





Research Article

Study into the physical chemistry and technology of alkali liquid metal coolants for nuclear and thermonuclear power plants^{*}

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Abstract

It is shown that, as the result of developing alkali liquid metal coolants, including sodium, eutectic sodium-potassium alloy, lithium and cesium, the scientific basis has been established for their application in nuclear power. The paper presents data from investigations of thermophysical, neutronic and physicochemical properties and characteristics of various alkali liquid metal coolants, the content of solid-phase and dissolved impurities in coolants, mass transport of impurities in circulation circuits with alkali liquid metal coolants, development of systems for removal of impurities, and control of the content of impurities in alkali liquid metal coolants. Alkali liquid metal coolants are considered as a part of a system that includes a structural material in contact with the coolant, and a gas space that compensates for the thermal expansion of the coolant. The state of the system is defined by the physicochemical properties of the system's components. And the coolant and the structural materials also represent subsystems consisting of a base material, a coolant and impurities contained both in the material and in the coolant. It has been shown that each alkali liquid metal coolant has its own set of impurities that define its technology. It depends on the physicochemical properties of the solution of the structural material impurities and components in the coolant. Objectives have been formulated for investigating further alkali liquid metal coolants, as stemming from the need to improve the efficiency, environmental friendliness, reliability and safety, and for extending the life of nuclear power plants in operation or under design. Alkali liquid metals are promising candidate materials for being used in thermonuclear power not only as the coolant but also as the tritium breeding medium. These include, first of all, lithium and its eutectic alloy with lead (17 at. % of lithium). The possibility for using lithium or a lithium-lead alloy as a coolant in the blanket of the international thermonuclear power reactor is compared.

Keywords

nuclear power plants, thermonuclear reactors, alkali liquid metals, physicochemical performance, mass transport, control of impurity content

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Introduction

It was the early stage in using liquid metals as coolants for nuclear power plants that saw the need for a new science as the physicochemical basis for application of liquid metal coolants and studies required to select and provide rationale for the techniques to clean up and control the content of impurities in coolants, develop all technologies that take into account the peculiarities of operating NPPs and liquid metal coolant circuit components, adopt methods for their safe and efficient operation, etc.

Works on physical chemistry and technology of liquid metal coolants became a top priority for the IPPE's thermophysical team led by V.I. Subbotin and, further on, for staff at other organizations and design bureaus in nuclear power industry. A contribution to their evolution was international cooperation with France, Great Britain, the USA and other countries supported by A.I. Leypunsky and V.I. Subbotin.

The theoretical basis for adopting the technology of alkali liquid metal coolants in Russia was laid by the classification of studies in a monography by Subbotin et al. 1970 and developed later by Kozlov et al. 1983, Subbotin et al. 1999, Arnoldov and Loginov 2005. The result has been the establishment of a scientific school, "Heat and Mass Transport, Physical Chemistry and Technology of Coolants in Energy Systems". It would not be an exaggeration to state that this school is a unique phenomenon not only in this country but also for the world community (Efanov et al. 2015).

The results of the above studies made it possible to develop and practically implement the instruments and systems to support successful operation of conceptually new NPPs (Rachkov et al. 2014). Alkali liquid metals have been used or considered as promising coolants in nuclear and thermonuclear power, including sodium, an eutectic sodium-potassium alloy, lithium, an eutectic lithium-lead alloy, potassium, and cesium. It should be added that under consideration are also alloys of some of the above metals with minor additions of other metals, e.g., sodium-lead, lead-potassium, and other alloys. Such alloys are expected to perform better.

To implement the strategy of two-component nuclear power with a closed fuel cycle using fast neutron sodium cooled reactors (Ponomarev-Stepnoi 2016), ensure a competitive edge and keep the lead enjoyed by Russia in NPPs with such reactors, it is required to continue studies in using new design and engineering solutions and development scenarios of emergency processes for fast reactor designs of a higher power, BN-GT and BN-VT.

Lithium and a sodium-potassium alloy with a eutectic composition have been selected for space nuclear power plants thanks to their nuclear physical and thermophysical properties. It should be noted that the eutectic sodium-potassium alloy is also used extensively at the IPPE as the model coolant for studies on heat exchange in the reactor core and heat-transfer components with lead and lead-bismuth coolants.

There are four types of liquid metal coolants discussed in literature that can be of interest for thermonuclear plants with a different degree of the rationale provided (Arnoldov et al. 2005). These are pure lithium and its eutectic alloy with lead (17 at. % of lithium, the eutectic point temperature is 235 °C), an eutectic lead-bismuth alloy (44 at. % of lead, the eutectic point temperature is 125 °C), and a eutectic lithium-tin alloy (5 at. % of lithium, the eutectic point temperature is 232 °C). The two latter alloys have been mentioned in literature a couple of times. At the same time, the two former coolants are considered as would-be coolants for thermonuclear plants for a number of reasons, which include radiation stability, excellent heat-transfer properties, etc. They also form a material for breeding tritium, a component involved in the reaction of synthesis in the thermonuclear plant, in the thermonuclear plant's radiation fields. Of undoubted interest is a ternary alloy of lead, bismuth and lithium.

A major contribution to the activities concerned with the use of alkali metals in nuclear and, specifically, thermonuclear power was made by Prof. M.N. Arnoldov who authored over 100 scientific publications summarized in monographs Subbotin et al. 1970, Subbotin et al. 1999, Askhadullin et al. 2021.

An alloy of lithium and lead was defined as the coolant and the breeder material in the materials dealing with the International Thermonuclear Experimental Reactor (ITER). It appears to be necessary to compare more thoroughly the advantages and drawbacks of each of these coolants.

An integrated test facility built at the IPPE allowed achieving the targets for the NPP designs with fast neutron sodium cooled reactors (BR-10, BOR-60, BN-350, BN-600, BN-800) and sodium-potassium cooled reactors for space vehicles (BUK, TOPOL, TOPAZ), and has been prepared for the experimental justification of innovative engineering solutions for the NPP designs of a new generation (Pershukov et al. 2016).

Thermophysical properties of alkali liquid metal coolants

The thermophysical properties of liquid metal coolants are summarized in a number of monographs, e.g., Shpi'lrain et al. 1970, Kirillov et al. 2007. Alkali liquid metals have a high boiling point and do not require high pressure to prevent their boiling (Table 1). Being highly heat-conductive thanks to their electron conductivity, alkali liquid metals have high heat transfer coefficients which ensures acceptable temperature conditions for the operation of the heat exchange surfaces at a high heat flux density.

Due to the combination of its properties, sodium has been selected as coolant for commercial and research nuclear power plants. Thanks to the low melting temperature (-12.5 °C), with comparatively satisfactory nuclear and thermophysical properties, the sodium-potassium alloy has been selected as coolant for space systems.

As compared with the lithium-lead alloy, lithium is highly advantageous practically in all thermophysical properties, including low density, high boiling point, low in-core activation, etc. (Shpi'lrain et al. 1970, Subbotin et al. 1999. This manifests itself as well in a smaller pumping pressure drop (therefore, smaller mechanical loads on the first wall), better heat-conductive properties, a higher thermal efficiency, etc. The drawback of the alloy, as compared with lithium, is its higher melting point (235 °C for the alloy, and 180.5 °C for lithium).

Physical chemistry of alkali liquid metal coolant melts

As a result of the studies undertaken at IPPE, rationale has been provided for the integrated approach to studying alkali liquid metal coolants as a complex multi-component heterogeneous system the state of which is defined by the interaction among the coolant, the impurities, the structural (process) materials, and the cover gas, and the key impurities have been identified that affect the serviceability of NPPs with alkali liquid metal coolants (Table 2).

The state of the impurities in coolants was studied using thermodynamic calculations for an equilibrium system, as well as mass exchange processes in the NPP circuits. Dedicated procedures (oxygen activity measurement, diffusion membranes, samplers) were used extensively in the experiments. Data have been obtained on the existence forms of impurities in coolants, their equilibrium concentrations and solubilities (oxygen, hydrogen,

Table 1. Thermophysical properties of liquid metals at 500 °C

carbon, nitrogen in lithium, etc.), kinetics of the reactions in such systems. For sodium and the sodium-potassium alloy, these are oxygen, hydrogen, carbon and their compounds, including products of the sodium reactions with air, water and hydrocarbons (motor oil), corrosion products of structural materials during long-term reactor operation (Fe, Cr, Ni, Mn, Mg), radionuclides, including tritium and nitrogen in cover gas.

Processes of physical chemistry and the technology for sodium and sodium-potassium coolants vary. The explanation is that, first, this is a two-components coolant and, second, complex forms are highly likely to result from the joint presence of impurities, such as oxygen and hydrogen (hydroxide forms), and oxygen and carbon (carbonate forms), in the alloy, as well as with formation of complex double oxides of the alloy components and the structural material components.

For lithium, along with the above impurities, one of the hazardous impurities is nitrogen expected to be present in coolant in large quantities and affect adversely its performance. Chemical and thermodynamic calculations have shown that nonmetallic impurities are present in lithium melt in the form of oxide, nitride, carbide and hydride, calcium and beryllium oxides, and so on.

The information acquired on the solubility of different impurities in liquid metals, their mutual effects on solubility, the kinetics of the reactions taking place in coolants, diffusive constants, the structure of melts, and the existence forms of impurities they contain, through being extensive, is not sufficient to describe the system in full. Based on the data obtained and as a result of studying the corrosion of structural materials in coolants, rationale

Melt	T _{melt} , ℃	$T_{\rm boil}, ^{\circ}{\rm C}$	R _{melt} , kJ/ kg	R _{boil} , kJ/kg	ρ, kg/m³	<i>Cp</i> , J/ (kg K)	λ, W/(m K)	v 10 ⁻⁸ , m ² /s	<i>a</i> 10 ⁻⁶ , m ² /s	Pr 10 ⁻³
Lithium (Li)	180.5	1347	66.3	19400	486	4178	49.98	59.2	25.6	2.3
Sodium (Na)	97.8	883	113	3870	883	1262	66.34	25.9	62.86	4.624
Potassium (K)	63.6	774	61.4	1983	739	763	41.9	24		3.2
Na-K (22%Na+78%K)	-12.5	784	96	2540	751.9	871.9	26.2	23.4	40.1	5.86
Na-KCs	-78	_	-	_	1235	384	13.7	15.4	-	5.3
Cesium (Cs)	28.5	678	15.73	495.9	1597	220	18.5	12.8	-	2.1
Lead (Pb)	327.4	1745	25	860	10470	147.3	15.45	17.62	9.82	17.8
Bismuth (Bi)	271.4	1533	50.15	857	9854	150	14.2	13		13.5
Pb-Bi (44,5% Pb+55,5% Bi)	125.0	1638	39	860	10050	145	14.2	13.6	9.7	14.7
* Pb-Li (83% Pb+17% Li)	235.0	-	33.9	-	9500	190	13.2	18.8	-	21.8

* Pb-Li alloy data are for 300 °C (Ponomarev-Stepnoi 2016) R_{melt} – melting heat, R_{boil} – boiling heat, ρ – density, Cp – heat capacity, λ – thermal conductivity, v - kinematic viscosity, a - temperature conductivity

Melt	$T_{\rm melt}$, °C	$T_{\rm boil},^{\rm o}{\rm C}$	$R_{\rm melt}$, kJ/kg	$R_{\rm boil}$, kJ/kg	ρ , kg/m ³	<i>Cp</i> , J/(kg K)	λ, W/(m K)	v 10 ⁻⁸ , m ² /s	<i>a</i> 10 ⁻⁶ , m ² /s	Pr 10 ⁻³
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has been provided for the allowable content of impurities in coolants and cover gas. This is defined by corrosion for oxygen, carbon, hydrogen and nitrogen, and by the radiation situation within and beyond the reactor plant for fission gases, cesium and strontium (Ohse 1985; Bystrov et al. 1988).

As these studies were becoming growingly profound, it was shown that the most important role in the processes taking place in coolants is played not only by the basic forms of impurity compounds with coolants (oxides, nitrides, carbides, hydrides) but also by complex compounds such as double oxides and doubles nitrides, hydroxides and carbonates.

The physicochemical properties of the lithium-lead alloy are underexplored as compared with those of lithium. More than that, some of the data on these properties for the alloy are simply insufficient or inaccurate.

Mass transport in circuits with liquid metal coolants

The results of the computational theoretical and experimental studies were used to propose and justify the models and codes to allow one to describe the behavior of hydrogen, carbon, hydroxides and carbonate, heat and mass transport, and physicochemical processes in the NPP anisothermic circuits: MASKA-LM, TURBO-FLOW, and TRICOD (Alekseev et al. 2020).

Models have been proposed for homogeneous (hydrogen, tritium) and heterogeneous (corrosion products of structural materials) mass transport in sodium circuits. Computer codes have been developed on their basis for calculating the mass transport of tritium in a three-circuit sodium cooled NPP, including the escape of tritium into the environment. The calculations have shown that most of the tritium accumulates in cold traps.

In normal conditions of operation, coolant not only transports impurities through the circuit but also plays an important role in processes of interaction with structural materials and suspended matter (Fig. 1).

The findings confirm that the corrosion rate for structural materials in sodium is low with normal concentrations of oxygen and hydrogen. However, these conditions fail to be fulfilled when water escapes into sodium in the SG in or near the outflow channel outlet. It has been shown that the corrosion rate exceeds by several orders of magnitude the corrosion rate during normal conditions of operation.

Recently, more experimental data have been obtained on the fundamental physicochemical regularities of mass exchange in a high-temperature anisothermic 'structural materials – sodium – impurities' system in a temperature range of 600 to 800 °C (Kozlov et al. 2014).

Lithium is more corrosively active than sodium but less corrosive than the lithium-lead alloy (by an order of magnitude, as occasionally reported). The corrosive activity of lithium has been studied in more details than that of the alloy (Subbotin et al. 1999).

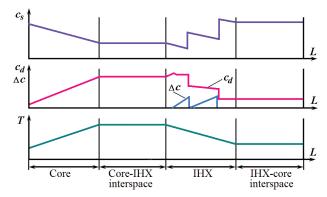


Figure 1. Quality flowsheet of impurity mass transport through the NPP circuit: c_s – suspended phase concentration in sodium; c_d – concentration of dissolved impurity; Δc – density of deposits on the flow path surface; T– coolant temperature; L – circulation circuit longitudinal coordinate; IHX – intermediate heat exchanger.

The available data show that the steels in alloy start to corrode intensively after a latent period of about one thousand hours. The oxide layers on the steel, which inhibit the lead corrosion, appear to be recovered by the alloy's lithium component for that time.

The suspended (insoluble) impurities have been found to redistribute across the flow and concentrate extensively on the walls in small-rate (stagnant) areas.

Removal of impurities and control of impurity content in alkali liquid metal coolants

One of the prerequisites for long-term failsafe operation of an NPP is fine cleanup of coolants to remove dissolved and undissolved impurities. Methods have been developed for physical, chemical and combined cleanup of circulating coolants. Cold traps (CT) (Kozlov et al. 1983) and chemical or hot (getter) traps (HT) (Kozlov et al. 2016) have been recommended for practical use, and carbon traps have been recommended cesium removal.

Cold traps. CTs have been selected as the key tool to clean up sodium, the sodium-potassium alloy and lithium. For present-day NPPs, the concentrations of oxygen and hydrogen in sodium and in the sodium-potassium alloy, following the removal of impurities (oxide, hydride, sodium hydroxide and, partially, other impurities) using a CT, ensure that the thermal-hydraulic parameters are as calculated and the corrosion rate of structural materials (steel, high-nickel steels) is low.

Thanks to exploring the thermal-hydraulic and mass exchange processes in CTs, scientific rationale has been formed to develop CTs for the BOR-60, BN-350 and BN-600 reactors. Thus, with sodium being present in a CT for more than 15 minutes, the CT impurity retention factor turns out to be close to unity. The minimum post-cleanup concentration of oxygen and hydrogen in sodium is equal to their solubility at the CT outlet temperature (120 to 150 °C). Such temperature leads to the oxygen and hydrogen solubility of 3 to 5 and 0.02 to 0.05 mln-1 respectively. The impurity capacity (one of the most important service characteristics) for Russian-designed CTs is 3 to 5 times as high as that of the best foreign counterparts. CTs are efficient in cleaning up sodium from the products of its interaction with water.

Flow-through and diffusive impurity CTs have been used successfully for continued and periodic cleanup of lithium coolants. For lithium and the sodium-potassium alloy, the minimum temperatures in CTs equal 200 °C and 80 °C respectively. In such modes, lithium was cleaned up to the concentrations of $5 \cdot 10-4$ mass % of oxygen, $5 \cdot 10-4$ mass % of hydrogen, and $1 \cdot 10-3$ mass % of nitrogen, and to the oxygen and hydrogen concentrations in the so-dium-potassium alloy equal to $1 \cdot 10-3$ mass % and 10-5 mass % respectively (Subbotin et al. 1999).

Literature lacks data on the allowable content of carbon in lithium. It follows, though, from the experience of operation that the carbon concentration in lithium cleaned up by a cold trap is not more than 5 mln⁻¹.

Emphasis was required to be placed on the development of a cold trap for a large reactor with its single-tank design (Alekseev et al. 2013). A conceptually new combined impurity removal system has been proposed and justified for a high-temperature hydrogen producing NPP with the sodium coolant temperature of ~ 900 °C, which allows all possible modes of operation (Kozlov et al. 2016).

Hot traps. Finer cleanup of coolants required in high-temperature plants to remove niobium, tungsten, vanadium, tantalum and their alloys is achieved with the use of HTs. Thermal-hydraulic calculations have justified the selection of getters for removal of alkali liquid metal coolant impurities (sodium, sodium-potassium, lithium), including oxygen (calcium, beryllium), nitrogen (zirco-nium, titanium, beryllium), carbon (titanium, zirconium, niobium), and hydrogen or tritium (lanthanum, calcium, cerium). Temperature and thermal-hydraulic modes have been experimentally justified for the getter cleanup of coolants.

In fast neutron reactors, HTs can be operated both in standby mode, using sodium heat-up at the HT inlet, and in 'high-rate cleanup' mode that allows coolant cleanup with simultaneous changeover from standby mode to nominal mode (Kozlov et al. 2016). Taking into account the need for the sodium cleanup to remove suspended matter (corrosion products of structural materials) using dedicated filters, it will be reasonable to consider combining these filters with HTs.

Removal of radionuclides. Studies into the distribution of radionuclides within the standard CTs in the BR-5 and BOR-60 reactors have shown these to be capable to accumulate radionuclides. It has been demonstrated that the ratio of the equilibrium bulk activities prior to and after the cleanup is equal to 100 for ¹³¹I, 7.2 for ⁶⁵Zn, 1.5 for ^{137, 134}Cs, and 1.3 for ⁶⁰Co and ¹²⁴Sb, and to 1 for the rest of the nuclides (no cleanup). Radioactive isotopes of iodine are retained and decay in the trap. The entrapment of cesium isotopes in the CT is explained largely by the cesium deposition on unpoisoned carbon impurities accumulated in the trap.

A sorption cleanup technique has been proposed and justified for the efficient cleanup of sodium from cesium using graphite materials (GMZ, ZBM and GMZ-3 graphite grades). Compact disposable adsorbers are installed in the core instead of FAs or the side shield assembly (Chechyotkin et al. 1983).

Control of impurity content. When justifying requirements for the techniques to control impurities in coolants and in cover gas in terms of efficiency, concentration range and sensitivity, one shall take into account not only the need for providing the conditions for long-term fail-safe operation of commercial plants and their prototypes, maintain the radiation situation and minimize exposure doses, but also to support experimental activities.

A variety of samplers have been designed and techniques were developed to analyze coolant samples for the content of hydrogen, oxygen, different carbon forms, and metallic impurities. The best results in terms of analysis accuracy and sensitivity were obtained with the distiller sampler. It was used to control impurities in the sodium-potassium alloy. The detection limits are 2 mln⁻¹ for oxygen (oxide, hydroxide and carbonate forms), 4 mln⁻¹ for carbon (nonvolatile forms), 1.6 mln⁻¹ for nitrogen (nitride forms), and 2 mln⁻¹ for chlorides.

Online impurity control in sodium placed emphasis on a plug indicator, sensors with diffusion membranes (nickel for hydrogen, Armco for carbon), and electrochemical methods.

Rationale has been provided for the selection of the materials, temperature and hydrodynamic modes for control of thermodynamic activity for oxygen, hydrogen and carbon in sodium by equilibrium standard sample (ESS) method. It has been shown to be possible to use this method for other coolants.

Characteristics have been studied and tests were conducted on experimental sodium benches for the IPPE-developed oxygen electrochemical cells (OECC) (Blokhin et al. 2017). Instruments of this type were used at test facilities with the sodium-potassium alloy and lithium. The current experience proves that prospects are good for the electrochemical method to be used to control the content of impurities in other media.

Much attention was given to exploring the processes caused by the alkali metal escape from circuits. The obtained results have shown that there are no internal causes for major defects. This is also confirmed by the many-year experience of operating such facilities. However, alkali metal cooled NPP designs take into account the potential for leakage. The experience in operating Russian fast reactors has shown the solutions adopted and introduced to provide reliable protection against sodium burning.

Methods and tools have been developed to clean up and control the purity of lithium. Requirements have been formulated with respect to the content of impurities in lithium. Various equipment has been tested for lithium circulation circuits (Subbotin et al. 1970; Subbotin et al. 1999; Arnoldov et al. 2005).

Further evolution of works on physical chemistry and technology of alkali liquid metal coolants

Emphasis shall be placed in further evolution of alkali liquid metal coolants on addressing issues that can be broken down into three groups:

- processes taking place during normal conditions of operation which define the dynamics of the impurity behavior in the NPP circuits;
- events leading potentially to abnormal situations to progress into emergencies;
- acquisition of fundamental data on the physiochemical processes for triple and more complex systems in sodium required to justify the models built in codes.

The key areas for investigating the performance of alkali liquid metal coolants are as follows

- exploring the physicochemical interaction of the components of structural materials and impurities in liquid metals, as well as the solubility, and the adhesive, diffusive and dispersive performance of complex liquid metal heterogeneous systems (coolant – impurities – structural materials – cover gas) and the behavior of such systems;
- investigating the effects of the coagulation processes in circulation circuits on the size distribution change;
- exploring the mechanism and kinetics for the formation and decomposition of complex oxides and carbon compounds in conditions of an anisothermic circuit;
- investigating the mass transport and tritium accumulation processes in fast neutron reactor circuits;
- exploring the separation of suspended matter in different portions of a sodium circuit, its entrainment from variously configured portions in the gas cavities of sodium circuits, and interaction of potential tertiary (secondary) coolants with sodium.

Conclusions

The current experience in adoption of alkali liquid metal coolants allows one to suggest that these have a niche of their own in nuclear power industry on a level with water coolant. Despite this, however, it cannot be assumed that all targets have been met, and it only remains to replicate the existing experience for newly built reactor facilities.

The metals and alloys listed have not been equally adopted. Those adopted and used to the greatest extent are sodium (commercial and experimental NPPs) and the eutectic sodium-potassium alloy (space NPPs). Lithium coolant is adopted as applied to thermonuclear plants. Some of the alkali metals and their alloys have not been adopted whatsoever.

A concept has been developed as a result of studies which treats coolant as a constituent in a multi-component, heterogeneous thermodynamic system: structural (process) materials – cover gas – impurities. The complexity of the system depends on the sources of the impurities, and its heterogeneity is defined not only by the existence of three phases (cover gas, coolant, structural material) but also by the presence of suspended matter in coolant and of aerosols in cover gas.

The key point in the technology of any liquid metal coolants is the content and state of the impurities the coolant contains. The existing experience shows that a technology of liquid metal systems is expected to minimize the corrosion of structural materials and the change in their performance, and to exclude the occurrence of excessive phases in the coolant flow.

It follows from comparing the findings for lithium and the lithium-lead alloy that lithium has been studied in much more detail than the alloy. Lithium is preferred to alloy in most of the aspects. Methods and tools have been developed to clean up and control the purity of lithium, and requirements have been formulated with respect to the content of impurities in lithium. Lithium is less aggressive than the alloy. The technology of alloy as coolant has been scarcely studied. At the same time, the key negative feature of lithium is that it is more chemically active and flammable than the alloy.

An important point is magnetohydrodynamic effects in the reactor with magnetic confinement of plasma. This requires further studies, including engineering solutions and search for insulating coatings.

Further work to justify incombustible coolant appears to be promising for future NPPs. It has been shown that the alloy Na+Pb 10 mol. % will not burn in a 'puddle' at 700 °C. It is necessary to prove the passive interaction of this alloy with a jet efflux into the atmosphere, confirm experimentally the expectedly low corrosive activity, due to physicochemical considerations, and study, in the event of positive results, the technology, the thermophysical properties, and the possibility for reducing the low-level operating temperature.

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