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Experimental and CFD study on influence of viscosity on layer melt crystallization

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ABSTRACT

In the present work, the influence of solution viscosity on growth kinetics and purification efficiency in layer melt crystallization was investigated. Melt crystallization experiments were conducted for three different types of aqueous sucrose solution as they are ideal solutions and a relatively wide viscosity range can be investigated with a moderate change of freezing points. The aqueous 10 wt%, 23 wt%, and 30 wt% sucrose solutions have a dynamic viscosity value of 2.01 mPas, 4.74 mPas, and 7.21 mPas at their respective freezing points of –0.63 °C, –1.78 °C, and –2.64 °C. The solution temperature distribution was predicted by computational fluid dynamics (CFD) simulations run in COMSOL Multiphysics 5.6 software. Experimental results showed that a higher solution viscosity caused a higher crystal layer impurity and lower crystal yields in static layer melt crystallization. The cooling process of different solutions predicted by a CFD heat transfer study showed that the supersaturation region is wider for less concentrated solutions as cooling proceeds more rapidly. Hence, the temperature gradients obtained follow the boundary layer theory, i.e., the thinner the boundary layer, the faster the heat transfer.

1. Introduction

Industrial production based on sustainable development and circular economy principles is imperative to ensure continuous economic growth with a minimized negative environmental impact by the industrial sector. Industrial energy consumption contributes to half of total global energy consumption and it is expected to increase by 30% by the middle of the century [1]. Industrial separation processes account for 22% of total energy consumption in the USA and high-energy separation such as distillation accounts for 10–20% of total energy consumption in the USA and high-energy separation such as distillation accounts for 10–15% of global energy consumption [2,3]. Hence, the development and application of industrial separation technologies that are less energy demanding and more environmentally friendly should be prioritized.

Melt crystallization is a low-energy industrial separation technique used for the purification and separation of compounds up to a very high level of purity. The low energy consumption of melt crystallization is the result of the property of the chemical, and that is a significantly lower latent heat of fusion than the latent heat of vaporization. It is an environmentally safe technology as there is no potential risk of leakage of vapors, nor is there any generation of waste streams because of the absence of vapor phase production and usage of additional substances [4].

The melt crystallization method is defined as the formation and growth of crystals from melt at a temperature close to its melting point with undercooling, or supercooling, as a driving force. Therefore, the progress of crystallization is affected by the difference between the temperature of the solution and the surface temperature of the growing crystalline layer [5].

The melt crystallization technique can be divided into two groups: layer crystallization and suspension crystallization. In layer melt crystallization, a crystalline layer forms and grows perpendicularly onto the undercooled surface of a crystallizer, whereas in suspension melt crystallization, crystals form and freely grow, dispersed in the solution. One special case of melt crystallization is freeze crystallization, which refers to the crystallization of water from aqueous solutions. Consequently, the freeze crystallization process can be performed either as layer freeze...
crystallization or suspension freeze crystallization. The simultaneous crystallization of water and solute that forms a eutectic system, at their eutectic point, is referred to as eutectic freeze crystallization.

Layer melt crystallization is usually applied for crystallization of melts with high viscosities, as high viscosity is a limiting factor for the application of suspension melt crystallization [6]. Some recently reported applications of layer melt crystallization include the separation of isomeric compounds [7–10], purification of nuclear waste fuel salts from radioactive ions [11], production of highly dry glycerol [12], and separation of phenolic compounds from carbohydrates [13]. Layer freeze crystallization has also been reported to be an efficient purification method for urban and industrial wastewater [14,15].

As a result of the broad range of layer melt crystallization applications, the present study investigates the influence of the thermophysical properties of the solution on layer melt crystallization and its kinetics. Layer growth kinetics affects the final purity of the formed layer. The crystal purity of single crystals depends mainly on the crystal growth rate, whereas in layer melt crystallization the secondary growth mechanism of the forming layer can also have a significant effect on the purity level. The latter growth mechanism occurs when the melt inclusions are entrapped in the voids of the layer formed by single grain crystals [16]. The solutes present in the solution, especially at higher concentrations, can affect the size and shape of single grain crystals [15]. According to the results of a previous study on the influence of viscosity on the crystal growth of single crystals, the growth rate of a high-viscosity solution was lower while the impurity concentration was higher than for a low-viscosity solution [17]. Moreover, the impurity of the crystalline layer increases as the growth rate increases [18,19].

Studies based on the mathematical modeling of different layer crystallization methods, such as static and falling layer crystallization with or without recirculation, have been reported [20–24] as well as the application of direct numerical simulation on static layer melt crystallization [25]. To the best of our knowledge based on a literature review, heat transfer modeling of layer melt crystallization by CFD has not been investigated more comprehensively.

In the present work, aqueous sucrose solution was selected as a model compound system for investigating the influence of solution viscosity on static layer melt crystallization. Aqueous sucrose solutions are ideal in a concentration range from 0.5 to 42 wt%, based on determination of water activity coefficients using the Van’t Hoff equation, which is in good agreement with previously reported data [26,27]. Ice layers were crystallized from aqueous sucrose solutions with three different solution concentrations to alter the viscosity in layer melt crystallization experiments initiated by ice seeding. Crystal yields and the distribution coefficient were used to compare the efficiency of melt crystallization for solutions with different viscosities. The temperature distributions of solutions prior to crystallization were predicted by CFD simulation.

2. Materials and methods

2.1. Experimental study of layer melt crystallization

Aqueous sucrose solutions containing 10 wt%, 23 wt%, and 30 wt% of sucrose were prepared by dissolving food grade sucrose in deionized water (0.56 µS/cm) under mixing conditions of 380 rpm for 60 min. Stagnant layer freeze crystallization experiments were conducted using a crystallizer consisting of a jacketed vessel equipped with a cylindrical cold finger. The outer diameter of the cold finger was 10 mm and the volume of the vessel was 250 mL with an inner diameter of 70 mm. The jacketed vessel and cold finger were connected to two separate Lauda ECO RE 1050 thermostats. The thermostats were used to control the temperatures of the coolant circulating through the jacket and the coolant circulating in the cold finger, as these temperatures represented the melting point and desired subcooling temperature of the solution, respectively. A detailed explanation of the layer melt crystallization set-up and experimental procedure was given in a previous study [28].

Experiments were conducted for degrees of subcooling in a range of 0.76–1.9 °C and for three crystallization times of 40 min, 60 min, and 80 min for aqueous 10 wt% and 23 wt% sucrose solutions and two crystallization times of 60 min and 80 min for an aqueous 30 wt% sucrose solution. The crystallization time of 40 min was disregarded in the case of layer melt crystallization of aqueous 30 wt% sucrose solution due to the formation of very delicate and fragile crystalline layers with a tendency to disintegrate.

For each experiment, a batch of 250 mL solution was poured into the vessel and the cold finger was immersed in the solution. When the set temperature values at the surface of the tip of the cold finger and in the solution were reached, layer crystallization was induced by ice crystal seeding. When a certain solution temperature was achieved, the cold finger was taken out from the crystallizer and an ice crystal seed was attached in the middle of the tip of the cold finger. Then, the cold finger was immediately re-immersed back into the solution and the layer was left to grow on the cold finger for a certain crystallization time. The moment when the ice seeding was added was considered as the beginning of the crystallization process.

At the end of crystallization, the formed ice layer was detached from the cold finger. The ice layer sample was washed with 2 mL of water at 0 °C to remove any remaining mother liquor.

Then, the dimensions (outer diameter and height) and mass of the ice layer sample were measured. Layer purity was determined based on the reflective index measurements of the melted ice samples. The measured reflective indexes of the melted ice layers were correlated with the available literature data on sucrose content in aqueous solutions [29] which represented the amount of impurities in the ice layer.

2.2. Crystallization parameters

The separation efficiency of layer melt crystallization was evaluated using two parameters: crystal yield and effective distribution coefficient. These parameters were determined based on experimentally obtained data on the amount of ice, the purity of the ice layer, and the concentration of the initial solution.

The crystal yield, Y, is expressed as a percentage of mass ratio between the crystallized solvent, (obtained ice quantity) and the initial solvent quantity (initial water amount). The amount of crystallized solvent is the mass of the pure ice layer determined based on the mass of the melted ice sample and reflective index purity analysis.

The effective distribution coefficient, K, is defined as the impurity concentration ratio between an impurity entrapped in the crystalline layer and the initial impurity concentration of the feed solution.

2.3. Dynamic viscosity and density

The dynamic viscosity of a solution is the highest at the crystallization temperature when the solution is undercooled down to a certain melt crystallization temperature, slightly lower than the equilibrium melting point. Therefore, the dynamic viscosities of three types of aqueous sucrose solutions at their freezing points [29] were predicted by the Pidoux formula [30] and measured with a Brookfield viscometer.

The Pidoux formula is given by Eq. (1) and the required parameters by Eqs. (2–4)

\[ \log \eta = a \phi + b \] (1)

\[ \phi = \frac{T - T_i}{T^2} \] (2)

\[ a = \log \eta_1 - \log \eta_2 \] (3)

\[ b = \log \eta_1 - a \phi_1 \] (4)
where: \( \eta \) calculated dynamic viscosity (mPas) 
\( T \) absolute temperature (K) at which the viscosity is calculated 
\( \eta_1, \eta_2 \) known viscosities (mPa s) at temperatures \( T_1 \) and \( T_2 \) 
\( T_0 \) absolute temperature (K) of melting ice

The extrapolated values were represented by the calculated viscosities of the solutions at their corresponding melting points because only the viscosity values for temperatures above 0 \(^\circ\)C were reported in the literature [29].

The dynamic viscosities of solutions at their freezing points were also measured with a Brookfield LVDV-E Viscometer, with standard uncertainty of 0.24 mPas (expanded uncertainty of 0.62 mPas). The viscosity measurement of samples in a low viscosity range at sub-zero temperature level was performed using a UL Adapter, consisting of a jacketed chamber tube and cylindrical spindle. The UL Adapter jacket was connected to a Lauda ECO RE 1050 thermostat that was used to adjust the coolant temperature to the freezing point of the solution. A solution sample of 16 mL was added inside the chamber tube and dynamic viscosity measurements were initiated by enabling spindle rotation when the temperature of the coolant inside the thermostat bath reached the value of the solution freezing point. The three types of aqueous sucrose solutions were proved to be Newtonian fluids, as the dynamic viscosities were constant for different shearing rates applied during measurement.

The density of the solution is an important property in free convection, which affects heat transfer in static layer melt crystallization. The measured density values of the studied sugar solutions at their freezing point were compared with the density values obtained by extrapolation from the values reported in the literature [30,31]. Densities were measured by weighing 100 mL of solution at its freezing point.

The calculated and measured dynamic viscosities and densities of the aqueous 10 wt%, 23 wt%, and 30 wt% sucrose solutions are presented in Table 1.

| Sucrose concentration, wt. % | Freezing point, \(^\circ\)C | Calculated dynamic viscosity, mPa s | Measured dynamic viscosity, mPa s | Calculated density, kg/m\(^3\) | Measured density, kg/m\(^3\) |
|-----------------------------|-----------------|---------------------------------|---------------------------------|-------------------------------|-------------------------------|
| 10                          | -0.63 \(^\circ\)C | 2.01                            | 2.65                            | 1041.98                       | 1040                          |
| 23                          | -1.78 \(^\circ\)C | 4.74                            | 4.62                            | 1099.63                       | 1100                          |
| 30                          | -2.64 \(^\circ\)C | 7.51                            | 7.52                            | 1133.31                       | 1133                          |

2.4. CFD simulation

The CFD heat transfer study was conducted for the three types of solution. In order to model the heat transfer inside the reactor caused by natural convection, the solution density was considered to be a temperature-dependent property. The 3D geometry model representing the solution domain inside the jacketed vessel equipped with a cold finger was created in COMSOL Multiphysics 5.6. As shown in Fig. 1, the model consists of two cylindrical elements that represented the outer surface of the cold finger and the inner wall of the jacketed vessel. The solution domain occupied the 3D domain between the two boundary surfaces. The typical dimensions of the geometry model are given in Table 2. These dimensions represent the laboratory-scale crystallizer dimensions used in the experimental study.

The generated tetrahedral mesh consisted of approx. 9.6 \( \times \) 10\(^5\) elements with an average quality of 0.677. COMSOL was used to solve the non-isothermal Multiphysics \( ^\text{®} \) problem that couples laminar flow and heat transfer in fluids. First-order linear discretization was used for the solution velocity, pressure, and temperature fields.

2.5. Theoretical study on heat transfer inside a melt crystallizer

The heat transfer of the stagnant melt inside the crystallizer was defined as a non-isothermal flow that combines the buoyancy-driven laminar flow and heat transfer inside the fluid. The general heat transfer equation is given by the following expression,

\[
\frac{\partial T}{\partial t} + \rho C_p \bar{u} \cdot \nabla T = \nabla \cdot (k \nabla T)
\]  
(5)

where \( \rho \) is the solution density \([\text{kg/m}^3]\), \( C_p \) is the solution heat capacity at constant pressure \([\text{kJ/(kg K)}]\), \( k \) is the solution thermal conductivity \([\text{W/(m K)}]\), \( T \) is the temperature \([\text{K}]\), and \( \bar{u} \) is the velocity \([\text{m/s}]\).
The heat equation for a solution that is not stirred, and is not exposed to external heat sources can be formulated as a case of pure conductive heat transfer:

$$\rho c_p \frac{dT}{dt} = \nabla \cdot \left( k \nabla T \right).$$  \hspace{1cm} (6)

The boundary conditions defined as the heat flux boundary conditions for the heat flux transferred from the walls of the jacketed reactor ($q_{0j}$ [W/m$^2$]) are presented in Eq. (7) and the heat flux transferred from the surfaces of the cold finger ($q_{0cf}$ [W/m$^2$]) in Eqs. (8),

$$q_{0j} = k \frac{dT}{dn} (r_j, \theta_j, z) = -U_j (T - T_j)$$  \hspace{1cm} (1)

$$q_{0cf} = k \frac{dT}{dn} (r_{cf}, \theta_{cf}, z) = -U_{cf} (T - T_{cf})$$  \hspace{1cm} (8)

where $T_j$ is the temperature of the cooling jacket surface which is equal to the melting point of the solution [K], $T_{cf}$ is the temperature of the cold finger surface which is equal to the subcooling temperature [K], and $U_j$ and $U_{cf}$ are the heat transfer coefficients related to the cooling jacket and cold finger, respectively [W/(m$^2$ K)].

The heat transfer coefficients of the circulating coolant – solid surface of the cooling jacket and cold finger were calculated by Eqs. (9) and (10)

$$U_j = \frac{1}{\frac{1}{h_{cj}} + \frac{\delta_j}{k_j}}$$  \hspace{1cm} (9)

$$U_{cf} = \frac{1}{\frac{1}{h_{cf}} + \frac{\delta_{cf}}{k_{cf}}}$$  \hspace{1cm} (10)

where $h_{cj}$ and $h_{cf}$ are the heat transfer coefficients of the circulating coolant inside the jacket and cold finger, respectively [W/(m$^2$ K)], $\delta_j$ and $\delta_{cf}$ are the thicknesses of the inner wall of the cooling jacket and outer surface of the cold finger [m], and $k_j$ and $k_{cf}$ are the thermal conductivities of the jacket and cold finger materials [W/(m K)].

The convective heat transfer coefficient of the circulating coolant inside the jacket ($h_{cj}$) was calculated from the Nusselt number for a fully developed laminar flow in a rectangular tube, whereas the convective heat transfer coefficient for the coolant inside the cold finger ($h_{cf}$) was calculated from the Nusselt numbers for the internal laminar flow in an annular pipe [32]. The heat transfer coefficients of the circulating coolant – cold finger surface ($U_{cp}$) were estimated to be around six times greater than the heat transfer coefficients of the circulating coolant – cooling jacket surface ($U_j$). The calculated values can be found in the Supplementary Materials (Tables 9). The thermophysical properties of the coolant (1:1 ethylene glycol: water, mass %) were considered constant at a mean temperature of $-1$ °C and were interpolated from literature data [33].

### 2.6. Parameters for heat transfer simulation

The thermophysical properties of aqueous sucrose solutions (density, viscosity, thermal conductivity, and specific heat capacity at constant pressure) were defined as temperature-dependent properties. Values of the thermophysical properties of aqueous sucrose solutions were not reported for temperatures below 0 °C. Thus, the values of these properties for the sub-zero temperature range were extrapolated from the values for the temperature range above 0 °C [30,31,34–36]. The CFD study included the simulation of the cooling process when solutions were cooled down from 3 °C to their respective freezing points and for two subcooling degrees of 0.85 °C and 1.85 °C for one hour. The subcooling degrees used in the CFD studies correspond to the lowest and highest subcooling degree achieved in the experimental study.

### 3. Results and discussions

In the following section the experimental results obtained in the layer melt crystallization studies and the CFD results of the heat transfer phenomena are presented.

#### 3.1. Layer melt crystallization

The crystal yield and distribution coefficients as a function of the
subcooling degree are shown in Fig. 2 for crystallization times of 40, 60, and 80 min. The experimental data can be found in the Supplementary Materials (Tables 1 to 8).

It can be concluded that the crystal yield is a linear function of the subcooling degree and decreases as the solution viscosity increases. The average value of the crystal yield decrease is approximately 20% for the 30 wt% sucrose solution in comparison with the 10 wt% sucrose solution. However, the effective distribution coefficients increase as the viscosity of the solution increases, which indicates that more impurities are entrapped in the crystalline layer. The average increase in the effective distribution coefficient is 29% for the 30 wt% sucrose solution in comparison with the 10 wt% sucrose solution. As the value of the effective distribution coefficient varies for different types of solutions, it can be presumed that the internal layer structures are different, and the volume fractions of impurity inclusions differ. This observation is consistent with conclusions of previous studies that the layer impurity of more concentrated solutions is relatively greater compared to more dilute solutions [37]. It is concluded that higher solute concentrations in solution lead to a higher volume fraction of voids in the layer where the solution can be entrapped. The layers formed from the most viscous solution of 30 wt% sucrose after 60 min of crystallization in the range of

Fig. 3. Freezing points and temperature lines for subcooling degree of 0.85 °C for 60 min of cooling: (a) 10 wt% sucrose, (b) 23 wt% sucrose, and (c) 30 wt% sucrose.

Fig. 4. Freezing points and temperature lines for subcooling degree of 1.85 °C for 60 min of cooling: (a) 10 wt% sucrose, (b) 23 wt% sucrose, and (c) 30 wt% sucrose.
the lowest subcooling degrees were very fragile and thin. Thus, it was difficult to perform the washing step after the crystallization for each layer as some of them were partially broken. This explains the untypical variation between the distribution coefficients of layers formed at four similar subcooling degrees in the range of 0.83–0.87 °C for 60 minutes of crystallization (Fig. 2). The ice layers crystallized on the surface of the cold finger from different solutions, increased thermal resistance of cold finger system for 2 to 3.75 times.

3.2. Heat transfer study

The results of the CFD study are presented as cooling curves and temperature distribution curves for a certain time period. The temperature distribution curves obtained show whether the solution was undercooled (supersaturated) after a certain period of cooling. For each solution the temperature distribution on the horizontal lines (y-axis, R-r) between the cold finger surface and crystallizer wall at different heights (z-axis), was evaluated for two cooling times of 30 and 60 min. In Fig. 1 (a), positions of the horizontal and vertical lines are shown. The temperature distribution curves after 60 min of cooling for subcooling degrees of 0.85 °C and 1.85 °C are shown in Figs. 3 and 4.

In the above figures, 3 and 4, it is apparent that after 60 min of cooling the studied sucrose solutions are undercooled and supersaturated at the subcooling degree of 1.85 °C, while the more concentrated sucrose solutions of 30 wt% and 23 wt% are not fully undercooled at the subcooling degree of 0.85 °C. After 30 min of cooling, solutions are not completely undercooled and there are distinguishable supersaturated and undersaturated solution zones. Therefore, the overall temperature distribution with velocity field vectors at the 30th minute of cooling at a subcooling degree of 0.85 °C is shown in Fig. 5.

A comparison of the cooling process for different solutions was made using the normalized temperature distribution. Normalized temperature is a dimensionless parameter that represents the ratio between the time-dependent difference of a solution temperature and its melting point and the respective subcooling degrees of 0.85 °C and 1.85 °C. The normalized temperature distributions, as a function of the horizontal distance between the cold finger and the wall of the crystallizer at height z = 40 mm (which corresponds to the bottom of the cold finger) after 30 min of cooling, are shown in Fig. 6.

As shown in Fig. 6, the zero-line represents the equilibrium state of solution (saturated solution), while the negative values of the normalized temperature represent the undercooled (supersaturated) solution region. The following observations can be made. First, for the more concentrated and more viscose solution, the supersaturation region is
narrower as the solution cools down more slowly. The boundary layer thickness increases from 20 to 29% for each subsequent decrease in solute concentration. Thus, for a subcooling degree of 0.85 °C the boundary layer thickness for the 10 wt% sucrose solution increases by 55% in comparison with the 30 wt% sucrose solution. Second, the subcooling temperature of the cold finger has a more prominent influence on the cooling process for less concentrated solutions. For the 10 wt% sucrose solution, the thickness is around 10 times greater when the subcooling degree increases from 0.85 °C to 1.85 °C as the solution becomes supersaturated for the former subcooling degree. For the 30 wt% and 23 wt% sucrose solutions, the increase in boundary layer thickness is 1.6 and 1.67 times, respectively. The normal total heat fluxes of cooling jacket and cold finger are presented in Fig. 7.

The heat fluxes transferred through the surface of the cooling jacket and cold finger are initially greater for the more concentrated sucrose solutions, as shown by the results for the lower melting points of the solution. The cooling jacket heat fluxes are slightly higher when the more concentrated solutions are cooled down, but as the solution temperature reaches equilibrium, the heat flux difference for the different solutions is less prominent. At certain moments of cooling, the temperature of all three types of solution becomes lower than the melting point, which corresponds to a negative value of heat flux through the cooling jacket, as shown in Fig. 7 (a). Sucrose solutions of 10, 23, and 30 wt% reach supersaturation after 27, 35, and 38 min of cooling respectively at a subcooling degree of 1.85 °C. In the case of the subcooling of 0.85 °C, supersaturation occurs 9–13 min later than in the case of the subcooling of 1.85 °C.

4. Conclusions

The experimental studies of static layer melt crystallization were conducted for three different aqueous sucrose solutions to investigate the effects of solution viscosity on separation efficiency. CFD simulation on heat transfer inside the melt crystallizer was used to predict how the solution cooling process is influenced by the thermophysical properties—specifically by the viscosity that changes more considerably than the other properties of the solution, such as solution density, with increase of its concentration. Hence, we predicted the undercooling conditions at the surface of the cold finger where the ice layer formation initiates. Based on this experimental study, it is apparent that the crystal yields obtained from more concentrated and more viscose aqueous sucrose solutions are lower and more impurities are entrapped inside the crystalline layers, resulting in a higher distribution coefficient. The more concentrated the solution, the slower the cooling process, and supersaturation naturally occurs later. Therefore, for a more concentrated solution, the boundary layer thickness that represents the thickness of the supersaturation region is narrower than for less concentrated solutions.

CRediT authorship contribution statement

Nahla Osmanbegovic: Investigation, Visualization, Writing – original draft, Writing – review & editing, Validation. Ville Alopaeus: Writing – review & editing, Supervision. Bing Han: Writing – review & editing, Supervision. Ville Vuorinen: Writing – review & editing, Supervision. Marjatta Louhi-Kultanen: Conceptualization, Writing – review & editing, Supervision, Project administration.
Declaration of Competing Interest
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data
Supplementary data to this article can be found online at https://doi.org/10.1016/j.seppur.2021.120170.

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