

Ice suspension crystallization pretreatment for a new hybrid seawater desalination process: modeling, simulation, and parametric sensitivity study

Ibtissam Baayyad, Naoual Semlali Aouragh Hassani*

Laboratoire d'Analyse et Synthèse des Procédés Industriels (LASPI), Ecole Mohammedia d'Ingénieurs, University Mohammed V in Rabat, BP 765 Rabat, Morocco, emails: semlali@emi.ac.ma (N. Semlali Aouragh Hassani), ibtissambayyad@research.emi.ac.ma (I. Baayyad)

Received 31 May 2017; Accepted 19 September 2017

ABSTRACT

Recent ice suspension crystallization in scraped surface heat exchanger (SSHE) developments and innovations in different industries (chemical, food, and water treatment) have enable this purification operation to become a promising application to seawater desalination. This paper focuses on modeling and simulation of ice suspension seawater SSHE crystallizer of a proposed new hybrid seawater desalination technology (freezing-reverse osmosis). The mathematical model that is developed is based on combining energy and population balance equations. Simulation results show that the developed mathematical model is able to predict physically interpretable trends of main ice suspension seawater crystallization phenomena in SSHE, essentially in terms of the evolution of ice suspension temperature and ice volume fraction. These results are in agreement with other studies conducted for ice suspension crystallization from other solutions. Furthermore, the developed model was used for a parametric sensitivity study to show its ability to quantify different freezing key parameters effects in order to improve the considered ice crystallizer seawater performance under investigation. Thus, the developed phenomenological model can be exploited as a promising tool for understanding ice suspension seawater crystallization phenomena so as identifying the significant variables and it allows to complete design of the new freezing pretreatment process. Hence, it can lead to improvements and optimization of the proposed hybrid seawater desalination performance.

Keywords: Seawater desalination; Pretreatment; Mathematical modeling; Simulation; Freeze crystallization; Population balance; Parametric sensitivity; SSHE

1. Introduction

Ice suspension crystallization process has recently seen growing developments and innovations in diversified industrial applications. It is essentially exploited in food processing for juice concentration [1,2]. It consists of water removal from a solution by freezing and separation of ice crystals formed from the concentrated fluid. Moreover, this process allows obtaining high purity ice crystals with high efficiency and low energy consumption [3,4]. A bibliographical analysis has shown that the majority of suspension freeze concentration applications in different industries (food, chemical, and water treatment) use scraped-surface heat exchangers (SSHE) [5–7]. This later design has been improved to enhance their specific application, whether for use as heat transfer units, crystallizers, or chemical reactors [8,9]. The interest in SSHE use is likely to increase in the future. This is due, firstly, to high viscosity and particulate fluids that can be processed and secondly, to its applicability as a research tool [9]. According to these above-mentioned arguments, ice suspension crystallization in SSHE is considered as a promising and leading technology candidate for the coming decade in freeze seawater desalination industry.

In the past, due to a lack of equipment and prejudice (cooling is always too expensive) have hampered freeze desalination development and implementation. However, these caveats have become recently less important. This resulted firstly in the increase of development of freezing separation technologies in several industries and secondly in

^{*} Corresponding author.

^{1944-3994/1944-3986 © 2017} Desalination Publications. All rights reserved.

the growing attention to exploring more effective solutions to potable water shortage that is actually a very critical problem in more and more regions worldwide.

Tremendous researchers have currently investigated a variety of freezing crystallization techniques for desalination. The use of this technology is still restricted due to the limited knowledge of the optimum technique for ice freezing and separation; it is almost impossible to obtain pure ice crystals by freezing method alone and consequently obtained water that did not comply with regulatory requirements for potable water [10–16].

Recently, a theoretical study of freeze concentration processes basic principles and future applications in desalination industry have been investigated [17]. The research concluded that in order to make this technology more attractive, a complete energy and economic processes study along with hybrid technologies development must be achieved. These hybrid technologies combine freezing process and other desalination methods such as reverse osmosis (RO). They permit to reduce both natural water supplies and waste use and increase total plant yield.

In the same context, a previously paper proposed analysis concerning a newly developed hybrid desalination technology that combines freezing process as seawater pretreatment upstream RO module [18]. The freezing pretreatment consists of (1) ice suspension crystallization in a scraped surface heat exchanger (SSHE), (2) separation and (3) fusion of ice crystals (Fig. 1).

The proposed freezing pretreatment process presents several advantages, such as relatively pure fed solution into RO module compared with conventional and membranes pretreatment processes. This permits less foulant and a lower operating pressure in RO module. Therefore, pump and membrane costs are reduced and high-quality water is obtained. A study of energy consumption evaluation of the designed hybrid system was investigated and it showed an energy savings of approximately 25%, and osmosis water quality improvement for about 71% when compared with conventional–RO desalination. These results were obtained on the basis of proposed freezing crystallization process mass and energy balances followed by RO desalination process simulation using non-commercial software [18].

The more important challenge in ice suspension crystallization processes for seawater pretreatment is to control both ice crystals quantity and quality in order to improve the obtained potable water. Indeed, this process modeling can help to gain deeper freezing pretreatment phenomena understanding and also identify new ways for optimizing its performance. Furthermore, it can reduce process scale-up



Fig. 1. Schematic representation of seawater freeze pretreatment process.

time from theoretical study to laboratory scale equipment and thereafter to industrial scale production. To our knowledge, no phenomenological model was ever suggested for seawater freezing process.

The objective of the present work is to develop a mathematical model of ice suspension seawater crystallization process for freezing pretreatment conditions in an SSHE for the newly proposed hybrid process. This model combines population and energy balances taking into account main phenomena involved in crystallization processes such as nucleation, growth, and breakage. Obtained simulation results showed that the developed model permits to predict main ice suspension seawater crystallization phenomena and parameters trends evolution in SSHE.

Furthermore, in order to show key parameters influence on ice seawater crystallization process, a parametric sensitivity study is conducted using the developed model.

This later can be exploited as a promising tool for phenomena involved comprehension, complete design of the new freezing pretreatment process and thus, improving and optimizing seawater freezing pretreatment process performance.

2. Model development

The phenomenological model for seawater freezing pretreatment in SSHE (Fig. 2) is developed based on population and energy balance equations. It takes into account the most occurred phenomena such as ice nucleation and growth kinetics, as well as breakage. It must be noted that generally, the breakage phenomena is neglected for simplification reasons in crystallization modeling studies.

The model is set up on the following usual assumptions.

- SSHE crystallizer is assumed to behave as a plug flow reactor [19–21].
- Radial diffusion is assumed to be negligible, because it has no significant influence due to the high radial mixing effect of the rotating blades [19–21].
- Refrigerant fluid or wall temperature is assumed to be constant over the whole length of the exchanger as it is suggested in many SSHE modeling studies [19–21].
- Growth rate is assumed to be independent of the crystal size [19–21].
- Attrition and agglomeration phenomena are generally neglected in crystallization processes. They are not usually significant for ice crystallization in SSHE [19–21].



Fig. 2. Schematic representation of the scraped surface heat exchanger.

36

2.1. Population balance equation

Population balance modeling has been applied to a number of industrial processes and forms an appropriate framework for ice crystallization process dynamic modeling. The population balance equation (PBE) is a hyperbolic partial differential equation, strongly non-linear, allows describing system state in term of the density function. It is generally given as follows:

$$\frac{\partial \mathbf{n}(L,t)}{\partial t} + G \frac{\partial \mathbf{n}(L,t)}{\partial L} = N \partial (L - L_c) + B_b \tag{1}$$

where n(L,t) is the density function (crystal distribution function), t is time, L is a characteristic dimension, δ denotes the Dirac function, G is the growth rate, N is the ice crystals nucleation rate, B_b is the ice crystals net production due to the breakage phenomena.

It is important to specify that in contrary of many works, the developed model takes into account breakage phenomena, which has a significant effect on ice crystallization mechanisms in SSHE due to the scraper.

2.1.1. Growth phenomenon G

Crystal growth determines the rate of single crystal diameter change with respect to the difference between ice/liquid interface temperature, T_{sat} , and ice suspension temperature, *T*. It is expressed as follows [19–21]:

$$G = \beta(T_{\text{sat}} - T) \tag{2}$$

where β is the growth rate parameter, T_{sat} is a threshold temperature.

If $T < T_{sat}$ the crystals are growing and G > 0. On the contrary, if $T > T_{sat}$ (in warm temperature zones) the crystals are melting and G < 0.

2.1.2. Nucleation phenomenon N

Nucleation represents ice crystals birth. In SSHE, this only occurs at the freezer wall where the temperature gradient is high enough to lead to new crystals birth characterized by the initial crystals size L_c [22].

Two types of ice nucleation mechanisms are distinguished: primary or secondary nucleation, with primary nucleation being either homogeneous or heterogeneous. In SSHEs, heterogeneous primary nucleation is the predominant mechanism [23]. It depends on the sub-cooling degree [22] that is defined as the difference between solution freezing temperature, $T_{\rm sat'}$ and refrigerant fluid temperature, $T_{\rm rf'}$.

Therefore, ice nucleation rate is expressed by [19-21]:

$$N = \alpha \frac{2 \times \pi \times R_e}{V} \left(T_{\rm sat} - T_{\rm rf} \right)^2 \tag{3}$$

where α is the nucleation rate parameter, R_e and V are the SSHE diameter and volume, respectively.

2.1.3. Breakage phenomenon B_h

Analyzing SSHE design, we noticed the presence of scraper blades that scrape the inner surface of the crystallizer. It permits to avoid particle deposition and to enhance the heat transfer rates. Nevertheless, the scraper blades can break the ice crystals formed. Thus, in the developed model, the breakage phenomena must be taken into account in order to better describe ice crystallization mechanisms inside the SSHE.

It is assumed that a particle of size L_0 is broken into two particles of the same size *L*. Ice total volume is considered unchanged by fragmentation [20]. Thus, the relation between L_0 and *L* is given as follows:

$$L_0 = 2^{1/3} L$$
 (4)

So the particles net increase by breakage B_b can be expressed according to the equation as follows [19–21]:

$$B_b = \varepsilon N_{\text{scrap}} \phi_i^{\upsilon} [22^{2/3} \text{Ln}(\sqrt[3]{2L}) - \text{Ln}(L)]$$
(5)

where ε is the breakage rate parameter, N_{scrap} is dasher rotation speed and ϕ_i is the ice volume fraction.

 ν , is the breakage power coefficient that is taken equal to 0 [19–21].

2.2. Energy balance equation

Energy balance is expressed in terms of volumetric internal energy, *U*, as follows [20,21]:

$$\frac{dU}{dt} = Q_{\rm cirs} + Q_{\rm visc} = h_{\rm e} S(T_{\rm rf} - T) + \eta \dot{\gamma}^2 \tag{6}$$

with

$$\dot{\gamma} = 2\pi \chi N_{\rm scrap} \tag{7}$$

and

$$S = \frac{\text{surface}_{\text{exchange}}}{\text{volume}_{\text{crystallizer}}} = \frac{2R_e}{R_e^2 - R_i^2}$$
(8)

where η is the flow viscosity, $\dot{\gamma}$ is the shear rate, χ is an adjustment coefficient, *S* is the heat exchange area per unit volume, R_e is exchanger radius and R_i is the equivalent scraper radius h_e , is the heat transfer coefficient between refrigerant fluid (temperature $T_{\rm rf}$) and seawater (temperature T). It is assumed constant along the crystallizer under the condition that the boundary layer is completely replaced after a scraper pass [24,25].

The internal energy, U, is related to seawater temperature, T, and ice volume fraction, ϕ_{i} , by the following equation:

$$U = -\Delta H \rho_i \phi_i + \rho_{sw} [C_0 C \mathbf{p}_s + (1 - C_0) C \mathbf{p}_w] T$$
⁽⁹⁾

Table 1

Characteristic parameters correlations used in the model

Designations	Expression of the correlation		Units	Reference
Ice suspension viscosity	$\eta = \eta_{sw} \left(1 + 2.5 \phi_{i} + 10.05 \phi_{i}^{2} + 2.73 \times 10^{-3} e^{16.6 \phi_{i}} \right)$	(10)	Pa s	21,24,26
Heat transfer coefficient on the seawater side	$h_e = \left(\frac{\lambda_{\rm SW} \rho_{\rm SW} C p_{\rm SW} N_{\rm scrap} N_b}{15\pi}\right)^{1/2} \cdot e^{16.6\frac{k}{2}}$	(11)	W/m ² K	24,25
Ice density	$\rho_i = 916.67 - 0.15T + 3 \times 10^{-4} T^2$	(12)	kg m ⁻³	27
Saturation temperature	With $0.01 < T < -100^{\circ}$ C $T_{sat} = -0.0575C + 1.710523 \times 10^{-3}C^{3/2} - 2.154996 \times 10^{-4}C^2 - 7.53 \times 10^{-4}P$	(13)	°C	28,29

where ΔH is the specific fusion latent heat, Cp_s and Cp_w are salt and water specific heat capacities, ρ_i is ice density, ρ_{sw} is seawater density and C_0 is the initial salt mass fraction. Characteristic parameters of the developed model are given by correlations summarized in Table 1.

2.3. Model discretization method

The developed mathematical model presented earlier is based on PBE, which is a first-order partial differential equation, where crystal size distribution depends on time and crystal size. PBE is coupled with energy balance described by ordinary differential equation (ODE).

Much effort has been invested in developing appropriate numerical resolution techniques for PBE such as finite difference method, finite volume method, finite element method and method of moments. This later is an alternative to direct discretization approach, commonly used for PBE numerical solution. It transforms PBE into a set of ODEs [20,21,30].

Since the moment *j* is defined as follows:

$$\mu_j = \int_0^\infty L^j n(L,t) dL \tag{14}$$

Multiplying the equation PBE (Eq. (1)) by *U*, and then integrating both sides with respect to *L* (from L = 0 to $L = \infty$), the transformed PBE leads to a set of the following linear algebraic equations in terms of moments:

$$\frac{d\mu_j}{dt} = jG\mu_{j-1} + NL_c^j + Bc_j\mu_{j+1}$$
(15)

with

$$B = \varepsilon N_{scrap}$$
(16)

And c_j is a constant depending on j and given by: $c_j = (2^{1-j/3} - 1)$.

The first four moments (j = 0–3) have physical signification and represent, for a unit volume, particles total number (μ_0), sum of diameters (μ_1), sum of their surfaces (μ_2), and sum of their volumes (μ_3), respectively [31–33]. Ice crystal is considered as spherical particles. Total ice volume fraction is given by [20,21]:

$$\phi_i = \frac{\pi}{6} \mu_3 \tag{17}$$

In the sequel, the mathematical model developed for ice suspension seawater crystallization in SSHE for freezing pretreatment of the new proposed hybrid desalination process is expressed as follows:

$$\frac{d\mu_0}{dt} = N + B\mu_1 \tag{18}$$

$$\frac{d\mu_1}{dt} = (G\mu_1) + (NL_c) + (c_1 B\mu_2)$$
(19)

$$\frac{d\mu_2}{dt} = (2G\mu_1) + (NL_c^2) + (c_2B\mu_3)$$
(20)

$$\frac{d\mu_3}{dt} = (3G\mu_2) + (NL_c^3)$$
(21)

$$\frac{dT}{dt} = \frac{1}{\rho_{sw}[C_0 C p_s + (1 - C_0) C p_w]} \left[S h_e(T_{rf} - T) + \eta \dot{\gamma}^2 + \frac{\pi}{6} \Delta H \rho_i[(3G\mu_2) + (NL_e^3)] \right]$$
(22)

The set of five differential equations [Eqs. (18)–(22)] of the developed mathematical model includes five state variables ($\mu_{i'}$ j = 0 to 3 and T) that are the model outputs. The model accounts 11 parameters ($T_{sat'} \eta$, $h_{e'} \rho_{i'} \alpha$, β , ε , χ , $N_{scrap'} T_{ri}$). The four ones ($T_{sat'} \eta$, $h_{e'} \rho_i$) are given by correlations according to Table 1. Analysis of degree of freedom shows that the parameters (α , β , ε , χ , $N_{scrap'} T_{ri}$) must be specified to uniquely determine a feasible solution to the proposed mathematical model. Therefore, these input parameters will be set in from literature in the following section.

3. Model simulation

3.1. Model numerical resolution method

The mathematical model developed above for ice seawater SSHE crystallizer is represented by a set of ODEs. Fourth order Runge–Kutta method is commonly used for solving such set of equations.

Fig. 3 presents the flow chart considered for solving the mathematical model equations. It was developed using FORTRAN.

3.2. Model simulation data

The developed phenomenological model concerning the seawater freezing pretreatment process of the new hybrid desalination technology was not suggested elsewhere. Simulation of such model allows understanding of



Fig. 3. Flow chart for solving the mathematical model developed.

all occurred phenomenon and it permits to describe and estimate ice crystallization characteristics in SSHE that aid the complete design of the new freezing pretreatment process since experimental plant is under development. Therefore, simulation data were extracted from the literature studies of the ice crystallization processes in the same crystallizer type used for other solutions.

3.2.1. Crystallizer characteristics

Characteristics of laboratory scale SSHE crystallizer employed for ice crystallization process of sorbet were used according to Table 2 [19–21,34].

3.2.2. Operating condition

Data related to operating conditions include inlet seawater characteristic quantities, refrigerant fluid temperature, and crystallization kinetics, as follow:

- Inlet seawater characteristic quantities are summarized in Table 3 [29].
- The freezing point of seawater at 3.5% salt concentration is equal to nearly -2°C, so the refrigerant fluid operating temperature, T_{rf} is taken equal to -6°C.
- Kinetic data are presented in Table 4. It is must be noted that kinetic data for ice seawater crystallization process in SSHE are not available in literature; they were extracted from studies conducted in the same kind of crystallizer for ice crystallization from sorbet [19–21].

Table 2 SSHE characteristics used

Symbols	Designations	Model
		values
<i>m_{sw},</i> kg/h	Mass flow rate	50
<i>V</i> , m ³	Available volume of the freezer	$3.87\times10^{\scriptscriptstyle -4}$
	to the fluid	
$S, { m m}^{-1}$	Heat exchange area per unit volume	135.5
<i>H,</i> m	Feature size of the freezer	0.4
$N_{_{ m scrap'}} m rpm$	Dasher rotation speed	450
$N_{\rm b}$	Dasher scraper blades number	2

		-		
Inl	et s	seawater	characteristic	quantities

Table 3

Symbols	Designations	Values
C_0	Initial salt mass fraction	0.035
<i>T</i> ₀ , K	Initial seawater temperature	273.15
$\rho_{sw'} \text{ kg/m}^3$	Seawater density (0°C, 35 g/L)	1,028.11
$\lambda_{sw'}$ W/m K	Thermal conductivity (0°C, 35 g/L)	0.563
μ _{sw} , m/s	Velocity (0°C, 35 g/L)	1,449
ΔH , J/kg	Specific fusion latent heat	334×10^3
Cp _{s'} J/kg K	Salt-specific heat capacity	880
Cp _w J/kg K	Water-specific heat capacity	4,226

Table 4 Kinetic data values [19–21]

Symbols	Designations	Values
$A, \mathrm{s}^{-1}\mathrm{m}^{-2}$	Nucleation coefficient	10^{8}
<i>B</i> , m s ⁻¹ K ⁻¹	Growth coefficient	3×10^{-6}
<i>E</i> , m ⁻¹	Breakage coefficient	5
X	Viscous dissipation	17
L _c , μm	Initial crystal size	5

3.3. Simulation results and discussion

The model developed in this paper is used to simulate ice suspension seawater crystallization process phenomena and parameters trends for seawater pretreatment in SSHE. Many simulation results have been obtained. The most important ones are ice volume fraction and ice suspension temperature.

Simulation results were obtained by integrating the five differential equations [Eqs. (16)–(19)] for total residence time, $t_{,r}$ expressed as follows:

$$t_s = \frac{V_{sw}}{V} \tag{23}$$

where t_s is total residence time, V_{sw} is volumetric flow rate and *V* is the available volume of the fluid in SSHE.

The model predicted variation of ice suspension temperature (ABCD) inside SSHE is depicted in Fig. 4.

Before point (A), it shows a rapid decrease of seawater temperature from the inlet temperature at $T_0 = 0^{\circ}$ C to reach the initial freezing point at $T_{sat} = -1.9^{\circ}$ C. Thus, within this first time, ice crystallization operating condition ($T > T_{sat}$) are not satisfied. Then, seawater temperature continues to drop until the point (B) at $T_B = -3.9^{\circ}$ C that represents the minimum temperature value reached by the suspension at about t = 0.3 residence time. This period (AB) corresponds to the beginning of ice crystallization mechanisms.

Subsequently, a gradual increase in ice suspension temperature can be observed in the time period from point (B) to (C). The suspension temperature, from point (C) to (D), increases further but at a slower rate. At this time period (CD), suspension temperature still lowers than T_{sat} and ice crystallization continuous. At the SSHE outlet, it can be seen that the ice suspension reaches nearly an equilibrium temperature.

Ice volume fraction evolution predicted by SSHE model is presented in Fig. 5.

Before point (A): $T > T_{sat}$ (cf. Fig. 4), so it can be deduced that ice crystals have not yet been formed. Ice crystallization starts when suspension temperature reaches initial freezing temperature value ($T_{sat} = -1.9^{\circ}$ C) at point (A). Thereafter, ice volume fraction shows an increase and hence, ice crystals continuous to be produced until ice suspension reaches SSHE exit.

The variation of suspension temperature accompanied by the increasing of ice volume fraction can be interpreted by the beginning of ice crystallization phenomena at $T \le T_{sat}$ that continuous until the SSHE inlet. Indeed, ice volume fraction increase resulted in a large amount of heat production and increasing of solute concentration in unfrozen seawater.

Fig. 4. Model predicted suspension temperature vs time.



Fig. 5. Model predicted ice volume fraction vs time.

These can explain the increase of suspension temperature at the time period from point (B).

Therefore, a positive effect is expected in terms of pretreated water quality and process performance. The suspension temperature increase implies lower growth ice crystallization rate and leads to smaller crystals; hence, high purity of ice produced [19,35].

The all obtained results using the developed phenomenological model allow predicting satisfactory and physically interpreted trends.

In the lack of industrial and experimental data for ice suspension crystallization from seawater in SSHE, a comparison of the obtained simulation results according to the developed model with other ice crystallization studies was performed. Two ice crystallization processes modeling case studies were exploited. The two models studies comprise of coupled kinetics, population, and energy balances.

The first case study is a computer-based dynamic model of KNO_3 -water solution eutectic freeze crystallization in Figs. 6(a) and (b) shows model predicted mean values of ice suspension temperature and ice volume fraction. In this case, appropriate study experiments were under development to show the model reliance. However, it has concluded that this model plays an important role to identify the significant variables which have to be measured and aids the design of the complete apparatus [36].

The second case study is a mathematical model of ice crystallization process of sorbet in a continuous SSHE [20].



Fig. 6. Model predicted mean values for KNO₃–water solution eutectic freeze crystallization of (a) ice suspension temperature and (b) ice volume fraction [36].

Figs. 7(a) and (b) show model predicted mean values of ice suspension temperature and ice volume fraction. A comparison of the calculated simulation results from this model with the experimental data for different operating conditions shows that the model predicted very satisfactorily tendencies within a 10% error limit.

It can be clearly shown from this comparison analysis that the obtained simulation results trends of the main key variables (i.e., ice suspension temperature and ice volume fraction), calculated from the developed ice seawater SSHE pretreatment crystallizer model, are confirmed by the trends obtained for the same kind of ice crystallization process conducted from other solutions. Therefore, the developed model allows adequate estimation and physically interpretable trends of ice suspension temperature and ice volume fraction.

It can be used to correctly describe and predict the SSHE behavior main aspects and phenomena for the proposed new seawater freeze pretreatment process.

4. Parametric sensitivity study

In this section, the developed mathematical model was exploited for a parametric sensitivity study in order to identify the main input variables having an appreciable impact on ice seawater crystallization process performance. This later is evaluated in our study by ice volume fraction and ice suspension temperature evolution within the studied SSHE crystallizer.

Generally, the principal input variables affecting the ice crystallization in SSHE are (1) freezing temperature (i.e., refrigerant fluid temperature), (2) residence time (i.e., seawater flow rate), and (3) scraping action (i.e., dasher rotational speed) [19,37]:



Fig. 7. Model predicted mean values for ice crystallization process of sorbet, of (a) ice suspension temperature and (b) ice volume fraction [20].

- 1. The refrigerant fluid temperature determines the heat removal rate of the crystallizer and provides the driving force for ice nucleation and growth.
- 2. The flow rate determines the time available to remove heat from the seawater, and consequently, affecting the ice nucleation and growth.
- 3. The dasher rotational speed improves the heat transfer rate between the crystallizer wall and the seawater, influencing ice nucleation and growth.

As it can be noted, all the above three variables affect both ice nucleation and growth and hence, ice crystallization process

4.1. Operation parameters intervals tested

The above three operating parameters variation intervals used for sensitivity study are taken from the same bibliography studies of similar systems already used in the simulation section. Table 5 summarizes the usual intervals considered.

4.2. Refrigerant fluid temperature effect

Figs. 8 and 9 present, respectively, the ice suspension temperature and the ice volume fraction evolutions during seawater freezing crystallization within the SSHE for different refrigerant fluid temperature. Dasher rotation speed and seawater flow rate are fixed, respectively, at mean values (450 rpm) and (50 kg/h) of the considered intervals.

Fig. 8 indicates that ice suspension temperature evolution curves have the same profiles for different refrigerant fluid temperatures, T_{rr} studied.

The seawater temperature profiles show a rapid decrease from the inlet temperature to reach the same initial freezing point T_{sat} in point (A) but at different times. Then, ice suspension temperature continues to drop until its minimum value at point (B) which differs among refrigerant fluid temperature considered. As it can be seen, when this later decreases from -6°C to -10°C, ice suspension temperature minimum (B) is rabidly reached with a decrease of about 1°C. While at SSHE outlet, the equilibrium temperature (point D) is reached at the same ice suspension temperature (-2.1°C).

Fig. 9 shows that ice crystallization starts (point A) at different moment under different refrigerant fluid temperature. When refrigerant fluid temperature decreases, ice crystallization begins more rabidly and ice volume fraction increases. The decrease of refrigerant fluid temperature from -6° C to -10° C leads to an increase of ice volume fraction from 29% to 67%. Thus, a decrease of 1°C of freezing temperature implies an increase of ice volume fraction evaluated by almost 10%.

Based on the above results, we can deduce that refrigerant fluid temperature, $T_{rf'}$ has a strongest effect on ice volume

Table 5

Process operating parameters data test

Operating parameters	Values
Freezing temperature (T_{rf}), °C	-6 to -10
Seawater flow rate (m_{sw}), kg/h	25–75
Dasher speed ($N_{\rm scrap}$), rpm	250-750



Fig. 8. Ice suspension temperatures evolution at different refrigerant fluid temperature.



Fig. 9. Ice volume fractions evolution at different refrigerant fluid temperature.

fraction more than on crystallizer outlet ice suspension temperature. Indeed, decreasing refrigerant fluid temperature improved heat transfer and implies that ice suspension temperature decreases more rapidly (Fig. 8, AB). That facilitates further ice crystallization and thus ice volume fraction (Fig. 9) and consequently solute concentration in unfrozen seawater solution increase. This explains the later increasing of suspension temperature at crystallizer outlet (Fig. 8, BC) leading to the same equilibrium temperature (*D*). This can be explained by the compensatory effect due to the ice volume fraction increase within the crystallizer.

4.3. Flow rate effect

Figs. 10 and 11 depict seawater flow rate variation effect on both ice suspension temperature and ice volume fraction, respectively, during seawater freezing crystallization in the system investigated. While Dasher rotation speed and freezing temperature are considered, respectively, to 450 rpm and -7° C.

Ice suspension temperature curves (Fig. 10) have practically the same variations (ABCD) regardless to the studied flow rate. Nevertheless, when the crystallizer seawater flow rate varies from 25 to 75 kg/h, outlet ice suspension temperature (equilibrium temperature, point D) decreases slightly by about 0.1°C.



Fig. 10. Ice suspension temperatures evolution with different seawater flow rate.



Fig. 11. Ice volume fractions evolution with different seawater flow rate.

Fig. 11 shows that lower ice volume fraction is obtained by increasing the seawater flow rate. Its increase from 25 to 75 kg/h leads to the ice volume fraction decrease from 62% to 23%.

Thus, we can deduct that there is no significant seawater flow rate, m_{sw} , variation influence on ice suspension temperature. Nevertheless, the slight decrease observed is due to the decrease of ice suspension residence time.

Furthermore, a decreasing of seawater flow rate and thus increasing contact time (residence time) permit enhancing heat transfer between seawater and refrigerant and consequently increasing ice crystallization rate, which allows considerably increasing the ice volume fraction.

4.4. Dasher rotation speed effect

Figs. 12 and 13 present, respectively, ice suspension temperature and ice volume fraction evolutions at different dasher rotation speed with similar flow rate and freezing temperature which are fixed, respectively, to 50 kg/h and -7° C.

Fig. 12 shows that ice suspension minimum temperature value (point B) changes clearly with the dasher speed variation. When this later varies from 250 to 750 rpm, ice suspension minimum temperature decreases by nearly 0.7°C. At SSHE outlet, ice suspension reaches the same equilibrium temperature.



Fig. 12. Ice suspension temperatures evolution at different dasher speed.



Fig. 13. Ice volume fractions evolution at different dasher speed.

Fig. 13 indicates that ice volume fraction increases with increasing of dasher speed. The variation of dasher speed from 250 to 750 rpm implies ice volume fraction increase from 28% to 49%.

Indeed, dasher speed increase avoids ice crystals deposition at freezer wall. This enhances the heat transfer rates and consequently ice suspension temperature decrease and ice volume fraction increase. Furthermore, higher dasher speed generates energy amount dissipated into the seawater suspension which leads to a slight warming effect. This warming partially compensates the heat transfer rate improvement. Thus, at SSHE outlet, ice suspension reaches the same equilibrium temperature regardless to the considered dasher speed.

Therefore, the dasher speed, $V_{\text{scrap}'}$ variation appears to has a very slight effect on SSHE outlet ice suspension seawater temperature than on ice volume fraction.

The overall parametric sensitivity study results obtained proves that the ice seawater produced quantity is strongly influenced by the variation of operating parameter studied. Whereas the ice quality has not significantly influenced, since the minimum temperature reached by ice suspension is still greater than -6° C, the temperature of (Na₂SO₄, 10H₂O) precipitation which would alter the purity of ice crystals produced.

5. Conclusion

A phenomenological model of ice suspension seawater SSHE crystallizer has been developed for a proposed new seawater pretreatment process. The mathematical model is based on combining energy and PBEs taking into account main phenomena involved in the SSHE crystallizer such as nucleation, growth, and breakage. To our knowledge, such phenomenological model was never suggested for freezing seawater pretreatment in such crystallizer.

Obtained simulation results showed that the developed model is able to describe SSHE behavior main aspects and predict physically interpretable trends. These results were confirmed by comparison with studies of the same ice crystallization process from other solutions since appropriate experiments are under development to show model's reliance. The developed model is able to satisfactorily describe SSHE behavior main key aspects and phenomena (i.e., ice suspension temperature and ice volume fraction). Further, subsequent parametric sensitivity study that consists of a precise identification of appreciable parameters impact on ice seawater crystallizer process is conducted using this model.

At the SSHE outlet, it can be seen that the ice suspension reaches nearly an equilibrium temperature (-2.1° C). Which means that the ice quality has not significantly influenced; since the minimum temperature reached by ice suspension is still greater than -6° C, the temperature of (Na₂SO₄, 10H₂O) precipitation which would alter the purity of ice crystals produced. Whereas the ice produced quantity is strongly influenced by the variation of freezing temperature (i.e., refrigerant fluid temperature), residence time (i.e., seawater flow rate), and scraping action (i.e., dasher rotational speed). The variation of these parameters has enabled to have up to 65% of produced ice.

We can conclude that the developed model can be useful to gain a deeper understanding of the involved phenomena so as identifying ice suspension seawater crystallization significant variables in SSHEs. Thus, it allows to complete design of the new freezing pretreatment process and to optimize the proposed hybrid seawater desalination process performance.

Symbols

п	—	Number of crystals per meter (of the
		crystallizer) per cubic meter of the solution
		at the outlet of the SSHE, m ⁻⁴
μ_i	_	<i>j</i> th order moment, m ^{<i>j</i>-3}
t	_	Time variable, s
t	_	Residence time, s
Ľ	_	Crystal size variable, m
ī.	_	Initial crystal size, m
R^{-c}	_	Crystallizer minimum diameter m
R	_	Crystallizer maximum diameter m
G.	_	Growth rate of the crystals $m s^{-1}$
N	_	Nucleation rate $m^{-4} e^{-1}$
B	_	Not increase of crystals number by
D_{b}	_	hreakage m ⁻⁴ e ⁻¹
\$		Europhicon direct
0 T	_	Function dirac
I sat	_	Saturation temperature, °C
I rf	_	freezing temperature, °C
T	—	Ice suspension temperature, °C
T_0	_	Inlet seawater temperature, °C
α	—	Surface nucleation constant, $m^{-2} s^{-1} K^{-2}$
β	—	Growth constant, m s ^{-1} K ^{-1}
V	—	Volume of the crystallizer, m ³
ϕ_i	—	Ice fraction
N _{scrap}	—	Dasher rotation speed, r s ⁻¹
N _b	—	Dasher scraper blades number
ν	_	Breakage power coefficient
∈	_	Breakage constant, m ⁻¹
С	_	Mass fraction of solutes in the unfrozen
		phase
C_{\circ}	_	Initial mass fraction of solute (salt)
ρ.	_	Mass density of ice, kg m^{-3}
D D	_	Mass density of seawater, kg m^{-3}
λ	_	Thermal conductivity, W/m K
11 II	_	Volumetric internal energy. I m ⁻³
0	_	Heat flow through the wall of the
$\kappa_{\rm cris}$		crystallizer W
0	_	Heat production caused by viscous
$\mathcal{Q}_{\text{visc}}$		dissipation W
И		Viscosity Pac
ν γ	_	r_{1}
1	_	Viacosity of the unfrozon phase. Dec
η _{sw}	_	Viscosity of the unirozen phase, Pas
χ	_	Viscous dissipation coefficient
ΔH	_	Specific fusion latent heat, J kg
Cp _s	_	Solute specific heat capacity, J kg ⁻¹ K ⁻¹
Cp _w	—	Water specific heat capacity, J kg ⁻¹ K ⁻¹
h_{e}	—	Convective heat transfer coefficient,
		$Wm^{-2}K^{-1}$
S	—	Ratio of the periphery over the surface of
		the section, m^{-1}
m _{sw}	_	Inlet mass flow rate, kg s ⁻¹
V _{sw}	—	Inlet volume flow rate, m ³ s ⁻¹
Nbpas	_	Number of steps
Pas Temps	_	Time step of calculation, s

References

- J. Sánchez, Y. Ruiz, J.M. Auleda, E. Hernández, M. Raventós, Freeze concentration in the fruit juices industry, Food Sci. Technol. Int., 15 (2009) 303–315.
- [2] J. Sánchez, E. Hernández, J.M. Auleda, M. Raventós, Freeze concentration technology applied to dairy products, Food Sci. Technol. Int., 17 (2011) 5–13.
- [3] M. Kapembwa, M. Rodriguez-Pascual, A.E. Lewis, Heat and mass transfer effects on ice growth mechanisms in pure water and aqueous solutions, Cryst. Growth Des., 14 (2013) 389–395.
- [4] L. Liu, O. Miyawaki, K. Nakamura, Progressive freeze concentration of model liquid food, Food Sci. Technol. Int., 3 (1997) 348–352.
- [5] S. Lemmer, R. Klomp, R. Ruemekorf, R. Scholz, Preconcentration of wastewater through the Niro freeze concentration process, Chem. Eng. Technol., 24 (2001) 485–488.
- [6] H. Jansen, M.A. Hernandez, A. Martinez, Concentración por congelación de disoluciones acuosas: un nuevo método para obtener productos innovadores de alta calidad, CTC Alimentacion, 10 (2001) 13–15.
- [7] M. Van Nistelrooij, Bridging the Cost Barrier to Freeze Concentration, Food and Beverage Asia, 2005.
- [8] M. Harrod, Scraped surface heat exchangers: a literature survey of flow patterns, mixing effects, residence time distribution, heat transfer and power requirements, J. Food Process Eng., 9 (1986) 1–62.
- [9] S.C. Rao, R.W. Hartel, Scraped surface heat exchangers, Crit. Rev. Food Sci. Nutr., 46 (2006) 207–219.
- [10] L. Vrbka, P. Jungwirth, Brine rejection from freezing salt solutions: a molecular dynamics study, Phys. Rev. Lett., 95 (2005) 148501.
- [11] M.V. Rane, Y.S. Padiya, Heat pump operated freeze concentration system with tubular heat exchanger for seawater desalination, Energy Sustain. Dev., 15 (2011) 184–191.
- [12] A. Attia, New proposed system for freeze water desalination using auto reversed R-22 vapor compression heat pump, Desalination, 254 (2010) 179–184.
- [13] R. Fujioka, L.P. Wang, G. Dodbiba, T. Fujita, Application of progressive freeze concentration for desalination, Desalination, 319 (2013) 33–37.
- [14] T. Mtombeni, J.P. Maree, C.M. Zvinowanda, J.K.O. Asante, F.S. Oosthuizen, W.J. Louw, Evaluation of the performance of a new freeze desalination technology, Int. J. Environ. Sci. Technol., 10 (2013) 545–550.
- [15] G. Pénghui, G. Zhi, Z. Donghai, Z. Xingye, Z. Guoqing, Performance analysis of evaporation-freezing desalination system by humidity differences, Desalination, 347 (2014) 215–223.
- [16] A. Rich, Y. Mandri, D. Mangin, A. Rivoire, S. Abderafi, C. Bebon, N. Semlali, J.-P. Klein, T. Bounahmidi, A. Bouhaouss, S. Veesler, Sea water desalination by dynamic layer melt crystallization: parametric study of the freezing and sweating steps, J. Cryst. Growth., 342 (2012) 110–116.
- [17] P.M. Williams, M. Ahmad, B.S. Connolly, D.L Oatley-Radcliffe, Freeze desalination: an assessment of an ice maker machine for desalting brines, Desalination, 356 (2015) 314–327.
- [18] I. Baayyad, N. Semlali Aouragh Hassani, T. Bounahmidi, Evaluation of the energy consumption of industrial hybrid seawater desalination process combining freezing system and reverse osmosis, Desal. Wat. Treat., 56 (2015) 2593–2601.
- [19] J.E. Gonzalez, M. Arellano, D. Leducq, G. Alvarez, H. Benkhelifa, D. Flick, Effect of Sorbet Freezing Process on Draw Temperature and Ice Crystal Size Using Focused Beam Reflectance Method (FBRM) Online Measurements, Proc. 23rd IIR International Congress of Refrigeration, Refrigeration for Sustainable Development, August 21–26, Prague, Czech Republic, 2011.
- [20] M. Arellano, H. Benkhelifa, G. Alvarez, D. Flick., Coupling population balance and residence time distribution for the ice crystallization modeling in a scraped surface heat exchanger, Chem. Eng. Sci., 102 (2013) 502–513.
- [21] C. Casenave, D. Dochain, G. Alvarez, M. Arellano, H. Benkhelifa, D. Leducq, Model identification and reduction for the control of an ice cream crystallization process, Chem. Eng. Sci., 119 (2014) 274–287.

- [22] R.W. Hartel, Crystallization in Foods, Aspen Publishers, Gaithersburg, 2001.
- [23] K.L.K. Cook, R.W. Hartel, Mechanisms of ice crystallization in ice cream production, Compr. Rev. Food Sci. Food Safety, 9 (2010) 213–222.
- [24] R.J.C. Vaessen, M.M. Seckler, G.J. Witkamp, Heat transfer in scraped eutectic crystallizers, Int. J. Heat Mass Transfer, 47 (2004) 717–728.
- [25] F.G.F. Qin, X.D. Chen, S. Ramachandra, K. Free, Heat transfer and power consumption in a scraped-surface heat exchanger while freezing aqueous solutions, Sep. Purif. Technol., 48 (2006) 150–158.
- [26] D.G. Thomas, Transport characteristics of suspension: VIII. A note the viscosity of Newtonian suspensions of uniform spherical particles, J. Colloid Sci., 20 (1965) 267–277.
- [27] W. Hyland, A. Wexler, Formulations for the thermodynamic properties of the saturated phases of H₂O from 173.15 to 473.15 K, ASHRAE Trans., 89 (1983) (2A) 500–519.
- [28] F.J. Millero, W.H. Leung, Thermodynamics of seawater at one atmosphere, Am. J. Sci., 276 (1976) 1035–1077.
- [29] A. Rich, Dessalement de l'eau de mer par congélation sur parois froides: aspect thermodynamique et influence des conditions opératoires, PhD thesis, Université Claude Bernard, France, 2011.
- [30] A. Randolph, Theory of Particulate Processes: Analysis and Techniques of Continuous Crystallization, Academic Press, New York, USA, 1971.

- [31] C. Costa, M. Maciel, R. Filho, Considerations on the crystallization modeling: population balance solution, Comput. Chem. Eng., 31 (2007) 206–218.
- [32] B.R. Diemer, H.J. Olson, A moment methodology for coagulation and breakage problems: Part 1 – analytical solution of the steady-state population balance, Chem. Eng. Sci., 57 (2002) 2193–2209.
- [33] R.B. Diemer, J.H. Olson, A moment methodology for coagulation and breakage problems: Part 2 moment models and distribution reconstruction, Chem. Eng. Sci., 57 (2002) 2211–2228.
- [34] E. Gonzalez, Contribution au controle par la modélisation d'un procédé de cristallisation en continu, PhD thesis, Agro Paris Tech, Paris, France, 2012.
- [35] Z. Zhang, R.W. Hartel, A multilayer for freeze concentration of liquid milk, J. Food Eng., 29 (1996) 23–38.
- [36] C. Himawan, R.J.C. Vaessen, H.J.M. Kramer, M.M. Seckler, G.J. Witkamp., Dynamic modeling and simulation of eutectic freeze crystallization, J. Cryst. Growth, 237–239 (2002) 2257–2263.
- [37] Marcela Patricia Arellano Salazar, Experimental Characterization and Modelling of Multiphase Systems During the Freezing Process at the Pilot Scale: Application to Sorbet Manufacturing in Scraped Surface Heat Exchangers, Food Engineering, AgroParisTech, 2012.

44