Impact of Suspended Solids on Coarsening of Ice

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ABSTRACT: Suspended solids, such as silica particles and cellulose fibers, were added to a sucrose aqueous solution, and ice crystals were coarsened at $-10 \degree C$. From the radius of the ice crystals, the coarsening rate constant was obtained using the Lifshitz–Wagner equation and the impact of the suspended solid on the coarsening of ice was evaluated. The results showed that the addition of the silica particle suppressed coarsening, but this behavior was not dependent on particle size. It was also shown that cellulose fibers suppressed coarsening more than silica particles. In order to clarify these causes, the present study investigated the correlation between $L_w - L_{mea}$ and the coarsening rate constants obtained from different suspensions. $L_w$ is the latent heat of fusion (calculated value) corresponding to the water content of the suspension, while $L_{mea}$ is the latent heat of fusion (measured value) obtained by thermal analysis. A correlation was observed between $L_w - L_{mea}$ and the logarithm of the coarsening rate constant. $L_w - L_{mea}$ represents the volume of water that did not form ice crystals on the addition of the suspended solid (volume of unfrozen water at $-10 \degree C$), with a larger $L_w - L_{mea}$ associated with greater inhibition of coarsening. The present findings suggest that suspended solids inhibit coarsening by promoting ice crystal melting.

1. INTRODUCTION

Coarsening (recrystallisation) of ice is the phenomenon in which small crystals melt and large crystals grow. Therefore, suppression of coarsening is an important issue for industries that handle ice such as the quality maintenance of frozen foods and storage of ice in ice thermal storage. Past studies have focused on antifreeze protein (AFP) as the substance that strongly suppresses the growth of ice crystals. For its suppression mechanism, the Gibbs–Thomson effect, wherein AFP adsorbs onto ice and lowers the melting point by increasing the curvature of the surface, is said to be effective.1 In addition, the suppression effect of AFP on the coarsening of ice has been confirmed and has also been indicated to be due to the Gibbs–Thomson effect.2,3 Meanwhile, suppression of crystal growth, coarsening, and nucleation of ice have been confirmed with various synthetic polymers.4,5 For example, polyvinyl alcohol is said to have a similar coarsening suppression effect as AFP and a high suppression effect has been shown when the large mean molecular weight and high degree of saponification is present.6 The coarsening of ice in low-molecular-weight aqueous solution showed that a solution with a higher concentration and lower diffusion coefficient of solute molecules had a lower coarsening.7 Among low-molecular-weight aqueous solution, there has been extensive research on aqueous solutions of sugar in ice cream. These studies have shown that the coarsening of the ice follows the Lifshitz–Slyozov–Wagner model8–10 and that the mechanism of coarsening is the melting and recrystallization of ice crystals depending on the holding temperature history.11,12 Furthermore, it has been suggested that the coarsening rate is independent of the type and concentration of sugars but depends on the spin–spin relaxation time ($T_2$) of water molecules13 and the glass-transition temperature ($T_g$) in the maximally freeze-concentrated state.14 There are many studies that target aqueous solution, but few studies have focused on the impact of suspended solids on the coarsening of ice in aqueous solution. For example, in the field of glaciology, there have been studies of coarsening when there are bubbles and solid particles in aqueous solution, but these studies target pure water.15 Thus, in the present study, silica particles and cellulose fibers were added in sucrose aqueous solution and ice crystals were allowed to coarsen at $-10 \degree C$. From the radius of these ice crystals, the coarsening rate constant using the Lifshitz–Wagner equation16,17 is obtained and the impact of the suspended solid on the coarsening of ice was examined. In addition, to understand the different coarsening trends based on the type and concentration of the suspended solid, the latent heat of fusion at $-10 \degree C$ was measured to determine using differential
scanning calorimetry (DSC). The results showed that the latent heat of fusion and coarsening rate constant presented similar dependencies on suspended solid concentrations. In order to clarify these causes, the correlation between \( L_w - L_{\text{mea}} \) and the coarsening rate constants obtained from different suspensions was investigated. \( L_w \) is the latent heat of fusion (calculated value) corresponding to the water content of the suspension, while \( L_{\text{mea}} \) is the latent heat of fusion (measured value) obtained by thermal analysis. As a result, it was found that there was a correlation between \( L_w - L_{\text{mea}} \) and the logarithm of the coarsening rate constant.

2. RESULTS AND DISCUSSION

2.1. Observation of Ice Crystals. Silica particles, 310P or 710, or cellulose fibers were suspended in 1.0 mol/kg sucrose aqueous solution and maintained for 6 h at \(-10^\circ C\). An image of ice crystals at \(-10^\circ C\) is shown in Figure 1. After 6 h, all ice crystals had coarsened. The ice crystal sizes, in the order of largest to smallest, were no additive \( \approx 710 > 310P > \) cellulose fibers. The addition of the suspended solid suppresses the coarsening of ice, but its degree varies depending on the type of suspension. Compositions of 310P and 710 were both silicon dioxide, and their mean particle size was the same; however, the impact on coarsening was notably different. Therefore, the coarsening behavior of ice with the addition of the silica particle does not simply depend on particle size but also on the structure of the suspended solid within the freeze-concentrated solution (difference in aggregation and diffusion).

In general, sedimentation volume measures the volume of the sedimentation layer after a certain period after the diffusion of the suspended solid and is used as the index of suspension characteristics. Usually, when the suspended solid forms a structure within the liquid, the sedimentation volume is high and when the suspended solid does not form a structure, the sedimentation volume is low. In addition, sedimentation volume is also a numerical value that reflects not only particle size and shape but also affinity to a dispersion medium (water). For reference, the sedimentation volume reported by the manufacturer (volume of the sedimentation layer when 5 g of particles is added) was compared. These values were 110 (mL/5 g) for 310P and 20 (mL/5 g) for 710. This indicates that, compared to 710, 310P is more widely distributed within the freeze-concentrated solution. Figure 2 shows a microscopic image of 310P within the freeze-concentrated state. Since the observation condition of the present study did not allow clear observation of suspended solids while frozen, 310P (5.0 wt %) was maintained at \(-10^\circ C\) for 6 h, and then, the image was presented taken after melting as a reference. The white circle in the image indicates where the ice was present, and multiple suspended particles are confirmed around this circle. Therefore, it is assumed that most suspended solids were present in the freeze-concentrated solution, impacting the coarsening from the surface of the ice.

Coarsening was done at \(-10^\circ C\) for 6 h, and the image was taken after melting. The white circle in the image indicates where the ice was present. The multiple suspended solids were confirmed around these circles.

2.2. Concentration Dependence of the Coarsening Rate Constant. When quantitatively evaluating the coarsening rate, the equivalent radius of ice was measured from the image analysis of ice crystals in each condition. Then, the coarsening rate constant, \( k \), was obtained from the Lifshitz–Wagner equation, as shown in eq 1

\[
r^3 = r_0^3 + kt
\]

where \( r \) is the radius of the ice, \( r_0 \) is the radius of ice with an elapsed time of zero, and \( t \) is the elapsed time. Conditions under which eq 1 can be used are when the crystal is spherical, distance...
to the adjacent crystal is larger than the particle diameter, and crystals are in equilibrium with their surroundings. The image of ice in Figure 1 shows that most of these conditions are met. Generally, the coarsening process of particles with time presents a proportional relationship between the square of the radius and time in the initial stage. Subsequently, the coarsening transitions to Ostwald ripening, wherein the cube of the radius is proportional to time. Figure 3 shows the relationship between the cube of the radius of ice crystals and elapsed time. Table 1 shows the coarsening rate constant and $r_0^3$ value obtained from this linear approximation. Figure 3 shows that, regardless of the type or concentration of the suspended solid, a linear
relationship is established. As such, it was shown that the coarsening behavior of ice with the addition of the suspended solid is Ostwald ripening. Compared to the coarsening rate constant for each suspension, it was found that when 2 wt % of cellulose fibers or 5 wt % of 310P was added, coarsening was mostly suppressed and the rate constant decreased by 1–2 orders of magnitude smaller than that of the sucrose aqueous solution. However, there was little change to the rate constant when 710 of any concentration was added; no value was notably different from the sucrose aqueous solution alone.

Furthermore, although it can be seen that the sucrose aqueous solution has the smallest \( r_0 \) value, this value tends to increase depending on the concentration of the suspended solid. That is, supercooling occurs readily when there are no suspended solids, which results in low \( r_0 \) because the ice nucleation temperature is low. However, when suspended solids are present, the value of \( r_0 \) is thought to increase because the ice nucleation temperature rises due to heterogeneous nucleation. The rate constant of Equation 1, \( k \), can be expressed with eq 2 as

\[
k = \frac{8\sigma_0 Dc_0 V_m^2}{9RT}
\]

where \( \sigma_0 \) is the interface energy of the freeze-concentrated solution and ice crystals, \( D \) is the diffusion coefficient of water molecules in the freeze-concentrated solution, \( C_0 \) is the equilibrium water concentration, \( V_m^2 \) is the molar volume of ice crystals, \( R \) is the gas constant, and \( T \) is the temperature. When considering the influence of the suspended solid on \( k \) with eq 2, as the suspended solid is water-insoluble, the influence on the diffusion coefficient of water molecules should be small. However, in terms of its impact on the interface energy, Ostwald ripening is a growth that uses the interface energy between the freeze-concentrated solution and ice crystals as the driving force. Therefore, considering that the decrease in \( \sigma_0 \) suppresses Ostwald ripening, it is possible that the existence of the suspended solid contributes to the decrease in \( \sigma_0 \). Figure 4 shows the concentration dependence of the coarsening rate constant. In the case of cellulose fibers and 310P, the rate constant does not monotonically decrease relative to the concentration, where there is a sudden decrease at a specific concentration. In other words, in the case of cellulose fibers, the rate constant clearly decreases at 0.5 wt %, but the decrease became more notable at 1.0–2.0 wt %. This trend was similar for 310P, where the rate constant notably decreased at a concentration of 3.0 wt % and higher. However, 710 had no clear concentration dependence.

Previous studies have confirmed the concentration dependence of additives in terms of coarsening suppression. For example, Hagiwara added type-I AFP to a sucrose aqueous solution by a method similar to the present study and maintained the solution at \(-10 \, ^\circ C\). He showed that with an additive of 1 \( \mu g/mL \) or more, suppression of coarsening could be observed. Furthermore, they indicated that if the coarsening suppression mechanism of AFP is adsorption to the ice crystal surface, \( 1 \, \mu g/mL \) is the amount equivalent to the minimum required to cover the surface. For example, Hagiwara added type-I AFP to a sucrose aqueous solution, the highest coarsening suppression was observed at 0.3 wt %. Therefore, the fact that the coarsening suppression was observed at a specific concentration of suspension indicates that the suppression mechanism of the suspension is similar to that of water-soluble polymers.

2.3. Concentration Dependence of Latent Heat of Fusion. Coarsening suppression by the suspended solid is assumed to have a direct impact on ice crystals. The measurement of latent heat of fusion is effective to clarify this point. In other words, the type and concentration of the suspended solid may lead to different amounts of ice crystals. Thus, under conditions similar to those used for the observation of coarsening, latent heat of fusion of suspension at \(-10 \, ^\circ C\) was measured and differences in the coarsening rate was discussed. Figure 5 shows the relationship between suspension concentration and latent heat of fusion for (a) a case where temperature was increased from \(-55 \, ^\circ C\) to room temperature and (b) a case where the temperature was maintained on the way at \(-10 \, ^\circ C\). Figure 5 also shows the latent heat of fusion (calculated value) equivalent to the amount of water in suspension at each concentration (dashed line) and the latent heat of fusion measured through thermal analysis (solid line). Figure 5a shows that when the temperature was increased immediately, there was no notable difference in the latent heat of fusion between suspensions and all plots are near the dashed line. Therefore, a decrease in the latent heat of fusion associated with increasing concentration (Figure 5a) indicates a decrease in water in response to the amount of added suspended solids. However, as shown in Figure 5b, when the sample was maintained at \(-10 \, ^\circ C\), the latent heat of fusion of cellulose fibers and 310P did not reach near the dashed line and there is a notable decrease compared to the dashed line at higher concentrations. In addition, when the latent heat of fusion was compared at the same concentration of the suspended solid, values were from smallest to largest, in the order of cellulose fibers < 310P < 710 ≈ no additive (sucrose aqueous solution). These results indicate that the latent heat of fusion varies greatly between different types of suspended solids.

The dashed line shows the latent heat of fusion (calculated value) equivalent to the amount of water in the suspension at each concentration. The solid line shows the latent heat of fusion measured through thermal analysis. When the sample was maintained at a low temperature, the impact of the suspended solid on ice crystals was limited. However, when the sample was maintained near the melting point, its impact was notable. Even a suspended solid could impact the melting of ice. The reason there were differences in
the impact of the suspended solid depending on the holding temperature may be as follows. When an aqueous solution is frozen, the freezing rate (the ratio of water that crystalized relative to the amount of water in the suspension) is determined depending on the temperature reached. Therefore, the latent heat of fusion in Figure 5 presents the freezing rate at $-55^\circ C$, but in this case, the freezing rate is higher than that when the sample was maintained at $-10^\circ C$. Since the suspended solid is strongly concentrated in the freeze-concentrated solution, its impact on ice crystals is reduced and the difference in the latent heat of fusion between suspensions is also reduced. However, when the sample is maintained at $-10^\circ C$, the freezing rate is lower than that at $-55^\circ C$ and the freeze-concentrated is less prominent. Therefore, the suspended solid was widely dispersed within the concentration phase and the direct impact on ice crystals increased. This, in turn, lowered the latent heat of fusion. The reason the latent heat of fusion varied between suspensions was likely due to the suspended solid structure within the freeze-concentrated solution (difference in aggregation and dispersion). In other words, compared to 310P, 710 has a lower sedimentation volume; thus, 710 was densely packed during freezing and did not redisperse when the temperature was maintained at $-10^\circ C$. This likely minimized the impact of 710 on the ice crystals. On the other hand, the concentration dependence of the coarsening rate constant (Figure 4) and the latent heat of fusion (Figure 5b) is similar. In other words, the addition of the suspended solid reduced ice crystals, which tended to suppress the coarsening accordingly. Though detailed data are not shown here, sample temperatures during DSC measurements remained within the range of $-9.4$ to $-9.6^\circ C$, thus, temperature fluctuation during the measurements was limited. The latent heat of fusion after additional 3 h of elapsed time was not different from that after 1 h of elapsed time. Therefore, the latent heat of fusion obtained with this measurement method has little impact, such as temperature fluctuations.

2.4. Relationship between the Coarsening Suppression Trend and Latent Heat of Fusion. These results show that the suspended solid with notable reduction in latent heat of fusion, such as cellulose fibers and 310P, strongly suppresses coarsening. Meanwhile, suspensions with limited reduction in latent heat of fusion, such as 710, had a limited suspended solid effect. This indicates that a suspended solid that reduces ice crystals has a stronger suppression effect on coarsening. In other words, when the difference between the latent heat of fusion equivalent to the amount of water in the suspension, $L_w$, and latent heat measured through thermal analysis, $L_{mea}$, $(L_w - L_{mea})$ is larger, coarsening is suppressed. Figure 6 shows the relationship between $L_w - L_{mea}$ and the coarsening rate constant, which indicates that a correlation is established regardless of the type and concentration of suspension. This correlation indicates that $L_w - L_{mea}$ increases with the increasing concentration of cellulose fibers and 310P and the coarsening rate constant decreases accordingly. 710 has a small $L_w - L_{mea}$ regardless of the concentration, and the coarsening rate constant barely changes. On the other hand, the freezing rate of an aqueous solution is derived from the melting equilibrium of a thermodynamically

Figure 5. Concentration dependence of the latent heat of fusion. (a) Case where temperature was immediately raised from $-55^\circ C$ to room temperature. (b) Case where the temperature was maintained on the way at $-10^\circ C$.

Figure 6. Relationship between the difference between latent heat of fusion equivalent to the amount of water in the suspension, $L_w$, and latent heat measured through thermal analysis, $L_{mea}$, $(L_w - L_{mea})$ and the logarithm of the coarsening rate constant.
ideal solution and its approximation can be expressed by Heiss’ equation, as shown by eq 3

\[ X = 1 - \frac{T_m}{T} \]  

(3)

where \( X \) is the freezing rate of the aqueous solution, \( T_m \) is the melting point of ice in the freeze-concentrated solution, and \( T \) is the holding temperature. The temperature here is not the absolute temperature but the temperature in degrees Celsius. Therefore, the nonfreezing rate is expressed by eq 4

\[ 1 - X = \frac{T_m}{T} \]  

(4)

Considering the relationship between eq 4 and the latent heat of fusion, \( X \) is \( L_{m,es} \) of the aqueous solution measured by thermal analysis and the total amount of water in suspension, \( L_w \), is equal to one; therefore, \( T_m/T \) is equivalent to \( L_w - L_{m,es} \). In other words, the more the melting point depression, the more the \( L_w - L_{m,es} \) increases, resulting in a lower coarsening rate constant. On the other hand, the Gibbs–Thomson effect has been indicated as the reason AFP suppresses growth of ice crystals. When molecules such as AFP are adsorbed on the interface with ice, the curvature of the interface increases and the melting point of ice locally decreases. This, in turn, suppresses the growth of ice crystals. The Gibbs–Thomson effect can be expressed by eq 5

\[ T_m(d) = T_m \left(1 - \frac{4\sigma_d}{d\Delta H f T} \right) \]  

(5)

where \( T_m(d) \) is the melting point of ice crystals with diameter \( d \), \( T_m \) is the melting point of the bulk body (in other words, an ice crystal with \( d = \infty \)), \( \sigma_d \) is the interface energy of the ice crystals and the freeze-concentrated solution, \( \Delta H_f \) is the latent heat of fusion of ice crystals (the bulk body), and \( \rho_s \) is the density of ice crystals. The essence of this study is not crystal growth but the coarsening of ice. Moreover, the suspended solid is water-insoluble. Thus, it is unclear if the melting point depression is by the same mechanism. However, if it is assumed that a decrease in the latent heat of fusion by the addition of the suspended solid is the Gibbs–Thomson effect, it is assumed that \( \sigma_d \) contributes to melting point depression since \( \Delta H_f \) is a physical property. In other words, it is possible that \( \sigma_d/\Delta H_f \rho_s \) is equivalent to \( X \) in eq 4. This shows that there is a relationship between a decrease in the interface energy by suspension addition and a depression in the freezing rate. On the other hand, particles in the suspensions used in the present study were all on the order of a micron; thus, it is unlikely that the suspended solid directly lowered the interface energy. Therefore, the surface morphology of a suspended solid is likely to have caused the decrease in \( \sigma_d \). That is, the diameters of cellulose fibers are in the range of 10–50 nm, and the average pore diameters of 310P and 710 are 21 and 2.5 nm, respectively. Therefore, it is thought that the coarsening rate decreases due to the ice melting when a suspended solid with a specific (on the order of 10 nm) surface morphology touches the ice.

3. CONCLUSIONS

The coarsening rate constant of ice crystals in sucrose aqueous solution, with added silica particles or cellulose fibers maintained at \(-10 \, ^\circ\text{C}\), was obtained by using the Lifshitz–Wagner equation. The results showed that, regardless of the presence or absence of the suspended solid, the coarsening of ice in the sucrose aqueous solution exhibited Ostwald ripening; with the addition of the silica particle, the coarsening rate constant decreases, though this trend does not depend on particle size; compared to silica particles, cellulose fibers more notably decreased the coarsening rate constant. On the other hand, when the latent heat of fusion of different suspensions maintained at \(-10 \, ^\circ\text{C}\) at the same concentration was compared, the values were in the following order: cellulose fibers <310P < 710 \approx \text{no additive} (\text{sucrose aqueous solution}). Thus, the addition of the suspended solid lowers the latent heat of fusion. The amount of latent heat of fusion reduction was consistent with the coarsening rate constant. Furthermore, the correlation between \( L_w - L_{m,es} \) and the coarsening rate constants obtained from different suspensions was investigated. As a result, the differences in the coarsening rate constant by the additions of the suspended solid could be uniformly interpreted by \( L_w - L_{m,es} \). Meanwhile, the decrease in \( L_{m,es} \) was considered to be due to a depression in the melting point by the addition of the suspended solid. The cause of the melting point depression was likely due to the Gibbs–Thomson effect.

4. EXPERIMENTAL SECTION

4.1. Preparation of Suspensions. The cellulose fibers (Sugino Machine Limited: BMa-10002) were added to 1.0 mol/kg sucrose aqueous solution at the concentrations of 0.5, 1.0, and 2.0 wt %. Similarly, silica particles (Fuji Silysia Chemical Limited: Sylsysia 310P or Sylsysia 710) were added to 1.0 mol/kg sucrose aqueous solution at the concentrations of 1.0, 3.0, and 5.0 wt %. The prepared suspension was diffused using an ultrasonicator immediately before measurement. Cellulose fibers used in the present study were obtained from plant cell walls that were physically processed. Although it is difficult to precisely measure the size of cellulose fibers because it is on the order of nanometers, the fiber diameter and fiber length are estimated by electron microscope observation to be 10–50 nm and 20 \( \mu \text{m} \), respectively. Silica particles were synthetic silica particles with 99% \( \text{SiO}_2 \), and the average particle size was 2.8–27 \( \mu \text{m} \), which are usually used to prevent pigment precipitation. The specific surface areas of 310P and 710 determined by the Brunauer–Emmett–Teller method are 300 and 700 \( \text{m}^2/\text{g} \), respectively, and the average pore diameters are 21 and 2.5 nm, respectively. Therefore, although the average particle diameter is the same, the surface morphology is different.

4.2. Observation of Ice Crystals. 1 \( \mu \text{l} \) of the suspension and a 15 \( \mu \text{m} \) thick stainless shim ring (Iwata MFG. Co., Ltd.: RS012016001) were placed between two cover glasses with thicknesses ranging from 0.012 to 0.017 mm and a diameter of 16 mm. The cover glasses were placed on the cooling stage of a microscope (Linkam Scientific Instruments: HP95). The temperature of the stage was cooled to \(-60 \, ^\circ\text{C}\) at a rate of \(-90 \, ^\circ\text{C}/\text{min}\), followed by immediately warming to \(-10 \, ^\circ\text{C}\) at a rate of \(+5 \, ^\circ\text{C}/\text{min}\), which was maintained for 6 h. Images were captured every 60 min using a digital microscope (KEYENCE Corporation: VHX-900).

4.3. Measurement of Ice Crystals. Image analysis software (Mitani Corporation: WinRoof 2013) was used to analyze the equivalent radius of the ice crystals. Then, the coarsening rate constant was calculated from the obtained equivalent radius, \( r \), using the Lifshitz–Wagner equation.

4.4. Measurement of Latent Heat of Fusion by Thermal Analysis. DSC (Shimadzu Corporation: DSC60) was used to determine the latent heat of fusion. The sample amount was 25.0 \( \pm \) 0.1 mg, and an empty aluminum cell was used as a reference. The temperature was cooled to \(-55 \, ^\circ\text{C}\) at a
rate of −5 °C/min, which was maintained for 10 min. Then, the sample was heated to −10 °C at a rate of +5 °C/min, which was maintained for 60 min. Subsequently, the sample was heated to 25 °C at a rate of +5 °C/min and the latent heat of fusion was then measured. Under the condition without the maintaining period at −10 °C, the temperature was increased from −55 °C to room temperature at a rate of +5 °C/min and the latent heat of fusion was then measured.

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Notes

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