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Active control of supercooling degree using two surfactants of different molecular sizes

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ABSTRACT

The authors have achieved active control of the supercooling degree by varying surfactant concentrations. To improve controllability of supercooling dissolution, the following experiments were carried out by different mixing methods using two kinds of surfactant of different molecular sizes. First, supercooling degrees were measured after dropping a “different” surfactant–pure water mixture with a fixed concentration onto a “base” surfactant–pure water mixture with a critical micelle concentration (CMC) in a glass test tube varying concentrations of the different surfactant-mixture. Second, supercooling degrees were measured after simultaneously mixing the mixture of a base surfactant at its CMC and a different surfactant of fixed concentration in a glass test tube varying the different surfactant-mixture concentrations. In both cases, adsorbed amounts of two surfactants to SiO_2 surfaces were measured by a quartz crystal microbalance. Correlations among the mixing method, surfactant concentration of the total mixture, the supercooling degree and the adsorbed amount were clarified.

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Contrôle actif du degré de surfusion en utilisant deux tensioactifs de différentes tailles moléculaires

Mots clés : Surfusion ; Deux types de tensioactif ; Différentes tailles moléculaires ; Différents procédés de mélange ; Adsorption

1. Introduction

Ice slurry has good fluidity and can maintain its low temperature, and it is therefore used for cold storage of food. One of the typical methods to form ice slurry is a supercooling method that forces dissolution of water’s or solution’s supercooling. However, supercooling has instability, so it is very possible that the formation of ice slurry must be stopped if supercooling unexpectedly results in dissolution. Furthermore, supercooling causes reduction of the coefficient of performance (COP) in the ice formation system due to a drop of the refrigerant’s evaporation temperature and the blocking of ice slurry flow in a tube. So, control of supercooling dissolution is so important.

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Nomenclature

A	cross-sectional area of surfactant molecular's hydrophobic group [Å^2]
Ae	electrode area [m^2]
C	target concentration of total mixture [wt%]
C_a	concentration of base surfactant [wt%]
C_b	concentration of different surfactant [wt%]
CMC	critical micelle concentration [wt%]
F_0	resonance frequency of quartz oscillator with SiO_2 deposition [Hz]
F_1	resonance frequency of quartz oscillator before adsorption [Hz]
F_2	resonance frequency of quartz oscillator after adsorption [Hz]
ΔF	$= F_2 – F_1$ [Hz]
INIC	International Nomenclature of Cosmetic Ingredients
m_a	mass of base surfactant [g]
m_b	mass of different surfactant [g]
m_{mix}	mass of total mixture [g]
m_w	mass of pure water [g]
m_{wa}	mass of base surfactant–pure water mixture [g]
m_{wb}	mass of different surfactant–pure water mixture [g]
Na	Avogadro constant [(=6.02214086 × 10^{23} mol^{-1}) QCM quartz crystal microbalance]
R	universal gas constant [(=8.31 J(mol K)$^{-1}$)]
T	absolute temperature [K]
T_f	temperature of total mixture immediately before supercooling dissolution [K]
Γ	adsorbed amount of surfactant molecules to gas-liquid interface at CMC [mol Å^{-2}]
Δm	adsorbed amount of surfactant molecules [ng]
γ	surface tension of surfactant–pure water mixture [mN m^{-1}]
μ_q	quartz shear modulus [kg m^{-1} s^{-2}]
ρ_q	quartz density [kg m^{-3}]

Several studies investigated various continuous ice slurry formation systems using the supercooling method (Tanino and Kozawa, 2001, Castaing-Lasvignottes et al., 2006, Bédécarrats et al., 2010). Validity of irradiation of ultrasonic waves to control supercooling dissolution of water was reported (Inada et al., 2001; Zhang et al., 2001). Supercooling dissolution was also investigated from the viewpoints of material and electrode shape (Hozumi et al., 2003, 2005). Furthermore, the influence of an additive such as polyvinyl alcohol on suppression of supercooling dissolution was investigated (Kumano et al., 2009). The influence of the surface roughness of aluminum on supercooling dissolution of an ethanol solution was investigated (Faucheux et al., 2009). The influences of the surface characteristics of test plates on suppression of supercooling dissolution were also studied (Jung et al., 2011). In addition, supercooling dissolution of a TBAB aqueous solution using DC voltage application was studied (Kumano and Goth, 2016). And the influences of AC and DC voltage impressions on the propagation of supercooling dissolution for W/O emulsions with higher water contents of 70% and 80% were also investigated (Matsumoto et al., 2010a, 2010b).

However, studies for control of supercooling dissolution have been limited to either its suppression or its promotion. Thus, the authors clarified that adsorption of the surfactant molecules to the interfaces suppressed supercooling dissolution, while, the surfactant molecular itself which could not be adsorbed or a micelle formed by those surfactant molecules promoted supercooling dissolution. Based on this result, the authors were able to actively control the average supercooling degree of a surfactant–pure water mixture (hereafter, "surfactant mixture") for the first time by varying the surfactant mixture concentrations (Matsumoto et al., 2013, 2015). But, for controllability of supercooling dissolution there is still room for improvement. So, in this paper, to further improve the controllability supercooling degrees of two kinds of surfactant mixture mixed in a glass test tube, they were measured with varying concentrations of the mixed mixture and mixing methods, however, two surfactants used have different molecular sizes. Validity of the measured supercooling degrees was investigated by comparison with the adsorbed amounts measured for both surfactants.

2. Experiment

2.1. *Selection of surfactants*

Two kinds of surfactant were used, and those of INICs (International Nomenclature of Cosmetic Ingredients) were Polyglyceryl-5 Dioleate (hereafter, Dioleate) (critical micelle concentration (CMC): 0.0171 wt% at 0 °C) and Polyglyceryl-10 Caprate (hereafter, Caprate) (CMC: 0.0177 wt% at 0 °C) which are nonionic surfactants. The CMC value was obtained by the relation between the surface tension and the concentration of the surfactant mixture. Two surfactants having different molecular cross-sectional areas were selected. The adsorbed amount of surfactant molecules to the gas-liquid interface at the CMC was obtained by Gibbs' adsorption isotherm corresponding to Eq. (1) (Kitahara and Furusawa, 1995), after which the cross-sectional area of each surfactant molecular was calculated by Eq. (2) (Kitahara and Furusawa, 1995).

$$\Gamma = -\frac{1}{RT}\left(\frac{d\gamma}{d\ln C}\right) \quad \left[\mathrm{mol}\ \text{Å}^{-2}\right] \tag{1}$$

$$A = \frac{1}{\Gamma} \times \left(\frac{1}{Na}\right) \quad \left[\text{Å}^{2}\right] \tag{2}$$

For each surfactant, INIC, the hydrophobic group, the CMC value at 0 °C and the cross-sectional area are shown in Table 1. As shown in Table 1, the cross-sectional area of the Dioleate was larger than that of the Caprate by about 4.6 times.

2.2. *Mixing method*

In this paper, two types of the mixing method, called "Pattern 1" and "Pattern 2", were adopted.

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