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ELEKTROSORBTION PHENOMENA IN LAYERS OF SHIELD-VACUUM HEAT INSULATION OF HYDROGEN RESERVOIRS IN EMERGENCY OPERATING CONDITIONS

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1. INTRODUCTION

Emergency conditions in big cryostats in the circumstances of their long-term operation have been discussed in this review. Emergency conditions in cryostats arise at the appearance of considerable heat flows into a cryoagent, which much more exceed the certificate flows and come in over heat bridges and through heat insulation [1-3]. During a long-term operation of big cryostats, especially in the end of the routine maintenance interval, residual hydrogen gets accumulated in heat-insulating cavities. Hydrogen, as a rule, appears as a result of inter-lattice hydrogen diffusion from thick and warm cryostat casing walls into the vacuum cavity. The residual medium of other gases is mainly formed due to the atmospheric air inflow through microloosenesses. Big cryostats always contain microloosenesses. In the beginning of the cryostat operation, they are, as a rule, insignificant, and then grow due to the processes in welds approaching by magnitude to the maximal permissible value.

The most spread strategies of the cryostat superinsulation operation are built on strictly determined heat insulation routine maintenance intervals. At this stage of cryogenic engineering development, designers, as a rule, determine the routine maintenance interval as one year. However such attitude leads to considerable operational and energy costs. It would be more expedient to build the planning strategy of the routine maintenance interval with the account of changes in the cryostat design condition. In a number of works [4, 5], a possibility is postulated to predict the beginning of the extreme cryostat operation period as well as to plan the optimal duration of routine maintenance intervals. At the same time, as the operational practice has shown, when the routine maintenance interval in cryostat superinsulation is exceeded under certain conditions, some phenomena impeding the normal cryostat operation may appear. However the conduction of extreme planned experiments on full-scale big cryostats has allowed to prove the possibility of management of these effects in order to eliminate a negative effect and obtain a positive effect in case when it is impossible to conduct an emergency superinsulation routine maintenance. In addition, the analysis of these phenomena has permitted to build models, which can be useful at the development of fundamentally new versions of superinsulation embodiments. These processes can be completely stopped and eliminate their extreme danger.

The proposed review covers discussions and analysis of the data being accumulated by the present time on proceeding of electro-sorption processes in screen-vacuum heat insulation (SVHI) layers of big cryogenic reservoirs and cryogenic pipelines [6-7]. Their influence on the cryogenic products volatility as well as on the safety reduction of thermostatically controlled objects has been demonstrated.

A special attentions has been paid to the field effect, the Bardeen-Brettain-Shockley gas-water cycle, the electro-adsorption effect, metastable states of superinsulation surface, kinetics and dynamics of the residual atmosphere of very big cryogenic reservoirs with insignificant effusion leaks, the cryogenic liquid volatility, the determination of the heat inflows to a cryogenic liquid in the conditions of ambient parameter changes. For the first time, the following recently discovered phenomena in big cryogenic reservoir superinsulation have been described in the references being reviewed [6-9]: an effect of effusion induced hydrogen superinsulation instability, an effect of effusion induced heat-conductive superinsulation instability in cryogenic-vacuum objects, an effect of multiplication of the number of desorbing hydrogen molecules in respect to the inflowing moist air molecule magnitude. The effects in heat insulation can be controlled. In order to create new heat insulation samples with a high exergy efficiency and a high safety degree, new heat-insulating structures and designs should be developed [8, 9].

The main tendencies of further superinsulation development have been determined. A fundamentally new approach to the superinsulation designing and calculation has been demonstrated, which, apart from radiation and convection heat conduction mechanisms, takes account of convection component composition variations. In addition, the convection component variations occur due to the change of residual water composition and concentration as a result of the electro-sorption process. The electro-sorption process arises at the availability of clearly expressed hydrogen residual atmosphere on superinsulation heat screens with water concentration changes in the air inflowing through microloosenesses into the heat insulation cavity [6, 7].

Theoretical models for the appearance of effusion induced hydrogen and heat conduction instabilities of the superinsulation have been proposed for the first time in this review. Thermodynamic description of these new effects has been carried out with the use of analytical thermodynamics mechanisms. On the grounds of variational description of heat and mass transfer processes for a heterogeneous system in the continuum approximation and with the account of electro-sorption processes according to the hydrogen-water cycle of Bardeen-Brettain-Shockley, a formulation of the mathematical model of molecular heat and mass exchange in superinsulation has been derived.

A fundamentally new approach to the superinsulation construction and calculation, which, apart from radiation and convection heat conduction mechanisms, takes account of the convection component variation mechanism. In addition, the convection component variations occur due to the change of residual water composition and concentration as a result of the electro-sorption process. The electro-sorption process arises at the availability of clearly expressed hydrogen residual atmosphere on superinsulation heat screens with water concentration changes in the air inflowing through microloosenesses into the heat insulation cavity.

2. CRYOGENIC OBJECT SUPERINSULATION CHARACTERISTIC

After the superinsulation invention by P. Petersen, quite a little time has passed – only several decades. However the concept of superinsulation operation mechanism has suffered multiple variations. In the course of time these models have allowed to develop a modern superinsulation.

P. Petersen placed screens made of aluminium foil in a vacuum volume and separated them by means of glass-fibre mats. Instead of foil, a polymer film with thin aluminium layers being applied on its both sides is most widely used now.

A number of competing concepts exists as to the heat transfer mechanism in superinsulation. These concepts were sufficiently true in order to develop a sufficiently effective superinsulation. However, in the process of operation of big cryogenic objects, researchers have noted that our ideas on thermal processes in superinsulation are not correspond to reality.

Using the latest views on superinsulation, one can make the following definition.

Screen-vacuum heat insulation (superinsulation) is a system of parallel or concentric (coaxial) gas-permeable metal films applied on a substrate being separated from other by a porous padding manufactured from a material with a high heat resistance coefficient providing a small degree of heat radiation absorption and a small degree of accommodation of the inter-screen gas molecule energy at a high and stable adsorption ability of the metal films.

At the present time, a polyethyleneterephthalate film with the thickness of 12-15 μm with thin layers of aluminium of 0.5 μm thick being applied thereon on both sides are widely used as screens [10, 11]. A low heat conductivity of the film and a small thickness of the aluminium layer reduce the heat transfer along the layers and increase the superinsulation effectiveness in industrial products. For the insulating pad thin-fibre (with the fibre thickness up to one micron) glass materials with low gas release are used. As the distance between the screens is sufficiently large (the packing density normally lies within 10-50 screens/cm), the screen-vacuum insulation operates most effectively at practically the same low pressure values as the pure vacuum insulation, i.e. at the pressure values below 10^{-2} Pa. However the effectiveness of such insulation is far higher than the vacuum and powder-vacuum insulation.

The present-day industrial superinsulation provides a heat flow at the level of 0.3-0.5 W/m^2. Such heat inflow values are realised at the screen number of 45-75, i.e. at the thickness values less than 0.1 m and a small insulation layer mass [11]. The best superinsulation samples within the temperature range of 10 - 350K are characterised by the effective heat conductivity coefficient equal to (2-3)*10^{-5}

W/(m*K), i.e. significantly less that with other heat insulation types. This parameter provides for the preferable superinsulation application for the protection against heat inflows of devices operating at cryogenic temperatures. [11].

A peculiarity of superinsulation is the non-additivity of thermal resistance in respect to the number of screens and the fact that the thermal resistance of insulation practically cease increasing when a certain number of layers has been reached [12].

3. SUPERINSULATION MODERNISATION URGENCY

A rapid growth of interest in superinsulation has been noted in 1970-80s and has been connected with the development of cryogenic engineering, space engineering, aviation, surface and underwater sea fleet. An interest in superinsulation was also resumed in the end of 1990s and in the beginning of the 21st century. The development of hydrogen power industry in the symbiosis with nuclear power industry is a part of prospective national programs of a number of developed states. The placement of a nuclear reactor in the world ocean water area for hydrogen production and liquefied hydrogen transportation to an island is one of the prospective projects of the Japanese power industry development. In order to store reserves of liquid hydrogen, oxygen and other liquefied gases, effective cryogenic reservoirs and pipelines will probably be required.

The volatility of most effective reservoirs is 0.8-1% per day of the total amount of liquid being stored.

Giant dimensions of present-day reservoirs determine a large amount of expenses on manufacture of the internal and external reservoir shells. In order to optimise expenses, the external shell is manufactured from a low-alloyed steel. The latter circumstance leads to increased gas releases of inter-lattice hydrogen into the heat-insulating cavity. The hydrogen content in the casing metal of most spread modern cryogenic reservoirs is 9.5–11 cm^3/100 grams of metal. Zeolites being most widely used in Russian cryogenic engineering in cryoadsorption pumps comparatively well absorb hydrogen in the range of 20.2K and considerably worse at higher temperature.

As a result of long-term reservoir utilisation without a possibility to conduct of obligatory process of technological TIP blowings of heat-insulating cavities (HIC) and KSN regeneration, the amount of residual hydrogen in HIC achieves significant values. The reason of the hydrogen concentration increase in HIC can be both the inter-lattice hydrogen of structural materials and hydrogen inflowing (or diffusing) through microleaks from the internal vessel at the storage of hydrogen therein as well as from the atmosphere. The consequence of it is a considerable increase of the cryogenic liquid volatility. The situation being considered is much more related to the emergency categories and manifests itself to the full extent quite rarely. However engineers dealing with the operation of such systems can rather often observe the phase of appearance of several occurrences of this situation at the normal functioning of the reservoir too, especially at the final stage of the routine maintenance interval.

4. SYSTEMATISATION OF HEAT TRANSFER MECHANISM MODELS IN SUPERINSULATION SYSTEMS

Model 1. It is intrinsic to the initial period of the superinsulation investigation. The superinsulation is considered as a system of parallel layers with surfaces reflecting thermal radiation according to the Stephen-Boltzman law. In addition, the thermal shunting of the layer is absent in the model. This model implied a small degree of surface blackness. The superinsulation according to Model 1 is considered as a system of poor or “defected” reflectors [13]. The following superinsulation damage sources have been proposed: adsorption of water vapours, working liquid and volatile components of the padding material [14] as well as tunnel-radiation phenomena in the area of contact between the padding material and the reflecting surface [15].

Model 2. It describes the superinsulation as a system of screen-reflectors with a small value of the emissivity factor and heat shunted by means of conductive-conduction pads located between them [16-18]. In accordance with this model, the pad's parameters, the fibre diameter, the total thickness of pad, the number of bonds in the padding should be of major importance. In addition, the fibre diameter and the pad's thickness is determined by the number of contact areas in the heat flow direction and the amount of the bundle, which is normally concentrated in the contact area, is determined by the heat resistance of this contacts.

Model 3. It describes the superinsulation as a system of effective screens of radiant energy shunted by pads, which entire heat resistance is concentrated in the area of their contact with the screens. According to this model, the pad thickness is of secondary value [19-21].

Model 4. It describes the superinsulation as a system of screens with a small degree of surface blackness shunted by the interlayer gas being in the free molecular mode (the Knudsen criterion >1). According to this model, the pad thickness is insignificant, the main importance belongs to the number of sections, into which the gas space is divided and the residual gas pressure in this space. The pad parameters are important only insofar as they influence on the residual gas pressure [22–28].

Model 5 (belonging to the author of this review). It describes the superinsulation as a system of screens with a small degree of surface blackness shunted by the interlayer gas being in the free molecular mode. According to this model, the pad thickness is insignificant, the most important are the number of sections, into which the gas space is divided, the screen material, the composition and pressure of residual gases in this space as well as the effusion magnitude and composition. The pad parameters are important only in connection with the fact that they influence on the residual gas pressure [6,7].

Model 6 (G. G. Zhun's model). It describes the superinsulation as a peculiar pump [29].

Model 7 (belonging to the author of this review). It describes the superinsulation as a “quasicapacitor”, in which the charge generator on the superinsulation screens is the evaporating cryoliquid [30].

Model 8 (belonging to the author of this review). It describes the superinsulation as a system being unstable in warm layers and having the number of adsorption centres, which varied depending upon the ambient conditions.

4. TYPES OF FUNCTIONAL INSTABILITY OF THE SUPERINSULATION

In works [6,7], on the grounds of thermodynamic description of non-equilibrium systems with the use of phase space formalism and the multi-dimensional fundamental equation at the availability of diffusion, the processes of **chemical instability generating fluctuations in a macroscopic system including the vacuum cavity of a cryogenic reservoir with the superinsulation inside it** have been explained. In addition, the macroscopic system consists of a large number of particles $N \rightarrow \infty$ and occupies the macroscopic volume $V \rightarrow \infty$ at finite density N/V. Moreover, an external influence is directed to the system – in the form of low-magnitude fluctuations of the diffusive flow with the donor gas and water vapour concentration changing in composition. In work [6], small and intermediate magnitudes of fluctuations have been considered, which most effectively deflect the system from the unstable state.

This review covers the consideration and analysis of the data being accumulated by the present time **on passing of electro-sorption processes in screen-vacuum heat insulation (superinsulation) layers** of big cryogenic reservoirs.

Periodic variations of the concentration of the effusion flow in the link with electro-sorption processes lead to the appearance of chemical pseudowaves. In a diffusion system with chemical reactions, information is transmitted at an infinitely high speed, as such a system related to the parabolic type. Therefore no delay periods are observed between the rate of change of the concentration parameters of effusion values and the variation of thermodynamic parameters in the thermodynamic open macrosystem being investigated. The influence of chemical pseudowaves on the cryogen product volatility as well as on reduction of the safety degree of thermally controlled objects have been analysed.

At the investigation of a non-adequate process, the variations of volatility in identical cryogenic reservoirs during purposeful reduction of the hydrogen concentration in a clearly expressed hydrogen residual atmosphere the following effects have been detected [6,7]:

1. Effect of effusion induced hydrogen instability of the superinsulation (EIHIS) [6,7],

2. Effect of effusion induced heat conduction instability of the superinsulation in cryogenic and vacuum facilities (EIHCIS) [6, 7],

3. Effect of multiplication of the amount of desorbed hydrogen molecules in respect to the magnitude of inflowing humid air molecules in the superinsulation of cryo-vacuum objects (MADHM) [6, 7].

The effects in heat insulation can be controlled. In order to create new heat insulation samples with a high exergy efficiency and a high safety degree, new heat-insulating structures and designs should be developed [8, 9].

As a rule, in the process of operation of big cryovacuum objects, when the heat insulation routine maintenance intervals are exceeded, a process of daily fluctuations of the residual pressure and volatility of the cryogenic liquid arises. The cryogenic reservoirs in question of the RS-1400/1.0 type (hydrogen, nitrogen, oxygen) [10] had an insignificant atmospheric effusion leak being within the tolerance by magnitude. Variations of the residual pressure and volatility of the cryogenic liquid occur in such heat-insulating cavities (HIC) with expressed symbate nature in respect to the variations of the atmospheric leak effusion component.

However for the residual pressure variations, the multiplication mode of desorption processes is characteristic as compared with the calculated effusion flow. The influence on the residual HIC atmosphere by a selective chemical hydrogen absorber based on palladinised manganese dioxide [30,31,34] has allowed to reduce the cryogenic liquid volatility from 1100 kg/day to 390 kg/day [6]. The oscillatory process of the volatility has stopped. The oscillatory process of the residual pressure occurred with the monotonously decreasing correlation coefficient (the variation of the residual medium pressure – the relative humidity) as the hydrogen was pumped out from its maximal value of 0.95 to negative values. After pumping out of the calculated amount of residual hydrogen, the correlation coefficient has changed its sign and has been monotonously increased by modulus up to the value of 0.95. The process of reduction of the correlation coefficient has been symbate in respect to the process of hydrogen removal from HIC. The change of sign of the correlation coefficient occurred at the moment when the whole calculated amount of hydrogen had been removed from HIC [6]. Such behaviour of the correlation coefficient has caused an idea of the electronically stimulated adsorption-desorption process of acceptor or donor gas depending upon the Fermi level of the metallised screen surface. Two hypotheses have been considered:

1. Adsorption-desorption process thermostimulated in micropores.
2. Electronically stimulated adsorption-desorption process of acceptor or donor gas (depending upon the Fermi level) by the inflow of ionised oxygen and water vapour.

5. SURFACE STATES AND PESHEV'S ADSORBON

The basis of the effects being observed is the Shockley surface states on semiconductor surfaces of metallised superinsulation screens and their interaction with acceptor and donor gases in the process of well-known Bardeen-Brettain-Shockley gas-water cycle [32, 33].

Generally the conditions of atom existence on the surface of solids differ from the conditions, in which they are in a volume. The surface states are characterised by a greater probability of electron staying near the surface. They have their own energy levels differing from the volume states. The appearance of Tamm's surface states [35] can be explained by means of the strong bond method (LKAO). Shockley [36] and Maue [37] have found another type of surface states known as Shockley's levels. These states arise when free valencies exist on the surface. The

Shockley's chemisorption states appear only in case of a weak interaction between the adsorbent and the solid. It is this state that is characteristic to the Bardeen-Brettain-Shockley gas-water cycle [32, 33].

The hydrogen adsorption occurred on disperse surfaces of a dimension-quantised semiconductor film [6]. As the main model for the adsorbate placing on the adsorbent, the Peshev's adsorbon can be successfully used. According to this model, the current carrier in the dimension-quantised film with an adsorbate consists of a conductivity electron (with x,y,z coordinates) and adsorbed molecules at the (y,z) surface section with the area of λ^2. In the transverse direction, the electron is connected with the adsorbed molecules by an interaction depending only upon *x*, whereas along the *y* and *z* axes it moves freely changing its adsorption framing. It is assumed that the longitudinal and transverse movements of the carrier are separated, as it takes place in the film without an adsorbate. The difference is that the transverse part contains adsorbed molecules now and therefore determines the carrier mobility by its state. This state is just the Peshev's adsorbon.

6. ELECTRIC CHARGE GENERATION MECHANISMS

The mechanisms amplifying the effects considered have been discussed. They include virtual mechanisms of electric charge generation on the screen surfaces and their accumulation. The "quasicapacitance effect" [30] can be related to these mechanisms, in which the charge generation is carried out according to the G. G. Thomson's mechanism (1896) as well as according to the film thermal electromotive force mechanism [38] arising in thermally strongly differentiated superinsulation layers or according to the semiconductor surface charging mechanism as a result of interaction between the screens and the gas medium [39]. The electret basis of the screens (the PETF-DA-12 film is used in electric capacitors) at low temperatures allows to keep the accumulated charges during a very long time.

The general picture of the given oscillatory process of the parameters being investigated is fully regulated by the ratio between acceptor and donor gases in HIC as well as by the magnitude of relative gas inflow from the environment, by the magnitude of developed dimension-quantised film surface and by the surface's Fermi level magnitude. It goes without saying that it is also assumed that the atmospheric medium with daily humidity and temperature variations is considered as the ambient environment.

7. EIHIS, EIHCIS, MADHM EFFECTS

The effects detected in the course of full-scale experiments [6,7] are as follows: **1. Effect of effusion induced hydrogen instability of the superinsulation (EIHIS), 2. Effect of effusion induced heat conduction instability of the superinsulation in cryogenic and vacuum facilities (EIHCIS), 3. Effect of multiplication of the amount of desorbed hydrogen molecules (MADHM) in respect to the magnitude of the moist air inflowing molecules** will allow to deeply understand the essence of phenomena occurring in foliated heat-insulating systems.

7.1. EFFECT OF EFFUSION INDUCED HYDROGEN INSTABILITY OF THE SUPERINSULATION (EIHIS-EFFECT)

The effect of effusion induced hydrogen instability of the superinsulation is the appearance of pressure oscillations of residual hydrogen medium in the vacuum space of the superinsulation caused by adsorption-desorption processes on metallised surfaces of the superinsulation. The genesis of these processes is caused by the occurance and the destruction of the surface states. The fluctuations of the residual hydrogen concentration can be described in the terms of birth-death processes [40]. The increase of the concentration of ionised oxygen and water in residual hydrogen medium up to the optimal level contributes to the appearance of hydrogen adsorption centres. With the water concentration in the residual atmosphere medium being increased, the destruction of these adsorption centres is observed. The desorption mechanism may be as follows: 1) surface exciton excitation, 2) volumetric exciton migration to the sample surface, 3) recombination processes of charging centres on the dispersoid surface of the SVHI dimension-quantised film.

When an effusive leak is available in the casing, the water concentration variation in the residual medium is symbate to the relative humidity variation in the ambient air [31]. The water amount being excessive in respect to the oscillatory process cycle in question is being frozen out on colder SVHI screens due to thermodiffusion through the porous screen frame.

7.2. EFFECT OF EFFUSION INDUCED HEAT CONDUCTION INSTABILITY OF THE SUPERINSULATION IN CRYOGENIC AND VACUUM FACILITIES (EIHCIS-EFFECT)

The effect is the appearance of heat conduction oscillations of the heat insulation being determined by the hydrogen concentration variations.

The hydrogen concentration in the residual medium changing in the EIHIS effect process dynamics is considered as a "thermal bridge" [6] carrying out the periodic switching of the heat flow from the cryogenic reservoir casing to the wall of the cryogenic reservoir vessel and, consequently, to the cryogenic liquid.

7.3. EFFECT OF MULTIPLICATION OF THE AMOUNT OF DESORBED HYDROGEN MOLECULES IN RESPECT TO THE MAGNITUDE OF THE MOIST AIR INFLOWING MOLECULES (MADHM-EFFECT)

In the course of experiments, it has been obtained that the pressure of desorbed hydrogen molecules is approximately by 21 times as much as the calculated pressure of moist air being supplied through the microleak.

So a multiplicative effect [6,7] is realised on the semiconductor surface of the superinsulation. It should be noted that similar multiplicative phenomena have been observed in the work [41] where the argon desorption stimulation has been investigated by means of the addition of oxygen. The relative enhancement of the desorption in the presence of oxygen has been increased approximately by 20 times as compared with pure argon crystals.

7.4. SELF-ARRANGEMENT OF STRUCTURAL AND DEFECTIVE STATES IN NON-EQUILIBRIUM SYSTEMS

At high hydrogen concentrations in the residual medium, the **(EIHIS, EIHCIS, MADHM) effects** play a **positive** role of a peculiar **safety device** against the excessive heat flow during hot time of the day.

These effects perfectly match the present-day concepts in the field of quantum chemistry [42] and classical thermodynamics [43-47]. In accordance with the classification of classical thermodynamics, a chemically active open complicated thermodynamic system has been considered in this review [45]. The set of dissipative processes being considered vividly demonstrates the non-equilibrium ability to serve as a source of ordering through fluctuations. The **(EIHIS, EIHCIS, MADHM) effects** are the manifestation of self-arrangement in non-equilibrium systems. The discovery of these effects [6,7] shows the non-suitability of the description formalism of similar thermodynamic systems with superinsulation in the field of instability based on the Boltzman principle. During the experiments on reduction of the hydrogen concentration in the vacuum cavity of the filled cryogenic reservoir, the consecutive oscillatory and bifurcation processes of the residual hydrogen pressure variation have been noted.

So some instabilities, secondary bifurcations of time-periodic cycles observed have been noted. These bifurcations in case of a three-molecular models can be obtained analytically.

The investigation conducted has shown that a gradual decreasing of the amplitude of superinsulation heat conduction oscillations occurs at reduction of the residual hydrogen concentration.

8. MAIN TRENDS OF SUPERINSULATION RESEARCH

Only a few works have been devoted to the matters of electro-adsorption interaction of SVHI screens with the residual gas [1-5]. At the same time, the F.F. Vol'kenshtein's monograph [48] is devoted to electronic process on semiconductor surfaces where the results of the 40-year work of the author and his colleagues in the field of physics and chemistry of semiconductor surfaces are summarised. The semiconductor surface in the work cited above is considered from the positions of the interface between two phases. The interaction between gas molecules of the residual medium and free semiconductor electrons and holes occurs at this interface. As the majority of metals have an oxide film, then their surfaces in most cases are semiconductor surfaces. Among works devoted to the physics and chemistry of semiconductor surfaces, the most substantial are monographs by K. Haufe [49] and S. Morrison [50], F. F. Vol'kenshtein [48], A. V. Rzhanov [51]. It is necessary to note a great contribution of the V. S. Kohan's school into the theory of interaction of active gases with metal films [52]. A very meaningful is the monograph of the Indian physicist K. L. Chopra [38] devoted to the investigation of physical processes passing in thin films of metals, semiconductors and dielectrics.

By the present time, the main tendencies of further development of the superinsulation have been carried out in the following directions. First of all, studies devoted to the optimal installation of the superinsulation [53], the process of vacuuming of the cryogenic equipment superinsulation [54-57], investigations of gas permeability of the superinsulation [58, 59], the determination of diffusion coefficients of residual gases in the superinsulation [60], investigations of the kinetics of gas release of heat-insulating materials in vacuum [61], investigations on studying of the distribution law of residual gases in superinsulation layers [62], investigations on studying if the unsteady pressure field in the superinsulation [63], investigations of the properties of superinsulation materials [64, 65], investigations of heat and mass exchange in the superinsulation [66-68]. The work [69] has a high theoretical and practical value, Mikhalchenko with colleagues have detected therein a hundred-fold pressure ratio in the middle of the sample over its thickness and on the outer insulation layer with the temperature of 77K. In the work [70], it has been obtained that the pressure in the superinsulation grows proportionally to the squared insulation thickness. In addition, it has been detected that the pressure distribution over the insulation thickness in the steady mode has a parabolic nature.

In order to improve the conditions of superinsulation vacuuming, it has been proposed to perforate screens [71]. Barron [72] has come to a conclusion that if the perforated hole area equals to 10% of the screen area, then the rate of gas pumping-out from the insulation increases approximately by 1000 times. In the work [73], it has been shown that gas releases of screens can be absorbed inside the insulation, for example, by means of the use of glass-fibre paper with filaments from activated charcoal as a pad's material. In the work [74], it has been determined that gas releases of screens can be absorbed inside the insulation, for example, by means of a low-temperature gas absorber.

Studies are known devoted to surface effects on a dimension-quantised screen-vacuum heat-insulation in the conditions of cryogenic temperatures and vacuum [75], influence of surface centres on the gas medium formation in HIC, selection of a metallised coating of the superinsulation, optimisation of the technology of manufacture of a metallised film with the account of electro-adsorption phenomena on the film surface: selection of the crystal size to be applied onto the polymer film substrate, values of the thickness of the metal spraying onto the film, management of the metallised film properties by the introduction of special alloying gases at the manufacture, etc.

Quite a few works have been devoted to operational methods and means of maintaining and monitoring of the optimal mode of the superinsulation functioning [76, 77].

The investigations devoted to surface effects on dimension-quantised films in the superinsulation, influence of the surface centres of superinsulation screens on the gas medium formation in HIC, selection of the metallised coating of superinsulation screens, optimisation of the technology of manufacture of a metallised film with the account of electro-adsorption phenomena on the film surface, selection of the crystal size to be applied onto the polymer film substrate, values of the thickness of the metal spraying onto the film, management of the metallised film properties by the introduction of special alloying gases at the manufacture, management of the value of the surface potential of superinsulation screens by means of connection of the superinsulation screen to an external potential, etc., are completely absent.

In addition, the matters of selection of the protective casing of the reservoir, introduction of an additional "dry" volume into the cryogenic reservoir design between the casing and the additional moisture-insulating shell are completely missed. The shell can be made with an overflow moisture-impermeable valve. As to the matter of exclusion of a moisture film on the outer heat insulation surface, we are aware of only one work [78].

It is well-known that the film alloying by gas impurities can lead to the improvement of their properties [79]. The task of management of the film properties by their alloying with impurities was stated in a number of works [80].

A special attention will be paid to the management of properties of screens on the polymer basis by means of their modification by the ion implantation. By the present time, a sufficiently large volume of work on the investigation of kinetic phenomena in conducting organic films has been carried out [81-86].

The complexity and diversity of processes being observed in heat-insulating cavities cause a necessity of the conduction of additional emergent experiments with the use of newest measuring apparatus.

The urgency of investigations on the development of a fundamentally new superinsulation is in the present time dictated by the beginning of intensive works on the development of ecologically clean transport on hydrogen in all developed countries.

9. MATHEMATICAL FUNCTIONING MODELS, CALCULATION AND OPTIMISATION OF THE SUPERINSULATION

At the present time, a considerable number of experimental and calculation and theoretical studies have been carried out, which consider heat exchange in the superinsulation both for steady heat modes [11, 16, 87-88] and for unsteady insulation system operation modes [11, 93-95].

9.1. THREE APPROACHES TO HEAT INSULATION CALCULATION

The authors of the monograph [11] have conditionally systemised three fundamentally different approaches to the heat insulation calculation as **homogeneous** and **discrete**.

The homogeneous approach assumes consideration of the superinsulation as the form of a homogeneous medium having reduced thermophysical properties (heat conductivity of the medium, specific heat capacity, etc.). The heat flow in this case is determined by the Fourier equation and the effective heat conduction coefficient being its part is deemed to be dependent upon temperature. A drawback of such approach is the determination of reduced thermophysical parameters of a homogenised insulation system. In the opinion of the authors of the monograph [11], the theoretical determination of these parameters is not seemed to be feasible. These parameters are determined on the basis of experimental data, which imposes considerable limitations on their practical use and impedes the conduction of the analysis of the influence based thereon upon the heat exchange of various factors.

The discrete approach assumes consideration of the superinsulation in the form of a discrete system. In the opinion of the authors of the monograph [11], only this approach allows to use real rather than reduced thermophysical parameters of the system in question at the mathematical description of the heat exchange process.

The homogeneous-discrete approach to the calculation of heat exchange in the superinsulation is used in a number of well-known works.

All these methods of the superinsulation calculation are based on approximate heat transfer process models at the simplified consideration of processes passing in the heat insulation.

9.2. VARIATIONAL DESCRIPTION OF HEAT AND MASS EXCHANGE FOR A HETEROGENEOUS SYSTEM WITH THE ACCOUNT OF ELECTRO-SORPTION PROCESSES IN THE CONTINUUM APPROXIMATION

The variational description of heat and mass exchange processes for a heterogeneous system with the account of electro-sorption processes in the continuum approximation has been carried out with the attraction of the basics of analytical thermodynamics. Analytical thermodynamics is a new trend in the development of classical phenomenological thermodynamics. Having the investigation subject and method being common with the classical thermodynamics, analytical thermodynamics differs from it at least by two peculiarities.

In the first place, it is based on the variational principle, which consequences are basic macrosystem laws – the first and the second laws of thermodynamics, and therefore has a broader scientific basis as compared with the classical thermodynamics. In the second place, the analytical thermodynamics differs from the classical thermodynamics by the analysis method, i.e. the vector analysis. The use of vector analysis reduces a body of mathematics of the classical thermodynamics to the generally accepted one for other macrophysical theories.

The outstanding works by Onsager have laid down the beginning of the deductive theory of irreversible processes and the establishment of variational principles of non-equilibrium thermodynamics [96]. The variational formalism provides a possibility of construction of the whole phenomenological theory of thermodynamics based on the variational principle. The heat and mass exchange problems in the system in question are characterised by an unsteady nature, a considerable non-linearity, the interconnection of heat and mass transfer processes, multi-dimensionality, non-homogeneity and electrochemical activity of heat-insulating system.

To main advantages of the variational description of irreversible processes, one normally relate the following [97-104]:

- a high degree of generality of the physical content of variational principles, as they express fundamental physical laws, and differential equations of irreversible processes in macrosystems and boundary conditions of their implementation can be obtained from variational principles,

- the use of direct methods, by means of which accurate, approximate analytical and numerical solutions in the problems formulated in the variational form can be obtained,

- an opportunity to obtain rough approximation to the accurate solution of the problem, which is especially important in engineering calculations, at that most various information on the process can be used for the selection of a trial solution, including empirical data or accurate solutions of simpler problems of this class;

- the use of extreme values of variational principles functionals for obtaining of integral estimates of the approximate solution accuracy;

- a property of functionals to express important characteristics of irreversible processes such as energy dissipation or entropy production, heat taking part in the process.

The variational principle developed by my teacher V.V. Chikovani [44,45] has been called basic variational principle of the classical phenomenological thermodynamics. Such name of the variational principle is connected with the fact that the basic laws (principles) of classical thermodynamics follow from it – the first and the second laws of thermodynamics.

The main differences of the V.V. Chikovani's variational principles from the well-known principles of thermodynamics of irreversible processes [100, 105, 106, 108, 109] as well as from the conditions of steadiness of functionals having a formal mathematical nature [101, 110-120] is that they, though being an expression of the basic variational principles of the classical phenomenological thermodynamics, have a simple and clear physical meaning expressed in terms of fundamental macrosystem properties. These principles have an integral form both by spatial variables and by time, which provides the use of not only the Kantorovich's method but also the most effective direct method of variational calculus, i.e. the Ritz method.

As it is well-known, at the description of heat and mass exchange processes in continua, an assumption on the local equilibrium is used, in accordance with which any differential volume of the continuum is an internal equilibrium thermodynamic system [45]. Each differential continuum volume can be considered as a multi-phase system characterised not only by thermodynamic parameters of state being equilibrium over the whole differential volume but also by parameters determining irreversible processes of interphase interaction inside the differential volume being non-equilibrium within the limits of the differential volume (but being equilibrium within the limits of each phase being a part of the differential volume.

A peculiarity of the proposed superinsulation model as a continuum is the concept of any differential volume in the form of a two-phase system (V.V. Chikovani, N.V. Dolgorukov,1991). The mass exchange occurs between the gaseous and solid phases due to sorption processes. The temperature within the limits of the differential volume is considered identical for both phases. We will describe the sorption system "adsorbent – adsorbate (solid phase)" by parameters characterising it on the whole, i.e. without account of the real structure of the adsorption phase. Such approach allows to use the gas release characteristics of the superinsulation materials being determined experimentally and obtain a mathematical model being suitable for the description of heat and mass exchange processes, both at the availability of gas adsorption in the surface layer and in the material micropores (V.V. Chikovani, N.V. Dolgorukov, 1991). In order to obtain the variational formulation of the mathematical model of molecular heat and mass exchange processes in the superinsulation, one can use the basic principle of the classical phenomenological thermodynamics [44,45]:

$$\delta Q = \delta \int_{1-2} \vec{P} d\vec{r} = \delta \int_{1-2} \sum_{n=1}^{N} P_n(x_1, x_2, x_3, ..., x_N) dx_n = 0, \quad (1)$$

where $\vec{P}$ is a vector in the N-dimensional space of macrophysical parameters of state $x_n (n=1,..., N)$ of a thermodynamic system characterising its interaction with the environment; $d\vec{r}$ is the radius-vector differential in the same space.The variational description of the heat and mass exchange processes for a heterogeneous system with the account of electro-sorption processes will be given in detail in the second part of the review.

10. RELIABILITY AND MAINTENANCE OF THE SUPERINSULATION

The advantages of heat-insulating systems are decidedly determined by the reliability and an opportunity to maintain them in good operating order. The growing degree of use of the superinsulation having an extreme functional vulnerability as compared with other heat insulation types gives these criteria a high value. So the reliability and maintenance for the superinsulation are of foremost importance.

Under reliability of the superinsulation, one understands a heat-insulating system property to perform pre-set functions at a certain time interval and at that to maintain the values of established performance characteristics within preset bounds at the corresponding conditions of operation, repair, storage and transportation [97].

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