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# Particle Migration on Ice Surfaces

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## Abstract

A novel phenomenon is described which could indicate a new mechanism of mass flow along an ice surface. Glass beads evenly and randomly scattered on an ice surface migrate and tend to form pronounced clusters when the ice surface is exposed to an unsaturated atmosphere. Time lapse motion pictures reveal two types of migration of glass beads, smooth continuous and intermittent. Both types of migration tend to form beads clusters. No movement is observed when the atmosphere is saturated with respect to ice.

Conventional mechanisms of surface mass flow such as evaporation-condensation and surface diffusion do not appear to explain the migration. Volume diffusion in ice seems to be too slow a process, and liquid film flow is a rather improbable mechanism. Enhanced diffusivity of the surface layer is one possible mechanism.

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## I. Introduction

The surface of ice sometimes shows peculiar phenomena such as regelation. Regelation has been discussed as evidence of a surface liquid layer by Faraday (1860), Tyndall (1858) or as proof of pressure melting by Thomson (1859). Nakaya and Matsumoto (1953) found that two ice spheres, suspended by strings and in contact, rotated without separation when they were pulled apart. They attributed the phenomenon to the existence of a liquid layer. From observations of the motion of a wire through ice Telford and Turner (1963) were inclined to conclude that a fluid layer does exist on the ice surface.

The following experiments could supply new knowledge about the ice surface. Particles on the subliming ice surface migrate along the surface and tend to coagulate into clusters. Though the migration resembles the motion of particles suspended on a water surface, several experimental results seem to indicate that mass flow in the relatively thick high diffusivity layer could be one possible mechanism. Some of the aforementioned phenomena could be explained by the high diffusivity layer.

## II. Experimental Procedure

### *Sample preparation*

The samples used in most of the experiments were ice single crystals produced in the Mendenhall Glacier. The crystal is oriented by the reflection of light from frost crystals grown on the ice surface (Nakaya, 1956), then cut by a band saw to the desired orientation using a goniometric jig. The ice slabs thus cut are frozen onto 2×3 inch glass slides applying slight heat. The top surface of the ice sample is then microtomed.

When the basal or prismatic plane is desired, the ice is oriented to obtain a more accurate plane by the frost, using a microscope with vertical illumination and adjustable microtome stage. The accuracy is within 0.3 degree for the basal and prismatic planes and about two degrees for the other orientations.

Glass beads, from 10 to 500  $\mu$  in diameter, supplied by Cataphote Corporation, Toledo, Ohio and Lapine and Company, Chicago, Illinois, are used. The glass beads are generally perfect spheres; however, their surfaces are covered by particulate dirt. Water used to wash the beads showed a high electrical conductivity, indicating that some of the dirt is water soluble and electrolytic. The glass beads are spread evenly and randomly on the ice surface immediately after the microtoming.

Two techniques are used to spread the glass beads. The method generally used is to shake the beads onto a piece of urethane resin foam and keep them in the pores until they are fillipped off onto the ice surface. The second method is used when contamination of the glass bead surface must be avoided or the beads are sticky because of surface treatment. The desired amount of beads is put into a shallow vessel such as the cap of a glass jar. A sharp fillipping blow on the bottom of the vessel about 10 cm aside from the ice surface spreads the beads evenly and randomly on the surface.

*Experiment 1: Untreated glass beads*

The general features of glass bead migration can be seen in Figs. 1 a-d which are obtained from a time lapse motion picture. The temperature was about  $-11^{\circ}\text{C}$  and the humidity measured by a dew-point hygrometer shows frost point of about  $-16^{\circ}\text{C}$ . A

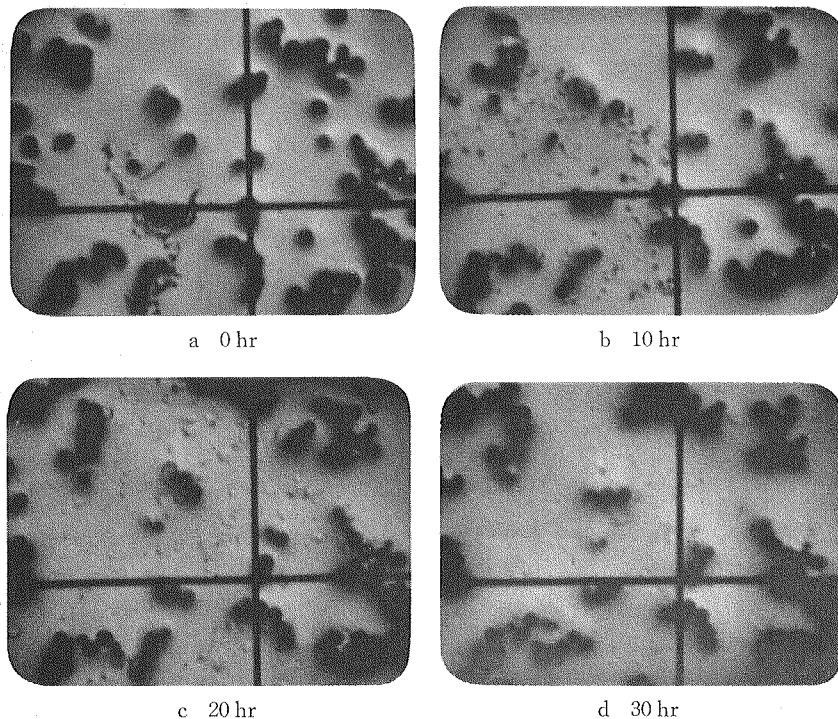


Fig. 1. Migration of untreated glass beads. ( $\times 55$ )

slight motion of the air was indicated by the movement of tobacco smoke.

The migration can be classified as three types. The steady motion of individual and clustered beads is observed generally, while there is a tendency for the single beads and clusters to come together and form larger clusters. This type of movement resembles the movement of particles on a water surface supported by surface tension. The third type is an intermittent motion. This can be expected if the supporting neck of the bead is broken or bent by its own weight.

A side view of the migration of the beads is shown in Figs. 2a-d. Three beads designated A, B and C are seen close to the cross hair sticking to the left surface of an ice block which appears as a dark field. Bead A, with a smaller bead sticking to it, serves as a good reference mark for rotation. The thin neck supporting bead A can be seen in Fig. 2a. Beads A and B migrate toward the top of the photographs as the ice surface recedes while C migrates toward the bottom of the picture until it disappears. Although gravitational force is acting perpendicular to the photographs no preferential migration occurs in the direction of the gravitational force because beads A and B remain in focus.

Note that the smaller beads on A remain in the same position, which shows that little rotation occurs. It is observed from the time lapse motion picture that A moves slowly but steadily while B moves rather intermittently.

Small specks appear on the ice surfaces after the microtome traces disappear; these did not exist when the glass beads were spread. Some of the specks could be related

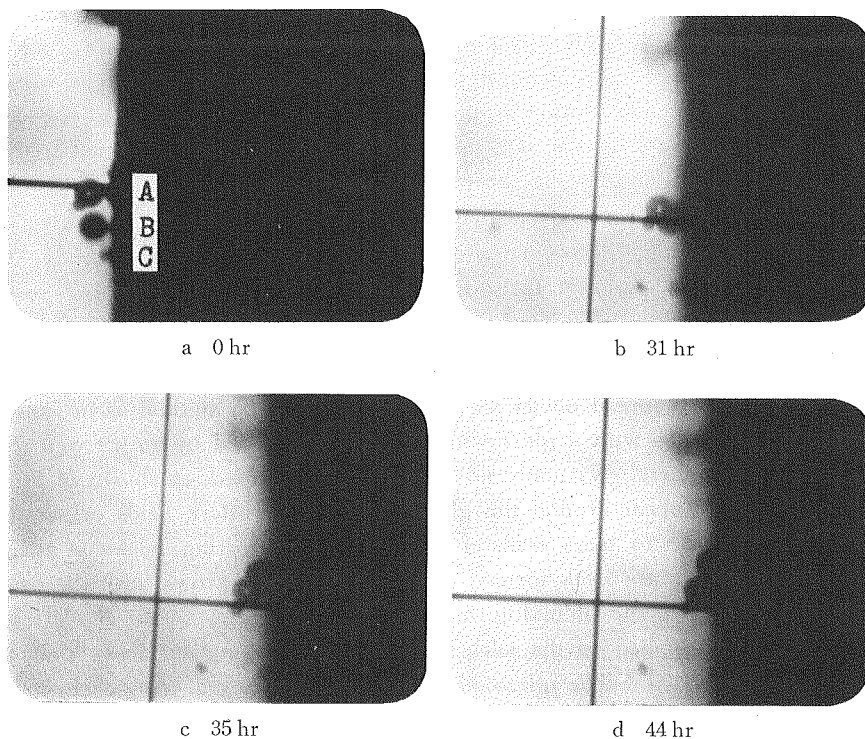


Fig. 2. Side view of migration. ( $\times 55$ )

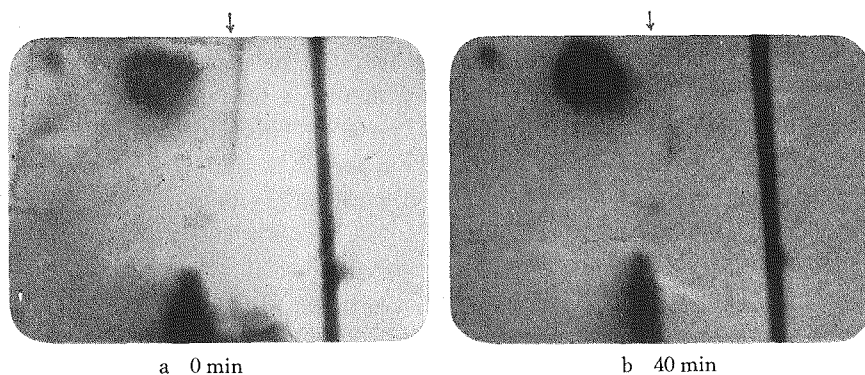


Fig. 3. Elongated speck. ( $\times 170$ )

to the microtomed traces, which disappear during the sublimation of the surface. They are rather uniform in size, but sometimes elongate to a short line as shown in Fig. 3 a. Figure 3 b shows the same field as Fig. 3 a 40 minutes later. The short line tends to resume its original shape.

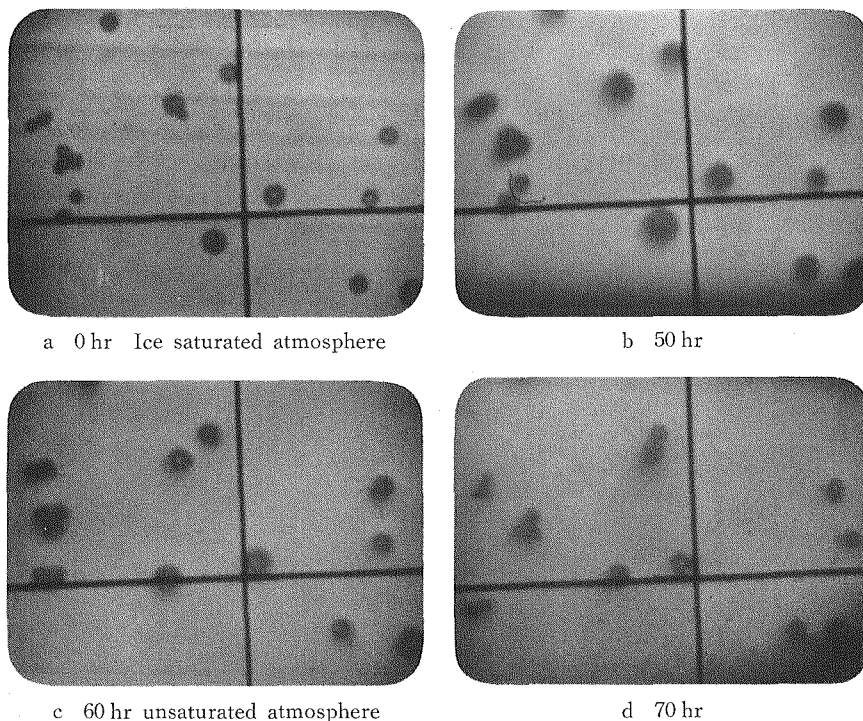
Removal of a surface layer about  $200\sim 500\mu$  thick by sublimation is required to produce these specks depending on the surface treatment and orientation. A very flat surface usually appears after the traces of microtoming or lathing disappear. Etch patterns which resemble the etch channels studied by Kuroiwa (1965) appear after  $200\sim 500\mu$  of sublimation. These patterns spread in a relatively short time and become individual specks. The final density of the specks is about  $10^5/\text{cm}^2$  which is the same order of magnitude as the dislocation density obtained by Muguruma and Higashi (1961, 1963).

Two typical modes of movement of these specks are 1) rather straight with occasional turning, and 2) circular motion around a fixed point. The number of specks does not increase after the final density is attained, indicating that the foreign particles deposited on the ice surface cannot be the main cause of these specks. No clustering of the specks is observed. These facts lead us to speculate that the specks are related to dislocation. Assuming that the specks are dislocation etch pits, their movement shows the time display of the sections of three-dimensional dislocation structures, because the ice is receding by sublimation.

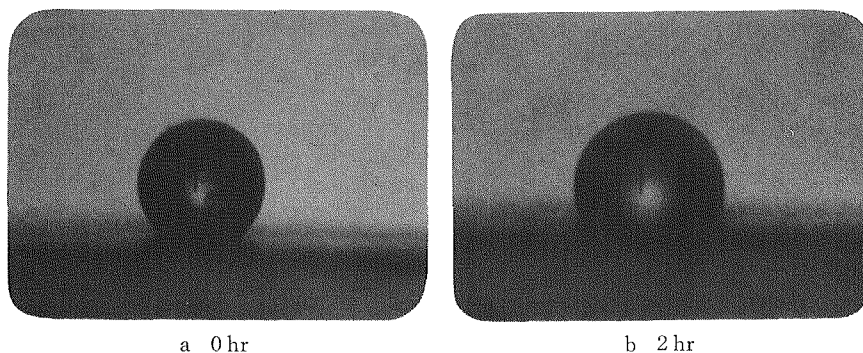
#### *Experiment 2: Experiment in an ice-saturated atmosphere*

The glass beads are spread on an ice surface by the same method as in experiment 1, then enclosed in a case with a glass window and surrounded by an ice wall to make an ice-saturated atmosphere. Virtually no migration is observed, as shown in Figs. 4 a-b, though dark fields appear around the glass beads. These dark fields originate from surface modification due to mass transfer toward the supporting necks of the beads. The large negative-curvature neck formed in the initial stage grows until the beads are partially buried by ice, as shown in Fig. 5.

The material is supplied to the neck from the surrounding surfaces; thus the ice surface is metamorphosed. The glass beads buried under the ice cover could not move unless they were exposed to some driving force such as a temperature gradient (Hoekstra, 1963).



**Fig. 4.** Effect of humidity. ( $\times 55$ )



**Fig. 5.** Partially immersed beads in ice saturated atmosphere. ( $\times 170$ )

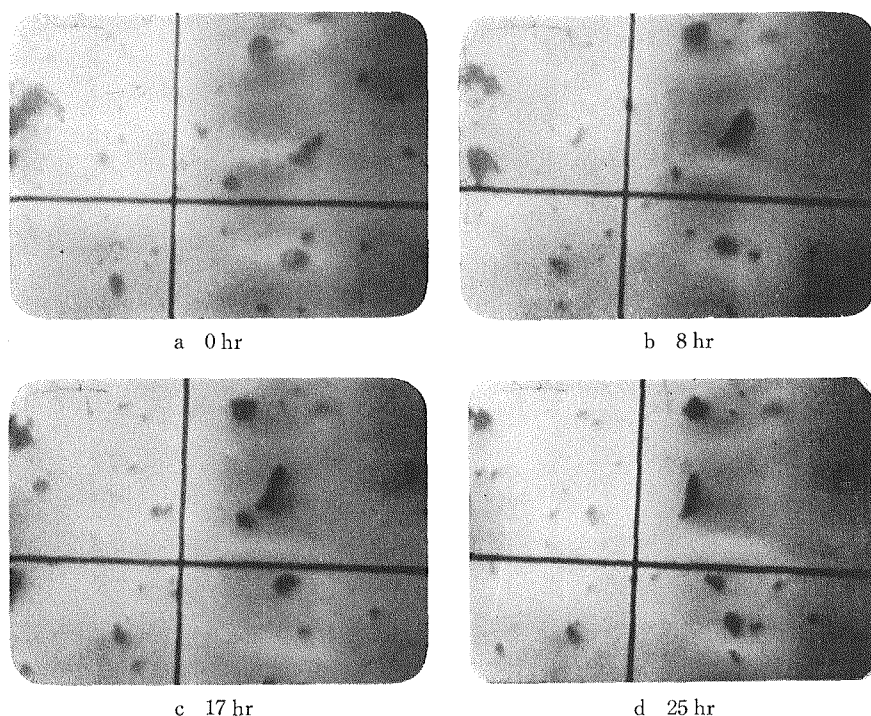
The migration of the glass beads starts only one hour after the removal of the ice in case as shown in Figs. 4c-d. The field is the same as Figs. 4a-b. The dark field around the beads reduces to a small shadow for the individual beads though it still remains between the beads constituting clusters.

*Experiment 3: Effect of surface treatment*

As mentioned earlier, considerable dirt is found on the surface of the glass beads as received. Some of this is water soluble substances. There is a possibility that these water soluble substances could dissolve into the ice surface and produce a liquid layer because of melting point depression and that the glass beads are suspended on the

liquid layer by surface tension. If this is the case the migration of the beads can be attributed to the same mechanism that moves particles suspended on a water surface by surface tension.

Glass beads are washed by bichromate-sulfuric acid and then by deionized water until the electrical conductivity of wash water becomes 1 micromho. Then they are dried in vacuum and spread on a microtomed ice surface. The migration of glass beads thus treated appears the same as that of the untreated beads, indicating that the impurity of the glass bead surface is not the major cause of glass bead migration. This is confirmed by other experiments. Larger glass beads are crushed above the microtomed ice surface as shown in Fig. 6. Most of the shards thus produced are scattered on the ice surface directly, thus there is little chance of contamination. As shown in Figs. 6a-d, the shards migrate as well as the glass beads though their shape is irregular.



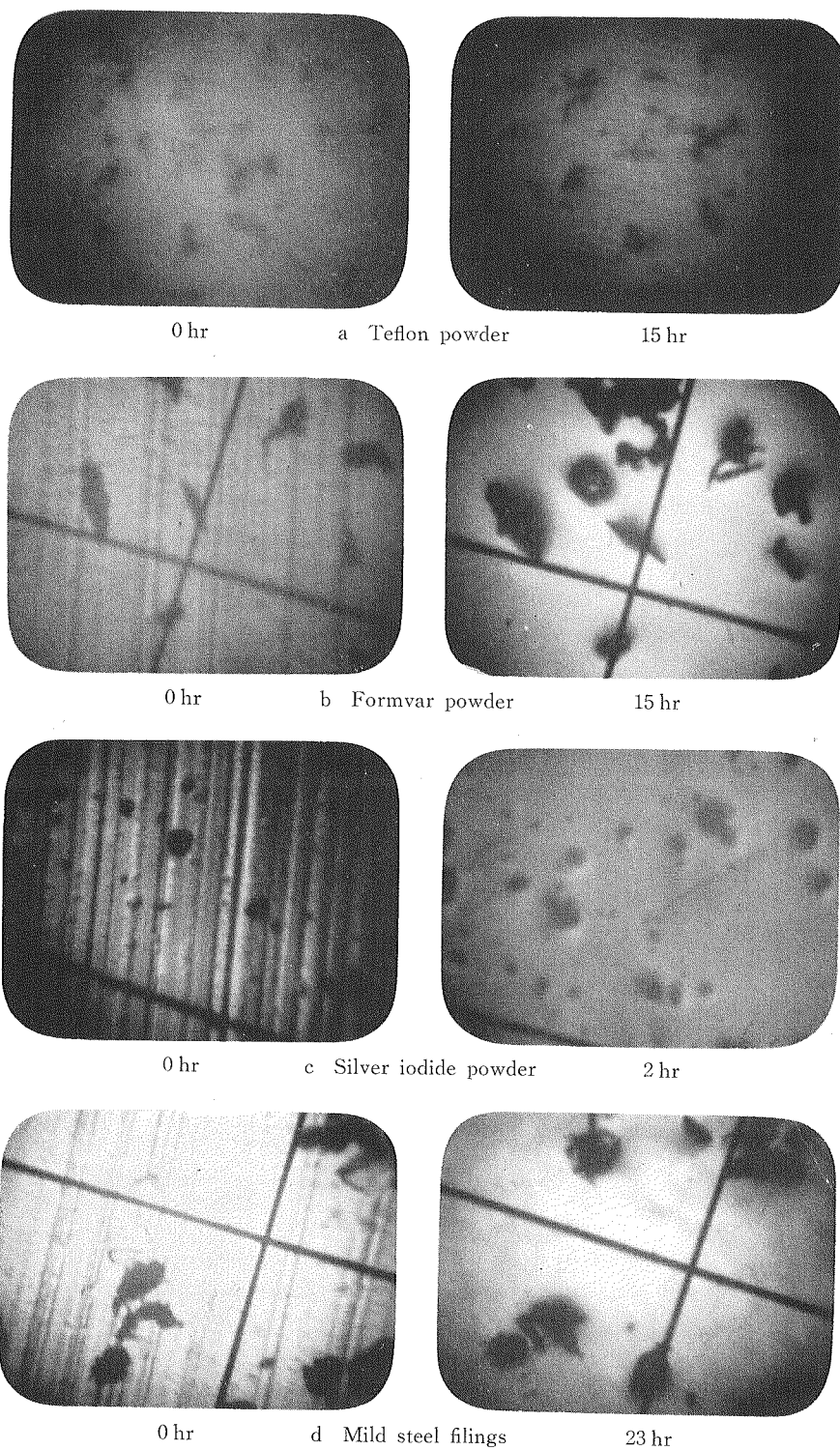
**Fig. 6.** Migration of irregular shaped shards. ( $\times 55$ )

After washing with deionized water, glass beads are coated with a very thin film of silicone grease by immersing them in a dilute toluene solution of DC-7 and then vacuum dried. The DC-7-coated glass beads migrate and form clusters like the untreated or bichromate-sulfuric acid treated beads; however, time lapse motion pictures reveal that the migration is rather intermittent. This is in contrast to the clean-surfaced glass beads such as those treated with bichromate-sulfuric acid or shards, which migrate continuously. The cause is not known.

#### *Experiment 4: Migration of particles other than glass beads*

To determine the effect of different materials on migration, experiments were





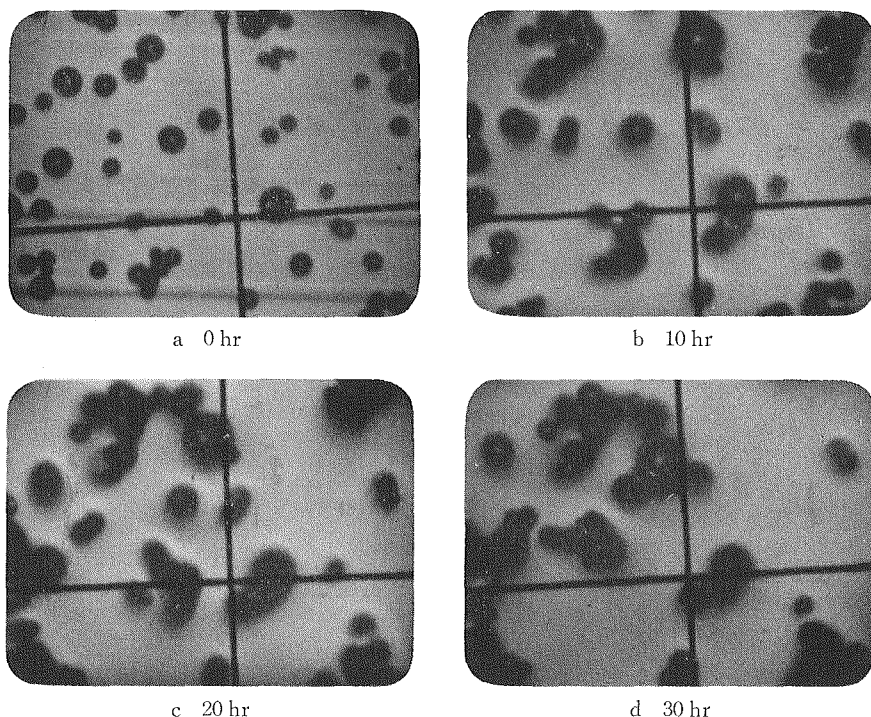
**Fig. 7.** Migration of various kinds of particles. ( $\times 55$ )



performed using Teflon powder, Formvar powder, silver iodide particles, filings of mild steel, cotton filament and carbon black. All materials migrated though there might be differences in migration velocity. The migration velocity is very difficult to define because the movement is random and the temperature and humidity in the room are not kept constant. Figures 7 a-d show migrating particles of various materials.

*Experiment 5: Migration of glass beads sticking to a downward-facing ice surface*

If migration is caused by the breaking or bending of the neck by the weight of the bead, the beads would not migrate when the ice surface is facing downward. Figures 8 a-d show the migration in this case. Note that no beads are lost. Time lapse movies show that movement is rather intermittent as with the DC-7-treated beads. The beads are probably contaminated by a small amount of grease. Intermittent migration probably caused by contamination of beads.



**Fig. 8.** Migration on a downward facing surface. ( $\times 55$ )

*Experiment 6: Effect of particle size on migration*

Rate of migration seems to depend on the size of the bead. Smaller single beads move actively while clusters and larger beads migrate slowly. As seen in Figs. 9 a-d, the tendency of smaller beads to migrate toward larger ones could indicate mass flow towards each bead by an unknown mechanism.

*Experiment 7: Migration of other than spherical particles*

Shards of larger glass beads crushed by a pair of pliers and spread on an ice surface migrate as fast as beads of the same size stated in experiment 3. As shown in Figs.

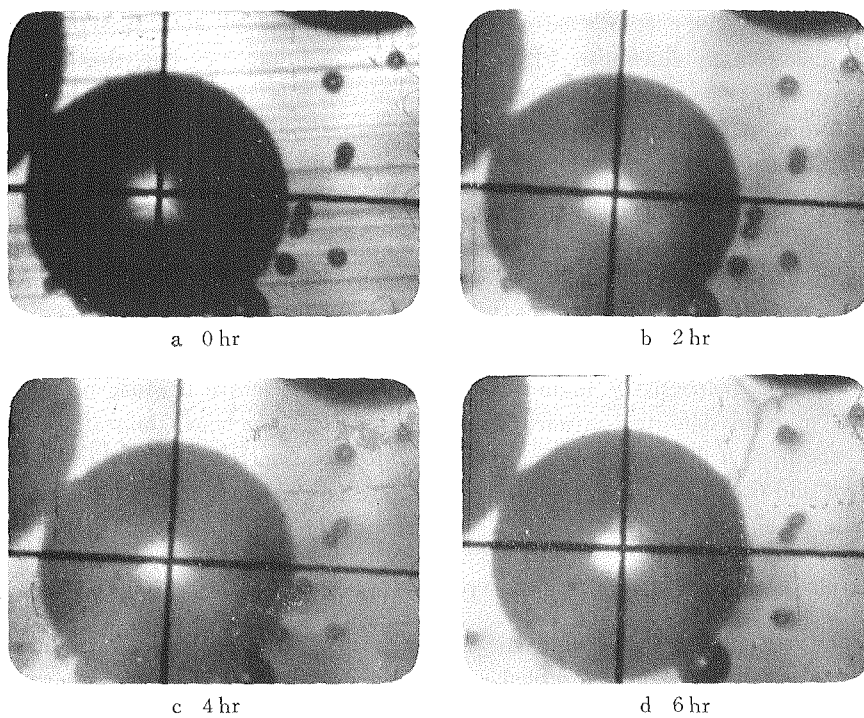


Fig. 9. Large and small beads

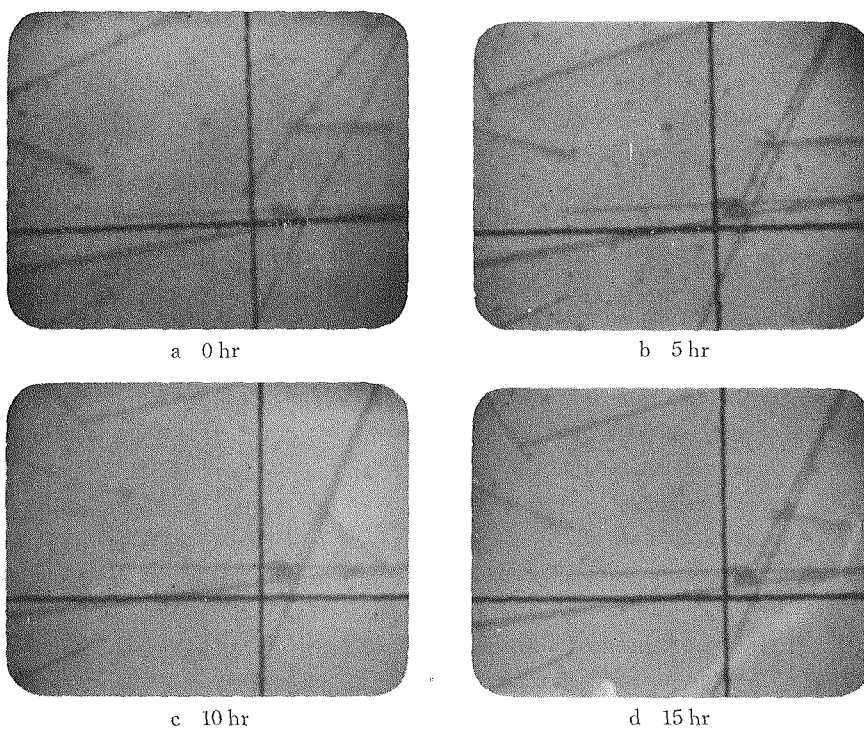


Fig. 10. Migration of glass fiber. ( $\times 55$ )

6 a-d long shards move somewhat sidewise. Short pieces of glass fiber also migrate somewhat sidewise as shown in Figs. 10 a-d. As mentioned earlier, the larger beads migrate more slowly. Lengthwise migration corresponds to migration of the larger beads and thus will be slower, while sidewise migration corresponds to migration of the smaller beads and thus will be faster.

*Experiment 8: Migration under a temperature gradient*

A temperature gradient was produced along the ice surface by sandwiching the ice sample between metal plates, one of which has a small electric heater. Experiments were made under temperature gradients of about 0.5 and 1.0°C/cm. Ice surface were exposed to an unsaturated atmosphere. The mean temperature between the plates is about 2 and 4°C respectively higher than ambient temperature because of the heater. The beads migrate definitely toward the colder plate. The mean migration rates for beads of about 50  $\mu$  diameter are approximately  $10^{-6}$  cm/sec for a temperature gradient of 0.5°C/cm and  $6 \times 10^{-6}$  cm/sec for 1.0°C/cm, though larger beads tend to migrate slower than smaller beads.

### III. Discussion

Eight possible mechanisms of particle migration were studied; some of these were excluded by experiments or theoretical considerations while the others remain possible explanations. The mechanisms are:

1. Rolling of glass beads on the ice surface following breaking or bending of their supporting necks.
2. Drifting of glass beads on a liquid layer on the ice.
3. Migration of the beads supporting necks by evaporation-condensation.
4. Neck migration by surface diffusion.
5. Diffusion through bulk ice.
6. Mass flow in the glass/ice interface layer.
7. Neck migration due to bulk diffusion in the neck.
8. Migration of the beads by enhanced surface layer diffusion.

*Mechanism 1.* As was seen in experiment 5, no breaking of the neck is observed when the ice surface is faced downward, yet the beads still migrate. If a bead made contact with the ice (other than at its neck) because of bending of the neck, it could be considered a possible mechanism. This, however, would require rolling of the bead, which was not observed in experiments 1 and 7. Thus the breaking or bending of supporting necks is not responsible for migration of the beads.

*Mechanism 2.* According to Fletcher (1962) an ice surface is covered by a 100 Å thick liquid film at 0°C, and hydrophilic glass beads will be wetted by this film. The force acting on a 40  $\mu$  diameter glass beads through the surface tension of this film is of the order of 1 dyne, which will pull the bead to the solid ice surface. About 0.1 dyne of force would be required to move the bead without rolling, assuming the coefficient of friction is 0.1. Applying Stokes' drag  $F = 3\pi\mu DU$  ( $F$ =force,  $\mu$ =viscosity,  $D$ =diameter,  $U$ =velocity), on this bead, and assuming it to be half immersed in the fluid, one obtains  $U = 3 \times 10^2$  cm/sec for  $\mu = 1.8 \times 10^{-2}$  dyne-sec/cm<sup>2</sup>,  $D = 4 \times 10^{-3}$  cm,  $F = 10^{-1}$  dyne. This is

an extremely fast flow that would fill a  $300 \times 100 \mu$  groove lying perpendicular to the flow in one second! Hydrophobic glass beads could be supported on this layer by surface tension and could drift with the flow. Observations, however, show that the speed of migration is not this great but is of the same order of magnitude as that of the hydrophilic beads, although the migration is intermittent. This mechanism is improbable.

*Mechanism 3.* The most operative mechanism of mass transfer on an ice surface above  $-10^\circ\text{C}$  is evaporation-condensation (Itagaki, 1966). The neck supporting the bead can be moved by evaporation from one side of it and condensation on the other side caused by some driving force such as thermal gradient. However, this mechanism does not move the neck as a unit but by transfer of individual molecules; thus it can cause no movement of the bead without rolling.

*Mechanism 4.* Most of the sintering processes of metals are ascribed to surface diffusion. However, the one-by-one process of surface diffusion, that is surface adsorbed molecules migrating, cannot move the neck of a bead as a unit, and thus cannot cause migration of the bead.

*Mechanism 5.* Several measurements have been made on marker movement in metals under a strong thermal gradient (Brammer, 1960; Meechan and Lehman, 1962; Shewmon, 1960; Swalin *et al.*, 1965). One-dimensional atom current density per unit time,  $J$ , (Meechan and Lehman, 1962, eq. (1),) is

$$J = \frac{D}{kT^2\lambda} \frac{dT}{dy} (\Delta E), \quad (1)$$

where  $D$  is the self-diffusion coefficient,  $\lambda$  is the lattice spacing,  $T$  is the absolute temperature,  $k$  is Boltzmann's constant,  $dT/dy$  is the temperature gradient, and  $\Delta E$  is the algebraic sum of certain energies. This can be positive or negative for the vacancy diffusion mechanism and negative for the interstitial mechanism. The maximum absolute value of  $\Delta E$  will be the activation energy of self-diffusion.

The presumable marker movement in ice due to this mechanism was calculated from eq. (1) using  $D = 10^{-11} \text{ cm}^2/\text{sec}$ ,  $T = 263^\circ\text{K}$ ,  $dT/dy = 1^\circ\text{C}/\text{cm}$ , and  $\Delta E = 15 \text{ kcal/mol}$ . The result is  $1.1 \times 10^{-12} \text{ cm/sec}$ , which is of the order of  $10^{-6}$  of the actual migration as seen in experiment 8. This mechanism is too slow to explain bead migration.

*Mechanism 6.* As long as the bead remains on the ice surface while ice recedes by sublimation, water molecules must be removed from the supporting neck to maintain it at a reasonable length. The molecules are probably removed from the interfacial layer between bead and ice. Flow in the interfacial layer is necessary because the removal of molecules must occur only from the edge of the interfacial layer, the surface of the ice. If there is a predominant flow in one direction in the layer, the beads supported by this layer may move with the flow. This layer could be 1) the liquid-like layer discussed by previous researchers, 2) the crystallographic structure transformed gradually to an amorphous structure in the layer, or 3) crystallographic periodicity in the layer up to the interface but with defect concentration greater than in the bulk of the ice.

*Mechanism 7.* The shape of the neck supporting the bead is symmetrical if no disturbance exists. However, nearby beads, etch pits, steps, etc. make the neck deviate from its symmetrical shape, therefore, the total curvature of the neck becomes asymmetric.

The difference in curvature around the neck requires mass flow in and around the neck toward the larger negative curvature because of the difference in surface free energy. Although most of the mass flows through evaporation-condensation, the bulk of the ice could contribute to the mass flow which would drift the neck as a whole and thus the bead along the flow. As the diameter of the neck supporting a  $40\mu$  bead is of the order of a few microns, the high diffusivity layer discussed in mechanism 8 could enhance the bulk diffusivity in the neck.

*Mechanism 8.* Diffusivity in the crystal could be different near the surface and in the bulk ice crystal. The surface could be the source or sink of point defects which are responsible for diffusion. A different concentration of defects near the surface would change the diffusivity in this layer.

The vapor pressure of ice at  $0^\circ\text{C}$  is  $6.13 \times 10^3$  dyne/cm<sup>2</sup> which is produced by  $2.3 \times 10^{21}$  molecules/cm<sup>2</sup>-sec striking the surface. Assuming the surface barrier to be  $E_b$ ,  $r = N \exp(-E_b/RT)$  will pass through the barrier and become interstitial where  $r$  is the production rate of excess interstitial molecules in the first layer,  $N$  is the number of molecules striking the ice surface ( $2.3 \times 10^{21}$ /cm<sup>2</sup>-sec at  $0^\circ\text{C}$ ),  $R$  is the gas constant and  $T$  is the absolute temperature. The number of interstitial molecules that jump inward from the first layer is  $[c_1 n \nu \exp(-E_j/RT)]/8$  where  $c_1$  is the concentration of excess interstitial molecules in the first layer,  $n$  is the number of molecules in the layer/cm<sup>2</sup>,  $\nu$  is the lattice vibration frequency and  $E_j$  is the energy required to jump into the next layer. Layers parallel to the (0001) plane are assumed in this case. The number of molecules that jump outward is  $[c_1 n \nu \exp(-E_b/RT)]/8$  and  $(6/8)[c_1 c_s n \nu \exp(-E_s/RT)]$  will be absorbed by the sink in the layer, where  $c_s$  is the concentration of sink and  $E_b$  and  $E_s$  are the energies required for the molecules to jump outward and into the sink respectively. Assuming  $E_b = E_j = E_s$  the exponential term can be written simply as  $\beta$  and the following equation is obtained:

$$N\beta + \frac{1}{8} c_2 n \nu \beta = \frac{1}{8} (c_1 n \nu \beta + c_1 n \nu \beta + 6c_1 c_s n \nu \beta), \quad (2)$$

where  $c_2$  is the concentration of excess interstitials in the second layer. General terms can be written by writing  $c_0 = \frac{8N}{n\nu}$  as

$$c_{i-1} - c_i = c_i - c_{i+1} + 6c_i c_s. \quad (3)$$

Using lattice spacing  $d$ , eq. (3) can be approximated as

$$\frac{d^2 c}{dx^2} - 6c \frac{c_s}{d^2} = 0. \quad (4)$$

The solution is

$$c = c_0 \exp\left(-\sqrt{\frac{6c_s}{d^2}} x\right). \quad (5)$$

From eq. (5) the effective thickness  $x_0 = \sqrt{\frac{d^2}{6c_s}} = \sqrt{\frac{10^{-15}}{6 \times 10^{-9}}} = 4.1 \times 10^{-4}$  cm assuming that the concentration of the sink is  $10^{-9}$  ( $\approx 10^6$ /cm<sup>2</sup>), which is the concentration of dislocations per lattice site in well annealed ice crystals (Muguruma, 1961; Muguruma and Higashi, 1963). The average defect concentration in this layer is

$$\frac{1}{x_0} \int_0^{x_0} c dx = \frac{1}{x_0} \int_0^{x_0} c_0 \exp\left(\frac{-x}{x_0}\right) dx = c_0 \left(1 - \frac{1}{e}\right) = \frac{8N}{n\nu} \left(1 - \frac{1}{e}\right). \quad (6)$$

Inserting the same values as before we obtain  $\frac{8 \times 2.3 \times 10^{21}}{10^{15} \times 10^{12}} \left(1 - \frac{1}{(2.7)}\right) = 1.16 \times 10^{-5}$ . This is more than  $10^4$  times larger than the equilibrium values obtained from the self-diffusion coefficient in bulk. Preliminary measurements of surface self-diffusion show it to be about  $10^5$  times greater than the bulk diffusion constant. Mass flow in this thick high diffusivity layer could move the beads with the flow.

Mechanisms 6-8 remain possible. The present author, however, feels that mechanism 6 is rather improbable. If a temperature gradient existed in the supporting interface, the mass flow would be from the colder side to the warmer side because evaporation from the warmer side would be faster. If there are no other local effects this flow would move the beads toward the warmer side, which is contradictory to the observations made in experiment 8.

Neither the materials of which the beads are made nor treatment of their surfaces seem to have any effect on their migration, with the exception of silicone grease treatment. This fact would also tend to exclude mechanism 6 because interfaces of different materials would have different characteristics which would affect migration. The effect of silicone grease could be attributed to uneven spreading of the grease on the ice surface, which might have produced an uneven temperature distribution to drive the beads.

A thermal gradient drives particles from the warmer to the colder side. Migration where there is no external thermal gradient could be attributed to local temperature differences due to an uneven surface around the beads. An uneven surface, *i.e.* one with etch pits or steps, would cause uneven heat dissipation during sublimation, resulting in an unequal temperature distribution. This random temperature distribution could supply the driving force for random migration of the beads.

The author feels that the most probable mechanism in accord with the existing experimental facts is mechanism 7 under the influence of enhanced surface layer diffusion as discussed in mechanism 8. However, a mechanism completely different from those discussed here is still possible.

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