

CONSTITUTIONAL SUPERCOOLING IN NATURAL ICE

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ABSTRACT

Theoretical examination of the thermal regime of crystallisation of natural ice demonstrates that constitutional supercooling is strongly favoured for sea ice formation, but is not likely to occur under normal conditions in the formation of fresh water ice. Apart from the salinity, the critical factor is the proportionality of the thermal gradient in the water just beneath the ice to the thermal gradient within the ice just above the freezing interface. Constitutional supercooling can occur in fresh water ice as the thermal gradient in the water beneath the ice approaches zero. Constitutional supercooling results in dendritic crystallisation and gives rise to horizontal c-axes in sea ice. Lack of constitutional supercooling makes plane front crystallisation more likely, and, since isothermal conditions are not usually found beneath fresh water ice, plane front crystallisation and vertical c-axes development is the normal case. Nevertheless, turbulence in the water immediately beneath growing ice could create an isothermal layer by mixing cold water downward to account for the occurrence of horizontal c-axes in fresh water ice.

INTRODUCTION

During the formation of ice at the surface of a natural body of water the conditions for constitutional supercooling and consequent dendritic crystallisation may appear at the lower surface of the ice. Such crystallisation results in horizontal orientation of the c-axes of the ice, and is the normal case in sea ice. On the other hand, it will be shown that, contrary to the conclusions of Weeks & Ackley (1982), constitutional supercooling is unlikely to appear in fresh water in the normal case, this conclusion being supported by the frequent observation of crystallisation of fresh water ice with vertical c-axes. Nevertheless, horizontal orientation of c-axes in fresh water ice has been observed in a significant number of cases, as reported, for example, by Lyons & Stoiber (1962), and this suggests that constitutional supercooling and dendritic crystallisation may take place in some circumstances in fresh water ice.

In the following pages, the freezing of fresh water and of sea water will be discussed to show the contrasting thermal conditions developed in each of these environments. The texture of the ice will be described to show the controls on c-axis orientation, and the principles of constitutional supercooling will be explained. Finally, theory will be developed to show that, whereas constitutional supercooling will always occur in the formation of sea ice, it occurs only in a very particular thermal environment in the formation of fresh water ice.

FREEZING OF FRESH WATER

The liquid-solid phase transition for water is unusual in that the density of ice is lower than that of water, the maximum density being reached at a temperature near 4°C . This has significant consequences for the freezing of natural bodies of fresh water where the chill boundary is at the upper surface of the water. While the temperature of the chilled surface remains above 4°C , the whole body of water loses heat to the surface by internal convective transfer which cools a substantial depth of the water isothermally to the surface temperature while this is above 4°C . However, once the chilling temperature at the surface falls below 4°C , the process of convective overturn ceases, since the density of the cooled water at the surface is now less than the maximum density already reached by the water below. Further cooling may be accomplished only by conduction of heat through the upper cooled layer of water which grows in thickness as more water is chilled. Eventually, if the chilling is severe enough, the freezing point is reached, and ice crystallises right at the surface of the water. The cover of ice thickens by conduction of heat away from the underlying water upwards through the

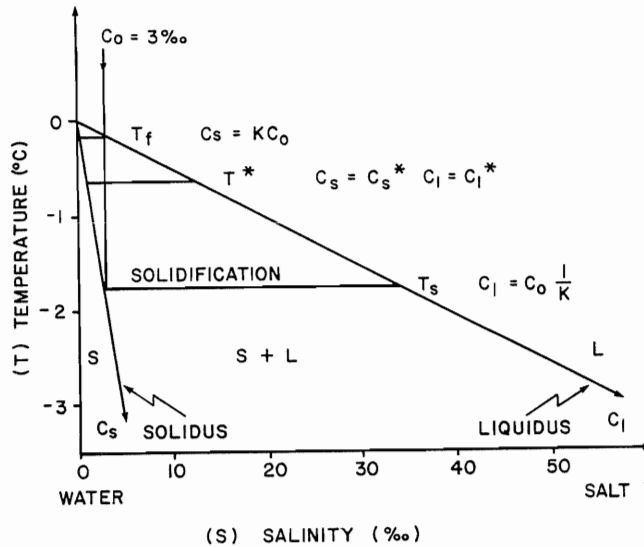


Figure 1. Simplified phase diagram - water/salt.

Notes.

1. The phase relations water/salt are shown by fields separated by the solidus and liquidus lines on which compositions are designated C_s and C_l respectively. The fields are:
 - S = solid ice (containing some salt):
 - S+L = ice in equilibrium with salt water:
 - L = liquid salt water,
2. Cooling of salt water of initial composition $C_0 = 3‰$ from above 0°C is portrayed.
3. At temperature T_f on the liquidus, ice crystals first appear, these being of composition $C_s = k.C_0$, where k is the equilibrium partition coefficient. For NaCl in ice $k < 10^{-4}$ (Harrison & Tiller, 1963).
4. Further cooling to T^* brings liquid of composition C_l into equilibrium with ice of composition C_s^* , this being the condition to develop constitutional supercooling, and $k = C_s^*/C_l^*$.
5. Ultimately, in a closed system, the liquid will solidify at the solidification temperature T_s , with ice of composition C_0 in equilibrium with the last liquid of composition C_l where: $C_l = C_0 \cdot 1/k$, but this point is not reached in natural open systems.
6. The slope of the liquidus is given by the equation $T = 0.052 S$, where T is the temperature, and S the salinity of the salt water. The value of k gives the relative slopes of solidus and liquidus.
7. The slope of the solidus for ice is inseparable from the temperature axis at any reasonable scale, so that a fictitious slope has been given in Figure 1 to illustrate the general principle.

ice, not only to keep the lower surface of the ice at the freezing temperature but also to remove the latent heat of crystallisation produced there. This statement may be put in the form of the energy balance equation for a planar solid-liquid interface where crystallisation is proceeding at an equilibrium rate, as follows:

$$K_s \cdot G_s - K_l \cdot G_l = \rho_s \cdot H \cdot R \quad (1)$$

where:

K_s and K_l are the thermal conductivities of the solid and liquid, in watts per metre per degree Kelvin,
 G_s and G_l are the corresponding temperature gradients, in degrees Kelvin per metre,
 ρ_s is the density of the solid, in kilograms per cubic metre,
 H is the heat of fusion of the solid, in joules per kilogram,
and R is the growth velocity, in metres per second.

This equation, given by Flemings (1974), demonstrates that the growth rate depends on the difference between the heat flow in the solid and in the liquid.

The temperature gradient existing in the ice depends upon the severity of the temperature at the upper surface of the ice. In fresh water, a steep thermal gradient normally also exists in the water just below the ice, the temperature rising from the freezing point at the bottom of the ice to 4°C in the isothermally cooled water a short distance below. This is in contrast to the situation in sea water which will be discussed in the next section.

The description given above has assumed, of course, that the water is quite still. If the water is agitated, particularly in the upper layer that has cooled enough to be at less than maximum density, then the coldest layer at the surface will be mixed downward by turbulence, and the cooling distributed through the upper agitated layer. Under severe conditions, a layer of significant thickness may be cooled to the freezing point, and freezing can then commence throughout this layer. The ice appears quite suddenly, as a blizzard of small discs of clear ice up to 2 to 3 millimetres across and a fraction of a millimetre in thickness, this being the process of formation of frazil ice. Since the ice of the frazil is less dense than the water, it will rise, but may well be kept in suspension by the turbulence. The blizzard is, in effect, upside down, the frazil ice behaving as a sediment in which the settlement field direction is upwards.

Turbulence is necessary to bring a significant depth of water to the freezing point all at the same time to create the conditions for formation of frazil ice in fresh water.

FREEZING OF SEA WATER

Sea water, in contrast to fresh water, is not a single component system, for it contains soluble ionic salts which have three very significant effects on the freezing process, each effect increasing in intensity proportionally to the amount of salt in solution.

First, the density of sea water is increased, and the contrast in density between ice and water is enhanced, particularly because the salts do not enter into the crystal lattice of the ice and are rejected to the water during crystallisation. Secondly, the temperature at which ice appears in sea water upon cooling is depressed below the freezing temperature of fresh water so that the temperature of appearance of ice is no longer invariant, as shown in Figure 1. This means that, for sea water, the term "freezing point" does not have the same clear meaning apparent for the freezing point of fresh water. At a given salinity, there is a temperature at which ice appears on cooling (the ice point shown as T_f in Figure 1), but, if the system is closed, further cooling does not solidify everything in the system as it does for fresh water at the freezing point. Further cooling of sea water merely produces more ice and concentrates the residual brine, which thereby obtains a lower freezing point. In the natural open system, however, the brine is dispersed into the larger volume of sea water, mostly by convection, but also by diffusion, and ice continuously forms at a temperature controlled by the bulk salinity of the sea water. A natural open system behaves therefore, as if it had a definite freezing point unless the dispersal of the brine is interrupted.

The third significant effect of the salts is that the temperature of maximum density is depressed below 4°C , so that, at about 25‰ salinity, the sea water reaches maximum density at the temperature at which ice appears, which is about -1.33°C . At greater salinities the density of the water increases right up to the ice point, and, supposing insufficient heterogeneous nucleation, the density of the water would continue to increase with supercooling.

These properties conveyed to sea water by the salts result in a process of production of ice from sea water (of salinity greater than 25‰) that differs markedly from that for fresh water. Chilled water at the surface is now always more dense than the water that lies beneath it, and the layer of low density water that appears prior to freezing to halt convective overturn in the case of fresh water does not appear, with the result that convective overturn is able to chill a substantial depth of water right down to the freezing point. By contrast to fresh water, sea water develops an isothermal layer of negligible temperature gradient lying directly below the ice when it forms.

This means that convective overturn has produced in sea water an effect very similar to that produced in fresh water by turbulence, and frazil ice should be able to appear throughout the chilled layer of sea water as it does in fresh water, turbulence not being necessary for frazil ice formation in sea water.

STRUCTURE, TEXTURE AND FABRIC OF NATURAL ICE

A core taken through a layer of naturally formed ice often reveals a structure of at least three layers in each of which a distinct texture may be recognised. The surface layer is usually composed of fine-grained equal-sized crystals of ice with a fabric in which most of the c-axes are aligned vertically. A few centimetres below the surface, a transition layer is recognised by its texture of slightly larger equant grains now with a fabric in which the c-axes are more or less equally divided between horizontal and vertical. The transition zone usually gives way within a few more centimetres in depth to a columnar zone recognised by the presence of vertically elongated crystals of relatively clear ice that increase in horizontal dimension as they extend downward to the bottom of the ice. In sea ice, the c-axes of these crystals are ubiquitously horizontal, although distributed randomly in azimuth in the normal case. In fresh water ice, on the other hand, orientation of c-axes is often vertical, although cases of horizontal orientation are also described as, for example, by Lyons & Stoiber (1962) and Michel and Ramseier (1971).

Michel and Ramseier (1971) recognise two types of crystallisation in the columnar zone in fresh water ice. In the first (designated S1), the c-axes are vertically oriented, preserving the vertical orientation of the crystallisation of the first thin primary sheet of plate ice, this type of ice forming in situations of low flow velocity in the underlying water. In the second type (designated S2), the c-axes are all horizontally oriented below a few centimetres of growth in the columnar crystals. This type of columnar ice is found beneath a layer of finer grained ice initially formed with a random orientation of c-axes by swift freezing or by growth from nucleating crystals of disturbed frazil or snow. The preferred orientation of c-axes comes about as the result of interference during growth by the mechanism described by Perey and Pounder (1958).

The explanation for the fabric of horizontally oriented c-axes of the columnar zone given by Perey and Pounder (1958) arises from understanding of the crystal structure of ice. The packing of the lattice is such that the reticular density promotes most rapid crystal growth in the basal plane. Growth along the c-axis is very much slower, with the result that the natural crystalline form of ice is a basal plate, which is in accord with the observed shape of frazil discoids and the observed planar extension of snowflakes. If the first ice on the surface of the water is composed of frazil ice rising from below, or of snowflakes falling from above, the natural floating position of these particles in still water will be with upright c-axes. A very simple mechanism therefore exists to explain the fabric of the surface layer. Wind or wave action may disturb the orientation of such initial fragments, and a more complex orientation of the c-axes will come about as a result of shingling effects. Relaxation of the disturbance, however, permits reorientation of the upper part of the layer to the natural floating position prior to freezing, and accounts for the development of the surface and transition layers found in the cores of natural ice.

New ice tends to nucleate on previously existing crystals, extending the lattice in the directions already established, particularly in the directions of most rapid growth. If the initial crystal orientations are random, crystal growth in the directions with a horizontal component is far more likely to result in interference between growing crystals than crystal growth in a downward direction. This is the basis for the explanation given by Perey and Pounder (1958), Pounder (1963) and Hobbs (1974) for the predominance of horizontal c-axis orientation in sea ice. The result appears to be a natural selection for continuing growth of those crystals with horizontally oriented c-axes and the eventual cessation of the growth of those crystals with c-axes not oriented horizontally. Striking examples of such growth in the laboratory are illustrated by Palosuo & Sippola (1963).

The explanation offered by Perey and Pounder (1958) is valuable to explain the establishment of horizontal c-axes after the initiation of columnar crystallisation just below the intermediate zone. But this mechanism should work just as well for fresh water ice as it does for sea ice and does not explain why much lake ice in the columnar zone has c-axis orientations that are vertical, whereas in sea ice they are horizontal.

The key to understanding these differences in texture between sea ice and fresh water ice comes from understanding the concept of constitutional supercooling.

CONSTITUTIONAL SUPERCOOLING

Development of the concept of constitutional supercooling starts with consideration of a solid crystal forming from a liquid of the same composition. A solute is now added to the liquid, but the condition is imposed that this solute be rejected and returned to the liquid by the crystallising lattice of the solid. Consequently, as the crystal face of the solid advances into the liquid, a concentration gradient of the solute is set up in the liquid, and the slope of this gradient depends on the rate of rejection of the solute by the crystal as compared to the rate of diffusion of the solute into the

liquid. If the rate of rejection is high, and the rate of diffusion is low, there will be a much higher concentration of the solute at the crystal boundary than exists further out in the liquid, as shown in Figure 2. The crystallisation of ice is marked by a high rate of rejection of salt by the crystallising ice and a low rate of diffusion of the salt into the water.

As the solid crystallises, a temperature gradient also develops in the region of the crystal interface which depends upon the relative rates of conduction of heat through the solid and the liquid, together with the latent heat of crystallisation, as described in Equation (1).

A hypothetical temperature gradient that corresponds to the ice points for the concentrations can now also be recognised extending through the concentration gradient along a line perpendicular to the growing crystal boundary. This temperature gradient (shown in Figure 2) will be that of the liquidus line in the appropriate part of the phase diagram for the system, (shown in Figure 1), and, if it does not correspond to the actual temperature gradient that has been set up by the heat transfer requirements (which are shown by Equation 1 to be independent of the factors setting up the concentration gradient), then constitutional supercooling exists where the actual temperature is lower than the hypothetical ice-point temperature, and ice will be a stable phase in such a region of constitutional supercooling, and this situation is shown in Figure 2.

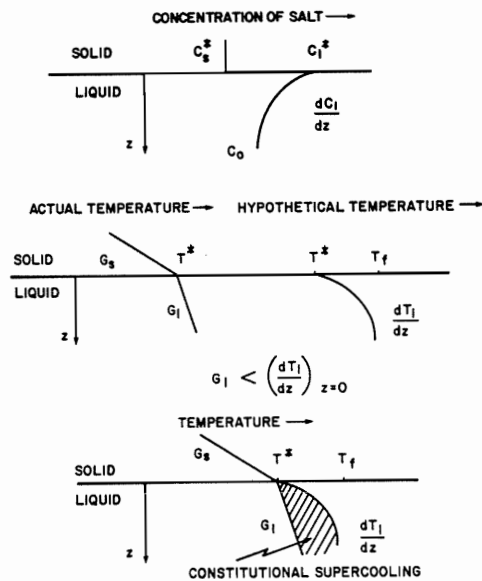


Figure 2. Constitutional Supercooling.

The result of constitutional supercooling is that any protuberance on the initial plane front of crystallisation will tend to advance into the constitutionally supercooled zone, and the rate of advance will be greatest where the crystallographic orientation presents a favourable growth direction towards the zone. The plane front is no longer stable, and, for reasons not yet fully understood, breaks into either a cellular or a rippled configuration, as described by Harrison & Tiller (1963). The outcome is dendritic crystallisation which develops in the columnar zone in the crystallisation of metal ingots. In natural ice, such dendritic crystallisation in the direction of most rapid growth will develop horizontal c-axes.

CRITERIA FOR CONSTITUTIONAL SUPERCOOLING

Consider ice freezing from saline water of composition C_0 , so that the composition of the ice (C_s^*) and brine (C_l^*) at the interface are those required by the phase diagram (Figure 1) at the temperature T^* . Rejection of salt by ice is almost complete, so that C_s^* is negligible, and need not be considered further.

Consider that diffusion is limited to the liquid, and that there is no convection, or, in the case of convection, a thin laminar layer close to the crystal surface will remain in the liquid, and this layer will be subject only to diffusion. The following discussion is limited to that layer.

During crystallisation, salt is rejected to the brine by the crystallising ice, and removed from the crystal surface by diffusion into the brine.

Let D_1 be the diffusion coefficient of the salt in the liquid, in metres per second,
 C_1 be the concentration of the salt in the liquid, in kilograms per kilogram,
and R be the rate of advance of the crystallisation front vertically downward along z , in metres per second.

All concentrations are given in kilograms per kilogram, and distances are measured downwards in the vertical direction, the point of origin being the interface between crystal and liquid, distance being designated by z' .

The requirement for solute conservation at the interface means that the resultant flux of solute at the interface is zero, so that;

$$D_1 \left(\frac{dC_1}{dz'} \right)_{z'=0} + R \cdot C_1^* = 0$$

Assuming equilibrium at the flat interface, the gradient of the hypothetical equilibrium liquidus temperature (T_1) with distance (z') away from the interface is related to the composition of the liquid given by the slope (m_1) of the liquidus line in the phase diagram, so that;

$$\left(\frac{dT_1}{dz'} \right)_{z'=0} = m_1 \left(\frac{dC_1}{dz'} \right)_{z'=0}$$

It might be recalled that, in the water/salt phase diagram (Figure 1), m_1 has a negative value, the ice-point temperature increasing as the concentration decreases.

Constitutional supercooling exists when the actual temperature gradient in the liquid at the interface (G_1) is less than the hypothetical liquidus temperature gradient, that is;

$$G_1 < \left(\frac{dT_1}{dz'} \right)_{z'=0}$$

and this is the condition for instability.

Making the appropriate substitutions, it follows that;

$$\frac{G_1}{R} < \frac{|m_1| C_1^*}{D_1} \quad (2)$$

where $|m_1|$ is the value of the slope of the liquidus line without regard to sign.

From the phase diagram shown in Figure 1, it can be seen that the term $|m_1| C_1^*$ gives the depression of the freezing point at C_1^* and corresponds to T^* . Equation (2) may therefore be rewritten:

$$\frac{G_1}{R} < \frac{T^*}{D_1} \quad (3)$$

For freshwater ice, the value of T^* will be very small; so that, if instability is to develop, the value of G_1/R must be very small indeed.

The value of m_1 may be found from the equation for the depression of the freezing point given by Kester (1974), the equation being;

$$T_f = -0.0137 - 0.051990 S - 0.00007225 S^2$$

where T_f is the depression of the freezing point below 0°C , and S is the salinity of the sea water between 4‰ and 40‰. The departure from linearity in this equation is not large, and the approximation:

$$T_f = -0.052 S$$

will not be very much in error at low salinities, and this equation has been used in constructing Figure 1.

A further modification can be made to equation (3) by eliminating the term R with substitution from equation (1), so that:

$$\frac{G_s}{G_1} > \frac{1}{K_s} \cdot \left(\frac{\rho_s \cdot H \cdot D_1}{T^*} + K_1 \right) \quad (4)$$

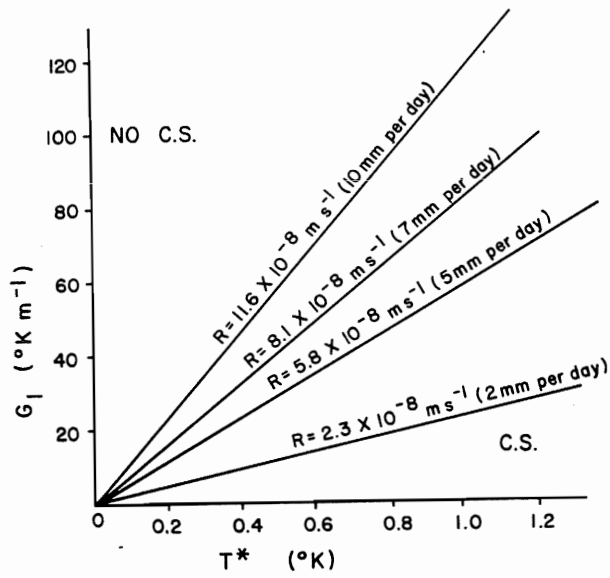


Figure 3. Criteria for constitutional supercooling (1).

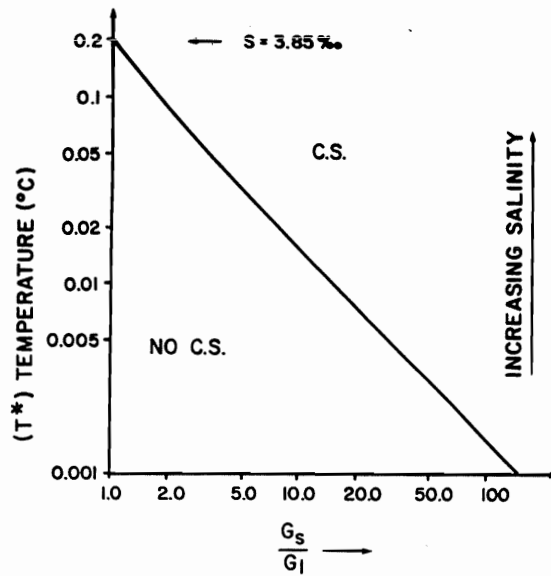


Figure 4. Criteria for constitutional supercooling (2).

It is not known how far T^* lies below the ice point T_f , but it is probable that the difference increases as the salinity increases, since there is more salt to reject. T^* must also be related to the original salinity C_0 since T_f depends on this. Therefore, it seems reasonable to suppose that a high value for T^* will be found in water of high salinity, and a low value in water of low salinity.

Again, it can be seen that a small temperature gradient in the underlying water together with a large salinity, provide the conditions for constitutional supercooling, and this last equation gives a dimensionless number that, for a particular depression of the freezing point, defines the conditions for constitutional supercooling.

The salinity of sea water, and the development of isothermal water beneath sea ice indicate that the conditions for constitutional supercooling are favoured in sea ice formation. On the other hand, neither of the mechanisms for developing instability works in the formation of fresh water ice, since there is little salt to be rejected to create the conditions for constitutional supercooling, and the temperature gradient beneath the ice is normally high. Crystallisation at the base of a cover of fresh water ice, therefore could be expected to be by planar growth, and, if this started on surficial crystals with c-axes oriented vertically, would preserve the orientation of the crystals through the whole period of freezing of the ice cover, particularly if the growth rate were not high.

The discussion has dealt so far only with the question of which state, solid or liquid, is thermodynamically stable in front of an initially plane crystallising surface. Other variables, such as surface energy and interface kinetics will finally decide if a perturbation will grow into the constitutionally supercooled liquid. Further equations can be developed to consider a more elaborate set of variables in the pursuit of instability theory. For example, a more complicated equation given by Flemings (1974) develops a term dependent on the ratio of the latent heat of crystallisation to the thermal conductivity of the liquid. For water/ice crystallisation, this term has a large value, indicating that the crystallising front will be stable, and cellular or dendritic textures are not likely to develop in fresh water. However, this resistance is obviously overcome in sea water.

TESTS OF CRITERIA FOR CONSTITUTIONAL SUPERCOOLING

Data for utilising relations (3) and (4) to outline possible fields of constitutional supercooling are listed in Table 1. The relations are put in the form of an equality which can be plotted as a line to separate the field of possible constitutional supercooling from the field of no constitutional supercooling.

TABLE 1

Data for Tests of Criteria for Constitutional Supercooling

| | | | |
|--------------------------------------|--------------------|-------------------|-----|
| $K_{ice} 0^\circ C$ | 2.0 | $W m^{-1} K^{-1}$ | (1) |
| $K_{water} 0^\circ C$ | 0.562 | $W m^{-1} K^{-1}$ | (1) |
| $\rho_{ice} 0^\circ C$ | 917 | $Kg m^{-3}$ | (2) |
| $H_{(water/ice)}$ | 334×10^3 | $J Kg^{-1}$ | (1) |
| $D_1 NaCl \text{ dilute}, 0^\circ C$ | 1×10^{-9} | $m^2 s^{-1}$ | (1) |

Sources

1. Gray, Dwight E. (Ed.) (1972)
2. Keast, Robert C. (Ed.) (1983)

For the relationship (3), values of the rate of growth of the bottom of the ice sheet have been considered as 0.002, 0.005, 0.007, and 0.010 metres per day, a line being drawn for each of these values in Figure 3. These data include the value of 7mm per day reported by Lake and Lewis (1972). The results shown in Figure 3 indicate that, if the temperature gradient in the underlying liquid is steep enough, constitutional supercooling is prevented even at relatively high values for T^* . On the other hand, if the thermal gradient in the liquid is low enough, constitutional supercooling is possible even at very low values for T^* , and therefore at very low salinities. The field of constitutional supercooling is expanded at higher growth rates.

For relationship (4) the equality between the dimensionless number G_s/G_1 has been plotted against T^* in Figure 4 to separate the fields of constitutional supercooling (C.S.) from non constitutional supercooling (NO C.S.). Using values from Table 1, the relationship (4) reduces to:

$$\frac{G_s}{G_1} = 0.153 \frac{1}{T^*} + 0.281$$

The results show that for salinities greater than about 4‰, constitutional supercooling is very likely to occur where $G_s > G_l$, and this is the normal condition in formation of sea ice. For constitutional supercooling to occur at lower salinities, the temperature gradient in the ice must be very much greater than in the liquid ($G_s \gg G_l$), so that, for constitutional supercooling to occur, the liquid must be nearly isothermal, the value of G_l approaching zero. Since G_l is unlikely to become negative under natural conditions, it can be seen from Equation (1) that, when $G_l = 0$, the rate of growth of the ice reaches a maximum where:

$$R_{\max} = \frac{K_s \cdot G_s}{\rho_s \cdot H}$$

and the maximum rate of growth calculated from the data in Table 1 is

$$R_{\max} = 6.7 \times 10^{-9} \times G_s \text{ m s}^{-1},$$

the temperature gradient in the ice (G_s) being in $^{\circ}\text{C}$ per metre.

CONCLUSIONS

Constitutional supercooling is likely to occur in the formation of natural ice, particularly in saline water with a low thermal gradient in the water beneath the ice. Constitutional supercooling promotes dendritic crystallisation with horizontal c-axes, which is the usual occurrence in sea ice. For fresh water, however, constitutional supercooling is inhibited for normal temperature gradients beneath the ice, so that crystallisation of ice from fresh water is not dendritic and c-axes are normally vertical.

Constitutional supercooling is possible in fresh water where the temperature gradient in the water beneath the ice approaches zero. In the normal formation of fresh water ice, this condition is unlikely, but could come about if turbulence beneath the ice causes mixing to create an isothermal layer of water. Turbulence associated with an isothermal layer creates the conditions for frazil ice formation beneath the ice, and fresh water ice with horizontal c-axes may therefore be associated with sedimentary structures such as ripples and dunes formed on the underside of the ice by sedimentation of frazil in an upward settlement field direction. Such sedimentary structures beneath fresh water ice have been reported by Carey (1966) and (1967).

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