New Coolant from Lead Enriched with the Isotope Lead-208 and Possibility of Its Acquisition from Thorium Ores and Minerals for Nuclear Energy Needs

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1. Introduction

In critical and subcritical fast reactors functional materials fulfill various tasks, including:

- heat transfer from pins to heat exchangers,
- heat removal from ADS target under dissipation of high energy intensive proton beam in the liquid metal – a source of spallation neutrons.

As such of materials molten heavy metals – mercury, lead, eutectic of lead (45%) and bismuth (55%) and others are using or to be used in future.

Heavy metals posses acceptable for FRs and ADSs neutron and physical characteristics while due to some their properties, for example chemical passivity to water, high boiling temperature, they are better as coolant in comparison to liquid light metal which is now used in sodium cooled FRs such as BN-600 and BOR-60 in Russia.

One of important parameters of functional material considered is a value of neutron absorption in coolant because it is desirable the neutron losses in the core of FR and ADS blanket have to be minimized.

Ways of minimization of neutron absorption in FR are well-known: it is offered to use wrapper less fuel assemblies, low neutron absorbing nitrogen isotope ^{15}N in nitride fuel contents, structural materials with low cross section of neutron capture, etc.

The authors of this paper are pointing out on one more possibility of reducing the neutron losses in the core cooled with lead: it is connected with enrichment of lead isotope, lead-208, from its value in the natural lead isotope mix, equal to 52.3%, up to the value of 99.0% [1-9]. Lead-208 as a twice magic nucleus possesses a very low cross section of neutron radiation capture. This unique feature leads to economy of neutrons in the core and other profitable factors which are listed in the Part I of this paper.

The limiting factor of usage highly enriched ^{208}Pb as the coolant is its high price in the world market. In the ISTC #2573 project [10], executed in the RF, the opportunity of creation of the plant for separation of lead isotopes using selective photoreactions was considered. The complex of calculations and theoretical works were carried out, the outline sketch of the separation installation was developed, and economic and technical estimations of industrial production of highly enriched ^{208}Pb were made. Developers of the ISTC #2573 project expect that at the scale of manufacture equal to 150-300 kg of ^{208}Pb per year its price will be of US $200/kg [11]. But these theoretical predictions have not been confirmed experimentally yet.

Presently lead isotopes are separated in gaseous centrifuges in using tetra methyl of lead $Pb(CH_3)_4$ as a working substance. According to estimations given in Ref. 12 the price of lead-208 with enrichment of 99.0% will be about 1000-2000 US $/kg, which is relatively high for nuclear power plants. For comparison, another heavy metal coolant, Pb-Bi costs approximately 50 US $/kg.

Meanwhile, in nature besides lead of usual isotopic content: 1.48% Pb-204, 23.6% Pb-206, 22.6% Pb-207, 52.32% Pb-208, it can be found lead with higher enrichment of lead-208. Such type of lead can be found in ores and placers containing thorium. Lead-208 is a final product of decay the radioactive nucleus Th-232 and that is why such type of lead is called as radiogenic lead. The period of half decay of Th-232 nucleus is $1.4 \cdot 10^{10}$ year. In ancient ores ($\sim 3 \cdot 10^9$ year) the total content of thorium of 3-5 wt% is usual. In this case concentration of radiogenic lead reaches approximately 0.3 wt%. The enrichment of lead-208 in radiogenic lead is about 85-93%, depending on uranium content in ores and minerals. Uranium-238 produces in isotope mix the input of lead-206 which is product of uranium-238 radioactive decay.

As known, thorium containing ores and minerals can be found in India, Brazil, Australia, Ukraine, Russia and other countries. In Part 2 of this paper the possibility of reprocessing thorium containing ores and minerals for production of thorium-232 and lead-208 for nuclear engineering needs is discussed.

2. Advantages of using lead enriched with lead-208 as coolant of FR and ADS

2.1 Reducing of neutron absorption in cores of FRs and ADSs

In Fig. 1 microscopic cross sections of radiation neutron capture, s(n, g), by the lead isotopes ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb and the natural mix of lead isotopes ^{nat}Pb in the ABBN-93 (Abagian-Bazaziants-Bondarenko-Nikolaev of 1993 year) system of 28 neutron energy groups [13] are given. The cross sections are cited on the basis of files of the evaluated nuclear data for the ENDF/B-VII.0 version.

As can be seen, the microscopic cross sections of radiation neutron capture by the lead isotope ^{208}Pb for all of the 28 neutron energy groups of the ABBN-93 system are smaller than the cross sections of radiation neutron capture by the mix of lead isotopes ^{nat}Pb, and this difference is especially large, by 3-4 orders of magnitude, for intermediate and low energy neutrons, E_n <50 keV.

In Fig. 2 microscopic cross sections of radiation neutron capture, s(n, g), by the lead isotope ^{208}Pb and the eutectic lead(45%) – bismuth (55%) in the ABBN-93 system of 28 neutron energy groups are given. The cross sections are cited on the basis of files of the evaluated nuclear data for the ENDF/B-VII.0 version.

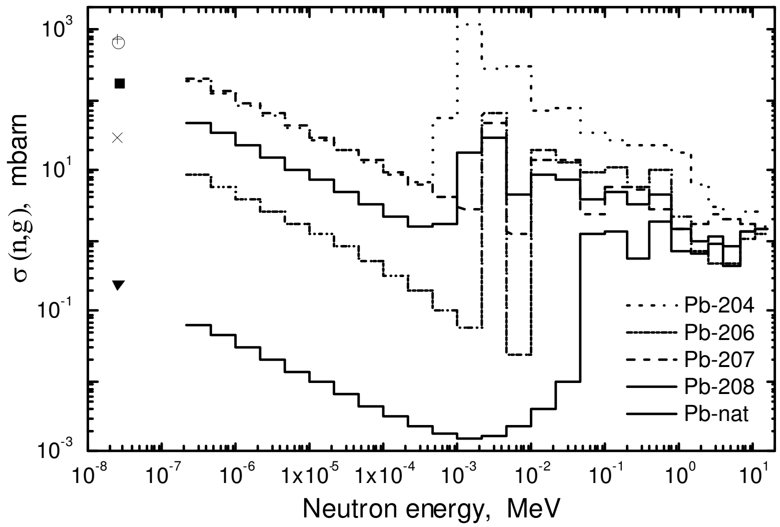

Fig. 1. Microscopic cross sections of radiation neutron capture s(n,g) by stable lead isotopes and by natural mix of lead isotopes taken from the ENDF/B-VII.0 library.
Cross sections are represented in the ABBN-93 system of 28 neutron energy groups.

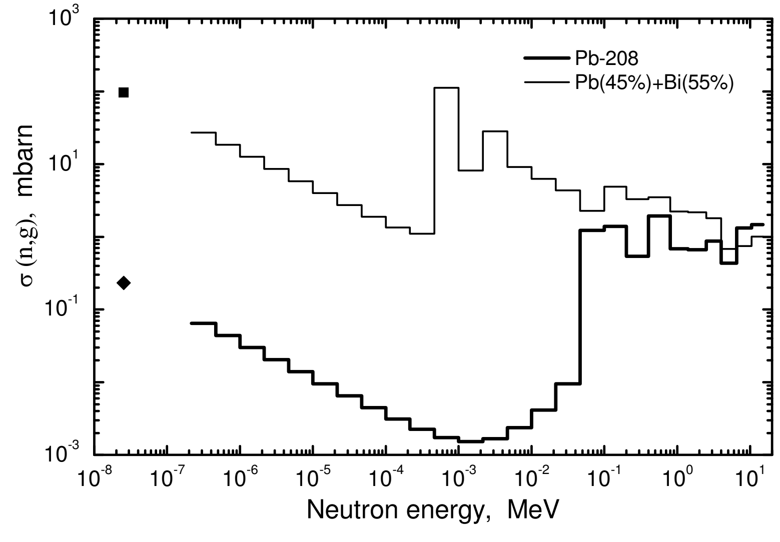

Fig. 2. Microscopic cross sections of radiation neutron capture s(n, g) by stable lead-208 isotope and by the eutectic Pb-nat(45%) – Bi (55%) taken from the ENDF/B-VII.0 library.
Cross sections are represented in the ABBN-93 system of 28 neutron energy groups.

As can be seen, the microscopic cross sections of radiation neutron capture by the lead isotope ^{208}Pb for all of the 28 neutron energy groups of the ABBN-93 system are smaller than the cross sections of radiation neutron capture by mix of lead ^{nat}Pb (45%) and bismuth, Bi

(55%), and this difference is especially large, by 3-5 orders of magnitude, for intermediate and low energy neutrons, E_n <50 keV.

Share of neutrons with energies less than 50 keV, E_n<50 keV, usually is about 20-25% of all neutrons in FR or ADS cores and it increases in lateral and topical blankets of the core.

In Table 1 one-group cross sections of neutron radiation capture by two types of coolants - Pb-208 or the eutectic of Pb-Bi – in the lead-bismuth fast reactor project named as RBEC-M and designed in the Russian Kurchatov Institute [14] are given.

Reactor and its coolant	Core 1 with small enrichment of fuel	Core 2 with middle enrichment of fuel	Core 3 with large enrichment of fuel	Lateral blanket	Topical blanket under core 1	Topical blanket under core 2	Topical blanket under core 3
RBEC-M, Pb-Bi	**3.71190**	**3.62388**	**3.66404**	**4.82878**	**5.32383**	**5.22481**	**5.40967**
RBEC-M, Pb-208	**0.93296**	**0.94187**	**0.93931**	**0.86595**	**0.80867**	**0.81212**	**0.79005**

Table 1. One-group cross sections of radiation neutron capture by various coolants in the fast reactor RBEC-M core consisted from core 1, 2, and 3. Data are given for the standard lead-bismuth coolant, as it has been designed at the Kurchatov Institute, and for lead-208 coolants, proposed by authors of this paper.
Cross sections in millibarns are given.

From Table1 follows that the coolant from lead-208 is characterized with minimum one-group cross section, about <s>=0.93-0.94 millibarns. In standard lead-bismuth coolant the value of the same one-group cross section is by ~4 times bigger, about <s>=3.62-3.71 millibarns. In lateral and topical blankets one-group cross sections for Pb-208 by ~6-7 times are less than for Pb-Bi. The small values of one-group sections in RBEC-M cooled with lead-208 and corresponding excess of neutrons can be used for minimization of fuel load of the core, increasing fuel breeding and transmutation of long-lived fission products in lateral and topical blankets.

2.2 Hardening of neutron spectra in FRs and ADSs cooled with lead-208

In Fig.3 neutron spectra for the core 1(small enrichment of fuel) of the reactor RBEC-M cooled with its standard PB-Bi coolant and proposed Pb-208 coolant are given. Spectra were calculated for cases when their total fluxes were similar and the neutron multiplication factors were equal to 1 in using both of coolants. It can be seen that replacement of standard lead-bismuth coolant in RBEC-M leads to neutron hardening: the mean neutron energy increases from the value of 0.402 MeV to 0.428 NeV , i.e. on 6.5%.

In Fig.4 the ratio of neutron fluxes in the core 1 of RBEC-M in linear scale is represented. It is shown the increasing of share of fast neutrons (E_n>0.4 MeV) and the increasing of the very low share (less than 1%) of neutrons with energies E_n <100 eV in the core cooled with lead-208. In whole the mean neutron increases on 6.5% as it has been mentioned above.

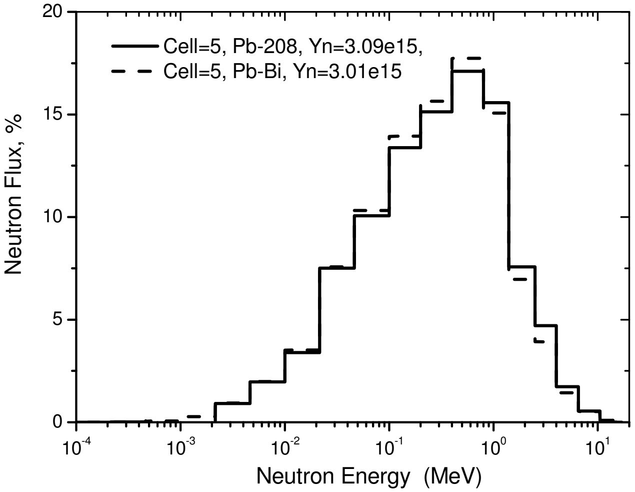

Fig. 3. Neutron spectra for the core1(small enrichment of fuel) of the reactor RBEC-M cooled with its standard Pb-Bi coolant (dash line) and Pb-208 coolant (solid line).
Yn- total flux of neutrons in core 1.

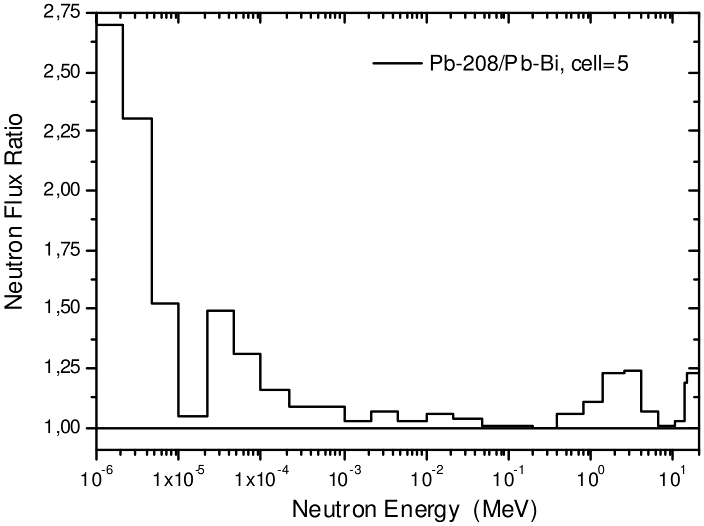

Fig. 4. The ratio of neutron fluxes in the core 1 of RBEC-M given in linear scale.The core is cooled with lead-208 leading to increasing the mean neutron energy on 6.5%.

2.3 Increasing effective neutron multiplication factor in FRs and ADSs cooled with lead-208

In the reactor RBEC-M in replacement its standard coolant to lead natural its effective neutron multiplication factor, K_{ef}, decreases from its standard value, K_{ef}=1.0096, to the value

K_{ef}=0.9815. But replacement of Pb-Bi to lead-208 leads to the value K_{ef}=1.0246, i.e. K_{ef} increases approximately on 1.5%. For reducing this increased value to the standard value, K_{ef}=1.0096, the plutonium enrichment must be decreased from its initial value equal to 13.7% as designed in lead-bismuth RBEC-M project to the value equal to 13.0%. It means that initial plutonium fuel loading must be decreased from 3595 kg to 3380 kg, i.e. on 215 kg. Thus, it means that economy of plutonium will be of 650 kg per 1 GW electrical power in using lead-208 as coolant instead of lead-bismuth in RBEC-M type reactors. It can be noted, that this quantity of power grade plutonium is comparable with the annual value of plutonium quantity, about 1 tone, which is now obtaining after reprocessing the spent fuel of Russian NPPs – VVER-440 and BN-600.

In the ADS with subcritical blanket of 80 MW thermal power [5] K_{ef} increases approximately on 1.7% in replacement lead natural as coolant to lead-208, from its value of K_{ef}=0.95289 for lead natural to K_{ef}=0.96997 for lead-208. In this case to liberate the nominal 80 MW thermal power in the blanket the power of the proton beam can be reduced from 2.59 MW to 1.68 MW, i.e. by 1.5 times.

2.4 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. Here, as an example, we assume the radiation capture of neutrons by uranium-238 leading to creation of plutonium-239. 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 large. In Fig.5 microscopic cross sections of radiation neutron capture by U-238 taken from ENDF/B-VII.0 library are given.

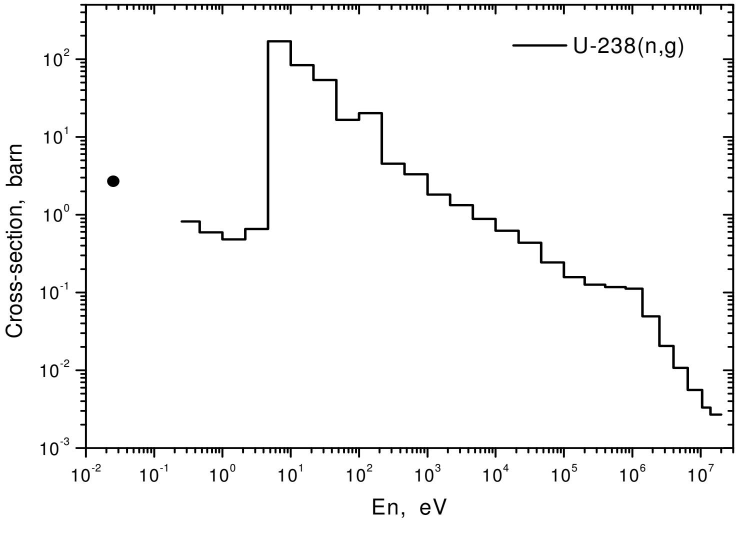

Fig. 5. Microscopic cross sections of radiation neutron capture by uranium-238 taken from ENDF/B-VII.0 library.

From Fig.5 it can be seen that at neutron energies near to E_n=5-10 eV these cross sections have maximum equal to 170 barns. That is why if the neutron spectra contains an increased share of neutrons of small and intermediate energies the corresponding one-group will be large enough.

In table 2 the one-group cross sections of radiation neutron capture by U-238 averaged over neutron spectra of the 80 MW ADS and various FRs (BREST, BN-600 and RBEC-M) are given.

Reactor	Coolant	One-group cross sections in barns
ADS-80 MW th.	Pb-208	**0.6393**
ADS-80 MW th.	Pb-nat	**0.4053**
BREST-300 MW el.	Pb-nat	**0.3089**
BN-600 MW el.	Na-23	**0.2965**
RBEC-M -340 MW el.	Pb-208	**0.1874**
RBEC-M-340 MW el.	Pb-Bi	**0.1886**

Table 2. One-group cross sections of radiation neutron capture by U-238 averaged over neutron spectra of the 80 MW ADS and various FRs (BREST, BN-600 and RBEC-M) cores. Cross sections in barns are given.

It can be noted that one-group cross section of neutron capture by uranium-238 in ADS spectrum is by 2.15 times bigger than for sodium reactor BN-600 spectrum and this fact indicates to the possibility of enhancing the breeding gain in the blanket of ADS 80 MW cooled with lead-208.

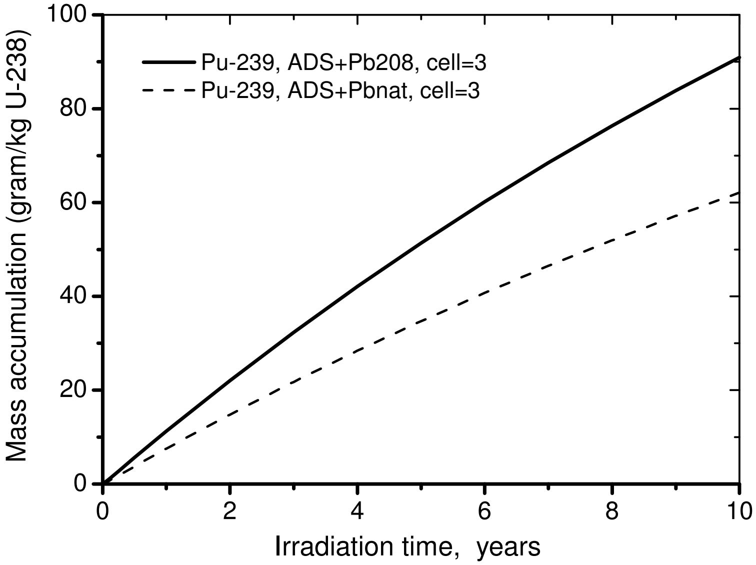

Fig. 6. Mass accumulation of Pu-239 in the ADS 80 MW subcritical blanket in inserting 1 kg of U-238. in the cell 3, near the blanket's far margin. The solid curve corresponds to the case, when the blanket is cooled with lead-208, the dash curve – to the case, when the blanket is cooled with lead natural.

As an illustration, in Fig. 6 and 7 the results of burning 1 kg of uranium-238 placed in the one part of ADS 80 MW subcritical blanket (cell 3 near the blanket far margin) and corresponding accumulation of plutonium-239 are given. Calculations have been performed on the basis of code ACDAM [15] developed at the IPPE Centre of nuclear data.

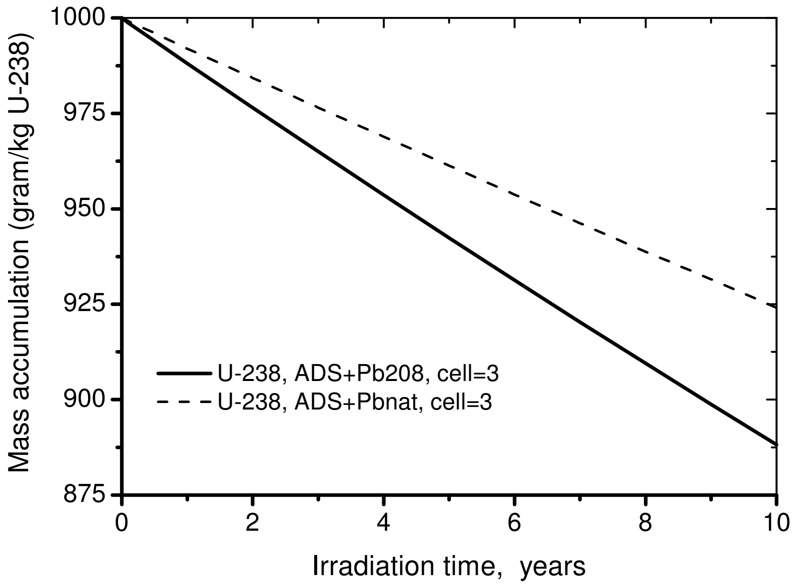

Fig. 7. Mass burning of 1 kg of U-238 in the neutron spectra of 80 MW ADS blanket, in the cell 3 which is near far blanket's margin. The solid curve corresponds to the case, when the blanket is cooled with lead-208, the dash curve – to the case, when the blanket is cooled with lead natural.

3. On the possibility of acquisition of radiogenic lead enriched with lead-208

3.1 On the sources of radiogenic lead enriched with lead-208 in Russia

The problem of acquisition of radiogenic lead enriched with lead-208 is coupled with perspectives of involving thorium into nuclear power engineering of Russia. As it is noted in Ref.16 to develop the thorium nuclear energetic it is necessary to obtain at least 10-13 thousand tones of thorium per year at the stage of 20-30 years of this century.

Content of lead-208 in thorium ores and minerals can reach 0.3-0.5% wt of thorium mass. In acquisition 10-13 thousand tones of thorium per year it will be possible to recover about 65 tones of radiogenic lead per year. This quantity of lead is insufficient to cover the needs in lead coolant of large scale nuclear power which requires approximately 2000 tones of lead per 1 GW of electrical power. But 65 tones of lead are sufficient to cool the blanket of 80 MW_{th} ADS. About 700 tones of lead can be enough to cool the reactor RBEC-M delivering 340 MW electrical.

As it is shown in Ref. 16, the main source of thorium in Russia is the Lovozerskoe deposit at Kola Peninsula. Estimations show that in reprocessing 2 mln tones of loparit ore per year 500-600 thousand tones of Ln_2O_3 and TiO_2, 100 thousand tones of Nb_2O_5, 10 thousand tones of Ta_2O_5, 13 thousand tones of ThO_2 and 65 tones of radiogenic lead can be produced. In Ref 16 the conclusion was made that is possible to extract in near future large quantities of thorium from the progress of industry and as co-product of rear metal raw.

The separate problem is the level of lead-208 enrichment of lead-208 in various deposits. It can be strongly different. For example, in Brazil monazites radiogenic lead is enriched by lead-208 up to 88.34% [17]. For FRs and ADSs it can be desirable the following isotopic composition of radiogenic lead: lead-208-93% and lead-206-6% with minimum content of lead-207 – the isotope with large cross section of neutron capture. In Ref. 18 the data concerning thorium-containing ores and monazites in the world scale are given. The authors of this paper pointed out that as a rule radiogenic lead contains very small quantities of lead-204 and lead-207–isotopes with large cross sections of neutron capture.

It can be noted that the advantages of lead-208 can be used, besides nuclear power plants, in other branches of nuclear science and technology. It seems that lead-208 as low moderating material will be preferable in the lead slowing down neutron spectrometers [19] and also in the spallation neutron sources to have the harder neutron spectra under interaction of high energy protons with liquid proton target from lead-208 [2, 20].

3.2 Prospects of ancient monazite from placers and bed-rock's deposits of Ukraine as the raw materials to produce highly enriched ^{208}Pb

Monazite is the phosphate containing mainly ceric rare earths ((Ce, La, Nd …, Th) PO_4) and is the main natural concentrator of thorium. It is widely spread (though usually in small amounts) in rocks and some types of ores. Owing to chemical and mechanical durability monazite is accumulated in placers.

The crystal structure of monazite can be presented as three-dimensional construction of oxygen nine apex polyhedron with rare-earth center atoms and oxygen tetrahedrons with the central atom of phosphorus. Nine-fold coordination allows a wide occurrence of relatively large ions of the light rare earths and thorium in mineral structure. The total content of thorium in a mineral can reach 28 wt%, and concentration of 5-7 wt% is usual. Though there are no experimental data about the form of radiogenic lead presented in the monazite structure, the numerous data, summarized for example in work [21], argued for its good stability in a monazite crystal matrix that allows monazite to be used for isotope dating.

In Ukraine monazite contains in developed fine-grained titanium-zirconium placers. By the explored easily enriched titanium-zirconium ores Ukraine comes to the forefront in Europe and in the CIS. The resources of zirconium in Ukraine make more than 10% of world ones. Now the largest Malyshevsky (Samotkansky) placer is developed and the working off of the Volchansky placer has been started.

Owing to the marked paramagnetism monazite at existing capacity of mines can be taken in passing by working out of placers in quantity of about 100 tons per year that corresponds approximately to 3.5 tons of thorium and 0.5 tons of the lead enriched with 208 isotope. Now monazite is considered as a harmful radioactive impurity and it is not produced.

The composition of monazite from the Malyshevsky placer as to the amounts of U, Th and Pb for dating purposes is well studied in work [21] by means of X-ray-fluorescent technique specially developed for individual grain analysis. In Table 3 the data about the contents of thorium, uranium and about isotope contents of lead for monazite of the Malyshevsky deposit is cited. The average composition of lead is confirmed by direct mass spectrometry determinations.

Average values from 224 X-ray-fluorescent determinations according to [21] data, wt %.			Isotopic composition of lead by mass spectrometry analysis of average sample, relative %%.				Average value of 70 uranium depleted samples. Elements – mass %%, Lead isotopes –relative %%.					
										Lead isotopes		
Th	U	Pb	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb	U	Th	Pb	^{206}Pb	^{207}Pb	^{208}Pb
3,52	0,23	0,30	0,04	13,11	1,43	85,42	0,06	3,63	0,33	3,8	0,4	95,7

Table 3. Contents of thorium, uranium, lead and isotopic composition of lead for monazite of the Malyshevsky placer (Ukraine)

As is seen from Table 3, enrichment by ^{208}Pb in the average for all monazite is insufficiently high. However, there is a probability of monazite separation by the flotation, magnetic or other characteristics with release of low uranium fraction of the mineral.

Extraction of total monazite concentrate by working out of the Malyshevsky placer scattering of an average almost won't demand additional costs and its price as at first approximation can be accepted as the equal to zircon concentrate, i.e. ~ 1 US $/ kg. Cost of hydrometallurgical emanation of lead from monazite by analogy with similar processes can be estimated as (24÷30 US $/ kg). The removal of differences with low U/Th ratio and the high content of ^{208}Pb from monazite concentrate will require additional researches and will cause some rise in price of a product.

In Ukraine there are insufficiently studied shows of monazite in ancient radical breeds, their barks of aeration and in placers, i.e. enriched ^{208}Pb. According to the available analytical data there is a possibility to detect monazite with highly enriched ^{208}Pb.

For extraction of thorium and the lead enriched with 208 isotope Russia has a great opportunities by preparation the fine-grained titanium-zirconium placers for development and by the extraction from raw materials in complex deposits.

4. Conclusions

The paper is dedicated to the proposal of using lead enriched with the stable isotope ^{208}Pb in FRs and ADSs instead of lead natural, ^{nat}Pb.

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

The main advantage of ^{208}Pb is its low neutron absorption ability: for neutron energies E_n=0.1-20.0 MeV the microscopic cross sections of radiation neutron capture by ^{208}Pb are by 1.5-2.0 times smaller as compared with ^{nat}Pb, and for energies, E_n<50 keV, the difference in the cross section values reaches 3-4 orders of magnitude. Averaged over neutron spectra of the LFR or ADS the one-group cross sections for a coolant from ^{208}Pb are by 5-6 times smaller than those for the coolant consisted from ^{nat}Pb.

The second advantage of using ^{208}Pb consists in achievement the core neutron spectra hardening on 5-6% due to low energy losses. Low neutron absorbing and moderating features of ^{208}Pb permit to reach the gain in the multiplication factor K_{ef} on 2-3% for critical or subcritical core fueled with U-Pu mix. In this case to have the multiplication factor K_{ef} =1.01 for the LFR or K_{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 on 0.7-0.8%.

The third important advantage of using ^{208}Pb is coupled with increasing the small share of neutrons of low energies, 5-10 eV in spite of the neutron spectra hardening in whole. In this region of neutron energies the microscopic cross sections for such nuclides as ^{238}U and ^{99}Tc are maximum and very high, and the one-group cross sections for these nuclides averaged over neutron spectra of LFRs and ADSs cooled with lead-208 are equal to 0.6 and 0.8 barn respectively which are comparable with the one-group cross sections for typical breeders and transmutters.

The possibility of using ^{208}Pb 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 permit to expect obtaining in future such a material in large quantities and under economically acceptable price. In the paper it is shown that principal possibility of acquisition of radiogenic lead containing high enriched lead -208, up to 93%, exists. Nowadays in Russian Federation and Ukraine thorium- containing loparit ores and monazite minerals are reprocessed for production of rare metal raw. Thorium and lead are not required now and they are deposited in sludge. Nevertheless, the scales of future thorium and radiogenic lead production for innovative nuclear reactors have some prospects in near-term future. The conclusion is made that to obtain the minimum amount of required in future radiogenic lead (65 t/year) for small sized FRs and ADSs the very large quantities of loparit ores or monazite minerals must be reprocessed and acquisition of radiogenic lead-208 can be economically acceptable as a co-product of rare metal raw.

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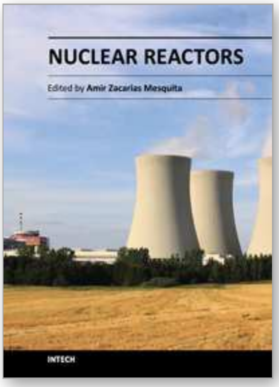

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This book presents a comprehensive review of studies in nuclear reactors technology from authors across the globe. Topics discussed in this compilation include: thermal hydraulic investigation of TRIGA type research reactor, materials testing reactor and high temperature gas-cooled reactor; the use of radiogenic lead recovered from ores as a coolant for fast reactors; decay heat in reactors and spent-fuel pools; present status of two-phase flow studies in reactor components; thermal aspects of conventional and alternative fuels in supercritical water?cooled reactor; two-phase flow coolant behavior in boiling water reactors under earthquake condition; simulation of nuclear reactors core; fuel life control in light-water reactors; methods for monitoring and controlling power in nuclear reactors; structural materials modeling for the next generation of nuclear reactors; application of the results of finite group theory in reactor physics; and the usability of vermiculite as a shield for nuclear reactor.

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