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Review

Water crystallization and its importance to freezing of foods: A review

Hossein Kiani and Da-Wen Sun*

FRCFT, University College Dublin, National University of Ireland, Agriculture and Food Science Centre, Earlsfort Terrace, Belfield, Dublin 4, Ireland (Tel.: +353 1 7167342; fax: +353 1 7167493; e-mail: dawen.sun@ucd.ie; URLs: http://www.ucd.ie/refrig, http://www.ucd.ie/sun)

In this review, different aspects of water crystallization including modelling approaches, process evaluation methods and the effect of novel freezing techniques is presented. There are different methods available to explain the nucleation and growth of crystals. The characteristics of ice crystals are studied by light and electron microscopy methods for many years, and recently a number of novel methods including magnetic resonance imaging, X-ray analysis, and infrared spectroscopy are employed. Several emerging techniques are developed to improve the crystallization of water during freezing, including ultrasound assisted freezing, high pressure freezing, ice nucleating proteins, and supersession of nucleation. Understanding the mechanisms of these new techniques and their relationship to the crystallization phenomenon can be helpful for improving freezing processes.

Introduction

Freezing is among the most popular and efficient methods of food preservation and consists of three stages, i.e., cooling the product to its freezing point (pre-cooling or chilling stage), removing the latent heat of crystallization (phase transition stage) and finally cooling the product to the final storage temperature (tempering stage). The phase transition part of the freezing process involves the conversion of water to ice through the crystallization process and is the key step determining the efficiency of the process and the quality of the frozen product (Alizadeh, Chapleau, de-Lamballerie & Le-Bail, 2009; Alvarez, Fernández & Canet, 2010; de Paula, Colet, de Oliveira, Valduga & Treichel, 2011; Fennema, Powrie, & Marth, 1973; Jin *et al*., 2010; Le-Bail, Nicolitch & Vuillod, 2010; Maity, Raju & Bawa, 2009; Steffolani, Ribotta, Perez, Puppo & León, 2011; Zaritzky, 2006). In the freezing of tissue foods, formation of large ice crystals which are mostly extracellular, results in significant damages to the tissue (Ahmad, Yaghmaee & Durance, 2010; Delgado, Zheng & Sun, 2009; Ming, Rahim, Wan & Ariff, 2009; Streit, Corrieu & Béal, 2010; Yu, Ma, Zheng, Liu & Sun, 2011). On the other hand, formation of fine crystals that are evenly distributed both inside and outside the cells, leads to the quality of the product to be better preserved due to less damages to the tissue (Sun & Zheng, 2006). However, in some processes such as freeze drying and freeze concentration, large crystals are more desired (Saclier, Peczalski, & Andrieu, 2010). Therefore, the control, understanding and prediction of the crystallization process and related phenomena in regard to the crystal characteristics are very essential for the improvement of freezing processes.

Crystallization is a general term used to describe several different phenomena related to the formation of a crystalline lattice structure (Hartel, 2001). This process consists of two main successive stages; nucleation and crystal growth. The interaction between these two steps determines the crystal characteristics, i.e., size, distribution and morphology of the crystals. Although the process of formation and growth of the crystals is complicated and therefore difficult to understand, some theoretical modeling approaches have been proposed for the description of both nucleation and growth (Hartel, 2001; Mullin, 2001; Martins, Castro & Lopes, 2011; Myerson, 2002a, 2002b). The theories employ different thermodynamic, mass transfer and heat transfer principles to explain the crystallization process. In addition to the theoretical modeling approaches, several experimental studies have been carried out to link the crystal characteristics to different processing parameters including cooling rate and heat transfer (Bald, 1986; Bevilacqua, Zaritzky, & Calvelo, 1979; Bevilacqua & Zaritzky, 1980; Woinet, Andrieu, Laurent, & Min, 1998) as well as mass transfer phenomena (Bae, Miyawaki, & Yano, 1993; Miyawaki, 2001; Miyawaki, Abe, & Yano, 1992).

Experimental evaluation of crystallization of water and its impacts on the texture of frozen products have been

* Corresponding author.

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Nomenclature

ΔG	overall excess free energy, J
ΔG_c	critical free energy for nucleation, J
$\Delta G_{c,het}$	nucleation energy of heterogeneous nucleation, J
$\Delta G_{max,s}$	nucleation energy, J
ΔG_s	surface excess free energy, J
ΔG_v	volume excess free energy, J
ΔG_v	free energy change of the transformation per unit volume, J m^{-3}
ΔT_s	supercooling, °C
a	constant
A	surface area, m^2
a_{for}	the volumetric surface of foreign particles, m^2 m^{-3}
b	constant
B	rate of nucleation, nuclei per unit volume per unit time
B_{het}	rate of heterogeneous nucleation, nuclei per unit volume per unit time
B_{hom}	rate of homogeneous nucleation, nuclei per unit volume per unit time
B_s	rate of surface nucleation, nuclei per unit area of crystal surface per unit time
C	actual concentration, mol L^{-1}
C_1	constant
C_2	constant
C_{ad}	concentration on the surface, mol kg^{-1}
C_C	the molar density of the solid, m^3 mol^{-1}
D	diameter of ice crystals, m
D_{AB}	diffusivity, m^2 s^{-1}
d_m	molecular diameter, m
D_{surf}	the surface-diffusion coefficient of units moving on the foreign surface
D_W	the diffusion coefficient of water, m^2 s^{-1}
$\frac{dT}{dt}$	cooling rate, °C s^{-1}
$\frac{dX}{dt}$	velocity of the moving freezing front, m s^{-1}
$\frac{dm}{dt}$	crystal growth rate, kg s^{-1}
$\frac{dq}{dt}$	rate of heat transfer, J s^{-1}
f	geometric correction factor for homogeneous nucleation
f	natural frequency, s^{-1}
g	constant
G	rate of crystal growth, kg s^{-1}
h	film heat transfer coefficient, W m^{-2} °C^{-1}
He_{ad}	adsorption constant
k	Boltzmann constant
K_1	thermal conductivity, W m^{-1} °C^{-1}
K_G	overall crystal growth coefficients
m	constant
n	constant
n_0	concentration of monomers in the supersaturated solution
N_A	Avogadro's number ($N_A = 6.023 \times 10^{23}$ mol^{-1})
n_c	number of stable nuclei
r	particle radius, m
r_c	critical nucleus radius, m
s	initial freezing front position, m
S	supersaturation ratio
$S_{met,s}$	relative supersaturation
T*	melting point, °C
T	temperature, °C
t	time, s
t_c	characteristic freezing time, s
T_f	initial freezing temperature, °C
T_i	interfacial temperature, °C
U	velocity, m s^{-1}
W	work, J
W_s	work required to form a surface, J
W_v	work required to form bulk of a particle, J
Z	imbalance factor
x	length, m
β	constant
ρ_1	density of frozen phase, kg m^{-3}
σ	surface energy of the particle per unit area, J m^2
ν	the number of molecules or ions
θ	the wetting or contact angle, deg
γ_{CL}	the surface tension

carried out by using visualization and monitoring methods including light microscopy (Chow, Blindt, Chivers, & Povey, 2003; Chow, Blindt, Kamp, Grocutt, & Chivers, 2004; Olmo, Baena, & Risco, 2008), electron microscopy (Delgado & Rubiolo, 2005; Fernandez, Otero, Guignon, & Sanz, 2006; Sun & Li, 2003) as well as non-invasive detection techniques (Bischof, Mahr, Choi, Behling, & Mewes, 2007; Hills & Remigereau, 1997; Hindmarsh, Wilson, Johns, Russell, & Chen, 2005; Lee, Kwon, & Ramamoorthy, 2008; Mahdjoub, Chouvenc, Seurin, Andrieu, & Briguet, 2006; Mousavi, Miri, Cox, & Fryer, 2005, 2007; Zelent & Vanderkooi, 2009). These kinds of evaluation have been considered as an important part of studies of freezing process and have resulted in practical knowledge of the ice formation mechanism and also the microstructure of frozen foods.

Improvements of the freezing process through the control of crystallization of water have also been an important subject of research. Along with rapid freezing techniques (Sun & Zheng, 2006), some new methodologies have been employed recently to control the crystallization of water (Inada, Zhang, Yabe, & Kozawa, 2001; Saclier *et al.*, 2010; Sun & Li, 2003; Zhang, Inada, Yabe, Lu, & Kozawa, 2001) and enhance the