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DEGREE OF NONEQUILIBRIUM OF THE VAPOR AND LIQUID PHASES DURING THE BOILING OF BINARY CRYOGENIC MIXTURES

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Results of a theoretical and experimental investigation are presented for liquid-vapor compositions during the boiling of binary cryogenic mixtures. Factors affecting the degree of phase nonequilibrium are set down.

The majority of processes utilized in cryogenic engineering is accompanied by heat and mass transfer during the phase transitions of binary and multicomponent mixtures. Realization of different technological operations during the storage, transportation, and retention in a cryostat is associated with the boiling of liquid mixtures and with their partial or total evaporation.

Mass transport of components within the phase and between the phases occurs together with phase transformation during the boiling of mixtures. One of the quantities exerting substantial influence on the integrated characteristics of the mixture boiling process is the distinction between the compositions of the liquid and vapor phases. By the equilibrium condition a greater low-boiling component is contained in the vapor than in the liquid. The gradual diminution of the low-boiling component in the liquid is associated with its excess concentration in the vapor and, as a result, so is the rise in the mixture boiling point. This exerts influence on the dynamics of the formation, growth, separation, and buoyancy of the vapor bubbles [1].

The change in composition of the liquid residue is determined by the concentration of the mixture components in the outgoing vapor, which is also related to the hydrodynamic and thermal conditions under which the boiling process progresses. The question of the vapor composition being formed here is studied extremely minimally and data in the literature are contradictory. Existing computational dependences of the change in mixture component concentration during boiling are obtained for equilibrium conditions, the deviation from which in real processes can be quite substantial [2, 3]. The determination of the nonequilibrium of the liquid-vapor phases is especially urgent for cryogenic mixtures since the accumulation of the fast-boiling components in the liquid residue is associated with the possibility of their deposition in solid form in different elements of the system and thereby creation of a dangerously explosive situation.

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At the present time there is no standard methodology for estimating the degree of deviation from equilibrium. The mechanism for origination of nonequilibrium in bubble formation processes during the boiling of binary mixtures and their subsequent movement to the free surface is not clear.

The composition of the vapor being formed during boiling of a mixture depends on the conditions of bubble generation and growth on the warm surface. If they are examined for the boiling of a pure component in many papers, they are barely studied for the case of mixture boiling at this time. The combination of the liquid and wall thermophysical conditions exerts noticeable influence on bubble growth. Analysis of many cryogenic-liquid warm-surface material combinations at almost atmospheric pressures enabled the authors of [4] to make a deduction about the preferred influence of microlayer evaporation. Among the specific features of cryogenic liquids should be the significant magnitude of the adhesion forces causing the "leakage" of liquid under the growing bubble. In this connection, a model of nitrogen bubble growth on a heat transmitting surface was proposed in [4] due to the evaporation of a liquid microlayer whose thickness does not vary in time. The dependences characterizing the dynamics of bubble growth during boiling of pure cyrogenic liquids that were obtained in [4] yield satisfactory agreement with experimental results. This permitted development of the proposed model for the case of the boiling of binary cryogenic mixtures.

The instant of bubble growth when the liquid microlayer composition under them agrees with the composition of the main mass of the binary mixture is considered as the initial state. The following assumptions are made here: the liquid of the microlayer under the growing bubble is mixed completely; the vapor being formed at each instant is in equilibrium with the liquid of the microlayer; liquid evaporation from the curvilinear bubble surface, the vapor condensation on it, and the diffusion mass transfer can be neglected exactly as in the model in [4]; and the concentration of liquid leaking under the vapor bubbles is constant during its growth, and we neglect the influence of adjacent bubbles.

Since the thickness of the liquid microlayer is considered a function only of the radius, then the liquid leaking under from the circumference minus the quantity going into the growth of the microlayer itself goes into the vapor. Therefore, the equation of material balance and of balance in the low-boiling component can be written for an elementary process in the form

$$dL_R = dG_R + dM_R, \tag{1}$$

$$xdL_R = y_R dG_R + M_R dx_R + dM_R x_R. \tag{2}$$

Let us express the differentials of the dependent quantities in terms of the derivative and differential of the independent variable

$$dG_R = G'_R dR, \quad dM_R = M'_R dR. \tag{3}$$

Taking (1) and (3) into account, (2) is written in the form

$$x[(G'_R + M'_R)\, dR] = y_R G'_R dR + M'_R dx_R + M'_R dR x_R, \tag{4}$$

from which

$$\frac{dx_R}{dR} = \frac{(x - y_R)\, G'_R + (x - x_R)\, M'_R}{M_R}. \tag{5}$$

To determine the values of the quantities in (5), we turn to the model in [4] (for pure cryogenic fluids) (Fig. 1). In connection with the fact that the angle α is small, part of the bubble volume, which is a truncated spherical segment bounded by a conical microlayer surface from below, can be neglected. Then the bubble volume equals the volume of a truncated sphere of radius R and

$$G_R = \pi R^3/3\, (1 + \cos\Theta_D)^2 (2 - \cos\Theta_D)\, \rho^V, \tag{6}$$

$$G'_R = \pi R^2 (1 + \cos\Theta_D)^2 (2 - \cos\Theta_D)\rho^V. \tag{7}$$

Thickness of the liquid microlayer under the bubble can be determined for small α in a circular section of radius R sin Θ_D from the formula

$$\delta_{(R\sin\Theta_D)} = \alpha R \sin\Theta_D. \tag{8}$$

Then

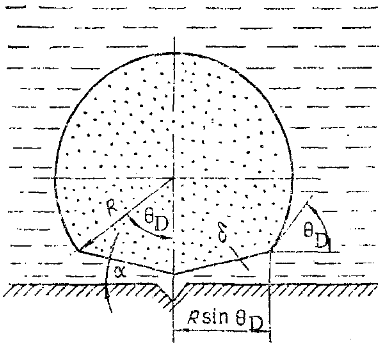

Fig. 1. Model of vapor bubble growth on a warm surface for cryogenic liquids [4]. Θ_D is the dynamic boundary angle connecting the interphasal surface and the solid wall, and δ is the liquid microlayer thickness under the growing bubble.

$$M_R = \frac{2}{3}\pi R^3 \rho^L \alpha \sin^3\Theta_D \tag{9}$$

and

$$M'_R = 2\pi R^2 \rho^L \alpha \sin^3\Theta_D. \tag{10}$$

Substituting the values of G'_R and M'_R from (7) and (10) into (5), we obtain

$$\frac{dx_R}{dR} = \frac{(x - y_R)(1 + \cos\Theta_D)^2(2 - \cos\Theta_D)\rho^V + 2(x - x_R)\alpha\sin^3\Theta_D\rho^L}{2/3\alpha\sin^3\Theta_D\rho^L R}. \tag{11}$$

An analysis, executed in [5], of this expression showed that as the bubble radius grew, the quantity dx_R/dR diminishes, i.e., the liquid microlayer under the bubble is depleted by the low-boiling component. Its concentration in the bubble was determined from (11) under the assumption that the vapor being formed at each instant from the microlayer liquid is in equilibrium with this liquid. Therefore, the composition of the vapor in the bubble deviates from the main equilibrium mass of the liquid during the whole period of its growth on the warm surface. The degree of deviation from equilibrium is determined by the parameters in (11) and depends on the physicochemical properties of the mixture, its composition, and the hydrodynamic and thermal conditions under which the boiling progresses.

After separation from the warm surface, the vapor bubble floats to the free level under the action of the Archimedes force. In the absence of thermodynamic equilibrium between phases in a heterogeneous system, diffusion fluxes occur for the components directed toward the phase with the lower value of the chemical potential. The heat transfer that occurs because of the temperature gradient between the phases coming into contact will exert influnce on the total effectiveness of the mass transfer.

The problem of a change in mass of the floating bubble and the low-boiling component concentration therein during boiling of a mixture in a cryogenic vessel with heating of the bottom is considered below. These assumptions are made:

The bubble rise occurs in a layer of heated liquid having a constant composition over its whole height; the change in liquid concentration occurs only in time; similarly for the temperature;

heat and mass transfer during rise of the bubble occurs only between the bubble and the heated liquid; with respect to the external medium this process is adiabatic;

we neglect the influence of adjacent floating bubbles on the process;

we also neglect the pressure change in the bubble as it rises to the free surface of the liquid;

the temperature of the vapor and liquid on the phase interface equals the saturation temperature T_s at the pressure of the liquid mixture; the concentration of the vapor in the bubble because of liquid evaporation corresponds to the equilibrium main mass of liquid for T_s. We denote it by y_x^p. Upon vapor condensation from the bubble, a liquid is obtained which is in equilibrium, in composition, with the vapor in the bubble $x_{y_h}^p$.

As the bubble rises, evaporation occurs of the heated liquid in it. The change in liquid mass in an infinitesimal time interval can here be determined for a layer of height dh by the formula

$$d^2l = \frac{\partial L}{\partial \tau} d\tau dh. \tag{12}$$

The vapor being formed as a result of liquid evaporation is partially expended in a change in the bubble mass in the separated layer at each time, and partially in an increase in the mass of the vapor passing through this layer. The change in vapor mass being contained in the layer is written analogously

$$d^2n = \frac{\partial N}{\partial \tau} d\tau dh. \tag{13}$$

The change of mass of bubbles passing through a layer of height dh in a time $d\tau$ is determined from the expression

$$d^2g = \frac{\partial G}{\partial h} dh d\tau. \tag{14}$$

The balance of the elementary mass fluxes for the isolated layer is

$$\frac{\partial L}{\partial \tau} d\tau dh = -\frac{\partial N}{\partial \tau} d\tau dh - \frac{\partial G}{\partial h} dh d\tau \tag{15}$$

or

$$\frac{\partial L}{\partial \tau} = -\frac{\partial N}{\partial \tau} - \frac{\partial G}{\partial h}. \tag{16}$$

A comparison of $\frac{\partial N}{\partial \tau}$ and $\frac{\partial G}{\partial h}$ for the case when $\frac{\partial N}{\partial \tau}$ has a maximal value shows that $\frac{\partial N}{\partial \tau}$ is ∿0.5% of $\frac{\partial G}{\partial h}$. Consequently, we later neglect the quantity $\frac{\partial N}{\partial \tau}$. Then

$$\frac{\partial L}{\partial \tau} = -\frac{\partial G}{\partial h}. \tag{17}$$

The change in mass of the bubbles passing through a layer 1 m high in 1 sec, that is in the right side of (17), is determined by the total of the heat and mass transfer because of the interaction of four flows: $\frac{\partial Q^{LB}}{\partial h}$ is the diffusion flux of the low-boiling component, $\frac{\partial G^{LB}}{\partial h}$ is caused by liquid evaporation during the heat transfer, $\frac{\partial Q^{HB}}{\partial h}$ is the contradiffusion flux of the high-boiling component, and $\frac{\partial G^{HB}}{\partial h}$ is associated with vapor condensation on the interphase boundary. The first two terms increase the bubble mass, while the two second terms decrease it. Therefore, the material balance equation of the elementary mass fluxes has the form*

$$\frac{\partial L}{\partial \tau} = \frac{\partial G^{LB}}{\partial h} - \frac{\partial G^{HB}}{\partial h} - \frac{\partial Q^{LB}}{\partial h} + \frac{\partial Q^{HB}}{\partial h}. \tag{18}$$

Taking account of the assumptions made, the balance in the low-boiling component is written as follows:

$$\frac{\partial L}{\partial \tau} x + \frac{\partial x}{\partial \tau} L = -\frac{\partial G^{LB}}{\partial h} y_x^{p} + \frac{\partial G^{HB}}{\partial h} x_{y_h}^{p} - \frac{\partial Q^{LB}}{\partial h}. \tag{19}$$

The first two members in the right side of (19) govern the change in the quantity of the low-boiling component in the bubbles that pass through the liquid because of heat transfer, while the last component is due to diffusion mass transfer. The density of the diffusion flux through the phase contact surface can be determined from the general theory of mass transfer. The main mass transport equation in a disperse gas–liquid system is written in differential form for the volume element in which the vapor and liquid fluxes are mixed completely:

$$\frac{\partial Q}{\partial F} = \frac{\rho^v}{\mu^v} k_{0y} (y^* - y), \tag{20}$$

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*The signs are reversed in (18) since $\frac{\partial L}{\partial \tau}$ is written with a plus sign.

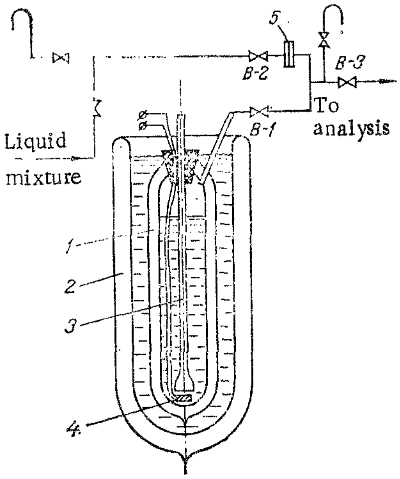

Fig. 2. Diagram of the experimental vessel.

where y* is the concentration of the low-boiling component in the vapor that is in equilibrium with the liquid of the flux nucleus, while y is its concentration on the phase interface. The difference in concentrations, in the parentheses, is the motive force of the mass transfer process. The total mass transport factor k_{0y} reflects not only the kinetics of the process but also the change in the phase contact surface F. Determination of F under complex phase interaction conditions is fraught with difficulty. Consequently, generalized characteristics, for instance the phase contact surface referred to the volume of the dispersed layer $a_V = F/V$, are ordinarily used in mass transport computations. For the volume element for which the base area is 1 m², the diffusion flux of the low-boiling component equals

$$\frac{\partial Q^{\mathrm{LB}}}{\partial h} = k_{0y}\frac{\rho^v}{\mu^v} a_V (y_x^{\mathrm{p}} - y_h), \tag{21}$$

where $(y_x^{\mathrm{p}} - y_h)$ is the difference in low-boiling component concentrations in the flux nucleus and on the phase interface according to the mass-transfer model taken. For the high-boiling component

$$\frac{\partial Q^{\mathrm{HB}}}{\partial h} = k_{0y}\frac{\rho^v}{\mu^v} a_V [(1 - y_x^{\mathrm{p}}) - (1 - y_h)]. \tag{22}$$

The total quantity of heat given off by the liquid to the vapor during its evaporation

$$r_L \frac{\partial L}{\partial \tau} d\tau dh \tag{23}$$

is the resultant magnitude of the following heat fluxes:

the heat given off to the vapor because of heat transfer from the heated liquid and its evaporation in bubbles:

$$\alpha_{\mathrm{e}} F_h (T - T_s)\, d\tau dh, \tag{24}$$

heat liberated during vapor condensation on the phase interface:

$$\alpha_{\mathrm{c}} F_h (T_h^v - T_s)\, d\tau dh, \tag{25}$$

heat perceived by the vapor during evaporation of the low-boiling component during diffusion mass transport:

$$k_{0y} r^{\mathrm{LB}} \left(\frac{\rho^v}{\mu^v}\right)^{\mathrm{LB}} a_V\ (y_x^{\mathrm{p}} - y_h)\, d\tau dh, \tag{26}$$

heat liberated during condensation of the high-boiling component during diffusion mass transfer:

$$k_{0y} r^{\mathrm{HB}} \left(\frac{\rho^v}{\mu^v}\right)^{\mathrm{HB}} a_V\ [(1 - y_x^{\mathrm{p}}) - (1 - y_h)]\, d\tau dh. \tag{27}$$

The heat-balance equation has the following form

$$r_L \frac{\partial L}{\partial \tau} = \alpha_{\mathrm{e}} F_h (T - T_s) - \alpha_{\mathrm{c}} F_h (T_h^v - T_s) + r^{\mathrm{LB}} k_{0y} \left(\frac{\rho^v}{\mu^v}\right)^{\mathrm{LB}} a_V (y_x^{\mathrm{p}} - y_h) + r^{\mathrm{HB}} k_{0y} \left(\frac{\rho^v}{\mu^v}\right)^{\mathrm{HB}} a_V [(1 - y_x^{\mathrm{p}}) - (1 - y_h)]. \tag{28}$$

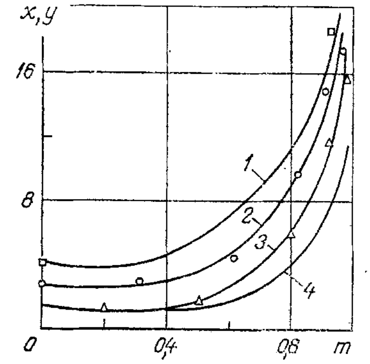

Fig. 3

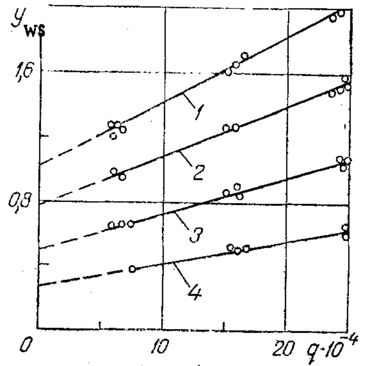

Fig. 4

Fig. 3. Change in oxygen concentration in the liquid and vapor phases (volume fraction, %) as a dependence of the relative quantity m of mixture being evaporated: 1) in the liquid phase x; 2) in bubbles being separated from the warm surface y_{ws}; 3) in the vapor above the free liquid surface y_{fs}; 4) in the vapor in equilibrium with the liquid phase y^p; $q = 15\cdot 10^4$ W/m².

Fig. 4. Influence of the heat flux density q(W/m²) on the oxygen concentration in bubbles growing on a warm surface y_{ws} (volume fraction, %). Liquid phase composition: 1) 3.2% O_2; 2) 2.5% O_2; 3) 1.6% O_2; 4) 0.8% O_2; dashed curves are the extrapolation of $y_{ws} = f(q)$ to $q = 0$.

The expression characterizing the change in vapor composition during its motion to the free level can be obtained during the combined solution of (19) and (28) with (21) and (22) taken into account.

The analytical dependences represented are obtained for a number of simplifying assumptions. Their validity is subjected to confirmation in an experimental study.

The vessel shown in Fig. 2 in which the mixture is boiled consists of two transparent (for visual observation) coaxial Dewars. The liquid being investigated is in the inner 1, while the outer Dewar 2 with liquid nitrogen is for shielding from external heat fluxes. The vapor composition in the bubbles was analyzed either directly after their separation from the warm surface 4 (with valve B-1 closed and B-2 open), or after they passed through valve B-1 through the liquid column. The presence of two regimes permitted investigation of not only the mechanism for origination of the deviation of the vapor and liquid from equilibrium but also the factors affecting the degree of phase nonequilibrium during boiling. The liquid rise in the bubble trap 3 (with the valve B-1 closed) was eliminated by using the shut-off washers 5 and the fine regulation valve B-3. A nitrogen—oxygen mixture with up to 20% O_2 concentration was investigated. The O_2 concentration was measured by using a LKhM-8MD chromatograph of second modification. The warm surface was a Nichrome plate of size 0.014 × 0.006 m.

Test data on the change in oxygen concentration in the bubbles y_{ws} at the time of their separation from the warm surface (curve 2) and after they have traversed a liquid column y_{fs} (curve 3) are shown in Fig. 3 as a function of the liquid level h or the relative quantity of mixture m being evaporated. Curves 1 and 4 characterize the equilibrium states of the vapor and liquid in this process. It is seen from the figure that the O_2 concentration in the bubbles being separated from the warm surface (curve 2) are substantially greater during the whole mixture boiling process than in the vapor which is in equilibrium with the main liquid mass (curve 4). The degree of deviation from equilibrium is determined by the bubble generation and growth conditions on the warm surface. It is shown in Fig. 4 how the vapor composition varies in the bubbles as a function of the heat flux density q. The curves $y_{ws} = f(q)$ are approximated with sufficient accuracy by straight lines, whose slope increases as the oxygen concentration increases in the liquid phase. The values of y_{ws} obtained by extrapolating the lines $y_{ws} = f(q)$ to $q \simeq 0$ (dashed lines in Fig. 4) should be the oxygen concentration in the vapor in equilibrium with the liquid, as is confirmed by tabulated data on the vapor-liquid equilibrium of a nitrogen—oxygen mixture (curve 4) in Fig. 3. The bubble composition changes as they rise. As is seen from Fig. 3, curves 3 and 4 agree, i.e., $y_{fs} = y^p$, for a high liquid level in the vessel. At the end of the mixture boiling process, the vapor given off from the vessel is close in composition to the bubble composition at the warm surface.

The degree of deviation from equilibrium is conveniently estimated by a parameter $\varkappa$, connecting the vapor and liquid phase compositions

$$\varkappa = \frac{y - y^{\mathrm{p}}}{x - y^{\mathrm{p}}}. \tag{29}$$

where $\varkappa$ varies between zero, when $y = y^{\mathrm{p}}$, and one when the vapor and liquid compositions are equal (maximal deviation from equilibrium). The degree of vapor nonequilibrium in the bubbles generated by the warm surface and the main liquid mass determined experimentally for the N_2–O_2 mixture is within the limits 0.5-0.8 depending on the composition, and reaches one for $q - q_{cr}$. The relative error in measuring the concentration did not exceed ±0.96% in the tests. The error in measuring the composition of the initial fluid mixture is ±4.5%. Comparison of the test values of the oxygen concentration in bubbles of the separation diameter and the computed values from dependence (11) was performed for the following values of the parameters: $\Theta_D = 47°$ is the mean value per period of bubble growth [4]; $\alpha = 0.003$ rad is determined by starting from the thickness of the liquid microlayer under the growing bubble; the Carslaw and Jaeger [6] solution for a semibounded solid on whose surface a thin constant-thickness film is superposed was used to compute the temperature distribution in the liquid microlayer. The deviation of the computed from the test values was from ±4% to ±9%, which increased as the values of q grew. The change in low-boiling component concentration in the bubble as a function of the liquid level was computed by the dependences presented by finite differences. The greatest deviation from the test values did not exceed ±13%.

NOTATION

x, y, low-boiling component concentrations in the liquid and vapor phases, respectively, in volume fractions, %; with subscript R, in the liquid microlayer under the bubble growing on the warm surface and in the vapor bubble; with subscript p, the equilibrium concentration; R, bubble radius, m; L_R, number of moles of liquid flowing from the circumference to the base of the bubble during the growth of its radius from 0 to R, kmoles; G_R, number of moles of mixture contained in the vapor bubble of radius R, kmoles; M_R, number of moles of mixture being contained in the liquid microlayer under the bubble of radius R, kmoles; ρ, mixture density, kg/m³; T, temperature, °K; h, height of liquid layer above the warm surface, m; l, number of moles of liquid in a layer of height dh, kmoles; g, number of moles of vapor that passed through a layer in a time dτ, kmoles; L, number of moles of liquid per unit length of layer, kmoles/m; G, number of moles of vapor that passed through the liquid in unit time, kmoles/sec; N, number of moles of vapor contained in unit length of the layer, kmoles/m; μ, molecular mass, kg/kmole; r, heat of vapor formation, kJ/kmole; α, angle between the generator of the conical microlayer surface and the work surface, rad; with subscript e, the coefficient of heat transfer from the liquid to the vapor, W/m²·K; with the subscript c, the coefficient of heat transfer from the vapor to the fluid during condensation on the phase interface, W/m²·K; F, phase contact surface, m². Subscripts: s, saturated state; cr, critical; h, height h above the warm surface; superscripts: L, liquid phase; v, vapor phase; LB, low-boiling component; and HB, high-boiling component.

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GAS-COOLED COMPOUND POROUS CURRENT LEADS FOR CRYOGENIC CABLES

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UDC 536.483

Studies have been made on porous current leads having a superconductor at the cold end. Estimates are made of the heat leak along the current lead in relation to the gas flow rate and current load.

Current leads are major components in cryogenic machines and cryogenic electrical transmission lines. There are many different designs for current leads for cryogenic machines, solenoids, and transmission lines handling currents from tens of amperes up to some kiloamperes. The leads in such a device work under conditions where one end (the warm one) is at room temperature and the other (the cold one) is at helium temperature, i.e., there is a temperature difference of about 300°K along the lead.

In designing such a lead one naturally has to provide not only the appropriate current carrying capacity but also the minimum heat leak to the cryogenic liquid in order to reduce the losses.

A current lead is a thermal bridge joining the warm zone to the cold one, and it also has a finite electrical resistance, which produces heat when current flows. To reduce the heat leak along the lead considered as a bridge transmitting heat by conduction, it must be made of material with the minimum thermal conductivity and be thin and long. On the other hand, to reduce the heat leak for a lead considered as a current carrier that produces heat when a current flows, it must be made of a material with a minimum electrical resistance, i.e., of a mterial having a fairly high thermal conductivity and fairly large cross section.

Therefore, the lead is subject to conflicting requirements in meeting the same purpose of providing the minimum heat leak to the helium zone. Therefore, there exists an optimum design providing the minimum leak on the basis of the heat production in the lead when a current flows and the heat leak along the material due to thermal conduction.

It has been pointed out in [1] that the minimum heat leak is also an important condition for reliable operation of cryogenic equipment. To ensure stable operation, one sometimes deliberately increases the helium flow rate above the calculated value.

It is desirable to cool the current leads to minimize the heat leak, and to make this cooling as effective as possible the heat transfer must be organized in such a way as to approximate to ideal, i.e., the temperatures of the cooling gas and lead should be equal in a given section. We therefore have to consider how to provide such conditions. It has been suggested that the surface area flushed by the gas should be increased by forming ribs on the lead and using porous metals, or else perforated metal sheets, metal grids, or sets of metal sheaths, or else to design compound leads, in which a superconductor is used to eliminate the heat production when the current flows at the cold end [2-9].

In most optimized leads, the minimum heat leak into the cold zone is 1 mW/A [10]. The calculation showed that use of a superconductor in a lead should roughly halve the leak into the cold zone, but the experimental results in the literature are very conflicting.

We have attempted to minimize both components of the heat leak along the lead. The first is due to thermal conduction and is reduced by using a material with a low thermal conductivity, while the second, which is due to the current, is reduced by using a superconductor, and that without a copper matrix. Calculations show that the leak along the lead can

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