THERMOPHYSICAL PRINCIPLES INVOLVED IN THE OPTIMIZATION OF THE BASIC PARAMETERS OF VACUUM-SUBLIMATION DRYING*

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Vacuum sublimation and condensation (desublimation) curves have been obtained with thermal flux probes. The results are used as a basis for optimizing the process parameters in vacuum-sublimation drying.

Feasibility studies concerning the wider use of sublimation drying have yielded quite favorable results, if one considers the forecast for the next few years in the development of fully automated high-tonnage [6, 8, 9], semicontinuous-duty [1, 2, 4, 5], and continuous-duty [3] apparatus.

The solution to this complex and difficult problem requires diverse thorough study, theoretical research, and engineering development.

Existing methods of automatically controlling [4, 5, 10] such a complex technological process as sublimation drying, which involves freezing and subsequent dehydration of the resulting solid phases as well as condensation (desublimation) of formed vapors, do not meet modern requirements. This is primarily due to the unavailability, at this time, of sufficiently simple means of continuous-duty automatic data acquisition with regard to desiccation and condensation (desublimation) in industrial sublimators. No method has been developed yet for reliably indicating the end of both the sublimation process and the condensation (desublimation) of vapor in a condenser.

Available weighing methods [4, 5] and electrical methods [10] based on the loss of mass, as well as known and already evaluated desiccation techniques applicable to some capillary-porous or colloidal products, are hardly adequate for the design and the operation of future high-tonnage sublimators.

Here the authors are proposing to use a thermal flux probe (TF probe) for the automatic control of the sublimation drying process; a probe which has been designed at the Institute of Engineering Thermophysics (Academy of Sciences of the Ukrainian SSR) [19] and which will make it feasible to let the complex desiccation process take place under optimum conditions of sublimation and desublimation.

Controlling the Technological Process of Contactive Sublimation. The authors experimented with the desiccation of hydroxidizing bacteria at the Moscow Power Institute. A specimen of the colloidal product was placed inside a cylindrical jar whose bottom constitutes a TF probe. This probe was heated electrically. During desiccation of the product inside, the loss of mass was measured on a model VLTK-500 balance. The amount of heat supplied to the product during sublimation was determined on the basis of the heater power and also by the TF probe reading at the same time. During spontaneous freezing under vacuum, the TF probe read the amount of crystallization heat released in the process.

In Fig. 1 we compare the desiccation curve (Fig. 1a) with the TF probe readings (the sublimation curve) (Fig. 1b) taken with a recording instrument. In this case the TF probe read the amount of heat

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*This engineering research was done as an extension of the studies in [13, 14].

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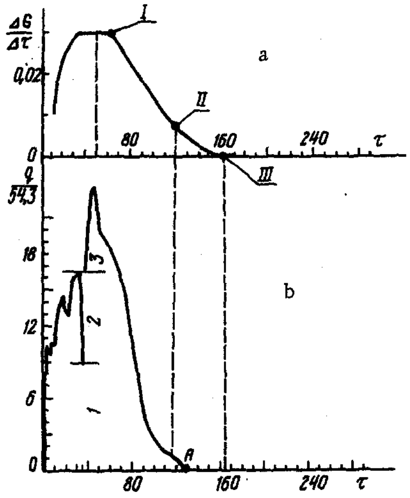

Fig. 1

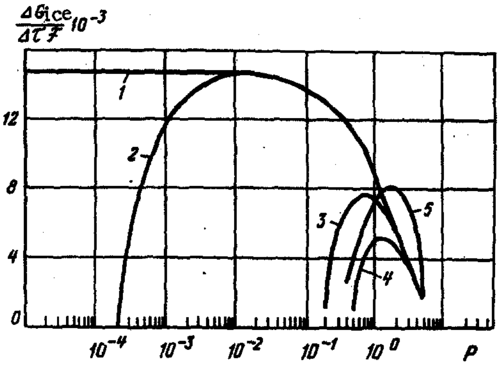

Fig. 2

Fig. 1. Desiccation curve (a) and sublimation curve (b) for the biomass: I, II, III) indicate the end of the first, the second, and the third stage of desiccation, respectively; change of the vacuum level in the chamber 0.1–0.5–1.0 mm Hg: 1) 0.1 mm Hg; 2) 0.5 mm Hg; 3) 1.0 mm Hg; O denotes start of desiccation; A denotes end of desiccation; time τ (min); ΔG/Δτ (g/min); q/54.3 W/m^2.

Fig. 2. Condensation (desublimation) rate of water vapor in vacuum, at various temperatures of the active surface, as a function of the vacuum level [17]: 1) $t_C = 196°C$; 2) $t_C = -74°C$; 3) $t_C = -34°C$; 4) $t_C = -25°C$; 5) test curve obtained by these authors ($t_C = -25.1°C$); pressure P (mm Hg); $\Delta G_{ice}/\Delta\tau F$ (g/h · m^2).

removed from the calorimeter as a result of sublimation of the frozen vapor in the product. These readings not only duplicated the basic stages of the sublimation process according to the desiccation curve almost exactly, but also yielded information about pressure fluctuations likely to occur in the vacuum chamber. According to Fig. 1, a change in the sublimator vacuum from 1.0 to 0.1 mm Hg ($\tau = 35$ min) appreciably decreased the sublimation rate, because of the higher thermal resistance of the material–calorimeter interface (higher degree of rarefaction in the voids within the contact zone). This short process occurring in the contact zone could not be picked up by the weighing method. These findings are especially important for explaining how various factors, such as air "injection" and noncondensing gases (helium, nitrogen, etc.), contribute to a higher rate of drying by contactive sublimation, insofar as they effect the contact zone directly [7]. On the basis of such TF probe data, then, it is possible to establish when the process of sublimation drying has come to an end. The completion of this process is also indicated by a null signal at the TF probe output.

An experiment with and an analysis of the contactive sublimation mechanism have shown [10, 11, 15], however, that an attenuation of the TF probe signal to zero corresponds merely to a complete deicing of the material (not the removal of adsorptively bonded moisture) and, in the final analysis, is determined by the predesiccation of its layer in contact with the heater surface (point A in Fig. 1).

The removal of adsorbed moisture (during the third stage of sublimation drying) can be ascertained on the basis of the total given drying time, from an evaluation of desiccation curves, and on the basis of a biological product analysis (Fig. 1). Tests have shown that the trend of a sublimation curve (Fig. 1), which determines the process of contactive sublimation drying, depends on the properties of the product, on changes in the vacuum level, on the profile of the contact surface (flat, corrugated, or other), on the thermal flux density, etc.

Controlling the Technological Process of Water Vapor Vacuum-Desublimation (Condensation). As has been shown in [13, 14], a TF probe placed on the cold surface during condensation (desublimation) reads the amount of heat released exactly during the phase transition.

The rate of that process depends on the properties of the raw product (in relation to moisture and uncondensable gases released from it), on the temperature of the condenser walls, on the vacuum (total pressure), on the surface condition, on the vapor rate (vapor stream velocity), etc.

The vacuum (total pressure) level in the sublimator and in the condenser is in many cases dictated by the necessity to conserve the product in the frozen state throughout the desiccation process. At every specified vacuum level the maximum process rate depends largely on the specified temperature of the desublimation surface (critical temperature) T_{cr}. In Fig. 2 we show our curve as well as curves taken from [17]. It is quite evident here that, in order to lower the critical temperature for pure vapor (twice distilled), it is necessary to deepen the vacuum in the condenser.

During the sublimation of various products, one most often deals not only with pure water vapor but also with various admixtures in the form of uncondensable and aromatic gases. Moreover, the composition and the concentration of these admixtures vary continuously throughout the desiccation process. This, in turn, may shift the maximum rate of the desublimation process toward a residual pressure which is higher or lower than stipulated.

It is well known [13, 14] that an increase in the area, per unit time, under a desublimation curve plotted with the aid of a TF probe represents an increase in the amount of released desublimation heat and this, in turn, indicates an increase in the icing rate at the active surface. As the ice formation rate increases, more heat is released and passes through the TF probe with the result that the absolute magnitude of the electrical output signal U_{out} for the regulator circuit becomes larger. In this case the regulation process consists in searching for the extremum (maximum) of the function

$$U_{\text{out}} = f\,[P_{\text{c}}, T_{\text{s}}, J, D_{\text{n}}, D_{\text{a}}, \delta(\tau)]\,,$$

(where U_{out} denotes the output signal from the TF probe, P_c denotes the total condenser pressure, T_s denotes the temperature at the condenser (desublimator) surface, J denotes the vapor current density in the condenser, D_n denotes the percentage of uncondensable gases, D_a denotes the percentage of aromatic components in the gas, and $\delta(\tau)$ denotes the thickness of the desublimated ice layer as a function of time) and, on this basis, finding the means of automatically maintaining the optimum process conditions. It must be taken into account, however, that regulating the desublimation process under vacuum becomes complicated because of the process instability, namely, by the fact that the process rate decreases almost to zero after the extremum has been reached. All this makes the design of the automatic regulation system more difficult and requires the development of an entirely special control circuit.

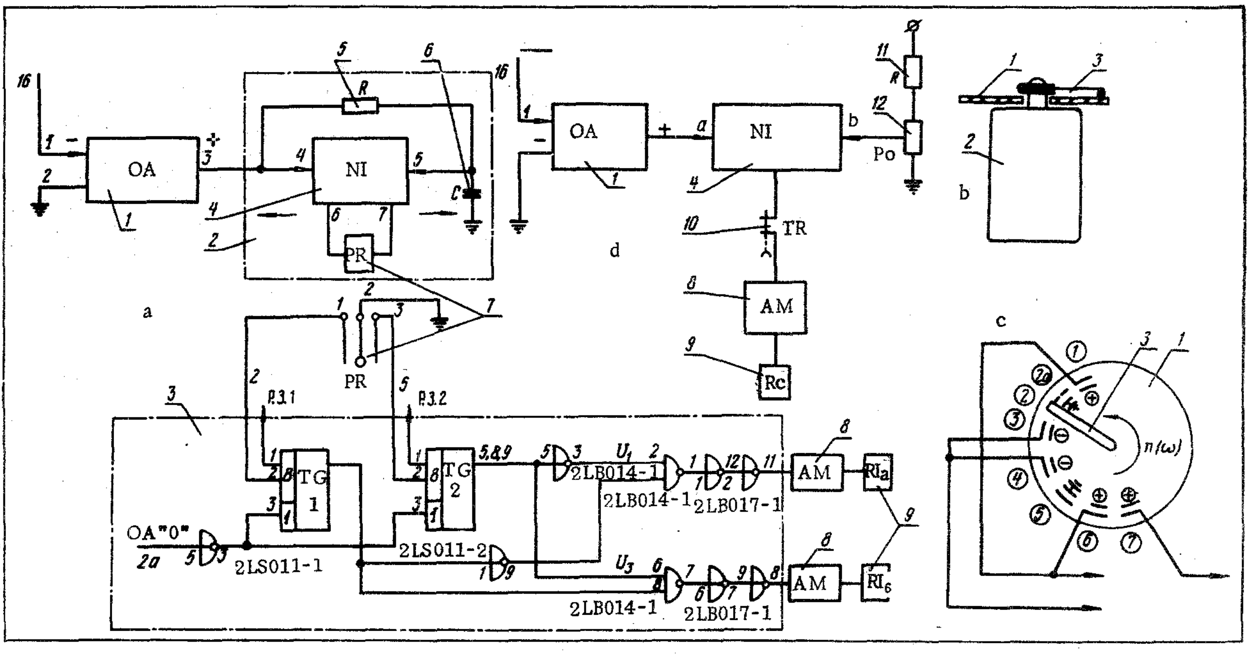

Fig. 3. (a) Schematic diagram of the automatic regulation system for processes with phase transitions: 1) operational amplifier OA; 2) comparator unit; 3) decoder; 4) null indicator NI; 5) resistor R; 6) capacitor C; 7) polarized relay PR; 8) amplifier AM; 9) relay RI. (b) Clocking transmitter: 1) contact plate; 2) electric motor; 3) contact arm. (c) Contact plate for clocking the regulatory signals: 1) contact plate; 3) contact arm. (d) Schematic diagram of the system for automatically deicing the active components in a vacuum condenser (desublimator): 1) operational amplifier; 4) null indicator; 8) amplifier; 9) relay; 10) time relay; 11) limiting reistor R; 12) potentiometer Po.

TABLE 1. Possible Modes of Actuating the Automatic Regulation System for Vacuum-Condensation (Desublimation) of Water Vapor

Row number	TG(trigger) 1	TG(trigger) 2	1 Signal reset	2 Signal reset	Preset
1	0	0	▬ ↓ ↑	↑ ▬	
2	0	1	▬ ↓ ↑	↑ ↓ ▬	
3	1	1	▬ ↓ ↑	↑ ↓ ▬	▬ ↓
4	1	0	▬ ↓ ↑	↑ ↓ ▬	

With all these considerations in mind, we have designed and built an automatic vacuum-desublimation regulator (Fig. 3) with integrated-circuit components and series 201 logic. While gas and vapor are forming in the sublimator, the proposed logic circuit searches for optimum P_c and T_s so as to bring the condensation rate to its maximum.

For clocking the time intervals between signals to the regulator we used a contact plate 1 (Fig. 3b) mounted on a model RD-2 electric motor 2, with a contact arm 3 on the rotor shaft. The angular velocity of the rotor was variable and a function of the voltage U_a applied to the motor: $\omega_m = f(U_a)$, the latter regulated through a slide-wire resistor and measured with a voltmeter.

On the contact plate we had spaced seven contact buttons which, in sequence, produced seven signals. The length and the frequency of these signals were functions of the motor speed ω_m and of the contact length $l_{c_1}-l_{c_7}$ (Fig. 3c). Contacts 2, 2*a*, and 5 could be made negligibly smaller than the others, since the trigger times of TG-1 and TG-2, as well as the stop "O" times, were shorter than 10 μsec. The lengths of the other contacts (makes and breaks) had been designed to match the inertia of the vacuum chamber and of the TF probe (the automatic regulation or AR probe). The transmitter in Fig. 3b, c periodically generated *a* and b signals to shift the values of the regulated parameters (during sublimation drying $(a_2) = (b_2) = \pm\Delta P$ and $(a_1) = (b_1) = \Delta T_s$). After such a perturbation by an *a* or b signal, moreover, the residual pressure and the temperature had to be returned to their initial levels.

A signal (from 0 to 80 mV) was transmitted from the TF probe (AR probe) 16 to the operational amplifier (OA) 1, as shown in Fig. 3a. The current gain of this operational amplifier was approximately 1000. Its maximum output voltage was 25 V.

From the OA output, the amplified voltage signal was transmitted to input 4 of the null indicator (NI) 4 and through resistor (R) 5 to input 5 of the null indicator (NI) and to capacitor (C) 6. The null indicator NI) was loaded with a model RP-5 polarized relay 7.

The initially steady pressure in the chamber $P_{ch} = P_i$ was perturbed with a signal $a = \Delta P$. As a result, $P_{ch} = P_i + \Delta P$ and the magnitude of signal U_{out} from the AR probe was stored in capacitor (C) 6, whereupon signal *a* was turned off and the system returned to its initial state $P_{ch} = P_i$.

The system was next perturbed with a signal $b = -\Delta P$. As a result, $P_{ch} = P_i - \Delta P$. Signal b was then turned off and the pressure in the vacuum chamber returned to its initial level $P_{ch} = P_i$. The temperature of the active condenser surface was varied in an analogous manner.

Depending on which voltage prevails, at input 6 or at input 7 of the null indicator 4, either contactors 12 or contactors 23 of the relay (PR) 7 close. Signals from these PR contactors are stored in the memories of triggers TG-1 and TG-2. They are then transmitted to the decoder consisting of a coincidence circuit which, through amplifiers AM and relays RI_a and RI_b (Fig. 3a), controls the motor driven valve of the water vapor evacuating system. When the extremum has been reached, the system can be operated in four possible different modes throughout the process (Table 1). Contactors 23 PR close when $U_4 < U_5$, contactors 12 PR close when $U_4 > U_5$.

Considering that the desublimation process in sublimators is concealed from the operator, there is always the possibility that the active desublimator surface will be deiced too early or too late, which will in each case ultimately downgrade the entire technological process of sublimation drying. It must be taken into account that a sublimator condenser often consists of several segments, each of them receiving a different amount of vapor and each of them with a different ice layer geometry [16, 17, 18].

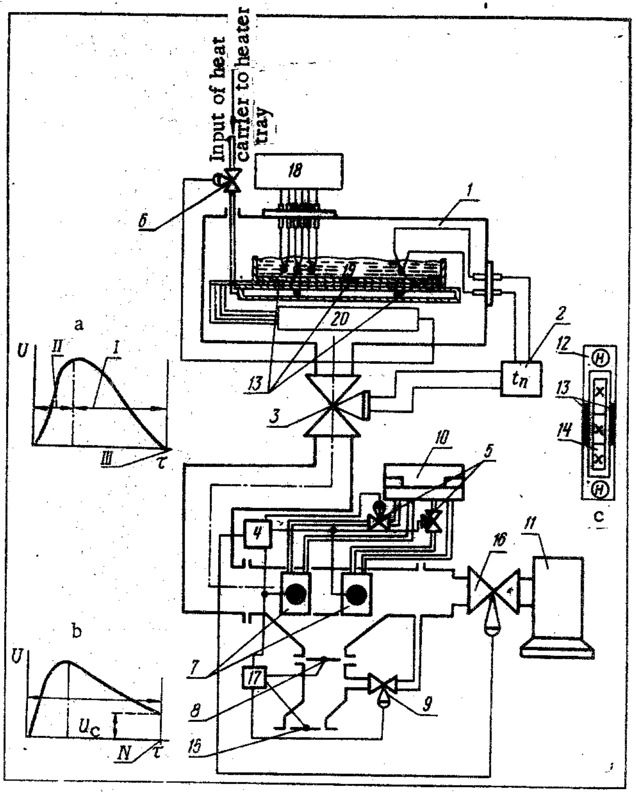

Fig. 4. Schematic diagram of an apparatus for contactive sublimation drying with TF probes: 1) sublimator; 2) temperature regulator; 3) vacuum valve; 4) automatic regulation system for the desublimation process; 5) coolant discharge valve; 6) regulator; 7) condenser segment; 8, 15) gate valves; 9, 16) vacuum valves; 10) cooler aggregate; 11) vacuum pump; 12) heater elements; 13) thermal flux probes; 14) coolant ducts; 17) gate valve actuator; 18) product temperature control; 19) product; 20) comparator; a) desiccation process [I) period of decreasing desiccation rate; II) period of heating and constant desiccation rate; III) end of desiccation]; b) condensation process [IV) end of condensation]; voltage U (V).

In order to eliminate this drawback too, the authors have developed a system for automatically deicing the active condenser components (Fig. 3d). With the aid of the potentiometer (Po) 12, a definite voltage level $U_p = U_f$ is established which corresponds to the minimum feasible (both technically and economically) electric signal from the TF probe 16, and this voltage is applied to the b input of the null indicator 4.

Inasmuch as desublimation is a twofold nonsteady process, i.e., since its rate decreases with time, because of the increasing thermal resistance of the product which forms on the active surface in a vacuum-type condenser, and this thermal resistance is again proportional to the process rate, hence the magnitude of signal U_{out} generated at the AR probe output and transmitted to the a input of null indicator 4 also decreases.

As soon as the magnitude of signal U_{out} at the AR probe output becomes smaller than the magnitude of the reference voltage U_p at the potentiometer (Po) 12, a signal from the null indicator 4 is transmitted to amplifier 8 to actuate relay (Rc) 9 and thus the servomechanism for deicing the given condenser segment.

Because the desublimation process is not effective enough during the period of crystal nucleation with corrugated ice film formation [13, 14] and $U_{out} < U_p$, we have added a time relay (TR) 10 to the circuit in Fig. 3d which actuates the deicing system only after a definite time delay, when U_{out} becomes greater than U_p.

Basic Automatic Regulation System for Sublimation Drying. For illustration, let us consider a desiccator with an automatically regulated contactive sublimation drying process* and with the TF probes installed directly on the bottom of the tray containing the raw material, as shown in Fig. 4. Test data and calculations indicate that a TF probe can be designed also for radiative–convective heat supply. In that case, probes are placed not only in the middle of the tray but also at its edges (within the zone of nonuniform thermal flux distribution), and charring of the product is thus prevented. Information is picked off each TF probe in terms of sublimation curves (Fig. 4a) and is continuously transmitted to a comparator. The control valve meters the amount of heat carrier supplied into a segmented heater and each respective sublimation curve thus becomes adjusted to the optimum shape for every given raw material. The sublimator vacuum is regulated through a temperature control 2 (with the product temperature below the cryoscopic point as the reference level [20]) and a valve 3.

The desublimation process is regulated by a system of valves 5, a source of low-temperature heat carrier with a vacuum valve 16, and the optimal-control system (described earlier) with TF probes 7. The condenser segments are switched on periodically as they become cleared of desublimated ice.

The system of gate valves 8, 15 and the valve 9, together with the gate valves actuator 17, ensure the removal of ice from the condenser (desublimator) by switching on the heaters 12 (Fig. 4b).

The proposed concept of an automatic regulating system for contactive sublimation drying can be developed further for the engineering design of continuous-duty apparatus.

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*Contact feed for sublimation drying is most widely used in large plants, on account of its favorable technoeconomic indicators.