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Effect of Magnetic Field on the Supercooling of Water Drops

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Abstract—The effect of a static magnetic field on the solidification behavior (critical supercooling, latent period, etc.) of water drops under atmospheric conditions was studied by thermal analysis. The critical supercooling was found to decrease with increasing magnetic field. In fields stronger than 0.5 T, supercooling is negligible and equilibrium solidification takes place.

Although the solidification kinetics of water have been the subject of many works [1–8], the origin of deep water supercooling and the mechanism of water solidification are still unclear. In this work, we present experimental studies of water solidification in a magnetic field during thermal cycling in a furnace with a considerable thermal mass. This method was used in our earlier works [9–12] to study other substances and was shown to have a satisfactory sensitivity to endo- and exothermic events.

Experiments were made on about 100 drops (0.5 g) of distilled water (USSR Standard GOST 6709-72) from different vendors in a purpose-designed resistance microfurnace, which was inserted in the 17-mm gap of an annular magnet 30.5 × 37 mm^2 in cross section. The water drop to be studied was enclosed in a copper or polyethylene crucible or applied to the glass-sheathed Chromel–Alumel thermocouple. The assembly was brought outdoors in windless, cloudless weather at an air temperature of –18 to –20°C. The measuring equipment and control panel were kept indoors. Up to 50 cooling/heating cycles were carried out for each sample. The cooling/heating rate was 0.06–0.3 K/s. The temperature (–20 to 50°C) was measured to within 0.15–0.20 K. The signal from the thermocouple was fed to a KSP-4 chart recorder (1-mV scale). The magnetic field was varied from 0 to about 0.5 T. The reproducibility of results was checked repeatedly for each drop at time intervals of up to a few days.

Experiments were made first in zero field. In the temperature-vs.-time curve shown in Fig. 1, line *ghkla* represents sample heating at 0.18 K/s. Points *h*, *b*, and *d* lie on the horizontal corresponding to the melting temperature of ice, 0°C. In all the heating curves, an additional endotherm was observed above 0°C. In Fig. 1, this endotherm lies at $T_c^+ = 2.6°C$ (point *l*). The corresponding heat effect is $L_x \simeq 8$ kJ/kg. Line *la* represents water heating to 9.8°C. After switching off the furnace at the point labeled ⊖, the temperature continues to rise, by virtue of thermal lag, to point *a* and then falls—at about 0.09 K/s to point *b* and at a somewhat slower rate to point *c* ($T^-_{\min} = -7.6°C$). Next, the sample temperature rises abruptly, at about 20 K/s, to 0°C (line *cd*), indicating that a reaction accompanied by a vigorous heat release (heat effect Q_1) occurs in the supercooled water. Portions *de* and *ef* represent water solidification and ice cooling, respectively. Three periods of time can be distinguished in the cooling curve: latent period τ_1, during which water is in a metastable,

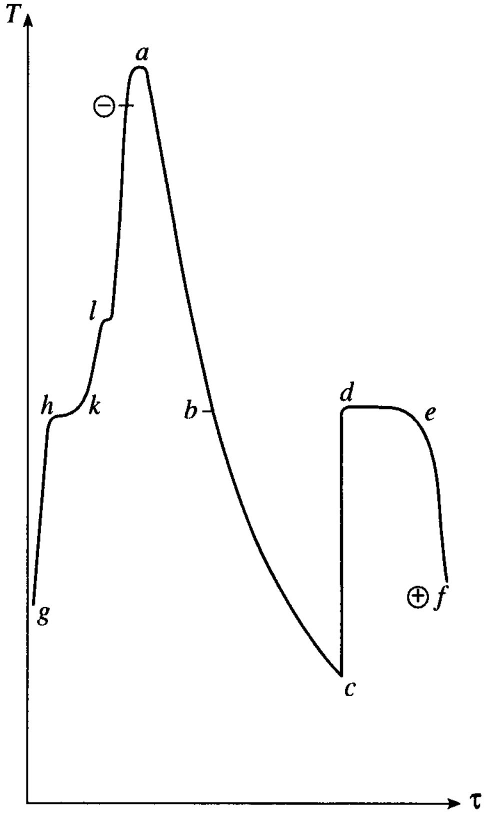

Fig. 1. Temperature curve recorded in the course of ice melting and water solidification in zero field.

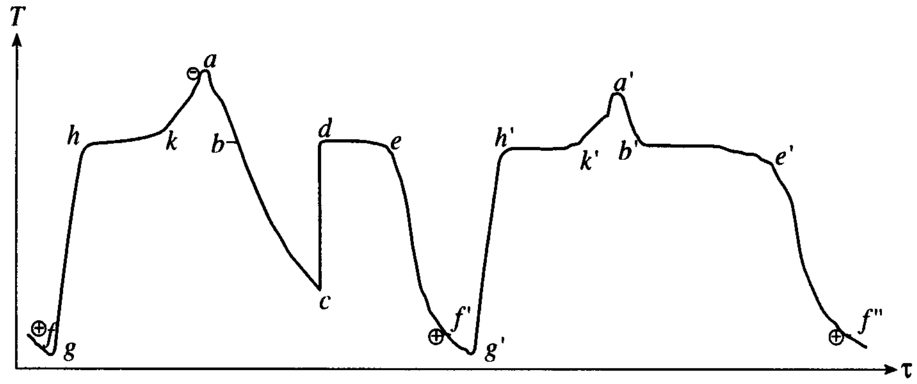

Fig. 2. Temperature curve illustrating the transition from solidification with supercooling to that without supercooling in the absence of a field.

supercooled state (*bc*); period τ_2 of the rapid temperature rise (*cd*); and solidification period τ_3 (*de*). Thus, the whole solidification process, including the latent stage, takes time $\tau = \tau_1 + \tau_2 + \tau_3$. For the curve shown in Fig. 1, we have $\tau_1 = 222$ s, $\tau_2 = 0.5$ s, $\tau_3 = 156$ s, and $\tau = 378.5$ s.

The endotherm at point *l* (at temperature T_c^+) is not an artifact. Similar features were observed for all the drops between 2.4 and 3.0°C, with an average temperature $\langle T_c \rangle \approx 2.7$°C. It is remarkable that, after heating from 0°C to a temperature $T_L < \langle T_c^+ \rangle$, cooling leads to equilibrium solidification of water drops, without supercooling ($\Delta T^- = 0$). After preheating to just above $\langle T_c^+ \rangle$, we observe explosive solidification following supercooling by 7.6–8.0°C. The transition between the two types of solidification behavior is very sharp and reversible.

The data shown in Fig. 2 illustrates the transition from explosive to equilibrium solidification. In the first cycle (*ghkabcdef'*), explosive solidification took place after supercooling. In the second cycle (*g'h'k'a'b'e'f''*), the drop was preheated to a temperature lower than that at point *a*, corresponding to point *l* in Fig. 1, that is, lower than T_c^+, and exhibited equilibrium solidification with $\Delta T^- = 0$. The time τ taken for equilibrium (*b'e'*) and explosive (*be*) solidification is nearly the same. Preheating to 50°C, holding for up to a few hours at a temperature above $\langle T_c^+ \rangle$, or varying the cooling rate had no effect on ΔT^-. The scatter in ΔT^- from cycle to cycle was ±0.4–0.5°C in all the experiments.

Given that the solidification behavior of water depends on sample overheating, the experiments in a magnetic field were carried out under identical temperature programs between −18° and +20°C, i.e., under conditions ensuring, in zero field, explosive solidification with $\Delta T^- = 7.6$–8.0°C ($\langle \Delta T^- \rangle \approx 7.8$°C).

The changes in solidification behavior caused by a static magnetic field of ≈0.5 T are illustrated in Fig. 3.

Note, first of all, that there is no heat effect at T_c^+ (Fig. 1, point *l*). Second, ΔT^- is decreased to about zero, and the latent period is much shorter than that in zero field. The field dependence of ΔT^- is shown in Fig. 4.

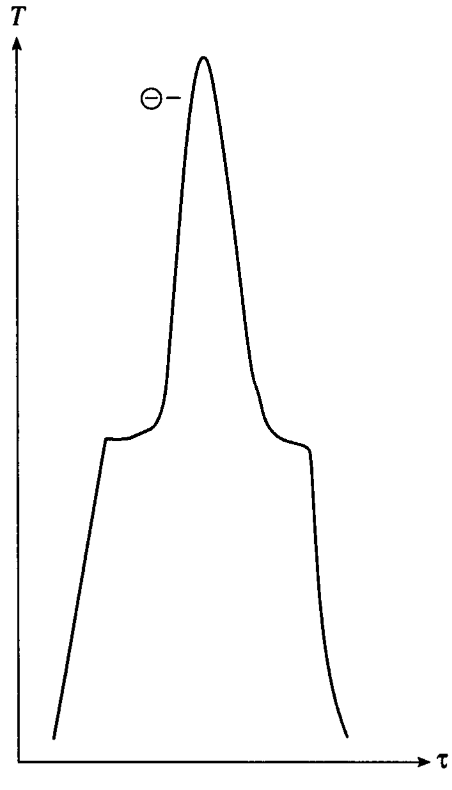

Fig. 3. Temperature curve recorded in the course of ice melting and water solidification under a magnetic field of 0.5 T.

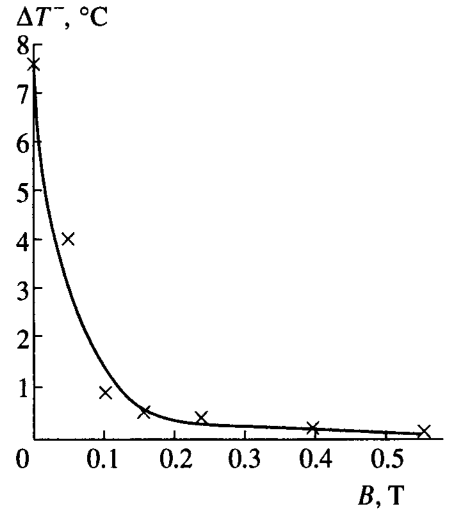

Fig. 4. Critical water supercooling as a function of magnetic field.

Our data on the solidification behavior of 0.5-g water drops in a magnetic field are summarized in the table.

The present results can be interpreted in terms of the cluster-coagulation model suggested in our earlier works [13, 14]. According to this model, the density of nuclei in a metastable, supercooled melt increases during the latent period τ_i (*bd* in Fig. 1). As soon as the critical density of nuclei is attained at a certain critical supercooling, ΔT_c^-, equal to the temperature difference between points *c* and *d* in Fig. 1, the nuclei undergo orientation ordering, approach one another, and coagulate rapidly. The process is accompanied by vigorous heat release, $Q_1 = m_x L$ (here, m_x is the total weight of nuclei just before coagulation, and L is the specific heat of solidification), leading to rapid sample heating (from point *c* to *d*).

The heat Q_1 raises the sample temperature by ΔT_c^-. With no allowance for heat losses, we have

$$m_x L = C m \Delta T_c^-, \tag{1}$$

where C is the specific heat.

Solidification behavior of water drops in a magnetic field

B, T	$\langle \Delta T^- \rangle$, °C	$\langle \tau_1 \rangle$, s	$\langle \tau_2 \rangle$, s	$\langle \tau_3 \rangle$, s	$\langle \tau \rangle$, s	η
0	7.6	220	0.5	158	378.5	≈0.1
0.071	2.5	75	1.5	304	378.5	0.03
0.157	0.7	20	3.0	357	380.0	0.009
0.235	0.4	10	5.0	361	376.0	0.005
0.505	≈0	≈0	0	379	379.0	≈0

Therefore, just before coagulation, the weight fraction of nuclei is

$$\eta = \frac{m_x}{m} = \frac{C \Delta T_c^-}{L}. \tag{2}$$

Under the assumption that the magnetic field has a negligible effect on C and L, η can be determined as a function of magnetic field by substituting the measured ΔT_c^- into (2). The results (table) show that η decreases with increasing field.

This finding suggests that the magnetic field favors the orientation ordering of nuclei, presumably by virtue of the diamagnetic effect, and ensures their coagulation at a lower supercooling.

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