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Comparative analysis of using natural and radiogenic lead as heat-transfer agent in fast reactors

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Abstract. Fast reactors with lead coolant have several advantages over analogues. Performance can be further improved by replacement of natural composition lead with radiogenic one. Thus, two main issues need to be addressed: induced radioactivity in coolant and efficient neutron multiplication factor in the core will be changed and need to be estimated. To address these issues analysis of the scheme of the nuclear transformations in the lead heat-transfer agent in the process of radiation was carried out. Induced radioactivity of radiogenic and natural lead has been studied. It is shown that replacement of lead affects multiplication factor in a certain way. Application of radiogenic lead can significantly affect reactor operation.

1. Introduction

Concept of the BREST reactor assumes replacement of sodium with lead – a chemically passive and poorly activated element with high boiling temperature (1740 °C). In addition, combination of the lead heat-transfer agent and dense nitride fuel creates possibility for the following main positive changes for ensuring safe operation of fast reactors [1 – 4]:

- at breeding ratio (BR) of the core above unity reactivity margin at fuel burn-out may be reduced down to the level not exceeding ratio of the delayed neutrons. As a result, power excursion on prompt neutrons is not possible at any emergencies;
- big emissions of radioactive substances into environment are not possible even if the reactor vessel and NPP containment are damaged;
- lead has lower moderation ability than sodium; that's why emergency removal of lead from the core will affect neutron spectrum less severely and, as a result, change of the reactivity will be lower than in case of the sodium moderator.

Isotopic composition of the lead used in the BREST reactors are to be estimated. Two opposite concepts exist which assume increase the ration of:

- light isotopes, which should reduce induced radioactivity in the heat-transfer agent;
- heavy isotopes, which will allow reducing absorption of neurons in the heat-transfer agent thus improving breeding properties of the core and increase value of the nuclear fuel reproduction factor.

Besides natural lead with sufficiently high content of light isotopes, there is also radiogenic lead, which is formed in uranium and thorium ores. Isotopes Pb^{206}, Pb^{207}, Pb^{208} are final products of the radioactive decay chains of U^{238}, U^{235} and Th^{232}, respectively. So, isotopic composition of the radiogenic lead depends significantly on elemental composition of the ore and age of the deposit [5-7]. For example, radiogenic lead from thorium or uranium-thorium ores with low uranium fraction contains mainly Pb^{208}, which is interesting because of the following reasons:

- isotope Pb[208] has unique neutron-physical properties. Nucleus of Pb[208] is double magic with closed neutron and proton shells. Levels of excitation of nuclei Pb[208] are in the area of higher energies than levels of excitation of other natural isotopes of lead. That is why Pb[208] has lower moderation power.
- isotope Pb[208] has extremely low section of radioactive capture of neutrons over the whole range of energies.

This means that use of the radiogenic lead with high content of Pb[208] as moderator in fast reactors may improve their neutron-physical characteristics. Poor absorption of neutrons will allow using uranium-plutonium fuel with smaller plutonium fraction. Poor moderation of neutrons will cause reduction of spectral component of the void reactivity coefficient (VRC). [8]

2. Change of the lead-based moderator nuclide composition

Lead moderator will be irradiated with neutrons, which will lead to production of radionuclides and thus increased activity. [9].

Figure 1 represents scheme of main nuclear reactions in the lead moderator, which change nuclide composition and accumulate radioactivity.

Figure 1. Process of lead moderator transmutation
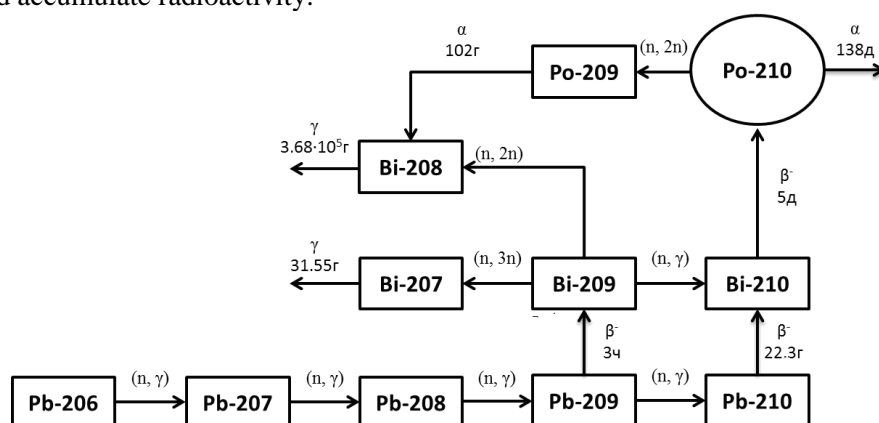
For determining dependence of the concentration values of different elements upon time of the moderator operation, it was necessary to determine values of microscopic cross sections of interaction of the neutrons of each group with the moderator nuclides. The TENDL-2012 database was used for the calculation.

The values of microscopic sections from the TENDL-2012 database were approximated by different functions, obtaining dependences of the microscopic cross sections on energy of neutrons for 26 groups. Then mean values of cross section in each group were determined taking into account spectrum of the neutron flow in a group using the ratio:

$$\sigma = \frac{\int_{E_1}^{E_2} \sigma_{\mathrm{app}}(E) \cdot \Phi(E) \cdot dE}{\int_{E_1}^{E_2} \Phi(E) \cdot dE} , \tag{1}$$

where E_1 and E_2 are the boundary values of energies for each group of neutrons; $\sigma_{\mathrm{app}}(E)$ are approximated dependences of the microscopic sections on energy of neutrons; $\Phi(E)$ is dependence of the neutron flow density on energy of the neutrons inside the considered group. These dependences were determined using the ratios:

a)   Watt spectrum for neutrons from 1st to 11th group inclusively:

$$\Phi(E) = e^{-E} \cdot sh\left(\sqrt{2E}\right) \cdot \sqrt{E} = \begin{cases} (0.913 + 0.75 \cdot E) \cdot \sqrt{E} \\ \left(0.944 + 0.0644 \cdot E^{-0.608}\right) \cdot \sqrt{E} \end{cases}, \qquad (2)$$

for $0.01 \leq E < 0,25$ MeV,

b)   Fermi spectrum for neutrons from 12th to 23[d] group inclusively:

$$\Phi(E) \sim \frac{1}{E}, \qquad (3)$$

for $0.25 \leq E < 10$ MeV;

As the BREST reactor is fast neutron reactor, we may not consider low energy groups.   The calculation is limited by first 17 groups inclusively, because fraction of neutrons in subsequent groups has negligibly low value.

Cross sections for different nuclides of moderator are shown given Table 1.

Table 1. Approximated values of microscopic cross sections from the TENDL-2012 database of micro-constants

| No of group	Pb^{204} σ_c, b	Pb^{206} σ_c, b	Pb^{207} σ_c, b	Pb^{208} σ_c, b	Pb^{209} σ_c, b	Bi^{209} σ_c, b	Bi^{209} $\sigma_{(n,2n)}$, b	Po^{210} $\sigma_{(n,2n)}$, b
1	0.00201	0.00089	0.00131	0.00080	0.00107	0.00100	0.41887	0.50067
2	0.00256	0.00045	0.00060	0.00072	0.00063	0.00100	0	0
3	0.00495	0.00056	0.00025	0.00188	0.00110	0.00150	0	0
4	0.00981	0.00078	0.00017	0.00125	0.00068	0.00200	0	0
5	0.01092	0.00344	0.00014	0.00086	0.00054	0.00250	0	0
6	0.00989	0.00604	0.03195	0.00384	0.00049	0.00250	0	0
7	0.02481	0.00632	0.03425	0.00275	0.00051	0.00250	0	0
8	0.10906	0.00612	0.02150	0.00560	0.00067	0.00250	0	0
9	0.67265	0.00810	0.00412	0.00117	0.00019	0.00200	0	0
10	0.06275	0.00801	0.03089	0.00001	0.00131	0.00200	0	0
11	0.08517	0.01679	0.01541	0	0.00219	0.00200	0	0
12	0.23902	0.00004	0.00094	0	0.00028	0.00500	0	0
13	0.25621	0.02949	0.13245	0	0.00042	0.02300	0	0
14	1.24659	0.00010	0.00252	0	0.00122	0.00150	0	0
15	0.04955	0.00015	0.00380	0	0.03667	0.19000	0	0
16	0.00611	0.00023	0.00573	0	0.52077	0.00070	0	0
17	0.00875	0.00035	0.00837	0	0.02953	0.00060	0	0
$\overline{\sigma}$	**0.13726**	**0.00659**	**0.02175**	**0.00237**	**0.00212**	**0.00378**	**0.00019**	**0.00022**

3. Results and discussion

Isotopic composition of the heat-transfer agent influences absorption of neutrons in it. Radiation capture of neutrons goes in direct proportion with fraction of light isotopes, since they have greater capture cross sections. Relative increase of k_{eff} at transition from natural to radiogenic lead will be about 2% ($k_{eff}^{rad} / k_{eff}^{nat} = 1.021$), thus significantly increasing reactivity margin in the reactor only due to use of the radiogenic lead.

Formation of radionuclides in the moderator causes increase of general radioactivity in it. In Figures 2–4 dependences of specific radioactivity in natural and radiogenic lead upon time of its operation in nuclear reaction and after the reactor shutdown are shown.
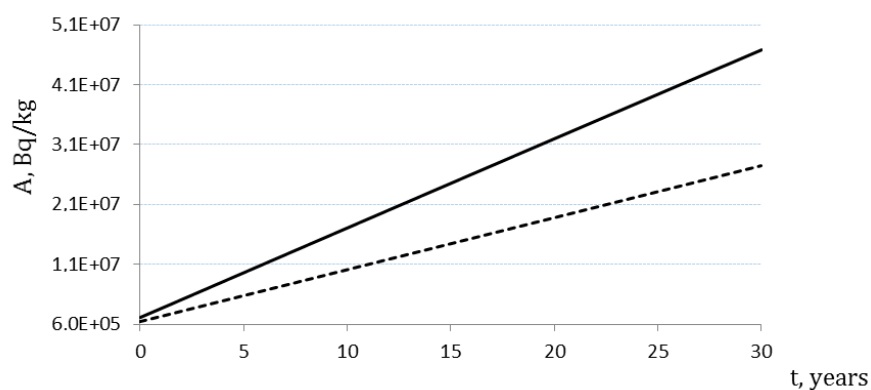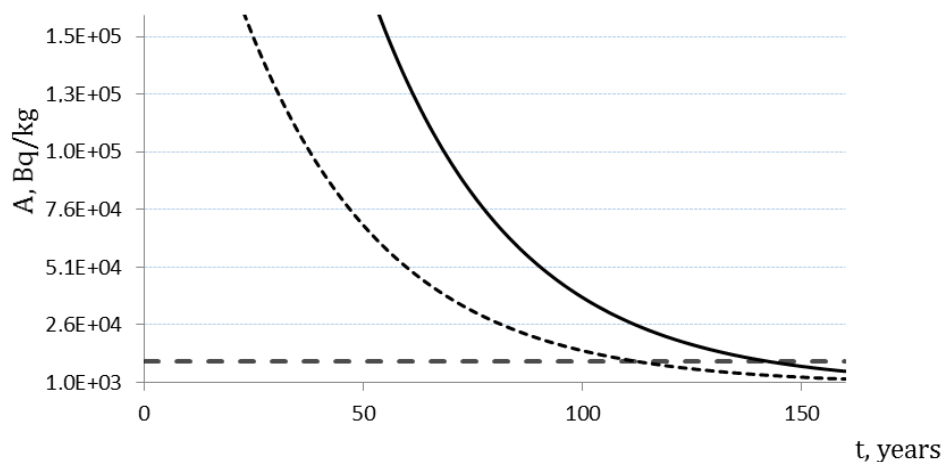

Figure 2. Change of specific alfa-activity of heat-transfer agent at 30-year operation of reactor:
────── − radiogenic; ------ − natural
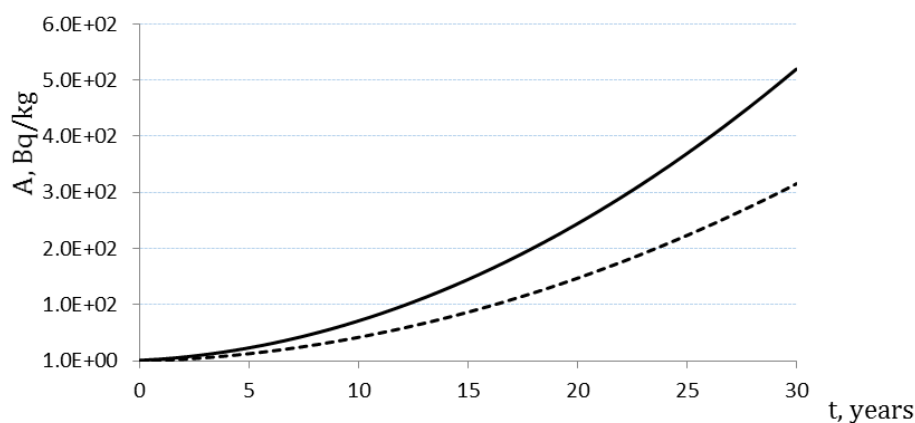
Figure 3. Change of specific gamma-activity of heat-transfer agent at 30-year operation of reactor:

────── − radiogenic; ------ − natural

Specific beta-activity of heat-transfer agent sharply increases during first hours of the rector operation and preserves its value at constant level: $7.6 \cdot 10^{12}$ Bq/kg and $1.3 \cdot 10^{13}$ Bq/kg for natural and radiogenic lead, respectively.

Figure 4. Change of specific beta-activity caused by Pb^{209}, Pb^{210}, Bi^{210} after shutdown of reactor: ▬▬ − radiogenic; ▪▪▪▪▪ − natural

Specific gamma-activity of the heat-transfer agent will be equal to 316 and 521 Bq/kg for natural and radiogenic lead, respectively. It will remain constant because of long half-life of Bi208, which makes main contribution into gamma-activity - about $3 \cdot 10^5$ years.

Conclusion

Radiogenic lead has greater induced activity compared to natural lead. This leads to increase of holding time. The results show that beta-activity is of primal importance. Due to accumulated beta-activity holding times are about 100 and 150 years for natural and radiogenic lead respectively.

However, the authors consider that increase of the reactivity margin by about 0.02 is more significant advantage of radiogenic lead in comparison with its disadvantage − the induced radioactivity. Since design value of reactivity margin in fast neutron reactors with lead heat-transfer agent does not exceed efficient share of the delayed neutrons (about 0.0063 for U^{235}), radiogenic lead may significantly affect implementation of the nuclear fuel cycle and nuclear safety in the course of the reactor plant operation.

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