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Prospects of Using Liquid Metal Coolants in Fast Reactors.

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ABSTRACT

Results of using sodium and lead-bismuth coolants in nuclear reactors of various applications were studied. Both positive and negative properties of these coolants are presented, as well as the results of development of engineering solutions aimed at the prevention of the impact of negative properties of the above two coolants and also lead coolant, whose characteristics are very close to those of lead-bismuth. Potential of further application of sodium, lead and lead-bismuth coolants for fast reactors was confirmed on condition of the availability of technical measures and devices designed for suppression of their negative properties impact. As regards some new coolants proposed for use, such as Na-Pb (instead of Na) and $Pb_{0.83}Li_{0.17}$, $Pb_{0.91}K_{0.09}$, and $Pb_{0.83}Mg_{0.17}$ eutectics (instead of Pb and Pb-Bi), these pose heavy, now insuperable, problems. The revealed problems are mainly caused by the fact that all these proposed liquid metals tend to interact intensively with water, steam and oxygen. However, it is impossible to reliably avoid such interactions taking into account existing reactor technologies and requirement to make both scheduled and emergency repairs and replacements of the reactor components. Besides, Li, K and Mg present in these eutectics are transformed into the oxides or alkalis, which may dissolve in the coolant or may be removed, thus causing significant changes in coolant composition and thermodynamics. The situation becomes even more difficult because the emerging oxides are very strong thermodynamically and cannot be regenerated under reactor conditions (in contrast to the oxides of traditional coolants, such as Pb and Pb-Bi, which are easily reduced by hydrogen). Therefore, the newly formed oxides may impact on hydraulic characteristics of the coolant circuit elements and heat transfer conditions in the circuits, causing irreversible changes in the reactor performance and safety characteristics. Thus, the declared advantages of the liquid metals proposed as advanced coolants have not been confirmed. So, there are no grounds now for acknowledging these liquid metals application as an advanced trend of development of fast reactor technology.

Keywords: nuclear reactor, liquid metal coolants, application prospects, safety, circuit, corrosion, impurities.

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INTRODUCTION

The share of power plants using non-carbonaceous fuel, including headmost nuclear power plants, in the global power industry is ever increasing (Rachkov and Kalyakin, 2014). In nuclear power area, the role of fast neutron reactors becomes noticeable (Poplavsky, 1999, Bagdasarov et al., 1999, Adamov and Orlov, 1999, Poplavsky, 2004, Oshkanov et al., 2004, Kozub and Sergeyev, 2008). In all fast reactors either in operation, or under design, liquid metal coolants, namely: sodium, lead or lead-bismuth eutectic are applied. These coolants (though, in common with all coolants known by now) are not ideal from the standpoint of their use under reactor conditions. For instance, sodium having many positive properties (low density and melting point, high thermal conductivity and thermal capacity, low corrosiveness, etc.) is characterized by very high chemical activity and relatively low boiling temperature. In this view, assurance of sodium cooled reactor safety requires technical and organizational measures preventing sodium negative properties impact on the reactor (Poplavsky, 2004, Kozub and Sergeyev, 2008, Bagdasarov et al., 2001, Poplavsky and Kozlov, 1990). Lead and lead-bismuth coolants are not so chemically active, they melt at relatively low temperatures and their boiling points are high. However, because of their high corrosiveness, these coolants can only be used on condition that concentration of oxygen dissolved in the coolant is maintained within strictly determined range. So, safe operation of reactors with these coolants requires design approaches aimed at the maintaining of the appropriate oxygen concentration in the coolant (Gromov et al., 1992, Beznosov et al., 2007, Gulevsky et al., 2008, Gromov et al., 1999). For the purpose of improving reactor performance and simplifying conditions of the reactor safe operation, there are many proposals on the use of various modified eutectics of the above traditional liquid metal coolants (Subbotin et al., 2002, Zasorin et al., 2008, Kuz'min and Okunev, 2000, Shimkevich and Shimkevich, 2014, Alexeyev et al., 1991, Alekseyev et al., 2015). Below presented is the background of such proposals.

Physical, chemical and technological properties of sodium are different from those of lead and lead-bismuth eutectic because of different charge polarity of these metals: sodium is electropositive element, while lead and lead-bismuth are electronegative elements. In chemical reactions, sodium being strong reducing agent donates electrons, and lead and led-bismuth, as oxidizing agents, take electrons. A combination of these opposites would give the new coolant, in which the components could complement each other fulfilling different technological functions: alkali metal would decrease corrosiveness of the coolant, while heavy metal would decrease its chemical activity. Boiling point of the coolant would be high owing to heavy metal additive. Besides, considerable decrease of its corrosiveness is assumed. It is proposed to replace heavy liquid metal coolants with the following eutectics: $Pb_{0.83}Li_{0.17}$, $Pb_{0.91}K_{0.09}$, and $Pb_{0.83}Mg_{0.17}$ having melting temperatures equal, respectively, to 230^oC, 272^oC and 248^oC. Liquid Na-Pb modifications with various heavy liquid metal contents were considered as alternative to sodium coolant (Shimkevich and Shimkevich, 2014, Alexeyev et al., 1991, Alekseyev et al., 2015). Furthermore, introduction of the modified eutectics of liquid metal coolants is proposed to consider as advanced direction of development of fast reactor technology.

In our opinion, the above proposals are, apparently, of interest from the standpoint of revealing fundamental properties of multi-component liquid metals and their useful application in various areas of science and technology. However, the possibility of practical use of modified liquid metal coolants under conditions of commercial reactors and, hence, acknowledgement of this modification as potential trend of fast reactor technology development is in a very big doubt. The arguments for this doubt are as follows.

Vast experience gained in operating reactors and various test facilities with both static and flowing sodium, lead and lead-bismuth eutectic coolants confirming the possibility of the long-term safe operation of nuclear reactors with the above coolants.

Results of development of advanced fast reactor designs and both analytical and experimental studies carried out in support of these designs, being demonstrative ground for the possibility of application of technical measures and devices, which would reliably prevent impact by the negative properties of sodium, lead and lead-bismuth eutectic coolants. Experimental results (mainly, negative) of studies on the possibility of application of the proposed modified coolants in nuclear reactors, and thermonuclear and other systems.

Present-day perceptions of fast reactors updating taking into account the absence in the foreseeable future of the possibilities of reliable prevention of liquid metal interaction with water, steam and air oxygen in the stages of reactor creation and normal operation, during repairs and scheduled replacements of the components, as well as under accidental conditions; in case of interaction, alkaline components of the liquid metals should oxidize with formation of the oxides and alkalis causing changes of the liquid metal composition, thermodynamic and other characteristics (Alabyshev et al., 1958, Hultgren et al., 1963).

Use of sodium, lead and lead-bismuth eutectic coolants

Sodium, lead and lead-bismuth eutectic were chosen as fast reactor coolants on the basis of the results of studies carried out for many years on their physical, chemical, and nuclear characteristics, proven natural reserves and production prospects, as well as of the overview of design approaches and comparison of the operating experience data from nuclear reactors and experimental facilities with various liquid metal coolants including the above coolants, almost all alkali metals, as well as mercury, gallium, and sodium-potassium, lead-magnesium, lead-lithium and other alloys (Rachkov and Kalyakin, 2014, Poplavsky, 1999, Bagdasarov et al., 1999, Adamov and Orlov, 1999, Poplavsky, 2004, Oshkanov et al., 2004, Kozub and Sergeyev, 2008, Rachkov et al., 2014, Gromov et al., 1992, Beznosov et al., 2007, Subbotin et al., 2002).

Practical assimilation of sodium as fast reactor coolant began in the early 1950-ies, when EBR-1 reactor in the USA and BR-1 and BR-2 reactors in the USSR were constructed. Then, in many countries, work was initiated on creation of experimental facilities simulating the main systems of the future NPP's, demo and commercial fast reactors (Poplavsky, 1999, Bagdasarov, 1999, Adamov and Orlov, 1999). Currently, Russia is the undoubted world-wide leader in the development of fast reactors with sodium coolant and the only country all over the world operating power fast reactors with sodium coolant (sodium cooled BN-600 and BN-800 reactors i.e., respectively, the third and the fourth power units of Beloyarsk NPP). Design of the larger size series sodium cooled fast reactor BN-1200 is under development. Construction of the first BN-1200 reactor is planned for the 5-th power unit of Beloyarsk NPP. Results of work performed on practical assimilation of sodium coolant confirmed its excellent thermal properties and good compatibility with many structural materials. It was shown that sodium could be practically ideal coolant if it was not for its high chemical activity with respect to the air oxygen and water, as well as relatively low boiling temperature. So, the assurance of sodium cooled reactor safety requires the use of some technical and organizational solutions preventing its negative impact (in addition to those, which are common for the majority of reactors with various coolants). Such solutions, implemented mainly in the BN-800 reactor and completely in the BN-1200 reactor design, are as follows (Poplavsky, 1999, Bagdasarov et al., 1999, Poplavsky, 2004, Oshkanov et al., 2004, Kozub and Sergeyev, 2008, Kashcheyev and Ashurko, 2014, Bagdasarov, 2001, Poplavsky and Kozlov, 1990):

- Three-circuit mono-bloc arrangement with the whole radioactive primary system having pressure lower than that in the non-radioactive secondary circuit, located in the reactor vessel, thus practically eliminating the possibility of radioactive sodium leaks and fires;
- Safety jackets for the secondary (non-radioactive) sodium circuit in order to decrease the probability and size of non-radioactive sodium leaks;
- Use of the cold traps as the simple method of maintaining required sodium purity level in order to practically avoid structural steel corrosion caused by sodium impurities;
- Special design of automatic protection system of the steam generators and its algorithm aimed at minimization of steam generator damage rate in case of water leak into the non-radioactive secondary sodium;
- Effective fire extinguishing system.

The use of lead and lead-bismuth eutectic in nuclear power technology was originated by the work on creation of reactors for naval construction needs (Rachkov et al., 2014, Gromov et al., 1992, Beznosov et al., 2007). For this purpose, properties of lead and lead-bismuth eutectic as potential coolants of nuclear power reactors for strategic submarines were studied along with those of sodium. Sodium was recognized inadequate for use under naval conditions for fire and explosion hazard reasons. As a result, several generations of ship reactors (on-ground prototypes, experimental and series reactors) got lead-bismuth coolant. Experience gained in creating and operating ship nuclear power plants turned most useful in the further developments of civil reactors (Rachkov et al., 2014, Gromov et al., 1992, Beznosov et al., 2007, Gulevsky et al., 2008). Currently, design of the small-size SVBR-100 reactor with lead-bismuth coolant is under development. This reactor is a standard module, and several SVBR-100 modular reactors can be installed on one site in order to achieve the required power of nuclear power plant. Dimensions of the reactor vessel meet the requirements of the railroad freight activity. By now, there is already a call for such small size power units. Demand for these reactors, as well as the number of their application areas are tremendous: from various prosperity countries to some enterprises being in need of power plants for various technologies including petro-chemistry, water desalination, industrial waste reprocessing, etc.

Design of the Russian reactor BREST-OD-300 with lead coolant nears its completion. Along with SVBR-100 reactor, BREST-300 belongs to the fourth generation reactors being the inherent safety systems. It means that owing to the approaches used in these reactor designs, the severe accidents are in principle improbable (Adamov and Orlov, 1999, Beznosov et al., 2007). Corrosiveness of the coolants against the structural steels is prevented by the steel passivation by oxygen, i.e. formation and maintenance of thin (1-10 µm thick), high density oxide films reliably bonded to the base on the steel surface. Such films cause significant increase of steel corrosion resistance. In order to provide conditions for protective oxide coating formation and their integrity control, certain value of oxygen thermodynamic activity in the coolant should be maintained during reactor operation. SSC RF – IPPE specialists have developed method and hardware for controlling oxygen thermodynamic activity in lead and lead-bismuth eutectic coolants based on solid phase lead oxide solution. This oxide is placed in the reaction vessel with the coolant flowing around (Beznosov et al., 2007, Gromov et al., 1999). Apart from the control of oxygen thermodynamic activity in the coolants, the following methods of coolant quality control have been developed (Gulevsky et al., 2008, Gromov et al., 1999):

- Periodic hydrogen purification of coolant circuits from slags based on coolant components oxides;
- Continuous coolant filtering for removal of impurities based on structural materials corrosion products.

The results of studies carried out for many years on substantiation of designs of facilities for the above methods implementation, as well as continued (over 50 th. hours) corrosion tests of reactor structural steels using the above methods, have shown the definitive effectiveness of these methods in preventing the impact of negative properties of lead and lead-bismuth eutectic coolants under reactor conditions.

Some properties of liquid metals proposed as fast reactor coolants

In the course of studies on physical, chemical and thermohydraulic properties of proposed liquid metals, some of their drawbacks were revealed, which could arise in case of using these liquid metals as coolants of the advanced fast reactors. In order to demonstrate these revealed properties, below presented are the results of studies carried out at the State Technical University, Nizhniy Novgorod and the Institute for Physics and Power Engineering, Obninsk.

In Nizhniy Novgorod, (Semyonov et al., 2006) studies were conducted on the experimental facility of State Technical University (Figure 1) with $Pb_{0.83}Li_{0.17}$ eutectic loop. Heat transfer to the eutectic and its corrosiveness were studied.

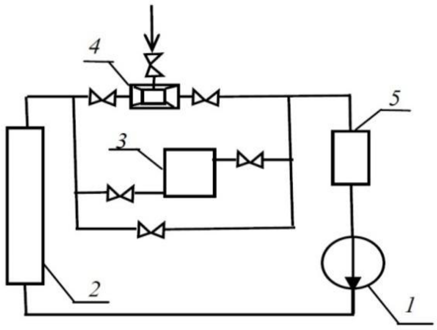
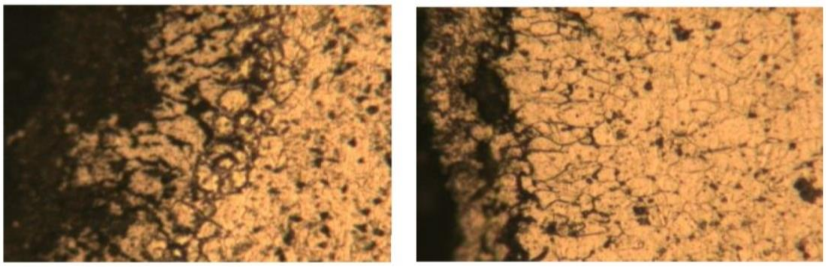


Figure 1: Test facility flow diagram

1-pump; 2-experimental section for heat transfer study; 3-measuring tank; 4-ejector; 5-heat exchanger

$Pb_{0.83}Li_{0.17}$ eutectic was circulating in the test facility during ~300 hours and was held under static conditions during ~100 hours within 450 - 520ºC temperature range. Studies revealed that heat transfer characteristics of this liquid metal were close to those of lead. Besides, practically not-reversible accumulation of slags that plugged circuit elements and filters was detected. Apparently, the slags were formed because of gas release from the circuit structural materials as a result of their temperature increase, as well as air ingress into the circuit in case of its transient opening (although excess inert cover gas pressure was maintained in the circuit in case of its opening).

Upon eutectic discharge, two samples of piping material were taken from the sections located upstream the ejector (sample 1) and downstream the ejector (sample 2) for the examination. Examination of these samples removed from the test facility showed corrosion damages of near-surface metal layer caused mainly by the inter-crystalline corrosion followed by detachment of crystals and their formations (see Figure 2). The depth of damage penetration varied from a few to 10 µm.



a) b)

Figure 2: Pictures of the piping inner surface sample 1 (a) and sample 2 (b). 630-fold enlargement

One of the conclusions made on the basis of studies results (Semyonov et al., 2006) was recommendation to provide mechanical filter on the main or by-pass pipeline of the circuit for removal of impurities particles from the eutectic during circuit operation. Since, in contrast to lead and lead-bismuth coolants, filter cannot be effectively cleaned from accumulated impurities by hydrogen reduction in order to restore its permeability, periodical replacement of the filtering element or the filter as a whole should be provided. The other research program was carried out at the Institute for Physics and Power Engineering in Obninsk within the framework of the Agreement with the Russian Ministry of Education and Science (Unique Identifier of applied scientific research (project) RFMEFI62514X0002). Behavior of $Pb_{0.91}K_{0.09}$ eutectic was studied in case of its interaction with water steam, as well as the effect of water leak into Na and Na-Pb on the steam generator components.

Studies on $Pb_{0.91}K_{0.09}$ eutectic behavior caused by its interaction with water steam

These studies were carried out in test facility (its flow diagram - see Figure 3).

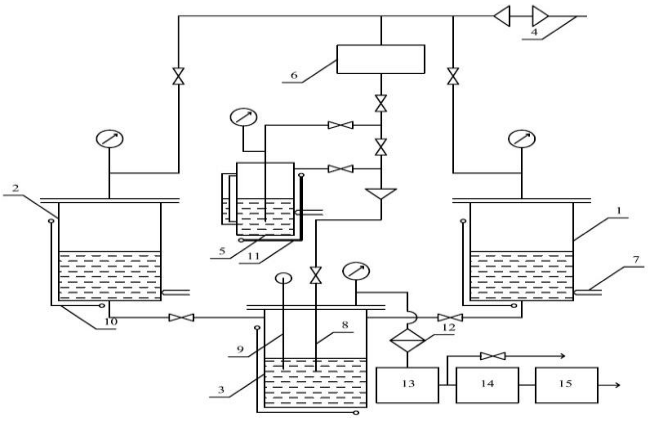


Figure 3: Flow diagram of test facility

1 – potassium tank; 2 – lead tank; 3 – Pb-K alloy tank; 4 – gas-vacuum system; 5 – steam generator; 6 – gas flow meter; 7 – thermocouple; 8 – sparge tube; 9 – sampling ampoule; 10 – nichrome electric heaters; 11 – fine control electric heater SNOL; 12 – filter; 13 – vapor condenser; 14 – gas counter; 15 – chromatograph.

The facility included storage tanks for Pb and K, as well as the tank for $Pb_{0.91}K_{0.09}$ alloy preparation equipped with the sparge tube for water steam supply to the eutectic and movable ampoule for eutectic sampling. Elaborately vacuumed tank was filled with potassium in the purified argon atmosphere in the glove box. Mass of the loaded potassium was determined by measuring tank mass before and after loading. After that, the amount of lead required for obtaining eutectic was evaluated, and the lead was loaded into the second tank. Then tanks containing K and Pb, and the empty tank for $Pb_{0.91}K_{0.09}$ were put under vacuum and heated, respectively, up to ~150°C, ~400°C, and ~500°C, and after that the total amounts of potassium and lead were transfused from their tanks to the third tank and intermixed by bubbling purified argon gas through the liquid metal at a rate of ~4 L/hour during 16 hours. Liquid metal sample was taken and its chemical analysis confirmed $Pb_{0.91}K_{0.09}$ eutectic formation, and then water steam supply to the eutectic started. Steam at ~500°C temperature was supplied from the steam generating device, i.e. heated bubble flask with water (located in the electric cabinet SNOL-3.5, 3, 5.3, 5/3, 5-I4M), through which purified argon flow was maintained at the variable flow rate.

The amount of water steam added to $Pb_{0.91}K_{0.09}$ alloy was measured using gage glass of the water steam generator. The experiment proceeded for about 27 hours. Approximately 277 g of water steam was supplied to the eutectic. The total water amount revealed in the condenser at the outlet of Pb-K tank was about 123 g. The remaining water interacted with the liquid metal by the following most probable reaction:

$$H_2O + K = KOH + 1/2H_2. \qquad (2)$$

This process finished in about 15 hours.

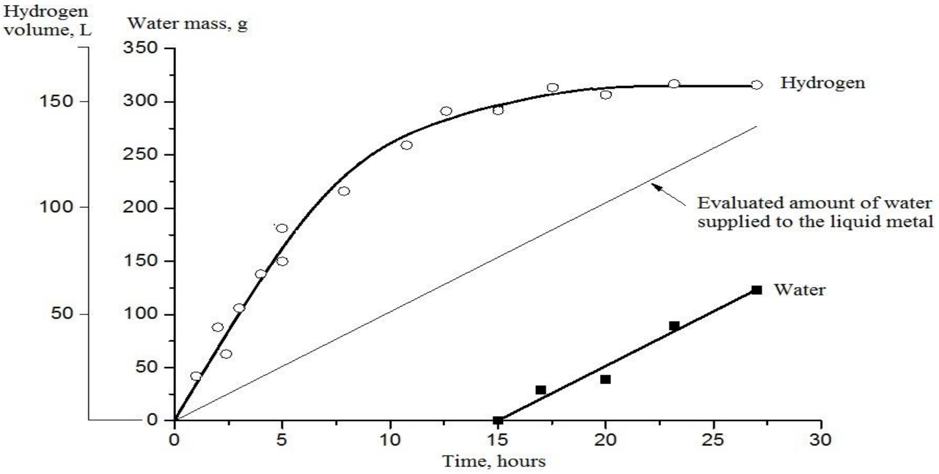


Figure 4: Amounts of released hydrogen and condensed water

This was confirmed by the fact of hydrogen release from the tank with Pb-K alloy (see Figure 4), which was detected by the chromatograph and evaluated by drawing up material balance: initial K mass in the alloy was ~544 g and amount of released hydrogen was ~158 L. According to the reaction (2) stoichiometry, the amount of released H_2 should be equal to 156 L, i.e. analytical and experimental results agreed within ~1%. The fact of complete conversion of K into KOH was confirmed as well by chemical analysis of the sample taken upon the end of the experiment. No K metal was revealed in the sample. After the tank was cooled down and opened, KOH traces were found on the liquid metal surface and on the inner surface of the tank wall. Appreciable amount of KOH (10 g) was only revealed in the filter. Small amount of KOH found outside of the liquid metal spoke for its good solubility in Pb-K.

During the experiment and upon its completion, condensed water samples were examined for pH value. The major amount of water (the first portion of ~100 g) seemed translucent, and its pH was equal to 7.3. Water in the last portion (23 g) was white and opaque with pH=12. Apparently, by the end of the experiment, $KOH{\cdot}nH_2O$ was formed and released from the liquid metal.

Studies on the effect of water leaks to Na and Na-Pb on the steam generator components

These studies were carried out in the facility shown in Figure 5. Facility includes vessel containing Na or Na-Pb (Pb ~ 2 at.%) (1), water injection device (2), tested material sample (3), and pump (4). Injection device (2) and sample (3) are imbedded into the tube (5). Water supply

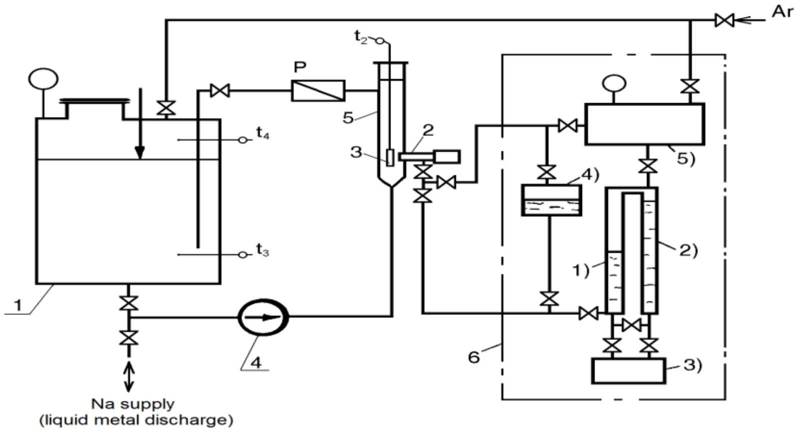


Figure 5: Test facility for studies on the effect of water leak on the growth of the steam generator initial defect

1 – vessel with liquid metal; 2 – water injection device for defect simulation; 3 – sample; 4 – pump; 5 – tube; 6 – water supply system: 6.1) – water flow meter tube; 6.2) – tube with constant water level; 6.3) – differential pressure gauge; 6.4) – vessel for water; 6.5) – gas vessel with pressure gauge.

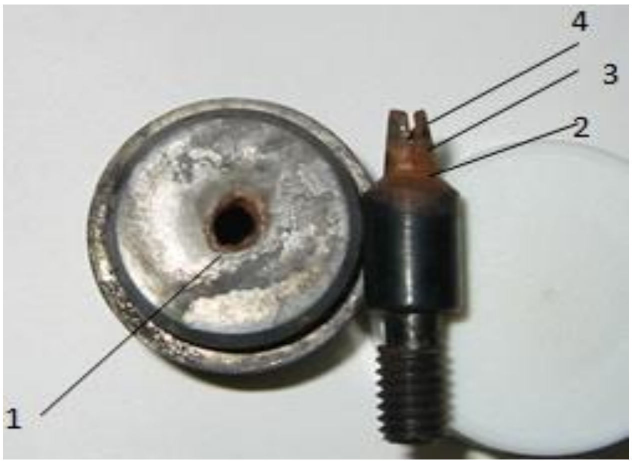


Figure 6: Picture of seat and valve of injection device upon test completion

1 – defect-side point on the seat end; 2 – initial defect channel on the valve; 3 – shut girth border facing seat inner surface; 4 – slot condition after test.

And its flow measurement system (6) is based on the monitoring of the water level change rate in the flow meter tube (6.1) relative to the water level in the reference tube (6.2). Water level change is measured by the differential pressure gauge (6.3). Tubes are filled with water taken from the vessel (6.4). Water is supplied under argon gas pressure in the vessel (6.5).

Samples of 10Cr2Mo type steam generator steel were tested. One of the samples was the simulator of the initial defect of ~0.15 mm size in 2.5 mm thick wall. The simulator was made as a component of water injection device (2), i.e. needle shut valve with bellows and extended pin. Both valve and seat were made of 10Cr2Mo steel, and a groove was provided on the valve to simulate the initial defect (Figure 6).

Additional target sample (3) was made of 10Cr2Mo steel and installed in the injection area at 8 mm distance from the injection device outlet hole. This sample was shaped as a disc of ∅ 30 mm with $\delta = 2.1$ mm.

Four chromel-alumel thermocouples were provided for temperature control.

Liquid metal flow rate was measured by the constant magnet flow meter (P). Liquid metal was heated by nichrome wire electric heaters provided on the outer surface of the test facility components.

Liquid metal flow was provided by ENIV 2/0.5 electromagnetic pump. In the water supply system, “Sapphire-22DD” differential pressure gauge was used.

Water was injected under the excess argon gas pressure of 5.1 at. The injection continued until the abrupt increase of the leakage flow rate was detected (about an order of magnitude as compared to the initial flow rate).

The basic parameters and the results of the first experiment with water injection into Na-Pb and two experiments with water injection into Na are presented in Table 1.

As follows from the table, in all three cases damage propagation to the whole wall thickness occurred after approximately the same water (steam) amount, namely: ~ 25 g had flowed through the defect. This characteristic, other factors being equal, is an integrated criterion used for the estimation of defects self-growth in case of water leak into the liquid metal coolant.

Table 1: Main parameters and results of the experiments on water injection into sodium (Bagdasarov et al., 1999) and alloy.

Parameters	Water injection into sodium		Water injection into alloy
	1	2	
Liquid metal temperature, °C	450	450	450
Initial defect: its depth/length, mm	0.12×2.5	0.12×2.5	0.15×2.5
Initial leak flow rate, g/hour	36	18	90
Final leak flow rate, g/hour	740	970	700
Injection duration, min	53	73	63
Water mass passed through the defect before the abrupt leak increase, g	26	25	23
Average leak rate observed in the tests, g/hour	30.2	22	24
Max dimension of the inlet channel by the end of discharge, mm	0.7	0.3	~1

No measurable difference was detected between the damages of the main and additional target samples in the experiments with water injection into Na-Pb alloy and Na. All samples had corrosion spots with rough surface in front of the injection hole. Within this corrosion spot, there was a point of ~1 mm^2 area bearing the marks of local material loss. Max rate of target damage propagation was ~$8.3 \cdot 10^{-3}$ mm/s.

Based on the obtained results, a conclusion can be made on that the small additives of lead to sodium (2 at.%) are not effective from the standpoint of decreasing damage rate of 10Cr2Mo type steels in case of water leak into the liquid metal.

CONCLUSION

Results of using sodium and lead-bismuth coolants in the nuclear reactors of various applications were studied. Both positive and negative properties of these coolants are presented, as well as the results of development of engineering solutions aimed at the prevention of the impact of negative properties of the above coolants and also lead coolant, whose characteristics are very close to those of lead-bismuth. Potential of further application of sodium, lead and lead-bismuth coolants for fast reactors was confirmed on condition of availability of technical measures and devices designed for suppression of the impact of negative properties of these coolants.

Results of experimental studies on physical, chemical and thermohydraulic properties of eutectic alloys of lead with alkali metals are presented. These alloys are proposed to use as coolants instead of Na, Pb and Pb-Bi eutectic. Some negative properties of these liquid metal coolants were revealed, which would pronounce under conditions of advanced fast reactors.

Available analytical and experimental results demonstrate that all proposed alloys ($Pb_{0.83}Li_{0.17}$,, $Pb_{0.91}K_{0.09}$, and $Pb_{0.83}Mg_{0.17}$ eutectics, as well as Na-Pb alloys) intensively interact with water, steam, and oxygen. These interactions are almost unavoidable because of leaks in the steam generators occurring at various frequency and gas release from structural materials taking place in the stages of creation and operation of the reactors, as well as in the course of both scheduled and emergency replacements and repairs of the components. The alkaline components of the alloys are converted into the oxides and alkalis, which may dissolve in the coolant or may be removed, thus causing significant changes in coolant composition and thermodynamics. The situation becomes even more difficult because the emerging oxides are very strong in terms of thermodynamics and cannot be regenerated under reactor conditions (in contrast to the oxides of traditional coolants, such as Pb and Pb-Bi, which are easily reduced by hydrogen).

Thus, available to date analytical and experimental results indicate practically insuperable problems related to the use of $Pb_{0.83}Li_{0.17}$, $Pb_{0.91}K_{0.09}$ and $Pb_{0.83}Mg_{0.17}$ eutectics as coolants of nuclear reactors and other technological systems. Their supposed advantages were not confirmed. Therefore, there is no reasonable basis today for considering eutectic modification of liquid metal coolants as a promising trend of fast reactor technology development.

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