



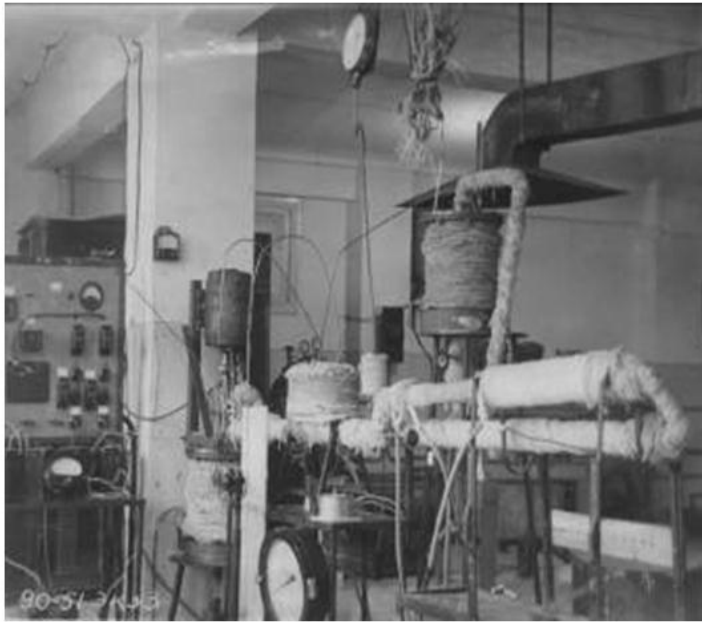
FSUE "SSC RF – Institute for Physics and Power
Engineering named after A.I. Leypunsky"

Technology of lead and lead-bismuth heavy coolants

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A.N.Storozhenko

Workshop "Fast neutron reactor coolants "
(Paris, February 19-20, 2013)

Phases of learning of heavy liquid metal coolants



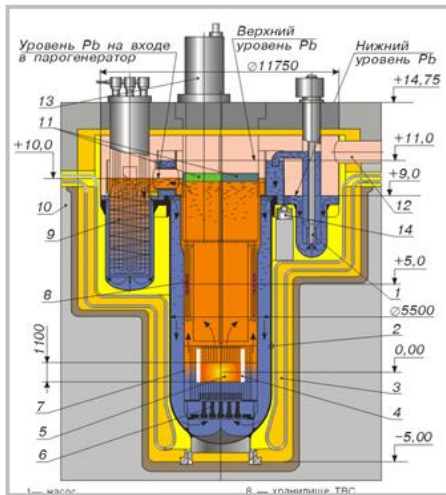
1951
First test
circulation facility
with Pb-Bi coolant
(IPPE)



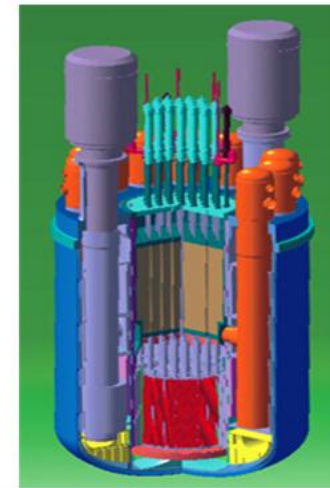
1963 – First NPS (Design 645)
with Pb-Bi coolant



1971 – NPS (Designs 705 and 705K)
with Pb-Bi coolant



2013
First in the world civil nuclear
reactors with Pb-Bi and Pb
coolants are under development





Several methods and instruments of coolant technology have been the first to be applied during startup and operation of NPS of Design 705 (1968 - 1990). This allowed avoid previous emergencies (having place in Design NPS K-27 and test facility 27 BT5). Positive experience available is taken into account in the development of modern methods and instruments of coolant technology for reactor facilities with Pb and Pb-Bi coolants.

Main goals of heavy coolant technology (Pb-Bi, Pb) in civil reactor facilities:

- 1. Provision of coolant purity and cleanliness of circulation circuit surfaces of tank arrangement to maintain the designed thermal and hydraulic parameters during long-term operation (several tens years when nuclear power plant operates at nominal power and up to 100%).**
- 2. Prevention of corrosion and erosion of structural materials during long-term operation. (several tens years when NPP operates at nominal power and up to 100%).**
- 3. Meeting of modern safety requirements at different phases of reactor facility operation (coolant preparation, RF startup, current operation, repair and reloading, depressurization, deviation of rated operation modes).**

Absence of coolant technology instruments can result in negative consequences: slag deposit on heat transfer surfaces, blocking of coolant flow rate along separate circuit sections; high corrosion severity; reduction of facility operation durability, deterioration of radiation situation, etc.



The first submarine “K-27” with Pb-Bi coolant
Startup - in 1963
Accident – in 1968
Cause of accident – absence of coolant technology systems

Result of test facility operation without application of methods and instruments of heavy coolant technology



Slag in pipeline



Slag deposit in the circuit during circulation pump tests



Slag deposit in heat exchanger

Such circuit surface conditions have not been observed since the introduction of coolant technology systems into circulation circuits.

Modern package of measures in technology of heavy liquid metal coolants

Coolant preparation
(Pb-Bi or Pb) and its loading

Coolant and circuit purification from
impurities during current operation

Preliminary out-of-circuit
passivation of RF units

Control and regulation of the coolant
oxidation potential

In-circuit passivation of internal
surfaces of RF primary circuit

Purification of cover gas in
liquid metal circuit

Coolant technology during
repair and reloading

Execution of technological operations
in cases of rated operation deviation

Technological operations for
coolant recycling

Equipment under development will make available the needed technological measures. It is an important component of safety system during the reactor facility operation at all phases of their life cycle

Main activity phases of technology of heavy liquid metal coolants (Pb, Pb-Bi)

**Phase 1. Selection of coolant initial composition.
Technology of coolant preparation for loading
into the primary circuit of reactor facility**

**Phase 2. Coolant technology processes
during initial (startup)
passivation of structural materials**

**Phase 3. Coolant technology
during current operation of reactor facility**

**Phase 4. Coolant technology
after reactor facility decommissioning**

Selection of lead engineering grade for its use as initial coolant

Lead coolant					
Element	C2	C2C	C1	C0	C00
Mass fraction, %					
Pb	99,95	99,97	99,985	99,992	99,9985
Ag	0,0015	0,002	0,001	0,0003	0,00001
Cu	0,001	0,002	0,001	0,0005	0,00001
Zn	0,001	0,002	0,001	0,001	0,0001
Bi	0,03	0,02	0,006	0,004	0,0005
As	0,002	0,002	0,0005	0,0005	0,0005
Sn	0,002	0,001	0,0005	0,0005	0,0005
Sb	0,005	0,005	0,001	0,0005	0,0001
Fe	0,002	0,001	0,001	0,001	0,0001
Mg	0,015	0,003	0,003	0,002	0,0001
Ca					0,0001
Na					0,0001
Tl	Not regulated	Not regulated	Not regulated	Not regulated	0,0001
Cd					0,0001
Al					0,0005
Hg					0,0001
In					0,0001

Selection criteria:

1. Impact of impurities in lead coolant on radiation situation during RF operation
2. Impact of impurities on nuclear and physical reactor properties
3. Generation rate of Po^{210} from Bi^{209} and Pb^{208} and migration rate of Po^{210} from RF gas circuit
4. Impact of impurities on structural material corrosion
5. Impact of impurities in the initial coolant on slag formation process

C1 – possible candidate lead grade for the use as coolant

Selection of lead and bismuth engineering grades for their use as components of initial coolant

Lead-bismuth coolant			
Element	Bi1	Bi00	C1
Mass fraction, %			
Pb	1,8	0,01	99,985
Ag	0,12	0,00002	0,001
Cu	0,01	0,0001	0,001
Zn	0,003	0,0005	0,001
Bi	98	99,98	0,006
As	0,0002	0,00007	0,0005
Sn	Not regulated	Not regulated	0,0005
Sb	0,005	0,00002	0,0005
Fe	0,001	0,001	0,001
Mg	Not regulated	Not regulated	0,002
Ca			
Na			
Tl			
Cd	(0,0001)	0,00005	Not regulated
Al	Not regulated	Not regulated	Not regulated
Hg			
In			

Selection criteria:

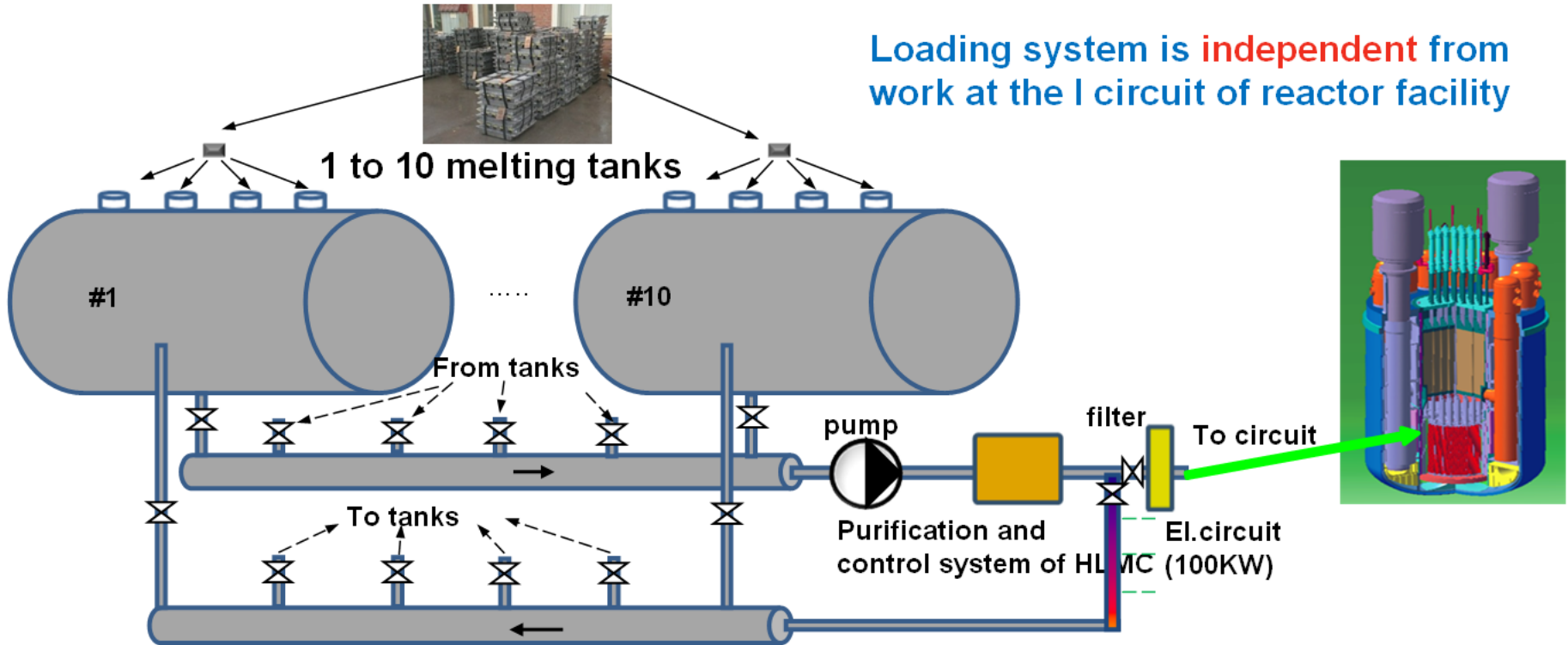
1. Impact of impurities in lead coolant on radiation situation during RF operation
2. Impact of impurities on nuclear and physical reactor properties
3. Generation rate of Po^{210} from Bi^{209} and Pb^{208} and migration rate of Po^{210} from RF gas circuit
4. Impact of impurities on structural material corrosion
5. Impact of impurities in the initial coolant on slag formation process

Bi00 – possible candidate bismuth grade for the use in Pb-Bi coolant

C1 – possible candidate lead grade

Tank system of coolant loading

Loading system is **independent** from work at the I circuit of reactor facility



Name	Value
Total coolant volume, m ³	<2000
Tank volume, m ³	1.5-250
Coolant volume in tank, m ³	1-200
Number of tanks, pcs.	1-10
Power of electric circuit, KW	<100
Pump efficiency, m ³ /h	<20

Elements of purification and control system:

- Hydrogen purification.
- Oxidizing refining.
- Filtration.
- Sampling, oxygen thermodynamic activity control.

Control and regulating systems of oxygen potential

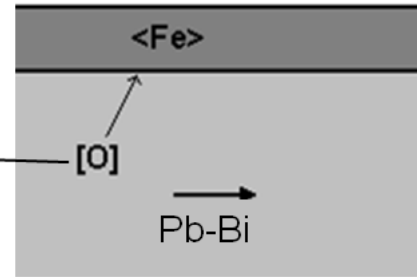
- 1. Provision of initial corrosion resistant passivation of structure material surfaces.**
- 2. Maintenance of the coolant oxygen potential during current operation of reactor facility.**

Solid phase dosing method (regulating) of dissolved oxygen in coolant

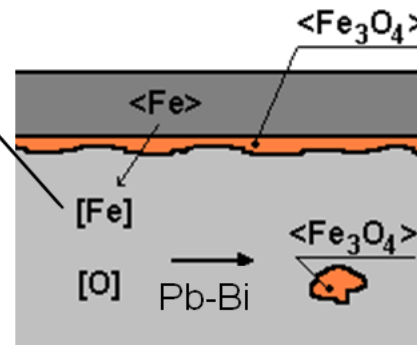


Dissolved oxygen

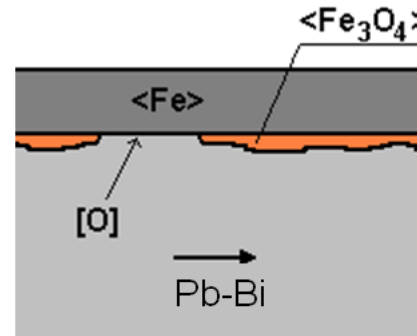
Dissolved iron



Initial condition of steel surface (without protective oxide film)



Start protective oxide film. Oxidation of diffusant impurities from steel



Recovery of damage in protective oxide film

Method options:
 $C_{[O]} = 1 \cdot 10^{-3} \div 1 \cdot 10^{-11} \% \text{mas.}$



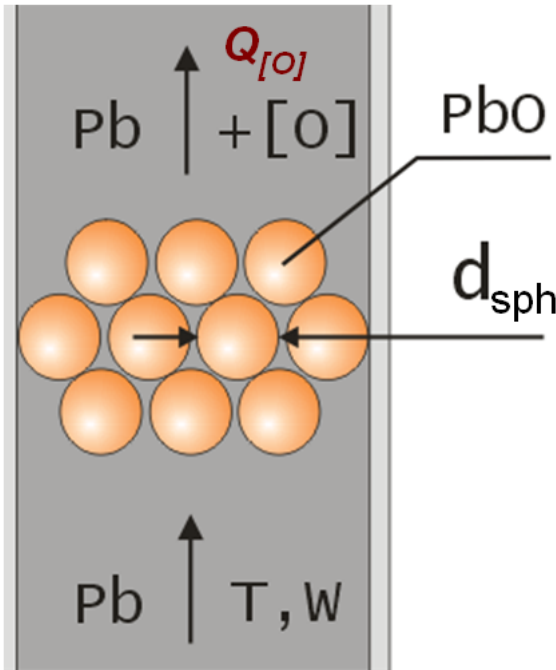
$$Q = K_p \cdot S \cdot (C_s - C_{[O]}),$$

$$K_p = f(T, Re, C_{[O]}),$$

where K_p – dissolution rate factor;
 S – dissolution surface;
 C_s – oxygen saturation concentration;
 $C_{[O]}$ – oxygen concentration in dissolved zone



Dependence of dissolution rate factor of PbO spheroids on temperature and rate of lead coolant



$$Q_{[O]} = K_p \cdot (1 - a_{[O]}) \cdot S_p, [g_{[O]}/h], (a_{[O]} \ll 1)$$

for $Re = 1000 \div 5000$, $Sc = 30 \div 200$

$$K_p = Sh \cdot D/l \cdot C_s \cdot \rho \cdot 360, [g_{[O]}/(cm^2_{<PbO>} \cdot h)]$$

$$Sh = 8,7 \cdot 10^{-4} \cdot Re^{1,4} \cdot Sc^{0,8}$$

$$Sh = \frac{\beta \cdot l}{D} - \text{Sherwood number}; \quad Re = \frac{w \cdot l}{\nu} - \text{Reynolds number};$$

$$Sc = \frac{\nu}{D} - \text{Schmidt number}; \quad l = \frac{2}{3} \cdot \frac{\varepsilon \cdot d_{sph}}{1 - \varepsilon} - \text{characteristic dimension, m}$$

β – mass transfer coefficient, m/s;

$a_{[O]}$ – oxygen thermodynamic activity;

ρ – lead density, kg/m³;

C_s – oxygen saturation concentration in lead, in mass fractions of 1;

ν – kinematic viscosity, m²/s;

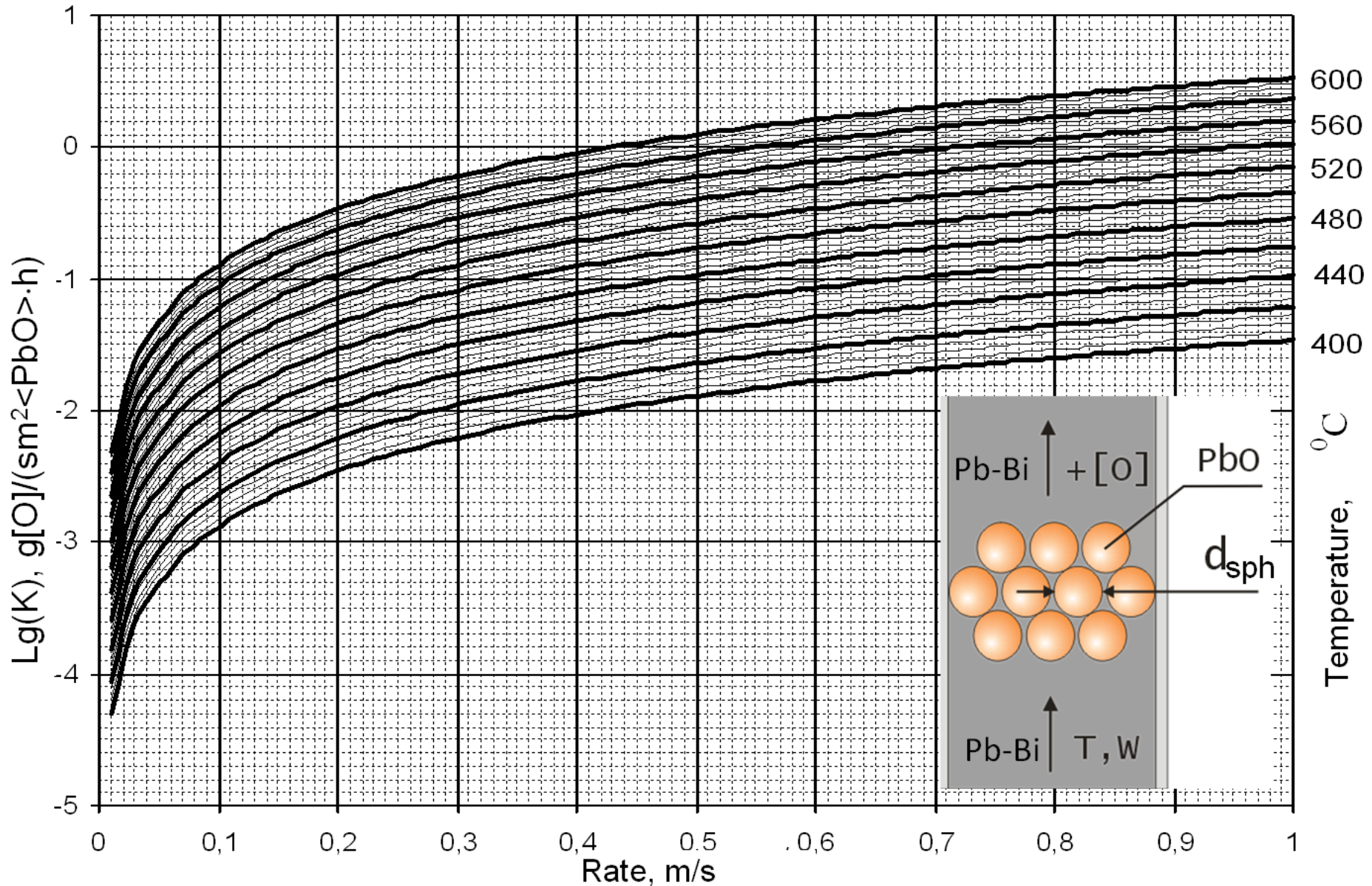
D – oxygen molecular diffusion coefficient in lead, m²/s;

ε – porosity of PbO spheroid layer (accepted as 0.4 for layer of spherical particles);

d_{sph} – spheroid diameter, m;

S_p – dissolution surface of PbO, cm².

Dependence of PbO spheroid dissolution rate factor on coolant temperature and rate

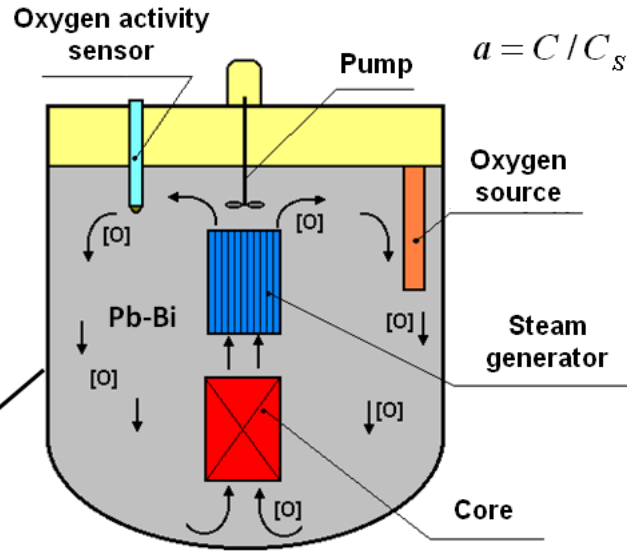


Control and regulating of oxygen potential in heavy liquid metal coolant

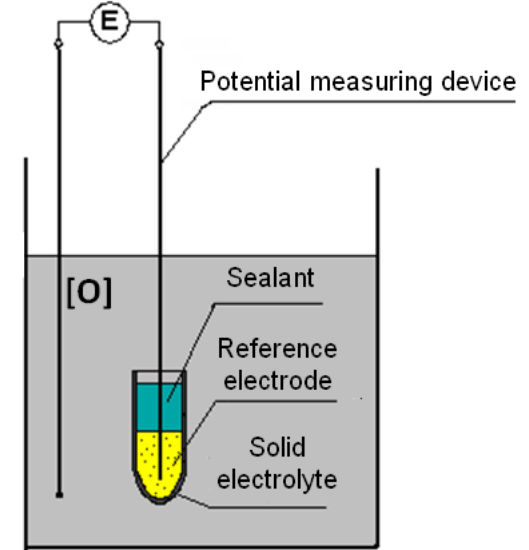
Oxygen is needed for generating and maintenance of strong protective coating on the surfaces of structural materials.

Excess of oxygen in the coolant results in slag deposition. Low oxygen can result in development of liquid metal corrosion.

Up to several tens thousand square meters of cover is needed for structure materials of primary circuit of NPP in the initial operation period



$$a = C / C_s \quad \lg C_s (\% \text{ mac.}) = 1,2 - 3400 / (T + 273)$$

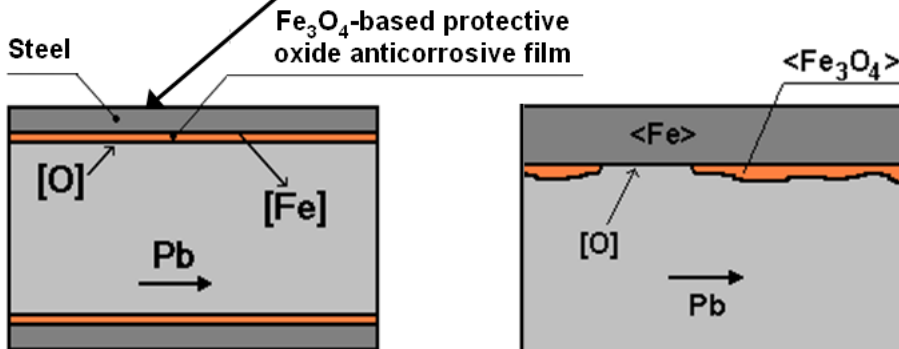
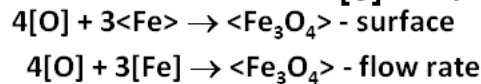


Dependence of EMF (E) of galvanic cell on variation of thermodynamic potential

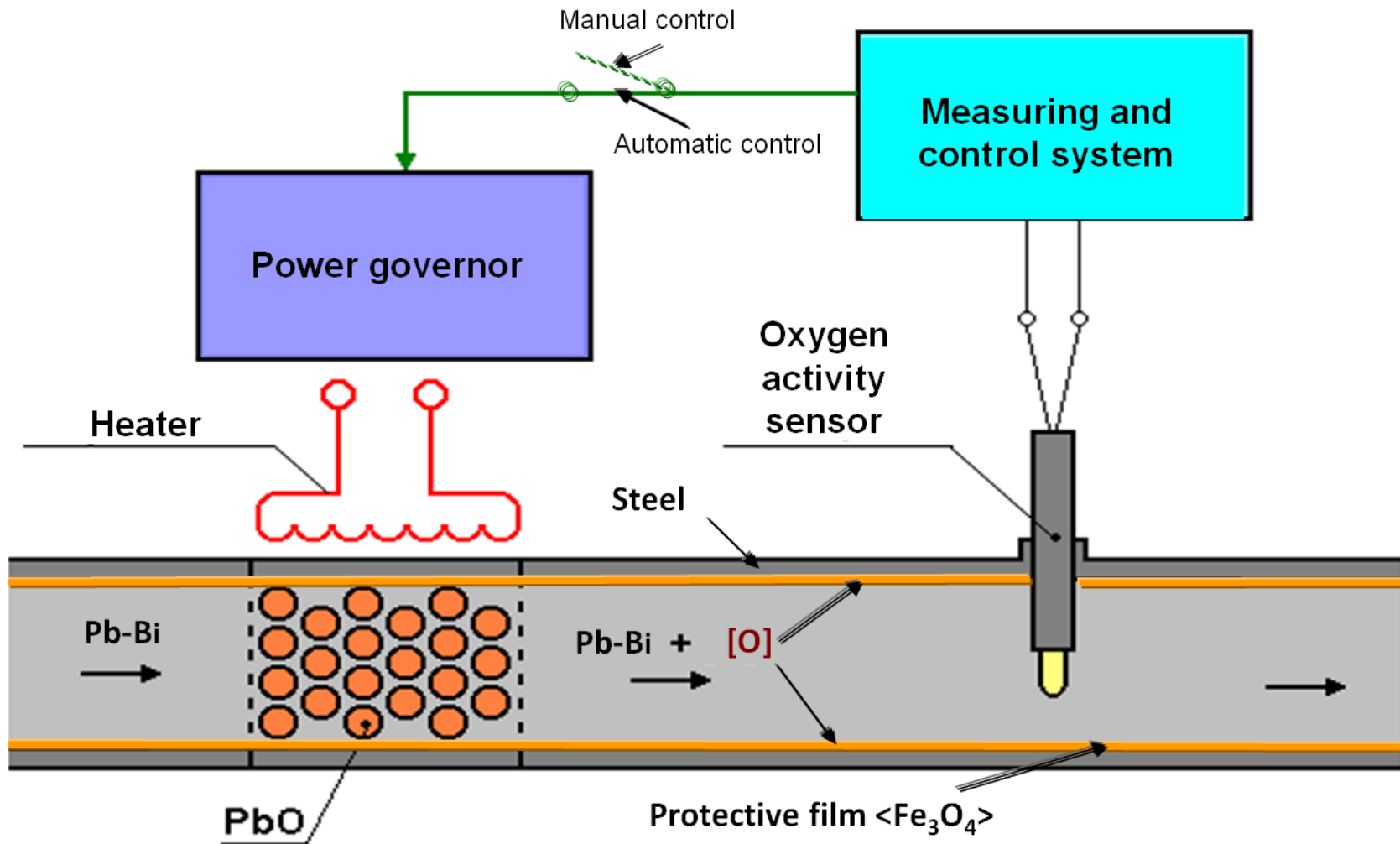
$$E = \frac{R \cdot T}{n \cdot F} \cdot \ln \left(\frac{a_{[O] \text{ r.e.}}}{a_{[O]}} \right),$$

n – number of electrons taking part in reaction;
 F – faraday constant;
 R – universal gas constant;
 T – temperature, K;
 $a_{[O]}$ – oxygen activity in coolant;
 $a_{[O] \text{ r.e.}}$ – oxygen activity in reference electrode.

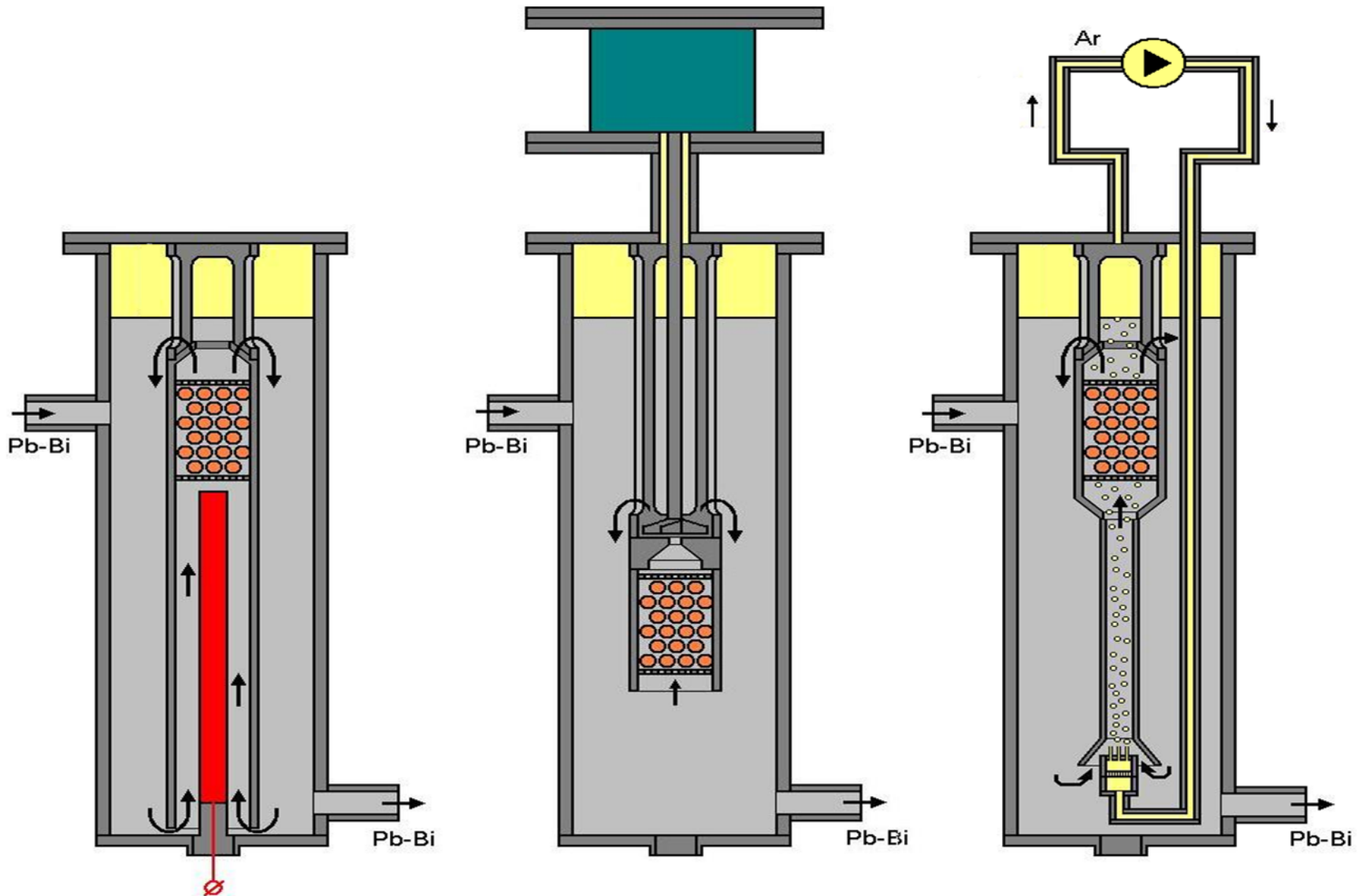
$$C_{[O]} = f(T, E)$$



Principle diagram of the oxygen potential regulating system in HLMC



Types of mass-transfer apparatus to control the coolant composition in circulation circuits



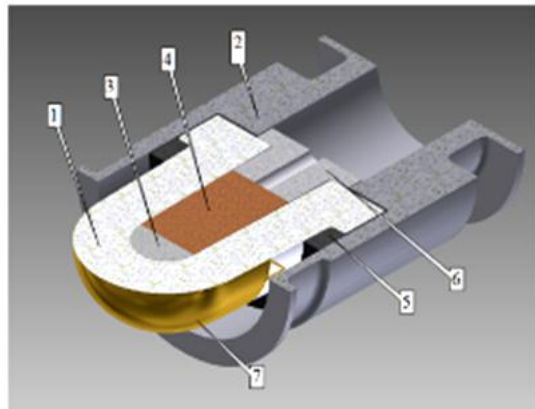
With in-built electric heater

With own axial-flow pump

With airlift pump

Design of sensors for oxygen concentration control in coolant

- Working medium – Pb-Bi;
- Range of oxygen TDA – $10^{-6} - 1$;
- Limit of permissible ratio error of sensor's EMF – 10%
- Range of working temperature – 350- 650°C;
- Working pressure – 0 – 1.5 MPa;
- Coolant rate in the main circuit – 0 - 1 m/s;
- Speed of coolant temperature variation – up to 100°C/s;
- Vibration and water hammers;
- Operational life – up to 30 000h.

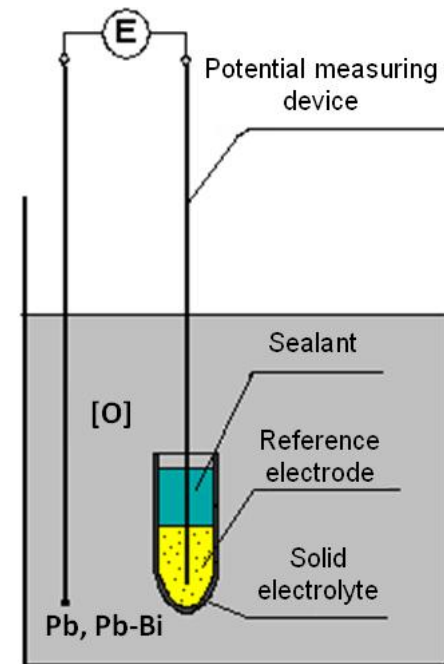


$$E = \frac{R \cdot T}{n \cdot F} \cdot \ln\left(\frac{a_{[O]_{э.с.}}}{a_{[O]}}\right),$$

Where:

$a_{[O]}$ – oxygen activity in coolant;

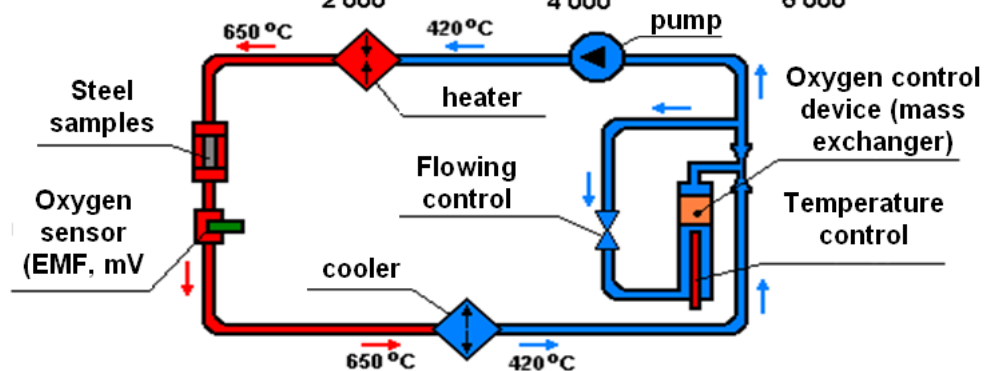
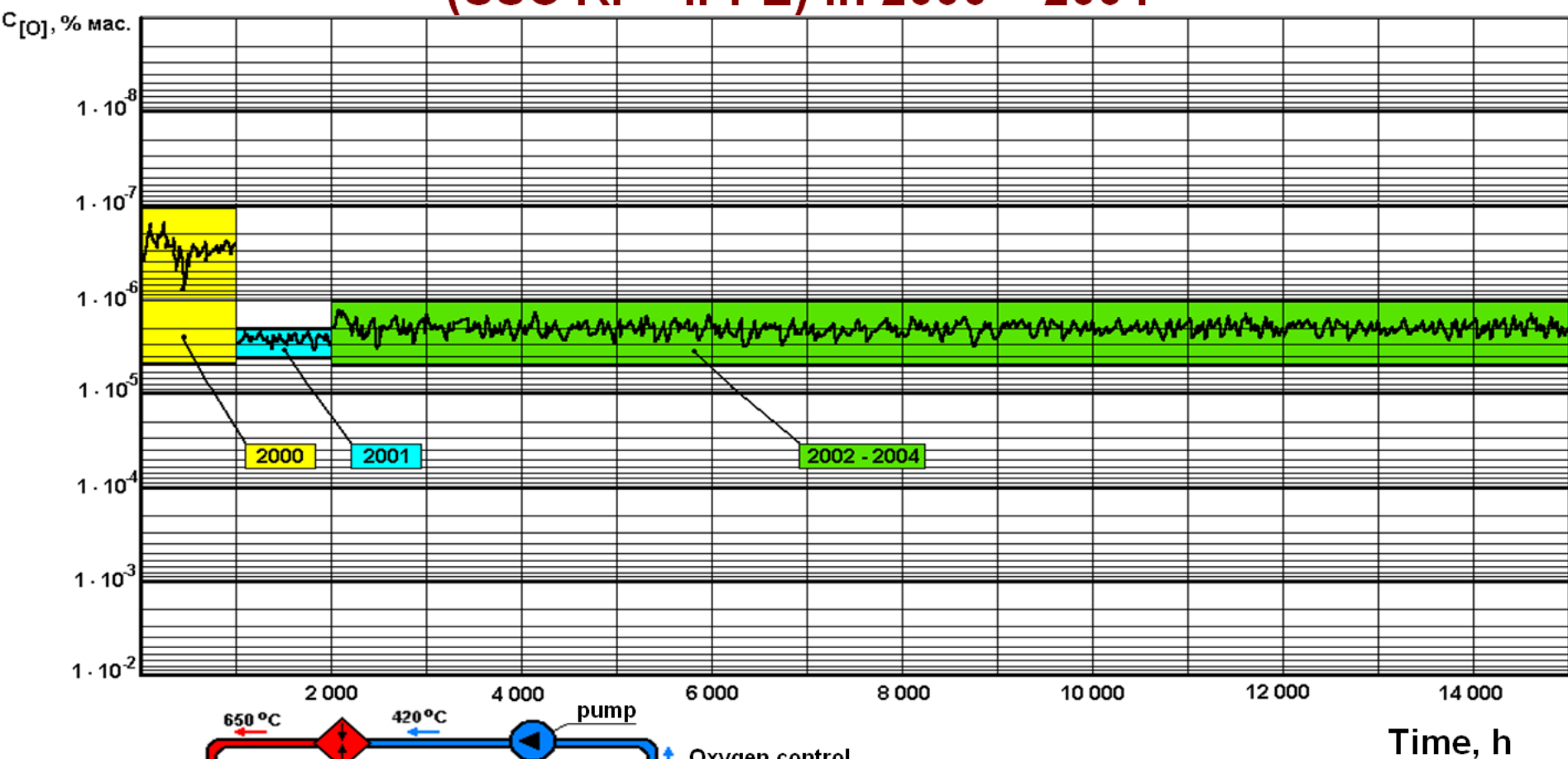
$a_{[O]_{э.с.}}$ – oxygen activity in reference electrode.



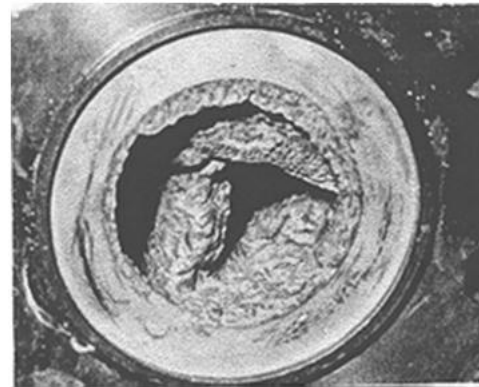
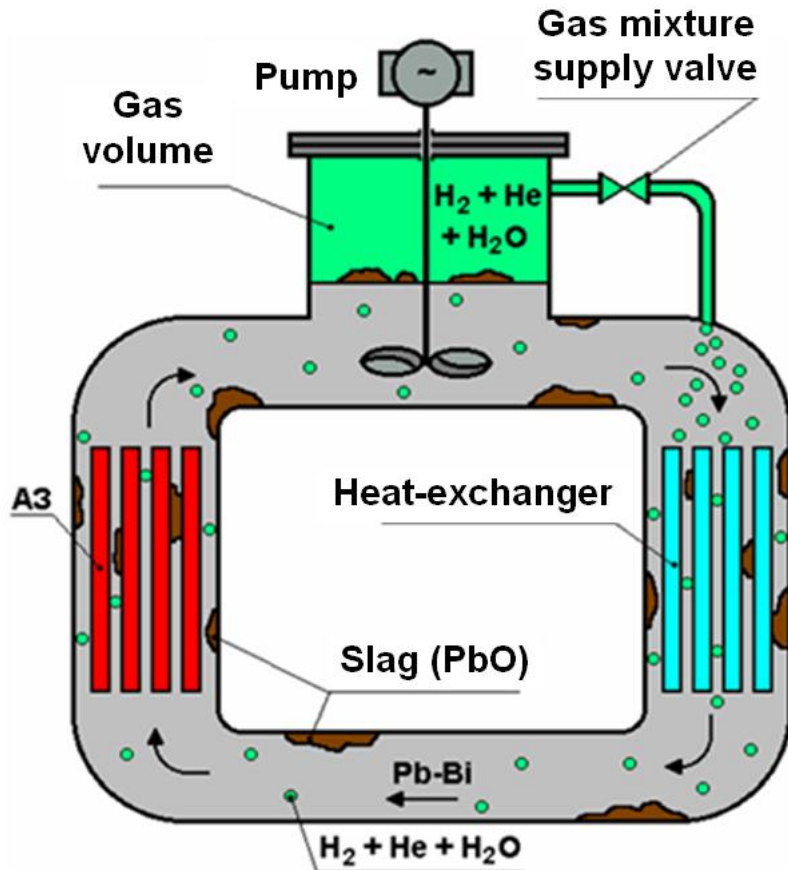
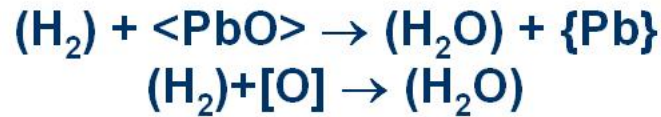
$$a = C / C_s$$

$$\lg C_s (\% \text{ mac.}) = 1,2 - 3400 / (T + 273)$$

Example of the oxygen potential control in coolant during long operation life at test facility CM-2 (SSC RF - IPPE) in 2000 – 2004



Coolant purification and surface cleaning from deposits with hydrogen-containing gas mixtures



Slag deposit in circuit during tests of MCP in МОЦКТИ (Moscow branch of the Central boiler-turbine Institute) not using CTS (1972, records)



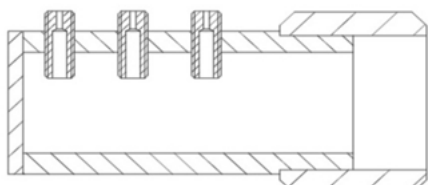
Circuit section with the use of CTS (after introduction of cleaning system)

Periodical coolant purification and cleaning of circuit surfaces from deposits with hydrogen-containing gas mixtures (1 per 5 – 10 years)

Experimentally proved disc gas handler as the main device for hydrogen purification

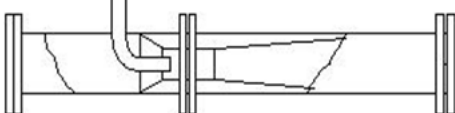
Prototypes of gas introduction devices and their drawbacks:

Nozzle extension



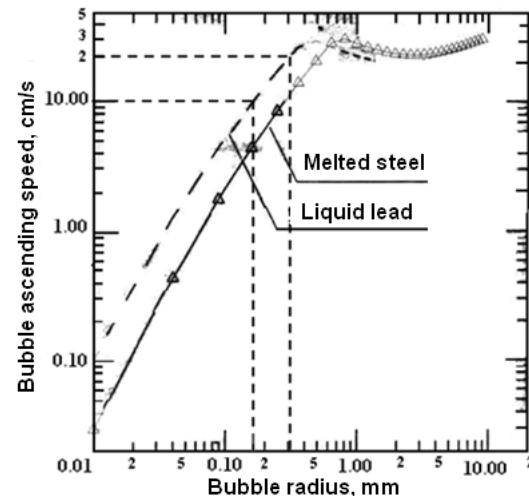
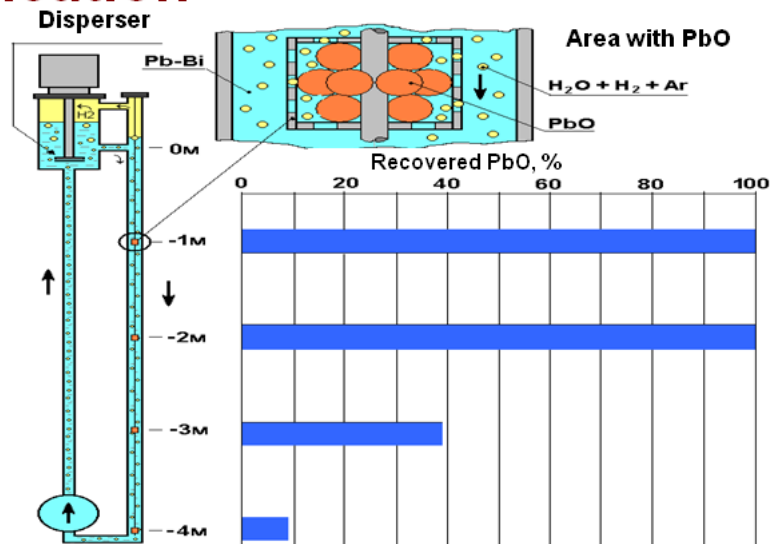
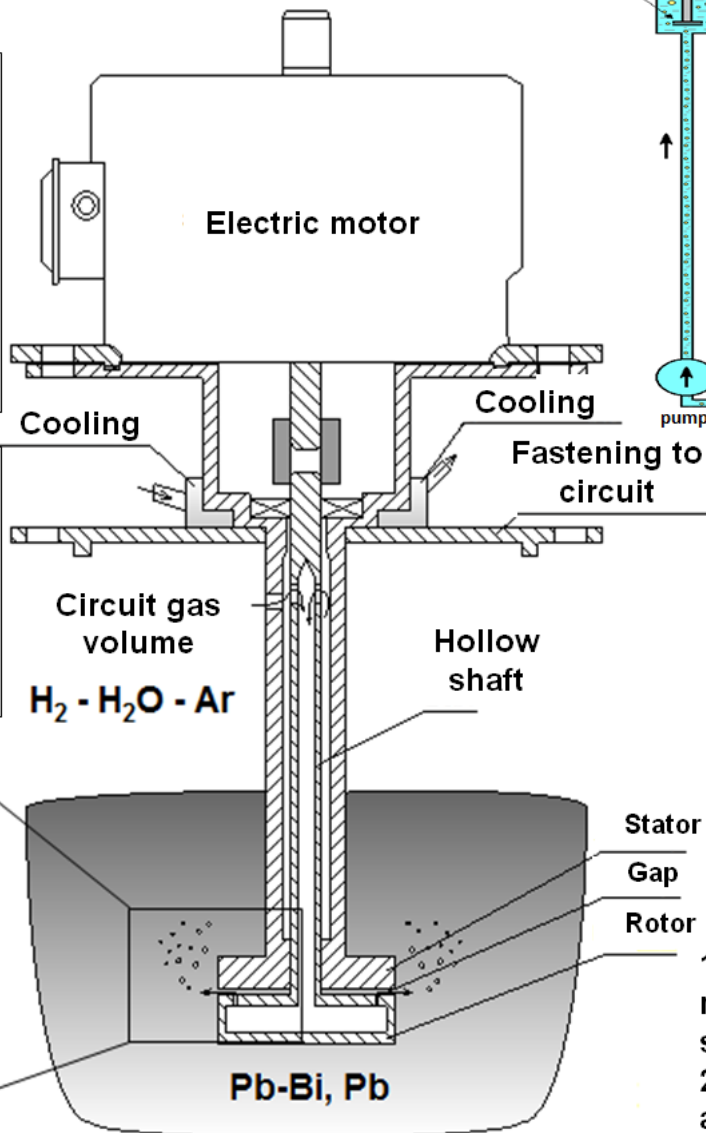
(problems with gas dispergation)

Ejector



(forced flow is needed in pipes)

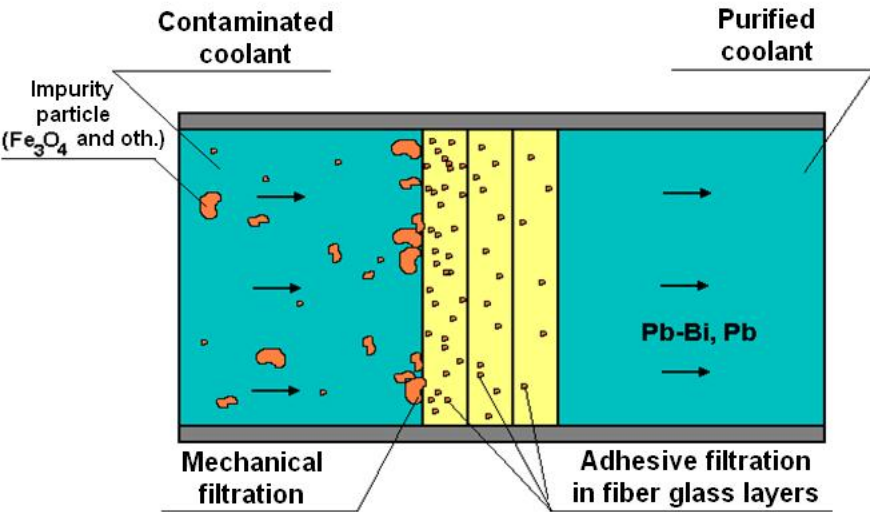
Accepted option – disc gas handler



Advantages:

1. Small bubbles generation (less than 100 mkm) with effective delivery to specified section of circuit
2. Possibility of assembling in circuit of any arrangement
3. Simple design

Filtration of HLMC

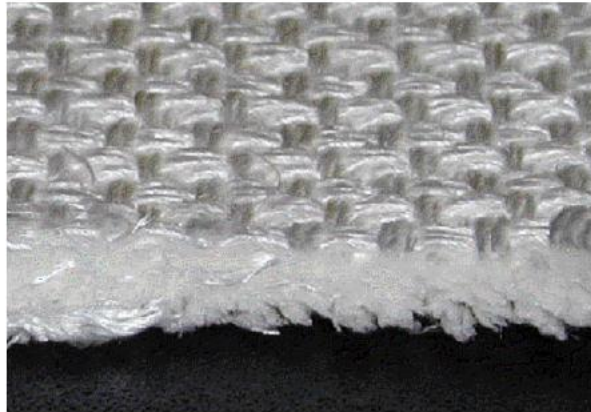


Continuous cleaning filter is intended for HLMC purification from mechanical impurities generated in primary circuit as result of HLMC interaction with constructional steels.

Filter operation principle is based on continuous retention of suspended impurities independently on their nature, concentration and sizes in the volume of filtering material.

Filter can be improved to make possible HLMC simultaneous purification from suspended polonium compounds.

Filtering materials of space-occupying type



Fiber glass MKTT-2,2A
 $\text{SiO}_2 > 95\%$,
Fiber diameter $\sim 6\text{mkm}$,
Heat resistance - 450°C



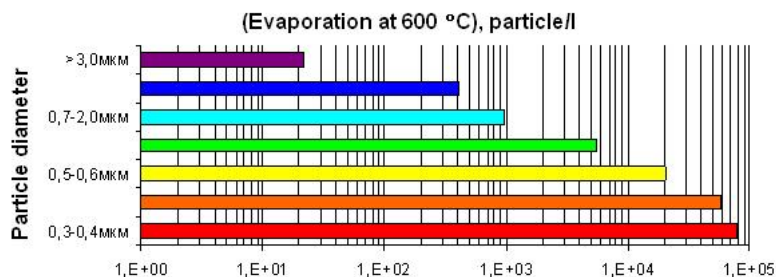
Metal fabric
thickness 2-6 mm,
Fiber diameter - 40mkm ,
Heat resistance - up to 600°C



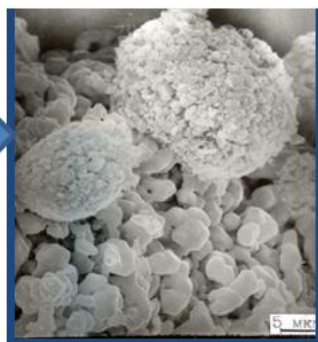
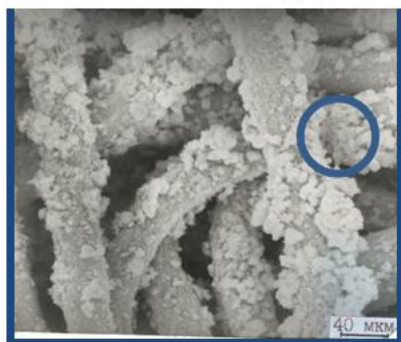
Fused corundum
(screened size Al_2O_3)
Granular size - 0.8-1.2 mm,
Heat resistance $> 1000^\circ\text{C}$

Purification of cover gas from aerosols of heavy liquid metal coolants

Aerosol size-consist and concentration

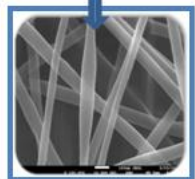
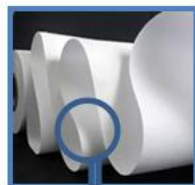
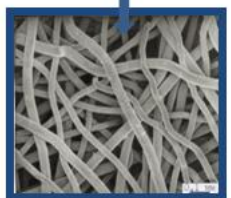


Micrograph of lead aerosol particles trapped by metal felt

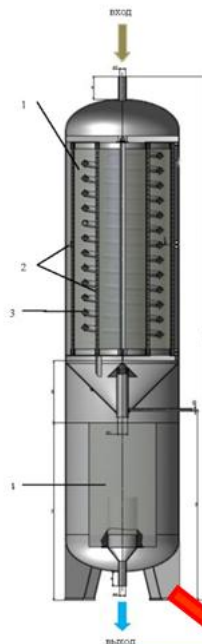


High-temperature filtering material

Low-temperature filtering material



Two-stage filter for gas purification during engineering operations in gas circuit

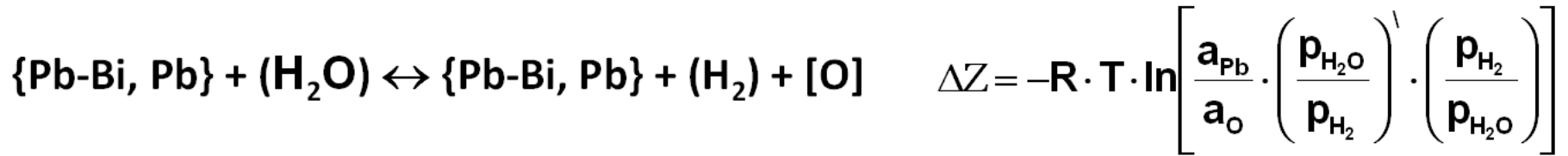


Name	Parameter
Rated capacity, not less than, m ³ /h	10
Air flow resistance, not more than, Pa	
- initial	300
- maximum permissible	1500
Purification efficiency for particles d ≥ 0.3mkm, not less than, %	99.9



Experimental research of gas filter at test facility CIAΦ-1 (test facility for aerosol filter testing)

Pb, Pb-Bi coolant interaction with water



[O] – dissolved oxygen

$$a_{[\text{O}]} = \frac{C_{[\text{O}]}}{C_{[\text{O}]}^s};$$

where $a_{[\text{O}]}$ – oxygen activity;

$C_{[\text{O}]}$ – current concentration of dissolved oxygen;

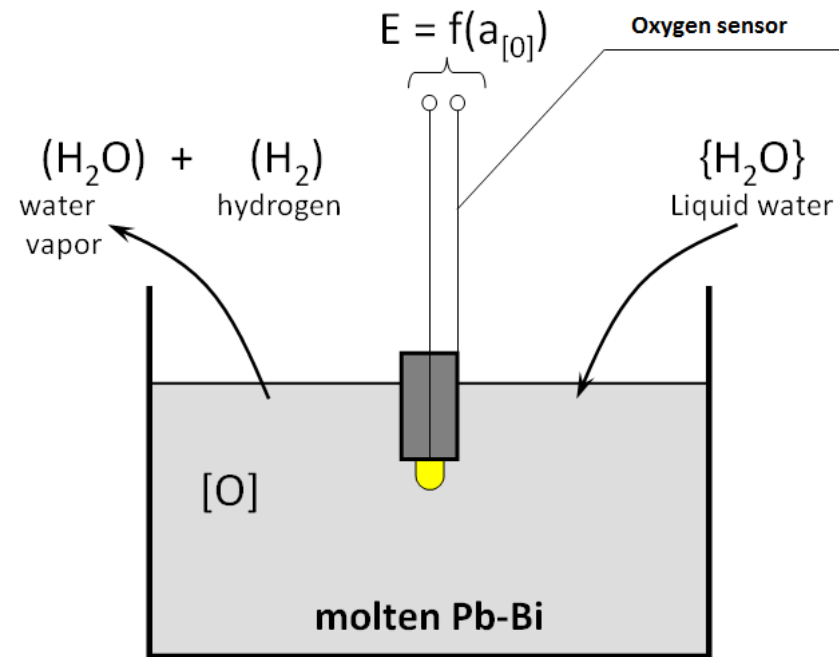
$C_{[\text{O}]}^s$ – concentration of melt saturation with dissolved oxygen;

ΔZ – actual variation of the reaction Gibbs free enthalpy;

T – temperature, K;

$\left(\frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2}} \right)'$ – ratio of partial water vapor and hydrogen in equilibrium with coolant with dissolved oxygen activity = 1;

$\left(\frac{P_{\text{H}_2}}{P_{\text{H}_2\text{O}}} \right)$ – ratio of partial hydrogen vapor and water vapor in initial water.



Thermodynamics of HLMC interaction with “H₂–H₂O” mixtures

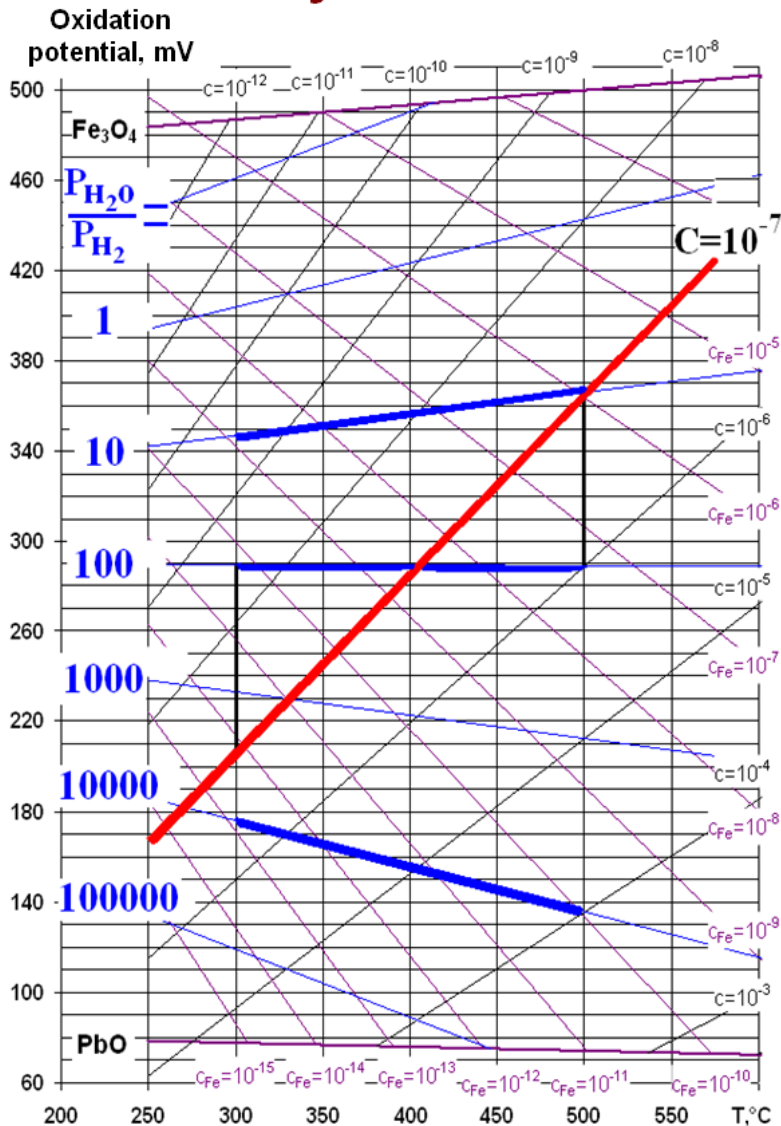
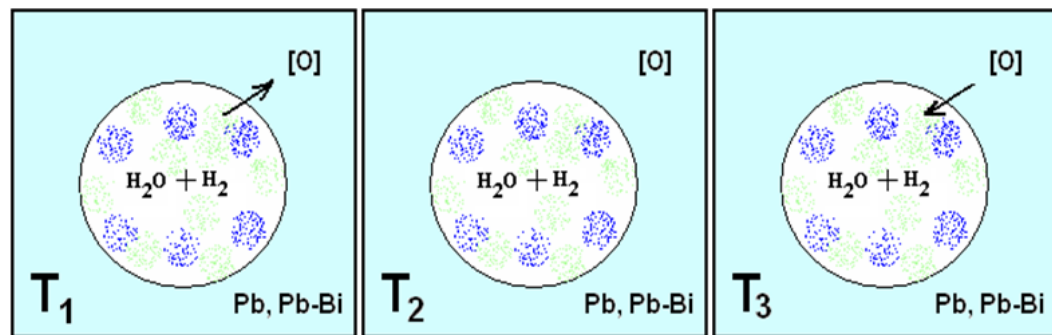


Diagram E – T – C – PH₂O/PH₂
for Pb-Bi melt

$$E (V) = 0,13 - 1,5 \cdot 10^{-5} \cdot T \cdot (1 + 6,6 \cdot \lg a_O) - Pb$$

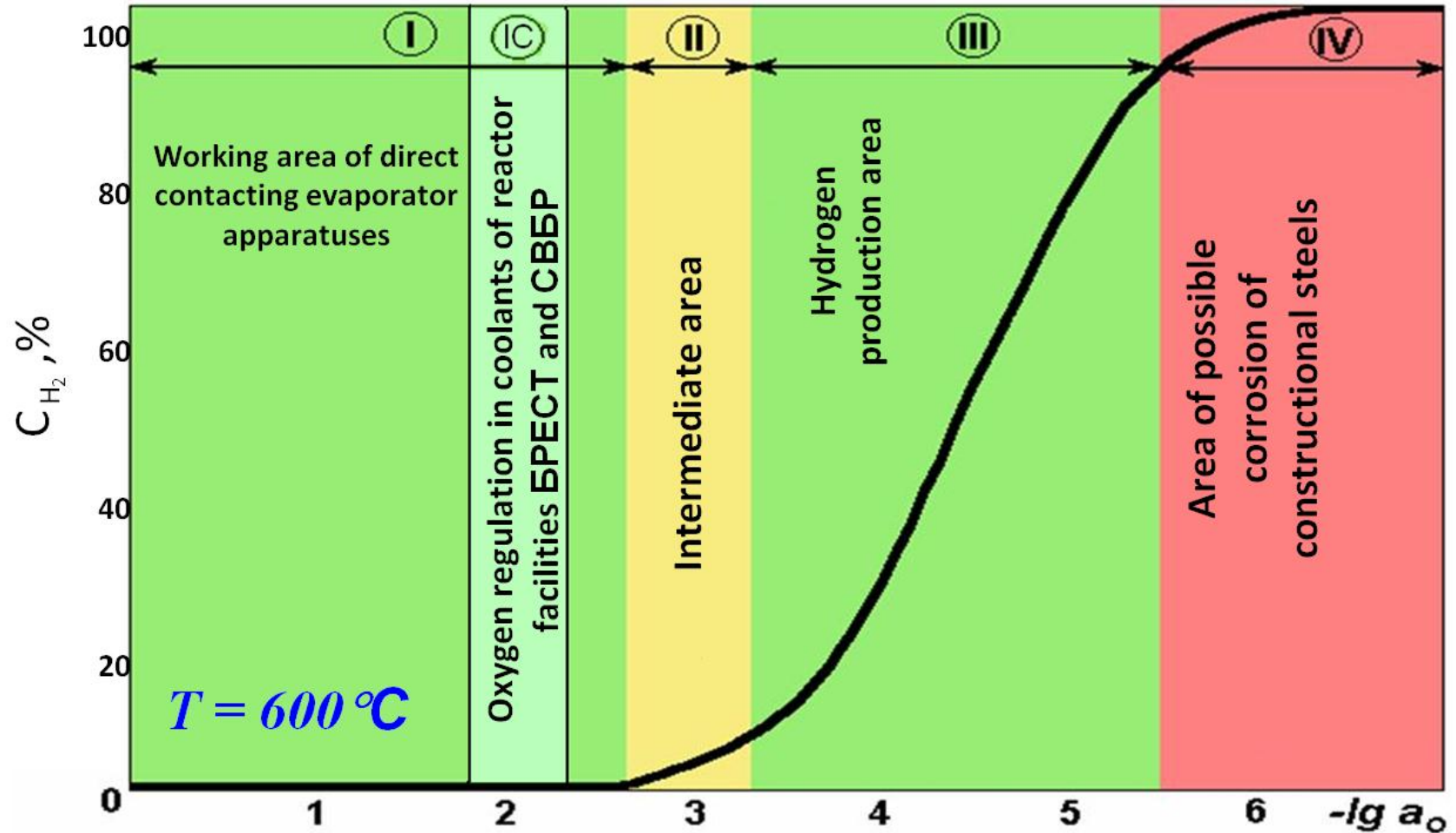
$$E (V) = 0,09 - 1,8 \cdot 10^{-5} \cdot T \cdot (1 + 5,6 \cdot \lg a_O) - Pb-Bi$$



$$C_O = f(V_{HLMC}, V_{gas}, T_{cold}, T_{hot}, F_{cold}, F_{hot}, \dots)$$

Thermodynamic properties of “H₂ – H₂O” and HLMC are different. Different interaction scenarios (oxidation, deoxidation, equilibrium) are possible during their joint transportation along different temperature zones of circulation.

Pb-Bi coolant interaction with water



$$a = \frac{C_o}{C_{os}}$$

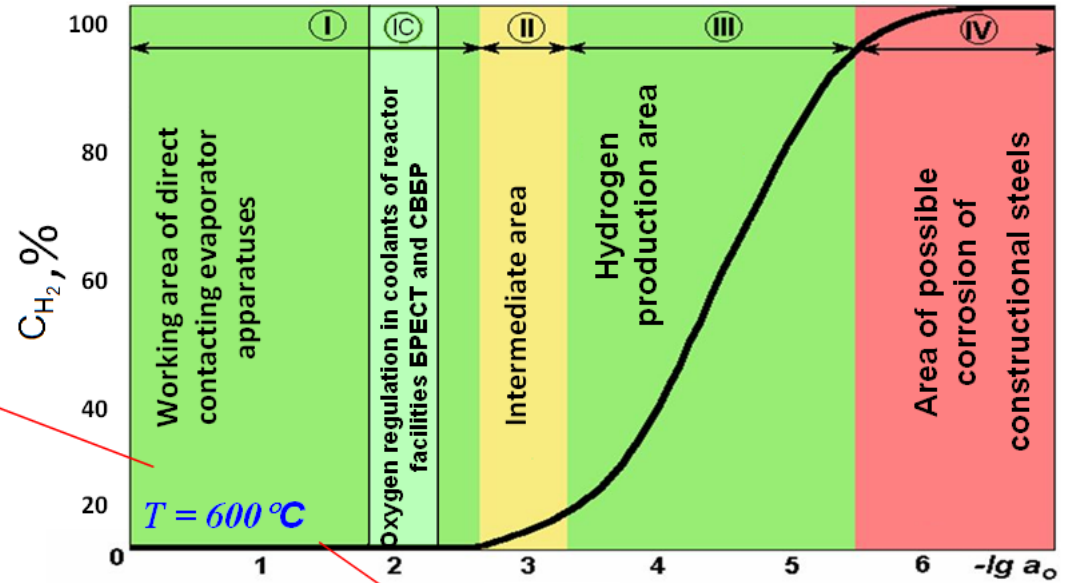
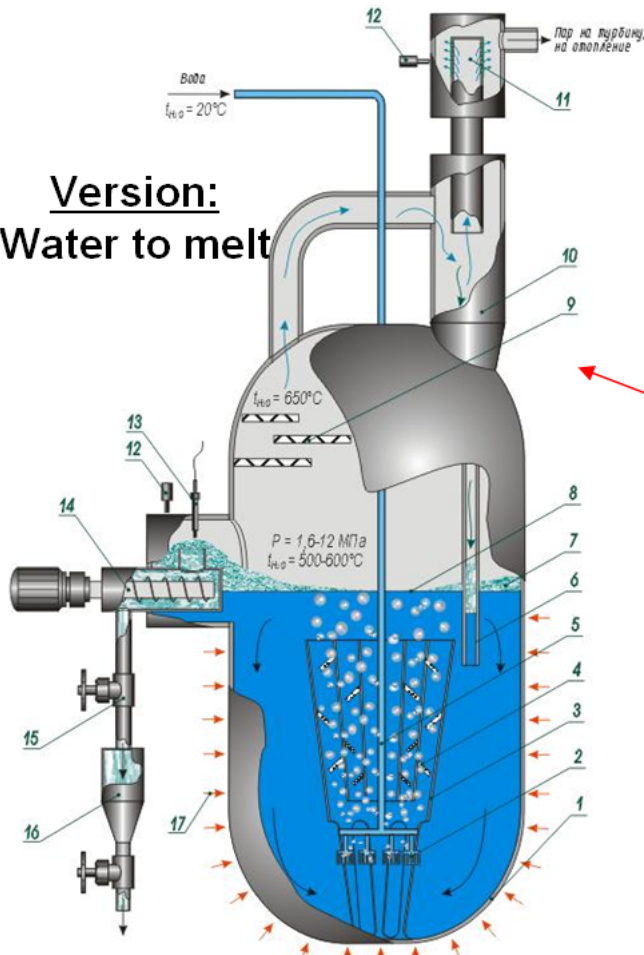
a – oxygen activity

C_o – oxygen concentration in Pb – Bi

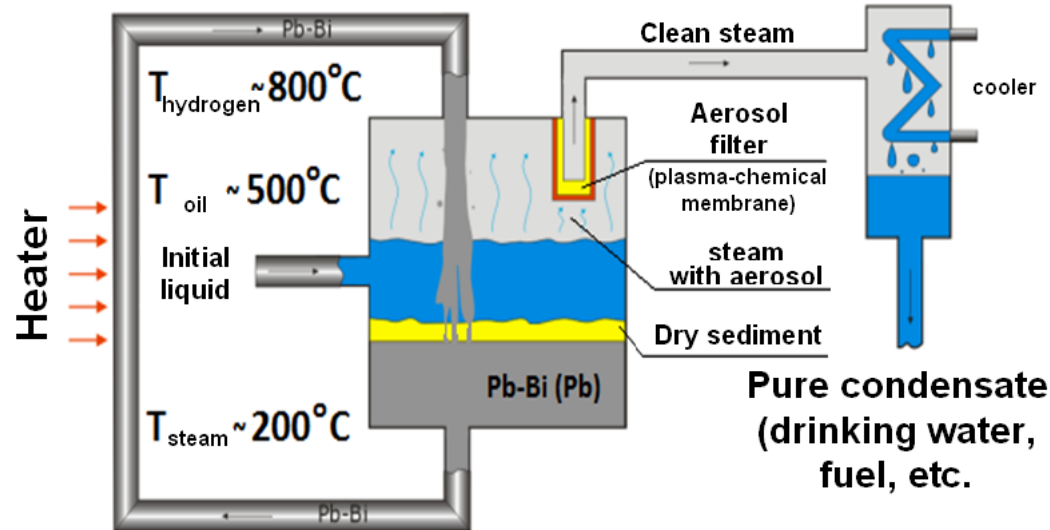
C_{os} – saturation concentration of Pb – Bi with oxygen

Heating and evaporation of liquids during direct contacting liquid metal heating

**Version:
Water to melt**



Version: melt to water



- 1 – body; 2 – bubbler; 3 – guiding ways;
 4 – swirler devices; 5 – water supply line; 6 – sediment discharge; 7 – sediment; 8 – LMC surface; 9 – impingement plates; 10 – cyclone; 11 – high-temperature filter; 12 – magnetostrictor; 13 – level sensor; 14 – conveyor; 15 – valve; 16 – storage device; 17 – heater

Coolant management after reactor facility decommissioning

The following concept is under consideration:

For Pb-Bi cooled reactor facilities

- coolant purification from radionuclides in reactor facility;**
- coolant unloading;**
- recycling.**

For lead cooled reactor facilities

- coolant unloading through filter to special tanks;**
- purification;**
- recycling.**

Summary

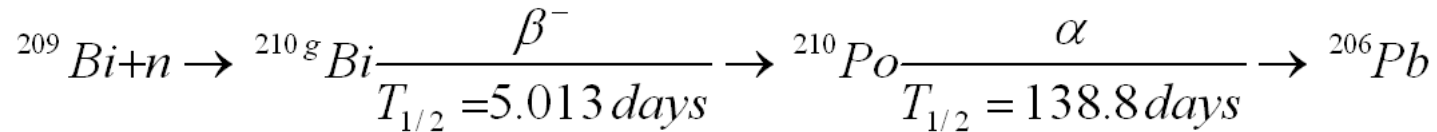
1. Problems of heavy coolant technology are solved within the whole life cycle of reactor facility. Coolant technology includes R&D to substantiate, design and develop the methods and instruments as well as direct execution of processes during the coolant preparation, startup of reactor or research facilities, their operation and decommissioning.
2. Selection of needed methods and instruments of heavy liquid metal coolant technology for reactor facilities under development is based on 62-years experience of Pb-Bi coolant investigation and results of R&D taking into account specificity of new generation of reactor facilities.
3. Methods and instruments of coolant technology under development include the hydrogen purification system of coolant and circuit from slag-making impurities, system of dissolved oxygen control in coolant for steel corrosion protection, filtering system for coolant and cover gas, coolant purification system from radioactive impurities, coolant control system both in reactor and out-of-reactor conditions and the other systems.
4. Methods and instruments of coolant technology under development provide thorough coolant preparation prior to its loading and during the loading to reactor facility circuit, execution of package of measures in startup modes and minimal diversion of RF personnel to the coolant technology process execution during the operation.

Thank you for attention

I will invite you to a fourth international conference
"Heavy Liquid Metal Coolants"
(October, 2013, Obninsk)
(mail to pmartinov@ippe.ru)

Polonium Po^{210} problem in NPP circuits with HLMC

Po^{210} GENERATION: reaction on Bi nuclei with radioactive β -decay of Bi^{210g} and next generation of α -radioactive Po^{210} :



RADIOACTIVITY: Po^{210} is pure α -emitter in practice, $E_{\alpha} = 5.3\text{MeV}$, its activity can be $4 \cdot 10^{10}$ Bq/kg (1.08 Ci/kg). By the end of run of CBEP-100 (50 thous. hours) total Po^{210} activity in coolant will become $3.2 \cdot 10^{16}$ Bq ($8.64 \cdot 10^5$ Ci), specific activity – $1.7 \cdot 10^{11}$ Bq/kg (4.5 Ci/kg), which corresponds to about 1mg of Po^{210} per 1 kg of coolant (10^{-6} rel.units); total activity in gas system is $5.7 \cdot 10^{10}$ Bq (1.54Ci), and concentration activity – $6.0 \cdot 10^5$ Bq/m³.

CONDITION of Po^{210} IN HLMC: Po^{210} is mostly molten in the form of lead polonide $PbPo$, its small amount only (less than 1%) transfers to gas phase ΓC in the form of radioactive aerosols; mentioned condition of Po^{210} in melt and gas allows its removal with the help of appropriate filters.

HAZARD: there is no hazard from Po^{210} under normal operation conditions of NPP and tight circuit. Hazard appears during inter-circuit leaks, primary circuit depressurization during scheduled repair, nuclear fuel reloading or emergency spillage of radioactive coolant into attended room. Calculation shows that in condition of emergency leak from gas system with relative constant speed 1 h^{-1} the level of Po^{210} aerosol concentration activity in the air of reactor box is $\sim 140 \text{ Bq/m}^3$, which is about 50 times exceeds the maximum permissible level for personnel.

TECHNICAL MEASURES: execution of comprehensive program accounting Po^{210} radiation danger in all NPP modes as well as initial events and different scenarios of beyond design basis accidents; development of thermally and chemically stable sorbents for non-reversible extraction of Po^{210} from HLMC with the use of experience accumulated at SSC RF – IPPE and other institutions; development of appropriate sorption-filtering devices for Po^{210} extraction from coolant and reactor gas vessel and local storage; development of Po^{210} α -radioactive detector.

FOREIGN INVESTIGATION: increased attention is paid to polonium problem in NPP with lead-bismuth coolant abroad, especially in Japan. Extensive research of coolant purification from polonium with selection of filtering materials are carried out at Technological Institute of Tokyo under supervision of professor Toru Obara. Several papers devoted to this problem were recently published in international journals.

Calculation assessment of polonium activity in reactor room (RR) of CBEP-100 reactor facility under design and beyond design basis accidents

- **Design basis accident:** loss of tightness of gas system with constant relative speed of gas leak 1 h^{-1}
- **Beyond design basis accident:** full pipe break of evaporator module (large inter-circuit leak) with simultaneous gas system emergency depressurization
- **Concentration activity of Po^{210} in RR air permissible for personnel** 2.7 Bq/m^3
- **Maximum concentration activity of Po^{210} in RR air under design basis accident** $1.4 \cdot 10^2 \text{ Bq/m}^3$
- **Maximum concentration activity of Po^{210} in RR air under design basis accident and availability of purification - HLMC and gas filters** 1.4 Bq/m^3
- **Maximum concentration activity of Po^{210} in RR air under beyond design basis accident** $2.8 \cdot 10^2 \text{ Bq/m}^3$
- **Maximum concentration activity of Po^{210} in RR air under beyond design basis accident and availability of purification - HLMC and gas filters** 2.8 Bq/m^3