EFRE 2020 | 7th International Congress on ENERGY FLUXES AND RADIATION EFFECTS

September 14 - 26, 2020 | Tomsk, Russia

NMHT 4th International Conference on New Materials and High Technologies

● Functional Materials and Coatings

N3-P-002802

RANKING OF LMFR COOLANTS BY DEGREE OF PREFERENCE

V.S. Okunev
Department of Physics
Bauman Moscow State Technical University,
Moscow, Russia

okunevvs@bmstu.ru

ABSTRACT

A new approach to ranking the coolants of liquid metal fast reactor (LMFR) by degree of preference is proposed and implemented. The approach involves comparing the optimal LMFR layouts with different coolants and compatible structural materials. The layouts were obtained in solving mathematical programming problems with restrictions in the same formulation. Permissible values of functionals and control parameters changed depending on the used coolant and structural material. The approach is implemented based on the use of operations research methods. Almost all stable metals of the periodic system of elements were considered as initial data. The problem is solved in three stages. At the first stage, on the basis of an elementary analysis of the physicochemical properties and cost, unacceptable metals with a high cost (rare metals), a small wide range of operating temperatures, a low boiling point, and high induced activity were excluded. The second stage involves obtaining the optimal layout of LMFR with different coolants. Then, using lexicographic methods for solving multicriteria problems, a coolant ranking procedure is carried out based on a comparison of the optimal LMFR layouts. The third stage involves the analysis of preferred options. The correction of the results obtained in the second stage is possible. An analysis is made of the possibility of using technological additives to the selected coolant, optimization of the composition of a multicomponent coolant, optimization of the isotopic composition of a coolant containing several stable isotopes of one chemical element. Depending on the power and purpose of the reactor, the most preferred coolants are different. For medium and high power reactors, lead extracted from thorium ores is most preferred. For low power reactors, lead from polymetallic and lead ores is preferred. In such reactors, the use of lead of uranium ores is possible. For high- and very-high-temperature low power reactors, gallium and its alloys, including gallium-lead alloys, are preferred. For space reactors, a coolant based on the eutectic Na-K-Cs alloy is more preferred.

INTRODUCTION

The world concepts of LMFR are developing in two main directions. The traditional direction involves the use of sodium coolant, an alternative direction involves the use of a coolant based on natural lead. Lead coolant has several advantages in ensuring the safety of the reactor.

The development of nuclear technology leads to the need to analyze new materials in nuclear technology. This is necessary in case of unforeseen difficulties in the transition to the new generation of reactors (including high power reactors). Based on the use of operations research methods, the ranking of potential coolants LMFR was conducted. Traditional and exotic coolants that have never been used in nuclear technics were considered. Not individual coolants or “coolant - structural material” pairs were compared, but the optimal reactors layouts (with different coolants and compatible structural materials) obtained by solving mathematical programming problems with restrictions in the same formulation (the modernized “DRAGON-M” code is used).

In 2018, there are **79 experimental facilities for researching reactors with sodium coolant** in the world, **72 facilities for researching reactors with lead coolant**, and **14 facilities for researching reactors with lead- bismuth coolant**.

(Interest only in the sodium coolant is shown by India. Interest in the study of only lead coolant is shown by Belgium, the Czech Republic, the European Union as a whole, Italy, Spain and Sweden. Interest in both sodium and lead coolant is shown by China, France, Germany, Japan, Republic of Korea, Latvia, Russia, and the USA. Facilities for studies of lead-bismuth coolant are in Russia, the USA, France.)

STAGES OF SOLVING THE PROBLEM

The task of ranking liquid metal coolants according to the degree of preference is solved in three stages.

- **At the first stage**, an elementary analysis of physical and chemical liquid metals from a wide range of source data was carried out. A wide range of potential coolants was considered: almost all stable metals of the periodic system of elements. Based on numerous literature data, stable metals were selected, characterized by a wide range of operating temperatures, that is, temperatures at which the metals are in liquid form. The properties of alloys of these metals were analyzed for various combinations and concentrations of components, i.e., the multicriteria problem had to be solved on an infinite number of objects (coolants). The least preferred metals by their physicochemical properties were excluded from consideration.
- **At the second stage**, the coolants are ranked according to the degree of preference. The approach does not consist in comparing the properties of individual coolants or pairs of "coolant - structural material". The optimal layout of the core, cooled by different coolants, including alloys, is compared.
- **The third stage** involves the analysis of preferred options. At this stage, correction of the results is possible, associated, for example, with the use of technological additives to the selected coolant, optimization of the composition of a multi-component coolant, optimization of the isotopic composition of a coolant containing several stable isotopes of one chemical element.

RESULTS AND DISCUSSION

FIRST STAGE

As a result of the analysis of the properties of all known stable metals, it was possible to reduce the initial multicriteria continuous problem to a discrete finite-dimensional problem with a relatively small number of objects: Li, Na, K, Cs, Ga, Pb, Bi, their alloys of a fixed composition (Table 1). These metals and their alloys were either used as LMFR coolants, or were considered as possible coolants, or were not used and were not considered as possible LMFR coolants. Sodium and lead of natural isotopic composition are considered as a coolant of power LMFRs of new generation. By natural is meant lead, which is close in its isotopic composition to lead extracted from galena or polymetallic ores. Its isotopic composition is averaged over all known deposits (of which about one and a half thousand). Average isotopic composition of natural lead: 1.4% ^{204}Pb - 23.6% ^{206}Pb - 22.6% ^{207}Pb - 52.4% ^{208}Pb. In nature, lead deposits with such an isotopic composition are absent. Natural lead is closest in its isotopic composition to that of the deep-water or Pacific Ocean (1.34% ^{204}Pb - 25.43% ^{206}Pb - 21.11% ^{207}Pb - 52.12% ^{208}Pb). Such lead is called modern.

Table 1. Illustration for the task of ranking coolants by degree of preference

Criteria	Coolant													
	Li	Na	K	Cs	Ga	Pb	Bi	90% Na-10% Li	Na-K	79% Na-21% K	Na-K-Cs	Pb-Bi	Na-Pb	Na-Ga-Pb
I. Wide range of operating temperatures (at 1 atm.):														
low freezing temperature	10	6	5	4	4	12	11	7	2	3	1	7	9	8
high boiling point and efficiency energy conversion cycle	5	8	11	13	1	2	3	7	10	9	12	4	6	4
maximum operating temperature range	6	9	10	11	1	4	5	8	9	9	9	2	7	3
II. Technology:														
main components of structural materials	Nb	Fe, Cr, Ni			W	Fe, Cr, Ni[1], Si		Fe, Cr, Ni				Fe, Cr, Ni, Si		W
steel corrosion problem	3	1	1	2	3	2	2	3	1	1	2	2	4	4
intermetallic formation	1	1	1	1	1	1	1	1	1	1	1	1	2	2
opportunity to solve problems within existing technologies	4	1	1	3	6	3	3	4	1	2	3	3	5	6
III. Neutron-physical properties:														
long-term induced activity	3	4	5	6	2	1	7	4	5	5	7	6	4	4
the possibility of VRE reducing	6	5	2	8	8	1	1	5	3	4	6	1	2	7
IV. Internal self-protection against ATWS:														
accidents with worsening heat sink conditions	2	6	9	11	1	1	1	5	8	7	10	1	4	3
accidents with increasing power	2	6	9	11	1	1	1	5	8	7	10	1	4	3
set of severe accidents:														
excluding restrictions for VRE[2]	4	7	10	12	1	4	4	6	9	8	11	2	5	3
subject to restrictions for VRE[2]	4	3	3	3	6	1	1	4	3	3	4	1	2	5
V. Prevalence in the Earth's crust	6	1	2	9	7	8	10	3	2	2	7	8	4	5

Note. The numbers from 1 to 13 reflect the degree of preference for the coolant according to this criterion (the most preferred corresponds to "1"); [1] it is supposed to use steel with a low nickel content in BREST type reactors; [2] consider the most dangerous scenario for the implementation of LOCA WS: drainage of the central part of the core.

SECOND STAGE

The optimal layouts of LMFRs, which are subject to further comparison, were obtained using the modernized DRAGON-M code with the same problem statement taking into account the characteristics of each specific coolant.

The optimal design problem is formulated as a mathematical programming problem with restrictions for functionals modeling the reliable (reliability functionals) and safe (safety functionals) reactor operation. As control parameters for the problem, the geometric parameters of the core and reflectors, the sizes of the zones, the geometry of the lattice of the fuel pins, the coolant flow rate, the fraction of the covers of the fuel assemblies, displacers, etc. were considered. Among the safety functionals, the maximum temperatures of the components of the reactor core, thermal power, and pressure in the cavity of the fuel pins (for collecting gaseous fission products), etc. First of all, anticipated transients without scram (ATWS) were analyzed. For each potential coolant and structural material compatible with it, the admissible values of the functionals and control parameters are different.

The whole variety of emergency processes in LMFRs boils down to various combinations of perturbations in reactivity ρ, flow rate G, and temperature T_{in} of the coolant at the inlet to the core. The emergency mode with the loss of coolant is considered separately: drainage (sometimes a decrease in the density of the coolant) of the active zone or its part (LOCA - Loss of Coolant Accident).

The most dangerous situations occur when emergency protection fails - situations like ATWS. Such processes are considered in the first place.

For example, for LMFRs, these are LOCA WS, LOF WS (Loss of Flow Without Scram, characterized by disturbance $\delta G < 0$), TOP WS (Transient Overpower Without Scram, $\delta\rho>0$), OVC WS (Overcooling Accident Without Scram, $\delta G > 0$ - the mode is caused by switching the pumps to increased performance; $\delta T_{in} < 0$ - the mode is caused by connecting a “cold” backup circulation loop, if it is provided for in the project), LOHS WS (Loss of Heat Sink Without Scram, $\delta T_{in} > 0$) and their combination.

None of the above emergency modes can be identified unambiguously. All emergency modes were combined into several groups. The unification criterion is the disturbance initiating the emergency mode ($\delta\rho$, δG, δT_{in}). Each group is characterized by several functionals. The most dangerous combinations of processes are considered. Each of them is characterized by several functionals.

Different purposes LMFRs requires consideration of different quality criteria. For medium and large power reactors, it is necessary to limit the void reactivity effect (VRE). VRE, implemented during the drainage of the entire reactor, was considered as the target functional. VRE, which is realized by draining the central part of the core (which is the most dangerous), was considered among the restrictions: VRE < 0 (or VRE < β, where β is the effective fraction of delayed neutrons). For low-power reactors, VRE is, as a rule, negative, and it can be excluded from the optimization problem. Additional requirements are imposed on multipurpose low power reactors and special purpose reactors. For high- and very-high-temperature reactors (for example, for the production of hydrogen, liquid fuel from coal, etc.), special attention should be paid to the restrictions for the maximum temperatures of the coolant and structural materials. At the same time, a coolant with a wide range of operating temperature is needed.

For space reactors, it is necessary to use a coolant operating in a wide temperature range at a low freezing temperature. In all optimization problems, constraints for the functionalities characterizing the nominal and emergency (ATWS) reactor operation modes were considered.

For example, consider the power LMFRs. As a criterion of optimality (target functional), the VRE, which is realized when the core is drained, is chosen. VRE, implemented during the drainage of the central part of the core (the most dangerous scenario), is included among the limitations of the problem.

All the resulting reactor layouts with different coolants are compatible in power and size with BN-800.

After obtaining the optimal configuration options for LMFRs, a discrete multicriteria problem was formulated. Objects of the task are potential coolants, including alloys with different content of components. After a preliminary analysis, we selected Li, Na, K, Cs (including the possibility of extracting it from spent nuclear fuel), Ga, Pb, Bi, and the alloys Na-Li, Na-K, Na-K-Cs, Pb-Bi, Na-Pb, Na-Ga-Pb with different concentrations of components.

- A wide range of operating temperatures (low freezing point, high boiling point, high efficiency of the energy conversion cycle, maximum operating temperature range). Fig. 1 illustrates this criterion.

● Technological development of the coolant (how much is the problem of corrosion of structural steels solved, the possibility of the formation of relatively refractory intermetallic compounds, the possibility of solving problems within the framework of existing technologies).

● Neutron-physical properties of the coolant in terms of induced activity, the possibility of reducing VRE. Fig. 2 shows in a histogram the values of VRE realized during the drainage of the central part of the core (the most dangerous scenario) for reactors with different coolants. The indices for the histogram columns (abscissa axis) correspond to the coolant: 1 - Na, 2 – 4.16% (wt.) Na – 22.08% K – 73.75% Cs, 3 – Na 22.8% -77.2% K, 4 – 43.5% Pb-56.5% Bi, 5 - Pb, 6 - Bi, 7 – 90% Na-10% Pb, 8 – 90% K-10% Pb, 9 – 90% K-10% Bi, 10 – 90% K-10% Bi (BN-1200 type reactor), 11 - Ga, 12 - 25% Na - 50% Ga - 25% Bi.

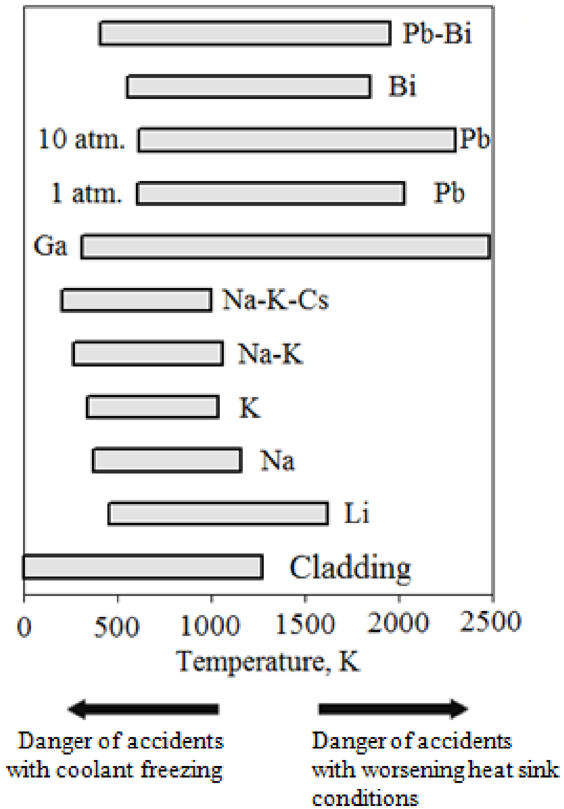

Fig. 1. Operating temperature range liquid metal coolants

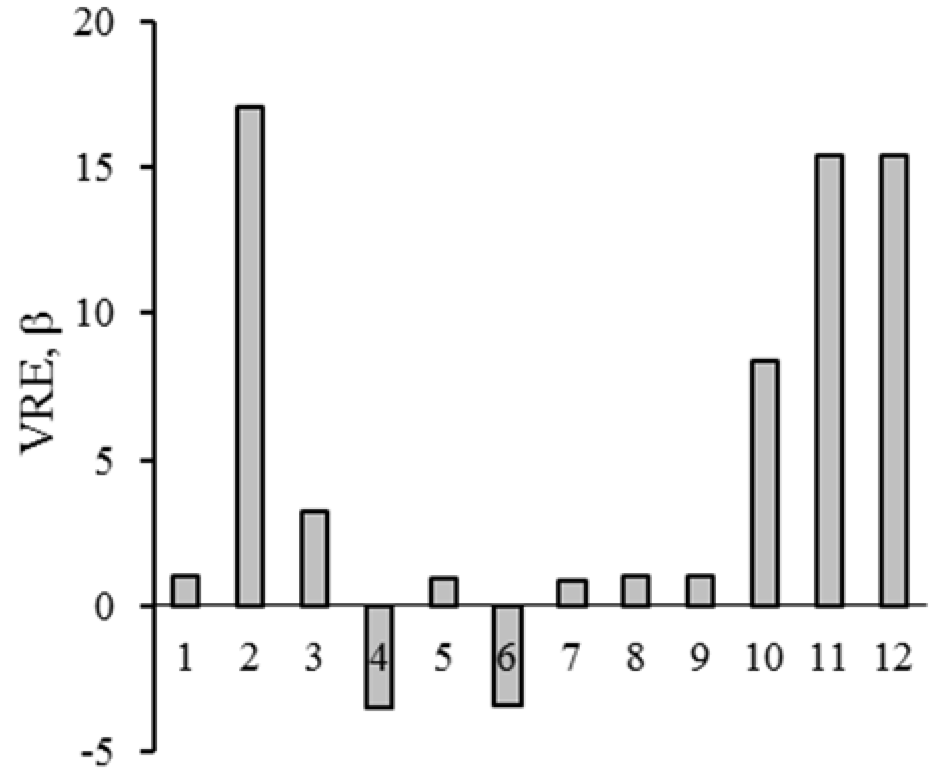

Fig. 2. Comparative analysis of VRE

● Possibility of ensuring the self-protection of the reactor (from ATWS). The LOF WS, LOHS WS, TOP WS, OVC WS, a set of severe accidents without taking into account the limitation of VRE and with the limitation are considered. Fig. 3 and 4 show the temperature reserves ΔT before the coolant boils in the LOF WS and LOHS WS, respectively. The numbering of coolants corresponds to Fig. 2.

● Cost-effectiveness. In the absence of mass production, the prevalence and concentration of metals in the earth's crust, the possibility of simplifying the design of a nuclear power plant (for example, due to the possibility of switching to a two-circuit energy conversion scheme) are accepted as an economic criterion. The prevalence in the earth's crust or sea water is given according to [1].

([1] V.A. Alexeenko, A.V. Alexeenko, "Chemical elements in geochemical systems. Clarks of soil in residential landscapes", Rostov-na-Donu, Publishing House of SFedU, 2013, 388 p.)

The lexicographic method of solving such problems is selected as the most preferred.

Tab. 1 illustrates the solution of the problem of ranking coolants by degree of preference.

For example, alkali metals are preferred by the first criterion, the lead coolant by the second and both criteria. Based on a comparison of the optimal layout of the core, cooled by different coolants, liquid lead was selected as the preferred coolant for high-power LMFRs.

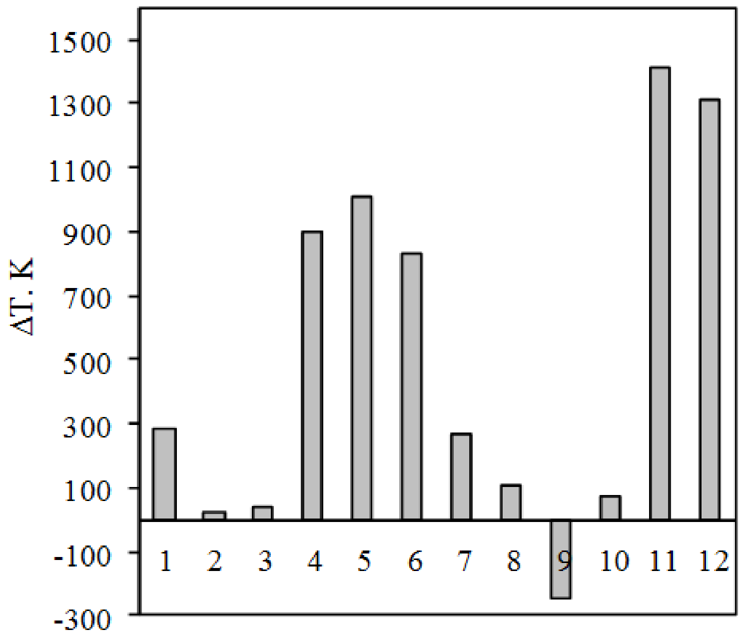

Fig. 3. The temperature reserves before the coolant boils in the LOF WS

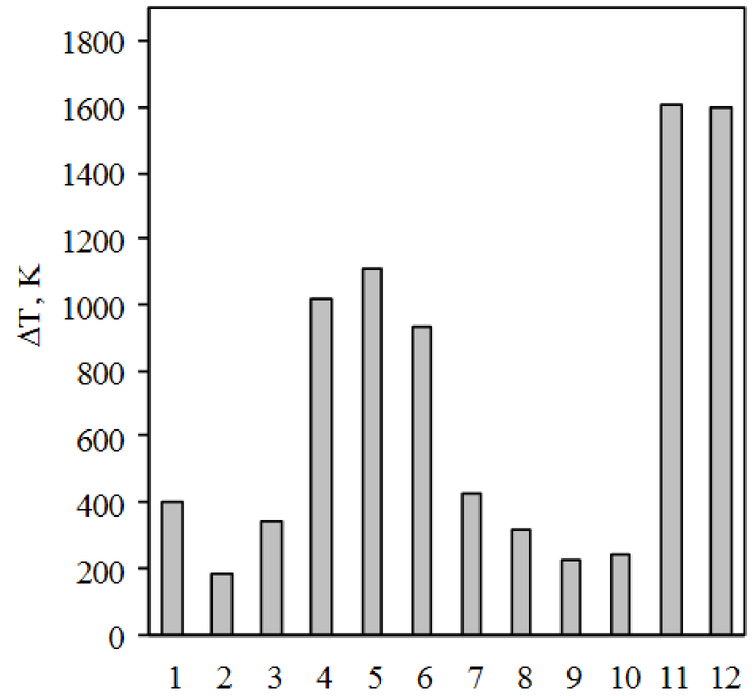

Fig. 4. The temperature reserves before the coolant boils in the LOHS WS

1 - Na, 2 – 4.16% (wt.) Na – 22.08% K – 73.75% Cs, 3 – Na 22.8% -77.2% K, 4 – 43.5% Pb-56.5% Bi, 5 - Pb, 6 - Bi, 7 – 90% Na-10% Pb, 8 – 90% K-10% Pb, 9 – 90% K-10% Bi, 10 – 90% K-10% Bi (BN-1200 type reactor), 11 - Ga, 12 - 25% Na - 50% Ga - 25% Bi.

For space reactors, a eutectic or near-eutectic Na-K-Cs alloy is more preferable. As components of such a coolant, you can use radioactive waste from nuclear energy. Spent nuclear fuel contains a high concentration of cesium isotopes. In fission of heavy nuclei, the yield of the stable isotope ^{133}Cs is from 6.61 to 7.05%, and the yield of the long-lived (half-life of 2.3 million years) isotope ^{135}Cs is from 6.37 to 7.54%. One of the possible concepts of a space reactor is a fast reactor with fuel based on minor actinides, cooled by the eutectic alloy Na-K-Cs.

For high-temperature and very-high-temperature, low-power ground-based reactors, gallium and lead, as well as alloys based on them, are of most interest.

(● V.S. Okunev, "Nuclear Power in Space: a Look into the Future", AIP Conference Proceedings **2171**, 040001 (2019); https://doi.org/10.1063/ 1.5133187 Published Online: 15 November 2019.

● M.B. Chadwick, M. Herman, P. Obložinský et al. "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data", Nuclear Data Sheets, Los Alamos National Laboratory Unclassified Report LA-UR 11-05121, **112-12**, 2887-2996 (Dec. 2011), htpps://t2.lanl.gov/ nis/data.shtml.)

THIRD STAGE

At the third stage, when the ranking of coolants is completed, the most preferred options are analyzed and specified. For example, coolants based on natural lead, lead of thorium and uranium ores, and lead containing small additives of alkali metals (lowering the freezing temperature and / or minimizing the corrosion rate) were considered as lead coolant.

Chemically pure sodium and sodium with small additions of heavy metal (reducing chemical activity) were considered as the sodium coolant.

The composition of Na-K-Cs alloys with reduced chemical activity (from the Rosebum triangle) was optimized for reactors for various purposes. The content of the components of the Na-K-Cs alloy may vary depending on the purpose of the rector. For ground-based reactors, the Na-K-Cs-based coolant is not optimal. The relatively low boiling point leads to a decrease the efficiency of the energy conversion cycle. For space reactors, a eutectic or near-eutectic Na-K-Cs alloy is more preferred. As components of such a coolant, you can use radioactive waste from nuclear energy. In particular, the stable isotope ^{133}Cs and the long-lived ^{135}Cs should be extracted from spent nuclear fuel.

In the task of correcting the composition of the coolant, the control parameters include cross sections of inelastic processes for the coolant material (which makes it possible to consider lead with small additions of other metals), the concentration of stable isotopes in the composition of the coolant (for example, lead), the coolant density, and other properties. Among the limitations of the problem are the boiling and freezing temperatures of the coolant, the temperature of destruction (depressurization) of the claddings of fuel elements.

In the analysis of heavy coolant, lead-based coolants with different isotopic compositions corresponding to different deposits (lead, polymetallic, thorium, uranium ores, etc.) were considered. In this case, the optimization of the composition of the coolant is possible due to mixing lead extracted from different deposits or different samples of the same deposit, and does not require isotope separation. For example, lead of thorium ores (extracted from monazites) is characterized by an isotopic composition: 0.01 ... 0.076% ^{204}Pb - 0.89 ... 26.43% ^{206}Pb - 0.35 ... 4.11% ^{207}Pb - 69.15 ... 97.74% ^{208}Pb. Lead of uranium ores (extracted from uraninite): 0.01 ... 0.076% ^{204}Pb - 0.89 ... 26.43% ^{206}Pb - 0.35 ... 4.11% ^{207}Pb - 69.15 ... 97.74% ^{208}Pb. Lead extracted from zircon: 0.101 ... 0.320% ^{204}Pb - 46.71 ... 75.58% ^{206}Pb - 7.74 ... 12.93% ^{207}Pb - 12.48 ... 45.40% ^{208}Pb. Lead of lead and polymetallic ores (extracted from galena): 1.09 ... 1.61% ^{204}Pb - 18.64 ... 25.17% ^{206}Pb - 21.36 ... 30.80% ^{207}Pb - 49.30 ... 52.49% ^{208}Pb.

Technological additives to lead coolant can solve some problems, but also create a number of additional problems. In particular, it is possible to reduce the freezing temperature of lead coolant by using technological additives of lithium or potassium. These additives in small quantities form a eutectic with lead, and also act as a deoxidizer, which helps to solve the corrosion problem. However, even small additions of relatively light alkali metals to lead to a significant elastic neutron moderation and an increase in VRE (Fig. 5).

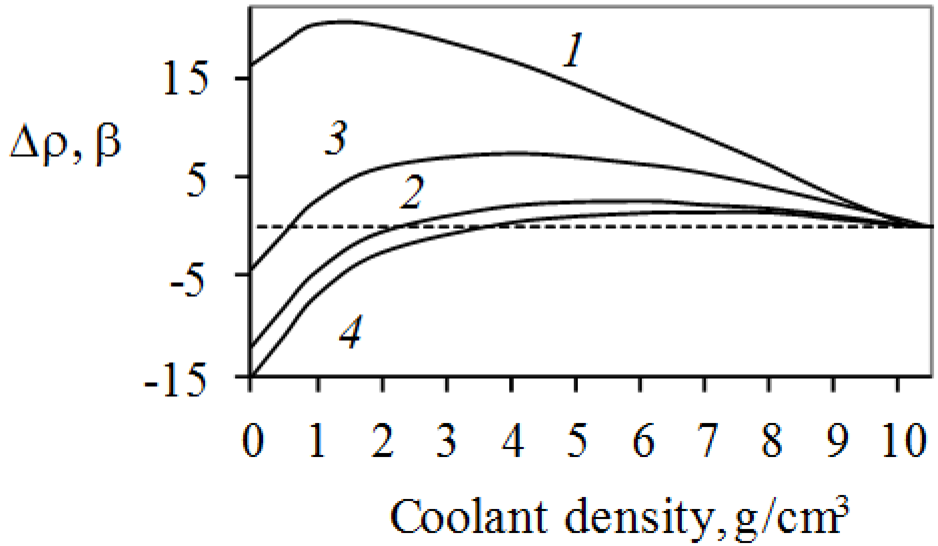

Fig. 5. The dependence of the void (corresponds to a density equal to zero) and the density effects of reactivity (Δρ) on the density of the coolant in the core and the lower reflector of the BREST reactor of high power:
1 - eutectic ^{7}Li-^{208}Pb; *2* - ^{nat}Pb; *3* - eutectic ^{nat}K-^{208}Pb; *4* - ^{208}Pb (calculations by "MCNP").

An actual task is optimization of the isotopic composition of lead, which does not require isotope separation. It is known that ^{206}Pb, ^{207}Pb, and ^{208}Pb are the decay products of ^{238}U, ^{235}U, and ^{232}Th, respectively. The ^{204}Pb isotope is of non-radiogenic origin. Its concentration in nature is low. Uranium ores are characterized by a high content of ^{206}Pb and ^{207}Pb and a low content of ^{208}Pb in lead, thorium ores are characterized by a high content of ^{208}Pb. By mixing lead from different deposits and (or) different samples of one deposit, we can obtain a continuous spectrum of changes in the concentrations of stable isotopes of lead, i.e., lead of any (given) isotopic composition.

Control parameters of the task are the concentration of stable isotopes of lead in the coolant. The criterion of optimality (target functional) for a local problem is determined by the volume of the core (or reactor power). For low power reactors, a coolant based on ^{206}Pb is preferred. Its use allows to minimize the polonium-210 production (criterion of optimality). In high-power reactors, a coolant based on the double-magic ^{208}Pb isotope should be used. This will significantly reduce the criterion of optimality - VRE (due to the small cross section of inelastic processes: absorption and inelastic scattering of neutrons), increase the lifetime of instant neutrons (due to the small cross section of absorption) and, as a result, reduce the risk of reactive accidents (second criterion of optimality). A larger neutron mean free path in lead-208 will require increasing the size of neutron reflectors or using materials with a larger absorption cross section than steel, for example, ^{99}Tc together with ^{14}C (slowing down neutrons in the reflector and increasing the transmutation efficiency of ^{14}C and ^{99}Tc) - long-lived radioactive waste from nuclear energy to be disposed of.

CONCLUSION

For medium and high power reactors, lead extracted from thorium ores with a concentration of ^{208}Pb isotope of at least 75...80% can be considered the most preferred coolant. This limits VRE (even in the most dangerous scenarios of its implementation) to safe values (< β) in reactors of arbitrarily high power.

For low power reactors, lead from polymetallic and lead ores is preferred. As the reactor power decreases, lead uranium ores with a low concentration of the ^{208}Pb isotope become most preferred. The use of such lead helps to limit the production of highly active ^{210}Po.

For high- and very-high-temperature low power reactors (small sized reactors), preference should be given to gallium and alloys based on it (including alloys with lead with a low content of the ^{208}Pb isotope). Gallium is characterized by high corrosivity. Tungsten and its alloys are most stable in liquid gallium.

For space reactors, a eutectic or near-eutectic Na-K-Cs alloy is more preferred. A eutectic alloy is characterized by a minimum freezing point for liquid metals over a wide range of operating temperatures. Costly cesium can be extracted from spent nuclear fuel.

THANKS FOR YOUR ATTENTION