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Accident Tolerant Materials for LMFR

Viacheslav Sergeevich Okunev

Abstract

Several concepts of fast reactors with liquid metal coolant (LMFR) are being developed in the world. Lead-cooled reactors are most preferred for the safe nuclear power of the future. Projects of such reactors are being developed in Russia (BREST), the European Union (the latest development is ALFRED), and the USA (a series of STAR low-power reactor designs). The potential capabilities of fuel, coolant, and structural materials considered for use in the core to increase safety have not been exhausted. There are still unused reserves that can significantly increase the self-protection of the reactor. This chapter presents the results of the analysis of the use of new types of nuclear fuel: based on ceramics and beryllium and ceramics and uranium nanopowder. Studies are being conducted on the possibility of optimizing the composition of lead coolant without isotope separation. The possibilities of improving the safety of LMFR with a coolant based on lead extracted from thorium ores are being investigated. The possibility of using tungsten coatings of the cladding of fuel pins deposited using low-temperature plasma spraying is analyzed. The composition of materials was optimized in terms of improving reactor safety. The proposed innovations will significantly increase the self-protection of the reactor from the totality of severe accidents.

Keywords: liquid metal-cooled fast reactor, mixed oxide fuel, mixed nitride fuel, uranium nanopowder, self-protection, anticipated transients without scram, void reactivity effect

1. Introduction

Currently, the world is considering two basic concepts of medium- and high-power liquid metal-cooled fast reactor (LMFR): sodium cooled and lead cooled. Solving the problem of core optimization with restrictions characterizing the safe completion of emergency conditions accompanied by a failure of emergency protection, formally it is possible to eliminate severe accidents in a reactor with lead coolant and mononitride fuel. (The author used the DRAGON-M code [1], which makes it possible to optimize the geometric characteristics of the LMFR for given materials.) For the sodium-cooled LMFR, it is impossible to exclude severe accidents with core destruction.

It is time to optimize core materials. First of all, within the framework of the systematic approach, the multicriteria problem of choosing the optimal coolant for a power LMFR and ranking coolants according to the degree of preference was solved [2, 3]. Then, the properties of core materials (density, thermal conductivity, etc.) were included in the control parameters of the DRAGON-M. Then it remains only to select materials in a given range of properties.

2. Background

2.1 Background of the development of new technologies

The recent decades have radically changed the opinion of scientists about laws linking the probability of an accident and its consequences and about ways to prevent technological disasters and, in particular, ways to ensure the safety of nuclear power facilities. This is based on the following objective factors:

1. As industrial society and nuclear power developed in the early 1980s, it was revealed that the dependence of the probability of accidents (including at nuclear reactors and facilities) on damage or consequences (human casualties or the costs of measures to prevent them) is close to the distribution Gauss. The most probable accidents are characterized by small damage and the unlikely ones—by large damage, that is, technological disasters should occur extremely rarely, and nonserious accidents—often. Accumulated by the end of the twentieth century, catastrophe statistics on industrial facilities and transport have somewhat changed the usual idea: the probability of accidents depends weakly on damage; in any case, this dependence is closer to the exponential distribution. There is reason to believe that in the near future, the exponential distribution will be replaced by a uniform distribution: severe accidents and minor emergency situations will be equally probable.
2. Prior to the first serious accidents at nuclear power plants (NPPs), there was confidence that all emergency situations with unacceptable releases of radioactive substances into the environment could be prevented by using technical (engineering) safety systems. After serious accidents at NPP, safety criteria came to the fore. The “engineering” philosophy has been replaced by the philosophy of “inherent safety.”
3. An increase in the number of potentially dangerous objects will increase the likelihood of accidents. By the middle of the twenty-first century, Russia is planning a transition to large-scale nuclear energy. If, for example, the number of power units increases by 10 times, then the probability of an accident (in a first approximation) will increase by 10 times. For this reason, when designing new-generation reactors and NPPs, a deterministic approach is preferred [4].
4. Nuclear energy is relatively young; emergency statistics are unrepresentative, which casts doubt on the appropriateness of using probabilistic analysis to justify the safety of reactors and opens up wide possibilities for using deterministic and (or) phenomenological approaches.

2.2 The history of the issue

After the Three Mile Island accident (1979), the work to eliminate severe accidents at NPPs was intensified in the USA and Europe. By the mid-1980s, concepts were proposed for the PIUS light-water reactor (ASEA-Atom, Sweden), high-temperature reactors KWU/Interction (Germany), GCRA (USA), DYONISOS (Switzerland), and MSGR liquid-salt reactor (China). The aim of the development was to eliminate the destruction of the core in severe accidents.

Among the technologically developed reactors, liquid metal-cooled fast reactors proved to be the most attractive, as far as possible to avoid severe accidents. In the early 1980s, in the USA research began on theoretical and experimental modeling of

accident transients without scram (ATWS). By early April 1986 (a few days before the accident at Chernobyl NPP in the USSR), the work on experimental modeling of ATWS regimes at the EBR-II reactor (Argonne National Laboratory, Idaho, USA) was largely completed, demonstrating high capabilities in ensuring self-protection of low-power reactors with sodium cooling. The results of these unique studies are published in [5–10]. Zirconium-doped metal fuel has been proposed as a possible accident-tolerant fuel for fast sodium reactors in the USA [11]. It is a high-density and heat-conducting fuel. By the early 1990s, projects of modular sodium-cooled fast reactors PRISM and SAFR with such fuel were considered as promising in the USA [11, 12].

After the accident at the Chernobyl NPP (1986), Soviet scientists began to solve the problem of excluding severe accidents at reactors. It was clear that experimental modeling was limited in relation to the problem of safe reactors. First of all, this applies to the full-scale modeling of the ATWS. Such experiments are expensive and difficult to perform. They pose a potential danger to the public and the environment.

Sodium is characterized by high chemical activity. Its use requires an intermediate circulation circuit. Reactor used a three-circuit cooling systems. The melting point of metal fuel doped with zirconium is relatively low. Russia went on the way of the development of the heavy coolant (lead), much less active chemically, and more heat-resistant mixed mononitride fuel (MN). MN fuel is a reasonable compromise between heat-resistant mixed oxide (MOX) and high-density and heat-conducting metal fuel (including zirconium doped).

By 1989, the Soviet Union had developed a conceptual design of the lead-cooled BRS-1000 power fast reactor fueled by MN. By 1993, another conceptual design of the BRS-300 pilot reactor was proposed. It was the predecessor of the BREST-OD-300 project (JCS “NIKIET,” Moscow, Russia), which has been developed to date [13–15]. The BREST uses a two-circuit cooling systems.

2.3 Specific hazards of fast reactors

In addition to the general hazard factors, fast reactors are organically characterized by reactivity accidents (the reactor is prompt supercritical) and the possibility of implementing a positive void reactivity effect (VRE) when using a liquid metal coolant. VRE is realized when the core or part of it is drained. Consider these two factors.

2.3.1 Risk of reactivity accidents

When entering reactivity $\rho > \beta$ (β is the effective delayed neutron fraction), the reactor is prompt supercritical. The change in reactor power or neutron density n over time t can be estimated using the point kinetics equation:

$$\frac{dn}{dt} \approx \frac{(\rho - \beta)nk}{l}, \quad (1)$$

where k is the effective multiplication factor, l is the average lifetime of prompt neutrons, $l = l_{\infty}P$, l_{∞} is the average lifetime of prompt neutrons in an infinite medium, and P is the non-leakage probability.

Average lifetime of prompt neutrons in an infinite medium

$$l_{\infty} = \frac{\Lambda_a}{v} = \frac{1}{v \Sigma_a}, \quad (2)$$

where Λ_a is the average path length of a neutron until absorption.

In a fast reactor, the average neutron velocity v is several orders of magnitude higher than in a thermal reactors, although the macroscopic absorption cross section

Σ_a is two orders of magnitude lower. As a result, in fast reactors $l \sim 10^{-7}$ – 10^{-5} s, in thermal reactors $l \sim 10^{-3}$ s, other things being equal (ρ , β), the dn/dt in fast neutron reactors is higher, i.e., the power or density of neutrons increases faster. Obviously, the use of a coolant with a small absorption cross section also contributes to the self-protection of a fast reactor from reactivity accidents. In this case, the advantages of thorium lead (^{208}Pb) are obvious.

Another feature of fast reactors is the presence of ^{239}Pu in the fuel as the main fissile nuclide. The effective delayed neutron fraction for ^{235}U is equal 0.68%. The effective delayed neutron fraction for ^{239}Pu is equal 0.217%. As a result, when the same reactivity is introduced, the difference ($\rho - \beta$) is greater in a reactor using ^{239}Pu as a fissile nuclide (compared to ^{235}U). In fast reactors, the fission of fertile nuclides (heavy nuclei with an even number of neutrons) plays an important role. Depending on the fission cross section of such nuclei (^{238}U , ^{232}Th) on the kinetic energy of the neutron that caused the fission, there is a threshold at $E \approx 1.4$ MeV. In the spectrum of the thermal reactors, the contribution of the fission of fertile nuclides to the effective neutron multiplication coefficient is small: 5–7%. In fast reactors, 1/4–1/3 of the fertile nuclides are fissioned. The value of β for ^{238}U is 1.61%, for ^{232}Th is 2.28%. When used in a fast reactor mixed U-Pu-fuel containing dump uranium and plutonium with a high concentration of ^{239}Pu , the effective fraction of delayed neutrons will be significantly higher than when fission isotope-pure ^{239}Pu : about 0.36–0.38%, and when using ^{232}Th —even higher.

A certain role is played by fast neutron fission of some nuclei from among the minor actinides, for example, long-lived radioactive waste with an even number of neutrons: ^{237}Np and ^{241}Am , contained in small quantities (up to 5%) in the fuel of new-generation fast reactors to recycle this waste and protect the fuel cycle from proliferation. The value of β for ^{241}Am is 0.16%. The fission threshold of these nuclei is slightly more than 1 MeV. In the fast reactor spectrum, up to 1/3 americium nuclei can fission, which leads to a decrease in the average value of β and an increase in the potential danger of reactivity accidents. From the standpoint of the deterministic approach to exclude this accident, it is necessary to prevent the input of reactivity ($\rho > \beta$). To do this, it is necessary to limit (minimize) the reactivity reserves and, first of all, the fuel burnup reactivity reserve ($\Delta\rho_b$). For a homogeneous core, the conditions breeding ratio in core $\text{BRC} = 1$ and $\Delta\rho_b = 0$ are equivalent. In practical cases, the total reactivity margin can be limited to β .

2.3.2 The problem of positive void reactivity effect

The VRE is characterized by a strong spatial dependence. The most dangerous is the drainage of the central part of the core [4, 16]. In this case, the neutron leakage from the core is small, and, as a rule, the VRE is maximum. Draining the reflector leads to an increase in neutron leakage from the core, with the $\text{VRE} < 0$. During the transition to high-power reactors, the volume of the core increases, and the leakage of neutrons from it decreases, which leads to an increase in VRE. In high-power reactors with a traditional core layout, the local VRE, which is realized when its central part is drained, is positive and, as a rule, several times higher than the effective fraction of delayed neutrons.

There are several components of VRE associated with spectrum changes and neutron leakage, reduction of parasitic absorption, and changes in self-shielding factors [4]. The spectral component of the VRE and the leakage component are maximal in absolute value and opposite in sign. They are mainly and determine the VRE. From the point of view of VRE minimization, a coolant based on double-magic nuclei (e.g., lead-208), fuel and structural materials with a high neutron capture cross section are of interest.

2.4 Problems of oxide fuel use

1. When using dioxide fuel in fast reactors of medium and high power, there are some problems associated with the opposite role of the Doppler coefficient of reactivity in emergency situations of different types and, as a consequence, the contradictory requirements for the Doppler coefficient in the design of self-protected reactors [16–18].
2. When using dioxide fuel (including with a reduced proportion of oxygen), traditional fuel assemblies, and fuel pin grids, the maximum value of BRC < 1 and does not exceed 0.86. This is due to the elastic scattering of neutrons on oxygen nuclei and, as a consequence, a decrease in the energy of neutrons and a decrease in the average number of neutrons produced per fission. The low thermal conductivity and density of the dioxide fuel reduce the self-protection of the reactor. Low BRC requires significant (several times greater than β) reserves of reactivity burnup, which does not allow deterministically to exclude reactivity accidents (with the reactor is prompt supercritical). Monoxides do not have the necessary temperature resistance and are not used as fuel for nuclear reactors.
3. At operating temperatures, oxygen is released from plutonium oxide and migrates to the cladding of fuel pin [18]. In the presence of free oxygen, the chemical interaction of nuclear fuel, fission products (cesium, iodine, tellurium, bromine, selenium, antimony), and technological impurities (chlorine, fluorine, carbon dioxide) with the cladding of fuel pin reduces the strength and plasticity of the cladding, increases the rate of corrosion of the inner surface of the cladding. Developers of oxide fuel for LMFRs in the USSR, USA, and Great Britain faced the problem of oxygen yield at the dawn of the development of fast reactors. To “bind” free oxygen, a getter is needed. In Russia, the first experiments on the use of vibrocompacted MOX fuel with uranium getter (3–10% by weight) for LMFRs were started at JSC “VNIINM” (Moscow), then continued at JSC “SSC NIIAR” (Dimitrovgrad) [19, 20]. Research was carried out at the BOR-60 reactor since 1981, BN-350 since 1982, and BN-600 since 1987. The transition to such fuel in BN-600 did not solve the problem of corrosion of the inner surface of the cladding.

2.5 Modern directions of development of power LMFRs

As noted, the world is considering two main concepts of medium-power and high-power LMFRs: with sodium and lead coolant. As a possible heat carrier for use in low-power reactors, the eutectic alloy Pb-Bi [15, 21, 22] is considered. In the 1990s, South Korea, China, Japan, and other countries showed interest in fast lead-bismuth reactors. In Russia, projects of such low-capacity reactors are being developed [15, 22]. Bismuth is significantly more expensive than lead. The use of bismuth leads to the working time of the short-lived highly active isotope polonium-210 in the heat carrier. In the 1990s, systems for purifying heat carrier from polonium were actively developed during reactor operation [23].

Sodium coolant is technologically developed. In many countries, it is preferred in the development of LMFRs [15]. In Russia in 2016, the power unit of the BN-800 was put into power operation; the project of the BN-1200 power reactor is being developed [24, 25].

Lead has not yet been used as a coolant. According to the developers of BREST projects, their implementation is possible within the framework of existing Russian

technologies [14]. The key works that initiated the development of the BREST direction can be considered articles [13, 26–28].

Works on the development of lead-cooled fast reactors are actively carried out in Russia (technical design and planned construction of BREST-OD-300, work on the concept design of MN fuel BREST-1200 is underway [27, 29]), European Union countries (concept projects ELFR, ELSY, LEADER, ALFRED with MOX fuel have been developed since 2006 [30]), and the USA by 2005–2007, and conceptual projects of STAR series of reactors of different target purpose with UN-fuel have been proposed [22, 30].

2.6 Problems of BREST concept during transition to high-power reactors

In terms of the possibility of avoiding severe accidents among fast reactors, the BREST-OD-300 project is most attractive [14]. It's a low-power reactor. It is well-known that it is much easier to ensure the safety of a low-power reactor. At reactor power increase (BREST-1200), there are problems with inherent safety provision. For example, the VRE is several times greater than β . The achievement of high economic efficiency of the NPP with BREST reactor may be hampered by the relatively high rate of corrosion-erosion of liquid lead on structural materials. If in BREST-OD-300 it is estimated at 39 kg of steel (for core only) per year [14, 31], for BREST-1200 it can be $39 \cdot 4 = 156$ kg/year. At the same time, economic estimates should correspond not to the cost of steel but to the cost of fuel assemblies, in which fuel elements will be depressurized.

The use of MN fuel will be accompanied by the release of nitrogen (from the PuN) and its migration to the cladding of fuel pin. In the presence of free nitrogen, the corrosion rate of the inner surface of the claddings increases. (The process is similar to the release of free oxygen from MOX fuel. The only difference is that the corrosion rate in the presence of nitrogen is less than in the presence of oxygen).

The absence of zirconium in reactor core allows deterministic elimination of steam-zirconium chemical reaction. However, hydrogen release is possible in other metal-based chemical reactions implemented at high temperatures. Chromium reacts with steam (water) in the absence of oxide film on the fuel element surface: $2\text{Cr} + 3\text{H}_2\text{O} \leftrightarrow \text{Cr}_2\text{O}_3 + 3\text{H}_2$. In this reaction, heat and free hydrogen are released. The surface of cladding must be carefully protected from possible contact with the steam involved in the core. (Chromium is present as part of the structural steel of fast reactors. The BREST uses a two-circuit cooling systems. Lead is primary circuit, steam is secondary circuit).

According to journalists, the interaction of nitrogen with lead (if there is a lead layer between the fuel and the cladding) can cause the formation of explosive lead azide. Lead does not react with nitrogen and such an event is highly unlikely. Lead azide production requires special conditions and reagents not present in BREST.

2.7 The considered emergency operation

As part of the deterministic approach to safety analysis [4], ATWS requires priority consideration. In fast reactors with liquid metal cooling, the whole set of emergency situations, including ATWS, is described (simulated) by perturbations of reactivity $\delta\rho$, coolant flow rate δG , and coolant temperature at the core inlet δT_{in} . When designing LMFRs, the following emergency situations (commonly used abbreviations are specified) and their combinations correspond to various disturbances:

- $\delta\rho > 0$ —TOP WS (transient overpower without scram).
- $\delta G < 0$ —LOF WS (loss of flow without scram).
- $\delta G > 0, \delta T_{BX} < 0$ —OVC WS (overcooling accident without scram).
- $\delta T_{BX} > 0$ —LOHS WS (loss of heat sink without scram).
- Separately consider the loss of coolant accident without scram (LOCA WS).

Simulation of emergency modes LOF WS, TOP WS, LOHS WS, and OVC WS was performed using a code version of FRISS-2D and DRAGON-M [1]. DRAGON-M and MCU codes [32] are used to analyze LOCA WS.

2.8 Problem definition

Modern projects of fast reactors with sodium and lead coolant have some disadvantages and, therefore, not yet used reserves in terms of increased safety. This is particularly evident in the transition to high-capacity power reactors (e.g., BN-1200, BREST-1200). There are opportunities to improve the fuel, coolant, and structural materials used. The greatest effect in improving the safety of the next-generation reactors should be expected with the complex (simultaneous) use of improved core materials.

This chapter offers some ideas for improving fuel, coolant, and structural materials to improve reactor self-protection.

3. Lead-208-based coolant

Among the realistic liquid metal coolants of fast reactors are sodium, lead, and eutectic alloy of lead and bismuth. **Table 1** shows data on the prevalence of lead, bismuth, and sodium, their world production, and world reserves (according to data [33]).

Natural lead is intended to be used in lead-cooled reactors. The isotopic composition of such lead (^{nat}Pb : 1.4% ^{204}Pb – 23.6% ^{206}Pb – 22.6% ^{207}Pb – 52.4% ^{208}Pb) is obtained by averaging all known deposits (about 1500). The isotopic composition of lead in different fields can vary significantly (see **Table 2**).

The large differences in the isotopic composition of lead from different deposits open up great opportunities for optimizing the composition of lead coolant without the use of expensive isotope separation technology. By mixing lead from different deposits, it's possible to obtain a coolant with a given concentration of stable

Element	Prevalence		World production, tons/year	World reserves, tons
	10 ⁴ %	Place		
Na	23,000	6	2 × 10 ⁵ (metal), 2.9 × 10 ⁷ (carbonate), 1.68 × 10 ⁸ (salts)	Are almost not limited
Pb	14	37	4.1 × 10 ⁶	8.5 × 10 ⁷
Bi	0.048	70	3 × 10 ³ (associated with lead and copper)	There are no data

Table 1.
World production, reserves, and prevalence of certain elements in the Earth's crust.

Ore	²⁰⁴ Pb	²⁰⁶ Pb	²⁰⁷ Pb	²⁰⁸ Pb
Monocyte of pegmatites of granite thorium ores	0.01–0.076	0.89–26.43	0.35–4.11	69.15–97.74
Uranitol of granite of uranium ore gneiss	0.12–0.15	85.53–85.81	10.49–10.54	3.58–3.69
Zircon of a pegmatite pink, granites, monocyte, magmatite	0.101–0.320	46.71–75.58	7.74–12.93	12.48–45.40
Lead and polymetallic ore galenite	1.09–1.61	18.64–25.17	21.36–30.80	49.30–52.49
Deep sea and Pacific Ocean	1.34	25.43	21.11	52.12
Natural lead (^{nat} Pb)	1.4	23.6	22.6	52.4

Table 2.
Isotopic composition of lead of different deposits, % (wt) [16].

isotopes, i.e., with given nuclear-physical properties. If it's necessary to minimize VRE, thorium lead with a high concentration of ²⁰⁸Pb (decay product of ²³²Th) is preferable, and if it is necessary to minimize the production of ²¹⁰Po, uranium lead with a high concentration of ²⁰⁶Pb (decay product of ²³⁸U), and small impurities of ²⁰⁷Pb (decay product of ²³⁵U), as well as non-radiogenic lead ²⁰⁴Pb are required. The use of uranium and non-radiogenic lead leads to an increase in VRE.

Figures 1 and 2 present statistics on the isotopic composition of lead in the monocytes of the Ukrainian shield. The age of the breed is about 2 billion years. An analysis of 49 samples was carried out: along the abscissa axis is the sample number according to [34].

Figure 3 presents the results of the MCU calculation of the void and density effects of reactivity ($\Delta\rho$) in a high-power BREST reactor. In **Figure 3(a)**, the dependence of VRE on the content of ²⁰⁸Pb in the coolant is given. **Figure 3(b)** shows the dependence of the effects of reactivity on the density of a coolant based on lead of polymetallic ores. Various drainage scenarios are considered: (1) the entire reactor (when calculating the density effect—the change in the density of the coolant in the entire reactor); (2) the core and the lower reflector (or a change in the density of lead); and (3) drainage of the core. The implementation of VRE involves the complete loss of coolant (drainage of the reactor, core or part thereof), the implementation of the density effect involves a change in the density of the coolant.

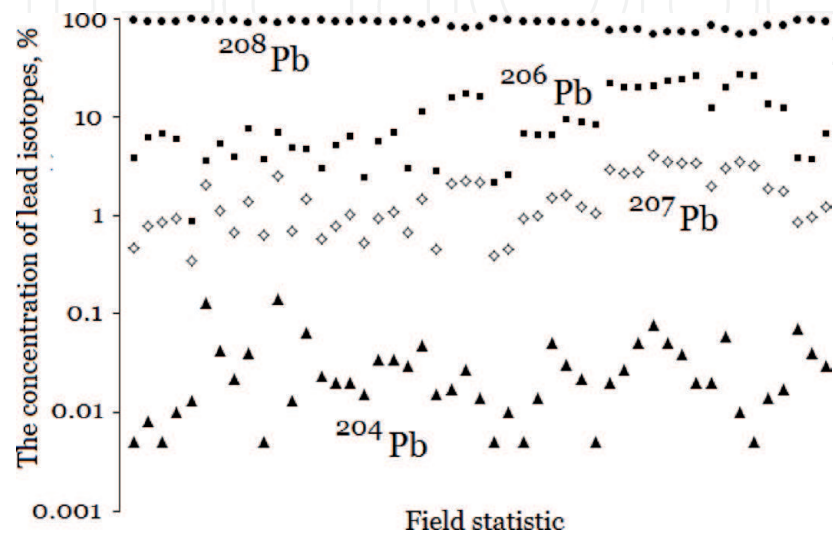


Figure 1.
The concentration of lead isotopes in different samples of monazite.

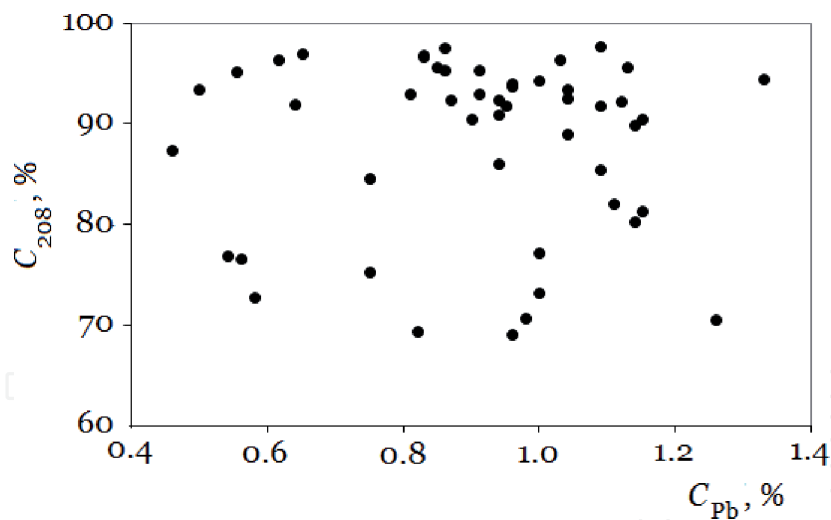


Figure 2.
The dependence of the content of the isotope ^{208}Pb in lead (C_{208}) on the lead content in monazite (C_{pb}).

Table 3 shows the VRE values (calculations according to the MCU code) for various scenarios of drainage of the BREST reactor. For comparison, the last line shows the VER values for the sodium-cooled reactor. The numbers indicate drainage scenarios (see **Figure 3**).

Mineral, ore, isotope	The isotopic composition of coolant, % (wt)				VRE (scenarios), \$		
	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb	1	2	3
Monazite	0.013	0.89	0.35	97.74	−31.7520	−15.1263	11.6096
Zircon	0.226	75.58	11.54	12.48	−14.2736	0.4048	22.6550
Uraninite	0.150	85.53	10.64	3.69	−12.8460	1.6599	25.0906
^{nat}Pb	1.4	23.6	22.6	52.4	−20.5426	−4.2157	−4.2151
^{204}Pb	0	0	0	100	−32.4267	−15.16023	11.2648
^{nat}Na	^{23}Na (100%)				−4.3644	2.5481	19.3386

Table 3.
VRE values for various drainage scenarios (LOCA WS).

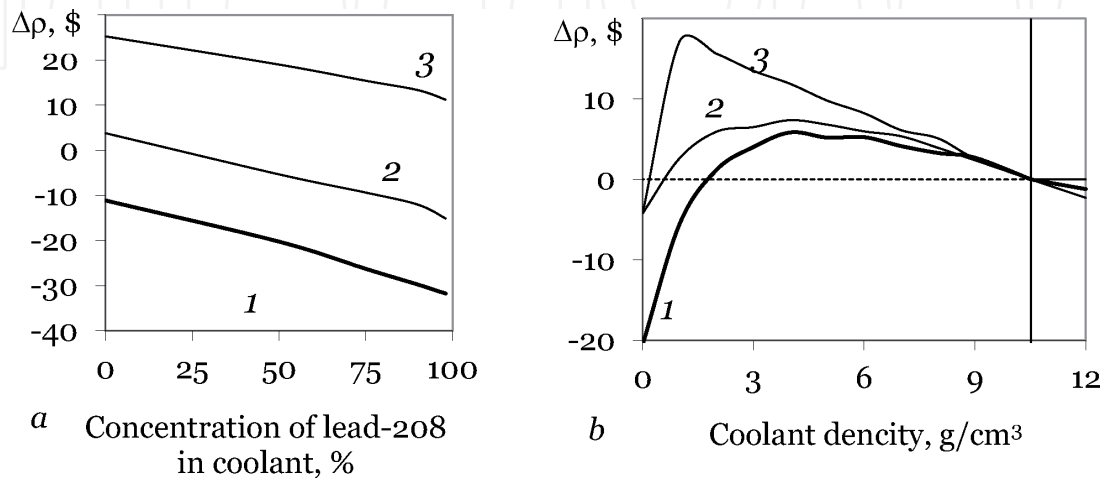


Figure 3.
Studies of the void and density effects of reactivity in a high power BREST reactor, (a) the dependence of VRE on the content of ^{208}Pb in the coolant is given, (b) the dependence of the effects of reactivity on the density of a coolant based on lead of polymetallic ores.

When using lead with a high concentration of ^{208}Pb as a coolant, the potential risk of reactivity accidents is reduced. The neutron absorption cross section of ^{208}Pb in the spectrum of the BREST reactor is 10^{-3} b ($1 \text{ b} = 10^{-28} \text{ m}^2$), that is, approximately three times lower than $^{\text{nat}}\text{Pb}$. The core of such a reactor is characterized by a large volume fraction of lead and a small fraction of fuel, which increases the role of the coolant in changing the neutron macroscopic absorption cross section Σ_a by core materials. When switching to thorium lead, the Σ_a decreases, which leads to an increase in the lifetime of prompt neutrons and limits the prompt supercritical when reactivity is introduced that is greater than β . When switching to uranium lead (^{206}Pb), the Σ_a increases (the neutron absorption cross section of ^{206}Pb is more than three times higher than $^{\text{nat}}\text{Pb}$); as a result, the potential danger of reactivity accidents (the reactor is prompt supercritical) increases.

One of the ways to increase the corrosion resistance of structural materials is the use of technological additives (inhibitors and deoxidants) to the coolant. The possible inhibitors are characterized by a high absorption cross section and quickly burn out in a neutron field, forming slugs in the coolant. All metallic impurities contained in liquid lead (except bismuth), and all metallic components that may be present in lead as a result of corrosion and erosion of structural materials, have a lower electrode potential than lead, i.e., these impurities play the role of deoxidizing agents. The deoxidizing agent may also be an alkali metal. Small additives of lithium or potassium form a eutectic with lead, reducing the freezing point of the coolant. Small sodium additives play the role of an alloying additive to lead, increasing the freezing point of the coolant. Lithium is the strongest reducing agent—the best getter of free oxygen. Experiments conducted at the JSC “SSC RF-IPPE” (Obninsk, Russia) show that the cardinal way of changing the initial oxidizing properties of heavy coolants is the use of additives of metal deoxidizing agents that can reduce the level of oxidative potential of the melt. Thus, a technological additive of 1.8% (wt.) potassium to lead allows the formation of a eutectic alloy with oxygen activity five orders of magnitude lower than that of a pure lead melt at temperatures of 500–550°C.

With a decrease in the mass number of nuclei, the role of elastic moderation increases, and the void and density effects of reactivity increase. The relatively high VRE with the use of a coolant based on the eutectic $\text{Pb}-^7\text{Li}$ alloy can be reduced by switching from natural lead to ^{208}Pb in the composition of this alloy (see **Figure 4**). However, in this case, VRE is significantly higher than when using lead polymetallic ores.

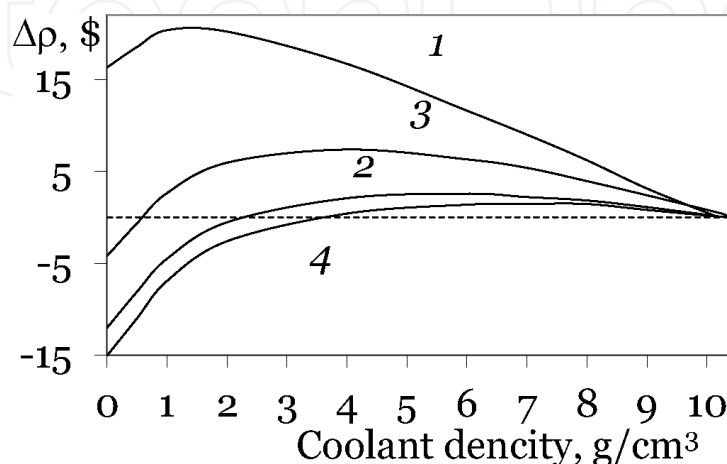


Figure 4.

Change in the reactivity effect $\Delta\rho$ depending on the density of the coolant in the core and the lower reflector during drainage of the BREST high-power reactor from the bottom up (involvement of bubbles in the core), (1) eutectic $^7\text{Li}-^{208}\text{Pb}$; (2) $^{\text{nat}}\text{Pb}$; (3) eutectic $^{\text{nat}}\text{K}-^{208}\text{Pb}$; (4) ^{208}Pb .

More detailed results of studies on the use of a coolant based on lead extracted from thorium ores are presented in [16, 35–38]. To ensure acceptable VRE in high-power reactors, it is not necessary to use isotopically pure lead-208. It is enough that the content of ^{208}Pb in the lead coolant is at least 75–80%. This makes it possible to use almost any thorium deposit for lead mining.

4. Construction materials

The greatest successes have been achieved in the field of creating new structural materials. For fast sodium reactors, specialists from Russia, the USA, Japan, China, France, and Ukraine consider radiation-resistant heat-resistant materials based on dispersion—hardened by thermally stable nanooxides Y_2O_3 , $\text{Y}_2\text{O}_3\text{-TiO}_2$ or Al_2O_3 (3–5 nm) ferritic-martensitic steels with good strength and mechanical properties capable of working at neutron fluences (with kinetic energies of more than 0.1 MeV) up to $2 \times 10^{16} \text{ cm}^{-2} \text{ s}^{-1}$ to damaging doses of 160–180 sleep at temperatures of 370–710°C (see [39–41] and links to these works).

While ensuring a good neutron balance in the core due to the use of innovative fuels, coolant, and structural materials, two advantages of additional absorbers in structural materials can be distinguished.

Firstly, the fast neutron absorption cross section of structural materials that cannot leave the core when it is drained favorably affects the development of the LOCA emergency: they contribute to the reduction of VRE. In the manufacture of structural materials, the content of alloying additives is regulated by regulatory documents. The content of alloying additives can vary in a given range. A negative factor may consist in a change in the sign of VRE depending on the content of alloying additives in the composition of structural steel (see **Figure 5**).

Secondly, the removal from the core of erosion and corrosion products (of structural materials) that strongly absorb neutrons helps to prolong the campaign of the reactor, increasing reactivity, similar to the action of burnable absorbers in thermal reactors (see **Figure 6**). The negative factor is a decrease in the thickness of the cladding of the fuel rods and slagging of the contour (increase in erosion rate) by corrosion and erosion products that have not settled on the “cold” section of the coolant circulation path. As a result, the processes of corrosion and erosion of structural steels contribute to solving the problem of minimizing the reactivity

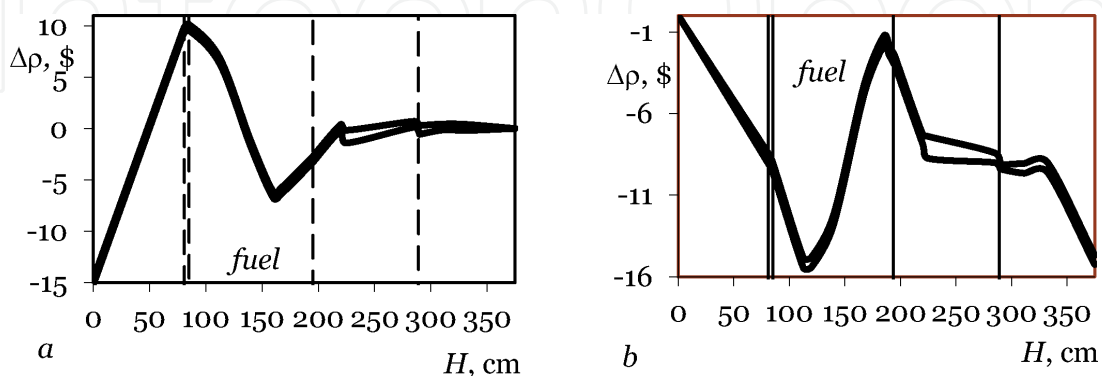


Figure 5. VRE in a BREST reactor of high power under conditions of uncertainty in the concentration of alloying additives in structural steel within the permissible (MCU). H—The boundary of the medium “lead-vacuum.” conservative assessment: Horizontal neutron leakage is equal to zero; 100% replacement of lead by vacuum are not taken into account, (a) the drainage boundary moves from top to bottom (lead discharge, in BREST the event is ruled out deterministically), (b) the drainage boundary moves from bottom to top (involvement of bubbles in the core during depressurization of the lead-water heat exchanger tubes).

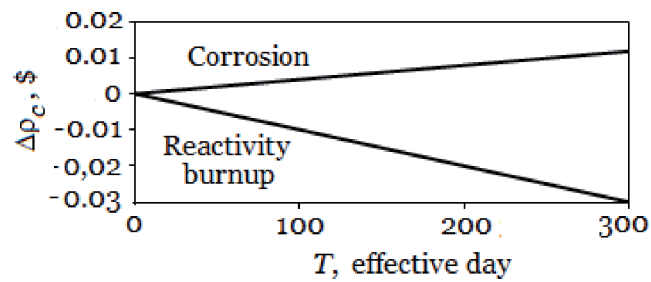


Figure 6.

Change in the reactivity $\Delta\rho_c$ for microcampaign T as a result of corrosion of the structural steels and the fuel burnup.

margin for fuel burnup. In BREST projects it is proposed to use steel without nickel but with increased silicon content [14]. Silicon promotes the formation of protective oxide films on the outer surfaces of cladding of fuel element.

The calculated analysis made it possible to conclude on the possibility of using tungsten coatings of fuel element cladding from the outside and inside. This will lead to the improvement of reliability and safety of BREST reactors without deterioration (and possibly with improvement) of economic characteristics of NPPs with such reactors [16, 42, 43]. The application of tungsten coverings will allow to reduce the corrosion rate and erosion in liquid lead and will lead to the deterioration in neutron balance (and perhaps, reactor costs); minimization will be promoted by the VRE (large absorption cross section of fast neutrons) and will open possibilities of use of the lead more polluted by impurity.

Tungsten does not react with nitride fuel at temperatures below 1485°C. This temperature is not achieved even in ATWS [14, 16]. Tungsten interacts weakly with lead. Conditions for the formation of lead (PbWO_4) tungsten in the reactor core are poor: insufficient oxygen, high-energy neutrons break chemical bonds.

Tungsten in combination with chromium improves mechanical properties of steel [44]. Due to the small (compared to iron) values of the cross section of neutron interaction with tungsten nuclei, accompanied by the yield of gaseous products (less than 10^{-2} b), and the high threshold of such reactions (10 MeV), tungsten does not swell and does not become brittle at high fluxes of fast neutrons.

Tungsten coatings produced by plasma spraying have a layered structure, do not crack, or peel even when bent [45]. Tungsten plasma spraying technology is well developed. The dense (nonporous) tungsten film completely covers the steel surface. Corrosion and erosion resistant coatings in a wide range of temperatures (higher than ATWS temperatures) in liquid metal melts, including under conditions of additional abrasive wear (practically excluded by tungsten coatings).

Like silicon, tungsten contributes to the formation of a protective oxide film on the surface of the shell (to a lesser extent because it holds oxygen slightly worse: bond energy $\text{SiO} = \text{Si} + \text{O}$ is 8.24 ± 0.10 eV, $\text{WO} = \text{W} + \text{O}$ is 6.94 ± 0.43 eV [46]). Due to the leakage of the circuit, oxygen will always be present in the lead. The oxide film (WO_2 , W_4O_{11} , WO_3) holds well (up to 923°C) and serves as an additional naturally formed protective coating.

The tungsten content of the sprayed layer is about 1%. As a result, the increase in the cost of the structural material may not be essential. Among the neutron-physical aspects of the use of tungsten-sprayed cladding, the following are noted. The deterioration of the neutron balance may not be essential when thin coatings are used. (At that, it is possible to reduce thickness of cladding).

The MCU code calculations showed that an increase in the proportion of tungsten in the structural material from 0 to 100% leads to a marked decrease in VRE: by 1.76 \$ with hypothetical drainage of the core and the lower end reflector and

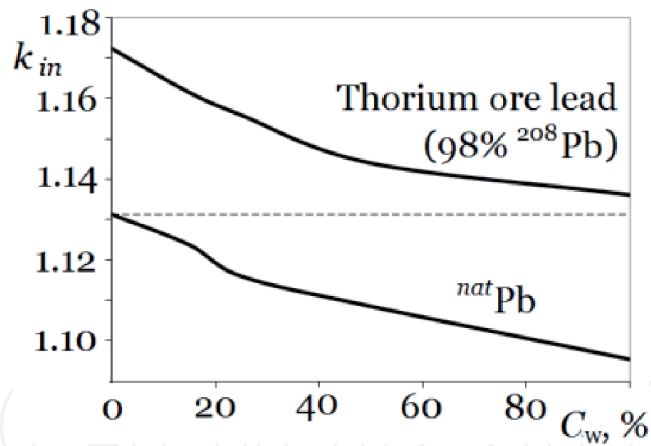


Figure 7.
 The dependence of k_{in} on C_w .

when 98% ^{208}Pb in the lead coolant. The most realistic and most dangerous scenario of such an emergency situation is not related to the complete drainage of the reactor but to the involvement of bubbles in the core during depressurization of the pipes of the “lead-steam (water)” heat exchanger. At that, reactivity effect is maximum at the reduction of lead density in the core and lower end reflector from 10.5 to 7.0 g/cm³ [35]. In such a scenario, the density effect of reactivity is \$0.06 when used as a structural material of corrosion-resistant steel and minus \$ 0.86 when switching to tungsten cladding. To obtain a conservative estimate, an infinite array of fuel elements is chosen, formally corresponding to the reactor of infinite power.

Figure 7 shows the dependence of the infinity multiplication factor k_{in} on the mass content of tungsten C_w in the structural steel of the BREST-OD-300 core. It is possible to fully compensate for neutron losses due to their absorption by tungsten nuclei by increasing the proportion of ^{208}Pb in the heat carrier even when using shells made entirely of tungsten.

The other way is connected with self-organizing coatings of internal surface of fuel element cladding. It is proposed to place liquid lead saturated with zirconium or liquid eutectic alloy 97.53% (wt.) Pb, 2.25% Mg, and 0.2% Zr [47]. When the reactor is operating, a protective coating based on zirconium nitride is spontaneously formed on the inner surface of the cladding. Self-healing of accidental damages of coating is provided.

5. Accident-tolerant fuel

5.1 General provisions

Fast reactors of the new generation are focused on the use of ceramic mixed uranium-plutonium fuel. The immediate prospects are associated with the use of mixed oxide (MOX). A more promising in terms of possible exceptions of severe accidents in NPP can be considered mixed mononitride uranium-plutonium fuel (MN fuel, mixed nitride). It is proposed to use in BREST reactors with lead cooled and, in the future, perhaps in power sodium-cooled reactors.

Nanotechnology, in relation to the creation of new materials for nuclear technology, is widely used since the early 1960s.

Thanks to the introduction of nanotechnology the unique fuel materials, characterized by high density and thermal conductivity, can be obtained. The task of creating high-performance nuclear fuel (mostly fast reactors), which is achievable for the high burnup, isolated two problems [39]:

- Creating a coarse structure with a given porosity (MOX with nanoadditives for BN-type reactors [40]), capable of holding gaseous and volatile corrosive fission products, preventing their migration along the grain boundaries to the cladding in order to slow down corrosion of the inner surface of the cladding.
- The activation process of sintering fuel pellets due to the nanoadditives [40].

The porosity of the ceramic nuclear fuel consisting of micrograins is approximately 25%. When making fuel from nanopowder, it is possible to significantly reduce porosity (up to 5–10%). For example, in order to increase the density of MN fuel to 95% of the theoretical density by reducing porosity, the inventors [48, 49] propose a fuel based on MN nanopowder. In terms of increasing reactor power, reducing porosity is equivalent to increasing the volume of fuel in the core or increasing the volume of the core.

It is known that, compared to ceramic fuel (MOX, MN, etc.), cermet fuel has some advantages. It is more attractive due to increased thermal conductivity and density, increased BRC (up to 1). The main disadvantage preventing the use of such a fuel is the reduction of the melting point. The use of a nanopowder of metallic uranium in conjunction with ceramic fuel micrograins will provide the same advantages, eliminating the disadvantage. Ideally, the pores between the micrograins can be filled with nanopowder. In this case, the melting point of the fuel will be determined by the melting point of the ceramic (MOX, MN, etc.). The nanopowder in the fuel composition can melt in emergency modes; there will be droplets of molten metal between the ceramic microloans, which will have practically no impact on the reactor safety. As a result, it is possible to obtain fuel having high thermal conductivity and density, which contributes to self-protection of the reactor. When using such fuel, the condition $BRC = 1$ is easily achievable.

At the beginning of the twenty-first century, in Russia fast reactors with sodium cooling was considered vibrocompacted MOX-fuel with uranium (up to 10% by weight) getter of free oxygen [19, 20]. Since it is difficult to control the uniformity of mixing MOX and uranium metal powders in a relatively large volume of fuel element (in case of vibration compaction), it makes sense to switch to small volumes (pellet fuel) [50, 51].

The finer the uranium powder particles, the better it exhibits getter properties. Nanodisperse powder is one of the best free oxygen getter. The ideal getter is a powder ground to size when any atom in the nanoparticles can be considered surface. (The best getter is thorium [46]. However, its use will require a reorientation of Russian nuclear fuel cycle enterprises, which is not economically feasible).

5.2 ATF based on ceramics and beryllium (a new look at the old concept)

The free oxygen getter may be a material having a high chemical affinity for oxygen (see **Table 4**). The first steps towards solving the problem of free oxygen binding were taken in the late 1950s and early 1960s [52]. Beryllium (and Be_2O) can also serve as a getter. The possibility of using oxide fuel with beryllium additives was considered at the dawn of the development of fast reactor technologies [4]. This was abandoned due to the decrease in BR.

Homogeneous placement of metal beryllium in MOX or MN fuel helps to solve the problem of corrosion of the inner surface of cladding and increase self-protection [51]. The spectral component of the VRE for an endless reactor with MN fuel and MN-5%Be fuel is 23.6 and 12.7, respectively [51]. The Doppler reactivity coefficient for infinite array of fuel elements is -7.99×10^{-6} and $-2.06 \times 10^{-5} \text{ K}^{-1}$, respectively.

Getter (bold type)	<i>E</i> , eV	Getter (bold type)	<i>E</i> , eV
ThO = Th + O	8.59 ± 0.22	TiO = Ti + O	6.85 ± 0.09
TaO = Ta + O	8.37 ± 0.43	ZrO ₂ = ZrO + O	6.68 ± 0.48
ThO ₂ = ThO + O	7.89 ± 0.30	VO ₂ = VO + O	6.51 ± 0.30
UO = U + O	7.81 ± 0.17	UO ₃ = UO2 + O	6.46 ± 0.30
ZrO = Zr + O	7.81 ± 0.43	VO = V + O	6.29 ± 0.43
YO = Y + O	7.37 ± 0.13	Be ₂ O ₂ = Be2O + O	5.94 ± 0.65
UO ₂ = UO + O	7.37 ± 0.30	BeO = Be + O	4.60 ± 0.13

Table 4.
Chemical bond energy E for some getter and oxides formed (as reported by [46]).

The increase in the thermal conductivity of the fuel (due to beryllium) leads to a decrease in the temperature of the fuel. As a result, the nature of the change in maximum fuel temperatures in medium or high power reactors (BN-800, 1200, 1800) in LOF WS and TOP WS is same. To increase self-protection against these accidents, a large absolute negative Doppler reactivity coefficient is required.

Additives of beryllium to MOX fuel contribute to the minimization of corrosion rate of fuel element shells from inside, the reduction of VRE, the elimination of known contradiction in requirement for Doppler reactivity coefficient in emergency modes LOF WS and TOP WS in high-power reactors, and the improvement of reactor self-protection. At the same time, beryllium supplements reduce BRC and BR. It's a major flaw.

5.3 ATF based on MOX and uranium nanopowder

For MOX-fueled reactors, it is possible to offer pellet fuel based on fine-grained MOX and nanopowder U. Fuel density (at 18% U) under normal conditions of about 12 g/cm³, thermal conductivity—12 W m⁻¹ K⁻¹. Increasing the density and thermal conductivity of MOX-U fuel with increasing U fraction leads to changing the role of Doppler reactivity coefficient in providing self-protection against accidents of LOF WS type. As a result, the role of Doppler reactivity coefficient in providing self-protection against accidents of type TOP WS and LOF WS becomes the same (or almost the same), which helps to resolve the conflict situation when optimizing the core layout. This makes it possible to significantly increase reactor safety.

Figure 8 shows the results of modeling LOF WS mode (FRISS-2D code) in LMFR using different fuels. The relationship between maximum temperature of fuel and heat carrier is presented when using pellet MOX fuel in BN-800 (a), MN fuel in reactor of type BREST-OD-300 [14] (b), and BN-800 (c). The thin line corresponds to the conservative approximation: constant coolant temperature at the core inlet in emergency mode. **Figure 8(d)** shows the time dependence of the maximum temperature of the MOX-based pellet fuel and uranium nanopowder in the TOP WS and LOF WS modes and when they are superimposed (dotted line in **Figure 8(d)**). For correct comparison of variants at use of different fuel, the same fuel assemblies and grids of the core BN-800 [53] are accepted.

LOF WS is initiated by de-energizing of all main circulation pumps of the primary circuit at coastdown time 30 s (for BN-800) or by reduction of flow rate in 20 s (for BREST-300). TOP WS is initiated by reactivity input \$0.3 for 10 s.

In order to increase the self-protection of reactors against the TOP WS accident, it is necessary to increase the negative Doppler reactivity coefficient by absolute value. In LOF WS mode, when MOX fuel is used, the maximum fuel

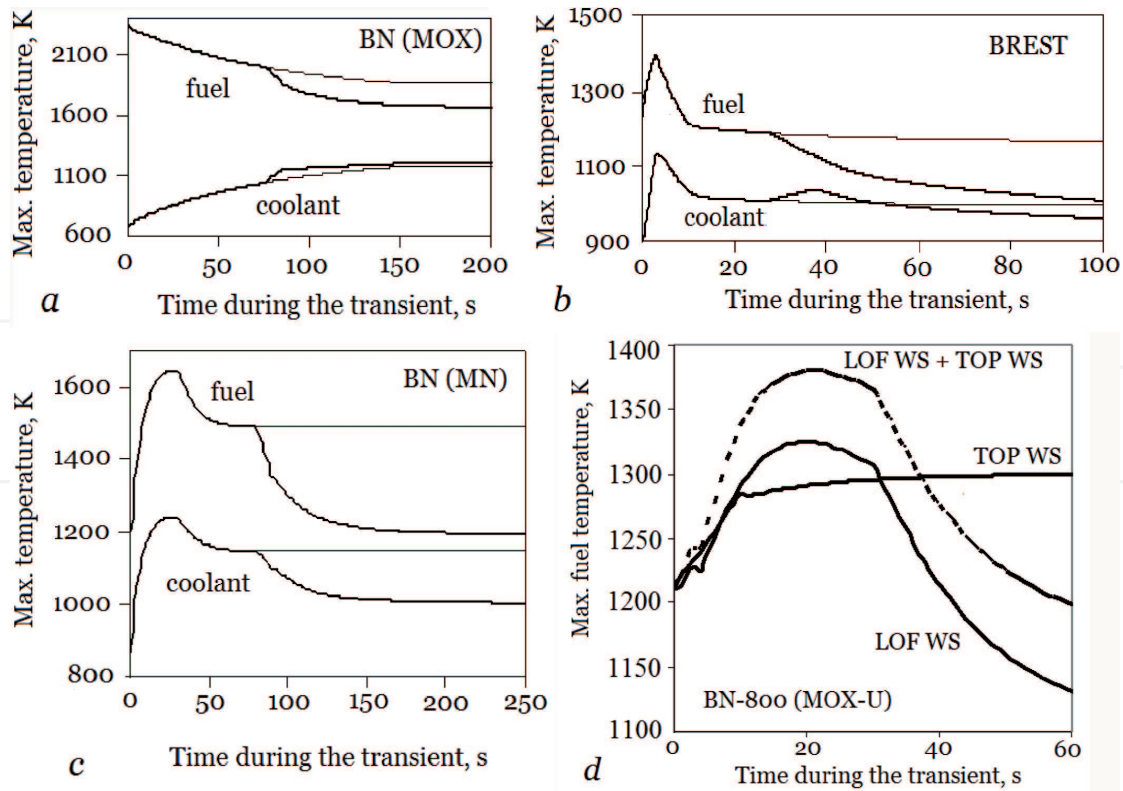


Figure 8.

Time dependence of pellet fuel temperature in LOF WS mode for BN-800 reactors with MOX fuel (a), BPECT-300 (b), and БН-800 (c) with MN fuel, BN-800 with MOX-U fuel (d).

temperature decreases with time. At the same time, it is possible to start boiling sodium. In order to increase self-protection, it is necessary to modulate the negative Doppler reactivity coefficient. The use of vibration-compressed MOX fuel with uranium getter [19, 20] does not significantly change these dependencies, and the role of Doppler reactivity coefficient in TOP WS and LOF WS modes remains the opposite.

When using MN fuel in LOF WS mode, an intermediate scenario between metal and oxide fuel is implemented. To minimize maximum temperatures, the negative Doppler reactivity coefficient must be increased modularly, as well as to ensure self-protection against TOP WS accidents. If the maximum fuel temperature is reduced to less than the nominal value, it is necessary to modulate the negative Doppler coefficient, but due to significant temperature reserves in this case, the role of Doppler coefficient has no determining value. This is the pattern (**Figure 1(d)**) observed in the emergency mode LOF WS when using a tablet fuel based on fine-grained MOX and nanopowder U (18% wt.). At its low U content, the scenario of emergency mode development is the same as when using tablet or vibrocompressed MOX fuel with finely dispersed (100 μm) U-getter. At high U content (up to 50% by weight), the nature of the change in maximum fuel temperature over time in the LOF WS mode is similar to that shown in **Figure 1(b)**. At mass content of uranium nanopowder, 18% fuel is slightly lower than the tablet MN in density and 1.5 times in thermal conductivity. As a result, when switching to MOX-U, the maximum fuel temperature is significantly lower than when using a tablet MOX fuel and approximately the same as the MN fuel (with larger fuel pellet diameter). The nature of the change of maximum temperatures in emergency modes TOP WS and LOF WS in fast reactors of medium and high power with MN fuel and fuel based on fine-grained MOX and nanopowder U are the same. In both cases $\text{BRC} = 1$.

The LOHS WS and OVC WS are not dangerous.

The temperature of MOX-U fuel even in ATWS is lower than the oxide when the reactor operates at rated power. This helps to reduce oxygen release from the Pu_4O_7 in nominal and emergency modes of reactor operation.

When the temperature increases (in emergency modes), the MOX-U fuel can contain nanodroplets of liquid metal between MOX grains. By neutron balance this fuel is closer to the pellet monoxide or MN, by the melting point—closer to UO_2 - Pu_4O_7 . So, when using nanotechnologies in the manufacture, MOX-U-fuel opens up large “reserves,” approaching it in properties to MN.

5.4 ATF based on MN and uranium nanopowder

Further the improvement of fuel characteristics while maintaining high melting temperature is possible at transition to fuel based on MN nanopowder (100–500 nm). To achieve a density of 85–95% theoretical, the required sintering temperature 1800–1900°C and a time of 10–30 hours. At the same time, up to 15% plutonium is evaporated from the fuel [48, 49]. When using such fuel, the nature of time dependence of the maximum temperatures in emergency modes does not change qualitatively.

The best and cheaper alternative is fuel based on micrograins MN and nanopowder U. The beginning of MN decomposition by 450–500°C exceeds the sintering temperature of fuel tablets during their manufacture. Significant improvement in reactor performance and safety can be expected with MN-U fuel due to high density and thermal conductivity, equal role of Doppler reactivity coefficient in TOP WS and LOF WS, and neutron balance improvement ($\text{BRC} = 1$). By neutron balance and nature of emergency modes, MN-U fuel is close to U-Zr and U-Pu-Zr [11] but much higher temperature.

The most relevant is the possibility of using fuel based on fine-grained MN and nanopowder U in lead-cooled reactors with a lead layer between the fuel and the cladding. The chemical bond energy in UN and PuN is markedly lower than in oxides (for UN, 5.464 ± 0.217 eV; PuN – 4.857 ± 0.651 eV; UO – 7.805 ± 0.174 eV; $\text{UO}_2 = \text{UO} + \text{O} - 7372 \pm 0.304$ eV; PuO – 7.459 ± 0.217 eV [46]). Although the operating temperature of MN fuel is much lower than MOX, nitrogen can exit MN. The absorption of nitrogen from MN fuel by metal uranium can slow corrosion of internal surface of cladding.

6. Conclusions

There is potential to further improve LMFR reliability and safety within the existing technologies.

In order to ensure acceptable VRE in lead-cooled LMFR of high power, the minimum content of isotope ^{208}Pb in the lead heat carrier should be about 75–80%. This is typical of thorium ores.

The use of tungsten coatings of fuel element cladding helps to reduce VRE, eliminate reaction of chromium with steam in emergency situations, and exclude hypothetical possibility of lead azide formation. With the simultaneous use of tungsten coatings and heat carrier based on lead extracted from thorium ores, it is possible to ensure a good balance of neutrons in the core.

The potential and reserves of MOX fuel in ensuring the safety of large LMFR with sodium or lead coolant have not yet been exhausted. Pellet fuel based on fine-grained MOX and nanoscale U has no advantage over pellet MN fuel.

The prospects for LMFR development may be related to the use of fuel based on fine-grained MN and nano-sized metal uranium powder.

Uranium nanopowder additives will not only increase the density and thermal conductivity of nuclear fuel, which is important in the problem of reactor safety,

but also significantly improve the neutron balance (by directing “excess” neutrons to ensure safety: achievement of condition $BRC = 1$ even on oxide fuel), almost completely eliminate the conflict character in the requirement for Doppler reactivity coefficient in terms of self-protection against ATWS in large LMFR, which opens up wide possibilities for the optimization of core layout without deterioration of safety.

The complex use of the proposed innovations allows to ensure the safety of large LMFR. So, pellet MN fuel with nanopowder of metallic uranium, a coolant based on lead extracted from thorium ores, and steel claddings of fuel elements with a tungsten coating together can significantly increase the safety of the reactor. This is accident-tolerant materials for LMFR.

The results presented can be easily summarized into other types of reactors. The research can be useful in the development of new-generation reactors and the improvement of technical solutions to existing nuclear reactors.

Conflict of interest

The author declares that he has no conflict of interest.

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