CONDENSATION RATE AND COEFFICIENTS OF REFRACTION AND REFLECTION OF VACUUM CRYOCONDENSATES OF WATER VAPORS ON A METALLIC MIRROR

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Specific features of heat transfer are investigated at the "gas–solid" interface at low temperatures for the case of water vapor condensation.

The present work is concerned with results of the experimental study of the growth rate of water cryocondensate layers at low temperatures and some properties of the formed precipitates. Such studies, being of obvious applied value, are also of pure scientific interest. This is first of all attributed to essential correlation between thermodynamic parameters of phase formation, mass transfer characteristics in the gas–surface system and a number of such properties of a solid-state phase as density, refractive index, reflectivity, etc. Obviously, such correlation must have a single basis of being most likely of a structural character.

Numerous recent studies in this field [1-3] point, directly and indirectly, to the presence of various structures of vacuum cryocondensates dependent on the condensation temperature and the pressure of the gaseous phase. The present authors make attempts to generalize results of the studies of three independent characteristics of water cryocondensates at temperatures ranging from 80-180 K in the pressure range 10^{-2}-10^{-4} Pa and compare them with data of other authors. In particular, the data are reported on the growth rate, refractive index and specular reflection coefficient of the test cryocondensate in the visible region as a function of the condensation temperature and pressure.

Investigations were conducted in accordance with the procedure described in [4] on an experimental set-up representing a multipurpose vacuum spectrophotometer. The main specific feature in obtaining experimental data lies in direct use of experimentally obtained refractive indices to calculate the condensation rate. This, in the present authors' opinion, is of importance since a possible change in the precipitate structure for different phase formation conditions is taken into account. After cessation of the growth process, we measured the substrate reflectivity (specular reflection coefficient for an angle of incidence of 50°) covered with the condensate in dependence on the wavelength. Then the condensate was subjected to stepwise heating during which reflectivity was measured at stationary temperatures. The flat surface from stainless steel 1Kh18N10T was used as the substrate.

All the results are presented in relative values with respect to the characteristic points of the relations.

Figure 1 displays the data on the temperature-dependent growth rate of water cryocondensate growth from a gaseous phase for three different pressures. As is seen, these dependences are of an essentially nonlinear character, namely, a condensation rate at first increases with decreasing a substrate temperature from 180 to 160 K, then drastically decreases and attains its minimum at a temperature of about 140 K. With subsequent decrease of the temperature, the growth rate monotonically increases. Analogous results for some other gases are obtained earlier in [5] where an explanation is offered from the viewpoint of substituting the "gas–metastable liquid–crystal" condensation mechanism by the "gas–crystal" mechanism which may be extended to the results discussed.

It is obvious that change in the phase transformation mechanism does not fail to influence a structure of the formed phase which is illustrated by the dependence of the water cryoprecipitate refractive index on the condensation surface temperature (Fig. 1d). As is seen, the refractive index increases within the temperature range 180-160 K and, remaining constant at 150 to 120 K, again decreases with a further decrease of the temperature.

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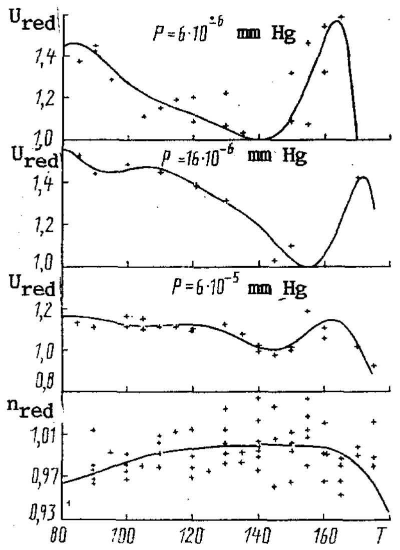

Fig. 1

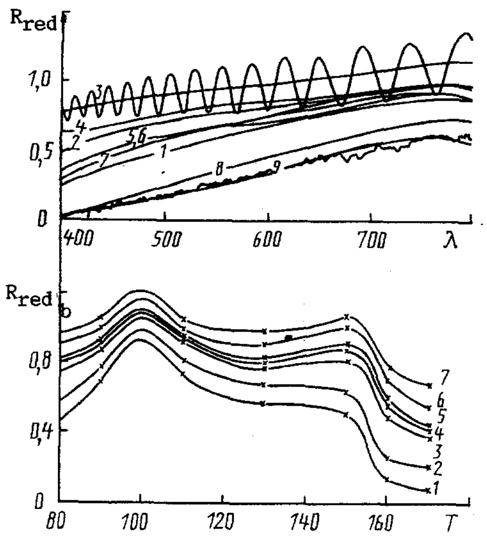

Fig. 2

Fig. 1. Reduced condensation rate of water vapors (a, b, c) and reduced refractive index (d) vs condensation surface temperature. T, K; U_{red}, n_{red}, dimensionless.

Fig. 2. Reduced refraction coefficient (dimensionless) of cryocondensate layers of water vapors formed at different temperatures {1) 80 K; 2) 90; 3) 100; 4) 110; 5) 130; 6) 140; 7) 150; 8) 160; 9) 170 K] vs wavelength (a) and temperature for different wavelengths [1) 450 nm; 2) 500; 3) 550; 4) 600; 5) 633; 6) 700; 7) 750 nm] (b).

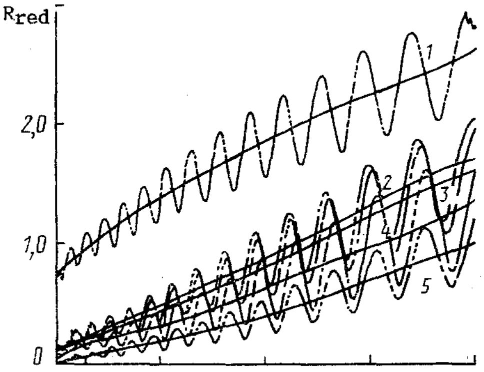

Fig. 3. Spectral dependence of the reduced reflection coefficient of cryocondensate layers of water vapors formed at 80 K with subsequent warming: 1) 80 K; 2) 80-100; 3) 80-120; 4) 80-140; 5) 80-160 K.

Figure 2a displays the reflectivity of a substrate covered with a 5 μm thick water cryocondensate layer for different condensation temperatures as a function of wavelength (reflectivity measurements are conducted at the same temperatures at which the condensates have been grown). Curves 1-9 represent the 5th-degree polynomial-smoothed dependences of the reflection coefficient on wavelength. For T = 100 K, the initial (unsmoothed) dependence of an interference character is shown which is typical of temperatures from 80 to 160 K. At 165 K and up, no interference is observed; a typical form of the unsmoothed reflectivity is shown for 170 K. To illustrate reflectivity variation with condensation temperature, Fig. 2b shows the temperature profiles of this family of curves for different wavelengths. The reflectivity of the substrate covered with a 5

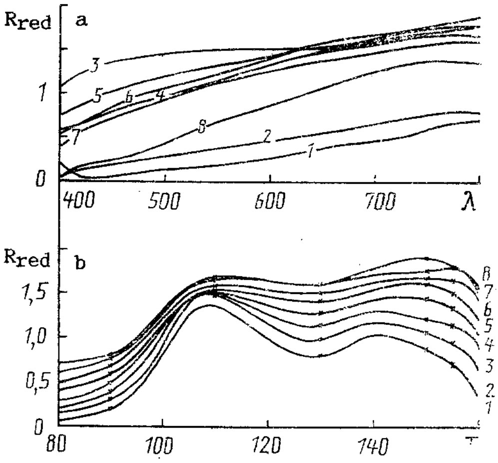

Fig. 4. Reduced reflection coefficient of water vapor cryocondensate layers formed at different temperatures [1) 80 K; 2) 90; 3) 110; 4) 130; 5) 140; 6) 150; 7) 155; 8) 160 K] with subsequent warming to 160 K as a function of wavelength (a) and condensation temperature for different wavelengths [1) 450 nm; 2) 500; 3) 550; 4) 600; 5) 633; 6) 650; 7) 700; 8) 750 nm] (b).

μm ice layer at a temperature of 100 K for a wavelength of 633 nm is assumed to be unity. Within the range 100-150 K, a change in the reflectivity variation with temperature at all wavelengths of the test range is observed. This specific feature obviously correlates with a behavior of the curves in Fig. 1.

An increase of the reflectivity at condensation temperatures of 100 and 150 K is so remarkable that, according to the data of different authors, just at these temperatures the transition from one crystalline structure to another proceeds. We cannot offer the convincing explanation to the presence of extrema at the indicated temperatures.

Data on the possible dependence of some properties of cryocondensed gases not only on the temperature of formation but also on the subsequent conditions of their existence, in particular temperature, have predetermined the choice of an experimental procedure, the results of which are shown in Figs. 3 and 4. Figure 3 depicts spectral dependences of the reflectivity of the substrate covered with a 5 μm thick cryocondensate layer at a condensation temperature of 80 K measured at different stationary temperatures in the course of subsequent warming. As analysis of the reported data reveals, the test layer reflectivity essentially depends on warming. In particular, warming from a condensation temperature of 80 K to 160 K causes more than a two-fold decrease in the reflection coefficient. Besides, warming from 80 to 120 K is seen to affect reflectivities more appreciably than the subsequent warming of this layer from 120 to 160 K. Most likely, this is connected with structural changes of the precipitate formed.

It may be logically assumed that thermocyclization within the temperature range of the existence of this or that structure will not result in such considerable changes of precipitate properties. This is supported by data depicted in Fig. 4. Here 5 μm layers of water cryocondensate have been formed at different temperatures and then warmed to the same temperature, i.e., 160 K. It is seen that spectral characteristics of cryocondensates (Fig. 4a) formed in the temperature range 120-155 K and warmed to 160 K are grouped in one region. Below, the curves for the condensation temperature $T = 160$ K and next for the cycles 90-160 K and 80-160 K follow. Thus it may be assumed that in the temperature range 120-155 K the water cryocondensate is sufficiently resistant to thermocycling within the same interval.

Analysis of the reported data yields the following conclusions.

1. In the considered temperature and pressure ranges of the gas–solid phase transition an anomalous behavior of the condensate growth rate as a function of condensation temperature is observed.

2. In different condensation temperature ranges, different-structure cryocondensate layers are formed due to the phase transformation. Comparison of our data with the results of [6] permit us to make the following assumption: the temperature range from 160 K and up is characterized mainly by growth of the phase having a hexagonal crystalline structure. At tempera-

tures of about 155 K, this structure is transformed into a cubic one for which is stable in the temperature range 150-120 K. The next decrease of the condensation temperature leads to formation of mainly amorphous densely packed structures [7, 8].

3. Owing to thermovariations in the layers formed at lower temperatures, the structure undergoes rearrangement thus causing a change in the reflectivity of the test surface. These changes are more essential, the larger the number of phase boundaries that intersect.

The increasing reflectivity observed at the boundaries of transition from one structure to another (see Figs. 2, 4) requires further investigation.

NOTATION

λ, wavelength, nm; n_{red}, reduced refractive index; α, angle of incidence, deg; r, thickness, μm; p, pressure, mm Hg; T, temperature, K; R_{red}, reduced reflection coefficient; U_{red}, reduced condensation rate.

REFERENCES

1. V. B. Yuferov, in: Nuclear Science and Engineering. Ser. Low-Temperature Adsorption and Cryogenic Vacuum [in Russian], Khar'kov (1973), Issue 1(4), pp. 41-47.
2. W. Schulze, D. Kolb, and G. Klipping, in: The Density of Cryopumped Gases, Berlin Dahlem (1975), pp. j6-j10.
3. K. E. Tempelmeyer and D. Mills, J. Appl. Phys., **39**, No. 6, 2068-2069 (1968).
4. N. V. Atapina, D. N. Garipogly, A. S. Drobyshev, et al., Inzh.-Fiz. Zh., **61**, No. 6, 960-966 (1991).
5. A. S. Drobyshev, "A dependence of a growth rate and thermal conductivity of gases cryocondensates on the temperature and pressure of the 'gas–solid,' phase transition" Candidate's Dissertation (Physics-Mathematics), [in Russian], Alma-Ata (1984).
6. M. Sagisaki, H. Suga, and S. Seki, in: Physics of Ice, New York (1969), pp. 329-335.
7. A. N. Delsemme and A. Wenger, Science, **167**, 44-45 (1970).
8. V. P. Koverda, N. M. Bogdanov, and V. P. Skripov, J. Non-Cryst. Sol., **57**, 203-212 (1983).