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FRAZIL-ICE NUCLEATION BY MASS-EXCHANGE PROCESSES AT THE AIR–WATER INTERFACE

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Abstract. The physical requirements for proposed frazil-ice nucleation theories are reviewed in the light of recent observations on frazil-ice formation. It is concluded that spontaneous heterogeneous nucleation in a thin supercooled surface layer of water is not a viable mechanism for frazil-ice nucleation. Efforts to observe crystal multiplication by border ice have not been successful. The mass-exchange mechanism proposed by Osterkamp and others (1974) has been generalized to include splashing, wind spray, bubble bursting, evaporation, and material that originates at a distance from the stream (e.g. snow, frost, ice particles, cold organic material, and cold soil particles). It is shown that these mass-exchange processes can account for frazil-ice nucleation under a wide range of physical and meteorological conditions. It is suggested that secondary nucleation may be responsible for large frazil-ice concentrations in streams and rivers.

Résumé. *Nucléation de la glace du "frazil" par des processus d'échanges de masses à l'interface air en eau.* On a revu les conditions physiques requises par les théories pour la nucléation de la glace du "frazil" à la lumière de récentes observations sur cette formation. On en conclue qu'une nucléation hétérogène spontanée dans un niveau d'eau superficiel mince en surfusion n'est pas un mécanisme viable pour la nucléation de la glace du frazil. Les efforts pour observer la multiplication des cristaux par de la glace de bordure n'ont pas eu de succès. Le mécanisme d'échanges de masses proposé par Osterkamp et autres (1974) a été généralisé de manière à inclure l'éclaboussement, la pulvérisation, l'échappement des bulles, l'évaporation et les matières dont l'origine se trouve à une certaine distance du courant (par exemple, neige, givre, particules de glace, matière organique froide et particules de terre froides). On montre que ces processus d'échanges de masses peuvent rendre compte de la nucléation du frazil dans une large gamme de conditions physiques et météorologiques. On suggère qu'une nucléation secondaire peut être responsable des grandes concentrations de glace de frazil dans les cours d'eau et rivières.

Zusammenfassung. *Keimbildung von Sulzeis durch Massenaustauschvorgänge an der Grenzfläche zwischen Luft und Wasser.* Die physikalischen Bedingungen für vorgeschlagene Theorien zur Keimbildung von Sulzeis werden im Lichte neuer Beobachtungen der Entstehung von Sulzeis überprüft. Es ergibt sich, dass spontane Fremdkeimbildung in einer dünnen, unterkühlten Oberflächenschicht von Wasser kein ausreichender Mechanismus für die Keimbildung von Sulzeis ist. Bemühungen, Kristallvermehrungen durch Randeis zu beobachten, waren erfolglos. Der von Osterkamp u.a. (1974) vorgeschlagene Mechanismus des Massenaustausches wurde verallgemeinert, um Spritzen, Zersprühen durch Wind, Bersten von Blasen, Verdunstung sowie Stoffe einzubeziehen, die in einiger Entfernung vom Bach entstehen (z.B. Schnee, Reif, Eisteilchen, kalte organische Stoffe und kalte Bodenteilchen). Es wird gezeigt, dass diese Massenaustauschvorgänge die Keimbildung von Sulzeis unter mannigfachen physikalischen und meteorologischen Bedingungen erklären lassen. Man kann annehmen, dass sekundäre Keimbildung zu grossen Ansammlungen von Sulzeis in Bächen und Flüssen führt.

Introduction

Frazil-ice production in streams and rivers causes many engineering problems. These include flooding by frazil-ice jams, blockage of water-supply intakes, reduced power generation in hydroelectric facilities, interference with ship and barge transportation, and damage to hydraulic structures and facilities. Development of rational design methods for alleviating or avoiding these frazil-ice problems has been hindered by deficiencies in the present state of knowledge of frazil-ice formation.

Several theories have been proposed to explain nucleation of frazil-ice crystals in streams and rivers. Early investigators, notably Al'tberg (1938[a], [b]) and Devik (1944), believed that frazil ice was formed by spontaneous heterogeneous nucleation and by secondary nucleation processes, i.e. reproduction of frazil-ice crystals from parent ice in the water. The freezing nuclei were supposed to be either solid crystalline particles (e.g. dust particles) in the water or ice particles (e.g. border-ice fragments or snow). Piotrovich (1956) brought attention to the fact that the highest known threshold nucleation temperature T_m for freezing nuclei was

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-3.5°C for silver iodide. Since the supercooling ΔT observed in rivers was generally $<0.1^\circ$C, he concluded that materials normally found in rivers could not nucleate ice at such small values of ΔT and that only ice particles could be active freezing nuclei in rivers. Yet, there are certain situations (e.g. under clear skies, calm wind, and in wide rivers) where it has been difficult to explain frazil-ice nucleation by ice particles in the form of border-ice fragments or snow. Thus, the problem has been to explain frazil-ice nucleation occurring at the small values of ΔT observed in rivers and for all physical and meteorological conditions.

Contemporary frazil-ice nucleation mechanisms include spontaneous heterogeneous nucleation in a thin supercooled surface layer of water (Michel, 1967), a crystal multiplication process (Chalmers and Williamson, 1965), which is a special form of secondary nucleation, and a mass-exchange process (Osterkamp and others, 1974). Secondary nucleation should also be discussed with these mechanisms, since it may be important for multiplication of frazil-ice crystal numbers in streams and rivers. It should be noted that none of the above mechanisms has been directly observed to produce frazil-ice crystals in streams or rivers but, in theory, they could act singly or in concert to produce frazil-ice crystals. The purposes of this paper are to review the physical requirements for these mechanisms in the light of recent experimental evidence, to generalize the mass-exchange process proposed by Osterkamp and others (1974) and to show that mass-exchange processes and secondary nucleation can account for frazil-ice production in streams and rivers under a wide range of physical and meteorological conditions.

NUCLEATION IN A THIN HIGHLY SUPERCOOLED SURFACE LAYER OF WATER

Michel (1967) proposed to use heterogeneous nucleation theory to explain frazil-ice nucleation by assuming that a thin, highly supercooled surface layer of water exists at the time of frazil-ice nucleation. This layer was thought to be extremely thin, although no estimates of its thickness were made. The value of ΔT at the surface was supposed to be $>|T_\text{m}|$ so that frazil ice could be spontaneously nucleated there. A very large temperature gradient was assumed for this layer such that ΔT immediately below the layer was only a few hundredths of a degree.

Devik (1944, 1949) showed that ΔT for the surface layer of calm water in a shallow pan could reach 1.5 deg. Recent measurements by Katsaros (1973) and Katsaros and Liu (1974) have confirmed Devik's results for fresh water and saline water in laboratory experiments where ΔT in the surface layer of very calm water was generally about 1 deg with a maximum of 2.3 deg and for a lead in the ice of the Arctic Ocean where ΔT was about 0.2–0.3 deg in the surface layer. Their measurements were made with a radiation thermometer which senses a layer of water $\approx 50\ \mu$m in thickness. The temperature sensed by this type of radiometer was the same as the temperature at about the 10 μm depth (Hill, 1972). It has also been shown in the laboratory experiments of Katsaros (1973) that turbulence produced by stirring or mixing the water destroyed the supercooled layer (i.e. the temperature sensed by the radiometer was the same as the bulk temperature of the water). Osterkamp and Gilfilian (1975) used the same type of radiation thermometer to measure the surface temperature of a stream that was producing frazil ice. When the air temperature was -11°C and the stream was supercooled, with frazil-ice crystals present in the stream, the measured surface temperature was $0^\circ\pm0.5^\circ$C. The bulk water temperature of the stream (measured with a precision mercury-in-glass calorimeter thermometer) was -0.010°C and decreasing. Frazil-ice crystals were first observed about 4 min prior to the surface temperature measurement when the bulk water temperature was 0°C. Additional information on the surface temperature of the water at an air–water interface has been obtained from temperature-gradient measurements. Michel and Hanley (1975) found that the surface temperature of the water in a stirred tank in a laboratory cold room was -0.1 to -0.3°C at the time of frazil-ice nucleation. These experiments and the above radiometer measurements show that ΔT at the surface of a supercooled stream is, at most, a few tenths of a degree.

The value of T_m for the water in the stream is also of interest since if it is near 0°C (within a few tenths of a degree) then spontaneous nucleation would still be possible at the surface of the stream. Osterkamp and Gilfilian (1975) have shown that $T_m = -4.3$°C for small water drops (≈ 1 mm diameter) taken from a supercooled stream at a time when the stream was producing frazil ice. Water samples of larger volume (about 4×10^3 mm^3) frozen in sealed glass tubes gave similar results ($T_m = -4.5$°C). Schnell and Vali (1972) and Schnell (1974) have performed drop freezing experiments on distilled water samples containing specially prepared material from decayed leaves (leaf-derived nuclei) and bacteria-derived nuclei from the genus *Pseudomonas* sp. These biogenic nuclei can nucleate ice at T_m as warm as -1.3°C and they have been found to be abundant in cold mountain streams (personal communication from R. C. Schnell). However, the experiments with biogenic nuclei were performed on diluted rain-water and sea-water samples, not water from streams, and involved artificial and elaborate methods of preparation of the nuclei. Therefore, the application of these results for biogenic nuclei to the problem of frazil-ice nucleation is questionable. Since there is no other information available on the nucleation of natural stream water, it is concluded that $T_m = -4.3$°C but could potentially be as high as -1.3°C. Therefore, ΔT would have to be at least 1.3°C for spontaneous heterogeneous nucleation in the thin supercooled surface layer of water. This is about an order of magnitude greater than the observed ΔT (a few tenths of a degree).

Much of the research on ice nucleation has been done on quiescent water and, since frazil ice is a product of turbulent water, the question of the effect of turbulence on frazil-ice nucleation is pertinent. However, the results of Dorsey (1948), Carstens (1966), Thijssen and others (1968) and Michel and Hanley (1975) have shown that turbulence does not significantly affect T_m. For example, as noted above, Michel and Hanley (1975) found that T_m was -0.1 to -0.3°C for all stirring rates which included an experiment with zero water speed where $T_m = -0.22$°C.

In summary, while an extremely calm water surface may supercool 1–2 deg, it appears that the surface of a turbulent stream does not supercool more than a few tenths of a degree. Since the highest potential value of $T_m = -1.3$°C, it is concluded that spontaneous heterogeneous nucleation in a thin supercooled surface layer of water is not a viable mechanism for frazil-ice nucleation. It should be noted that it may be possible to nucleate ice spontaneously in an extremely calm water surface (puddle, pond or lake) but that the resulting ice form would be sheet ice rather than frazil ice.

MASS-EXCHANGE PROCESSES AT THE AIR–WATER INTERFACE

Osterkamp and others (1974) have shown that ice-crystal concentrations from 6×10^1 to 6×10^4 m^{-3} of air existed in the air near the surface of a stream during periods of supercooling. These ice crystals were in the form of hexagonal plates ranging in size from 60 to 350 μm with an average size of 180 μm. Ice crystals were always present in the air near the stream under both clear and cloudy conditions when the air temperature was <-8°C, while none was detected when the air temperature was >-8°C. After the stream surface was completely covered with ice, no ice crystals were detected near the stream. It was concluded that the ice crystals originated in the air near the stream. These airborne ice crystals were observed falling toward the surface of the supercooled stream and it was proposed that they became frazil-ice crystals after falling into the stream. This mass-exchange mechanism for frazil-ice nucleation is generalized below.

The air–water boundary is a complex region with the area of interaction extended by air bubbles in the water and water droplets in the air. For our purposes, mass exchange at the air–water interface can be divided into material that originates at some distance from the stream and material that originates in the stream.

Material which originates at a distance includes ice particles (snow, frost, ice particles from trees, shrubs, etc.), cold organic material and cold soil particles. This material may be introduced into the stream by precipitation (snow), wind or it may simply fall from the trees or banks. Many studies have shown that when ice particles are introduced into turbulent supercooled water, large numbers of frazil-ice crystals are produced (Al'tberg, 1938[a], [b]). The situation is not so clear when cold organic materials or cold soil particles are introduced into turbulent supercooled water. Al'tberg and Lavrov (1939) found that non-cooled sand introduced into water with $\Delta T = 4$ deg produced instantaneous crystallization but when $\Delta T = 1$–1.5 deg the crystallization process was much slower. Tesaker (1966) found that sand and sawdust reduced ΔT substantially. However, Kumai and Itagaki (1956) found that fine particles of silver iodide, kaolin, clay, and carbon (at room temperature) scattered on the surface of slightly supercooled water did not cause nucleation. They concluded that the above materials (at temperatures $> T_\mathrm{m}$) were not active freezing nuclei for slightly supercooled water.

Organic materials are well known for their ability to nucleate ice crystals at small ΔT and recent information has been summarized by Mason (1971) and Schnell (1974). Due to their abundance and availability, biogenic nuclei (Schnell, 1974) could easily nucleate frazil-ice crystals in certain situations (e.g. when cold organic material containing these nuclei comes in contact with the water).

While it is plausible for soil particles and organic material at temperatures $< T_\mathrm{m}$ to nucleate frazil-ice crystals in supercooled water, there does not appear to be any information from carefully controlled experiments on this point. Mass exchange involving material that originates at a distance can certainly account for frazil-ice nucleation when it is snowing and under windy conditions but it cannot explain frazil-ice nucleation in wide rivers under clear skies with calm wind conditions.

Mass exchange of material originating in the stream includes water droplets transferred to the cold air above the water surface by splashing, wind spray, bubble bursting, and evaporation. If the air temperature is $< T_\mathrm{m}$, some of these water droplets may be spontaneously nucleated in the air above the stream and the resulting ice crystals may reach the water where they become frazil-ice crystals.

The mechanical processes of splashing and wind spray may be important methods for introducing water droplets into the cold air above the water under certain conditions. In highly turbulent reaches of streams and rivers (rapids), splashing produced by water impinging on rocks and boulders and by waves is pronounced. Under windy conditions, a spray of water droplets may be introduced into the air above the water surface by mechanical disruption of waves and by air-bubble bursting at the surface of the water (Blanchard, 1963; Monahan, 1968).

Mass exchange at an air–water interface by air-bubble bursting in the water has been known for many years (see reviews by Blanchard, 1963; Mason, 1971; MacIntyre, 1974). Bubbles may be introduced into the water when air pockets are trapped in breaking waves, by splashing and by precipitation. The rising bubbles burst at the water surface when the thin film on top of the bubble ruptures and film drops and jet drops are ejected into the air. A 1 mm diameter bubble may produce 10^1–10^2 film drops from 1 to 20 μm in diameter ejected a few millimetres into the air and 1–5 jet drops with a diameter of ≈ 100 μm ejected 0.10–0.15 m into the air.

Water vapor produced by evaporation of supercooled water into the cold air may condense and form a supercooled fog (fogs have been reported over the water surface of supercooled streams (Osterkamp and others, 1974)). These fog droplets generally range in size from a few microns up to ≈ 60 μm in diameter (Mason, 1971).

When water droplets produced by any of the above mass-exchange processes are introduced into the cold air above the water surface, some of them may freeze into ice crystals if the air temperature is $< T_m$ for these droplets. To assess the relative importance of this mass-exchange process for eventually producing frazil-ice nuclei, it is necessary to determine the time required for a droplet to freeze. K. O. L. F. Jayaweera (unpublished research) has calculated that the time required for a water droplet with a 10 μm radius, initially at 0°C, and falling freely in a cold room at -10°C to reach -5°C is $\approx 10^{-3}$ s. A water droplet of 100 μm radius requires $\approx 10^{-1}$ s. This result is similar to that obtained by Hobbs and Alkezweeny (1969). It is clear that water droplets introduced into cold air at -10°C with $T_m = -5$°C would be nucleated in a very short time and therefore very close to the water surface. The resulting ice particles may be carried into the water by small-scale air turbulence or they may grow in the super-saturated environment until they are sufficiently large to overcome buoyancy forces in the air and fall into the stream where they become frazil-ice nuclei. Growth of these ice particles in the air can take place by vapor condensation and by collisions with other ice particles and with supercooled water droplets. The ice crystals collected above the surface of a stream by Osterkamp and others (1974) were all in the form of symmetrical hexagonal plates, indicating they grew from the vapor phase. Growth rates of ice crystals under these conditions are high (Mason, 1971) and the size range of ice crystals found by Osterkamp and others (1974) indicate residence times in the order of minutes for the ice crystals in the supercooled fog.

Ice particles nucleated in the air above a stream that subsequently enter the supercooled stream and become frazil-ice crystals may have various forms (e.g. hexagonal plates, ice spheres, irregularly shaped ice particles). However, the form of frazil-ice crystals in streams and rivers is usually that of thin circular discs. Schaefer (1950) and Arakawa (1954) have described the evolution of ice particles of various shapes in supercooled water. Their observations made at the small ΔT characteristic of streams and rivers showed that ice particles of all shapes evolved rapidly into disc forms.

These mass-exchange processes leading to frazil-ice nucleation are very general and can account for frazil-ice nucleation over the range of physical and meteorological conditions found on streams and rivers that produce frazil ice. Unfortunately, it is not possible to assess their relative importance because of a lack of field observations.

SECONDARY NUCLEATION

Secondary nucleation involves nucleation of ice crystals by parent ice already present in the water. It is known to occur in certain types of laboratory experiments (Al'tberg, 1938[b]; Arakawa, 1956; Garabedian and Strickland-Constable, 1974; Williamson, unpublished) in crystallizers used for freeze desalination (Estrin, 1970) and during solidification of metals (Flemings, 1974). The secondary ice crystals are produced by mechanical fragmentation and thermal dissolution of dendrites on the parent ice (Arakawa, 1956). Mechanical fragmentation by collisions (Garabedian and Strickland-Constable, 1974) appears to be very efficient for producing large numbers of secondary ice crystals. Secondary nucleation may be important once frazil-ice crystals exist in the stream water.

Al'tberg (1938[a]) appears to have been the first to recognize the importance of secondary nucleation. Arakawa (1956) observed the formation and growth of secondary ice crystals into disc crystals from parent ice in the water. Chalmers and Williamson (1965) proposed a special form of secondary nucleation which involved growth of disc crystals from border ice, although Michel (1971) has criticized this mechanism on the basis that lateral diffusion and transport is a slow process in large rivers.

It should be noted that there are no direct observations of secondary ice nucleation in streams or rivers. I have searched unsuccessfully for evidence of the crystal-multiplication process proposed by Chalmers and Williamson (1965) and higher concentrations of frazil-ice crystals near the shores (implied by their mechanism) have not been reported. Nevertheless, the high frazil-ice concentrations reported in the literature ($\approx 10^6$ m^{-3}; Schaefer, 1950) would seem to imply that secondary nucleation exists in streams and rivers. Again, field observations are necessary to support this conclusion.

SUMMARY

Water-surface temperature measurements show that for a very calm water surface ΔT may be as large as 2.3 deg while for turbulent water ΔT is not more than a few tenths of a degree. Laboratory measurements for stream water show that $T_m = -4.3$°C but that it may potentially be as high as -1.3°C. Turbulence does not appear to affect T_m. It is concluded that spontaneous heterogeneous nucleation in a thin highly supercooled surface layer of water is not a viable mechanism for frazil-ice nucleation, but that it may be possible to nucleate sheet ice spontaneously in an extremely calm water surface (e.g. puddle, pond or lake) by this mechanism. Efforts to observe crystal multiplication by border ice have not been successful. The mass-exchange mechanism proposed by Osterkamp and others (1974) has been generalized.

Mass exchange in the form of snow or ice particles which fall into the stream can easily nucleate frazil-ice crystals, but the situation is not so clear for cold soil particles or cold organic materials that may be introduced into the water. It appears that cold soil particles or cold organic materials at temperatures $< T_m$ could act as nuclei for frazil-ice crystals.

Water droplets can be introduced into the cold air above the stream by splashing, wind spray, bubble bursting, and evaporation. Some of these water droplets can freeze into ice particles in a very short time and the resulting ice particles can fall into the stream and become frazil-ice crystals. When ice particles and frazil-ice crystals exist in the stream, secondary nucleation can cause them to multiply, producing the high frazil-ice concentrations observed in streams.

The above evidence and considerations strongly suggest that mass-exchange processes and secondary nucleation are responsible for frazil-ice production in streams and rivers. However, more experimental work in streams and rivers that produce frazil ice and carefully controlled laboratory experiments are necessary to assess the relative importance of these processes.

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