrons does not exceed 0.5 MeV, while the mean energy of fission neutrons emitted by ^{235}U , for example, is equal to 1.98 MeV. Figure 9 presents the typical spectrum of neutrons in the core of lead fast reactor and spectrum of fission neutrons emitted by ^{235}U .

Figure 9: Neutron spectrum in the fuel zone of the 700 MW_{thermal} LFR (2) and spectrum of ^{235}U fission neutrons (1) in the ABBN-93 neutron-energy group system

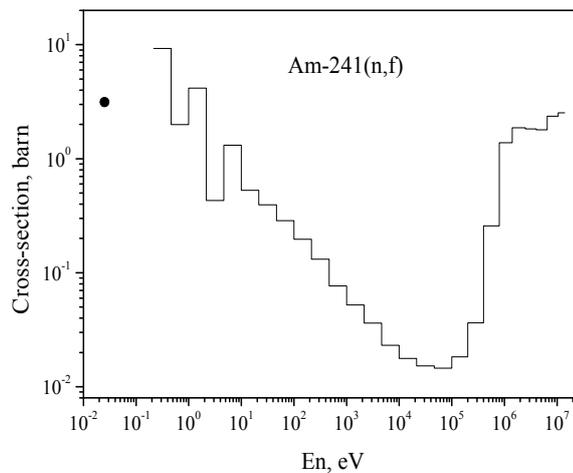


It can be seen that LFR neutron spectrum is strongly moderated as compared with the spectrum of fission neutrons. Neutron moderation is due to interaction of neutrons with fuel, structural materials and coolant.

Meanwhile hard spectrum of neutrons in ADS and FR core is preferable for the incineration of minor actinides. Incineration of long-lived radiotoxic MA – neptunium,

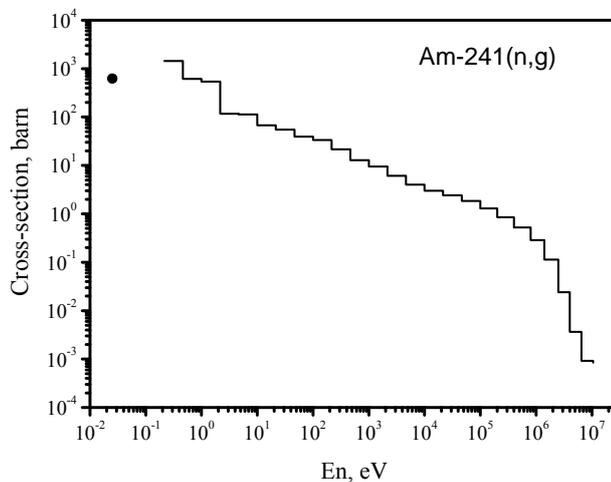
americium and curium – is one of the key problems of the nuclear power engineering. The world fleet of light water reactors (LWR) produces about 3.2 tonnes of MA per year as wastes. It is expected to incinerate MA in future ADSs which will be able to consume MA in quantities of 40% of fuel heavy atoms (h. a.). But ADS installation creation is a very expensive way and it takes a long time. An opportunity exists to incinerate MA in FR core but from reactor safety point of view the FR core can be loaded with MA in quantities not more than 2.5% of h. a. As an alternative it is possible to load a radial blanket with the fuel having MA of 10% of h. a. But, to avoid ^{242}Cm accumulation during americium transmutation it is desirable to incinerate MA via their fission. Figure 10 shows microscopic fission cross-section for one of MA, namely ^{241}Am .

Figure 10: Microscopic fission cross-sections for ^{241}Am taken from ENDF/B-VII.0 library



It can be seen that for fast neutron region there is a threshold equal to 0.1 MeV above which ^{241}Am fission cross-sections are growing up to 2-3 barns at $E_n=1-10$ MeV. In the range of thermal and intermediate neutron energies, $E_n < 10$ keV, ^{241}Am fission cross-sections are also large enough but at these energies neutron capture cross-sections are too large as shown in Figure 11.

Figure 11: Microscopic neutron radiation capture cross sections for ^{241}Am taken from ENDF/B-VII.0 library



For the estimation of ^{241}Am incineration capability in ADSs, the neutron spectra in 80 MW ADS subcritical blanket were calculated, using MCNP code and nuclear data library ENDF/B-VII.0. Calculations were performed in the ABBN-energy group structure system with 28 neutron energy groups.

As initial data the following ones were taken [4]:

- annular core with a source of neutrons – a target on its axis;
- calculated with the Monte-Carlo technique a spectrum of spallation neutrons in the target consisting of the modified lead-bismuth eutectic – ^{208}Pb (80%)-Bi (20%);
- proton beam energy – $E_p=600$ MeV;
- the effective multiplication factor for the subcritical core cooled by Pb-208 – $K_{ef}=0.970$;
- thermal capacity delivered in the subcritical core in the rated regime – $N=80$ MW;
- Core coolant – lead-208, ^{208}Pb (100%), or natural lead, Pb-nat (100%).

For the reduction of the core dimensions and minimisation the quantity of the coolant, a mix of mononitrides of the depleted uranium, ^{238}U , and plutonium from the PWR spent nuclear fuel and MA as the ADS core fuel was considered. Pu and MA contents in the uranium-plutonium mix were accepted equal to 15%. Table 4 indicates the calculated basic technical parameters of the 80 MW_{thermal} ^{208}Pb /Pb-nat cooled ADS core, satisfying the initial data.

Table 4: Parameters of the 80 MW_{thermal} Pb-208/Pb-nat cooled ADS core

Parameters	Values
Subcritical core thermal power	80 MW
Annular core outer diameter	123.7 cm
Annular core inner diameter	56.0 cm
Annular core height	110.0 cm
Core fuel	(U+Pu+MA) ¹⁵ N
Total fuel inventory	5410 kg
Total heavy metal inventory	5 090 kg
Total Pu and Minor Actinides inventory	810 kg
Mean pin linear power	188 W/cm
Mean volume power density	118 W/cm ³
Effective multiplication factor for the core cooled by Pb-208/Pb-nat	$K_{\text{eff}}=0.970$ for Pb-208 $K_{\text{eff}}=0.953$ for Pb-nat
Proton beam energy	600 MeV
Proton beam current required to deliver 80 MW _{thermal} core power	$I_p=2.8$ mA for Pb-208 $I_p=4.3$ mA for Pb-nat
Proton beam power required to deliver 80 MW _{thermal} core power	$N_p=1.68$ MW for Pb-208 $N_p=2.58$ MW for Pb-nat

On the basis of the calculated neutron spectra for 9 subzones of the ADS blanket the mean energies of neutrons were calculated, as shown in Table 5.

Table 5: Neutron energies averaged over the neutron spectrum of subzones 1-9 of ADS blanket cooled with ^{208}Pb (bold) or $^{\text{nat}}\text{Pb}$

Target source of neutrons	Subzone 1 0.4252/0.4404	Subzone 2 0.4438/0.3408	Subzone 3 0.3346/0.2362
	Subzone 4 0.4820/0.5576	Subzone 5 0.5377/0.4929	Subzone 6 0.3723/0.3754
	Subzone 7 0.3362/0.2533	Subzone 8 0.3732/0.3812	Subzone 9 0.3182/0.3287
0	28	39	51
Blanket radius, cm			

Note: Energies in MeV are given.

The mean neutron energies averaged over subzones 1-9 of the blanket are equal to $\langle E_n \rangle = 0.4025$ MeV for coolant from ^{208}Pb and $\langle E_n \rangle = 0.3785$ MeV for coolant from Pb-nat. Thus, replacement of coolant leads to neutron energy hardening by 6.34%.

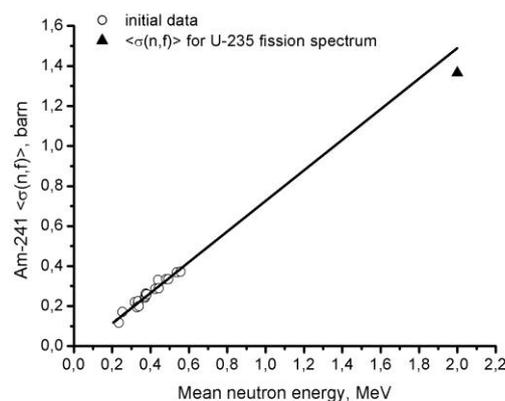
Table 6 presents one-group ^{241}Am fission cross-sections for subzones 1-9 of the 80 MW subcritical blanket cooled with ^{208}Pb or Pb-nat.

Table 6: One-group ^{241}Am fission cross-sections in barns for subzones 1-9 of the 80 MW subcritical blanket cooled with ^{208}Pb (bold) and Pb-nat

Target source of neutrons	Subzone 1 0.2854/0.3310	Subzone 2 0.2886/0.1984	Subzone 3 0.2197/0.1168
	Subzone 4 0.3335/0.3725	Subzone 5 0.3681/0.3334	Subzone 6 0.2429/0.2630
	Subzone 7 0.2245/0.1711	Subzone 8 0.2483/0.2594	Subzone 9 0.2194/0.1932

As follows from these calculations, the replacement of coolant from Pb-nat with ^{208}Pb in ADS blanket leads to increasing the mean neutron energy averaged over 1-9 subzones from its value 0.3785 to 0.4026 MeV, i.e. by 6.4%. In this case the one-group ^{241}Am fission cross-section averaged over 1-9 subzones increases from 0.2488 to 0.2700 barns, i.e. by 8.5%. Figure 12 presents the calculated dependence of one-group fission cross-sections for ^{241}Am upon mean neutron energy. It can be seen that these cross-sections grow as the neutron energy increases. It should be noted that there is a relatively high reserve to reach the maximum available mean neutron energy equal to 1.98 MeV, which corresponds to mean neutron energy of neutrons emitted by ^{235}U .

Figure 12: The calculated dependence of one-group fission cross-sections for ^{241}Am upon mean neutron energy



Conclusion

This paper presents the proposal of using lead enriched with the stable lead isotope, lead-208 in FRs and ADSs as coolant instead of natural lead or lead-bismuth eutectic.

It seems that unique neutron features of lead-208 make it one of the best among the molten metal coolants now assumed for FRs and ADSs: sodium, lead-bismuth, natural lead and others.

The main advantage of lead-208 is its low neutron absorption ability: for neutron energies $E_n < 50$ keV, the difference in the microscopic cross-section of neutron capture values reaches 3-5 orders of magnitude as compared with natural lead or lead-bismuth. Averaged over neutron spectra of the LFR the one-group cross sections of neutron capture for a coolant from lead-208 are by 5-6 times smaller than those for the coolant consisting of natural lead [7].

The second advantage of using lead-208 consists in achieving core neutron spectra hardening by 6% due to low energy losses in it. Low neutron absorbing and slow moderating features of lead-208 permit reaching the gain in the multiplication factor K_{eff} on 1.5% for critical or subcritical core fueled with U-Pu mix. In this case, to have the neutron multiplication factor $K_{\text{eff}} = 1.01$ for the LFR or $K_{\text{eff}} = 0.97$ for the ADS, both cooled with lead-208, the enrichment of power grade Pu in the U-Pu fuel can be reduced approximately by 0.7-0.8%.

This paper shows that the replacement of coolant from Pb-Bi to ^{208}Pb in fast reactor RBEC-M, designed in the Russian Federation, leads to increasing the core mean neutron energy from its standard value 0.3992 MeV to 0.4246 MeV, i.e. by 6.4%. As regards Pu isotopes, their one-group fission cross-sections become slightly changed. More

dramatically, by 9.6%, ^{241}Am one-group fission cross-section is changed as a result of neutron spectrum hardening.

A similar situation occurs in the lateral blanket containing small quantities of minor actinides. It is shown that, as a result of lateral blanket mean neutron energy increasing from 0.2509 to 0.2662 MeV, the one-group fission cross-sections of ^{237}Np , ^{241}Am and ^{243}Am increases up to 8-11%. This result is very important as it allows reducing the time of minor actinides burning in FRs.

The replacement of coolant from Pb-nat with ^{208}Pb in small sized ADS blanket proposed by authors [4] leads to increasing the mean neutron energy averaged over ADS subzones from its value 0.3785 to 0.4026 MeV, i.e. by 6.4%. In this case, the one-group ^{241}Am fission cross-section averaged over ADS subzones increases from 0.2488 to 0.2700 barns, i.e. by 8.5%.

The possibility of using lead-208 as coolant in commercial fast critical or subcritical reactors requires a special consideration, but the relatively high content of this isotope in natural lead, 52.3%, and perspectives of using the centrifugal or high performance photochemical technique of lead isotope separation allow expecting to obtain in future such a material in large quantities and at an economically acceptable price [8][9].

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