## 5. Observation of freezing of dispersed glass beads in water (Exp. 3)

In this chapter, an ice growing surface in water with dispersed glass particles was directly observed by using unidirectional freezing apparatus equipped with microscope, to clarify how the growth surface and each particle behave and what is the factor that affect to the behavior. In order to observe the behavior of each particle, the microscope of this experiment had higher resolution than that used in the **Exp. 1** and **2**

### 5.1 Sample and method

Samples used here were three types of glass particles with different diameters of 2.2, 5.3 and 9.7 μm, which were mixed with distilled water and deaired. Water content of each mixture prepared was 1000g-water per 1g-glass particles. In this experiment, sample cell had polyester spacers with thickness of 100 μm instead of 3 mm-thick spacer shown in figure 14. The mixture was poured into the sample cell by capillary attraction. After the temperature gradient of 0.33 ˚C/mm was attained to the sample for half an hour, the cell was moved at a constant rate by the unidirectional freezing apparatus. Then the vicinity of ice growing surface was observed with a microscope (100 magnification) until the cell was moved 5-mm distance. Table 3 lists the condition of the sample moving rate $V_s$ and the particle diameter d. The experiments were performed in the cold room with ambient temperature of 5.0 ˚C.

Table 3. Experimental conditions (**Exp. 3**).

| Sample | d (μm) | $V_s$ (μm/sec) | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 2 | 9.7 | | | 4.0 | | 3.0 | | 2.0 | 1.5 |
| 3 | 5.3 | 5.0 | | 4.0 | | 3.0 | 2.5 | 2.0 | |
| 4 | 2.2 | 5.0 | 4.5 | 4.0 | 3.5 | 3.0 | 2.5 | 2.0 | |

## 5.2 Experimental results

Figure 27 shows the vicinity of ice growing surface in water with dispersed glass particles. In this figure, white points are glass particles and dashed line indicates the growth surface of ice. The right side of a dashed line is frozen area and the left is unfrozen. In this experiment, the growth surface of ice stayed at the same location in the visual field of microscope. The behavior of the growth surface showed that the growth rate of ice was equal to the freezing rate and that the growth surface of ice kept the same temperature.

Two kinds of ice formations were observed in this experiment. Ice grew with pushing particles ahead at a low freezing rate (figure 27a) while it grew with encapsulating particles at a high freezing rate (figure 27b). The former ice formation shown in figure 27b might correspond to ice lensing in soil or fine particles bearing water. The latter ice formation shown in figure 27b might correspond to pore ice formation in soil or fine particles bearing water.

Table 3 lists freezing rates for three kinds of glass particles to examine criteria whether particles were encapsulated in ice or not. Underlines denotes the freezing rate that particles were pushed ahead by ice. For the sample 2, ice grew with encapsulating particles at the freezing rates of 4.0, 3.0 and 2.0 μm/sec while ice grew with pushing particles ahead at the rate of 1.5 μm/sec. The critical rate $V_c$ was affected by the particle size.

In the freezing as shown in figure 27a, it was also observed that particles, which had been pushed by ice growing surface, accumulated on the ice surface. And a new ice often started to grow beyond the layer of accumulated particles, which had a certain thickness.

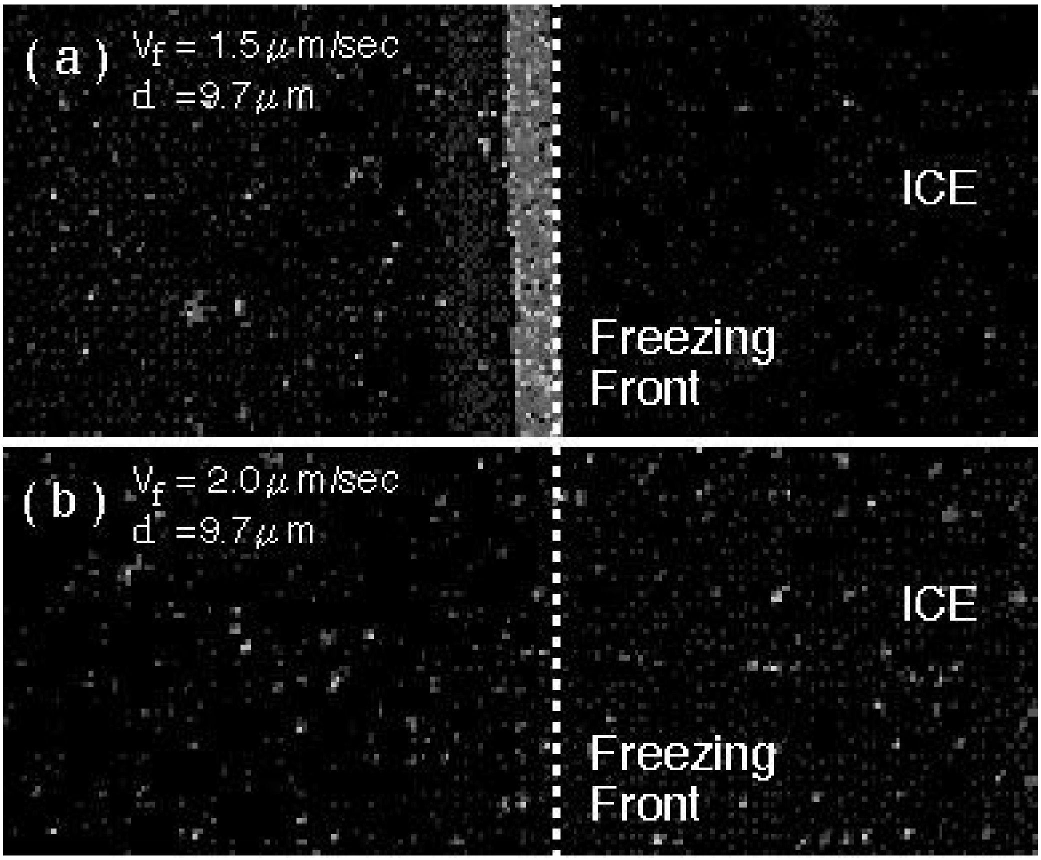


Fig. 27. An image of the vicinity of ice growing surface in water with dispersed glass particles. The white points are glass particles and dashed line indicates the growth surface of ice. The right side of a dashed line is frozen area and the left is unfrozen. Ice grew with pushing particles ahead at a low freezing rate (a) while it grew with encapsulating particles at a high freezing rate (b).

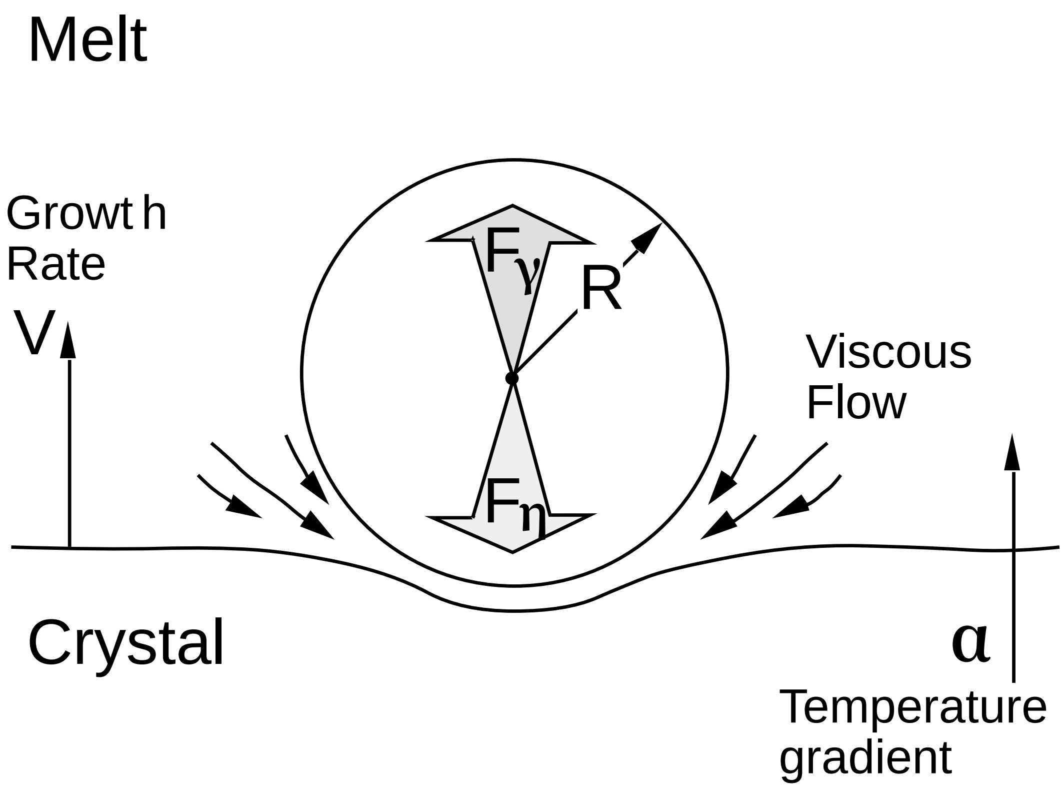


Fig. 28. Physical factors on a spherical particle ahead of the growing surface of ice crystal.

5.3 Discussion

The relationship of particle diameter, freezing rate and solute concentration to the freezing of water containing particle has studied by Chernov *et al*. (1977), Pötschke and Rogge (1989), Körber *et al*. (1992). Körber *et al*. has shown the criterion for such a particle trapping phenomena. According to their theory, an isolated particle pushed to an advancing ice front is subject to two counteracting forces (figure 28): an attractive and a repulsive force. The attractive force comes from viscous drag due to fluid flow around the particle, which favors entrapment (d <<R):

$$F_\eta = 6\pi\eta V_g R \frac{R}{d - a_0} \quad , \tag{26}$$

where $\eta$ is coefficient of viscosity, $V_g$ is the rate of ice growing, $a_0$ is average molecular distance in liquid, d is the distance between ice growing surface and a particle, and R is radius of the particle. The repulsive force originates from van der Waals forces:

$$F_\gamma = 8\pi a_0^2 \, \Delta\gamma \frac{R^3}{d^2 \, (d+2R)^2} \quad , \tag{27}$$

where $\Delta\gamma$ is the difference of surface energy between particle-liquid water and liquid water-ice. The balance of the two forces determines whether the particle is encapsulated or not. When $F_\gamma > F_\eta$, ice will grow without encapsulating of a particle. In this case, liquid water film between the particle and ice growing surface will keep constant thickness. If we assume that these forces are equal (equation (26) = (27)), critical freezing rate $V_c$ whether the particle is encapsulated or not would be given by

$$V_C = \frac{4 \, ( d - a_0) \, a_0^2}{3\eta \, d^2 \, (d + 2R)^2} \Delta\gamma \, R \quad . \tag{28}$$

The critical freezing rate $V_c$ is increased with increasing the particle radius R.

Our experimental results can be explained by Körber's theory, although the sample 4 was out of range to apply it. It can be considered that ice formation with pushing particles ahead is essentially the same ice formation as shown in ice lensing observed in **Exp. 1** and **2**. In fact, we often found that a new ice started to grow beyond the layer of accumulated particles shown in Figure 27a after the particles had accumulated to a certain thickness. Although details of the phenomenon were not examined, the new ice growth in the **Exp. 3** must be related to the ice lensing in **Exp. 1** and **2**. Therefore, it is suggested that the critical rate whether ice lensing will occur or not, can be derived from the critical freezing rate $V_c$. However, this theory deals with the forces around only one particle. In soil or micro-particles, where particles interacts each other, multi-body effect should be considered. Since these reasons, equation (28) does not explain the ice lensing as it is.

## 5.4 Summary

In this chapter, the behavior of growth surface of ice and particle near the surface was observed by using the unidirectional freezing apparatus. The criteria for exclusion and encapsulation of glass particles during ice formation with respect to particle size and freezing rate was shown. The relations of particle size and freezing rate to the critical freezing rate was explained by Körber's theory. Since the ice lensing is considered to be essentially same as the ice formation with pushing particle ahead, it is suggested that critical freezing rate is important factors to make ice lensing model.