

ANALYSIS OF THE MAIN REGULARITIES OF OBTAINING FROZEN GRANULES OF PRODUCTS AS APPLIED TO SUBLIMATION DRYING

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Results of theoretical and experimental investigation of the process of continuous production of frozen granules of liquid products by introducing them through a porous element into a vacuum chamber for subsequent sublimation drying of the frozen granules have been presented.

At the current stage of development of science and technology, one of the central problems is the improvement of production in any possible way and the development of new methods of treatment of materials and products which guarantee high quality and economy. This is particularly true of the food and pharmaceutical industries.

It is universally known that drying from the frozen state in vacuum (sublimation drying), in which changes in products are minimum and the products recover their properties after soaking almost completely, is the most efficient and progressive method of conservation of moist products. Large companies dealing with sublimation drying of valuable products have been established in many foreign countries (USA, England, France, Canada, Japan, etc.), the technology of sublimation drying has been developed, and the manufacture of industrial equipment has been arranged. Nonetheless, sublimation drying still remains an energy-intensive and expensive process, and traditional ways of enhancement have been exhausted, in practice. To improve its profitability one should pass to a continuous manufacture of dry product, including freezing, grinding (granulation), and drying from the frozen state.

At present, organization of the process of continuous sublimation drying of liquid products involves their preliminary freezing and granulation by different refrigerators installed inside or outside the sublimator and subsequent grinding of frozen sheets to granules and introduction of them into the sublimator. However, implementation of this technique leads to a significant increase in the energy consumption and degradation of the quality of the dried product because of the polydispersity of the granules obtained. "Self-freezing" due to the intense evaporation of the products themselves in the course of continuous introduction into the vacuum chamber, where the pressure is lower than at the triple point of the introduced product, is the most energy-profitable method of obtaining frozen granules of liquid products.

There have been attempts at introducing liquid products in the form of a flow or separate droplets into the vacuum chamber through heated nozzles of different design [1–4]. But in this case, due to the superheating of the liquid introduced at exit from the nozzle and entry to the vacuum chamber, we have boiling-up, spraying, "instantaneous" freezing, and, as a result, blocking of the nozzles with the frozen product. None of these devices have found application in sublimation technology.

A technique of continuous introduction of liquid products into the vacuum chamber for obtaining frozen granules (RF Patent No. 2022631) and a device for its implementation (RF Patent No. 2017052) have been developed and tested at the A. V. Luikov Heat and Mass Transfer Institute. This technique is based on the fragmentation of a liquid-product flow into droplets distributed in the horizontal plane by introducing it into the vacuum chamber through a flat porous element which consists of a set of numerous capillary nozzles and is installed in the upper part of the sublimator. Throughout the process of granulation, the difference between the temperature of storage of the solution on the upper plane of the element and the temperature of its freezing on the lower plane is created and maintained over the thick-

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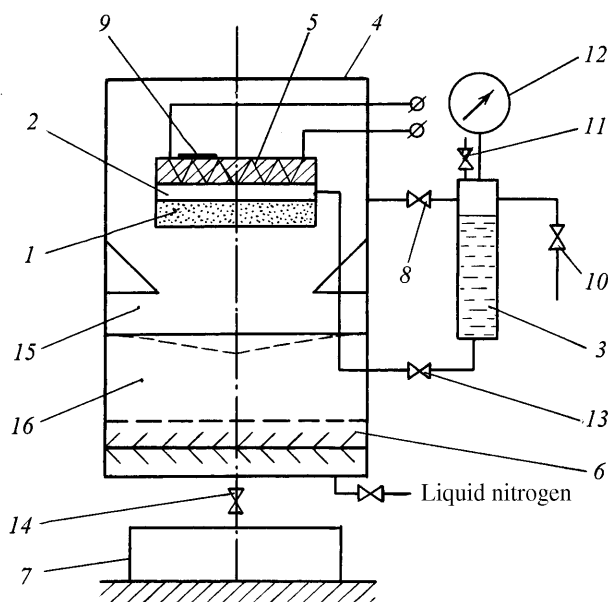


Fig. 1. Schematic of the experimental bench for introduction of a liquid into vacuum: 1) porous element (plate); 2) vessel of liquid supply; 3) tank for storage of the liquid; 4) vacuum chamber; 5) electric heater; 6) condenser; 7) vacuum pump; 8, 10, 11, 13, and 14) stop cocks; 9) heat-flux converter; 12) vacuum gauge; 15) accumulator of frozen granules; 16) sublimer.

ness of the porous element in the direction of product supply. For the valuable properties of the product to be preserved, this temperature difference must not exceed 20° .

Thus, the supply of energy to the porous element is organized so that the liquid product is gradually cooled as it moves over the thickness of the porous element and the freezing of droplets itself occurs in the vacuum chamber with intense evaporation of moisture during the motion of the droplets from the porous element to the sublimer. The porous element is connected to the vibrator to speed up the detachment of the cooled droplets. Thus, blocking of the nozzles with the frozen product is excluded.

To implement such a process one must preliminarily determine the temperature of freezing of the pumped liquid product T_{fr} .

The suggested technique of obtaining frozen granules in vacuum which has been tested in the sublimation chamber includes four simultaneous and continuous processes: introduction of liquid products into the sublimation chamber, their fragmentation into monodisperse granules, freezing without freezers and grinders, and sublimation dehydration.

This granulator can find wide application not only in drying of valuable thermolabile products but can also be used as stationary and portable ice-generators in cooled containers for transportation of food, pharmaceuticals, and other products to large distances.

In the engineering solution under consideration, two successive temperature zones of cooling are formed to obtain frozen granules due to the self-evaporation of the liquid introduced into vacuum. In the first zone, cooling from T_{st} to T_{fr} is carried out in pumping the liquid through the porous element (plate) on which a temperature gradient ∇T is maintained in the direction of solution supply such that the temperature reaches T_{fr} only at the site of liquid outflow into vacuum. The temperature gradient is created due to the supply of heat Q at the inlet to the porous element and to the natural cooling effect caused by the intense evaporation in vacuum of the introduced liquid itself at the outlet from the porous element (Fig. 1).

Liquid droplets formed in outflow from the pores "bounce" off the surface of the porous element under the effect of the total head ΔP and, moving down toward the sublimer where their drying is carried out, they are cooled (due to continuing evaporation) from T_{fr} to the evaporation temperature T_{ev} corresponding to the pressure of a saturated vapor $P_{s,v}$ in the vacuum chamber (second zone). Sublimation drying is usually performed at a pressure of 133–

13 Pa in the chamber. It is evident that the partial pressure of the vapor in the chamber cannot exceed this pressure; therefore, at worst, in the second zone granules are cooled to a temperature of 254–234 K and at this temperature they are supplied to the sublimator for further drying.

In designing the granulator, one must, first of all, study the processes of heat and mass transfer which occur in the first zone of cooling in pumping of the working liquid through a porous element of thickness δ (Fig. 1).

When the rate of heat supply to the surface of the porous element, which is in contact with the liquid, and the rate of heat removal due to the evaporation of the liquid at exit from it are equal, heat equilibrium is established and the temperature of the porous-element surface facing the vacuum remains constant throughout the period of granulation. Thus, the process in question can be considered to be stationary.

Since, according to the conditions of the formulated problem, the temperature difference over the thickness of the porous plate does not exceed 20° , we can assume that the physical parameters of the liquid and the rigid skeleton do not depend on temperature, i.e., do not change along the coordinate y . The temperature field inside the plate can be considered to be one-dimensional, since the dimensions of the plate in the direction x , y are much larger than its thickness, and the temperatures of the rigid skeleton and the pumped liquid are the same at any point of the porous material. This holds for low rates of filtration which are determined by the inequality

$$\text{Re}_f = Gd/(F_{\text{liq}} \mu_{\text{liq}}) \leq 1 .$$

Then the equation for the region $0 \leq y \leq \delta$ (heat transfer in the porous plate) and the boundary conditions are written as [6]

$$\frac{d^2 T}{dy^2} - \xi_{\text{solid}} \frac{dT}{dy} = 0, \quad 0 \leq y \leq \delta, \quad (1)$$

where $\xi_{\text{solid}} = Gc_{p\text{liq}}/(F_{\text{liq}}\lambda_{\text{ef}})$. When

$$y = 0: \quad T = T_{\text{liq}}; \quad (2)$$

$$y = \delta: \quad -\lambda_{\text{ef}} \frac{dT}{dy} + \alpha_c (T_{\text{med}} - T_\delta) = \frac{G_{\text{ev}}}{F_{\text{liq, ev}}} L. \quad (3)$$

Here G_{ev} is a portion of the pumped liquid which evaporates per unit time upon its introduction into vacuum. Since the liquid evaporates only at the outlet from the pores of the porous element, to determine the evaporation rate we relate G_{ev} to the evaporation surface. For a metallic porous element we can take with sufficient accuracy that $\lambda_{\text{ef}} \approx \lambda_{\text{solid}}(1 - \Pi)$ [5].

When the pressure of the medium is less than 133 Pa the convective flux is absent, in practice, and the temperature difference at the walls of the vacuum chamber and the granulator is small (less than 20°); therefore, in the boundary condition at $y = \delta$ we can neglect the second term as compared to others. Then, solution of Eq. (1) satisfying boundary conditions (2) and (3) has the form

$$T = T_{\text{liq}} + \frac{G_{\text{ev}} L (1 - \exp(\xi_{\text{solid}} y))}{Gc_{p\text{liq}} \exp(\xi_{\text{solid}} \delta)}. \quad (4)$$

We write the equation for the region of the incoming liquid in a similar way:

$$\frac{d^2 T'}{dy^2} - \xi_{\text{liq}} \frac{dT'}{dy} = 0, \quad -\infty \leq y \leq 0, \quad (5)$$

where $\xi_{\text{liq}} = Gc_{p\text{liq}}/(F_{\text{liq}}\lambda_{\text{liq}})$.

At the liquid–porous plate boundary, as has been indicated above, there is a heat source with density $q_h = Q/F_{\text{liq}}$. We write the boundary conditions for this region in the form

$$y = -\infty : T' = T'_{\text{liq}}; \quad y = 0 : -\lambda_{\text{liq}} \frac{dT'}{dy} + q_h = -\lambda_{\text{ef}} \frac{dT}{dy}. \quad (6)$$

Here T'_{liq} is the liquid temperature at the inlet to the granulator.

The solution of (5) satisfying boundary conditions (6) has the form

$$T' = T'_{\text{liq}} + \left[\frac{q_h F_{\text{liq}}}{Gc_{p\text{liq}}} - \frac{G_{\text{ev}} L}{Gc_{p\text{liq}} \exp(\xi_{\text{solid}} \delta)} \right] \exp(\xi_{\text{liq}} y). \quad (7)$$

We eliminate the unknown temperature T_{liq} from (4), relying on the fact that

$$T'_{y=0} = T_{\text{liq}} = T'_{\text{liq}} + \frac{q_h F_{\text{liq}}}{Gc_{p\text{liq}}} - \frac{G_{\text{ev}} L}{Gc_{p\text{liq}} \exp(\xi_{\text{solid}} \delta)}. \quad (8)$$

Having substituted (8) into Eq. (4), we finally obtain

$$T = T'_{\text{liq}} + \frac{q_h F_{\text{liq}}}{Gc_{p\text{liq}}} - \frac{G_{\text{ev}} L}{Gc_{p\text{liq}}} \exp[\xi_{\text{solid}}(y - \delta)]. \quad (9)$$

Equation (9) involves the temperature of the pumped liquid at the inlet to the granulator, which can be measured with high accuracy. The power of the exponent includes the criterion of cooling of porous bodies

$$k_p = \xi_{\text{solid}} \delta = \frac{Gc_{p\text{liq}} \delta}{F_{\text{liq}} \lambda_{\text{ef}}},$$

which characterizes the ratio of the quantity of heat transferred by the convective motion of the liquid inside the porous plate (filtration) to the quantity of heat transferred by heat conduction at the same temperature head. As k_p decreases, the temperature distribution approaches the linear one.

The amount of the liquid G_{ev} evaporated in unit time upon introduction into vacuum can be expressed in terms of the rate of evaporation in vacuum determined by the Hertz–Knudsen formula [7]:

$$I_m = \alpha \sqrt{\frac{M}{2\pi RT_\delta}} (P'_{\text{s.v}} - P'_v) = \frac{G_{\text{ev}}}{F_{\text{liq.ev}}}, \quad (10)$$

where T_δ is the temperature at the outlet from the pores of the porous plate, which is equal to T_{fr} according to the conditions of the problem and P'_v is the partial pressure of the vapor in the granulator near the surface of the porous plate.

As is seen from Eq. (10), the rate of evaporation in vacuum is limited by the velocity of removal of the vapor from the surface of the porous element due to diffusion transfer or when $P < 133$ Pa due to effusion transfer rather than by the value of the heat supply. Consequently, the evaporation rate depends to a greater extent on the value of the partial pressure of the vapor in the chamber P'_v , which is controlled by the mechanical removal of the vapor with the aid of a vacuum pump or a condenser cooled to negative temperatures.

In calculations, we take the evaporation coefficient α to be unity, as obtained under dynamic conditions [7].

On the basis of the conducted experimental study and analysis of material-heat balance we have found that 15% or more of the pumped liquid evaporates upon its introduction into vacuum. Consequently, to calculate the evaporation surface we can use experimental data on determination of the amount of liquid that evaporated upon introduction. Then we assume that an average of 20% of the liquid pumped through the porous element is evaporated, i.e., $G_{\text{ev}} \approx 0.2G$. Then, from (10) we find the value of the evaporation surface:

$$F_{\text{liq.ev}} = 0.2G \left[\alpha \sqrt{\frac{M}{2\pi RT_{\text{fr}}}} (P_{\text{s.v}} - P'_{\text{v}}) \right]^{-1}. \quad (11)$$

We calculate $F_{\text{liq.ev}}$ for the operating mock-up of the granulator used in the experiments.

The porous element is a flat 0.05-m-diam. and 0.0065-m-thick disk on which 26 cylindrical channels (pores) with a diameter of 0.0011 m and a 0.0008-m-diam. inner cylindrical insert made of D16T aluminum alloy are distributed uniformly. The amount of the liquid pumped varied from 1.4 to 5.2 kg/h.

Visual observations have shown that the optimum rate of granulation in this granulator, provided that the temperature of the liquid at the inlet to the porous element does not exceed the storage temperature of the liquid (283 K), is 4–4.5 kg/h.

From formula (11) we calculated the value of the evaporation surface $F_{\text{liq.ev}}$ at which the removal of heat due to evaporation in vacuum is sufficient for the set temperature gradient to be established over the thickness of the porous element and for the temperature at the outlet from the porous element to be equal to the temperature of the pumped liquid T_{fr} .

The working pressure in the chamber is taken to be 107 Pa. In granulation, it may be considered to be optimum for most liquid products [8].

Under these conditions, the values of the evaporation surface for the minimum ($G = 0.4 \cdot 10^{-3}$ kg/sec), maximum ($G = 1.4 \cdot 10^{-3}$ kg/sec), and optimum ($G = 1.25 \cdot 10^{-3}$ kg/sec) flow rates of the pumped liquid are, respectively, $F_{\text{liq.ev}}^{\text{min}} = 1.41 \cdot 10^{-4}$ m², $F_{\text{liq.ev}}^{\text{max}} = 4.92 \cdot 10^{-4}$ m², and $F_{\text{liq.ev}}^{\text{opt}} = 4.41 \cdot 10^{-4}$ m² and the values of the evaporation surface for each cylindrical channel ($F_{\text{liq.ev}}/26$) are $F_{\text{liq.p}}^{\text{min}} = 0.54 \cdot 10^{-5}$ m², $F_{\text{liq.p}}^{\text{max}} = 1.89 \cdot 10^{-5}$ m², and $F_{\text{liq.p}}^{\text{opt}} = 1.69 \cdot 10^{-5}$ m².

At the outlet from the pores at the instant when the cylindrical inserts were in the upper position, near the pores there formed droplets cooled to the freezing temperature and connected to the pore walls; as the insert went to the lower position the droplets, due to a sharp push, bounced off the outer surface of the porous element, thus making room for the formation of new droplets. Assuming these droplets to be spherical, we approximately determine their diameter d_d for different flow rates of the pumped product, $d_d = 2r_d = \sqrt{F_{\text{liq.p}}/\pi}$, and obtain $d_d^{\text{min}} = 1.3 \cdot 10^{-3}$ m, $d_d^{\text{max}} = 2.45 \cdot 10^{-3}$ m, and $d_d^{\text{opt}} = 2.32 \cdot 10^{-3}$ m. It is seen from the calculations that at a low flow rate of the pumped product, when it is introduced into the vacuum chamber virtually due to the capillary potential of the porous element, the droplet diameter is approximately equal to the diameter of the cylindrical channel, and the droplet size increases with the flow rate and is more than twice as large as the diameter of the cylindrical channel for the chosen optimum flow rate.

Direct measurements of the granules obtained have shown that they have a distorted shape, cubic rather than spherical, and, depending on the flow rate of the pumped liquid, their size is within 2–3.5 mm.

Thus, comparison of the results presented indicates that the calculated dimensions of granules are in agreement with those obtained experimentally.

Clearly, it is precisely the formation of droplets at the outlet from the pores and their periodic detachment from the surface of the porous element that limit the velocity of introduction of the product into vacuum; therefore, the flow rate of the pumped product entering into the coefficient ξ_{solid} (formula (1)) should be related to the determined surface of evaporation rather than to the surface of open pores (cross-sectional surface of the cylindrical channel). We also relate the supplied heat flux Q (in determining its density q_h) to the determined evaporation surface, since heat removal occurs only due to evaporation from the surface $F_{\text{liq.ev}}$.

We estimate the limiting heat flux which must be supplied to the surface of the porous element in order to maintain the required temperature gradient over its thickness. Having determined from (9) the temperature of the introduced product at the inlet to the porous element at $y = 0$ and having taken in calculation that T_{liq} must not exceed T'_{liq} (which must be maintained equal to the optimum temperature of product storage ($T_{\text{liq}} = T_{\text{st}}$)), we obtain

$$Q = q_s F_{\text{liq.ev}} = \frac{G_{\text{ev}} L}{\exp(\xi_{\text{solid}} \delta)}, \quad (12)$$

or having substituted $G_{\text{ev}} = I_m F_{\text{liq.ev}}$ and $\xi_{\text{solid}} \delta = \frac{G c_{\text{pliq}} \delta}{F_{\text{liq.ev}} \lambda_{\text{ef}}}$, we have

$$q_s = \frac{I_m L}{\exp \left[\frac{G c_{p\text{liq}} \delta}{F_{\text{liq.ev}} \lambda_{\text{ef}}} \right]}. \quad (13)$$

The same equation is also obtained in determination of the heat flux from the Fourier equation at $y = 0$.

It is seen from formula (13) that the heat flux depends on the rate of product evaporation in vacuum, the flow rate of the pumped liquid, and the value of the evaporation surface and also on the thickness and thermal conductivity of the porous element.

The last two parameters are specified by the design of the granulator and the choice of material for its manufacture. It is evident that in order to maintain the set temperature gradient, at which $T_{\text{liq}} = T_{\text{st}}$ at the inlet to the porous element and $T = T_{\text{fr}}$ at the outlet from, with decrease in the thermal conductivity of the porous element, it is necessary to decrease its thickness or the supplied heat flux. When λ_{ef} is constant, it follows from (13) that the larger the thickness δ , the smaller must be the heat flux supplied for maintaining the specified boundary conditions. It has been found from the analysis of material-heat balance and the experiments with metal-ceramic porous elements made of titanium ($\lambda_{\text{ef}} = 5 \text{ W/(m-deg)}$) that the granulator operates, though unstably, even at a thickness of the element of 1 mm.

On the basis of the results of the experimental investigation and estimating calculations we can draw the conclusion that in designing the granulator, one must choose the minimum admissible thickness of the porous element, proceeding from the conditions of stability of the process of granulation and of strength and reliability of the structure in operation in vacuum and at a pressure difference to 1 atm over the thickness of the porous element.

In the operating mock-up of the granulator used in the experiments, $\delta = 6.5 \cdot 10^{-3} \text{ m}$ and $\lambda_{\text{ef}} = 120 \text{ W/(m-deg)}$.

We calculate Q for the chosen optimum flow rate $G = 1.25 \cdot 10^{-3} \text{ kg/sec}$ and the limiting value $F_{\text{liq.ev}} = 4.41 \cdot 10^{-4} \text{ m}^2$, $P = P_v = 107 \text{ Pa}$. With account for (12) and (13) we have $Q \leq 328 \text{ W}$ and at $\delta = 5 \cdot 10^{-3} \text{ m}$ we obtain $Q \leq 380.6 \text{ W}$. Here we use the inequality sign since in the process of granulation $F_{\text{liq.ev}}$ can have an even higher value.

We determine what pressure head ΔP must be maintained in the granulator for granulation to be stable and continuous.

According to the Darcy law, the flow rate of the liquid through a porous element of thickness δ must be 5-fold higher than the evaporation rate, since during the granulation more than 20% of the product is evaporated upon its introduction and self-evaporation in vacuum:

$$\chi \frac{\Delta P}{\delta} \geq 5I_m. \quad (14)$$

Here χ is a coefficient that is dependent on the properties of the porous body and the type of liquid (determined experimentally) and ΔP is the pressure difference on the porous element ($\Delta P = P_2 - P$, where P_2 is the pressure in the cavity (Fig. 1)). Since the liquid is introduced into vacuum, we can take $\Delta P = P_2$, allowing for the smallness of the pressure above the granulator surface. Then condition (14) is written in the form

$$P_2 \geq \frac{5I_m \delta}{\chi} = \frac{5}{\chi} \frac{G_{\text{ev}}}{F_{\text{liq.ev}}} \delta. \quad (15)$$

The main relations of the stationary process of obtaining frozen granules of liquid products by introduction of them into vacuum and self-freezing are given in (9)–(15). These relations hold only with account for the assumptions taken in the formulation of the problem; however, they can successfully be used for estimation of the most important parameters in designing granulators.

In the dense flow of the formed granules from the input device (granulator) to the sublimator, where they are dried, the mutual position of these granules exerts a substantial effect on the evaporation rate, and consequently, the final temperature of the granules which entered the sublimator. This is caused by the fact that, due to the self-evaporation of droplets, vapor envelopes are formed near the droplets introduced into the vacuum chamber; at small distances

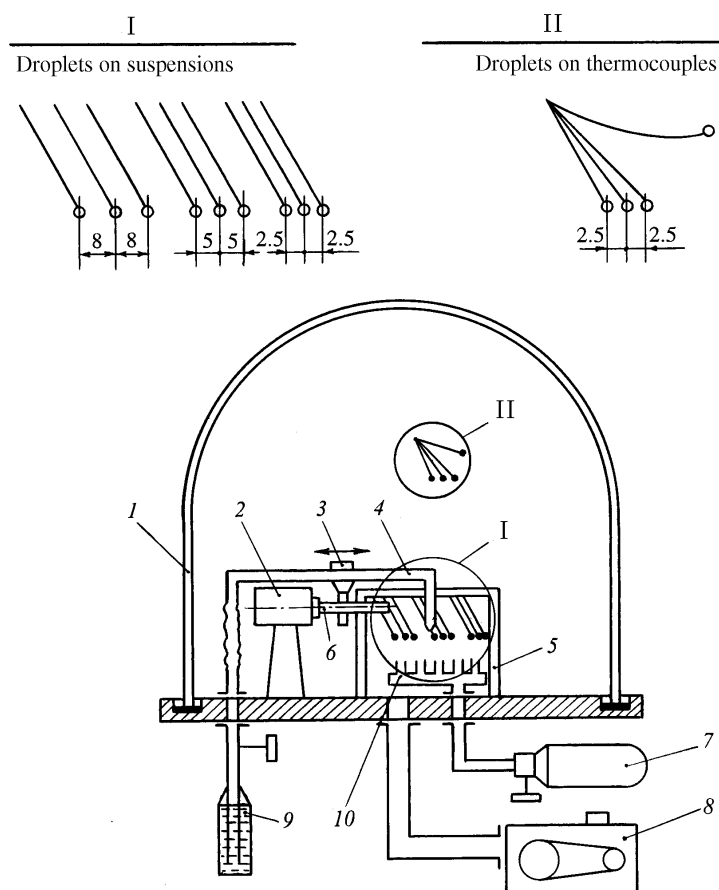


Fig. 2. Schematic of the experimental bench for determining temperatures of supercooling and crystallization of associations of droplets in rarefied medium: 1) vacuum chamber; 2) reversible engine; 3) joint; 4) capillary; 5) holder for suspensions; 6) shaft; 7) tank with ammonia; 8) vacuum pump; 9) vessel for the studied liquid; 10) comb of capillaries.

between the nozzles these envelopes are overlapped, which results in an increase in the density of the vapor flow between the droplets and in their defreezing and coalescence in motion to the sublimator. In this case, the quality of the dried product is degraded due to the distorted monodispersity of the granules. Therefore, to implement reliable operation of the granulator one should study the distinctive features and regularities of group evaporation of droplets in vacuum (effect of the mutual position and distance between droplets, the droplet size, the pressure of the medium, the conditions of power supply, and other factors on the intensity of heat and mass transfer with changing thermodynamic parameters of the medium during the self-freezing of liquid products in vacuum) and develop a technique for determining the temperatures of freezing (crystallization) of different solutions, the knowledge of which is necessary for implementation of the process of granulation in vacuum in designing a granulator.

In the adopted technique of investigation of the self-freezing of an association of droplets due to intense evaporation in vacuum, the process of cooling of the droplets was considered in continuous delayed reduction of the pressure of the medium in the vacuum chamber during a certain period of time. Thus, the process of freezing was protracted, which made it possible to study the main regularities of it.

Experimental investigation was conducted in a vacuum chamber of volume 0.2 m^3 (Fig. 2, I). The pressure in the chamber was changed by the vacuum pump and the needle leak from atmospheric pressure to 1 Pa. Between the chamber and the pump, a liquid-nitrogen condenser is mounted. The partial pressure of the vapor of the evaporating liquid in the chamber equipped with measuring devices for monitoring the temperature and the total pressure of the medium was controlled by changing the condenser temperature. The partial vapor pressure was measured by a psychrometric microthermocouple.

In order to determine T_{fr} of liquid products in the vacuum chamber, we tested an association (a group) of three droplets suspended on the heads of Copper–Constantan microthermocouples (Fig. 2, II). The distance between the thermocouples was 2.5 mm. Here, at a distance of 30 mm from them, an isolated microthermocouple was installed, which did not get into the vapor cloud around the group of droplets under all the experimental conditions.

The technique adopted allowed comparison of heat and mass transfer processes in the group of droplets and in an isolated droplet under the same external conditions.

Three groups of suspensions (three suspensions in each) made of 40-mm-long quartz filaments with a round loop at the end (the diameter of the loop varied from 0.5 to 2 mm) were positioned inside the vacuum chamber (Fig. 2, I). The comblike suspensions were fastened to holder 5. The distance between the droplets was changed by moving the suspensions in the horizontal plane. After preliminary studies we chose the version with a distance between the centers of the droplets of 8, 5, and 2.5 mm.

Droplets of the liquid product under study were suspended on the suspensions using a device consisting of capillary 4, which was connected to the shaft 6 of reversible engine 2, and joint 3. The capillary communicated hydraulically with the vessel for the tested liquid 9 and the pneumatic system, which allowed control of the pressure in the vessel and supply of the liquid to the end of the capillary at the given velocity. The capillary moved above the combs with suspensions so that its end was above their loops. Droplets of the same size formed at the end of the capillary were suspended successively on all the suspensions; then the capillary was moved clear of the holder and liquid supply stopped.

Energy was supplied to the droplets by heat transfer from the suspensions and by radiation from the chamber walls whose temperature was changed within wide ranges by an electric heater mounted on them. It should be noted that the material of suspensions affects the absolute rate of the process of evaporation of a single droplet and the relative rate of evaporation in the group is virtually independent of the suspension material and is determined only by the position of the droplets relative to each other.

The operating pressure of the medium in the vacuum chamber was set, and changes in the parameters of the process were recorded for all the versions of the position of droplets under the same external conditions.

The size of the droplets in the groups in the vertical and horizontal planes and its change in the process of evaporation and sublimation was determined by videorecording and filming of the entire group of droplets in certain constant time intervals.

The processes studied were also visualized by the "smoke method." For this purpose, droplets of an aqueous solution of hydrochloric acid were suspended on the suspensions and an ammonia vapor was fed from cylinder 7 through the comb of capillaries 10 installed under the droplets to the chamber, which was evacuated to the assigned operating pressure.

Distilled water and natural juices (beet, carrot, cranberry, and apple) were used as tested liquids.

Droplets of liquid product were suspended on three thermocouples combined into a group and on a single isolated thermocouple (Fig. 2, II). The initial diameter of the droplet was 2 mm. Then the chamber was evacuated.

Figure 3 presents a typical graph of the changes in the temperature of beet-juice droplets with pressure P in the vacuum chamber.

As the pressure in the chamber decreased to $1.3 \cdot 10^4$ Pa (in this case, P'_v in the chamber approached a value of the pressure at the triple point of water equal to 610.5 Pa), the temperature of the droplets T_d of all the liquids studied decreased virtually linearly much below the temperature at the triple point of the liquid under study, i.e., the liquid was supercooled. Thus, for distilled water the temperature of supercooling $T_{supercool}$ of the isolated droplet reached 256.7 K and 259.9 K on retarded and fast, respectively, decreases in the pressure in the vacuum chamber (Nos. 3a and 1a in Table 1). Here, in all the cases considered, the temperature of the central droplets remained higher than that of the isolated droplet and the last one of the three droplets (cf. Nos. 1–3). This regularity was preserved for all the liquid products under study.

In self-freezing of juices, $T_{supercool}$ depends on the physicochemical properties of the product, the geometric dimensions of a droplet, the concentration of dry substances in the product, the pressure in the chamber and the rate of its decrease, and the conditions of energy supply. The higher the viscosity of the juice and the content of dry substances in it, the lower the supercooling (cf. $T_{supercool}$ of water and cranberry or carrot juices).

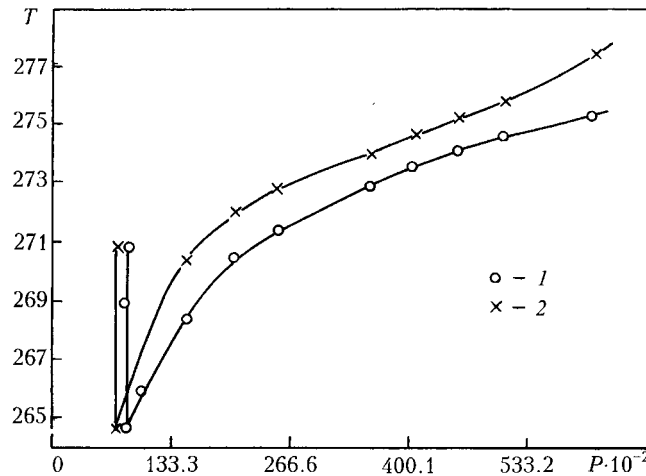


Fig. 3. Change in the temperature of the association of droplets with decrease in the pressure of the medium and determination of the supercooling and crystallization temperatures of the droplets: 1) isolated droplet; 2) central droplet of the three droplets.

It is seen from Fig. 3 that the phase transformation of liquid droplets to ice begins in removal of heat by evaporation at the instant when the supercooling state of the droplets is disturbed. The droplet temperature increases in a jumpwise manner to the temperature of freezing. Instantly (in hundredth fractions of a second), a droplet freezes completely and its temperature begins to decrease, tending to an equilibrium temperature which corresponds to the partial pressure of the vapor in the medium. Then the temperature of the frozen granule obtained slowly increases until it is completely dried.

The temperature at which the formation of ice begins is called the temperature of crystallization of liquid solutions T_{cr} (see Table 1) and for each product it depends on the nature and properties of the product.

From the study conducted, we have found that more than 20% of the liquid evaporates from the product during the self-freezing, which causes a decrease in the freezing temperature. For water this effect was not observed. In all the experiments, the freezing point of distilled water was $273.15 \pm 0.1K$.

The obtained values of the freezing temperature of some liquid products are necessary for developing the technology and designing a device for introduction of liquids into vacuum.

As is seen from Table 1, the value of the supercooling and the temperature of droplet crystallization are affected by the mutual position of the droplets in the association.

To reveal the effect of the distance between the droplets in the association on the intensity of heat- and mass-exchange processes, we conducted simultaneous investigations in three groups of droplets fixed on one holder (Fig. 2, D). Similar droplets were suspended on all groups of the suspensions. As the pressure in the chamber was decreased, observations of all the droplets were carried out simultaneously using filming.

It has been found that in horizontal arrangement of droplets and energy supply by radiation from the chamber walls the effect of the distance between the droplets l and their mutual position on the evaporation rate increases with decrease in the pressure of the medium.

As the pressure in the chamber decreased to 6665 Pa ($P'_v \sim 933$ Pa), at $l = 2.5$ mm the last droplets evaporated about 1.4 times more quickly than the central one, at $l = 5$ mm they evaporated 1.14 times more quickly, and at $l = 8$ mm we observed no difference in the rates of evaporation of droplets in the group. With further decrease in pressure ($P'_v = 147$ Pa), in the group with $l = 8$ mm all the droplets froze simultaneously, and at $l = 5$ mm only the last droplets froze. When $P < 107$ Pa, the droplets on all suspensions froze. However, at this pressure the difference in the rate of evaporation of the central and last droplets becomes noticeable even in the group with $l = 8$ mm. A similar regularity holds for all the liquid products under study. This dependence becomes weaker with increase in the distance between the droplets.

TABLE 1. Determination of the Supercooling and Crystallization Temperatures of Water and Liquid Food Products

No.	Liquid	$T_{\text{supercool}}$, K	T_{cr} , K	P_b , Pa	P'_v , Pa	τ , sec	Droplets
1	Distilled water	259.9	273.15	1333	122	240	a
		262.6	273.15	1333	122	240	b
2	Same	258.2	273.15	1333	122	260	a
		261.0	273.15	1333	122	260	b
3	Same	257.9	273.15	1333	122	360	a
		256.7	273.15	1333	122	490	a
4	Carrot juice	264.9	271.1	3931	124	240	a
		265.8	271.1	3931	124	240	b
5	Juice of "Ranet" apples	258.8	267.6	2666	97	240	a
		260.0	270.3	2666	97	240	b
6	Juice of "Ananas" apples	264.2	269.9	3999	122	240	a
		264.5	270.4	3999	122	240	b
7	Juice of "Krasnye" apples	261.8	269.4	3500	122	240	a
		260.1	269.1	3500	122	240	b
8	Cranberry juice	266.4	271.5	3331	122	240	a
		268.1	272.5	3331	122	240	b
9	Beet juice	264.7	271.1	2666	153	240	a
		264.3	271.0	2666	148	240	b

Note: P_b and P'_v , total and partial pressures of the vapor in the chamber at the instant of crystallization-temperature measurement; τ , time from the switch-on of the pump to the crystallization of a droplet; a, isolated droplet; b, central droplet.

The results of the investigations allow us to draw the conclusion that the mutual effect of droplets in their evaporation is, first of all, determined by the physical properties of the liquid, the coefficients of diffusion, which are functions of the pressure of the medium and the temperature of the droplets, and the velocities of motion of the vapor and the surrounding medium.

Visual observations have shown that in evaporation of distilled water whose molecular weight is 1.5 times lower than the molecular weight of air filling the chamber, at the beginning of the process of evaporation the density of the vapor-gas medium near the droplet surface is lower than the density of the surrounding medium; therefore, here we observed ascending vapor flows. With further decrease in the pressure, the outer flow of the medium is rejected from the droplet surface. Near the droplet, there appears a layer with a sharply outlined "vapor envelope" interface whose thickness increases with decrease in the medium density.

Joining of the vapor envelopes of neighboring droplets causes a change in the temperature and rate of droplet evaporation. In this case, the effect of the distance between the droplets on the kinetics of the entire association is especially noticeable.

Similar regularities are observed for all the juices studied, since water is the main component of them.

Mass transfer in the direction perpendicular to the surface (transverse flow of a substance) causes changes in the hydrodynamic and thermal boundary layers, which leads to a decrease in the heat-transfer coefficient and thus a decrease in the rate of evaporation of the droplets in the association.

In the present experiments, when $P < 1.3 \cdot 10^4$ Pa, we did not observe vortex formations in rarefied-gas flow past evaporating droplets, which allowed consideration of the transverse and longitudinal flows as potential ones. Change in the size of a vapor envelope with the evaporation rate and the velocity of the longitudinal flow is confirmed by computations assuming the potential character of motion of the interacting flows.

In [9], the limiting streamlines in evaporation of spherical bodies in vacuum as functions of the ratio u/v were calculated on a computer by the method of superposition of longitudinal u and transverse v potential flows [10]. These

theoretical computations were used in estimating the limiting distances between the droplets in the association at which their mutual effect on the processes of heat and mass transfer is insignificant.

At the initial instant of droplet formation, the longitudinal flow of a vapor-gas mixture in the process under consideration is caused only by the operation of the vacuum pump. According to the estimates, $u \sim 1.9$ m/sec. For the characteristic parameters of the process of introduction ($P_b = 133$ Pa, mean temperature of the medium 293 K, droplet size $2r_d \sim 2 \cdot 10^{-3}$ m, and time of droplet formation 1.2 sec) we have

$$v_v = I_m / \rho_v = 22.7 \text{ m/sec}.$$

Then, at $\varepsilon = u/v_v = 0.084$, by solution of the integral equations [10] we find $\delta_{eq}/r_d = 3.9$. Consequently, for the given droplet size we have $\delta_{eq} = 4.9$ mm.

Thus, in designing the device for introducing liquid products into vacuum and obtaining frozen granules by the method of self-freezing, proceeding from the conditions of overall dimensions and reduction in the consumption of materials, one should take the distance between the centers of the nozzles in a porous element equal to 10 mm.

This conclusion was confirmed experimentally.

NOTATION

T , temperature, K; T'_{liq} , temperature of the liquid at the inlet to the granulator, K; Q , heat flux, W; q , specific heat flux, W/m²; λ , thermal conductivity, W/(m·K); L , heat of evaporation, J/kg; c_p , heat capacity, J/(kg·deg); I_m , evaporation rate, kg/(m²·sec); P , pressure, Pa; P'_v , partial pressure of the vapor, Pa; $P_{s,v}$, pressure of the saturated vapor, Pa; ρ , density, kg/m³; μ_{liq} , coefficient of dynamic viscosity, N·sec/m²; G , mass flow rate, kg/sec; α , coefficient of evaporation; δ , thickness of the porous element, m; Π , porosity, %; R , universal gas constant, J/(deg·kmole); F_{liq} , surface area of the pores at exit of the liquid from the granulator, m²; $F_{liq,e}$, evaporation surface, m²; M , molecular weight, kg/kmole; d , mean diameter of the pores of the porous element, m; Re_f , filtration Reynolds number; k_p , criterion of cooling of porous bodies; α_{conv} , coefficient of convective heat exchange, W/(m²·deg); x, y, z , coordinates; u , velocity of longitudinal gas flow, m/sec; v , velocity of the transverse vapor flow, m/sec; $2r_d$, droplet diameter, mm; τ , time, sec; l , distance between the centers of droplets, mm; δ_{eq} , distance between the limiting streamline and the surface of an evaporating droplet in potential flow in a rarefied gas, mm; $T_{supercool}$, temperature of droplet supercooling, deg; G/F_{liq} , liquid flow rate related to a unit surface of open cylindrical pores, kg/(m²·sec). Subscripts: v, vapor; d, droplet; fr, freezing; supercool, supercooling; cr, crystallization; med, medium; max, maximum; min, minimum; opt, optimum; ev, evaporation; st, storage; liq, liquid; eq, equilibrium; s.v, saturated vapor; solid, solid; ef, effective; f, filtration; h, heating; m, mass; δ , at the outlet from the porous element; p, pore; b, barometric; in, initial; c, convective.

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