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EXPERIMENTAL AND NUMERICAL INVESTIGATION OF DESALINATION BY INDIRECT DIRECTIONAL FREEZING

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ABSTRACT

Only 20% of energy consumed by conventional thermal desalination processes is needed for a successful freeze desalination process. Freeze desalination (FD) is a freezing-melting technology where freshwater in form of ice blocks is separated by cooling from salt-water solution. Freezing in directional configuration found to be advantageous for better relocation of impurities away from ice crystals. In this work, desalination of artificial seawater using radial freezing at the side walls of cylindrical crystallizer is assessed experimentally and numerically. A 2-D axisymmetric computational fluid dynamics model of multispecies flow with solidification and melting model is utilized to simulate the experimental apparatus. Results showed a successful radial freezing where salt diffusion towards the center is observed. Removal efficiency and effective partition coefficient were investigated experimentally and numerically with a very good agreement. Nonetheless, higher efficiencies are achievable when optimum freezing temperature and stirring at ice front are present.

Keywords: freeze desalination, CFD, directional freezing, indirect freeze crystallization, removal efficiency.

1. INTRODUCTION

The use of freezing processes in desalination has gained significant attention recently [1-8]. The key interest in freeze desalination (FD) emerges from the much lower fusion enthalpy of freezing water to ice, i.e. 333 kJ/kg, compared to the enthalpy of evaporation of water, i.e. 2,500 kJ/kg, which is the case in the conventional thermally-based desalination technologies [9]. Fundamentally, when crystallization by cooling takes place in an aqueous solution, ice crystals made up of pure water expelling any impurities or salts thus concentrated in the remaining fluid. Indirect freeze crystallization (IFC) is the most common technique used in desalination application as it owes several advantages due to the indirect interaction between the brine and refrigerant.

Most importantly, IFC has the capability of one-dimensional crystal growth (or called directional freezing) which as a result lowers impurities and eases the separation process [2, 10]. For instant, Gao, et al. [11] have experimentally investigated directional downward freezing for industrial wastewater treatment where the removal efficiency of chemical oxygen demand and organic carbon ranged between 45-65% when no stirring is involved. Additionally, they have highlighted the importance of mixing where the removal efficiency raised to 95% in some cases. Gay, et al. [12] have also considered wastewater treatment using indirect freezing in radial movement. Similarly, removal efficiency of metals and nitrate has ranged between 98.24-99.97% with the aid of stirring. The group of Shafique, et al. [13, 14] and Mushtaq, et al. [15] has extensively studied the effect of slow directional freezing on the removal efficiency of salts, bacteria, acids and bases from solutions. Freezing direction was alternatively changed to horizontally, vertically, and radially. They've successfully proven the migration of impurities away from the freezing front with high removal efficiencies that reached up to 95% when radial freezing of bacterial solution was involved [15].

The authors of the current work have studied earlier the upward directional freezing of a rectangular crystallizer using computational fluid dynamics (CFD) model, where different parametric studies were performed [7, 8]. In this work however, radial freezing is implemented and investigated for seawater desalination. Evaluation of the process will be done using a cylindrical crystallizer that will be further simulated numerically using CFD. Metrics as removal efficiency and effective partition coefficients will be evaluated and validated in both experimental and numerical assessments.

2. METHODOLOGY

2.1 Experimental Setup

The directional freezing desalination process is assessed using a laboratory scale apparatus. Commercial table salt and deionized water are used to prepare the synthesized seawater at different salt concentrations. Freezing is performed using a circular tray seen in Fig 1, with an inner diameter of 20 cm and a height of 5 cm. The top and bottom sides of the container are sealed with double insulation after placing water sample, whereas side wall is left with no insulation to ensure the radial freezing from the sides. The experiment lasts for 24 hours in a freezer at temperature of around -20°C. Conductivity of different samples at different locations are measured and recorded using the *XL60 Fisher Scientific* conductivity meter.

Fig 1 Cylindrical apparatus serves as the crystallizer for directional freezing experiment

2.2 CFD Model Setup

The development of a multispecies solidification/melting CFD model of the crystallizer is performed. The freezing process is simulated using an axisymmetric 2D computational domain (see *Fig 2*) that is governed by transient incompressible system of Navier-Stocks and energy equations. The 2D laminar flow is considered to simplify the problem where the computational domain (10x5cm^2) of the cylindrical crystallizer filled with salt-water mixture at seawater salinity of 3.5% and subjected to indirect radial freezing from sides.

2.2.1 Navier-Stocks and energy equations

The continuity and momentum equations are defined for a single domain instead of multi-domains when solidification/melting model is involved. Where the solidified (ice) fraction of the fluid is tracked implicitly using the enthalpy-porosity technique [16]. Hence, liquid fraction (β) is linked with each cell in the domain to specify the fraction of the cell volume that is in the liquid form. Based on the local temperature (T) at each computational cell, the liquid fraction is therefore can be defined as:

$$\beta = \begin{cases} 0 & if\ T<T_{solidus} \\ 1 & if\ T>T_{liquidus} \\ \frac{T-T_{solidus}}{T_{liquidus}-T_{solidus}} & if\ T_{solidus}<T<T_{liquidus} \end{cases} \quad (1)$$

Where $T_{solidus}$ and $T_{liquidus}$ are the lower and upper boundary temperatures where the phase-change zone (also called the mushy zone) exists in a multicomponent mixture, as in the case of salty water.

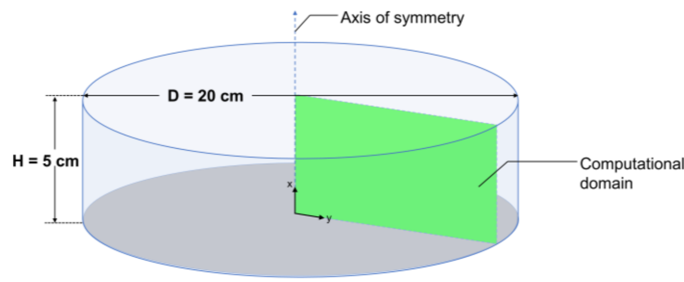

Fig 2 Geometry of the crystallizer and the axisymmetric modelling domain

Meanwhile, the redistribution of solutes (i.e. salt) from the solid phase (i.e. ice) into the liquid phase (i.e. concentrated brine) during the freezing process is quantified by the limiting partition coefficient of salt (K_0). It is defined as the mass fraction of salt in the solid phase ($Y_{s,sol}$) to the mass fraction of salt in the liquid phase ($Y_{s,liq}$) at the freezing front:

$$K_0 = \frac{Y_{s,sol}}{Y_{s,liq}} \quad (2)$$

For species segregation from the solid to the liquid phase, the non-linear Scheil rule is used in determining the relation between the liquid fraction and temperature.

The resulting continuity equation justified for solidification/melting Scheil rule species model is expressed as:

$$\frac{\partial}{\partial t}\left(\rho Y_{s,liq}\right) + \nabla\cdot\left(\beta \vec{v}_{liq} Y_{s,liq}\right) = \nabla\cdot\left(\rho\beta D_{s,m,liq}\nabla Y_{s,liq}\right) - K_0 Y_{s,liq}\frac{\partial}{\partial t}\left(\rho(1-\beta)\right) + \frac{\partial}{\partial t}\left(\rho(1-\beta)Y_{s,liq}\right) \quad (3)$$

Where ρ is density, $\vec{v}_{liq}$ is the liquid velocity, $D_{s,m,liq}$ is the mass diffusion coefficient of salt ,i.e. sodium chloride (NaCl), in water and it's found experimentally to be 2.2E-09 m^2s^{-1} [17].

In addition, the momentum equation is modified to comprise the additional pressure drop term caused by water solidification to ice. The resulting momentum equation can be written as:

$$\frac{\partial}{\partial t}(\rho\vec{v}) + \nabla\cdot(\rho\vec{v}\vec{v}) = -\nabla p + \mu\nabla^2\vec{v} + \rho\vec{g} + \frac{(1-\beta)^2}{(\beta^3+\varepsilon)}A_{mush}\vec{v} \quad (4)$$

Where p is the local static pressure, μ is the molecular viscosity of water, $\vec{g}$ is the gravitational acceleration, A_{mush} is the mushy zone constant (set to 1E+05), ε is a small value used to avoid division by zero at fully solidified regions (when β=0).

In addition to the Navier-Stocks system, the following energy equation is enabled:

$$\frac{\partial}{\partial t}(\rho H) + \nabla\cdot(\rho\vec{v}H) = \nabla\cdot\left(k\nabla T - h_s\vec{J}_s\right) \quad (5)$$

The two terms in the right-hand side represent the energy transfer due to conduction and species diffusion, where k is the thermal conductivity, h_s is the enthalpy of the salt specie, and $\vec{J}_s$ is the diffusion mass flux of salt. On the other hand, H is the overall sensible enthalpy. Hence, temperature is solved iteratively using the energy (5) and liquid fraction (1) equations.

2.2.2 Mesh discretization, model setup, and solution methods

The 2D geometry is structurally meshed using quadratic face meshing method. The top and bottom walls and the radial wall are refined at their boundaries with curvature size function using bias factor of 5. The final mesh shown in Fig 3 resulted in having 14,700 computational elements with perfect orthogonal quality of 1 and maximum face area of 1.339E-03 m^2.

ANSYS Fluent 17.2 is used to carry out the simulation and model setup. The left-hand side boundary of the domain is set to axis of rotation to simulate the axisymmetric. All other walls are stationary and subjected to no-slip shear condition and zero diffusive flux of salt specie. Thermally, top and bottom walls of the crystallizer are insulated by subjecting a zero-heat flux Neumann boundary condition. Whereas the radial side wall is subjected to Dirichlet boundary condition with a specified freezing temperature of -20°C to meet the experimental conditions.

The fluid zone is occupied with two species mixture, i.e. salt and water, where mixing law formula is enabled to calculate the equivalent thermo-physical properties of the mixture at each time step. The mixture is initially set with salt mass fraction of 0.035 resulting in overall mixture density of 1017 $kg.m^{-3}$, thermal conductivity of 0.83 $W.m^{-1}.K^{-1}$, and specific heat capacity of 4065.38 $J.kg^{-1}.K^{-1}$ at initial temperature of 20°C.

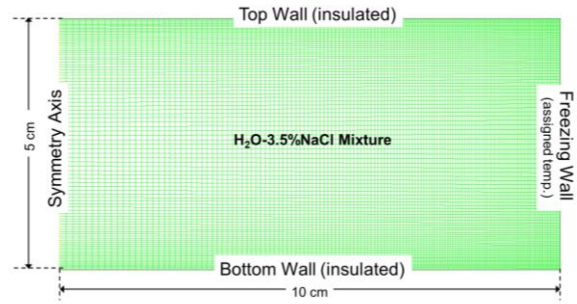

Fig 3 Discretized mesh and assigned boudary conditions

The governing equations are solved with spatial discretization (second order) using the velocity-pressure -based SIMPLE (Semi-Implicit Method for Pressure Linked Equations) algorithm. Accuracy of 1E-20 for continuity and 1E-06 for energy and species convergence is set to achieve reasonable accurate solution. Calculations are made with a fixed time step size of 0.01 sec and 20 iterations per time step.

2.3 Performance Evaluation

Several metrics can be used to evaluate the performance of the desalination by directional freezing process. Meanwhile, salt removal efficiency (R) and effective partition coefficient (K) are frequently spotted in literature as the most simple and functional parameters to evaluate a desalination process [2, 18]:

$$R = \left(1 - \frac{C_i}{C_0}\right) \times 100 \quad (6)$$

$$K = \frac{C_i}{C_L} \quad (7)$$

Where C refers to the salt concentration. The subscripts 0, i, and L refers to the initial solution, ice phase, and concentrated final liquid phase, respectively. Removal efficiency and effective partition coefficient are therefore inversely proportional.

3. RESULTS AND DISCUSSION

3.1 Experimental Results

In this work, two seawater solutions were prepared at salinity of 3.5% (Sol. A) and 7% (Sol. B), respectively. The solutions were further used in the experimental apparatus as explained earlier. After 24 hours of freezing, seven different samples each with rough diameter of 2 cm and height of 5 cm were extracted from the crystallizer as seen in Fig 4. As expected, the dissolved salt diffused towards the center while freezing resulting in highly concentrated brine at the middle and pure ice production at the boundaries of the crystallizer as seen in the figure. Samples were then left to reach ambient conditions, i.e.26°C, and tested for their conductivity. Conductivity results compared to the initial solution conductivity are displayed for both seawater salinities in Fig 5. Results of both experiments showed successfully the diffusion of salt towards the center due to the radial freezing and are in a very good agreement with the work of Mushtaq, et al. [15].

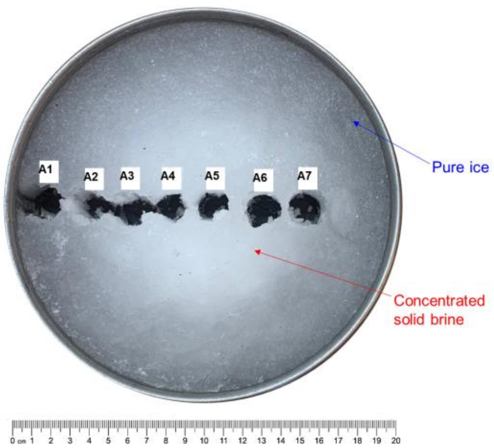

Fig 4 Radially frozen salt-water solution (Sol. A) and samples locations, total volume ~1570mL frozen at -20°C for 24 hours

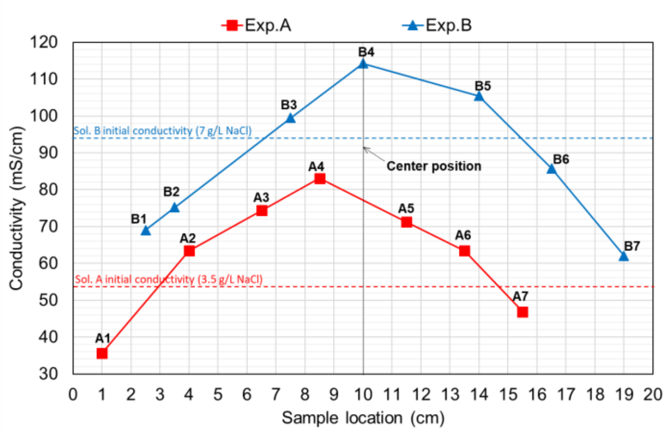

Fig 5 Conductivity results of samples of Sol. A and Sol. B taken at different locations

3.2 *Numerical Simulation Results*

The numerical CFD model was used to get more insight to the freezing process and its parameters by validating the experimental results of the 3.5% seawater solution (Sol. A). Fig 6 represents the distribution of temperature in the salt-water mixture at different flow time. The model was successful in simulating freezing initiated from side walls and directed towards the center of the crystallizer. Ice growth was also recorded in Fig 7 using liquid fraction distribution which clearly decreased directionally with time. Due to the time-intensive computational power required, simulations were stopped before full solidification of the salt-water mixture where the maximum physical time reached was 1.5 hours.

The directional diffusion of salt specie towards the center of the crystallizer is obviously seen in Fig 8. This indicates that the ice crystals formed at the side walls are relatively pure with salt content of 2.68%. In average, this makes the highest removal efficiency to be 23.39% and partition coefficient of 0.401. The relatively low removal efficiency is mainly due to the absence of stirring and mixing which as a result caused severe concentration of salt species at the crystal growth front as seen clearly in the figure. Those findings are in agreement with Gao, et al. [11] and Gay, et al. [12] who emphasized the importance of stirring for efficient separation. Additionally, the extremely low freezing temperature may also causes higher entrapment of salts due to the fast crystal growth as investigated in earlier work of authors [8].

Average removal efficiency (Eq.6) and effective partition coefficients (Eq. 7) were compared and validated with experimental data of solution A, i.e. 3.5% saline water, and tabulated with their relative error in **Error! Reference source not found.**. Operating at identical conditions, the CFD model results showed a very good agreement with the experimental results with almost negligible error. This indicates the validity of the simulation model to be used in further studies.

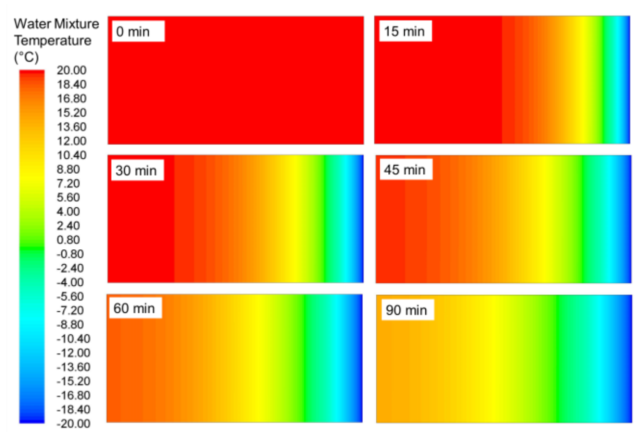

Fig 6 Temperature contours of water mixture in a crystallizer with radial directional freezing at a temperature of -20C°

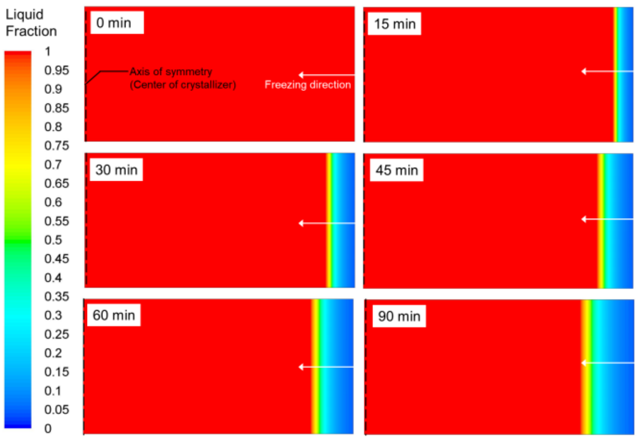

Fig 7 Contours of liquid fraction varying from 0 to 1 captured at the symmetrical section of the crystallizers showing the radial directional freezing at different flow time

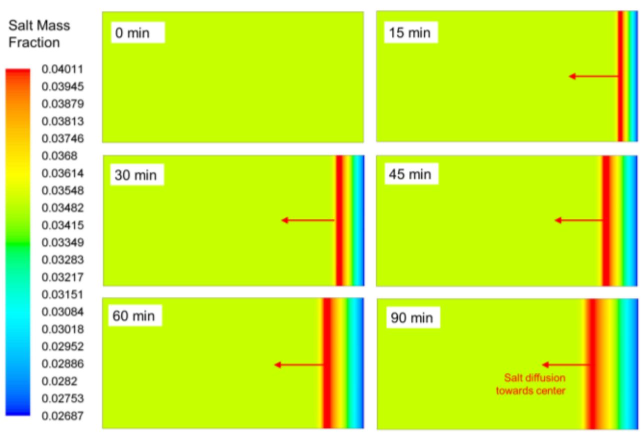

Fig 8 Contours of salt mass fraction showing the salt diffusion towards the center of the crystallizer running at freezing temperature of -20°C

Table 1 Parametric experimental and numerical results of directional freezing process with relative error percentages

	Experiment	CFD Model	Rel. Error (%)
Removal Efficiency, R (%)	23.677	23.390	-1.2093
Effective Partition Coefficient, K	0.398	0.401	0.7870

4. CONCLUSIONS

A 2-D axisymmetric steady-state computational fluid dynamics (CFD) model of indirect freeze desalination with radial freezing was implemented and validated experimentally. At exactly similar operational conditions, simulation and experimental results were in a very good agreement where the removal efficiency and the effective partition coefficient of the process were evaluated. Radial directional freezing was successfully performed; however, the process did not demonstrate superior desalination efficiency due to the absence of stirring and lack of operation optimization.

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