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Benefits in Using Lead-208 Coolant for Fast Reactors and Accelerator Driven Systems

Georgy L. Khorasanov and Anatoly I. Blokhin

Additional information is available at the end of the chapter

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1. Introduction

The chapter is dedicated to the analysis of benefits in using lead enriched with the stable lead isotope, Pb-208, instead of natural mix of stable lead isotopes, Pb-204, Pb-206, Pb-207 and Pb-208, as heavy liquid metal coolant for core and top-lateral blankets of critical reactors on fast neutrons (FR) and for subcritical blanket of accelerator driven system (ADS). Pb-208 has very low cross sections, less than 0.3 mbarn, of neutron radiation capture below 50 keV of neutron energy and high threshold, around 2.6 MeV, for inelastic interaction of neutrons with Pb-208 nuclei. These features of Pb-208 as coolant allow reaching economy of neutrons in FR and ADS core and blanket, hardening neutron spectra, favorable conditions for performance of fuel breeding and minor actinides incineration.

2. Small neutron absorption in FR and ADS coolant from lead-208

In the range of neutron energies below 20 MeV microscopic cross sections of radiation neutron capture by the lead isotope Pb-208 are smaller than the cross sections of radiation neutron capture by natural lead, Pb-nat, which is proposed as heavy liquid metal coolants for future lead fast reactors (LFRs). This difference is especially large, by 3-4 orders of magnitude, for intermediate and low energy neutrons, $E_{\rm n}$ <50 keV. Share of neutrons with energies less than 50 keV is usually about 20-25% of all neutrons in FR or ADS cores and it increases in lateral and topical blankets of FRs. In Fig. 1 cross sections of radiation neutron capture by the lead isotopes Pb-204, Pb-206, Pb-207, Pb-208 and the natural mix of lead isotopes Pb-nat in the ABBN-93 system [1] of 28 neutron energy groups are resulted. The cross sections are received on the basis of files of the evaluated nuclear data for the ENDF/B-VII.0 version library.



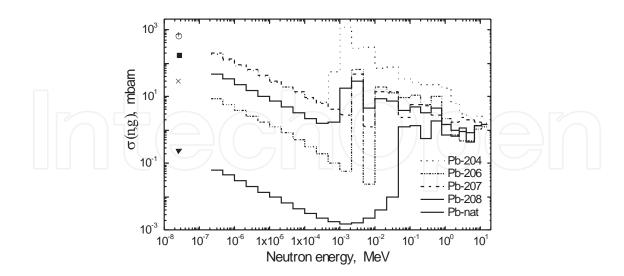


Figure 1. Microscopic cross sections of radiation neutron capture, $\sigma(n,g)$, by stable lead isotopes and natural mix of lead isotopes taken from the ENDF/B-VII.0 library. Cross sections are represented in the ABBN-93 system of 28 neutron energy groups.

It is visible, that practically for all of the 28 neutron energy groups of the ABBN-93 system, the cross sections of radiation neutron capture by the lead isotope Pb-208 are less than the cross sections of radiation neutron capture by the mix of lead isotopes Pb-nat, and this difference is especially strong, of 3-4 orders of magnitudes, for intermediate and low energy neutrons, E_n <50 keV.

In this chapter section benefits of the use of molten Pb-208 instead of Pb-nat as coolant are demonstrated on the model of the ADS blanket offered for transmutation minor actinides (MA) in a number of nowadays developed scenarios of destruction nuclear plant waste products. Calculations of neutron and physical characteristics of the ADS subcritical core have been performed by means Monte Carlo technique with the MCNP/4C code [2] and NJOY program specially developed with the help of the library of cross sections on the basis of ENDF/B-VII.0 evaluated nuclear data files. As initial data the following ones were taken [3]:

- 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 consisted of the modified lead-bismuth eutectic Pb-208 (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, Pb-208 (100%), or natural lead, Pb-nat (100%).

For reduction the core dimensions and minimization the quantity of the coolant, a mix of mononitrides of the depleted uranium, U-238, 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%.

The calculated basic technical parameters of the $80 \text{ MW}_{\text{thermal}}$ Pb-208/Pb-nat cooled ADS core, satisfying the initial data, are resulted in Table 1.

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	5090 kg	
Total Pu and Minor Actinides inventory	810 kg	
Mean pin linear power	188 W/cm	
Mean volume power density	118 W/cm³	
Effective multiplication factor K _{ef} =0.970 for Pb-208		
for the core cooled by Pb-208/Pb-nat	K_{ef} =0.953 for Pb-nat	
Proton beam energy	600 MeV	
Proton beam current required to deliver 80 MW _{thermal} core	I _p =2.8 mA for Pb-208	
power I _p =4.3 mAfor Pb-nat		
Proton beam power required to deliver 80 MW _{thermal} core	N _p =1.68 MW for Pb-208	
power	$N_p=2.58$ MW for Pb-nat	

Table 1. Parameters of the 80 MW_{thermal}Pb-208/Pb-nat cooled ADS core.

On the basis of the microscopic cross sections, $\sigma(n, g)$, in 28 group approximation and neutron spectra calculated by means the MCNP/4C code for subcritical core cooled with molten Pb-208 or Pb-nat the one-group cross sections, $\langle \sigma(n, g) \rangle = \sum \sigma_n \phi_n / \sum \phi_n / \phi_n /$

	Subzone 1 0.86 /5.08	Subzone 2 0.85 /5.22	Subzone 3 0.74 /5.86
Target-	Subzone 4	Subzone 5	Subzone 6
source of neutrons	0.92 /4.50	0.90 /4.83	0.83 /5.50
	Subzone 7	Subzone 8	Subzone 9
	0.82 /5.62	0.83 /4.74	0.73 /5.24

Table 2. One-group cross sections of neutron radiation capture by coolants:Pb-208 (bold) /Pb-nat in the ADS subcritical core subzones. Cross sections in mbarns are given.

The mean value of $\langle \sigma(n, g) \rangle$ for Pb-208 calculated from the data given in the Table 2 for subzones 1-9 is equal to 0.83 mbarn, that is by 6.2 times smaller than the mean value of $\langle \sigma(n, g) \rangle$ for Pb-nat (5.18 mbarn). This is evidence of small neutron absorption in the coolant from lead-208. This allows using the neutron surplus for reducing initial fuel load, increasing plutonium breeding, etc.

A possibility of using a low neutron absorbing coolant from lead-208 in the project of critical FR RBEC-M having 900 MW thermal power and fueled with uranium-plutonium [4] was also analyzed. In this reactor project the eutectic of lead (45%) and bismuth (55%) is envisaged. Pb-Bi coolant is characterized by relatively low melting temperature but has no potential in its using in full scaled nuclear power due to low stocks of bismuth and its high post radiation radio toxicity via polonium-210. As is known next generation of LFRs and ADSs, for example the BREST and the EFIT, are planned to be cooled with natural lead.

Neutron and physical characteristics of two reactors were calculated. Reactor RBEC-M is cooled with lead-bismuth as it has been firstly designed at the Kurchatov Institute, the Russian Federation. The second version of the reactor, RBEC-M, is cooled with lead-208 [5, 6]. These reactors are distinguished only by materials of coolant and coolant's temperatures are closely spaced. All other characteristics of these two reactors are similar. In these conditions the masses of uranium-plutonium fuel to reach criticality were calculated. The economy of the fuel load in the critical reactor RBEC-M cooled with lead-208 has been recognized. Then neutron fluxes in the core, lateral and low topical blankets of these two critical reactors were calculated. Calculations have been performed with the Monte Carlo code MCNP/4C, the special program NJOY and in using the neutron cross section data from the library ENDF/B-VII.0.

In Fig. 2 the microscopic cross sections of radiation capture of neutrons by lead-208 and Pb-Bi are given. The cross sections are taken from the ENDF/B-VII.0 library and they are represented in the ABBN-93 28 neutron energy group's approximation.

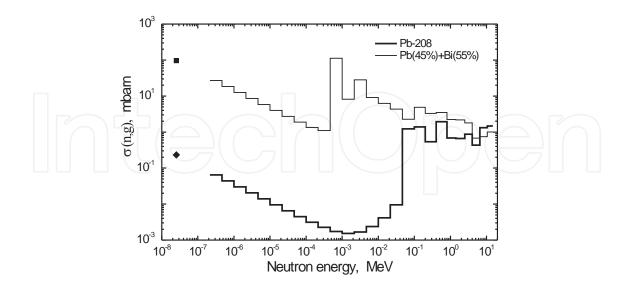


Figure 2. Microscopic cross sections of radiation capture of neutrons by lead-208 and Pb-Bi. Data are taken from the ENDF/B-VII.0 library.

From Fig. 2 follows that for all of neutron energies represented, the microscopic cross sections of neutron capture by Pb-Bi are larger in comparison with Pb-208. For energies below E_n <50 keV the cross sections of neutron radiation capture by lead-208 are by 3-5 orders of magnitude smaller than the cross section of neutron capture by Pb-Bi.

In Table 3 the one-group cross sections of neutron radiation capture by two various coolants, Pb-Bi and Pb-208, in RBEC-M neutron spectra are presented.

Name of reactor	Small fuel enrich-ment zone	Middle fuel enrich- ment zone	Big fuel enrich-ment zone	Lateral blanket zone		Topical v blanket below middle enrich- ment zone	big
RBEC-M (lead 45%- bismuth 55%)	3.7119	3.6239	3.6640	4.8288	5.3238	5.2248	5.4097
RBEC-M (lead-208 100%)	0.9330	0.9419	0.9393	0.8660	0.8087	0.8122	0.7901

Table 3. One-group cross sections of neutron radiation capture by coolants (Pb-Bi or Pb-208) of FR RBEC-M. Cross sections in millibarns are given.

From Table 3 follows that reactor RBEC-M with coolant from lead-208 is characterized with minimum value of mean one-group cross sections of neutron capture by the coolant in the core, <o>=0.94 millibarns. The corresponding mean cross section for the RBEC-M reactor core cooled by Pb-Bi is equal to 3.67 millibarns. As concerns the lateral and topical blankets, the one-group cross section of neutron capture by Pb-Bi exceeds the value of corresponding cross section for Pb-208 by 6-7 times. The small value of cross sections and corresponding excess of neutrons in the zones of the reactor RBEC-M cooled with Pb-208 can be used for reducing the fuel load in the reactor core, conversion of depleted uranium into plutonium and transmutation of minor actinides and long-lived fission products immersed in the lateral and topical zones.

3. Gain in core fuel loading due to excess of neutrons

As it follows from Table 1, in the ADS with subcritical blanket of 80 MW thermal power the calculated effective neutron multiplication factor, $K_{\rm ef}$, increases from $K_{\rm ef}$ =0.953 for coolant from Pb-nat to $K_{\rm ef}$ =0.970 for Pb-208 as coolant. In this replacement of the coolant proton beam power required to deliver 80 MW thermal blanket power will be reduced from 2.58 MW for Pb-nat to 1.68 MW for Pb-208. The gain in proton beam power equal to 0.9 MW arising in using low neutron absorbing coolant from Pb-208 is very valuable taking into account the high cost of 600 MeV energy proton beam power, approximately \$100 millions/MW according Ref. 7.

In LFR conditions the behavior of K_{ef} is very similar. In LFR RBEC-M of 900 MW thermal power in replacement its standard lead-bismuth coolant with lead-208 K_{ef} increases from its standard value, K_{ef} =1.00957, to the value K_{ef} =1.0246. It was calculated that to leave this coefficient at the standard level, i.e. to ensure the criticality, K_{ef} =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%. The economy in initial core fuel loading equal to 215 kg power grade plutonium per 340 MW of electrical power is very valuable taking into account that by this time the power grade plutonium stockpile reprocessed from light water reactors (LWRs) is expected to be insufficient for possible introducing in future a plenty of FRs in the countries quickly increasing their nuclear power. Now in France, the most advanced country on nuclear fuel reprocessing; only 8.5 tons/year of power grade Pu are available after reprocessing 1150 tons of spent fuel from LWRs.

4. Increasing the fuel breeding gain in FRs and ADSs cooled with lead-208

The excess of neutrons due to their small absorption in lead-208 can be used for fuel breeding and transmutation of long-lived radiotoxic fission products. As an example, we assume the radiation capture of neutrons by uranium-238 leading to creation of plutonium-239 [9].

The affectivity of this process will be as large as the value of one-group cross section of radiation neutron capture by uranium-238 nucleus is great. At neutron energies near to E_n =5-10 eV microscopic cross sections of neutron capture by uranium-238 have maximum equal to 170 barns as it can be seen from Fig. 3.

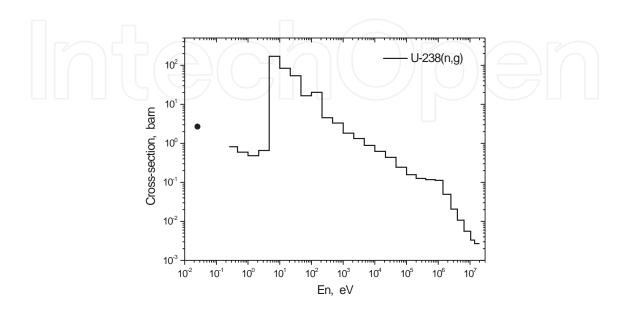


Figure 3. Microscopic cross sections of neutron radiation capture by U-238; taken from ENDF/B-VII.0 library.

That is why if the neutron spectra contains an increased share of neutrons of small and intermediate energies the corresponding one-group will be larger. In Table 4 the one-group cross sections of neutron radiation capture by U-238 in the ADS subcritical blanket cooled with Pb-208 or Pb-nat are given.

Target-source of neutrons	Subzone 1 0.34 /0.32	Subzone 2 0.45 /0.43	Subzone 3 1.23 /0.58
	Subzone 4 0.27 /0.23	Subzone 5 0.22 /0.28	Subzone 6 0.67 /0.35
	Subzone 7	Subzone 8	Subzone 9
	0.55 /0.46	0.46 /0.33	1.18 /0.61

Table 4. One-group cross sections of neutron radiation capture, $<\sigma(n,g)>$, by U-238 in the ADS subcritical core subzones cooled with Pb-208 (bold) /Pb-nat. Cross sections in barns are given.

The mean value of U-238 one-group radiation neutron capture cross section averaged over subzones 1-9 is equal to 0.6 barns for coolant from Pb-208 and to 0.4 barns for coolant from Pb-nat. The difference is due to excess of neutrons in the subzones 3, 6, 9 with smaller neutron energies at blanket's periphery and corresponding greater share of low energy neutrons.

In Fig. 4and Fig.5 the results of estimation of Pu-239 accumulation and U-238 incineration in the subzone 3 of the 80 MW thermal ADS are given. These calculations were performed using ACDAM code developed in the IPPE [8].

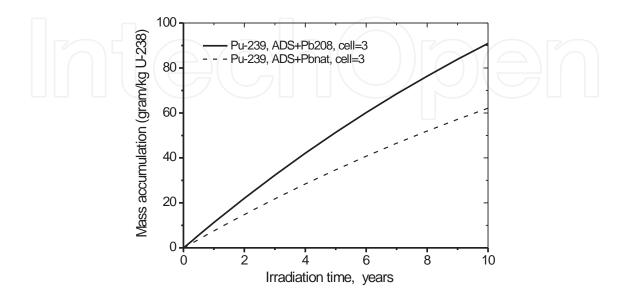


Figure 4. Accumulation of Pu-239 in grams in inserting 1 kg of U-238 in the neutron spectrum of the 80 MW ADS subzone 3. The bold line corresponds to ADS cooled with Pb-208 and the dash line - to ADS cooled with Pb-nat.

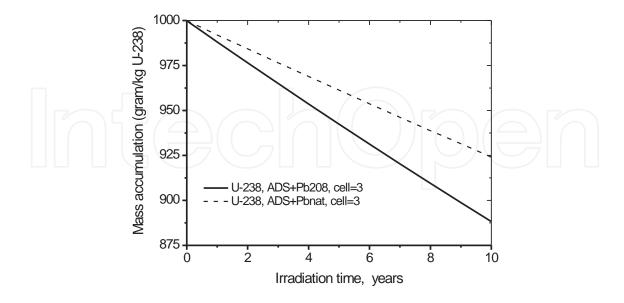


Figure 5. Mass decreasing of U-238 from its initial mass of 1 kg inserted in the neutron spectrum of the 80 MW ADS subzone 3. The bold line corresponds to ADS cooled with Pb-208 and the dash line - to ADS cooled with Pb-nat.

Similar behavior of U-238 one-group neutron capture cross section can be seen for LFR RBEC-M. The RBEC-M core is divided into 4 cells, its positions are shown in Table 5.

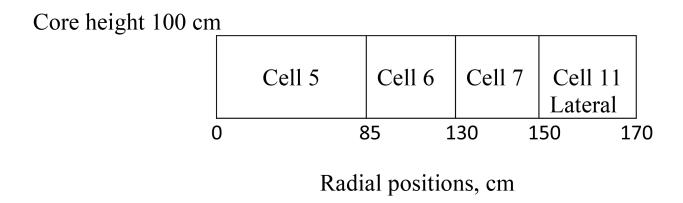


Table 5. Cell positions in the RBEC-M core.

In Fig.6 and Fig.7 neutron fluxes ratio acting in the cell number 5 and lateral cell number 11 cooled with Pb-208/Pb-Bi are given.

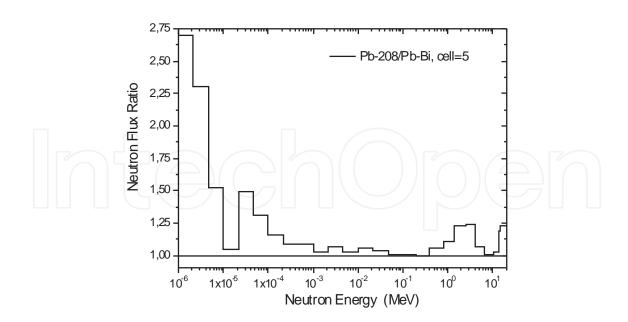


Figure 6. Neutron fluxes ratio in linear scale for RBEC-M cell 5 cooled with Pb-208/Pb-Bi.

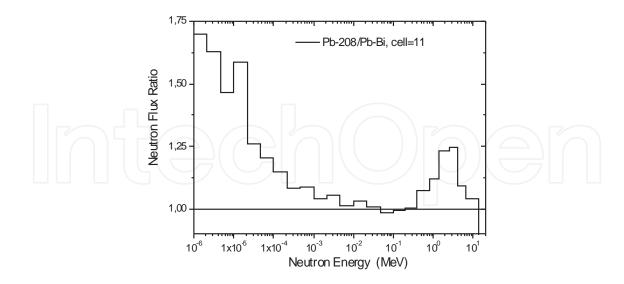


Figure 7. Neutron fluxes ratio in linear scaleforRBEC-M cell 11 (lateral blanket) cooled with Pb-208/Pb-Bi.

It can be seen that in using Pb-208 as coolant the share of main part of neutrons is increasing as well as the share of relatively small part of neutrons of low energy is also increasing. The last circumstance is due to small absorption of neutrons below their energy of 50 keV.

In Table 6 the one-group cross sections of neutron radiation capture by U-238 are given.

	Cell 5	Cell 6	Cell 7	Cell 11 - Lateral
<σ(n,g)/>, barns	0.273 /0.273	0.262 /0.261	0. 271 /0.269	0.575 /0.532

Table 6. One-group cross sections of neutron radiation capture, by U-238 in the RBEC-M core cells cooled with Pb-208 (bold) /Pb-Bi.

It is visible that one-group cross sections of neutron radiation capture, $\langle \sigma(n,g) \rangle$, by U-238in the RBEC-M lateral blanket is equal to 0.575 barns which is very close to the value of $\langle \sigma(n,g) \rangle = 0.6$ barns for ADS blanket.

The one-group cross sections for this nuclide averaged over neutron spectra of LFRs and ADSs cooled with lead-208 are approximately of 0.6 barns which are comparable with the one-group cross sections for typical breeders.

5. Hardening of ADS and FR neutron spectra in using lead-208 instead of natural lead or lead-bismuth

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

uranium-235, for example, is equal to 1.98 MeV. In Fig. 8 typical spectrum of neutrons in the core of lead fast reactor and spectrum of fission neutrons emitted by uranium-235 are given.

It is visible 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 incineration of minor actinides (MA). Incineration of long-lived radio toxic MA – neptunium, americium and curium – is one of the key problem of the nuclear power engineering. The world fleet of light water reactors (LWR) produces about 3.2 tons of MA per year as wastes. It is expected to incinerate MA in future ADSs which will be able consuming MA in quantities of 40% of fuel heavy atoms (h. a.). But ADS installation creation is a very expensive way and it needs a long time. It exists an opportunity to incinerate MA in FR core but from reactor's safety point of view the FR core can be loaded with MA in quantities not more than 2.5% of h. a. As alternative it is possible to load a radial blanket with the fuel having MA of 10% of h. a. But to avoid curium-242 accumulation during americium transmutation it is desirable to incinerate MA via their fission. In Fig. 9 microscopic fission cross section for one of MA, namely americium-241, is given.

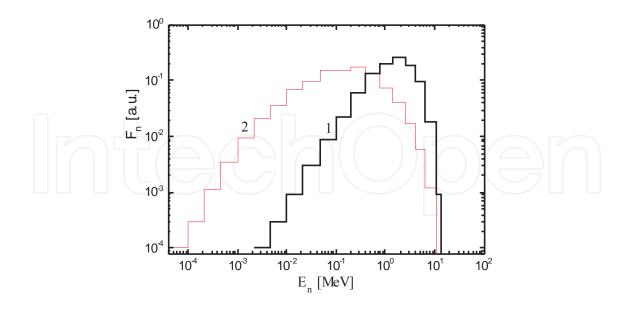


Figure 8. Neutron spectrum in the fuel zone of the 700 MW $_{thermal}$ LFR (2) and spectrum of U-235 fission neutrons (1) in the ABBN-93 neutron energy group system.

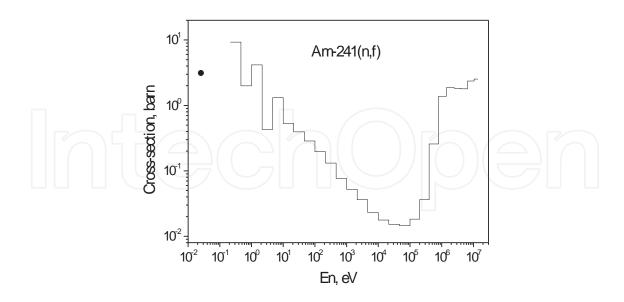


Figure 9. Microscopic fission cross sections for americium-241 taken from ENDF/B-VII.0 library.

It is visible that for fast neutron region there is a threshold equal to 0.1 MeV above which Am-241 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, Am-241 fission cross sections are also large enough but at these energies neutron capture cross sections are too large as it is shown in Fig. 10.

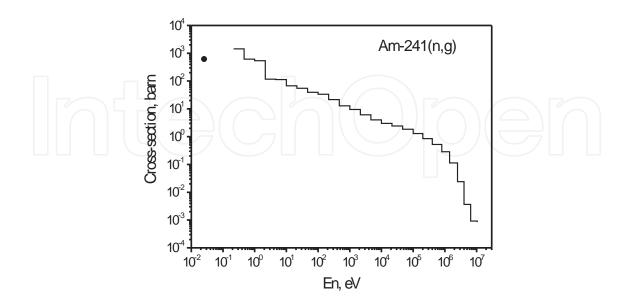


Figure 10. Microscopic neutron radiation capture cross sections for Am-241 taken from ENDF/B-VII.0 library.

For estimation of Am-241 incineration capability, the neutron spectra in 80 MW ADS subcritical blanket were calculated using MCNP-4C code and nuclear data library ENDF/B-VII.0. Calculations were performed in the ABBN-energy group structure system with 28 neutron energy groups. On the base of calculated neutron spectra for above mentioned 9 subzones of the ADS blanket the mean energies of neutrons and one-group fission cross sections for Am-241 were calculated. In Table 7 one-group fission cross sections calculated for Am-241 are given.

	Subzone 1	Subzone 2	Subzone 3
	0.2854/ 0.3310	0.2886 /0.1984	0.2197 /0.1168
Target-	Subzone 4	Subzone 5	Subzone 6
source of neutrons —	0.3335 /0.3725	0.3681/ 0.3334	0.2429 /0.2630
	Subzone 7	Subzone 8	Subzone 9
	0.2245/0. 1711	0.2483 /0.2594	0.2194 /0.1932

Table 7. One-group Am-241 fission cross sections in barns for subzones 1-9 of the 80 MW subcritical blanket cooled with Pb-208 (bold) and Pb-nat.

As follows from these calculations, the replacement of coolant from Pb-nat with Pb-208 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. on 6.4%. In this case the one-group Am-241 fission cross section averaged over 1-9 subzones increases from 0.2488 to 0.2700 barns, i.e. on 8.5%. In Fig. 11 the calculated dependence of one-group fission cross sections for Am-241 upon mean neutron energy is given. It is visible that these cross sections are growing as the neutron energy increases. From this Figure it can be concluded 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 uranium-235.

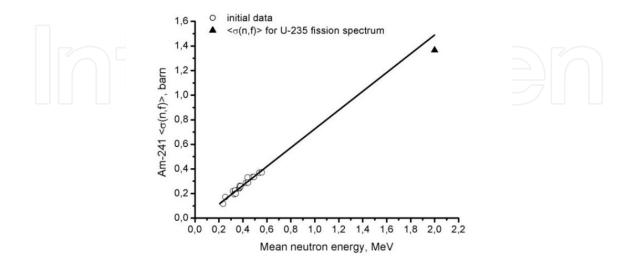


Figure 11. The calculated dependence of one-group fission cross sections for Am-241 upon mean neutron energy.

Similar calculations for LFR RBEC-M have been performed. For RBEC-M core and its lateral blanket the neutron spectra were calculated using MCNP5 code and nuclear data library ENDF/B-VII.0. Calculations were performed in the ABBN-energy group structure system with 28 neutron energy groups. On the base of calculated neutron spectra for several zones of the LFR core and lateral blanket the mean energies of neutrons were calculated. As follows from these calculations, the replacement of coolant from Pb-Bi with Pb-208 in RBEC-M leads to increasing the core mean neutron energy from its standard value 0.3992 MeV to 0.4246 MeV, i.e. on 6.4%. The one-group fission cross section for Am-241 included in core fuel in small quantities is increasing under these conditions from 0.2716 to 0.2981 barns, i.e. on 9.8%.

As concerns the RBEC-M lateral blanket, in switching coolant from Pb-Bi to Pb-208, its mean neutron energy increases from 0.2509 to 0.2662 MeV, i.e. on 6.1%. Under these conditions the one-group fission cross section for Am-241 is increasing up to 10%.

Thus, it can be concluded in nuclear installations with fast neutrons the replacement of natural lead or lead-bismuth with lead-208 as coolant leads to increasing the mean energy of neutrons approximately on 6% and corresponding increasing the one-group fission cross section of Am-241 on 8-10%. It must be mentioned once more that incineration of Am-241 via fission is more preferable than its transmutation because it allows avoiding creation of Cm-242 which has relatively high thermal emission that creates some difficulties in handling spent fuel containing MA.

6. Conclusion

It is shown that the one-group cross sections of neutron radiation capture, $\langle \sigma(n,g) \rangle$, for Pb-208 used as ADS and FR core coolant are equal to 0.8-0.9 mbarns, which are by 4-7 times smaller than the mean value of $\langle \sigma(n,g) \rangle$ for Pb-nat or Pb-Bi used as ADS and FR core coolant and by 2-3 times are smaller than for sodium coolant.

The mean value of $\langle \sigma(n, g) \rangle$ for U-238 leading to conversion into Pu-239 for ADS and FR core cooled with Pb-208 is equal to 0.6 barns which is comparable with the value of the same nuclide one-group cross section $\langle \sigma(n, g) \rangle$ for neutron spectrum of the FR core cooled with sodium.

In the ADS with subcritical blanket of 80 MW power the calculated effective neutron multiplication factor, $K_{\rm ef}$, increases on 1.7% in replacement of coolant from Pb-nat to Pb-208. In this replacement the proton beam power required to deliver 80 MW blanket power will be reduced to 0.9 MW.

In FR conditions the behavior of K_{ef} is very similar. In LFR RBEC-M of 900 MW thermal power in replacement its standard lead-bismuth coolant with lead-208 K_{ef} increases on 1.5%. This replacement leads to decreasing power grade Pu enrichment in the fuel from its initial value of 13.7% to 13.0% that means economy in fuel loading equal to 630 kg of power grade plutonium per 1 GW electrical.

The replacement of coolant from Pb-nat with Pb-208 in ADS blanket leads also to increasing the mean neutron energy averaged over the blanket from 0.3785 to 0.4026 MeV, i.e. on 6.4%. In the FR RBEC-M the replacement of coolant from Pb-Bi with Pb-208 leads to increasing the core mean neutron energy from its standard value 0.3992 MeV to 0.4246 MeV, i.e. also on 6.4%.

The harder spectrum of neutrons in ADS and FR core is for example preferable for incineration of Am-241 via its fission to avoid Cm-242 accumulation. It is shown that in neutron spectrum hardening on 6.4% the one-group Am-241 fission cross section averaged over ADS blanket or FR core increases on 8-10% from its initial value of 0.24 barns. In further increasing FR core neutron energy, if possible, the Am-241 fission cross section might be increased up to the value of 1.4 barns.

The advantages of the neutron and physical characteristics of molten Pb-208 allow considering it as a perspective material as coolant for next generation fast reactors.

The possibility of using Pb-208 as coolant in commercial fast critical or subcritical reactors requires a special considering but relatively high content of this isotope in natural lead, 52.3%, and perspectives of using high performance photochemical technique of lead isotope separation [10, 11] allow expecting to obtain in future such a material in large quantities and under economically acceptable price. Besides, the principal possibility of acquisition of radiogenic lead containing high enriched lead-208, up to 93%, exists [12].

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Author details

Georgy L. Khorasanov* and Anatoly I. Blokhin

*Address all correspondence to: khorasan@ippe.ru

State Scientific Centre of the Russian Federation – Institute for Physics and Power Engineering named after A.I. Leypunsky (IPPE), Russia

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