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POLONIUM ISSUE IN FAST REACTOR LEAD COOLANTS AND ONE OF THE WAYS OF ITS SOLUTION

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INTRODUCTION

One of the main issues in using materials for nuclear facilities is to minimize the production of the most hazardous radionuclides. In the ideal case, all nuclear reactor materials, except a fuel, should be low-activation. The term "low-activation material" means that this one loses its induced activity in a short time after removal from irradiation. Proposals for building a fusion reactor using low-activation materials are given in Ref.1, 2. For this purpose, low-activation structural materials based on V-Ti-Cr alloys are in the stage of R&D in several countries [3,4]. Another technique to avoid the hazardous activity is in using isotopically enriched materials [5-7]. Although isotopical tailoring option requires tremendous technical efforts and it is too expensive, its application can be first of all assumed for those structural and functional materials which generate very hazardous radionuclides under irradiation.

In modern projects of next generation NPPs the preference is given to fast reactors (FRs) with a lead coolant [8]. As it known, the coolant circulating through a FR core is activated, and in the future we should have problems with handling a completed coolant after FR decommissioning or at realization of repair or emergency activities. There, it is desirable to have a low-activation coolant with the low contents of hazardous radionuclides.

In papers [9,10] presented at the previous ICONE conferences it was proposed to use lead isotope, Pb-206, as a coolant instead of lead natural, Pb-nat. This paper is devoted to more detailed calculations of accumulating stable bismuth, Bi-209, and polonium radioisotopes, Po-209 ($T_{1/2}$=102 y) and Po-210 ($T_{1/2}$=138 d), in 1 kg of Pb-nat or Pb-206 placed in the core of the BOR-60 type FR.

1.POLONIUM ACCUMULATION IN FR LEAD COOLANTS

As it known, natural lead (Pb-nat) is a mixture of 4 stable lead isotopes. Their contents in Pb-nat are shown in Fig.1.

Usually, in a typical FR neutron spectrum, such nuclear reactions as (n,γ), (n,2n) and (n,3n) are mostly responsible for the transmutation of nuclei. Lead isotope, Pb-208, contained in Pb-nat in a quantity of 52%, forms radioisotope, Pb-209, via (n,γ) reaction, as it is shown in Fig.1. This short-lived isotope is responsible for generation of several long-lived and radiotoxic nuclides. Stable bismuth, Bi-209, is produced via fast beta decay of Pb-209. Under FR neutron irradiation concentration of stable bismuth in Pb-nat is increasing year by year in a rate of 0.1 g/kg/y. Bi-209, in its turn, is a source of producing such nuclides as Bi-207 and Bi-208 via (n.2n) and (n,3n) reactions. Both of these nuclides are long-lived and hazardous enough. Bi-207 decay half-time is equal to 31.55 years and its gamma radiation energy approaches to 1 MeV. Bi-208 is a very long-lived isotope ($T_{1/2}$=3*10^5 years) and its gamma radiation energy is equal to 2.6 MeV. That is why their clearance level for specific activity is of 1*10^4 Bq/kg.

Another hazardous radionuclide induced in Pb-nat immersed in FR neutrons is alpha active polonium isotope, Po-210. As it can be seen in Fig.1, its producing is also connected with accumulation of Bi-209. In Table 1 quantities of Bi-209, Po-210 and Po-209 generated in Pb-nat after its irradiation in FR during 30 years are given.

Table 1

The contents of stable bismuth and polonium isotopes, polonium activities in 1 kg of Pb-nat after its irradiation in a FR during 30 years.

Nuclide	Nuclide contents in 1 kg of Pb-nat, g/kg	Activity of nuclide contained in 1 kg of Pb-nat, Bq/kg
Bi-209	3.027	-
Po-209	$3.05*10^{-6}$	$1.89*10^{6}$
Po-210	$6.68*10^{-4}$	$1.11*10^{11}$

From data represented it follows that activity of Po-210 induced in Pb-nat in a FR core exceeds the clearance level by 7 orders of magnitude. Taking in account that lead circulates in reactor vessel and the ratio of coolant and core zone volumes is of 500:1, the real Po-210 activity in Pb-nat coolant will be decreased to the level of $2*10^{8}$ Bq/kg, but even in this case it remains much higher than the Po-210 clearance level, $A=1*10^{4}$ Bq/kg.

In Table 2, taken from Ref.11, the release of radioactivity from a lead coolant of the BREST-type FR in the case of a severe accident involving the failure of reactor vessel is shown. Activity of Po-210 is due to the Pb-nat activation and activity of other nuclides is connected with penetration of fuel fission products into a coolant. It can be seen that in spite of a small Po-210 radioactivity release, about 3 Ci, its radiological hazard is of 40% of the total risk. Thus, the polonium problem is a very actual one for a FR with a lead coolant.

Table 2

The release of radioactivity from a lead coolant of the BREST-type FR in the case of a severe accident involving the failure of reactor vessel taken from Ref. 11.

Name	Radionuclide						
	Zn	As	Cd	I	Cs	Hg	Po
Release, Ci	500	300	100	60	100	1200	3
Relative risk	0.1	0.044	0.006	0.16	0.08	0.2	0.4

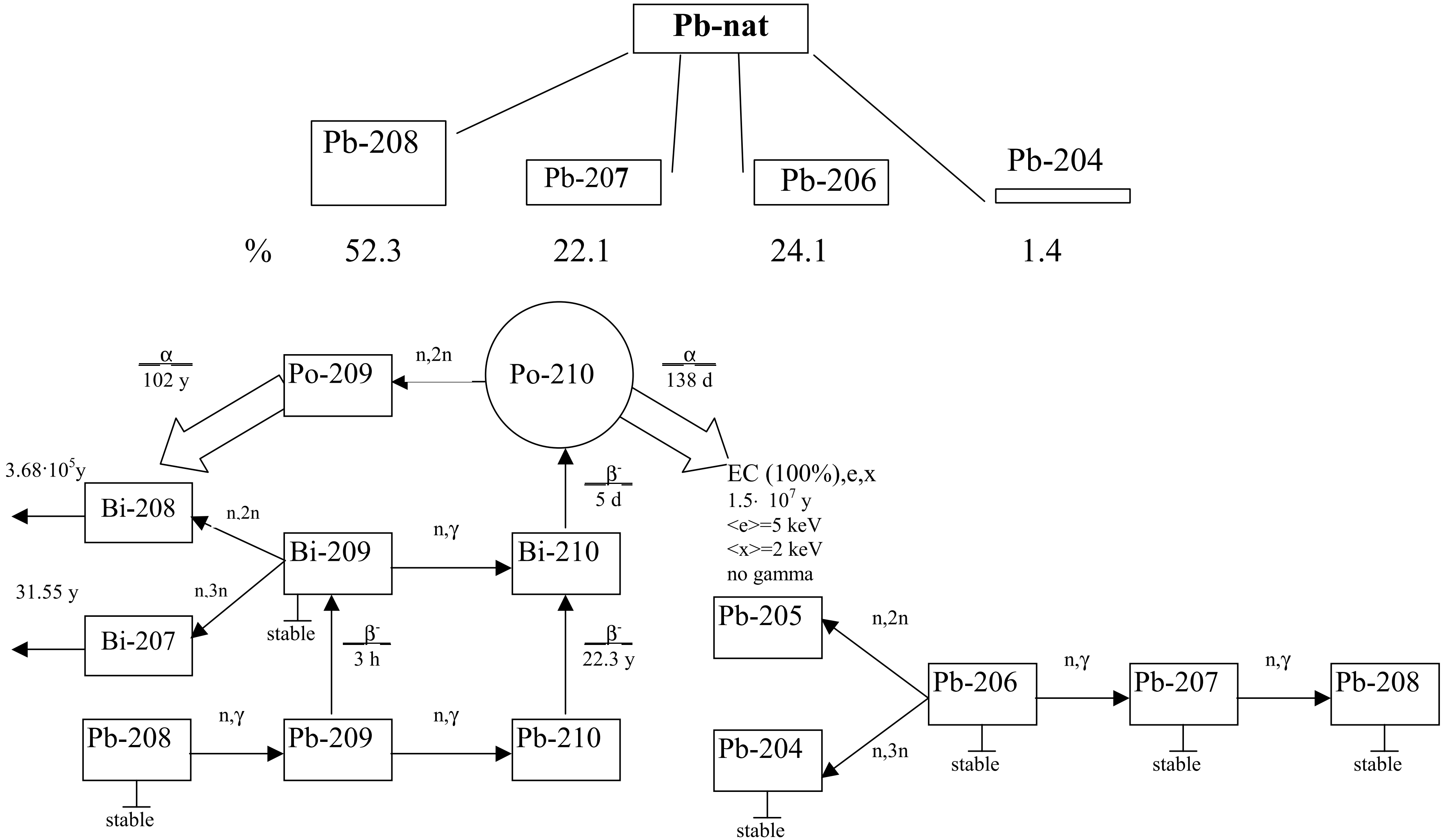

Fig.1. Pb-nat isotopical contents and schemes of Pb-208 and Pb-206 nuclei transmutations due to (n, γ), (n,2n) and (n,3n) nuclear reactions in a FR neutron spectrum.

In Table 3 results of calculation of Po-210 activity induced in lead isotope, Pb-206, after its irradiation in a FR during 30 years are given.

Table 3

The contents of stable bismuth and polonium isotopes, polonium activities in 1 kg of Pb-206 after its irradiation in a FR during 30 years.

Nuclide	Nuclide contents in 1 kg of Pb-206, g/kg	Activity of nuclide contained in 1 kg of Pb-206, Bq/kg
Bi-209	$1.87*10^{-4}$	-
Po-209	$9.33*10^{-11}$	$5.79*10^{1}$
Po-210	$3.94*10^{-8}$	$6.54*10^{6}$

It is clear from these data that the use of Pb-206 instead of Pb-nat practically solves the polonium problem in lead cooled FRs.

2.ISOTOPICALLY TAILORED LEAD TARGET WITH REDUCED POLONIUM AND BISMUTH RADWASTE

Presently, lead is considered as one of candidate materials for a target of ADS. Lead is a low cost material, it is a good liquid coolant and proton-to-neutron converter. Meanwhile, when natural lead is irradiated with fast protons a few hazardous long-lived nuclides are produced.

In such a target nuclides are produced due to spallation, fission, (n,γ), (n,xn) and other nuclear reactions. The alpha-active polonium isotopes, Po-210,209,208, and hard gamma emitting bismuth isotopes, Bi-208,207,206,205, are known as very hazardous and relatively long-lived radwaste. Calculated with the SNT code the residual activity of these nuclides produced in a target of the full-scaled ADS is represented in Figs. 2 and 3. It can be seen that activity of the Pb-nat-based target exceeds the clearance level by many orders of magnitude. The main contribution to this target radiotoxicity brings bismuth isotope, Bi-207. Its activity is of $5\cdot 10^9$ Bq/kg during 100 years of decay time. It seems that in a hard proton spectrum the main channel for Bi-207 producing is the ^{208}Pb(p,2n)^{207}Bi reaction It is clear from Fig. 3 that activity of Bi-207 when Pb-206 is used as a target decreases to the level of 10^5 Bq/kg under the same irradiation conditions as from Pb-nat.

Correspondingly, activity of other heavy hazardous nuclides, such as Po-210, Po-209, Po-208, can be suppressed essentially when Pb-206 is used in ADS targets instead of Pb-nat. As it follows from Fig. 3, in this case production of radiotoxic alpha-active isotopes of polonium can be practically excluded.

In Fig. 4 time-dependent residual doze rate for Pb-nat target after 1 year irradiation with 0.8 GeV, 30 mA proton beam is given. It is clear that radiotoxicity of a such target along 200-300 years of decay time does not permit its remote recycling mainly due to the high Bi-207 doze rate. When Pb-206 is used as a target the hands-on recycling becomes possible after only 1-2 years of decay time. It should be pointed out that in using lead isotope, Pb-206, the situation with producing lighter fragments of spallation and fission reactions does not change essentially, while generation of the heavier nuclides, such as isotopes of Po and Bi, decreases to a great extent.

Thus, although isotopic tailoring option requires tremendous technical efforts, it is still the attractive option which provides a low-activation functional material for future ADSs and FRs.

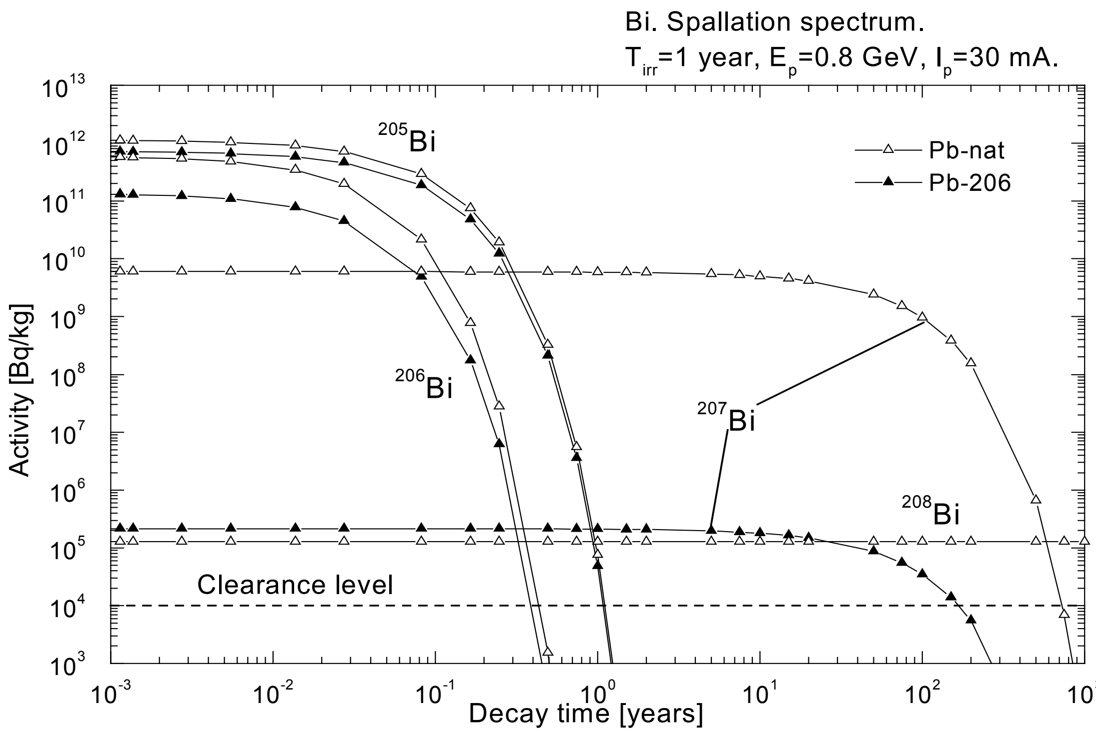

Fig.2. Residual activity of Bi-208, Bi-207, Bi-206 and Bi-205 produced in Pb-nat and Pb-206 after irradiation with proton beam E_p=0.8 GeV, I_p=30 mA, and T_{irr}=1 year.

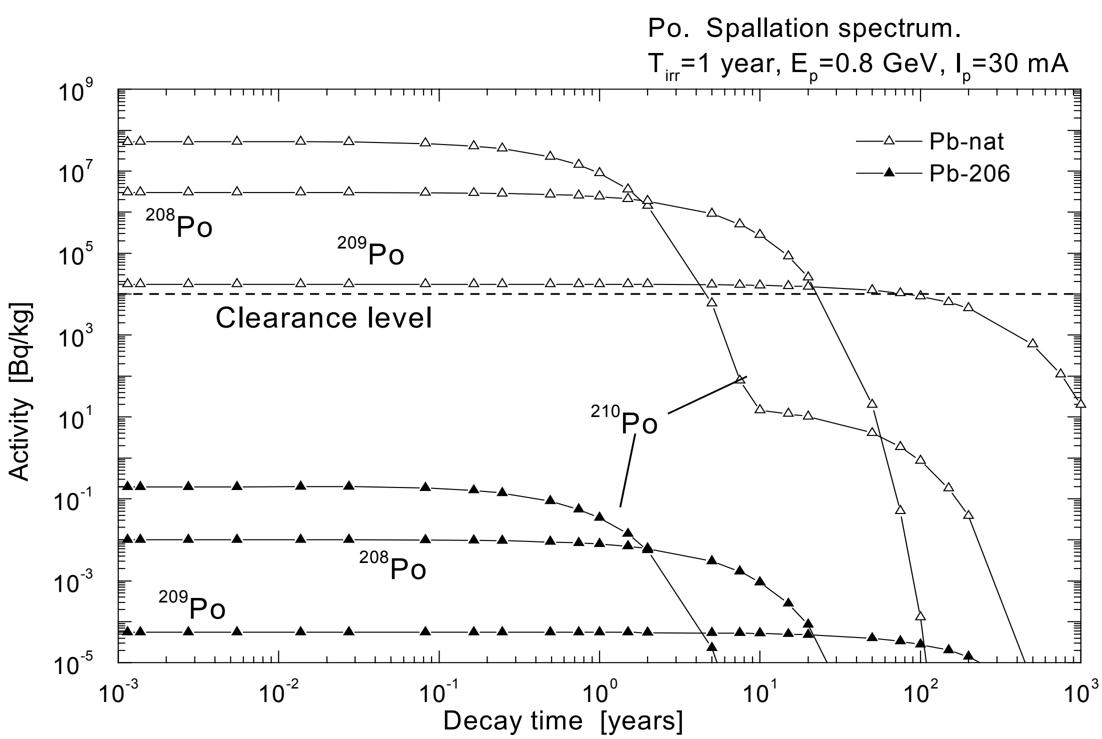

Fig.3. Residual activity of Po-210, Po-209 and Po-208 produced in Pb-nat and Pb-206 after irradiation with proton beam E_p=0.8 GeV, I_p=30 mA, and T_{irr}=1 year.

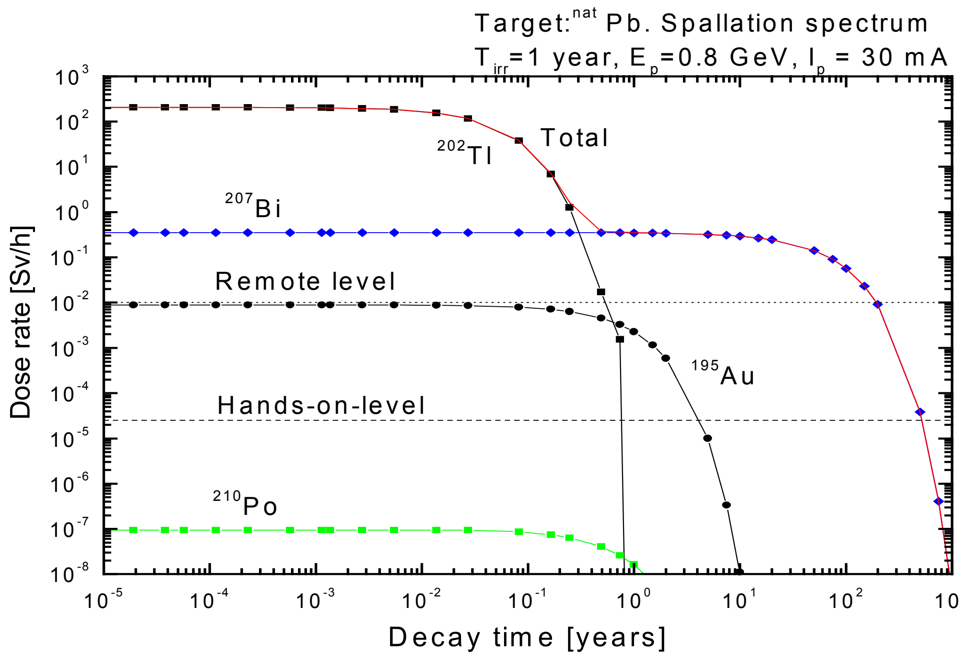

Fig.4. Time-dependent residual radiotoxicity of 1 kg of Pb-nat after 1 year its irradiation with a proton beam of 0.8 GeV energy and 30 mA current.

3.THE POSSIBILITY OF REDUCING CHARGES FOR THE FR SPENT LEAD COOLANT UTILIZATION

Now at designing advanced FRs it is recommended to envisage such decisions which reduce expenses for removal radwaste after FR decommission. Charges for disposing spent coolants and structural materials are of $20/kg [12]. Thus, for a FR such as the BN-600 expenses for utilization of the sodium coolant (800 tons) and containment materials (3800 tons) will cost $90 mln. For the FR of the same capacity but exploiting lead as a coolant, charges for spent lead storage will demand $200 mln as lead is 11.7 times heavier than sodium. Estimations performed specify the impossibility of clearing a lead coolant from radiation control after its exploiting in FRs during 30 years. It is due to an accumulation of long-lived bismuth radioisotopes, Bi-207, Bi-208, Bi-210m, and alpha-active polonium, Po-210.

The using of Pb-206 allows to take off the problems concerning the coolant residual activity. In this case, cost of lead enriching must be comparable with charges for spent lead utilization.

In Ref. 13 it is shown that for a FR designed correctly from the point of view of coolant amount minimization, demands of a coolant are of 0.7 tons/MW thermal. Thus, for the 700 MW thermal BREST-type FR it is demanded about 500 tons of lead enriched with isotope Pb-206 up to 90%. It should be mentioned that lead of Pb00 grade which is used in the BREST-type FR amounts bismuth to 0.5 g/kg. There are some difficulties to obtain pure lead or to clear it from induced bismuth during the FR operation. As it follows from Table 1, after 30 years FR operating time a coolant will include about 3 grams bismuth per 1 kg of lead natural. The usage of lead enriched with Pb-206 solves the bismuth and its radwaste accumulation issue.

CONCLUSIONS

- Residual activity of natural lead irradiated in a FR strongly depends on irradiation time due to the generation of stable bismuth, Bi-209, which is partly transmuting into Bi-207, Bi-208, Bi-210m, Po-209 and Po-210 via (n,xn) and (n,γ) reactions.
- In ADS with natural lead target a large portion of Bi-207 can be additionally produced via ^{208}Pb(p,2n)^{207}Bi and ^{207}Pb(p,n)^{207}Bi reactions.
- It is shown that for FR and ADS spectra the heavy lead isotope, Pb-208, is a source of production of the most hazardous alpha and gamma active nuclides.
- A new isotopically tailored coolant consisting of lead isotope, Pb-206, is proposed. By using this material, it is possible to reduce essentially production of the most radiotoxic isotopes of Pb, Bi and Po and to avoid disposing a large amount of spent lead.
- To provide the next generation systems with a low-activation coolant or converter, the obtaining large quantities (several hundred tons) of lead enriched with lead isotope, Pb-206, is needed.

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