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Research Article

Radiogenic Lead with Dominant Content of ^{208}Pb: New Coolant and Neutron Moderator for Innovative Nuclear Facilities

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As a rule materials of small atomic weight (light and heavy water, graphite, and so on) are used as neutron moderators and reflectors. A new very heavy atomic weight moderator is proposed—radiogenic lead consisting mainly of isotope ^{208}Pb. It is characterized by extremely low neutron radiative capture cross-section (0.23 mbarn for thermal neutrons, i.e., less than that for graphite and deuterium) and highest albedo of thermal neutrons. It is evaluated that the use of radiogenic lead makes it possible to slow down the chain fission reaction on prompt neutrons in a fast reactor. This can increase safety of the fast reactors and reduce as well requirements pertaining to the fuel fabrication technology. Radiogenic lead with high ^{208}Pb content as a liquid-metal coolant of fast reactors helps to achieve a favorable (negative) reactivity coefficient on coolant temperature. It is noteworthy that radiogenic lead with high ^{208}Pb content may be extracted from thorium (as well as thorium-uranium) ores without isotope separation. This has been confirmed experimentally by the investigations performed at San Paulo University, Brazil.

1. Introduction

In the nature there are two types of elemental lead with substantially different contents of four stable lead isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb). The first type is a natural, or common, lead with a constant isotopic composition (1.4% ^{204}Pb, 24.1% ^{206}Pb, 22.1% ^{207}Pb, and 52.4% ^{208}Pb). The second type is a so-called "radiogenic" lead with very variable isotopic composition. Radiogenic lead is a final product of radioactive decay chains in uranium and thorium ores. That is why isotopic compositions of radiogenic lead are defined by the ore age and by elemental compositions of mixed thorium-uranium ores including content of natural lead as an impurity.

The paper presents some results obtained in neutronics and thermal-hydraulics evaluations of the benefits from the use of radiogenic lead instead of natural lead as a coolant of fast breeder reactors. The paper demonstrates that substitution of radiogenic lead for natural lead can offer the following benefits for operation of fast breeder reactors.

(i) Improvement of the reactor safety thanks to the better values of coolant temperature reactivity coefficient (CTRC).

(ii) Improvement of some thermal-hydraulic reactor parameters.

(iii) Possibility for development of high-flux accelerator-driven systems (ADSs).

(iv) Upgrade of proliferation resistance for advanced Pu-containing fuels.

These benefits are described below in detail. At the end, the paper presents some information about natural resources of radiogenic lead and about its isotopic compositions.

2. Neutron-Physical Properties and Advantages of Radiogenic Lead

The term "radiogenic lead" is used here for designation of lead that is produced in radioactive decay chains of thorium and uranium isotopes. After a series of alpha- and beta-decays, ^{232}Th is transformed into stable lead isotope ^{208}Pb, ^{238}U—into stable lead isotope ^{206}Pb, ^{235}U—into stable lead isotope ^{207}Pb. Therefore, uranium ores contain radiogenic lead consisting mainly of ^{206}Pb while thorium and mixed thorium-uranium ores contain radiogenic lead consisting

Table 1: Neutron-physical characteristics of some materials.

Nuclide	σ_{elastic} (barn)	Number of collisions (0.1 MeV → 0.5 eV)	$\sigma^{th}_{(n,\gamma)}$ (mbarn)	$RI_{n,\gamma} + 1/V$ (mbarn)
^{1}H	30.1	12	332	149
^{2}D	4.2	17	0.55	0.25
^{9}Be	6.5	59	8.5	3.8
^{12}C	4.9	77	3.9	1.8
^{16}O	4.0	102	0.19	0.16
Pb_{nat}	11.3	1269	174	95
^{208}Pb	11.5	1274	0.23	0.78

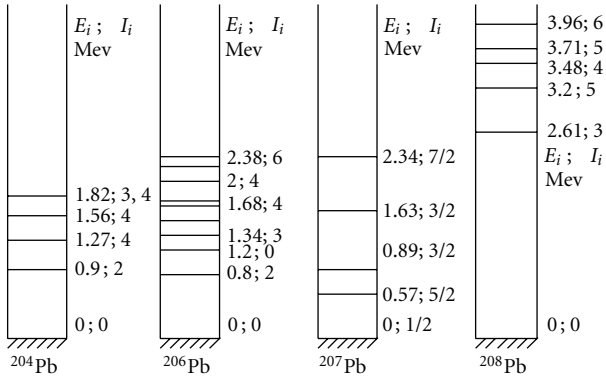

Figure 1: The excitation levels of lead nuclides.

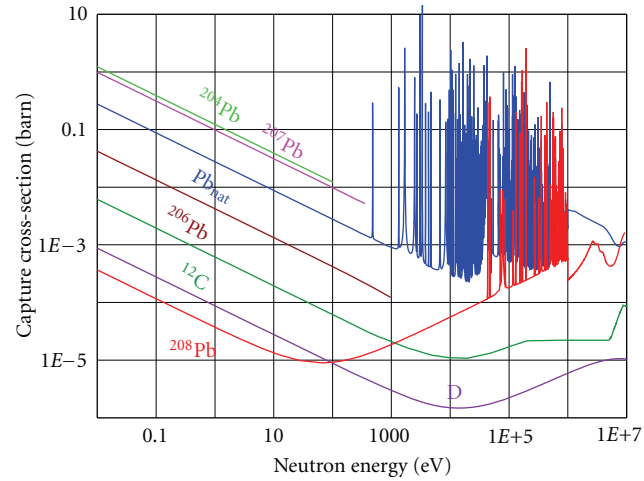

Figure 2: Capture cross-section of various nuclides as a function of neutron energy.

mainly of ^{208}Pb. Sometimes, the presence of natural lead in uranium and thorium ores can change isotope composition of radiogenic lead. Anyway, isotope composition of radiogenic lead depends on elemental composition of the ores where this lead is extracted from.

Radiogenic lead consisting mainly of stable lead isotope ^{208}Pb can offer unique advantages, which follow from unique nuclear physics properties of ^{208}Pb. This lead isotope is a double-magic nuclide with completely closed neutron and proton shells. Therefore, the excitation levels of ^{208}Pb nuclei (see Figure 1) are rather high (2.61–3.96 MeV) while the excitation levels of other lead isotopes are placed in relatively lower energy range (0.57–2.38 MeV). ^{208}Pb is an extremely weak neutron absorber.

Unusual nuclear properties of ^{208}Pb can produce the following effects on main parameters of chain fission reaction in a nuclear reactor core. Firstly, since energy threshold of inelastic neutron scattering by ^{208}Pb (~2.61 MeV) is higher substantially than that by natural lead (~0.8 MeV), ^{208}Pb can soften neutron spectrum in the high-energy range to a remarkably lower degree. Secondly, neutron radiative capture cross-section of ^{208}Pb in thermal point (~0.23 mb) is lower by three orders of magnitude than that of natural lead (~174 mb) and even lower than that of reactor-grade graphite (~3.9 mb). These differences remain large within sufficiently wide energy range (from thermal energy to some tens of kilo-electron-volts). Energy dependencies of neutron absorption cross-sections are presented in Figure 2 for natural lead, stable lead isotopes, graphite, and deuterium [1].

If radiogenic lead consists mainly of ^{208}Pb (about 90% ^{208}Pb plus 9% ^{206}Pb and about 1% ^{204}Pb + ^{207}Pb), then such a lead absorbs neutrons as weak as graphite within the energy range from 0.01 eV to 1 keV. Such isotope compositions of radiogenic lead can be found in thorium and in mixed thorium-uranium ores.

Thus, on the one hand, ^{208}Pb, being heavy nuclide, is a relatively weak neutron moderator in elastic scattering reactions within full neutron energy range of nuclear reactors because of heavy atomic mass and in inelastic scattering reactions with fast neutrons because of high energy threshold of these reactions. On the other hand, ^{208}Pb is an extremely weak neutron absorber within wide enough energy range. Weak neutron absorbing and moderating properties of ^{208}Pb make it possible to develop an advanced reactor design with very flexible neutron spectrum (from fast to thermal), with good neutron economy and without highly pressurized coolant in the reactor core.

Some nuclear characteristics of light (hydrogen, deuterium, beryllium, graphite, oxygen) and heavy materials (natural lead and lead isotope ^{208}Pb) are presented in Table 1 [1].

Table 2: Properties of neutron moderators at 20°C.

Moderator	Average logarithmic energy loss, ξ	Moderating ratio, $\xi\Sigma_s/\Sigma_a^{th}$	Neutron age τ (cm^2) (0.1 MeV → 0.5 eV)	Diffusion length L (cm)	Mean lifetime of thermal neutrons T_{th} (ms)
H_2O	0.95	70	1.35	3	0.2
D_2O	0.57	4590	58	147	130
BeO	0.17	247	66	37	8
^{12}C	0.16	242	160	56	13
Pb_{nat}	0.00962	0.6	3033	13	0.8
^{208}Pb	0.00958	477	2979	341	598

One can see that elastic cross-sections of natural lead and ^{208}Pb do not differ significantly from the others, being between the corresponding values for hydrogen and other light nuclides. Neutron slowing down from 0.1 MeV to 0.5 eV requires from 12 to 102 elastic collisions with light nuclides while the same neutron slowing down requires about 1270 elastic collisions with natural lead or ^{208}Pb. The reason is the high atomic mass of lead compared to the other light nuclides. From this point of view neither natural lead nor ^{208}Pb is an effective neutron moderator.

Taking into account that capture cross-section at thermal energy and capture resonance integral of natural lead are much larger than the corresponding values of the most light nuclides, it is safe to say that neutrons are captured during the slowing-down process in natural lead with a higher probability compared to the slowing-down process in light materials. So, only a small part of neutrons will slow down to thermal energy. This means that thermal neutron flux in natural lead will be much lower than that in the light materials.

Since ^{208}Pb is a double magic nuclide with closed proton and neutron shells, radiative capture cross-section at thermal energy, and resonance integral of ^{208}Pb is much lower than the corresponding values of lighter nuclides. Therefore, it can be expected that even with multiple scattering of neutrons on ^{208}Pb during the process of their slowing down, they will be slowed down to thermal energy with a high probability and thus create a high thermal neutron flux.

Some properties of neutron moderators at 20°C are presented in Table 2 [1–3]. It can be seen that the average logarithmic energy loss of neutrons in their elastic scattering by natural lead and ^{208}Pb is many times less than that for light nuclides. The reason is a much heavier atomic mass of lead compared to atomic mass of other light moderators. However, thanks to very low neutron capture cross-section, the moderating ratio, that is, the average logarithmic energy loss times scattering cross-section divided by absorption cross-section, of ^{208}Pb is much higher than that for light moderators. This means that ^{208}Pb could be a more effective moderator than such well-known light moderators as light water, beryllium oxide and graphite.

Age of neutrons slowed down in lead is significantly higher than that in light moderators. This parameter defines a mean distance between the place where neutrons were generated and the place where they were slowed down. Therefore, the use of lead in an ADS blanket may allow formation of thermal neutron spectrum at a longer distance from the target than that in light moderators. So, it may allow obtaining more space for placing the radioactive wastes to be transmuted. Also, the problem of neutron leakage may be weakened.

3. Improvement of the Reactor Safety

Spectral component of CTRC in sodium-cooled fast breeder reactor loaded with mixed uranium-plutonium oxide fuel has a large positive value [4]. The situation is nearly the same in the case when natural lead is used as a coolant. The positive CTRC values can produce an unfavorable effect on safety of the reactor operation because reactivity can rise when coolant temperature increases. Increase of coolant temperature can result in the following:

(i) decreasing effective neutron multiplication factor (K_{eff}) due to larger neutron leakage;

(ii) increasing K_{eff} due to smaller capture of neutrons;

(iii) increasing K_{eff} due to harder neutron spectrum.

The use of ^{208}Pb as a coolant helps to weaken the unfavorable contributions of the last two components into CTRC. Indeed, smaller values of ^{208}Pb capture and inelastic scattering cross-sections decrease respective CTRC components while the favorable component, associated with neutron leakage, ensures negative coolant temperature feedback (Figure 3, fast reactor, content of minor actinides in fuel—30%).

To find out if the theoretical prerequisites on potential advantages of ^{208}Pb as a neutron moderator, coolant, and reflector are well grounded, the lifetime of prompt neutrons was calculated in the fast reactor core (simplified model).

Neutron-physical calculations have been performed using the computer code TIME26 [5], where one-dimensional model of fast reactor in 26-group diffusion approximation is considered. Evaluated nuclear data file BNAB-78 was used, which was processed by auxiliary program ARAMAKO-C1 (preparation of self-shielded microstants for every reactor zone) [6].

One-dimensional axial model of central region in BREST-300 reactor was analyzed [7]. Main parameters for square elementary cell of fuel rods are presented in Table 3.

The initial goal was to determine such neutron-physical parameters of one-dimensional axial model for central

Table 3: Main parameters of the calculation model.

Parameter	Value
Pitch of fuel lattice (mm)	13.6
Diameter of fuel meat (mm)	7.7
Thickness of contact layer (mm)	0.2
Thickness of cladding (mm)	0.5
Diameter of fuel rod (mm)	9.1
Fuel	(U, Pu)N; $\gamma = 14.32$ g/cm^3
Uranium fraction	Natural uranium Composition: ^{235}U—0.007, ^{238}U—0.993
Plutonium fraction	Reactor-grade plutonium ^{239}Pu-60%; ^{240}Pu—25%; ^{241}Pu—11%; ^{242}Pu—4%
Plutonium fraction content (%)	13.84
Core breeding ratio (CBR)	1.038
Contact layer and coolant	Lead; $\gamma = 10.47$ g/cm^3
Cladding	Stainless steel EP-823; $\gamma = 8$ g/cm^3 Composition: 85% Fe, 12% Cr, 3% Si
Core height (cm)	110

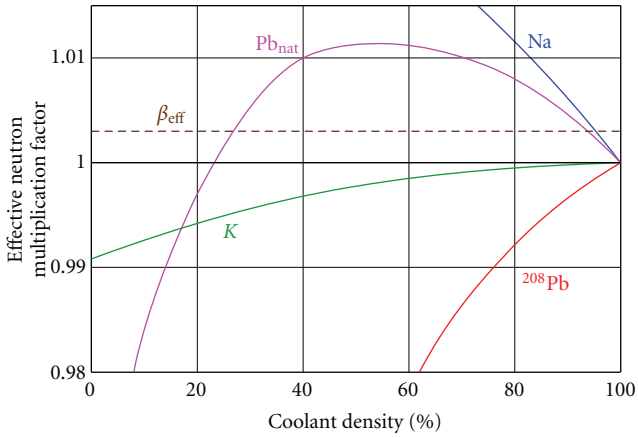

Figure 3: Comparison of coolants: dependence of K_{eff} on coolant density.

Table 4: Replacement of natural lead by ^{208}Pb: its influence on the reactor parameters.

Parameter	Natural lead	^{208}Pb
Core height (cm)	110	298
Pitch of fuel lattice (mm)	13.6	23.6
Content of Pu-fraction (%)	13.84	13.58

region of BREST-300 core cooled by natural lead which are equivalent to the parameters of two-dimensional model for the same region. The effects of the replacement of natural lead by ^{208}Pb were analyzed in subsequent calculations. As a result, the ^{208}Pb-cooled core was characterized by the same values of K_{eff}, core breeding ratio (CBR), and CTRC as the natural lead-cooled core but the values of the core height, pitch of fuel lattice, and content of Pu-fraction were properly changed (Table 4).

If natural lead is replaced by ^{208}Pb, then K_{eff} increases by 7%; 80% of this effect is caused by change of neutron spectrum while the other 20% by smaller neutron absorption. This circumstance facilitated a significant increase of fuel lattice pitch (from 13.6 mm to 23.6 mm).

Prompt neutron lifetime l_{prompt} in BREST-300 reactor core is about 0.5 μs [8]. Replacement of natural lead by ^{208}Pb, that is, the use of ^{208}Pb as a coolant, neutron reflector, and moderator, extended prompt neutron lifetime to 1.35 μs, that is, by almost 3 times longer. This became possible due to wider fuel lattice and better reflecting properties of ^{208}Pb. If thickness of ^{208}Pb reflector is extended from 0.5 m to 5 m together with addition of 2-meter graphite reflector, then prompt neutron lifetime increases to 1 ms (i.e., by 2000 times!) and becomes comparable with typical prompt neutron lifetimes in thermal CANDU-type reactors.

Such a drastical extension of prompt neutron lifetime is caused by the following effect. Fast neutrons from the reactor core penetrated deeply into ^{208}Pb reflector, multiple neutron-nucleus collisions slowed down these neutrons, and they came back to the reactor core after an essential time delay (due to small absorption and effective albedo of ^{208}Pb). Since these returning neutrons, in the terms of their origin, are prompt neutrons, it can be spoken about the slowed progression of chain fission reaction on prompt neutrons. Let us consider the influence of this circumstance on the reactor safety parameters.

We consider the reactor kinetics for the stepwise insertion of positive reactivity that exceeds fraction of delayed neutrons and no feedbacks are taken into account. Time evolution of neutron density can be described by the following equation where two summands define contributions of prompt and delayed neutrons, respectively,

$$\frac{n(t)}{n(0)} = \frac{\rho}{\rho - \beta} \cdot \exp\left[+\frac{t}{T}\right] - \sum_{i=1}^{6} \frac{\beta_i}{\rho - \beta} \cdot \exp\left[-\frac{\lambda_i \rho}{\rho - \beta} \cdot t\right]. \quad (1)$$

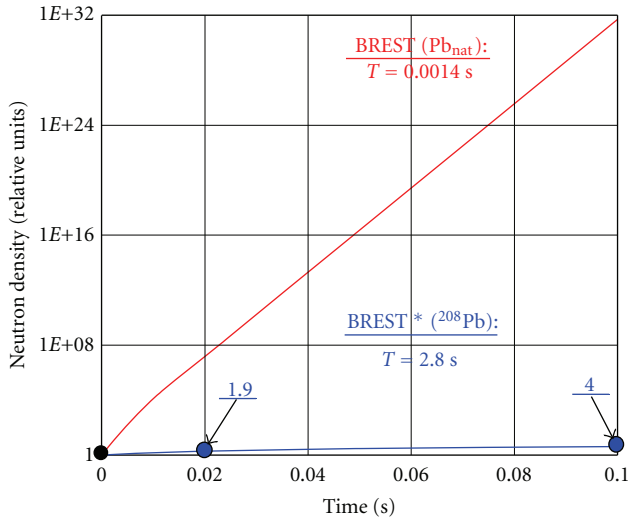

FIGURE 4: Reactor runaway without accounting for feedbacks induced by the stepwise insertion of positive reactivity $\rho/\beta = 1.1$ dollar.

$n(t)$—neutron density at time moment t; ρ—reactivity; β—fraction of delayed neutrons; β_i—fraction of delayed neutrons in the ith group; T—reactor period; $T \equiv l_{\text{PROMPT}}/(\rho - \beta)$; λ_i—decay constant of nuclei-emitters of delayed neutrons in the ith group.

If transient time is relatively short, then the second summand (delayed neutrons) may be neglected. The feedbacks to work, the reactor period T should be comparable with thermal inertia parameter of fuel rod, which is within the time range from 0.1 s for metal fuel to 3 s for oxide fuel [4]. This means that permissible stepwise insertion of positive reactivity could exceed delayed neutron fraction by no more than ~0.001 dollar for BREST-300 reactor core cooled by natural lead ($l_{\text{prompt}} \approx 0.5\,\mu$s) and up to several dollars if ^{208}Pb is used thanks to a much longer prompt neutron lifetime ($l_{\text{prompt}} \approx 1$ ms).

Let us consider the case when the stepwise insertion of positive reactivity $\rho/\beta = 1.1$ dollar. Under these conditions the reactor runaway without feedback effects is presented in Figure 4 for two cases: natural lead or ^{208}Pb is used as coolant and neutron reflector.

Neutron density upgrades by 32 orders of magnitude just in 0.1 s in the first case (BREST project), that is, this is actually an explosion, and only by 4 times in the second case (the use of ^{208}Pb as a coolant, neutron reflector, and moderator). So, the use of ^{208}Pb as a coolant, neutron reflector and moderator in fast reactors could drastically slow down progression of chain fission reaction on prompt neutrons and thus essentially improve the reactor safety.

4. Enhanced Proliferation Resistance of Pu-Containing Fuel

It was mentioned previously that, if lead or lead-bismuth coolant contains high fraction of ^{208}Pb, then unique properties of ^{208}Pb can allow us to reduce significantly unfavorable

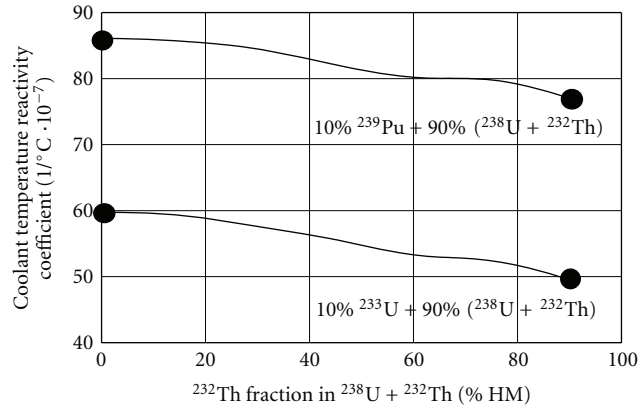

FIGURE 5: Dependences of spectral CTRC component on ^{232}Th fraction in fertile component of fuel.

spectral component of CTRC. Negative effect of such unfavorable CTRC component may be compensated by another CTRC component related with neutron leakage. Besides, the problem of plutonium protection against unauthorized proliferation arises here, like for any nuclear reactor loaded with plutonium-based fuel [9]. Here we consider the ways towards resolving these problems by using (^{238}U-Pu-Th-^{233}U)-mixture as a fuel and radiogenic lead as a coolant.

Spectral component of CTRC was calculated for an elementary fuel cell of fast, natural lead-cooled reactor BREST-300 [10] with application of the computer code SCALE-4.3 [11] and 44-group cross-section library generated from the evaluated nuclear data file ENDF/B-V. Since numerical analysis of elementary fuel cell does not take neutron leakage effects into consideration, these computations are able to determine just only spectral component of CTRC, main goal of the investigation. The computations were carried out for U-Pu, U-Th, and U-Pu-Th nitride fuel. Dependencies of CTRC on fraction of ^{232}Th in fertile component (mixture of ^{232}Th with ^{238}U while fissile component consisted of ^{233}U or ^{239}Pu) of fuel are shown in Figure 5. The following conclusions may be derived from these curves.

(i) Firstly, replacement of fertile isotope, that is, substitution of ^{232}Th for ^{238}U, resulted in remarkable reduction of unfavorable CTRC component (on 11% for ^{239}Pu and on 17% for ^{233}U).

(ii) Secondly, replacement of fissile isotope, that is, substitution of ^{233}U for ^{239}Pu, also resulted in remarkable reduction of unfavorable CTRC component (on 31% for ^{238}U and on 36% for ^{232}Th).

(iii) In total, transition from (^{238}U-Pu)-fuel to (Th-^{233}U)-fuel resulted in reduction of unfavorable CTRC component on 43%. These effects are caused by specific peculiarities in neutron-physical properties of involved fertile and fissile isotopes.

Some neutron-multiplying properties (the number of fission reactions per one neutron absorption as a function of neutron energy) of ^{238}U, ^{239}Pu, ^{232}Th, ^{233}U [1], and

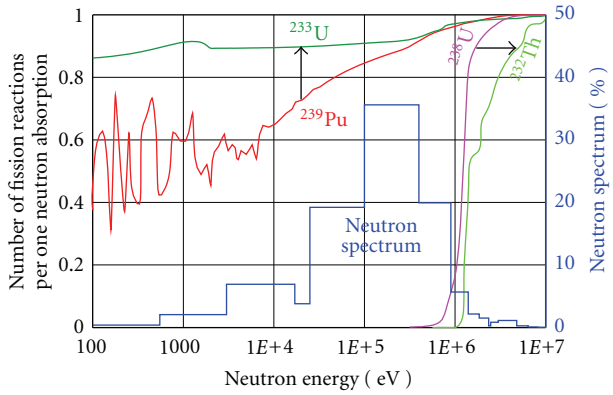

Figure 6: Neutron-multiplying properties of ^{238}U, ^{239}Pu, ^{232}Th, ^{233}U, and neutron spectrum in lead-cooled fast reactor BREST-300.

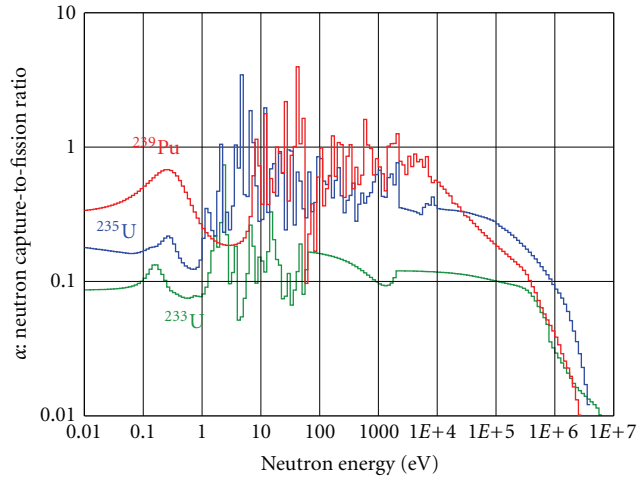

Figure 7: Ratios of capture-to-fission cross-sections for ^{239}Pu, ^{233}U, and ^{235}U.

neutron spectrum in lead-cooled fast reactor BREST-300 are presented in Figure 6.

The higher lead temperature results in the lower lead density. This negative density effect reduces neutron absorption and slowing-down rate, thus, shifting neutron spectrum towards higher-energy range. Under such a spectral shift, neutron-multiplying properties of ^{239}Pu become better to a relatively larger degree than those of ^{233}U. In addition, energy threshold for neutron-induced fission reactions of ^{238}U is lower than that of ^{232}Th. As computations of BREST-300 neutron spectrum have demonstrated, fraction of neutrons in the energy range close to fission thresholds of ^{238}U and ^{232}Th (about 1 MeV) were relatively small. Just that is why replacement of fertile isotopes in fuel has changed spectral component of CTRC to a relatively lower degree than replacement of fissile isotopes [4]. So, (Th-^{233}U)-fuel can provide more favorable spectral component of CTRC than (^{238}U-Pu)-fuel. Moreover, the favorable margin of CTRC may be used for giving back to fast reactors their initial function of nuclear fuel breeders by restoring the fertile blankets around of the reactor core.

So, mixed (^{238}U-Pu-Th-^{233}U)-fuel can decrease unfavorable spectral component of CTRC as compared with (^{238}U-Pu)-fuel. In addition, the ternary mixed fuel enhances protective barriers against unauthorized diversion of fissile materials [12] because fissionable isotope ^{233}U is diluted by fertile isotope ^{238}U, and fraction of plutonium component in mixed fuel is lower than in (^{238}U-Pu)-fuel. The latter circumstance is related with two following reasons. Firstly, mixed (^{238}U-Pu-Th-^{233}U)-fuel maintains the reactor criticality basically due to the presence of fissionable isotope ^{233}U and its build-up from thorium. Secondly, plutonium breeding rate is suppressed by the reduced content of fertile isotope ^{238}U.

Moreover, plutonium proliferation protection may be enhanced by creating favorable conditions for intense build-up of even plutonium isotopes. These conditions may be formed just by using radiogenic lead as a coolant and graphite as a structural material and neutron moderator. Both these materials, radiogenic lead and graphite, are extremely weak neutron absorbers. As is known [1], neutron-multiplying properties of ^{239}Pu in softened neutron spectrum become substantially worse than in hardened neutron spectrum. At the same time, neutron-multiplying properties of ^{233}U remain practically unchanged under transition from hard to soft neutron spectrum. Such a behavior of neutron-multiplying properties can be illustrated by the curves presented in Figure 7, namely, by energy dependencies of capture-to-fission ratios for appropriate cross-sections of ^{239}Pu, ^{233}U, and ^{235}U (only for comparison). So, the use of radiogenic lead as a coolant in softened neutron spectrum can protect plutonium against unauthorized proliferation at the isotopic level while keeping constant the breeding rate of secondary fuel. The main reason consists in a good neutron economy because of extremely weak neutron absorption by radiogenic lead. It should be noted that the isotopic level of proliferation protection remains still insurmountable barrier for those countries, which have not mastered yet the isotope separation technologies.

A possibility to create low-activated lead coolant by its isotopic tailoring was studied also in [13]. The investigation has revealed that, in neutron spectrum of fast reactor, radiative capture of neutrons by ^{208}Pb is a main channel for build-up of radioisotopes in lead coolant. So, basic and residual activity of lead coolant may be reduced on several orders of magnitude by preliminary lead enriching with ^{206}Pb. Numerical evaluations have demonstrated that, after 30-year usage period of lead as a coolant of fast reactor, lead coolant consisting mainly of ^{206}Pb may be withdrawn from the radiation control in a 1 ÷ 2-year cooling time while natural lead coolant only in a 100-year cooling time.

It should be noted here that softening of neutron spectrum makes it possible not only to provide plutonium proliferation protection at the isotopic level but also to reduce radically neutron-induced activation of radiogenic lead with high content of ^{208}Pb. Low activation rate of radiogenic lead is explained by the fact that micro-cross-sections of neutron radiative capture by ^{208}Pb, that is, main

Table 5: Transition from (U,Pu)N-fuel to $(U,Pu)O_2$-fuel.

Parameter	Fuel	Natural lead	^{208}Pb
Content of Pu-fraction (%)	(U,Pu)N	13.84	13.58
	$(U,Pu)O_2$	14.81	14.45
Pitch of fuel lattice (mm)	(U,Pu)N	13.6	23.6
	$(U,Pu)O_2$	12.2	20.4
Height of the reactor core (cm)	(U,Pu)N	110	298
	$(U,Pu)O_2$	125	314
Coolant velocity (relative units)	(U,Pu)N	1	0.59
	$(U,Pu)O_2$	1.60	0.87
Number of fuel rods (relative units)	(U,Pu)N	1	0.41
	$(U,Pu)O_2$	0.89	0.39
Pressure drop for coolant flow through the reactor core (relative units)	(U,Pu)N	1	0.18
	$(U,Pu)O_2$	4.02	0.57

channel for neutron-induced activation, are lower by several orders of magnitude in softened neutron spectrum than those in high-energy range of neutron spectrum, that is, in resonance region of neutron absorption (see Figure 2). So, activation rate of radiogenic lead coolant may be reduced without application of sophisticated, dangerous, and expensive isotope separation technologies.

5. Thermal-Hydraulic Advantages of the Reactor Core Cooled by ^{208}Pb

The use of ^{208}Pb as a coolant in the fast reactor core leads to much wider fuel lattice compared to the use of natural lead thanks to favorable neutron-physical properties of ^{208}Pb. This opens up a possibility to improve essentially the thermal-hydraulic characteristics of the fast reactor core.

One else important issue is connected to the influence of a wider fuel lattice and higher core (see Table 4) on a pressure drop for coolant flow through the reactor core. Evidently, it is easier to create a regime for natural circulation of coolant in the case of smaller pressure drop needed for coolant flow through the reactor core.

Let us assume that, upon replacement of natural lead by ^{208}Pb, the coolant temperature parameters and the core thermal power remained the same.

The calculations showed (see Table 5) that application of ^{208}Pb as a coolant in the fast reactor core loaded with mixed uranium-plutonium nitride fuel allows us to achieve a noticeable gain in the reactor parameters. It was transpired that replacement of natural lead by ^{208}Pb while retaining the same values for K_{eff}, CBR, and TRC by introducing proper changes into content of Pu-fraction, pitch of fuel lattice, and height of the reactor core made it possible to achieve the following effects:

(i) the same coolant heating up with a lower coolant velocity (about 2 times lower);

(ii) an essential reduction of pressure drop for coolant flow through the reactor core (5 times lower);

(iii) the same thermal power with a smaller number of longer fuel rods (2.5 times smaller).

6. Replacement of Nitride by Oxide Uranium-Plutonium Fuel

BREST-300 reactor cooled by natural lead, as proposed by the developers [8], is characterized by improved natural safety and small reactivity change during the reactor lifetime (within delayed neutron fraction). This is achieved mainly through the application of high-density uranium-plutonium nitride fuel.

Since oxide fuel is widely used in nuclear power reactors, the question arises: could ^{208}Pb allow us to come back from nitride fuel to the commonly used and industrially assimilated oxide fuel? Evidently, the transition from high-density (U,Pu)N-fuel to low-density $(U,Pu)O_2$-fuel can reduce, to a certain extent, the advantages gained by replacement of natural lead by ^{208}Pb.

To answer the question, numerical evaluations were carried out in which the previous approach was applied to definition of the model parameters, that is, the models are made equivalent on the values of K_{eff}, CBR, and CTRC by introducing the proper changes into content of Pu-fraction, pitch of fuel lattice and height of the reactor core. Table 5 shows how content of Pu-fraction, pitch of fuel lattice, and height of the reactor core had to be changed so that the replacement of (U,Pu)N-fuel by $(U,Pu)O_2$-fuel would not change the values of K_{eff}, CBR, and CTRC.

With ^{208}Pb application, replacement of nitride fuel by oxide fuel increased content of Pu-fraction, increased height of the reactor core and decreased pitch of fuel lattice. Nevertheless, fuel lattice is still much wider compared to that if natural lead is used.

As is mentioned above, replacement of natural lead by ^{208}Pb results in a remarkable decrease (almost in 2 times) of coolant velocity mainly thanks to the wider fuel lattice. Replacement of nitride fuel by oxide fuel when natural lead is used as a coolant requires increasing the coolant velocity (by 60%) because of tighter fuel lattice and larger height of the reactor core. Finally, however, the losses caused by transition from nitride fuel to oxide fuel are not so large, and they cannot nullify the gains from ^{208}Pb application instead of natural lead. Transition from (U,Pu)N-fuel to $(U,Pu)O_2$-fuel accompanied by replacement of natural lead by ^{208}Pb

Table 6: Influence of axial blanket thickness on reactor parameters.

Parameter	Thickness of axial blanket (cm)				
	0	10	20	40	60
Pu-fraction part (%)	13.58	13.51	13.46	13.39	13.35
Pitch of fuel lattice (mm)	23.6	22.5	21.9	21.4	21.3
Height of the reactor core (cm)	298	270	260	256	255
CBR	1.038	1.038	1.038	1.038	1.038
BBR	0.000	0.042	0.080	0.124	0.142

Table 7: Thermal-hydraulic parameters of models with axial blanket.

Parameter	Thickness of axial blanket (cm)				
	0	10	20	40	60
Coolant velocity (relative units)	1	1.02	1.05	1.08	1.10
The number of fuel rods (relative units)	1	1.10	1.15	1.16	1.17
Pressure drop required for coolant flow through fuel assembly (relative units)	1	1.11	1.28	1.61	1.78

nevertheless results in a desirable effect, that is, coolant velocity could be decreased by 13%.

A substantial decrease in the number of fuel rods by switching from natural lead to ^{208}Pb due to considerably larger height of the reactor core is an important effect. The 2.5 times fewer longer fuel rods are required to obtain the same thermal power.

Transition from nitride fuel to oxide fuel with natural lead as a coolant significantly increases (by 3 times) the pressure drop required for coolant flow through the reactor core. But the gain obtained by replacing natural lead by ^{208}Pb is so large that, finally, application of oxide fuel leads to a remarkable (almost in 2 times) decrease of the pressure drop required for coolant flow through the reactor core.

7. Possibility of Fuel Breeding in Axial Blankets

Application of blankets around the reactor core is not envisaged in BREST-300 project. So, this reactor is not regarded as a fuel breeder [7]. More favorable negative feedbacks, when using ^{208}Pb as a coolant, allow recovery of the axial and radial blankets in order to return fuel breeding property to lead-cooled fast reactors.

In this section the possibility of using an axial blanket in the examined model of BREST-300 reactor is evaluated, when natural lead is replaced by ^{208}Pb. Geometrical model of the reactor included a core loaded with mixed uranium-plutonium nitride fuel, an axial blanket loaded with natural uranium nitride as a fertile material, and a layer of lead after the blanket. The results reported above, obtained for a core cooled by ^{208}Pb, are the input data for the calculations.

The use of an axial blanket containing natural uranium nitride can essentially change the model parameters. Therefore the task involved the following: first, to find a variant of the model with the axial blanket which would be equivalent to the initial model on the values of K_{eff}, CBR, and CTRC by varying content of Pu-fraction, height of the reactor core, and pitch of fuel lattice; second, to evaluate blanket breeding ratio (BBR) depending on its thickness. Results of the calculations are presented in Table 6.

It can be seen that the increase of axial blanket thickness leads to a gradual increase in BBR, reaching a stable value at about 0.14. At the same time, content of Pu-fraction decreases, fuel lattice becomes tighter, and height of the reactor core decreases. Using the simple physical correlations, one can evaluate the influence of axial blanket on coolant velocity and the number of fuel rods when coolant heating up and thermal power are kept at the same level. The use of an axial blanket exerts an influence on the value of pressure drop required for coolant flow through the reactor core, axial blanket, and gas cavity. Results of the evaluations are presented in Table 7. It is assumed that the contribution of the axial blanket to coolant heating up is negligible.

One can see that the appearance of the axial blanket slightly worsened some reactor parameters in comparison with those in initial variant: coolant velocity and the number of fuel rods, when coolant heating up and thermal power are kept at the same level, somewhat increased. This effect is mainly caused by a tighter fuel lattice. The pressure drop for coolant flow through fuel assembly changed the most (almost twofold increase in relative units). The reason is a combined effect from longer fuel rods (core plus axial blanket plus cavity for accumulation of gaseous fission products) and from tighter fuel lattice.

Thus, the use of an axial blanket allowed to increase the total breeding ratio (by 0.10–0.14), but at the cost of a certain deterioration of some other parameters. It was necessary to increase slightly coolant velocity, the number of fuel rods and, above all, to increase significantly the pressure drop required for coolant flow through the reactor core, axial blanket, and gas cavity, which could weaken a role of natural circulation under emergency conditions.

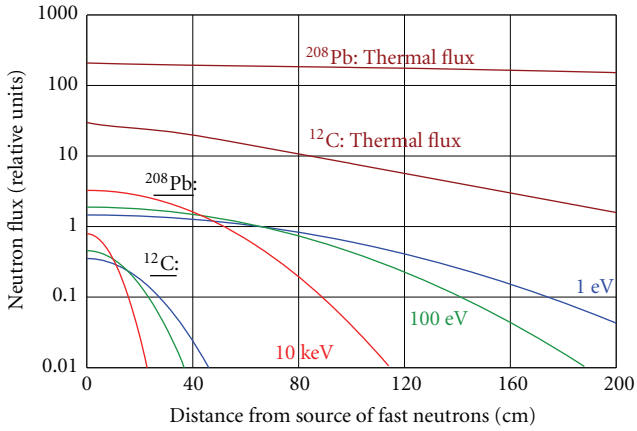

Figure 8: Fluxes of thermal and slowing-down neutrons in ^{208}Pb and graphite depending on distance from ADS target.

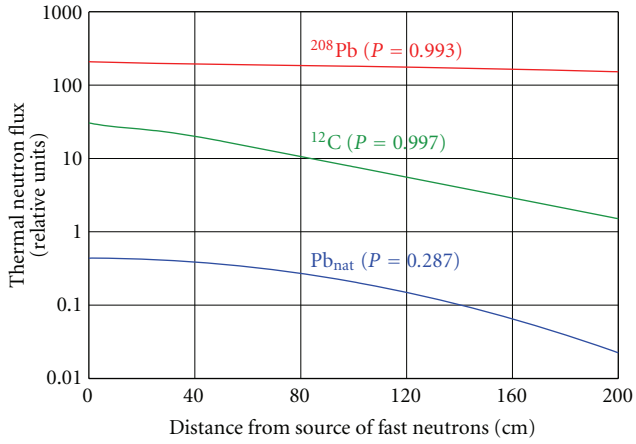

Figure 9: Thermal neutron flux in ^{208}Pb, graphite, and natural lead depending on distance from the target.

8. High Neutron Flux in an ADS Blanket

Extremely small capture cross-section and small average logarithmic energy loss open up a possibility to obtain high neutron flux in large volumes of an ADS blanket.

To find out if theoretical preconditions on advantages of ^{208}Pb application correspond to the facts calculation research was conducted of the space-energy distributions of neutron flux in an ADS blanket consisting of different materials: oxide of beryllium, graphite, isotopes of lead, natural lead, and bismuth.

Light and heavy water were not studied since their use in the vicinity of a liquid-metal target in high-temperature ADS is quite questionable. The blanket was modeled by an infinite homogeneous nonmultiplying medium with flat source of fast neutrons at the beginning of the coordinates.

Results of the calculations showed that neutron fluxes reached their maximum values in ^{208}Pb and graphite blankets. So, neutron flux distributions are comparable in these media. One can see from Figure 8 that near the target the fluxes of both thermal and slowing-down neutrons in ^{208}Pb is several times higher than those in graphite, and advantage of ^{208}Pb is strengthened very rapidly when moving off the target.

Probabilities for neutrons to avoid capture in the slowing-down process to thermal energy in ^{208}Pb and graphite are close to each other and approach unity, being 0.993 and 0.997, accordingly, and significantly less unity in natural lead (0.287). It is explained by the fact that in the slowing-down process in ^{208}Pb and graphite the neutrons are being scattered and slowed down with much more probability than they are captured. The opposite case occurs in natural lead. Note that the average logarithmic energy loss and capture cross-section of ^{208}Pb is approximately 17 times less, while the scattering cross-section is 2 times greater than that for graphite. This means that, on average, the neutron slowing-down process in ^{208}Pb must have 17 times more elastic collisions than in graphite. However, at each elastic neutron-^{208}Pb collision the probability of neutron scattering and slowing-down is 34 times higher than the probability of neutron capture in comparison with graphite. As a result, it appears that higher thermal neutron flux can be created in ^{208}Pb than that in graphite.

Indeed, near the target the slowing-down neutron fluxes in ^{208}Pb are higher than those in graphite, and, when moving off the target, the fluxes decrease slower than in graphite because ^{208}Pb has a greater atomic mass and, accordingly, lower average energy loss, which defines both the amplitude of slowing-down neutron fluxes near the target and their shape at a distance from the target. Near the target, thermal neutron flux in ^{208}Pb is higher than in graphite since ^{208}Pb has a larger value of scattering-to-capture cross-section ratio. When moving off the target, thermal neutron flux in ^{208}Pb decreases significantly more slowly than in graphite because diffusion length in ^{208}Pb is considerably longer than in graphite. Note that thermal neutron flux in natural lead is almost 3 orders of magnitude less than that in ^{208}Pb and decreases when moving off the target essentially quicker than in ^{208}Pb (Figure 9).

Thus, it can be supposed that ^{208}Pb is a number-one candidate for the role of neutron moderator for creating a high-flux ADS blanket in both resonance and thermal energy spectra. At the same time, a blanket with ^{208}Pb can have a sufficiently large volume to place necessary quantity of the materials to be transmuted and solve the problem of neutron leakage.

9. Transmutation in Resonance Region of Neutron Energy

High-energy neutrons leaving the target are slowing down as a result of elastic and inelastic scattering on nuclides of the medium. Inelastic scattering has a threshold nature and stops acting, below certain rather large value of neutron energy. Elastic scattering acts at any values of neutron energy.

Mean part of energy which neutrons lose as a result of elastic scattering is defined by atomic mass of medium. In mediums of heavy atomic masses, for example, in ^{208}Pb, at each elastic scattering the neutrons lose only a small part of

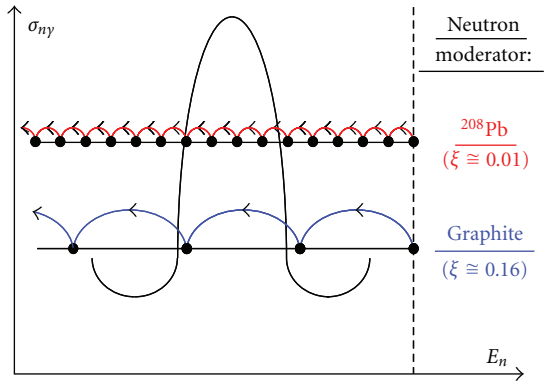

Figure 10: Passage of neutrons through resonance in heavy and light mediums.

their energy and in light medium, for example, in graphite, they lose a significantly larger part of their energy. As a result, the same resonance of transmutation cross-section (capture for long-lived fission products (LLFP) or fission for minor actinides (MA)) can be either "wide" for slowing-down neutrons if they have multiple scattering in the energy region of this resonance or "narrow" if they are going through the resonance region with almost no scattering [9] (Figure 10).

In the first instance the neutrons remain in the resonance region of cross-section for a comparatively long time and so the probability of their capture with subsequent transmutation of LLFP and MA increases. The opposite situation is observed in the second case. Thus use of a very heavy neutron moderator, radiogenic lead, for example, consisting mainly of ^{208}Pb, can result in a radical increase of neutron capture in the resonance region of energy. This could be attractive for transmutation of LLFP and MA in the resonance neutron spectrum.

10. High-Flux ADS with Transmutation Zone

The principal scheme of high-flux ADS with cryogenic zone for transmutation of radioactive wastes (RAWs) is shown in Figure 11. According to the scheme, proton beam bombards lead target, thus initiating spallation reactions and generating high-energy neutrons which go into ADS blanket and gradually slow down.

A lead-bismuth wall separates the lead target from the ADS blanket. The wall may be made as a system of Field tubes filled up with molten lead-bismuth eutectics at 250°C (Figure 12). The surrounding lead has a solid form and creates a frozen wall. When the wall gains a significant damage dose induced by high-energy spallation neutrons coming from the target, it is sufficient to warm lead-bismuth eutectics up to 400°C and the frozen wall melts. If lead-bismuth eutectics is cooled back to the previous 250°C, the wall freezes again but with zero damage dose.

When maximum permissible damage dose is reached in Field tubes, they may be withdrawn. Thus a problem of replacing a single hermetic wall is simplified and transformed into a problem of its replacement in parts (with sections of Field tubes). Note that a target and a blanket zone surrounding it are made of identical material (lead). Therefore, when ADS is in operation, the liquid target and the solid blanket can be separated without the wall by supporting the proper temperature regime of blanket cooling. Before starting ADS, the target can be melted by filling the Field tubes with lead-bismuth eutectics at the temperature higher than the melting-point of lead. So, when ADS is in operation, no metal structure is near the target and all procedures on replacing channels for transmutation and sections of Field tubes may be conducted from above the target and blanket.

11. Natural Resources of Radiogenic Lead

Lead isotopes ^{208}Pb, ^{206}Pb, and ^{207}Pb are the final products of the radioactive decay chains starting from ^{232}Th, ^{238}U, and ^{235}U, respectively,

$$
\begin{aligned}
&^{232}\mathrm{Th} \xrightarrow{\alpha \;\cdots\; 6\cdot\alpha \;\cdots\; +\;\cdots\; 4\cdot\beta} {}^{208}\mathrm{Pb} \quad \text{(14.6 billion years)}\\
&^{235}\mathrm{U} \xrightarrow{\alpha \;\cdots\; 7\cdot\alpha \;\cdots\; +\;\cdots\; 4\cdot\beta} {}^{207}\mathrm{Pb} \quad \text{(0.7 billion years)}\\
&^{238}\mathrm{U} \xrightarrow{\alpha \;\cdots\; 8\cdot\alpha \;\cdots\; +\;\cdots\; 6\cdot\beta} {}^{206}\mathrm{Pb} \quad \text{(4.6 billion years)}
\end{aligned}
\tag{2}
$$

Therefore radiogenic lead with large abundance of ^{208}Pb could be extracted from natural thorium and thorium-uranium ores [14–18] without any isotope separation procedures.

The relative contents of lead isotopes in radiogenic lead depend on the ore age and on the content of natural lead as an impurity. The contents of ^{208}Pb and ^{206}Pb in natural lead are 52.4% and 24.1%, respectively. It should be noted that neutron capture cross-sections of ^{206}Pb, although larger than those of ^{208}Pb, are significantly smaller than those of ^{207}Pb and ^{204}Pb. So, at the first glance, it appears that the ores containing about 93% ^{208}Pb and 6% ^{206}Pb (Table 8) could provide the necessary composition of radiogenic lead. However, the first estimations showed that the content of only 1% ^{204}Pb and ^{207}Pb (these isotopes have high values of capture cross-sections) in radiogenic lead could significantly weaken the advantages of radiogenic lead.

So, radiogenic lead can be taken as a byproduct from the process of uranium and thorium ores mining. Till now, extraction of uranium or thorium from minerals had been followed by throwing radiogenic lead into tail repositories. If further studies will reveal the perspective for application of radiogenic lead in nuclear power industry, then a necessity arises to arrange by-extraction of radiogenic lead from thorium and uranium deposits or tails. Evidently, the scope of the ores mining and processing is defined by the demands for uranium and thorium. However, the demands of nuclear power industry for thorium are quite low now and remain

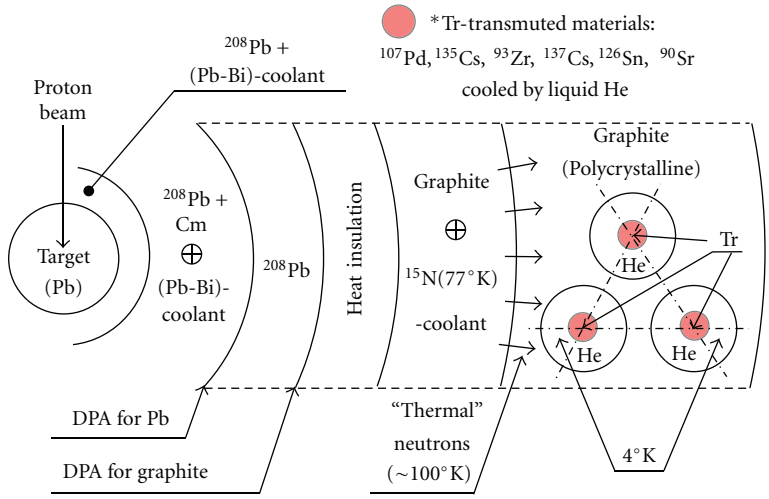

Figure 11: The principal scheme of high-flux ADS with zone for RAW transmutation.

Table 8: Main deposits of uranium, thorium, and mixed uranium-thorium ores. Elemental compositions of minerals and isotope compositions of radiogenic lead.

Deposit	U/Th/Pb (wt.%)	$^{204}Pb/^{206}Pb/^{207}Pb/^{208}Pb$ (at.%)	Age, 10^9 years
Monazite (Guarapari, Brazil)	1.3/59.3/1.5	0.005/6.03/0.46/93.5	0.52–0.55
Monazite (Manitoba, Canada)	0.3/15.6/1.5	0.010/10.2/1.86/87.9	1.83–3.18
Monazite (Mt. Isa Mine, Australia)	0.0/5.73/0.3	0.038/5.44/0.97/93.6	1.00–1.19
Monazite (Las Vegas, USA)	0.1/9.39/0.4	0.025/9.07/1.13/89.8	0.77–1.73
Uraninite (Singar Mine, India)	64.3/8.1/8.9	—/89.4/6.44/4.18	0.885
Monazite (South Bug, Ukraine)	0.2/8.72/0.9	0.010/6.04/0.94/93.0	1.8–2.0

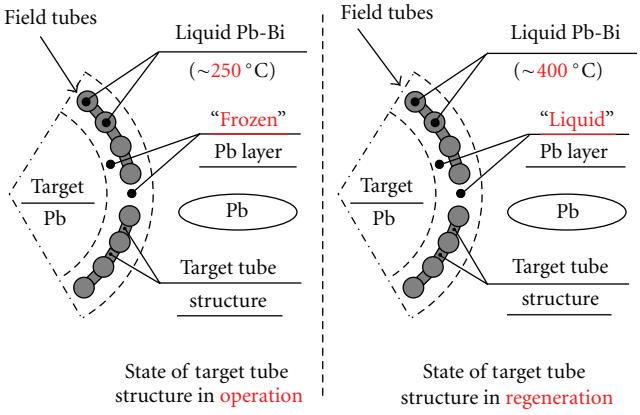

Figure 12: The structure of target tube for high-flux ADS.

so in the nearest future. However, there is one important factor which can produce a substantial effect on the scope of thorium and mixed thorium-uranium ores mining. In the majority of cases, uranium and thorium ores belong to the complex-ore category, that is, they contain minor amounts of many valuable metals (rare-earth elements, gold, and so on). The paper [19] has demonstrated that the presence of useful accompanying elements (some elements of cerium group, in particular) in uranium and thorium ores might be a factor of high significance for making cheaper the process of natural uranium and thorium production. By-extraction of some valuable elements from uranium ores can drop down the lower limit (industrial minimum) of uranium content in ores to 0.01–0.03% under application of the existing technologies for natural uranium extraction.

If necessary, radiogenic lead could be extracted from the available tail repositories or as a byproduct of the processes applied for extraction of the accompanying valuable metals from uranium and thorium ores.

12. Conclusions

The use of radiogenic lead as a coolant of power nuclear reactors makes it possible to improve their nuclear safety, enhance proliferation resistance of fissionable materials, intensify nuclear fuel breeding rate, and improve some thermal-hydraulics parameters. In addition to safe, secure, and effective energy production, radiogenic lead-cooled fast breeder reactors and accelerator-driven systems could be used for effective neutron transmutation of radioactive wastes with extremely low cross-sections of neutron absorption. If necessary, radiogenic lead could be extracted either from the available tail repositories or from the production technologies of the accompanying valuable metals as a byproduct.

Finally, the use of radiogenic lead in nuclear power opens up new opportunities for upgrading the neutron-physical properties of nuclear power reactors and for widening the areas of their applicability.

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