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Coupled population balance and heat transfer model for the description of ice recrystallization during long-term storage of ice cream

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ABSTRACT

A coupled population and energy balance model describing separately growth and dissolution of the ice crystals was developed to simulate recrystallization by Ostwald ripening during long-term storage of ice cream. The model reproduced accurately ($R^2 > 0.90$) measurements of the ice crystal average diameter for two ice creams (ICA and ICB) stored at temperatures between −5 and −18 °C for 104 days. Simulations indicated that ice crystal dissolution controls recrystallization during the early stages of storage, after which both growth and dissolution occur at decreasing rates. Carrageenan, ICB primary stabilizer, seemed to better preserve small ice crystals than locust bean gum, ICA primary stabilizer, by reducing ice crystal growth and dissolution rates. A sensitivity analysis indicated that the activation energy for ice crystal dissolution is the most significant model parameter and that the impact of heat transfer parameters is negligible because of the rapid change of the ice cream temperature.

1. Introduction

During storage of frozen desserts such as ice cream, ice recrystallization can significantly modify the state of the water in the product. Ice recrystallization involves modification of the number, size, shape and/or orientation of ice crystals following initial solidification and can affect the product microstructure and texture (Donhowe and Hartel, 1996a; Hartel, 2013). In ice cream, recrystallization generally promotes an increase of the average size of ice crystals, causing an unappreciated coarse and grainy texture when ice crystals become sufficiently large to be detected in the mouth (Russell et al., 1999; Adapa et al., 2010; Goff, 2008). It is critical to select appropriate storage conditions, mainly regarding the storage temperature, for an adequate preservation of the ice cream during long-term storage.

Several mechanisms are implicated in ice recrystallization and are driven by the minimization of the surface free energy of the crystal phase and the equalization of the phase chemical potential (Donhowe and Hartel, 1996a; Hagiwara et al., 2006). Ostwald ripening, also known as migratory recrystallization, has been identified as the main recrystallization mechanism during storage of ice cream (Flores and Goff, 1999). Ostwald ripening is characterized by the growth of large ice crystals from the integration of liquid water provided by the dissolution of small ones. Large ice crystals, having a low surface to volume ratio, are more stable than small ice crystals because the well-ordered and packed water molecules in the interior of ice crystals have a lower energy state than water molecules at the surface. During storage, water molecules at the surface of small ice crystals tend to diffuse through the unfrozen phase of the ice cream to deposit on the surface of larger ice crystals (Adapa et al., 2010; Hagiwara et al., 2006). The net impact of ice recrystallization in ice cream during storage is an increase of the ice crystal average size at a rate which depends on the sum of these growth and dissolution components.

Models have been developed to describe modification of the distribution of crystal size by Ostwald ripening during storage of food with the aim to better understand the impact of Ostwald ripening on food preservation and to streamline the selection of appropriate storage conditions. Donhowe and Hartel (1996a, 1996b) studied the effect of ice recrystallization in ice cream during storage at stable and fluctuating temperatures. The recrystallization rate increased with increasing temperature and temperature

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Nomenclature

A	Heat transfer surface between the ice cream and the air, m^2
A_{ice}	Total surface of the ice crystals in the ice cream, m^2
$A_{ice,avg,i}$	Average surface of a crystal in the interval $L_{avg,i} - \Delta L_i/2$, $L_{avg,i} + \Delta L_i/2$, m^2
C_{32}	Sauter average chord length of the ice crystals, m
$C_{p,app}$	Apparent heat capacity of the ice cream, J kg^{-1} °C^{-1}
D	Dissolution rate of the ice crystals, m s^{-1}
$\overline{D}$	Average dissolution rate of the ice crystals, m s^{-1}
E_a	Activation energy of ice crystal dissolution, J mol^{-1}
G	Growth rate of the ice crystals, m s^{-1}
H	Enthalpy of the ice cream, J
H_{ref}	Reference enthalpy of the energy balance, J
k_d	Coefficient of dissolution of the ice crystals, m^2 s^{-1}
k_g	Coefficient of growth of the ice crystals, m s^{-1} °C^{-1}
L	Diameter of an ice crystal, m
L_{43}	[4, 3] average diameter of the ice crystals, m
$L_{avg,i}$	Average diameter of an ice crystal in the interval $L_{avg,i} - \Delta L_i/2$, $L_{avg,i} + \Delta L_i/2$, m
L_{crit}	Critical diameter of the ice crystals in the ice cream, m
L_{max}	Maximum diameter of the ice crystals in the ice cream, m
$\overline{L}$	Average size of the crystals calculated using the LSW model (Eq. (1)), m
$\overline{L}_0$	Initial average size of the crystals for the LSW model (Eq. (1)), m
n_1	Recrystallization rate of the LSW model (Eq. (1))
n_2	Coefficient of the LSW model (Eq. (1))
N_i	Number density of the ice crystals in the interval $L_{avg,i} - \Delta L_i/2$, $L_{avg,i} + \Delta L_i/2$, m^{-3}
p	Number of classes for the numerical implementation of the recrystallization model
q	Number of time intervals for the numerical implementation of the recrystallization model
R	Ideal gas constant, J mol^{-1} °C^{-1}
t	Storage time, s
T	Temperature of the ice cream, °C
T_a	Temperature of storage, °C
T^*	Freezing temperature of the ice cream, °C
U	Global heat transfer coefficient between the ice cream and the air, J m^{-2} s^{-1}
V	Volume of ice cream, m^3
$V_{avg,i}$	Average volume of an ice crystal in the interval $L_{avg,i} - \Delta L/2$, $L_{avg,i} + \Delta L/2$, m^3
w	Mass fraction of sugar in the unfrozen phase of the ice cream

Greek letters

γ	Surface tension between the ice crystals and the unfrozen phase, J m^{-2}
ΔH_S	Enthalpy of solidification of the ice, J kg^{-1}
ΔL_i	Diameter interval for class *i*, m
Δt	Time step for the numerical implementation of the recrystallization model, s
θ	Input parameter of the recrystallization model
ρ_{app}	Apparent density of the ice cream, kg m^{-3}
ρ_{ice}	Density of the ice in the ice cream, kg m^{-3}
ϑ_{ice}	Volume fraction of the ice in the ice cream
ϕ_{ice}	Mass fraction of the ice in the ice cream
ϕ_w	Mass fraction of water (liquid + ice) in the ice cream
ψ	Ice crystal size distribution in the ice cream, m^{-4}

fluctuations and was well fitted by the Arrhenius and Williams–Landel–Ferry equations. The experimental results of Donhowe and Hartel (1996a, 1996b) were later used by Ben-Yoseph and Hartel (1998) to simulate ice recrystallization in ice cream during transportation and storage. They determined that the evolution of the average ice crystal size was described accurately using a general asymptotic model developed by Lifshitz and Slyozov (1961) and Wagner (1961) (Pronk et al., 2005a), also referred as the LSW model:

$$\overline{L}(t) = \overline{L_0} + n_1 t^{1/n_2}, \tag{1}$$

where $\overline{L}$ is the average size of the crystals after a storage time t, $\overline{L_0}$ the average size at the beginning of storage, n_1 the recrystallization rate and n_2 a coefficient generally between two and three depending on the limiting (diffusion or convection) mass transfer mechanism (Pronk et al., 2005a). The LSW model may represent the most widely used model to describe average crystal size in recrystallization processes governed by Ostwald ripening and has recently been successfully applied to describe the average ice crystal size during long-term storage of ice cream at temperatures between −5 and −18 °C by Ndoye and Alvarez (2015). However, the LSW model has significant limitations, notably that it does not provide information regarding the distribution of crystal size and may inaccurately describe the average crystal size during short-term storage because it does not consider the initial distribution of crystal size (Iggland and Mazzotti, 2012).

In the last few years, models describing the complete distribution of crystal size have been developed using population balance equations. Population balance equations are partial differential equations describing all or some of the accumulation, growth, dissolution, diffusion, convection, birth and death phenomena occurring in the product. Population balance equations have been successfully applied to describe the size of ice crystals during freezing of sucrose solution, sorbet and ice cream (Lian et al., 2006; Benkhelifa et al., 2008; Dorneanu et al., 2010; Arellano et al., 2013; Casenave et al., 2014) accounting nucleation, growth, breakage and/or agglomeration processes. However, these models have not been extended to recrystallization or Ostwald ripening which can take place in crystallization process when the supersaturation is reduced. Several population balance models that account Ostwald ripening phenomenon but concerning applications other than ice crystallization can be found in the literature (Tavare, 1987; McCoy, 2001; Madras and McCoy, 2001; Stahl et al., 2004; Iggland and Mazzotti, 2012; Vetter et al., 2013). From these studies, two Ostwald ripening modelling approaches may be highlighted: description of growth and dissolution processes using a single rate coefficient or separately. When growth and dissolution phenomena are considered together, the rate coefficient is often expressed as size-dependent based on the Gibbs–Thomson equation (Stahl et al., 2004; Iggland and Mazzotti, 2012; Vetter et al., 2013). Such an approach results in a rate coefficient that can be sufficiently accurate when Ostwald ripening is negligible compared to nucleation and growth phenomena. However, gathering growth and dissolution in a single expression provides limited insight into how these two competing phenomena interact when Ostwald