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ScienceDirect
International Journal of Refrigeration 29 (2006) 1218–1224
REVUE INTERNATIONALE DU FROID
INTERNATIONAL JOURNAL OF
refrigeration
www.elsevier.com/locate/ijrefrig

Influence of surface roughness on the supercooling degree: Case of selected water/ethanol solutions frozen on aluminium surfaces

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Received 10 October 2005; received in revised form 28 November 2005; accepted 12 January 2006
Available online 5 June 2006

Abstract

This paper presents a study on the impact of the roughness of a metallic surface on the magnitude of the supercooling during freezing of an aqueous solution. Aqueous solutions of ethanol (5%, 10% and 15% w/w) were used as model solutions. Five tubes of aluminium (internal diameter 8 mm) were machined to obtain a roughness between 0.63 and 13.3 μm. These tubes were immersed in a refrigerated bath with a programmable temperature scan. Thermocouples located at the inner surface of the tubes and in the solution were used to measure the magnitude of supercooling. Crystallisations were monitored and supercooling released calculated for each experiments. Our experimental results reveal that roughness is the influencing parameter of the supercooling released: larger the roughness, lower the supercooling. Moreover, a power law correlation between the roughness and supercooling was deduced.

Keywords: Ice slurry; Binary mixture; Water; Ethanol; Two-phase flow; Roughness; Wall; Aluminium

Influence de la rugosité de la paroi sur le degré de surfusion: étude de différentes solutions eau/éthanol congelées sur des surfaces en aluminium

Mots clés : Coulis de glace ; Mélange binaire ; Eau ; Éthanol ; Écoulement ; Diphasique ; Rugosité ; Paroi ; Aluminium

1. Introduction

Intense research effort is carried out in many laboratories and industries to develop environmental friendly secondary refrigerant, with the objective of reducing the mass of refrigeration fluids. Among these secondary refrigerant fluids, ice slurry offers many advantages, among which a high specific

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doi:10.1016/j.ijrefrig.2006.01.002

Nomenclature

G	free energy (J)
L	latent heat of fusion for pure water at 273.15 K (333.3×10^3 J kg^{-1})
n	number of experiments
$r(x)$	local roughness at location x (μm)
Ra	roughness (μm)
SC or ΔT	supercooling value (SC $= T_{IFP} - T_{RSC}$) (K)
T	temperature (K)
T_{IFP}	temperature of initial freezing point (K)
T_{RSC}	temperature at rupture of supercooling (K)
t	time (s)
X, x	length (m)
Greek symbols	
θ, α	angle (rd)
γ	surface tension (N m^{-1})
$\bar{\sigma}$	error (K)

energy and a reduced temperature shift over the melting domain. One of the major bottlenecks of the ice slurry technology lies in the development of reliable ice slurry generator. The complexity and cost of the existing devices still represents a main drawback of this technology. Researchers are now investigating different type of ice slurry generators such as generator using supercooled water with nucleation initialisation [1]. In this kind of generator, the mother solution is first refrigerated until a supercooling state. Then nucleation is initialized with different methods such as momentum decrease, ultrasonic field or bubble nucleation … The control of ice crystallisation and therefore of the supercooling remains a key aspect of this kind of generator. Research effort is thus needed to better understand the impact of roughness on the supercooling release in order to have a total control in this kind of generator.

Nucleation occurs spontaneously if and only if the associated change in free energy G for the system is negative ($\Delta G < 0$). Likewise, a system reaches equilibrium when the associated change in G for the system is zero ($\Delta G = 0$), and no spontaneous process will occur if the change in G is positive ($\Delta G > 0$). The creation of a nucleus implies the formation of an interface at the boundaries of the new phase. Some energy is expended to form this interface, based on the surface energy of each phase. If a hypothetical nucleus is too small, the energy that would be released by forming its volume is not enough to create its surface, and nucleation does not proceed ($\Delta G > 0$). As the phase transformation becomes more and more favorable, the formation of a given volume of nucleus frees enough energy to form an increasingly large surface, allowing progressively smaller nuclei to become viable. Heterogeneous crystallisation is characterised by the fact that a crystal is initiated from impurities or from specific surface conditions. The associate free energy variation needed to this phase transition is called ΔG_n^* (Eq. (1)). Above this threshold, ice crystals are unstable; they appear and disappear but cannot grow [2]. In this case, the ΔG_n^* threshold is function of the wetting angle θ (Fig. 1 and Eq. (1)). This angle is linked to the roughness of the surface and to other specific characteristics such as surface tension.

$$\Delta G_n^* = \frac{16\pi\gamma^3 T_{IFP}^2}{3L^2\Delta T^2} f(\theta) \quad (1)$$

$$f(\theta) = \frac{1}{4}(2 + \cos\theta)(1 - \cos\theta)^2 \quad (2)$$

It has been demonstrated for a long time that the roughness of a surface is supposed to interact with the level of supercooling. Besides, a limited amount of literature is available on this subject. Saito et al. [3] showed that the surface characteristics such as roughness influenced the freezing temperature of supercooled water. Copper surfaces were considered with different surface roughnesses; nevertheless, no values of the roughness of the surfaces were

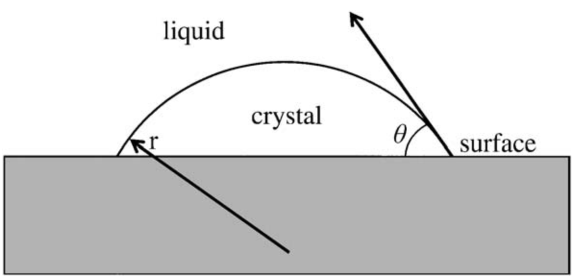

Fig. 1. Wetting angle θ of a droplet of water of radius r being frozen onto a surface.

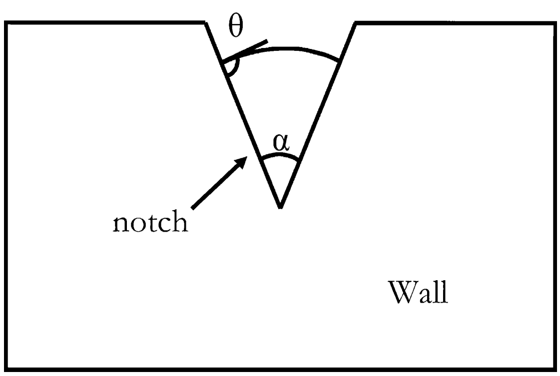

Fig. 2. Schematic view of a droplet of water undergoing crystallisation on notch.