

Strategies of maintaining appropriate technology of heavy liquid metal coolants in advanced nuclear power plants

R.Sh. Askhadullin, A.Yu. Legkikh, A.N. Storozhenko, and V.V. Ulyanov

¹ State Scientific Centre of the Russian Federation – Institute for Physics and Power Engineering named after A. I. Leypunsky (SSC RF – IPPE), Obninsk, Russia

E-mail contact of main author: raskhadullin@ippe.ru

Abstract. Up to date, the following three main problems can be emphasized concerning technology of Pb-Bi and Pb coolants used in the civilian reactors: 1) maintaining high purity of coolant and cleanness of the surfaces of NPP circuits in order to assure design thermal-hydraulic performance of the plant operating on power up to 100 % during several decades; 2) long-term prevention of corrosion and erosion of structural materials (plant operating on power up to 100 % during several decades); 3) meeting of up-to-date safety requirements in the various stages of reactor operation (coolant preparation, reactor start-up, day-to-day operation, repair and refueling operations, loss of integrity, and abnormal operating conditions). All necessary technological measures can be implemented using special equipment created on the basis of the modern realities, which would be the essential part of the plant safety system in all stages of its operation.

Key Words: heavy liquid metal coolant, nuclear reactor, technology

1. Introduction

It can be considered that development of the technology for heavy liquid metal coolant (HLMC: lead, lead-bismuth) started in 1951, when at the Institute for Physics and Power Engineering (Obninsk, Russia) the first circulation test facility with lead-bismuth coolant was built up. In 1963 the first nuclear submarine was constructed with the reactor cooled by lead-bismuth. Nuclear submarines commissioned under the “Lira” project (according to the NATO classification – “Alfa” class) were in successful operation from 1970 to 1990 [1].

Currently three main tasks related to Pb and Pb-Bi coolant technology can be distinguished for civil reactor plants: 1) maintaining high purity of coolant and cleanness of the surfaces of NPP circuits in order to assure design thermal-hydraulic performance of the plant operating on power up to 100 % during several decades; 2) long-term prevention of corrosion and erosion of structural materials (plant operating on power up to 100 % during several decades); 3) meeting of up-to-date safety requirements in the various stages of reactor operation (coolant preparation, reactor start-up, day-to-day operation, repair and refueling operations, loss of integrity, and abnormal operating conditions).

It follows from the above-stated tasks that the current strategy pertaining to Pb and Pb-Bi coolant technologies should provide the following [2]:

- 1) preparation of coolant (Pb-Bi or Pb) and its loading;
- 2) preliminary off-circuit passivation of reactor components;
- 3) in-circuit passivation of inner surfaces of the primary reactor circuit;
- 4) coolant technology in case of repair and refueling;
- 5) coolant purification and circuit cleaning from impurities during day-to-day operation;
- 6) monitoring and control of the coolant oxidation potential in the course of NPP operation with HLMC;
- 7) cover gas purification in the liquid metal circuit;
- 8) application of processing procedures in case of deviations from normal operation conditions;
- 9) processing procedures for the coolant reuse.

The equipment that is under development in light of the current reality will be able to ensure implementation of all the required technological measures [3, 4] and will be an important

component in the general safety system during reactors operation at every stage of their service life.

1. Preparation of coolant and its loading to the circulation circuit

When the source material (metal) is received for the future coolant, the following impurities are subject to control: Ag, Cu, Zn, As, Sb, Sn, Mg, Fe, Tl, Hg, Al, In, Cd, Na, Ca, Ni, Cr, Mn, Te, Co, Au, and if the lead coolant is prepared, the Bi impurity is controlled as well.

The sources of these impurities can be very diverse: genetic impurities from the source raw material (Ag, Cu, Zn, As, Sb, Sn, Bi, Fe, Tl, Ni, Mn, Co, Au); process impurities that come from reagents in the course of raw metal production (Si, Ca, Fe, C); and finally, these can be process impurities that come in the course of raw metal purification (Na, B, F, Cl, Si).

In view of above-mentioned impurities, when engineering grades of lead and bismuth are selected for their subsequent use as a loaded coolant, the following criteria are used: the impact of impurities in the coolant on the radiological situation during the reactor operation; the impact of impurities on the nuclear and physical properties of the reactor; the rate of Po^{210} production from Bi^{209} and Pb^{208} and the rate of Po^{210} migration from the gas circuit of the reactor; the impact of impurities on structural material corrosion; the impact of impurities in the start-up coolant on the process of slag formation. The C1 grade for lead and VI00 for bismuth meet all the criteria to the most extent. For start-up coolant loading to the reactor primary circuit, the most suitable systems are a montejus system (Figure 1) or a tank system (Figure 2). Out of the tank systems the optimum ones for today are the systems that consist of one or six tanks.



Figure 1. An example of a montejus loading system (with montejus preparation at the factory).

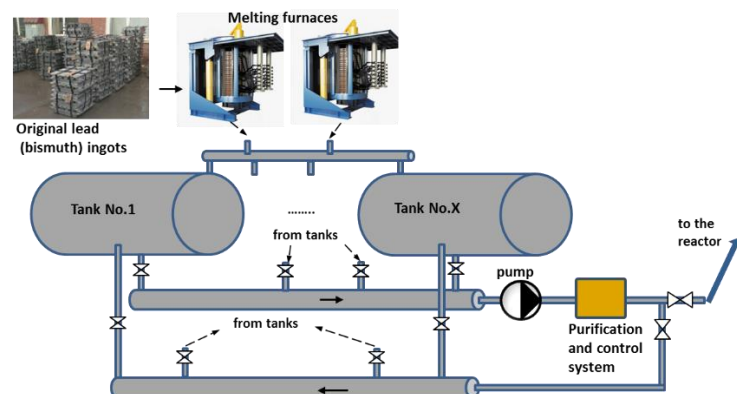


Figure 2. Loading tank system lay-out

The following stages are required to control the coolant composition:

- input control of the supplied tanks when metal is received;
- control in the process of final purification in the course of coolant preparation;
- control in the process of equipment surface preparation during the pre-commissioning work;
- control of specified and diagnostic impurities in the coolant during the normal reactor operation, reactor operation disturbances, decommissioning and after decommissioning.

In the process of coolant loading to the reactor, specific attention is paid to the presence of impurities in the cover gas in order to rule out any coolant contamination in the course of its loading.

2. Monitoring and control of the coolant oxidizing potential

The primary impurity in the HLMC is oxygen dissolved in the coolant. If the sufficient amount of oxygen is dissolved in the lead (lead-bismuth) coolant, on the surface of structural steel of the circulation circuit and equipment the oxide film is formed, which provides protection of surfaces against corrosion-erosion impact of the coolant. Due to the oxide nature of protective films, their state in the course of reactor operation, to a significant extent, is determined by the oxygen condition, i.e. by the concentration of oxygen dissolved in the coolant.

Oxide protective coating on the steel surfaces of the circuit components and equipment can be formed either in advance, off-circuit, or directly in the circuit of coolant circulation.

The range of allowable oxygen conditions can be determined on the basis of thermodynamic calculations. As an example, a diagram of oxygen conditions for lead melt is given in figure 3 [5].

The upper limit of the range is chosen proceeding from non-admittance of the conditions for formation of the solid lead oxide phase in the “cold” part of the circuit. The lower limit is determined proceeding from the oxygen concentration that corresponds to the level of potential destruction of the outer layer of protective oxide coating on the structural steel surfaces.

Thus, a theoretically admissible range of oxygen concentration was determined. When the concentration of oxygen dissolved in the coolant goes below critical values, i.e. below the lower limit of the concentration range, a reliable anti-corrosion protection of structural steel is not ensured. When the upper limit of the concentration range is exceeded, i.e. a significant amount of oxygen is present in the circuit; it may result in accumulation of prohibitive amounts of slag deposits on the heat-transfer surfaces of the circuit and the equipment.

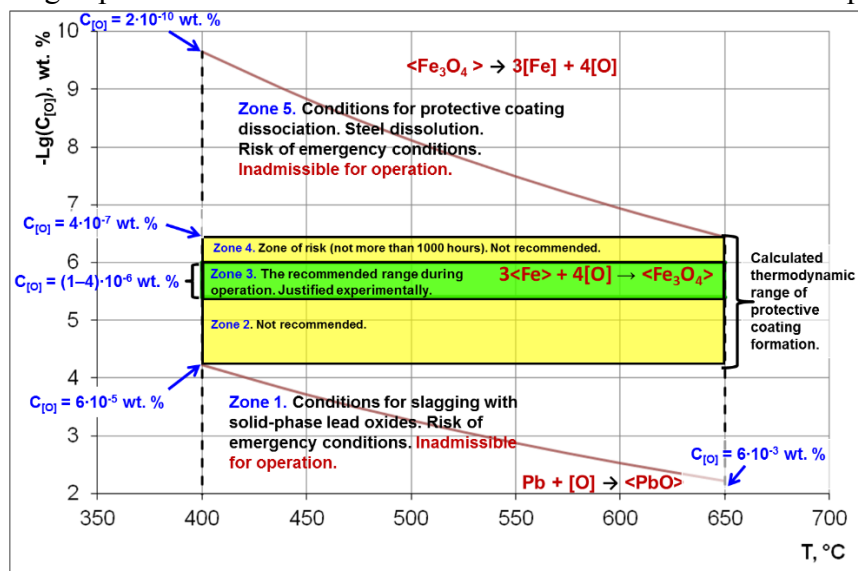


Figure 3 – Diagram of oxygen conditions for lead melt

Based on the results of long structural steel corrosion tests (for tens of thousands of hours) a narrower oxygen regime was determined for the coolant, the maintenance of which guarantees steel corrosion resistance at a long operation lifetime.

If the heavy coolant circuits are operated without special coolant makeup with dissolved oxygen, there occurs a spontaneous coolant deoxidation to the level at which no reliable structural steel anticorrosion protection is provided. A typical change in the concentration of oxygen dissolved in the coolant in the course of HLMC circuit operation is given in figure 4. So, a standard oxygen potential control system should be envisaged as part of the HLMC plant to maintain the preset oxygen conditions.

The SSC RF – IPPE specialists developed the methods for coolant oxidation potential monitoring and control and the tools for their implementation. Currently automatized heavy

coolant oxygen potential control and monitoring systems both for reactors and for test facilities are under development, which includes a set of equipment.

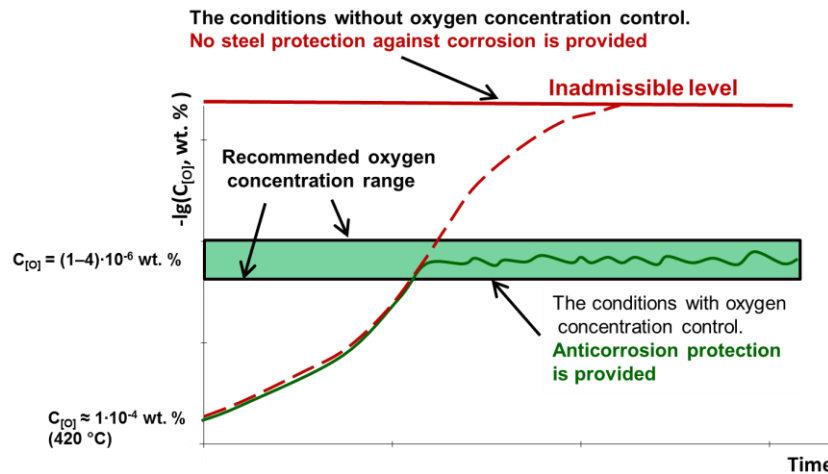


Figure 4 – Typical change in concentration of oxygen dissolved in the coolant during the HLMC circuit operation

Oxygen thermodynamic activity sensors (OAS) based on the solid oxide electrolyte have been developed to control oxygen dissolved in the coolant (Figure 5).

The OAS operation principle is based on EMF variation in the solid electrolyte concentration galvanic cell depending on oxygen concentration in the medium under analysis.

The paramount component of the sensor is a ceramic sensitive element that determines such OAS operation parameters as thermal and vibro-resistance, strength under increased pressure. During the studies performed the optimal chemical and phase composition of the ceramic material based on zirconium dioxide was developed with the required thermo-mechanical properties.

The developed ceramic sensitive elements made of partially stabilized zirconium dioxide make it possible to perform oxygen measurements in the lead and lead-bismuth melt within the thermodynamic activity range from 10^{-7} to 1 at the temperatures from 300 to 700 °C.

The performed studies on ceramic sensitive elements' thermal resistance demonstrated that ceramics can stand temperature variations at a rate up to 20 °C/s.

The sensors with three reference electrodes are currently under development to provide high reliability and validity of measurements for reactors with HMLC.

On the whole, OASs are characterized by fast response, high sensitivity, capability of working for a long time under the conditions of elevated temperatures and thermal shocks, reliability and stability of conducting and mechanical properties within a wide range of temperature and partial pressure of oxygen.

The solid-phase control method developed by the SSC RF – IPPE specialists is the optimum one to control injection of dissolved oxygen into the coolant to maintain the preset oxygen conditions.



Figure 5 – Capsule-type oxygen thermodynamic activity sensors

This control method consists in controlled dissolution of lead oxide granules fabricated by special technology and located in a specific reaction tank in the form of filling, through which the HLMC flow is arranged. Lead oxide granules contact heavy liquid metal and dissolve, thus enriching the melt with oxygen, which is transported further on throughout the whole circuit with the coolant flow.

By now, oxygen solubility in lead and lead-bismuth melts and lead oxide solubility kinetics in these melts have been studied. Empirical dependencies were obtained to determine oxygen solubility in the heavy liquid metals considered and to calculate kinetic characteristics of the oxide dissolution process under different temperature and hydrodynamic conditions. The availability of above-mentioned data allows the development of effective devices for implementation of the solid-phase control method, i.e. mass-transfer apparatus (MTA) with an oxygen solid-phase source.

As of today, a number of mass-transfer apparatus have been developed, these are devices of various designs and different principles based on which the process of solid-phase lead oxides dissolution is arranged (Figure 6). A significant experience has been gained in a long-term operation of various mass-transfer apparatus at the test facilities with lead and lead-bismuth coolants. It testifies to their reliability, capability of fine control of the oxygen input rate and no negative effect on the circuit on the whole.



Figure 6 – Mass-transfer systems to implement the solid-phase method for HLMC oxygen potential control

The process of preset oxygen conditions maintenance in heavy coolants is automated by means of a special automated oxygen concentration control system developed at the SSC RF – IPPE.

This system should meet the following requirements:

- to provide thermodynamic activity control for the oxygen dissolved in HLMC within a range of 10^{-7} – 1;
- to provide continuous maintenance of dissolved oxygen concentration in the coolant within a preset range under all operation conditions of the facility;
- to introduce oxygen to the circuit with the rate sufficient for maintenance of oxygen concentration within the preset range, with consideration of oxygen consumption by the circuit;
- not to allow formation of the solid phase of lead oxide-based slag in the circuit under all the conditions of dissolved oxygen generation and transportation;
- the system should reliably perform its functions for a long operation lifetime (tens of thousands of hours);
- a possibility of self-diagnostics and performance testing should be envisaged in the system for all its main components, namely, sensors, mass-transfer devices, software and hardware.

This system carries out the required makeup of the coolant with dissolved oxygen, with control of the rate of oxygen input from the mass-transfer device by means of the hardware-

software complex. There is a signal feedback from oxygen thermodynamic activity sensors, which makes it possible to ensure a high accuracy of the control process, actually without the intervention of an operator.

3. Hydrogen purification of coolant and circulation circuit surface from lead oxide-based slags

The necessity to perform hydrogen purification of heavy liquid metal coolants (lead and lead-bismuth) and circulation circuit surfaces is determined by the following.

When the circuit is filled with the coolant and is in operation, impurities can be formed in it. Impurities can primarily be formed as a result of coolant - air oxygen interaction. Oxygen can go to the circuit in case of its depressurization in a cold or heated-up state. In case of loss of tightness of the heated-up circuit, oxygen can directly interact with the coolant mirror. In case of loss of tightness of the cold circuit, oxygen can be adsorbed by the circuit and coolant surfaces. Then, when the circuit is heated up, oxygen can desorb and oxidize the coolant. With this, solid oxides are formed; they deposit on the circuit surface and the coolant mirror. The deposits can block heat-transferring surfaces (first of all, clear openings of the core), disturb the operation of pumps, valves, increase hydraulic resistance of flow paths, etc.

Usually, deposits most of all contain lead (for the circuits with liquid lead) and eutectic components, lead and bismuth (for the circuits with lead-bismuth alloy). Lead oxides and structural material component oxides (primarily that of iron) are present in deposits in significant amounts.

The ratio of lead (or eutectic components), lead oxides and iron oxides may vary significantly in different types of deposits, but their specific features remain the same. First of all, in spite of a high lead mass fraction (or that of eutectic components), deposits represent non-transportable, non-pumpable mass of a complex composition that blocks different parts of the circulation circuit. Lead oxide plays the role of the main binding material in deposits. It is the lead oxide with the impurity of structural component oxides that provides a porous spatial structure whose inner volumes (cells) are filled with unoxidized lead or eutectic components.

The indicated deposits are eliminated by means of a special technological procedure (hydrogen purification), which results in removal of lead or eutectic components from the deposits. In this case deposits are destroyed, and lead and bismuth go back to the coolant. Hydrogen purification is performed by means of gas mixtures $H_2 - H_2O - He (Ar)$. Gas mixtures may be added to the circuit in two ways: 1) to the gas volume of the circuit only; 2) simultaneously to the gas volume of the circuit and directly to the circulating HLHC flow. In the first case, hydrogen interacts with deposits that are only present on the free coolant surfaces. In the second case, the gas mixture can be spread over the entire circuit, and hydrogen can interact with deposits in all the parts of the circuit. Thus, hydrogen purification arranged in the second way is the most effective.

The following processes go on in the course of hydrogen purification:

- a) hydrogen reduction of metal (Me) from oxides (Me_xO_y) in deposits on the surfaces of the coolant and circuit structures by the reaction of: $Me_xO_y + y H_2 \leftrightarrow x Me + y H_2O$ (1), it results in deposit destruction and the subsequent entrainment of its particles by the HLHC flow;
- b) hydrogen reduction of Me by the reaction of (1) from oxide formations circulating together with the coolant;
- c) removal of dissolved oxygen from the coolant by the reaction of: $[O] + H_2 \leftrightarrow H_2O$ (2), where $[O]$ is dissolved oxygen;
- d) dissolution in the deoxidized coolant of the oxides from the surface deposits and from the oxide formations that circulate together with the coolant: $Me_xO_y \leftrightarrow x [Me] + y [O]$;
- e) mechanical destruction of the deposits on the circuit surfaces by means of increasing the dynamic two-component flow effect on them. The dynamic impact is increased through

creation of additional (as compared to one-component flow) tangential and normal stresses close to solid surfaces of impurities.

For the circuits of the loop-type facilities, the mixture of $H_2 - H_2O - He(Ar)$ can be added to the coolant circulating with a velocity of $U \geq 1$ m/s by any practically feasible way that provides the size of the gas bubbles injected to the coolant with a radius of $r \sim 1$ mm. Further on, in view of insignificant length of the pipeline connecting the circuit elements, small area of free coolant surfaces and effective gas fragmentation in the core, steam generators and pumps, these bubbles are effectively transported by the HLMC flow throughout the entire circuit.

In the circuits of upgraded loop-type and pool-type facility designs there are long channels with low ($U \geq 0.2-0.3$ m/s) velocities of the coolant flow, and large free surfaces of the coolant. So they have certain conditions for effective agglomeration and separation of gas bubbles with a rate of $r \geq 1$ m/s. To provide the gas-phase supply to all the parts of the circuit it is necessary to use only such ways of gas injection into the coolant that would ensure its fragmentation to the bubbles with a size of $r \leq 0.3$ mm.

So far, the following measures are envisaged to completely rule out any clear opening plugging with lead oxides in the core, steam generators, pumps and other equipment in the reactors with heavy liquid metal coolants:

- to purify the source lead and bismuth at the manufacturing plant and directly in the course of lead (or lead-bismuth) loading to the reactor primary circuit;
- to minimize, up to complete elimination, the possibility of air ingress into the reactor primary circuit at all the stages of reactor operation;
- to use special solid-phase feeders (mass-transfer apparatus considered above) of dissolved oxygen to form protective coatings on steel, with elimination of excess lead oxide formation on heat-transfer surfaces in the reactor ;
- to perform early monitoring of any indication of air ingress into the reactor primary circuit in order to timely detect the causes and eliminate them;
- to use an effective system of hydrogen purification of the coolant and the circuit surfaces from lead oxides in case of any indication of their formation;
- to set oxygen-free water-chemical conditions in the secondary circuit to rule out the formation of lead oxides in case of a probable microleak of the steam generator;
- to filter the coolant and gas circuit in order to remove impurities and aerosols that cannot be removed by hydrogen purification;
- to use the refueling and repair equipment that rules out any air ingress into the coolant, and thus, eliminates formation of excess lead oxide phase in the course of refueling and repair.

As it has already been stated, as a result of the use of hydrogen purification, no negative situations related to excess accumulation of lead oxide-based slag were observed either at any lead-bismuth cooled nuclear submarine or at test facilities with lead or lead-bismuth coolant.

4 Coolant filtration

Filtration of heavy coolants is required for the following reasons. Heavy liquid metal coolants represent a low concentrated finely-dispersed suspension in which the suspended finest solid particles of lead, bismuth, iron, chromium, manganese oxides and other impurities may be present. Depending on the operation conditions, their total concentration in the coolant may go up to the value of $\sim 10^{-3}$ % mass and higher.

A potential danger of the indicated impurities consists in their possible separation from the liquid metal to the surface of heat-exchange equipment, valves, etc., with all the adverse consequences that come with it. The indicated impurities are primarily formed as a result of slow corrosion-erosion processes in the circuit, circuit depressurization, ingress of

atmospheric oxygen, oil, etc., into it. Thus, a continuous removal of solid impurities from the coolant by filtration becomes extremely important.

As the inner surface of the circulating loop should be considered competitive with regard to the surface of the filter, the effectiveness of the latter should be high enough to prevent intensive formation of deposits on various components of the thermal engineering equipment, in the narrow clear openings of the channels, etc.

In the general case, in the closed circulation circuit with a filter, variation in the slurry concentration under the impact of its source can be presented with the equation $\frac{dc}{d\tau} = \frac{q}{V} - \frac{(C - C_B)G}{V} - R_K C$, where: C is current slurry concentration in the coolant, kg/m^3 ; C_B is slurry concentration at the filter output, kg/m^3 ; q is an impurity source intensity, kg/h ; τ is time, h ; V is a coolant volume in the loop, m^3 ; G is a coolant volumetric flow rate through the filter, m^3/h ; R_K is an experimental constant that characterizes the intensity of slurry removal from the coolant flow to the circuit surface, $1/\text{h}$. In this case, for the circuit without a filter the following correlation is valid: $C \approx C_0 \cdot e^{-R_K \cdot \tau}$, where C_0 and C are initial and current slurry concentrations, kg/m^3 . The R_K value can be presented as: $R_K \approx \frac{2,31q^{C_0/C}}{\Delta\tau}$, where $\Delta\tau$ is a time period before measurement of impurity concentration C in the coolant.

A large array of experimental data from commercial and laboratory circulation loops was processed. It made it possible to estimate, in terms of the particles lifetime, the values of R_K for the slurry that primarily consists of lead, bismuth and iron oxides formed as a result of oxygen gas supply to the liquid metal or artificially supplied to the circuit. It turned out that the R_K values are within the following ranges [6]: $R_K = 0.29 - 0.72$ ($1/\text{h}$) – for lead and bismuth oxides and $R_K = 0.11 - 2.16$ ($1/\text{h}$) – for iron oxides.

As it can be seen, there is a significant scattering of R_K values, which is evidently related to the difference in geometrical characteristics of circuits, circulation conditions and particle-size distribution of impurities that determine the intensity of their hydrodynamic separation.

If we require that the filter should improve its intensity of trapping the impurities by an order of magnitude as compared to the circuit, we obtain: $n = \frac{G}{V} \geq \frac{10R_K}{E}$, where: n is a circulation rate, $1/\text{h}$; E is coolant purification efficiency of the filter, $E = 1 - \frac{C_B}{C} = 1 - \frac{1}{K}$, K is a purification factor: $K = \frac{C}{C_B}$.

Thus, it is clear that the n value, to a large extent, determines the efficiency of the HLMC purification process by means of a filter (with equal R_K values, the E value variation is proportional to n variations), and for heavy liquid metal coolant filtration it is necessary to use filtering materials with higher filtration parameters, to use filters with a bigger volume and (or) larger surface area of filtering materials and their less hydraulic drag as compared to those recommended earlier for nuclear submarine reactors cooled with lead-bismuth.

5 Cover gas purification from coolant aerosols.

Operation of HLM-cooled reactors is related to the regimes accompanied with a yield of aerosol products to the reactor gas loop. As a result of these processes, the coolant aerosol particles are formed, transferred and deposit on the equipment, pipelines and instrumentation of the gas system.

The problem of gas system contamination in experimental facilities and reactors with heavy coolant is not new. As far back as in the initial period of lead-bismuth coolant development,

contamination of gas system pipelines and equipment were observed at the test facilities and later, at the commercial ones.

In particular, there is a well-known fact of radioactive dust particles influencing the representativeness of readings of the sensors in the fuel element cladding tightness control system. The level of impact differed from facility to facility; however, it remained noticeable for actually the entire period of their operation. [3]. Dust-like deposits in the gas system pipelines were also detected in the course of inspections of these components during the plant repairs.

It was noticed that the rate of cover gas dust pollution increased significantly under the conditions of treatment of the coolant and circulation circuit with “coolant-gas” two-phase mixtures. It can be explained by the fact that due to flotation transport of impurity particles on gas bubbles, the speed of their delivery to the coolant - cover gas interface surface and further on directly to the gas volume increased. It was usually observed during hydrogen recovery, i.e. the main process regime of the circuit cleaning in which hydrogen-containing gas mixtures were injected into the coolant flow. Under these conditions a significant dust concentration in the gas system resulted in respective contamination of water steam condensation devices and the condensed water as well.

In view of the available knowledge of aerosol formation as applied to the heavy-liquid metal-cooled reactor conditions under the reactor normal operation, contamination of the cover gas at the gas system tract inlet at the level of $C \approx 0.1-1.0 \text{ mg/m}^3$, that corresponds to the experimental data for the coolant temperature of 450°C and the minimum distance from the circuit, can be assumed as an estimator. Under the conditions of primary circuit treatment with gas mixtures and steam generator leakages, an increase in the dust particles concentration up to $10-40 \text{ mg/m}^3$ should be expected.

The level of impact of the processes of cover gas pollution with dust-like impurities on the serviceability of the equipment located in the gas system, to a significant extent, depends on the principle of operation of this equipment and its design peculiarities.

The gained knowledge of concentration and particle-size distribution in the ventilation system of the reactors with heavy liquid metal coolants allows filtering devices with separate purification sections to be considered necessary. The first “high-temperature” section for preliminary purification, $E \approx 80 \%$, is intended to purify the gas line from the coarse particles with a size exceeding $5.0 \mu\text{m}$. This section is assumed to be installed directly at the reactor outlet, where the gas temperature goes up to $450-600^\circ\text{C}$. In order to get these parameters, it is reasonable to use a needle-punched metal fabric made of heat-resistant steel as filtering material (Fig. 7).

The second “low-temperature” section for fine purification is meant to perform final purification, $E = 99.9 \%$ of gas, and is installed in the cold side of the circuit ($20-40^\circ\text{C}$). It is reasonable to use high-efficient fiber paper as filtering material (Figures 8, 9).

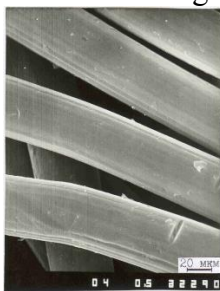


Figure 7. Micrograph of metal felt fibers



Figure 8. Highly-efficient fiber paper

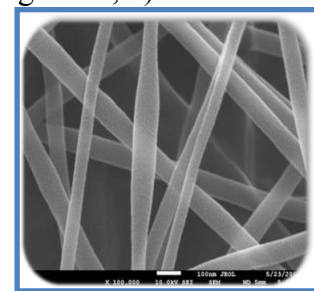


Figure 9. Micrograph of glass fibers

Conclusion

The issues related to heavy liquid metal coolant technology are to be solved during the entire reactor life cycle. The activities pertaining to the coolant technology include R&D developments aimed at justification, designing and creation of coolant technology methods and tools, and direct implementation of technological processes during the coolant preparation, power plant start-up, its lifetime operation and decommissioning.

Selection of the required coolant technology methods and tools for advanced reactor projects is justified both in terms of the gained 60-year experience in the coolant development, and based on the results of the performed R&D project designs that take into account the specific features of reactors of a new generation.

The coolant technology methods and tools, which are under development, include a system of hydrogen purification of the coolant and circuit from slag-forming impurities, a system for dissolved oxygen control in the coolant to ensure anti-corrosion protection of steel against corrosion, a system of coolant and cover gas filtration, a coolant control system both in reactor and non-reactor conditions, and other systems.

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