COOLING OF CRYOGENIC FLUIDS BY EVACUATION OF THE VAPOR SPACE

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Questions of cooling and freezing cryogenic fluids by evacuating the vapor space are examined. The results of a theoretical investigation are compared with experimental data.

The problem of cryostating objects at temperature levels which cannot be achieved by using cryogenic fluids which boil at atmospheric pressure often occurs in low-temperature investigations. Production of such temperatures can be realized most simply by reducing the pressure over the cryogenic fluid [1, 2].

Using such a method is associated with the discharge of a definite quantity of fluid which must be evacuated in the form of vapor in order to obtain a given temperature.

An analytic solution of this problem in generalized form is given in [1] by using the Watson equation for processes of fluid cooling to the triple point.

However, practical utilization of the dependences in [1] is constrained because of the discrepancies in the magnitudes of the heat of vapor formation r obtained by means of the Watson equation and from experiment for real gases at low temperatures.

Moreover, freezing processes of the cryogenic fluid are not examined in [1].

A mechanism and the kinetics of the process of cryogenic fluid solidification are given in [2] and the equation for the solidification process is derived, but cooling the fluid to the triple point is not considered. Hence, the question arises of creating a refined single engineering method for computing the cooling and freezing processes for cryogenic fluids on the basis of the method in [1] by using temperature dependences of the heat of evaporation (fusion) found experimentally in [3].

The process of cryogenic fluid cooling by evacuating the vapor space until a solid cryogenic agent is obtained can provisionally be separated into two stages shown on the T vs s diagram (Fig. 1):

1–2) fluid cooling in the limit to the triple point (proceeds with the reduction of the pressure from $p_1 = p_{am}$ to $p_{tp} = p_2$);

2–3) freezing of the fluid for p_{tp} = idem. Cooling of the solid along the line 3–4 will occur under further evacuation.

Considering an adiabatically isolated system, we can write the differential equation of the process 1–2 of fluid cooling [1]. Equation (1) is also valid for cooling of the solid cryogenic agent in the process 3–4:

$$Gc_s dT - r dG = 0. \tag{1}$$

Solving (1) for G (in the case of fluid cooling), we obtain

$$\ln \frac{G_2}{G_1} = \int_{T_1}^{T_2} \frac{c_s dT}{r}. \tag{2}$$

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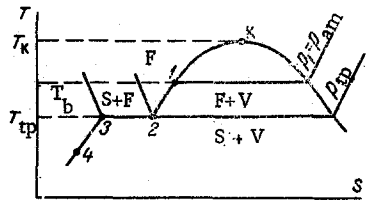

Fig. 1

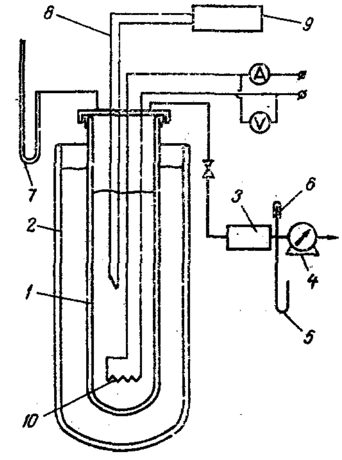

Fig. 2

Fig. 1. Cryogenic fluid cooling and solidification processes.

Fig. 2. Diagram of the experimental setup: 1) experimental Dewar vessel with cryogenic fluid; 2) shielding Dewar vessel with liquid nitrogen; 3) VN-4 vacuum pump; 4) GSP-400 gas counter; 5) liquid manometer; 6) thermometer; 7) mercury manometer; 8) thermocouple; 9) potentiometer; 10) electric heater.

For the domain of cryogenic fluid solidification (the process along the line 2–3), we can write the equation

$$(G_2 - G_3)\, xr_f = r_{tp} G_3. \tag{3}$$

Solving (3) for G_3 we obtain

$$G_3 = \frac{G_2 x r_f}{r_{tp} + x r_f}. \tag{4}$$

Under real conditions the process of cryogenic fluid solidification, exactly as its cooling, is nonadiabatic. Hence, a correction for the heat influx must be introduced. The quantity of vapor removed to cancel the heat influx can be determined from the ratio

$$G_h = \frac{Q_h}{r}. \tag{5}$$

Let us consider the analytic solution of (2) for the most widespread cryogenic fluids such as nitrogen, oxygen, neon, and para hydrogen, and let us compare it to the experimental results obtained for some of them.

The specific heats c_s of nitrogen, oxygen, and neon can be taken as 2.02 kJ/kg · deg, 1.68 kJ/kg · deg, 1.82 kJ/kg · deg, respectively, in the T_0–T_{tp} temperature range with an error not exceeding 3% of the experimental values [5-7].

The temperature dependence of the specific heat c_s of liquid para hydrogen, as well as the heat of vapor formation r_s of nitrogen and oxygen in the T_0–T_{tp} range, can be assumed linear with an error not exceeding 5% of the experimental values [4, 6, 7] and can be expressed, respectively, by Eqs. (6), (7), and (8):

$$c_s = 7.02 + 0.415\,(T - 13.8) \text{ kJ/kg·deg}, \tag{6}$$

$$r = 292 - 1.19\,T \text{ kJ/kg}, \tag{7}$$

$$r = 288 - 0.833\,T \text{ kJ/kg}, \tag{8}$$

To 3% accuracy, the values of r for neon and para hydrogen can be considered constant in the same temperature range and equal, respectively, to 86 kJ/kg and 452 kJ/kg [7, 3].

Substituting the values of c_s and r for each of the substances into (2), we obtain expressions (8) for nitrogen, (10) for oxygen, (11) for neon, and (12) for para hydrogen, respectively:

$$\ln \frac{G_2}{G_1} = 1.96 \int_{T_1}^{T_2} \frac{dT}{292 - 1.19T}, \tag{9}$$

$$\ln \frac{G_2}{G_1} = 1.68 \int_{T_1}^{T_2} \frac{dT}{288 - 0.833T}, \tag{10}$$

$$\ln \frac{G_2}{G_1} = 0.0211\,(T_2 - T_1), \tag{11}$$

$$\ln \frac{G_2}{G_1} = \frac{1}{452} \int_{T_1}^{T_2} [7.02 + 0.415\,(T - 13.8)\,dT. \tag{12}$$

The setup shown in Fig. 2, was used for an experimental confirmation of these dependences during the cryogenic fluid cooling and freezing processes.

The cryogenic fluid being investigated was poured into the experimental Dewar vessel (1). To diminish the heat influx, the vessel (1) was shielded by liquid nitrogen which was poured into the guard vessel (2). Visual observations were conducted through a transparent strip in each of the vessels. Vapor of the cryogenic fluid was evacuated by means of a VN-4 vacuum pump; the quantity of evacuated vapor was recorded by a GSB-400 counter. The vapor pressure above the fluid surface was measured by using an absolute mercury manometer with a 1 mm Hg error.

The mean fluid temperature in the experimental vessel was measured by using a thermocouple of gold + 0.02% iron–copper.

The percentage content of solid phase in the mixture was determined by means of the vapor evacuated and was compared selectively with the quantity found by the method of melting the "ice" formed during cooling. To this end, an electric heater 10, whose power was determined from readings on an ammeter A and voltmeter V of 0.2 class, was placed on the bottom of the experimental vessel.

The quantity of solid phase in the mixture, which was determined by the melting method, was computed according to the equation

$$G_{so} = \frac{(Q_H - W_f)\,\tau_f}{r_f}. \tag{13}$$

The values of the percentage content of solid phase in the mixture, obtained by measuring the quantity of evacuated vapor, differed from the quantities found by means of the melting method by 3-15%. The mean value of the solid phase content in hydrogen and nitrogen hence varied within 10-70% limits.

It is difficult to assure and control the uniformity of the temperature field over the whole volume of cryogenic agent during melting of the "ice." Hence, the error in determining the percentage content of solid phase by means of this method can be greater than the determination of this content by means of the quantity of evacuated vapor.

We henceforth used the method of determining the solid phase content in a mixture by means of the quantity of evacuated vapor as it was more simple and reliable.

Graphs of the dependences of the cryogenic fluid temperature T on the quantity G_2/G_1, computed by means of (9)-(12), are presented in Fig. 3.

Values of $T = f(G_2/G_1)$ obtained in our experiments for nitrogen and para hydrogen, respectively, under nonadiabatic and adiabatic cooling conditions are shown by points in the graphs. In the general case the magni-

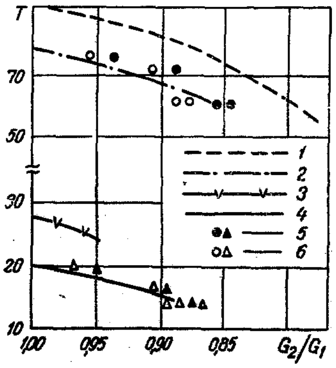

Fig. 3

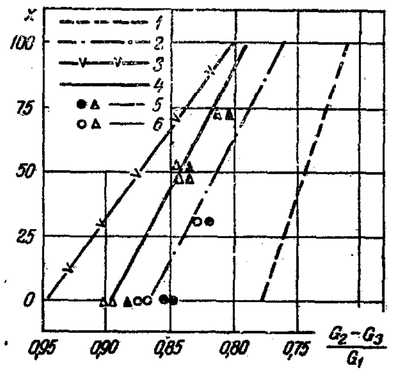

Fig. 4

Fig. 3. Dependence of the cryogenic fluid temperature on the quantity G_2/G_1: 1) oxygen; 2) nitrogen; 3) neon; 4) para hydrogen; 5) experimental points for nitrogen and para hydrogen, respectively (nonadiabatic conditions); 6) experimental points for nitrogen and hydrogen, respectively (adiabatic conditions), T, °K.

Fig. 4. Dependence of the percentage content of solid phase in a mixture on the quantity $(G_2-G_3)/G_1$: 1) oxygen; 2) nitrogen; 3) neon; 4) para hydrogen; 5) experimental points for nitrogen and para hydrogen, respectively (nonadiabatic conditons); 6) points referring to the nitrogen and para hydrogen solidification processes, reduced to adiabatic conditions. x, %.

tude of the heat influx to the cryogenic agent depends on the vessel shape, the quality of the insulation, etc.

The quantities of nitrogen and para hydrogen being evaporated because of heat influx were determined specially in the experiment.

By knowing the experimental values of G_2 and G_h, the quantity of vapor $G_{cool} = G_1(G_2-C_T)$, which was evacuated just in order to cool the fluid remaining in the vessel, can be determined. Therefore, the real cooling process can be reduced to adiabatic conditions for comparison with the results obtained by means of (9)-(12) by taking account of the experimental values of G_1, G_2 and G_h.

The discrepancies between the computed values by means of (9)-(12) and the experimental values reduced to adiabatic conditions do not exceed 5%.

Graphs of the dependence of the percentage content of solid phase x in the mixture on the quantity $(G_2-G_3)/G_1$ computed by means of (9)-(12) and (4) are shown in Fig. 4.

Experimental values of $x = \varphi[(G_2-G_3)]/G_1$, respectively, for nitrogen and para hydrogen under nonadiabatic and adiabatic conditions are shown by points in the graphs.

The discrepancies between the results computed by means of (9)-(12) and (4) and the experimental values $x = \varphi(G_2-G_3)$ reduced to adiabatic conditions also do not exceed 5%.

The kinetics of the process of solid phase formation was also studied during the experiments and the optimal rates of cryogenic fluid vapor evacuation were determined.

The experiments qualitatively confirmed the distinct picture of the solid-phase formation process for hydrogen and nitrogen. Thus, if a finely dispersed mixture of liquid and solid phase was obtained successfully for nitrogen by using continuous vapor evacuation, then a similar mixture could be obtained for hydrogen just by using agitation or pulsating evacuation.

Under adiabatic conditions, the rate of vapor pressure reduction in the vessel is proportional to the weight evacuation rate from a unit evaporation surface. Under real conditions this proportionality is spoiled because of the heat influx to the cryogenic fluid. Since the fluid temperature depends only on the pressure, it is expedient to check its cooling in the general case by means of the rate of pressure reduction rather than by means of the weight evacuation rate [2, 8], determined by the quantity of vapor evacuated from 1 m^2 of the evaporation surface per second.

The quantity of vapor evacuated from 1 m^2 in 1 sec depends on the magnitude of the heat influx to the cryogenic agent under real conditions. The higher the heat influx, the greater the optimal evacuation rate.

Hence, the optimal evacuation rates recommended in [2, 8] are not suitable for vessels having other heat influxes.

The rate of pressure reduction uniquely determines the behavior of the process; by maintaining its optimal value we assure the most favorable cooling mode. The quantity of vapor evacuated from unit surface during the same time will hence naturally be distinct for different vessels.

Experiments on the cooling of a cryogenic fluid in vessels of distinct volume and shape permitted the establishment of optimal pressure reduction rates for the cooling of para hydrogen and nitrogen. They are 0.3-1 mm Hg/sec. for para hydrogen and 0.2-0.5 mm Hg/sec. for nitrogen.

NOTATION

G, G_1, G_2, running value of the cryogenic fluid weight, its initial and final weights, respectively; G_3, weight of vapor removed from the system in obtaining x solid phase at the triple point temperature; p_{oc}, p_{tp}, pressure of the ambient medium and at the triple point, respectively; c_s, specific heat of the fluid; r, r_{tp}, r_f, running value of the heat of vapor formation in the range between the normal boiling point and the triple point, heat of vapor formation at the triple point, heat of fusion, respectively; quantity of heat influx; T, running value of the temperature; dT, change in fluid temperature during evaporation of dG vapor; W_H, electric-heater power; τ_f melting time.

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