Investigation of the processes of heat and mass transfer during cooling and crystallization of droplet nuclei of hail

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Abstract: The regularities of crystallization of water droplets in an air stream and the processes of heat and mass transfer have been investigated. Theoretical calculations for determining the time of complete solidification of a drop are presented. Calculation formulas are given for determining the Reynolds number and wind coefficient, as well as comparative curves of theoretical and experimental studies of the average time of complete freezing of drops. It is shown that the theoretical calculations are in agreement with the experimental data. It was found that with intense blowing, the droplet crystallizes in a regime in which its surface is maintained at a constant temperature. An ice crust forms in the form of a dendrite when supercooled droplets crystallize. On the basis of numerous experiments, it has been established that the character of droplet crystallization substantially depends on the external conditions under which the crystallization process takes place.

1. Introduction
In works [1, 2], devoted to the study of the physics of hail clouds and active influences, it is noted that many issues related to the formation of microstructural characteristics of hail, until now remain insufficiently studied. These include the conditions for the formation and growth of hail nuclei, the processes of formation of primary drops and crystals in clouds, the growth of crystals of various shapes, the electrification of cloud particles, and some other processes [3, 4]. In work [2] it is also noted that, despite the long-term study of microphysical processes in clouds, leading to the formation and growth of hail, the long-term practice of actively influencing hail processes, still in the theory of both natural hail formation and the management of its development remains there is still a lot of unclear what hinders the achievement of further successes in this area.

2. Results and discussion
Studies of heat and mass transfer processes during crystallization of water droplets hovering in an air stream are of great interest for influencing hail processes. In this regard, in the course of experiments in the laboratory of microphysics of clouds of the High Mountain Geophysical Institute, a study was made of the regularities of crystallization of water droplets in an air stream and the processes of heat and mass transfer during cooling and crystallization of droplets. To implement the set task, an experimental setup was used, a detailed description of which is given in [5], which makes it possible to study the processes of cooling, solidification and precipitation of hailstones floating in the air flow. In [6, 7], two types of crystallization of droplets, determined by the degree of their supercooling, are
described in detail. According to the data [7], on the basis of numerous experiments, it was established that the nature of crystallization of drops significantly depends on the external conditions under which the process of their crystallization occurs.

When supercooled droplets crystallize, an ice crust forms in the form of a dendrite. The thickness of this crust is proportional to the degree of supercooling of the droplets. This circumstance makes it possible to estimate the temperature of the medium at which the crystallization of droplets takes place. In some cases, an explosive splitting of droplets occurs, in which many ice fragments are formed.

When the temperature of the droplets is close to 0°C, the crystallization process begins from their surface and moves slowly, and the branched crystal is located near the surface of the droplets. In this case, the ice shell turns out to be more transparent, and the probability of explosive splitting of drops increases. Since the drop is in a suspended state, its surface will be cooled symmetrically, the temperature $T$ of the drop throughout the entire volume of the drop remains uniform, it gradually decreases until a solid phase appears on the surface of the drop.

Under these conditions, the heat balance equation looks like this:

$$\frac{d}{df}\left( \frac{4}{3} \pi R^3 \rho c T \right) = -4\pi R \left[ \lambda (T - T_\infty) + LD (\rho_1 - \rho_s) \right] \left( 1 + \beta \text{Re}^{1/2} \right),$$  \hspace{1cm} (1)$$

where $\rho$; $c$ – density and heat capacity of water respectively; $\lambda$ – coefficient of thermal conductivity of water; $D$ – air vapor diffusion coefficient; $L$ – specific heat of vaporization; $\rho_s$ – saturating vapor density on the droplet surface (at temperature $T$); $\rho_\infty$ – saturating density of vapor in the air stream (at temperature $T_\infty$); $\beta$ – an empirical parameter of approximately 0.6; $\text{Re}$ – Reynolds number.

$$\text{Re} = \frac{2RV}{V},$$

where $V$ is the flow rate, $\nu$ is the coefficient of the kinetic viscosity of air.

The multiplier $1 + \beta \text{Re}^{1/2} = F$ is commonly referred to as the wind multiplier; it takes into account the effect of the flow on the heat and mass transfer of the droplet with the environment, where $\beta$ is an empirical parameter, approximately equal to 0.6.

In equality (1), the first term in square brackets $\lambda (T - T_\infty)$ takes into account droplet cooling due to heat exchange of its surface with the environment, and the second term $LD (\rho_1 - \rho_s)$ takes into account droplet cooling due to evaporation from its surface.

According to the Clapeyron - Clausius equation:

$$\rho_s = \frac{\rho_1 T_\infty}{T} \exp \left[ \frac{L}{R_s} \left( \frac{1}{T_\infty} - \frac{1}{T} \right) \right],$$

where $R_s$ – gas constant of water vapor ($R_s = \frac{R_0}{\mu}$, where $R_0$ – universal gas constant, $\mu$ – molecular weight of water).

Usually supercooled droplets crystallize at temperatures $-10 \div -12^\circ C$, while

$$\frac{L}{R_s} \left( \frac{1}{T_\infty} - \frac{1}{T} \right) \approx \frac{L}{R_s T_\infty} (T - T_\infty) \ll 1.$$ 

Therefore, we can write:

$$\rho_s - \rho_\infty = \delta (T_\infty - T_\infty).$$  \hspace{1cm} (2)
where \( \delta = \frac{\rho c L}{R \rho^*} \approx 2.7 \cdot 10^{-2} \text{m}^2 \cdot \text{s} \).

Taking into account relation (2), equation (1) takes a simpler form:

\[
\frac{dT}{dt} = -a(T - T_{\infty}),
\]

where

\[
a = \frac{3\lambda}{\rho c R^2} \left(1 + \frac{L \delta D}{\lambda}ight) \left(1 + \beta \text{Re}^{-\frac{1}{2}} \right) (c^{-1}).
\]

A solution to this equation satisfying the initial condition that for \( t = 0 \) \( T = T_0 \), looks like:

\[
T = T_{\infty} - (T_0 - T_{\infty}) e^{-at},
\]

that is, the temperature drops exponentially; also for, i.e. the droplet temperature gradually approaches the air temperature exponentially.

Let the ice phase appear on the droplet surface at a certain temperature \( T = T_k \geq T_{\infty} \), i.e. the crystallization process of the drop begins. The time of the onset of crystallization can be determined from the formula:

\[
t_k = \frac{1}{a} \ln \frac{T_0 - T_{\infty}}{T_k - T_{\infty}}.
\]

At the moment of time \( t = t_k \), the second stage of the droplet crystallization process begins. The surface of the drop is covered with a thin ice shell. After the formation of a thin ice shell on the surface of the drop, the third stage of the crystallization process begins - the crystallization of the drop itself.

Since the thermal conductivity of water is approximately two orders of magnitude greater than the thermal conductivity of air, it can be assumed that the entire amount of heat released during the formation of the ice shell completely goes into the inside of the drop. The thickness of the ice shell \( h \) can be estimated using the following formula:

\[
h \approx \frac{\rho^*}{\rho} \cdot \frac{cRT_k}{3L} \approx \frac{cRT_k}{3L},
\]

where \( \rho^* \) is the density of ice; \( L^* \) - specific heat of water crystallization; \( T_k \) - droplet hypothermia temperature with a «+» sign. The formation time \( \tau^* \) of the ice shell on the droplet surface is of the same order of magnitude as the relaxation time of the temperature field in the droplet. After the formation of a thin ice shell on the surface of the drop, the third stage of the crystallization process begins - the crystallization of the drop itself. In this case, the temperature of the liquid core remains equal to zero all the time until the complete crystallization of the drop. In figure 1 schematically shows a crystallizing drop: \( \xi(t) \) – the radius of the liquid core (crystallization front). Taking into account that \( \frac{h}{R} \ll 1 \), we can practically identify the initial radius of the liquid core with the radius \( R \) of the drop.

As the crystallization front moves to the center of the drop, the surface temperature \( T_s \) of the ice shell will decrease. Thus, two quantities are to be determined: \( T_s \) and \( \xi(t) \). They are determined from the heat balance equation on the surface of a spherical shell and from the so-called Stefan condition, which expresses the heat balance at the moving interface.
Let us now formulate the condition for the balance of heat fluxes on the surface of the ice shell. Taking into account that evaporation from the ice surface does not play a significant role in the heat balance, and that the heat flux into the liquid core is zero, we can write:

$$-\lambda^* \frac{dT}{dr} \bigg|_{r=R} = \frac{\lambda^*_a F}{R} \cdot (T_a - T_n),$$

where $\lambda^*_a$ is the thermal conductivity of air, $F = 1 + \beta \text{Re}^{1/2}$ is the wind factor.

Taking into account that,

$$\frac{dT}{dr} \bigg|_{r=R} = \frac{2 T_a}{R \left( \frac{R^2}{\xi^2} - 1 \right)}$$

we get:

$$T_n = \frac{T_a}{1 + \frac{2 \lambda^*}{\lambda^*_a F} \left( \frac{R^2}{\xi^2} - 1 \right)} = \frac{T_a}{1 + \frac{2 \lambda^*}{\lambda^*_a F} \cdot \frac{\xi^2}{R^2} \cdot \frac{1 - \xi^2}{R^2}}$$

From this it immediately follows that at the moment of complete freezing of the drop, at $\xi = 0$, $T_n = T_\infty$. Those the surface temperature of the ice shell becomes equal to the temperature of the air flow.

If during the time $dt$ the crystallization front moves to the center by $d\xi$, then per unit time an amount of heat is released equal to $-\rho L \frac{d\xi}{dt}$. The released amount of heat must be removed from the crystallization front into the ice shell through the thermal conductivity of the ice. Then we get:

$$\rho L \frac{d\xi}{dt} = \lambda^* \frac{dT}{dr} \bigg|_{r=R}$$

Figure 1. Schematic representation of a crystallizing drop.
\[
\frac{dT}{dr} |_{r = \xi} = -\frac{2T_u}{R^2} \left( \frac{\xi^2}{R^2} - 1 \right) = -\frac{2T_u}{\xi^2 \left( \frac{\xi^2}{R^2} - 1 \right)} \]

Substituting this expression in (7) we get:

\[
\xi \frac{d\xi}{dt} = \frac{2\lambda^* T_u}{\rho c L}, \quad \frac{1}{1 + \left( \frac{2\lambda^*}{\lambda_u^*} \right) \xi^2} \cdot \frac{4\lambda T_u}{R^2 \rho L} \frac{1}{1 + \left( \frac{2\lambda^*}{\lambda_u^*} \right) \xi^2}.
\]

We denote \( \frac{\xi^2}{R^2} = x \): then \( \xi \frac{d\xi}{dt} = \frac{1}{2R^2} \frac{dx}{dt} \), and equation (8) will take the form:

\[
\frac{dx}{dt} = \frac{4\lambda T_u}{R^2 \rho L} \frac{1}{1 + \left( \frac{2\lambda^*}{\lambda_u^*} \right) x - 1} x;
\]

or

\[
\left[ 1 + \left( \frac{2\lambda^*}{\lambda_u^*} \right)(x - 1) \right] dx = \frac{4\lambda T_u}{R^2 \rho L} dt
\]

from where:

\[
x + \frac{\left( \frac{2\lambda^*}{\lambda_u^*} - 1 \right) x^2}{2} = \frac{4\lambda T_u}{R^2 \rho L} t + c_1,
\]

where \( c_1 \) – arbitrary constant. When \( t = 0 \) \( \xi = R \), \( x = 1 \), from where \( c_1 = \frac{1}{2} + \frac{\lambda^*}{\lambda_u^*} \). The law of displacement of the crystallization front is determined from the equation:

\[
\frac{\xi^2}{R^2} + \left( \frac{\lambda^*}{\lambda_u^*} - \frac{1}{2} \right) \left( \frac{\xi^2}{R^2} - 1 \right) = \frac{4\lambda T_u}{R^2 \rho L} t + \frac{1}{2} + \frac{\lambda^*}{\lambda_u^*}.
\]

For researchers, the most interesting is to determine the time of complete freezing of a drop, assuming that \( \xi = 0 \) it will be equal to:

\[
\tau = -\frac{\rho L R^2}{4\lambda T} \left( \frac{1}{2} + \frac{\lambda^*}{\lambda_u^*} \right).
\]

To compare the theoretical values of the time of complete freezing of a drop, obtained by formula (10) and the values obtained in laboratory conditions, a series of experiments was carried out.

During the experiments, the following values were measured: droplet size \( (d, \text{mm}) \); air flow temperature at the location of the drop \( (T, \circ\text{C}) \); air flow velocity in the same place \( (v, \text{m/s}) \); time of the processes of complete solidification of the drop \( (\tau, s) \).

Due to the fact that the droplets do not have a spherical shape during motion, their equivalent size is used as the characteristic droplet size, i.e. the diameter of a sphere with the same volume as the drop. In the experiment, the droplet diameter was determined from a known volume. Water drops were
introduced into the working tube with a 0.2 ml micropipette with a graduation of 0.002 ml. Knowing the volume of water (v) for the formation of a drop, its equivalent diameter is calculated according to the known dependence:

$$d = \sqrt[3]{\frac{6v}{\pi}}.$$  \hspace{1cm} (11)

To measure the air flow velocity directly in the region of the droplet location, the method of measuring by differential Pito tubes was used [9]. The air flow rate at this point was determined by the formula:

$$V = \frac{-2(P_{10} - P_1)}{\rho_a},$$  \hspace{1cm} (12)

where $P_{10}$ and $P_1$ are dynamic and static pressure, respectively; $\rho_a$ - air density.

The air flow temperature and the droplet temperature were measured using copper-constant thermocouples [9].

The processes of cooling and subsequent solidification of a water droplet are unsteady; therefore, the task of the experimental study was to obtain the dependence of the droplet temperature on time during the entire experiment.

The results of experimental modeling of the crystallization process of water droplets were compared with the works of M. K. Zhekamukhov [6, 9], the relative error of the series of measurements is 6–7%. In figure 2 comparative curves of theoretical and experimental studies of the average time of droplet solidification are presented.

Thus, the studies have shown that the theoretical calculations are in agreement with the experimental data.

![Figure 2](image_url)

**Figure 2.** Dependence of the time of water drop complete freezing on the size: a - according to the results of our experiments; b - according to the results of theoretical calculation [9].

### 3. Conclusion

With intensive blowing of the droplet, it crystallizes almost as in the case when a supercooled droplet crystallizes in a regime in which its surface is maintained at a constant temperature. It should be noted that the temperature of spontaneous crystallization of droplets in convective clouds is approximately $-10^\circ C$, which coincides with the natural crystallization of large droplets at the isotherm level $-8 \div -11^\circ C$, as is customary in [10] and intensive growth of hailstones in the temperature range $-10 \div -25^\circ C$ in the monograph [1].

### References

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