SELECTION AND REALIZATION OF METHODS OF REDUCING HEAT FLOW INTO CRYOGENIC VESSELS WITH LIQUID NITROGEN

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The effect of different design and service factors on heat flow into cryogenic vessels is examined. Results are presented from tests of cryogenic vessels equipped with new containers with vacuum-shield thermal insulation.

Cryogenic vessels have come into wide use in different areas of the economy for the storage of biological products in liquid nitrogen. These vessels take the form of an evacuated cylindrical or spherical body equipped with an internal container having a vacuum-shield insulation (VSIC). The container is suspended on the thin-walled throat of the vessel. With an increase in the scale of production of the vessels, it becomes necessary to make them more efficient. Methods of reducing total heat flow to the liquid nitrogen and, accordingly, increasing the service life of the vessel before it requires the addition of cryogenic agent can be found by analyzing the effect of the geometric and thermal characteristics of the vessel's structural elements and the coldness of the outgoing vapors on the magnitude of the resulting heat inflow.

To solve this problem, we developed a method for the thermal design of 35-liter vessels of the "Khar'kov-34B" type [1] and we measured the total heat inflow for cryogenic vessels of different configurations. The design method was based on the solution of conjugate nonlinear two-dimensional problems of heat conduction in the assembly consisting of the VSIC and the plug of the vessel, as well as the solution of the unidimensional problem in the throat with allowance for its cooling by vapors of the cryogenic agent and radiative—convective heat transfer through the gap or the layer of glass-paper between the VSIC and throat.

The heat flux through the thermal insulation of a Kh-34B cryogenic vessel is the sum of the heat flow through the VSIC assembly, the wall of the throat, the plug, the column of gaseous cryogen, and the gap between these elements:

$$Q_{\mathrm{in}} = F_{\Sigma}\left(\lambda_{\mathrm{in}}(T_{\mathrm{in}})\frac{\partial T_{\mathrm{in}}}{\partial r}\right)_{r=r_{\mathrm{ves}}} + F_{\mathrm{t}}\left(\lambda_{\mathrm{t}}(T_{\mathrm{t}})\frac{dT_{\mathrm{t}}}{dx}\right)_{x=0} + F_{\mathrm{pg}}\left(\lambda_{\mathrm{pg}}(T_{\mathrm{pg}})\frac{\partial T_{\mathrm{pg}}}{\partial x}\right)_{x=0} + F_{\mathrm{gs}}\left(\lambda_{\mathrm{gs}}(T_{\mathrm{gs}})\frac{dT_{\mathrm{gs}}}{dx}\right)_{x=0}.$$

It was shown in [1] that the character of the change in thermal conductivity with temperature is not important for calculating the total flux in vessels of the Kh-34B type and their components. What is important here is the mean integral value of temperature and the thickness of the insulation assembly (packet). This means that we can assign the change in the thermal conductivity of the VSIC in the form of the relation $\lambda_{\mathrm{in}}(T) = \alpha f(T)$, where α is an arbitrary constant; $f(T)$ is the calorimetric dependence of the thermal conductivity of the VSIC on temperature. Empirical data on mean integral values of thermal conductivity in the temperature range 77-300 K and their temperature dependences is available for the materials of the throat and plug and for nitrogen vapor. Thus, in designing a vessel with different compositions for the insulation packet, the only unknown is the thermal conduc-

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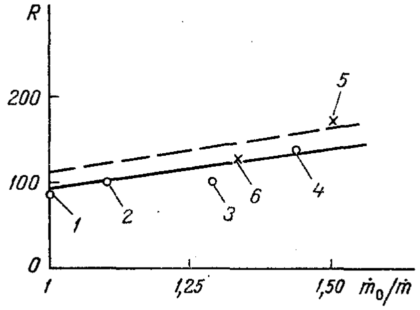

Fig. 1. Effect of the efficiency of the use of cold of the vapors $\dot{m}_0/\dot{m}$ on the service life R (days) of cryogenic vessel Kh-34B; the extreme points are for modifications of the vessel: 1) with vacuum plug having a central channel and a gap between the VSIC and the throat; 2) same, without gap; 3) vacuum plug, gap between VSIC and throat; 4) vacuum plug; 5) polystyrene plug; 6) same, with gap between VSIC and throat. The theoretical curves: solid) for vessel with vacuum plug; dashed) for vessel with polystyrene plug.

tivity of the VSIC. The mean integral value of this quantity is determined by the value of the constant α. In the course of calculations by the method in [1], the constant α is chosen so that the theoretical and experimental heat flows through the thermal insulation coincide for a given modification of the vessel Kh-34B. This approach makes it possible to use the results of tests and calculations for other vessel models to evaluate the reliability of the value found for thermal conductivity.

We used the above-described procedures to perform thermal calculations for six experimental Kh-34B vessels. The vessels differed in the material and configuration of the plug, the width of the gap between the VSIC and the insulation of the throat, and, accordingly, the degree of utilization of the cold of the nitrogen vapors. We used the same composite as the insulation in all of the vessels — a 6-μm-thick film of PET DA with intervening layers of glass cloth EVTI-7. The rate of evaporation of liquid nitrogen from the vessels was determined from the results of their periodic weighing. We used the mass rate to calculate the total heat flow to the liquid nitrogen and the service life of the vessel.

Comparison of experimental and theoretical values of total heat flow obtained using the same function $\lambda_{in} = \lambda_{in}(T)$ showed good agreement within 5-12% for all six vessel models. We therefore used the above method to analyze the effect of the cold of the vapors and the geometric and thermal parameters of vessel elements on the resulting heat flow into Kh-34B cryogenic vessels.

It was established on the basis of the theoretical study that one of the best ways to improve the thermal characteristics of the vessel is to make fuller use of the outgoing nitrogen vapors. Figure 1 shows experimental and theoretical values of the service life of Kh-34B vessels as a function of the efficiency of vapor use. For these vessels, 100% use of the cold of the vapors makes it possible to increase service life by a factor of 1.67 relative to a vessel in which the cold is not utilized (experimental point 1 in Fig. 1). The studies showed that the "shield-less" method is optimal for Kh-34B vessels. Here, service life is increased by a factor of 1.5, which corresponds to 90% use of the cold of the vapors. To realize this method, it is necessary to distribute the layers of the VSIC over the entire length of the throat, eliminate the gap between the VSIC packet and the throat, and provide good thermal contact between the layers of the VSIC and the throat. The last step is accomplished by tightly winding layers of glass cloth 1-5 μm thick around the throat and pressing the layers of the VSIC against them. As was shown by measurements and calculations of the temperature fields, radiative-conductive heat transfer through the wound layers of glass establishes nearly the same heat flow as would exist in the case of ideal contact between the VSIC layers and the throat.

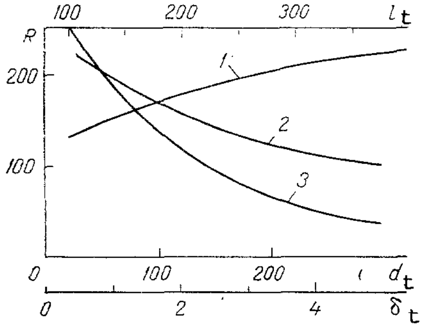

Fig. 2

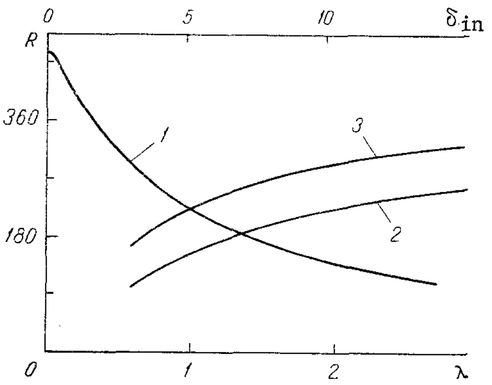

Fig. 3

Fig. 2. Effect of the parameters of the throat on the service life of a Kh-34B vessel: 1) dependence of service life R (days) on throat length ℓ_t (mm); 2) on the thickness of the throat wall δ_t (mm); 3) on the diameter of the throat d_t (mm).

Fig. 3. Dependence of the service life R (days) of a Kh-34B cryogenic vessel on the thermal conductivity of the VSIC λ (μW/(cm·K)) (1) and the thickness of the packet δ_{in} (cm) at $\lambda = 1.34$ (2) and 0.76 μW/(cm·K) (3).

It is evident from Fig. 1 that the possibilities for improving the thermal parameters of a Kh-34B cryogenic vessel with liquid nitrogen by using the cold of the vapors are nearly limitless. To investigate the further prospects for reducing heat inflow, we performed calculations to determine the effect on volatility (service life) of the length, diameter, and wall thickness of the throat (Fig. 2), the volume of the vessel, and the thermophysical characteristics of the VSIC (Fig. 3). In standard Kh-34B vessels, the thickness of the throat is 1.2 mm, the working length is 210 mm, and the diameter is 60 mm. It is evident from Fig. 2 that a reduction in the thickness of the throat to 1 mm can increase service life by 15 days, a reduction in throat diameter to 40 mm can increase it by 30 days, and an increase in length to 300 mm can increase it by 25 days. Increasing the volume of the vessel is also effective. For example, doubling of the volume increases service life by 85 days.

However, operational considerations and other requirements make it difficult to reduce the thickness of the throat wall and the diameter of the throat, increase its length, or increase the volume of the vessel. Thus, the main route to reducing heat inflow in cryogenic vessels of the Kh-34B type is to improve the VSIC.

The direction currently being taken to improve the VSIC is to reduce the gaseous and contact components of heat transfer [3]. Also important is the effort being made to improve sanitary-hygienic conditions in VSIC assembly by developing materials which do not generate silicon particles. An analysis of preliminary experiments showed the expediency of using new types of lining materials of synthetic and mineral fibers made by the "wet" method traditionally used in the paper industry. Advantages of this method not only include high productivity, but also the possibility of introducing any type of fiber into the paper to change its properties. In particular, for small vessels it is best to introduce the adsorbent directly into the VSIC layers and then subject them to vacuum treatment in the vessels at temperatures of 100-120°C. The use of crumpled film made of polyethylene terephthalate PET is also promising as a lining material that does not give off any kind of particles during assembly and use [4].

From the great variety of thin paper materials (8-10 g/m^2) we chose the following four composites on the basis of calorimetric tests: SNT-10, USNT-10, UNT-10, and ANT-10. The first two are based on synthetic fibers, while the other two are based on microthin basalt fibers. The materials USNT-10 and UNT-10 include carbon-bearing adsorbents, while ANT-10 includes vinyl chloride-vinyl acetate copolymer fibers. Table 1 shows the strength characteristics of the new lining materials and the result of thermal tests of VSIC packets with these materials on a plane calorimeter. We used a PET DA film 5 μm thick as the shield in

TABLE 1. Characteristics of Lining Materials

Lining material	Ultimate tensile strength, MPa	Thermal conductivity of the VSIC on the calorimeter, μW/(cm·K)
EVTI-7	3,5	0,31
SNT-10	3,7	0,35
USNT-10	3,3	0,34
UNT-10	0,8	0,33
ANT-10	0,9	0,29
NT-10	1,0	0,30
PET film	190	0,35

TABLE 2. Results of Tests of VSIC's on Kh-34B Vessels

Lining material	Life of vessel, days	Effective thermal conductivity of VSIC, μW/(cm·K)
EVTI-7	180—210	1,41—1,1
SNT-10	203—218	1,17—1,03
USNT-10	235—250	0,92—0,83
UNT-10	235—254	0,92—0,81
ANT-10	218—262	1,03—0,76
PET film	217	1,04

all of the specimens. The density of the lay of the specimens was 25-28 shields/cm. The error of determination of the thermal characteristics on the calorimeter was no greater than 10% and was for the most part determined by the accuracy with which we maintained adiabatic conditions on the lateral surface of the specimen. The specimen was 6 mm thick and 160 mm in diameter. All of the tested composites had fairly similar characteristics in the range 0.29-0.35 μW/(cm·K). It can be seen from Fig. 3 that having a VSIC with such characteristics on a Kh-34B vessel could increase its service life to 360 days.

On standard vessels with a VSIC based on glass fabric EVTI-7, its effective thermal conductivity is 3.5-4.5 times greater than on the calorimeter (see Tables 1 and 2). This result can be attributed not only to the high level of gas evolution from this material and the poor vacuum in the layers of the VSIC, but also to possible adhesion of the EVTI-7 to the surface of the shield after heating of the vessel. Such adhesion increases contact heat transfer. Another shortcoming of the standard EVTI composition is the harmful effect of silicon particles on the breathing passages and mucous membranes of workers. These problems are largely overcome with the use of new lining materials in the VSIC.

Table 2 shows the results of thermal tests of new VSIC composites for Kh-34B vessels with liquid nitrogen. The VSIC shields were made of PET DA film 5 μm thick. The insulation was formed by the orbital machine method with a strip of material 75-90 mm thick. The mean thickness of the VSIC packet on the vessel was 70 mm, while the density of the lay was 9-11 shields/cm.

It is evident from Table 2 that all of the new VSIC's have a thermal conductivity greater than the standard VSIC based on glass fabric EVTI-7. The best results were obtained with a VSIC which contained adsorbents (USNT-10 and UNT-10) in the layers. Equally good results were obtained with the VSIC containing vinyl chloride-vinyl acetate copolymer fibers (ANT-10). Having a thermal conductivity 1.5-1.6 times greater than the standard VSIC, they make it possible to increase vessel service life by 25% (see Fig. 3). It should be noted that the use of materials UNT-10 and USNT-10 requires vacuum treatment of the vessels with heating at a temperature no lower than 100-120°C.

The liners based on synthetic fibers (SNT-10) and the film linings of PET are also thermally efficient and very adaptable to commercial use. Another advantage they possess is the lack of harmful dust particles during assembly. These materials are strong (see Table 1) and are thus quite suitable for machine assembly. The PET film lining generally does not generate any particles during use, which makes it promising for application in cryogenic devices containing optical elements. Film PET materials and synthetic papers SNT-10 and USNT-10 (TU-13-7308 001-73-85) are already being produced commercially.

Industry has also mastered the production of new paper materials NT-10 and NT-8 (TU-13-7308001-695-84) based on basalt fibers. Due to insufficient strength (see Table 1), these materials are not suitable for machine assembly of VSIC's but are promising for large cryogenic units with VSIC's having a packet structure.

The use of new VSIC's has made it possible to increase the service life of Kh-34B vessels from 200 to 260 days and to reduce the difference in thermal conductivity on the vessel and calorimeter from a factor of 3.5-4.5 to a factor of 2.5-3. Also promising is the use of VSIC's with layers made of crumpled PET film in the case where the VSIC can be heated in the vessel to 130-140°C for 1-1.5 h [5]. By heating the PET film to the softening point at these temperatures and subjecting the shields and linings to thermal relaxation to remove compressive strains in the layers from assembly, it is possible to significantly reduce contact heat transfer in the VSIC. In this case, there is an additional possibility for a reduction in the radiative component of heat transfer in the VSIC. This can be done by increasing the number of shields per unit thickness of the insulation to 20-25 shields/cm. Here, the compressive forces in the VSIC remain low.

NOTATION

Q_Σ, total heat flux; F, cross-sectional area; T, temperature; λ, thermal conductivity; ℓ, length; d, diameter; δ, thickness; $\dot{m}$ and $\dot{m}_0$, rate of evaporation of liquid nitrogen with and without the use of the cold of the vapors; R, service life of vessel before replenishment of cryogen; r, radius; x, coordinate along the throat. Indices: t, throat; pg, plug; in, insulation; gs, gas; ves, vessel.

LITERATURE CITED

1. G. G. Zhun', V. F. Getmanets, R. S. Mikhal'chenko, et al., Inzh.-Fiz. Zh., 54, No. 4, 600-607 (1988).
2. V. F. Getmanets, R. S. Mikhal'chenko, and P. N. Yurchenko, Inzh.-Fiz. Zh., 47, No. 4, 676 (1984).
3. B. J. Verkin, R. S. Mikhalchenko, V. F. Getmanets, and V. A. Mikheev, Proc. of 10th Int. Cryog. Eng. Conf., 529-538 (1984).
4. Low-Temperature Insulation, Inventor's Cert. No. 970025 SSSR: M. Kl.^{3} G-17S3/02//F16 59/06.
5. Method of Thermally Insulating Cryogenic Devices: Inventor's Cert. No. 1262183 SSSR: 4F17S3/00.

TRANSPORT CALCULATIONS IN HYDROGEN STORAGE AS METAL HYDRIDES

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A method is described for analyzing reactive transport in a metal hydride layer, which has been checked on a model for a hydrogen accumulator.

Hydrogen storage in hydrides formed by intermetallides has some advantages over traditional methods [1]; the hydrogen in these hydrides is stored under conditions close to normal, while the density may exceed that of liquid hydrogen. Compressed gases or cryogenic storage can produce analogous effects but require extreme conditions (high pressures or low temperatures). Additional advantages are that the hydrogen released from the hydrides is very pure and at high pressure.

These advantages are accompanied by limitations due to the delay in the transport in the layers, which means that the hydrogen accumulator is charged at a restricted rate (the device that provides the bound storage), and the same applies to the release rate. The desire to accelerate the exchange leads either to overheating the heat carrier or to complicated accumulator design, which increases the metal capacity because it is necessary to extend the interior surfaces. This means that one needs a careful analysis of the heat and mass transfer in the layers to optimize the design.

Hydrogen reacting with a hydride involves various transport mechanisms such as transport through the metallide layer, hydrogen-molecule dissociation at active surfaces (or

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