Radiogenic lead as coolant, reflector and moderator in advanced fast reactors

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Abstract. Main purpose of the study is assessing reasonability for recovery, production and application of radiogenic lead as a coolant, neutron moderator and neutron reflector in advanced fast reactors. When performing the study, thermal, physical and neutron-physical properties of natural and radiogenic lead were analyzed. The following results were obtained:

1. Radiogenic lead with high content of isotope $^{208}$Pb can be extracted from thorium or mixed thorium-uranium ores because $^{208}$Pb is a final product of $^{232}$Th natural decay chain.

2. The use of radiogenic lead with high $^{208}$Pb content in advanced fast reactors and accelerator-driven systems (ADS) makes it possible to improve significantly their neutron-physical and thermal-hydraulic parameters.

3. The use of radiogenic lead with high $^{208}$Pb content in advanced fast reactors as a coolant opens the possibilities for more intense fuel breeding and for application of well-known oxide fuel instead of the promising but not tested enough nitride fuel under the same safety parameters.

4. The use of radiogenic lead with high $^{208}$Pb content in ADS as a coolant can upgrade substantially the level of neutron flux in the ADS blanket, which enables effective transmutation of radioactive wastes with low cross-sections of radiative neutron capture.

1. Introduction. Thorium, uranium and radiogenic lead

As it was written in the IAEA Red Book-2014 [1], the evaluated global thorium resources were 6.4 million tons in total. The largest thorium resources are placed in the following countries: India – 0.85 million tons, Brazil – 0.63 million tons, Australia and the USA – about 0.6 million tons each.

Demands of the world industry for thorium are very small, mainly because of its radioactivity. According to information from the US Mining Bureau [2], in 2009 only about 5000 tons of monazite ores were mined in India, 1200 tons in Brazil and 600 tons in Malaysia. In total, only about 1200 kg of thorium were consumed by the world industry in 2007.

The monazite sands constitute the most important source of rare-earth elements and thorium. General chemical formula for the monazites is usually written as (Ce, Th)PO$_4$ though the monazites contain, in addition to cerium, lanthanum, praseodymium, neodymium and some other rare-earth metals, as well as uranium and radiogenic lead in addition to thorium. As a rule, thorium content in the monazites covers the range from 2.5% to 12%, uranium content – from 0.1% to 0.4% content of rare-earth metals – from 55% to 68% and relatively low percentage of radiogenic lead.

The radiogenic lead in the monazites is a final material produced by consecutive radioactive decays of main thorium and uranium isotopes. As is known, $^{232}$Th is the start isotope for the chain of isotopic
transformations caused by $\alpha$- and $\beta$-decays of intermediate chain members. Totally, decay chain of $^{232}\text{Th}$ numbers twelve members, products of seven $\alpha$-decays and five $\beta$-decays. The decay chain ends with stable lead isotope $^{206}\text{Pb}$.

Similarly, uranium isotopes in the monazites are the start isotopes for their own decay chains. The decay chain of $^{238}\text{U}$ numbers fourteen members (eight $\alpha$-decays, six $\beta$-decays) and ends with stable lead isotope $^{206}\text{Pb}$. The decay chain of $^{235}\text{U}$ numbers eleven members (seven $\alpha$-decays, four $\beta$-decays) and ends with stable lead isotope $^{207}\text{Pb}$. So, final product in the decay chains of main thorium and uranium isotopes is the radiogenic lead. However, uranium-produced lead significantly differs from thorium-produced lead because of different final isotopes. Indeed, finally $^{232}\text{Th}$ transforms into lead isotope $^{208}\text{Pb}$, $^{238}\text{U}$ and $^{235}\text{U}$ – into other lead isotopes, $^{206}\text{Pb}$ and $^{207}\text{Pb}$, respectively.

That is why the natural minerals can contain, in addition to the “common” lead with a certain isotopic composition, a whole family of the radiogenic leads in uranium, thorium and mixed uranium-thorium ores. Isotopic compositions of the radiogenic leads can cover a rather wide range in dependence on elemental structure and age of the ore deposits. It is evident that the radiogenic leads extracted from mixed uranium-thorium ores with dominant thorium fraction (the monazite sands) are characterized by high content of $^{208}\text{Pb}$.

In the authors’ point of view, just the radiogenic lead obtained as a by-product of the monazite reprocessing plants represents a special interest for advanced projects of various nuclear facilities thanks to some unique neutron-physical properties of stable lead isotope $^{208}\text{Pb}$. The radiogenic lead with high content of this lead isotope is contained in the monazite sands where thorium fraction is substantially larger than uranium fraction.

Currently, the radiogenic lead can be extracted as a by-product of the existing technologies used for recovery of rare-earth metals from mixed deposits of uranium-thorium ores. Some experimental data were published in Ref. 3 on recovery of the radiogenic lead with high content of $^{208}\text{Pb}$ from the reprocessing tails of Brazilian monazites with application of ion-exchange and electrochemical technologies. It was demonstrated that ion-exchange refinement of the thorium reprocessing tails allowed them to extract only 33% of radiogenic lead while the more expensive electrochemical refinement was capable to extract up to 98% of radiogenic lead with content of $^{208}\text{Pb}$ at the level of 88%.

The table 1 presents experimental data about elemental structure of some monazites and about isotopic compositions of the radiogenic leads extracted from these monazite deposits.

| Ore deposits          | U / Th / Pb [weight %] | $^{204}\text{Pb}$ | $^{206}\text{Pb}$ | $^{207}\text{Pb}$ | $^{208}\text{Pb}$ [atomic %] |
|-----------------------|------------------------|-------------------|-------------------|-------------------|-------------------------------|
| Guarapari, Brazil     | 1.3 / 59.3 / 1.5       | 0.005             | 6.03              | 0.46              | 93.5                          |
| Manitoba, Canada      | 0.3 / 15.6 / 1.5       | 0.010             | 10.2              | 1.86              | 87.9                          |
| Mt. Isa Mine, Australia | 0.0 / 5.73 / 0.3     | 0.038             | 5.44              | 0.97              | 93.6                          |
| Las Vegas, USA        | 0.1 / 9.39 / 0.4      | 0.025             | 9.07              | 1.13              | 89.8                          |
| South Bug, Ukraine    | 0.2 / 8.72 / 0.9      | 0.010             | 6.04              | 0.94              | 93.0                          |
| “Common” lead         | ——                    | 1.4               | 24.1              | 22.1              | 52.4                          |

Today Russia possesses thorium ores with evaluated resources of 0.15 million tons of thorium dioxide [1]. About 85 000 tons of thorium-bearing monazite concentrate are stockpiled in Sverdlovsk region. These thorium reserves were partially mined in the Russian Federation and partially imported from Mongolia, China and Vietnam. The monazite concentrate is under reprocessing now only in order to reduce its radiation effects on the environment. If we suppose that Russian monazite concentrate contains 1.5% of the radiogenic lead, like Brazilian monazite sands, then total amount of the radiogenic lead in the stockpiled monazite concentrate is equal to about 1200 tons.

In Russia the rare-earth metals, molybdenum and titanium are extracted from uranium and thorium-bearing ore deposits at Kola Peninsula [4]. Full-scale industrial plants are in operation there for recovery of these useful materials. Besides, several technological lines are in operation there for
recovery and isolation of all radioisotopes to prevent their negative radiation effects on population and the environment. So, only one technological stage should be introduced into the existing technological lines, namely extraction of the last material in uranium and thorium chains of radioactive decays, i.e. extraction of the radiogenic lead. Obviously, the radiogenic lead could be extracted with application of the well-developed chemical technology which is currently in usage for recovery of “common” lead from natural lead-bearing ores.

2. Radiogenic lead in fast reactors
In the 1980s, on initiative and under leadership of Professor Victor Orlov, the works under the project of fast lead-cooled reactor BREST have started [9]. The use of lead as a coolant offers the following evident advantages: high boiling point, weak chemical interactions with air and water, low neutron-induced activation, good neutron-reflecting properties, relatively small neutron absorption and moderation. The radiogenic lead with high $^{208}\text{Pb}$ content is able to strengthen additionally these positive properties. Isotope $^{208}\text{Pb}$ is a double-magic nuclide with closed neutron and proton shells. Thanks to this fact, micro cross-sections of neutron absorption by $^{208}\text{Pb}$ are substantially lower than those of other lead isotopes and a lot of light nuclides.

The closure of neutron and proton shells explains the fact that excitation levels of $^{208}\text{Pb}$ nuclei are significantly higher than those of other lead isotopes. Therefore the energy threshold of inelastic neutron scattering by $^{208}\text{Pb}$ is significantly larger than that of other lead isotopes. This fact explains weak inelastic neutron scattering by $^{208}\text{Pb}$. As a result, operation of fast reactors could become safer due to the favorable reactivity effect of coolant density caused by the lower spectral component of this effect. Moreover, small micro cross-sections of neutron capture by $^{208}\text{Pb}$ decreases one else component of the coolant density reactivity effect, namely the component related with neutron absorption [10].

In order to verify theoretical prerequisites on potential advantages caused by the use of radiogenic lead with high $^{208}\text{Pb}$ content as a coolant, neutron reflector and neutron moderator, appropriate numerical studies were carried out for fast lead-cooled reactor BREST-OD-300 [11]. The computer code TIME26 [12] was used in the numerical studies. The computer code TIME26 can determine main neutron-physical parameters of fast reactors within the frames of 26-group diffusion approximation and one-dimensional geometrical models. Micro cross-sections of neutron reactions were taken from the evaluated nuclear data library ABBN-78 and processed by the auxiliary computer code ARAMACO-C1 [13] to take the self-shielding effects into account for each reactor zone.

The computations have demonstrated that the use of $^{208}\text{Pb}$ as a coolant of the fast reactor with mixed uranium-plutonium nitride fuel made it possible to obtain some remarkable improvements [10]. For example, if $^{208}\text{Pb}$ is substituted for “common” lead, then the values of effective neutron multiplication factor $K_{\text{EFF}}$, fuel breeding ratio in the reactor core (BRC) and the coolant density reactivity effect (CDRE) could be remained at the same level by appropriate variations of plutonium fraction in fuel, pitch of fuel lattice and height of the reactor core. The computations have shown that the former values of $K_{\text{EFF}}$, BRC and CDRE may be preserved by insignificant reduction of plutonium fraction (from 13.8% to 13.6%), but by substantial widening of fuel lattice (pitch of fuel lattice must be increased from 13.6 mm to 23.6 mm) and by substantial increasing the height of the reactor core (from 110 cm to 300 cm). These changes have led to the following results:

- The same coolant heating-up in the widened lattice of elongated fuel elements can be achieved with significantly lower coolant velocity (about two-fold reduction).
- Pressure drop for coolant pumping through the widened fuel lattice can be five times lower.

Project of fast reactor BREST-OD-300 presumes the use of high-density mixed uranium-plutonium nitride fuel in order to provide small variations of the reactor criticality at full-power operation, i.e. reactivity must be well below effective fraction of delayed neutrons during full time of the reactor operation. The computations have demonstrated that substitution of the radiogenic lead for “common” lead could allow us to come back to the more assimilated and widely used MOX-fuel without substantial losses in the advantages listed above.
Design of fast reactor BREST-OD-300 does not include radial and axial blankets (fuel breeding zones) to enhance safety of the reactor operation. The favorable negative feedbacks, which could appear if $^{208}\text{Pb}$ is used as a reactor coolant, allowed us to bring back these fuel breeding zones. The computations have demonstrated that addition of axial blanket fueled with natural uranium nitride could increase total fuel breeding ratio roughly on 0.14 with insignificant worsening of other parameters.

One other advantage of the radiogenic lead is a possibility for substantial elongation of prompt neutron lifetime. The value of prompt neutron lifetime in project of fast reactor BREST-OD-300 is equal to about 0.5 $\mu$s. If $^{208}\text{Pb}$ is used as a coolant and neutron reflector (50-cm thick), then prompt neutron lifetime becomes longer by a factor of 3. If this reflector is thickened up to 5 m, then prompt neutron lifetime elongates up to 1 ms (i.e. 2000 times longer!) and becomes comparable with typical values of prompt neutron lifetime in thermal heavy-water reactors CANDU. This drastic elongation of prompt neutron lifetime can be explained by the following considerations. Fast neutrons generated in the reactor core can migrate to $^{208}\text{Pb}$-reflector, penetrate deeply into reflector, lose their initial energy after many collisions with $^{208}\text{Pb}$ nuclei and, at last, come back to the reactor core after a certain time period (delay). The opportunity for fast neutrons to come back to the reactor core from $^{208}\text{Pb}$-reflector is provided by weak neutron absorption and effective albedo properties of $^{208}\text{Pb}$. As these returned neutrons are prompt neutrons on their origination, it may be concluded that the chain fission reaction on prompt neutrons slows down, and, as a consequence, safety of the reactor operation improves.

So, the use of $^{208}\text{Pb}$ as a coolant, neutron reflector and neutron moderator in fast reactors can slow down drastically the chain fission reaction on prompt neutrons and, thus, improve significantly safety of the reactors operation.

3. Radiogenic lead in ADS

Let us note that neutron flux is much higher in ADS compared to thermal reactors that could also be considered for transmutation purposes [14]. Very low cross-sections of neutron absorption by $^{208}\text{Pb}$ together with small step of neutron slowing down can open an opportunity to form zone with high flux of slow neutrons (FSN) in blankets of ADS.

The probabilities for neutrons to avoid absorption in the process of their moderation to thermal energy in $^{208}\text{Pb}$ and in graphite are approximately the same and close to unity. On the contrary, analogous probability in “common” lead is remarkable lower than unity (~0.29). Therefore, the probability for neutrons to be scattered and slowed down by $^{208}\text{Pb}$ nuclei is substantially larger than the probability for neutrons to be absorbed. The inverse situation takes place in the case of “common” lead.

Step of neutron slowing down and cross-sections of neutron absorption by $^{208}\text{Pb}$ nuclei are seventeen times lower than those by graphite nuclei while cross-sections of neutron scattering by $^{208}\text{Pb}$ nuclei are two times larger than those by graphite nuclei. This means that neutron moderation by $^{208}\text{Pb}$ nuclei requires seventeen times larger number of elastic collisions than neutron moderation by graphite nuclei. However, at each neutron collision with $^{208}\text{Pb}$ nuclei probability of neutron absorption is thirty-four times lower than that in neutron collision with graphite nuclei. As a result, the higher FSN can be achieved in $^{208}\text{Pb}$ than that in “common” lead or in graphite.

In order to verify theoretical prerequisites on potential advantages of $^{208}\text{Pb}$, appropriate numerical studies were carried out to determine radial FSN distributions in ADS blanket composed of $^{208}\text{Pb}$ or graphite. The computations were carried out for infinite plane model of ADS blanket and demonstrated that maximal values of FSN were achieved in $^{208}\text{Pb}$-blanket (figure 1). As is seen, thermal and slow neutron fluxes in the vicinity of ADS target are remarkably higher in $^{208}\text{Pb}$-blanket than those in graphite blanket. As far as distance from ADS target increases, this advantage of $^{208}\text{Pb}$-blanket becomes larger and becomes very quickly. FSN increases for low-energy neutrons because Fermi spectrum with inverse proportionality to neutron energy establishes in low-absorbing medium. So, it may be concluded that, in the case of plane geometry, $^{208}\text{Pb}$-blanket excels graphite blanket in FSN values at any distance from ADS target.
Similar computations were carried out for more real cylindrical and spherical models of infinite ADS blanket (figure 2).

As is seen, $^{208}$Pb-blanket excels graphite blanket in FSN values at any distance from ADS target in plane geometry, at distances longer than 10 cm in cylindrical geometry and at distances longer than 40 cm in spherical geometry. As far as distance from ADS target increases, this advantage of $^{208}$Pb-blanket becomes substantially larger. This means that it is possible to form peripheral large-volume zones in $^{208}$Pb-blanket with high FSN values.

Thus, isotope $^{208}$Pb may be regarded as a candidate on the role of neutron moderator in high-flux ADS blanket with resonance or thermal neutron spectra. $^{208}$Pb-blanket can be large enough in volume to place sufficiently large amounts of radioactive wastes for neutron transmutation and to solve problem of neutron leakage.

Let us note that there is one more possible opportunity to use $^{208}$Pb for arising controlled fusion chain reaction supported by neutrons in fusion facilities with magnetic plasma confinement [15].

4. Summary
This paper has demonstrated that recovery of the radiogenic lead from thorium and thorium-uranium minerals for needs of nuclear industry could be reasonable because the use of the radiogenic lead as a
coolant, neutron moderator and neutron reflector could upgrade safety, neutron-physical and thermal-hydraulic parameters of fast lead-cooled reactors as well as upgrade ADS efficiency on neutron transmutation of radioactive wastes.

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