Study of Lead Coolant Technology

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Abstract

Calculated and experimental work was fulfilled in IPPE that allowed to analyze the lead coolant features concerning its technology treatment. The coolant technology procedures that have been developed for lead-bismuth eutectic and tested with lead allow the following: removal of the excess of the solid phase based on the lead oxides; control and regulation of the coolant quality; formation of the protective oxide films on the steel surfaces.

Tests have revealed the closeness of the main physical and chemical processes taking place in the loops with lead and lead-bismuth coolants.

At present time lead and lead-bismuth eutectic are considered most frequently as coolants for nuclear reactors and as materials for liquid metal targets in accelerator-driven systems.

The choice of **lead and lead-bismuth eutectic** is determined by the number of favorable thermalphysic and physical-chemical properties.

Low chemical activity of lead and bismuth exclude the possibility of explosion and fire when their interaction with air, water and vapor. High boiling point of coolant ($1737^{O}C$ - lead and $1670^{O}C$ - lead-bismuth eutectic) exclude the possibility of coolant boiling out in the loop sections with high energy deposition. The low working pressure of the coolant in the loop increases the facility reliability and safety, simplifies the equipment design and manufacture, significantly simplifies the operation conditions for the equipment of the primary loop.

Lead-bismuth eutectic is used successfully as a coolant for nuclear reactors of transport purpose for many years. A big volume of data on thermalphysic, physical-chemical and other properties of alloy is obtained. The technology of this coolant treatment and equipment for its realization are developed [1].

In the few last years the study of **lead** as a coolant for fast reactors is being carried out.

The main favorable properties of lead are similar to lead-bismuth eutectic favorable properties. Besides, lead is attractive by the following properties:

- polonium activity of lead is ~ 3 oders of magnitude less than one of lead-bismuth eutectic;
- lead is easily accessible material, comparatively cheap and its production is well organized.

Basic defect of lead is high melting point ($327^{O}C$) that creates difficulties during its use as a coolant in nuclear power plants.

- amount of the impurities and the intensity of the physics-chemical processes and mass transfer are increased, this affects the maintenance procedures of the coolant quality;
- the use of the elaborated structural materials decreases the working range of the temperatures;
- difficulties connected with coolant technology significantly increase during facility starting, its operation and repair.

The main tasks of lead coolant technology as well as of lead-bismuth one are the following:

- insuring of the purity of the loop;
- maintenance of the concentration of oxygen dissolved in lead coolant at the determined level;
- controlling of coolant quality.

The number of calculated and experimental work were realized in IPPE that allowed to analyze the lead coolant features in the field of its technology treatment.

Oxygen solubility in lead and lead-bismuth eutectic is studied. Experiments confirmed the earlier obtained dependence of limit oxygen solubility in lead-bismuth eutectic on temperature.

$$lg\,C_S = 1.2 - 3400/T$$

The improved limit oxygen solubility in lead has been obtained.

$$lg\,C_S = -1.36 - 1274/T$$

Comparative study of **solubility kinetics** of solid phase lead oxide (PbO) of spheroid shape in lead and lead-bismuth eutectic in the range of temperature

400-600^{O}C revealed that solubility of lead oxide is more intensive in lead-bismuth eutectic.

Dependence of constant of the process velocity on temperature is determined:

for lead-bismuth

$$lg\,K_{p(Pb-Bi)} = 2.85 - 4898/T$$

for lead

$$lg\,K_{p(Pb)} = 1.72 - 4198/T$$

Complex of efforts to find out the possibility of using the procedures of the coolant technology developed for lead-bismuth eutectic as a coolant technology for lead was carried out. Experiments were fulfilled at the circulation facilities with lead-bismuth coolant (ÒÒ - 2Ì facility) and lead coolant (ÈÑÐ facility). The principle diagrams of these facilities are given in the fig.1

The technical characteristics of these facilities allow testing practically under identical hydrodynamics and temperature conditions. The both facilities are fitted with systems of hydrogen purification, filters for coolant purification, systems of regulating and controlling of coolant quality.

The procedure of hydrogen purification of coolant was studied under isothermal mode at the t $\cong$ 450^{O}C.

Argon-hydrogen gas mixture was supplied into the circulating coolant by ejector 5 (fig.1), interacting as with coolant so with slag deposits on the loop surface.

The process of hydrogen purification was under control using the changing of hydrogen concentration in gas mixture (6, fig.1) and thermodynamic oxygen activity in coolant (9, 14, fig.1).

In initial state both facilities were significantly contaminated by slag based on the lead oxide.

The results of study of the process of hydrogen purification of the lead coolant and the loop are given in fig. 2.

It is clear from fig. 2 that the process of hydrogen reduction of lead oxides was sufficiently effective.

$$PbO + H_2 = H_2O + Pb$$

Concentration of hydrogen in the gas mixture decreased from 30% vol. down to 5-10% vol. during 50 hours.

However, in spite of intensive purification of the loop and the lead coolant from oxygen, the thermodynamic oxygen activity

$$a = C_i/C_S$$

kept the level of a = 1 during first 450 hours.

Concentration of oxygen dissolved in the lead coolant corresponded to the oxygen saturation concentration. This fact indicated the significant initial contamination of the loop by the lead oxides.

After 450 hours of hydrogen purification, the process of purification of the lead coolant from dissolved oxygen started. Thermodynamic oxygen activity decreased down to the value equilibrium with Fe_3O_4 oxide which is the base of protective anticorrosive films covering the structural materials.

The process of hydrogen purification of the lead-bismuth loop kept the same way.

The experiment results showed the possibility to use the procedure of hydrogen purification for purification of lead loops.

At present time corrosion resistance of the materials used in the loops with heavy liquid metal coolants (Pb, Pb-Bi) is provided due to formation of the protective oxide films covering the materials. In the frames of this conception, the conditions of normal operation of structural materials require to maintain the presence of the definite amount of the oxygen impurity in the coolant. This insures the impossibility of dissociation of protective films as well as accumulation of solid phase lead oxides (slag).

In these experiments **the procedure of regulation** (increasing) of oxygen concentration dissolved in the coolant, using the solid phase lead oxides previously treated in a special technological way, as an agent for oxidation was studied. Changing the temperature and (or) the coolant flow rate in the test tank where the solid phase oxidizing agent is placed it is possible to regulate the velocity of oxide dissolution in accordance with the loop need of dissolved oxygen.

Experiments were carried out under non-isothermal mode of the loop operation - 420 - 540^{O}C.

The readings of the oxygen sensors N1 and N3 (fig.1, ÈÑÐ) placed in the loop sections having the coolant temperature of 460^{O}C and 540^{O}C accordingly are shown in fig. 3. As it is seen in all cases, when the power of the source of the metal impurities increasing or the power of the oxygen source decreasing, the noticeable coolant reduction takes place. When decreasing of the power of the iron source and increasing of the power of oxygen source, the coolant oxidation takes place.

The possibility of the accurate regulation of the dissolved oxygen concentration in the interval of $1\cdot10^{-6}$ - $5\cdot10^{-7}$ %mass during a long time (more then 1000 hours), using the solid phase source of oxygen was shown at one of the non-isothermal (420-650^{O}C) IPPE facilities.

The results of experiments confirmed the possibility of maintenance of the oxygen concentration dissolved in lead coolant at the required level, using the solid phase oxidizing agent, and efficiency of the equipment design.

It is necessary to give explanation of some difference in the readings of the oxygen sensors located in different temperature zones (fig. 3).

To explain, let's choose readings of all sensors (see fig. 1) at the time moments indicated by arrows in fig. 3. The number of the readings distribution of oxygen sensors

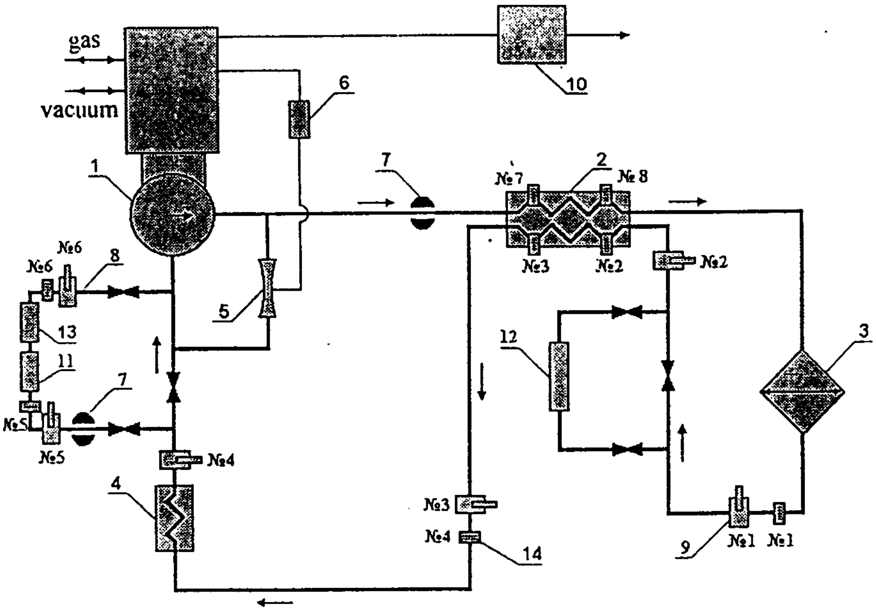

Fig. 1. Principal diagram of circulation contours of ÒÒ-2Ì and ÈÑÐ facilities.
1 - circulation pump, 2 - section for heat recuperation, 3 - heating section of loop, 4 - heat exchanger "metal-water", 5 - ejector, 6 - system for gas moistening, 7 - flow meter, 8 - technological bypass, 9 - sensor of oxygen thermodynamic activity at the ÒÒ-2Ì facility, 10 system for gas composition controlling, 11 - mass transfer device, 12 - source of metal impurities, 13 - filter for coolant purification, 14 - oxygen thermodynamic activity sensor at the ÈÑÐ facility.

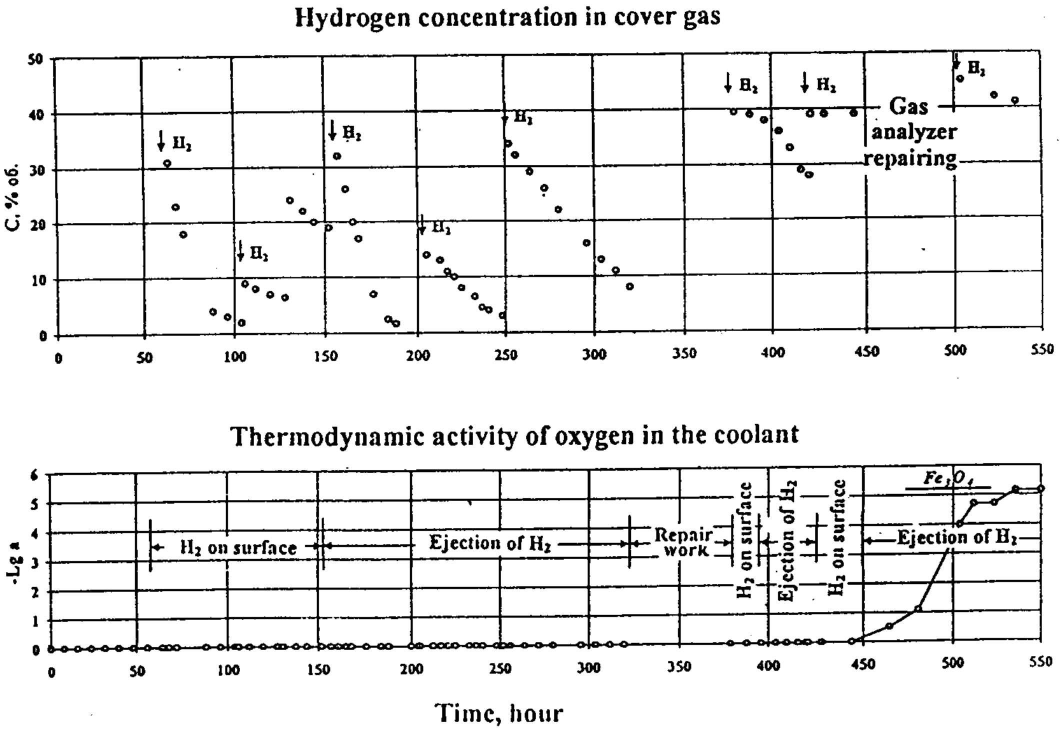

Fig. 2. Changing of lead coolant characteristics during the process of its regeneration by hydrogen (T = 450 ^{O}C) in the ÈÑÐ facility.

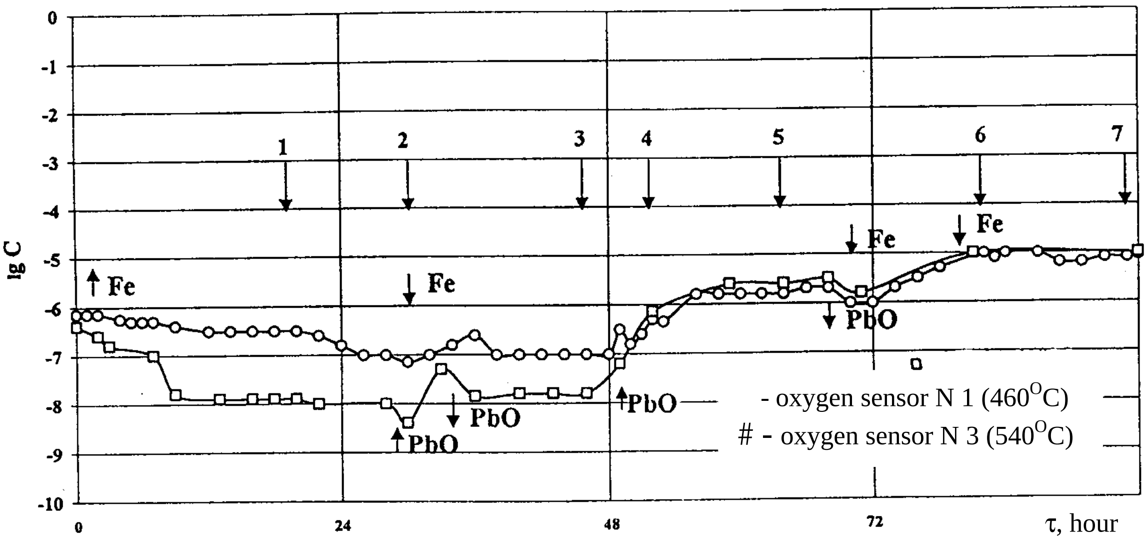

Fig. 3. Regulation of oxygen concentration (lgC) in lead coolant under non-isothermal mode (420-540^{O}C) in the ÈÑÐ facility.

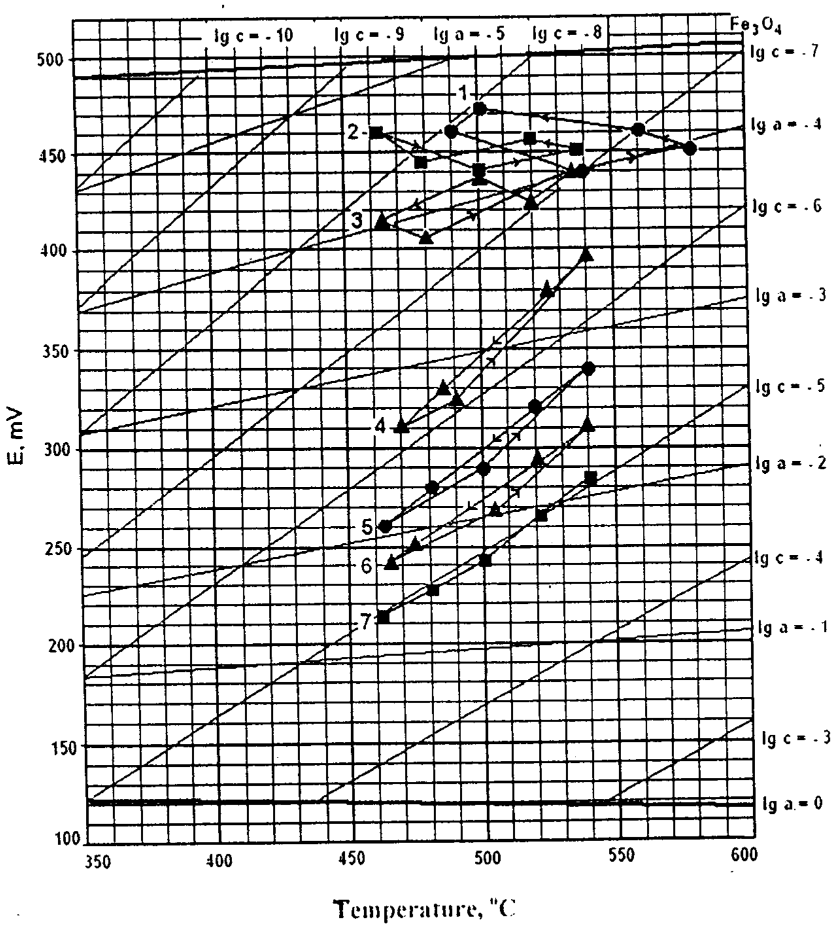

Fig. 4. Distribution of readings of oxygen activity sensor in dependence on oxygen concentration in lead coolant in the ÈÑÐ facility.

located in different temperature zones of the loop and shown in fig. 4 corresponds to every arrow number shown in fig. 3. As is seen in figures, when oxygen concentration in coolant is higher than ~10^{-6} %mass, readings of the sensors correspond (or approximately correspond) to the iso-concentration distribution. When oxygen concentration decreases down to ~10^{-7} %mass, distribution of the sensors readings changes sharply. First the distribution closed to the isoactivity is observed then when the coolant gets more reduction, the distribution is significantly different.

Changing of the distribution character of the oxygen sensors readings can be explained by the interaction of the impurities existing in the coolant, mainly of iron and oxygen. Any changing of iron (oxygen) activity in any point of the loop affects an oxygen (iron) activity. The sources of impurities presented in the loop affects the field of activities.

The results of study of the process of coolant quality regulation at the ÒÒ-2Ì lead-bismuth facility are given in fig. 5, 6. These results coincide with results obtained using lead coolant and given in fig. 3, 4. It is necessary to point out that changing of the distribution character of oxygen activity as in lead coolant so in lead-bismuth coolant takes place at the same oxygen concentration ($C_O \approx 10^{-7}$ %mass) measured in the "hot" point of the loop. This fact indicates that the ratio of amounts of interacting impurities in the coolants, apparently, is equal.

Thus, the practical possibility of operation of non-isothermal circulation loops with lead coolant is confirmed.

Experimentally confirmed the closeness of basic physical-chemical processes taking place in the loops with lead and lead-bismuth coolants.

At the same time existing calculated and experimental data are not sufficient for full substantiation of the technology of treatment with lead coolant as applied to the large-scale stationary plants. It is necessary for this to fulfill the complex of research efforts using the experience of the mastering of the lead-bismuth coolant.

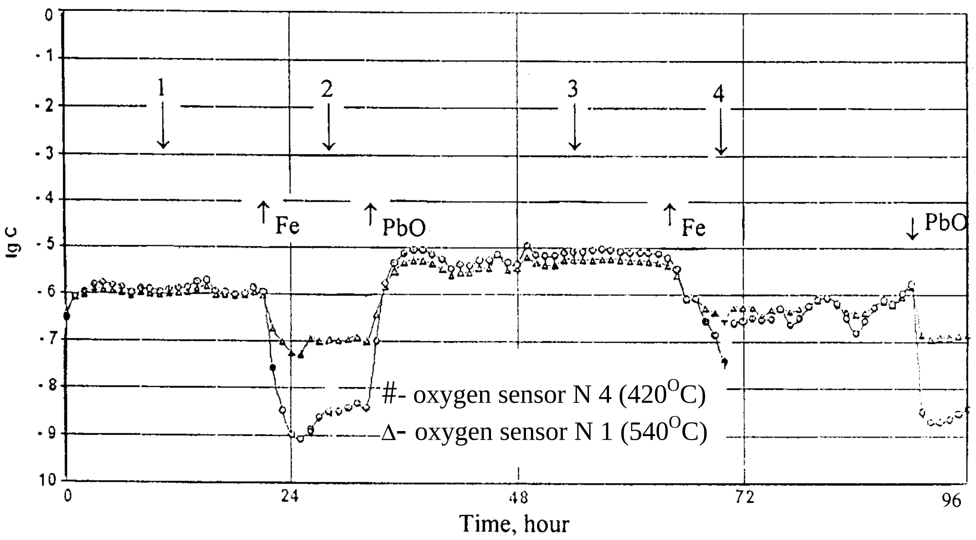

Fig. 5. Regulation of oxygen concentration (lgC) in lead bismuth coolant under non-isothermal mode (420-540^{O}C) in the ÒÒ-2Ì facility.

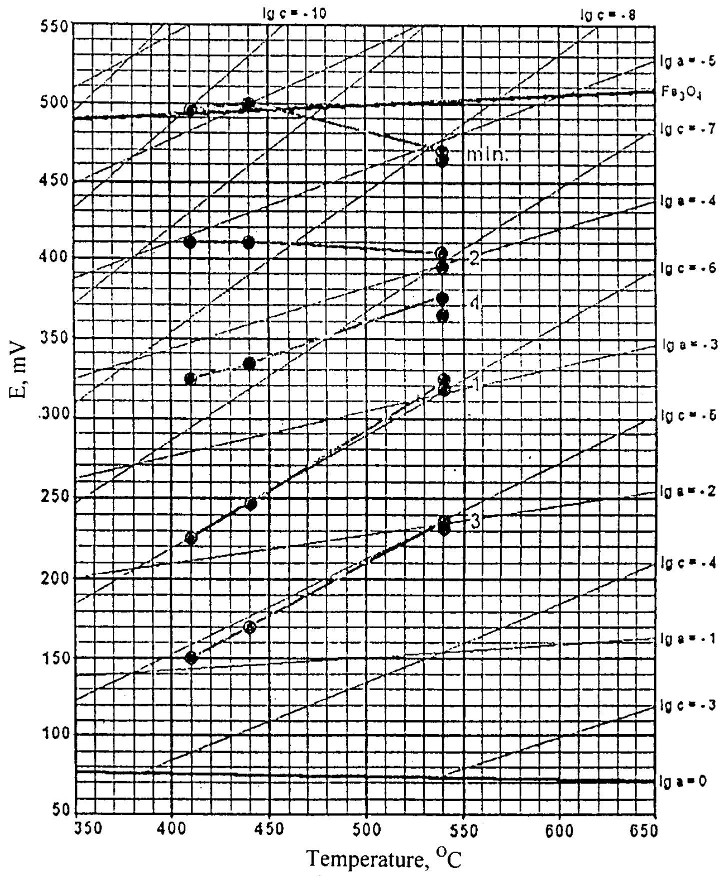

Fig. 6. Distribution of readings of oxygen activity sensor in dependence on oxygen concentration in lead-bismuth coolant in the ÒÒ-2Ì facility.

References

1. B.F. GROMOV, Y.I. ORLOV, P.N. MARTYNOV, V.A. GYLEVSKY
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