Desalination of seawater through progressive freeze concentration using a coil crystallizer

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ABSTRACT

In this century, the shortage of clean water supply is an issue of concern and the problem is expected to become more serious in the future. Consequently, researchers are trying to find the best solution to address this problem by introducing new desalination technologies that are able to accommodate the increasing demand for clean water. One of the new technologies introduced is desalination of seawater through freeze concentration. In this study, progressive freeze concentration (PFC) was implemented to produce pure water in the form of an ice crystal block, leaving behind higher concentration seawater using a coil crystallizer. The effect of operating parameters such as initial concentration and coolant temperature were investigated. Meanwhile, the efficiency of the system was reviewed based on the value of the effective partition constant $K$ which is defined by the ratio of solute in ice and liquid phase. A low value of $K$ indicates when the system is most efficient. In addition, the results for the overall heat transfer coefficient are also presented to observe the heat transfer involved in the system.

Key words | crystallization, desalination, freeze concentration, heat transfer, ice crystal, progressive freeze concentration

INTRODUCTION

Water shortage has become a major issue, particularly in developing countries. The existing water resources are unable to meet the water supply demand based on the latest trend of human population growth which is increasing enormously (Al-Subaie 2007). The tremendous growth of industrialization and urbanization also leads to a higher demand for water supply (Fujikawa et al. 2013). According to a report by Global Water Intelligence, desalination plants have a capacity of 78.4 million m$^3$/day all over the world, an increment of 64.7% compared to the capacity at the end of 2008.

Desalination technology can be developed to produce fresh water for industry, agriculture and also human consumption. The technology has now been practiced on a large scale for more than 50 years. For many years, thermal technologies were the only viable option, and multi-stage flash (MSF) desalination was established as the baseline technology. Multi-effect evaporation is now the modern thermal technology, but it has not been widely implemented. With the growth of membrane science, reverse osmosis has overtaken MSF as the leading desalination technology and should be considered as the baseline technology, while others are alternatives. The search for improved desalination methods has led to the use of the freeze concentration method (Shafiu Rahman & Ahmed 2007).

In freeze concentration, the solution is made concentrated by applying the progressive freeze concentration (PFC) process. The ice crystal is formed on the surface of the conducting material where cooling is supplied. Its separation from the solution is easier to handle and at a lower cost as only a single crystal is formed (Jusoh et al. 2009).

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According to previous reports, one of the main advantages of the freeze concentration desalination process is its energy consumption. In this method, only 420 kJ of energy are required to remove salt and produce 1 kg of fresh water (Attia 2010). This amount is six times lower than the energy required by MSF (Attia 2010). Additionally, as this method is operated at low temperatures where corrosion is not significant, this system has lower operational costs and requires less maintenance (Englezos 1994). However, one of the difficulties in dealing with this system is handling the ice after the process is completed (Rane & Padiya 2011).

Therefore, elements such as ice sampling and observation are important in designing the apparatus. In fact, the apparatus design is a critical factor in influencing the system efficiency. Previously, the coil crystallizer has been successfully used for glucose concentration (Jusoh et al. 2009). This study is intended to explore the possible application of this device for desalination. There are several factors affecting the efficiency of the system and the thawed ice quality (Luo et al. 2010) including initial concentration and coolant temperature. In addition, the overall heat transfer as affected by the parameters was also investigated because this factor has an impact on the ice crystal formed on the inner wall of the crystallizer which also represents the system efficiency (Habib & Farid 2006).

MATERIALS AND METHODS

Materials

A saline solution was used throughout the experimental work to represent seawater. In order to make saline water, salts were well-mixed with pure water for the desired concentrations. A 50% (v/v) ethylene glycol solution was used for the coolant in the water bath.

Equipment

The crystallizer used, as shown in Figure 1, is made of aluminum in a hollow helical structure. The three layers or crystallizer stages could be split into two and are attached by flanges at five points where the ice formation could be seen. In order to determine the temperature profile of the process, nine thermocouples were used by engaging them on each stage of the crystallizer to detect the temperature of the solution, aluminum wall and coolant. These temperatures were then displayed by PicoLog recorder software. A pump was used for solution movement. The waterbath acted as a cooling chamber for the process to maintain the process at the desired temperature. A salinometer was used to determine the salinity of the ice produced and saline concentrate.

Experimental procedure

Seawater was kept in the freezer between 2 and 3 °C as the initial temperature of the sample should be close to the freezing point of water. Cubes of seawater were mixed with the sample to maintain the temperature during the feeding process. Pure water was then pumped into the crystallizer to provide seed ice or form the ice lining. This step is necessary to avoid initial super cooling, which could cause serious contamination in the ice crystal initially formed (Liu et al. 1998; Luo et al. 2010).

The seawater was fed into the crystallizer, using a peristaltic pump, through a silicone tube until the solution had completely filled the crystallizer. Each end of the silicone tube was then connected carefully to avoid the presence of bubbles in the tube. In order to allow the crystallization process to occur, the filled crystallizer was then immersed into the waterbath at the desired temperature and circulation flowrate for about 10 minutes. After the designated time,
the circulation was stopped and the crystallizer was taken out to be thawed. The concentrated seawater in the silicone tube was drained out as a concentrate sample. The whole volume of the concentrated seawater left in the crystallizer was collected by disassembling the flanges. A sample of the ice layer produced was collected. The salinity of each sample was then measured.

**RESULTS AND DISCUSSION**

Exclusion of solute molecules from the moving ice front and the interface between the ice and solution phases is the main mechanism of concentration in PFC (Liu et al. 1997). The effective partition constant, $K$, was reviewed to determine the system performance. $K$ is related to the quality of the ice produced which can be determined by the following equation (Liu et al. 1997):

$$K = \frac{C_S}{C_L}$$

(1)

where $C_S$ is the concentration of the ice and $C_L$ is the concentration of the remaining solution (concentrate). In this case, the concentration is represented by salinity. Based on the equation of solute mass balance (Liu et al. 1997), the equation of effective partition constant $K$ can be integrated as follows (Liu et al. 1997):

$$(1 - K) \log \frac{V_L}{V_o} = \log \frac{C_o}{C_L}$$

(2)

From Equation (2), $V_L$ is defined as the concentrate volume, $V_o$ is the initial volume of solution, $C_o$ is the initial concentration of the solution and $C_L$ is the concentration of the concentrate. Equation (2) serves as the evaluation method in this research. In any condition, a lower $K$ value indicates greater efficiency of the system. According to Fujioka et al. (2013), the effect of desalination increases when the $K$ value is smaller. On the other hand the purity of ice produced can be determined based on its salinity: the lower the salinity the higher the purity of ice produced. Thus, a low $K$-value and low salinity of ice are considered as the favorable results for the system. At the end of the experiment, an ice layer was formed on the wall of the crystallizer. The thickness of ice is closely related to the operating conditions. The thickness of ice varied all the way through the experimental works as the operating conditions varied.

**Effect of operating parameters**

One of the most significant parameters in PFC is the initial concentration, which represents the total solute in the sample solution. The studied range of initial concentrations for the newly designed PFC system was 20–60 g/L which is the average range of salinity of sea water around the world. While the initial concentration was varied, the other operating conditions were kept constant.

From the graph in Figure 2, it can be observed that at the lowest initial concentration, the value of $K$ is also the lowest as compared to higher concentrations. This is in line with theories from previous studies for the following initial concentration where the $K$ value increases as the concentration increased. This means that the $K$ value depends on the initial amount of solute in the solution to be concentrated. At a lower initial concentration, only a small amount of salt is contained in the solution, resulting in lower accumulation of solutes at the ice-liquid interface in order to produce more concentrate (Miyawaki et al. 2005).

Solutions with higher initial concentrations have higher amounts of solutes, which will cause greater accumulation of solutes at the ice-liquid interface. As a result, more solute is trapped into the ice. Consequently the ice layer concentration is higher and results in a higher value of $K$. The graph in Figure 2 also shows the effect of initial concentration on salinity reduction. The graph shows that when the lower initial concentration was introduced, the highest salinity reduction was observed which signifies the highest purity of ice formed. This is because the low-concentration solution contains a small amount of solute resulting in less contaminant being trapped in the ice compared to the high concentration where there is more chance for contaminants to be entrained in the ice (Liu et al. 1998).

The studied range of coolant temperature for the experiment was $-10$ to $-16$ °C, which was chosen based on the freezing points of pure water and saline water. It was found that the freezing point of saline water decreased as
the salt concentration increased. As the freezing point of water is 0 °C, pure water will obviously become ice at the studied temperatures, leaving behind saline water which has a lower freezing point. While the coolant temperature was varied, the other operating conditions were kept constant. After examining the samples and determining their salinity, the effect of coolant temperature on $K$ is illustrated in Figure 3.

From Figure 3, a coolant temperature of $-12$ °C is considered as the best condition due to the lowest value of $K$ occurring at this temperature. The $K$ value decreases between $-10$ and $-12$ °C showing that the lower coolant temperature resulted in a lower value of $K$, which means higher efficiency for the system (Miyawaki et al. 2005). However, as the coolant temperature was decreased to $-14$ and $-16$ °C, the $K$-value started to increase. This means that the efficiency of the
system would decrease if the coolant temperature is too low. At $-14 \, ^\circ C$, there is a possibility that the saline water would also freeze and the salt would get trapped in the pure ice formed, resulting in an increase in the salinity of the ice. Thus, the $K$-value would also increase, resulting in reduced efficiency of the system.

In addition, the coolant temperature will influence the ice growth rate, whereby a low growth rate will result in high purity of ice produced. According to Flesland (1995), when the difference between the solution and the surface temperature increases, the ice growth rate will increase as well. In the same way, when the coolant temperature is decreased, a higher growth rate of the ice front will be observed (Flesland 1995). This situation is undesirable as it will produce a low $K$ for the system. The higher the ice growth rate, the more impurities that would be entrained in the ice. In addition, the solute outward movement will be blocked by the high speed of the ice moving front, resulting in promotion of solute inclusion in the ice crystals (Chen et al. 1998).

Therefore, the salinity of ice would increase as the coolant temperature is reduced to $-14 \, ^\circ C$ because of the high amount of ice impurities present at the low temperature applied, resulting in a faster freezing rate; hence, the impurities are easily trapped into the ice phase. This again shows that the coolant temperature of $-12 \, ^\circ C$ is the optimum condition for producing thawed ice with the lowest value of salinity, under the present study.

**Heat transfer analysis**

Heat transfer deals with energy transportation due to temperature difference. It occurs when there is an existence of temperature difference either in a medium or between media. In essence, a temperature difference driving force is the main factor that influences the occurrence of heat transfer. Basically, heat will flow from the high- to the low-temperature region (Geankoplis 2003). In this case, it is between the saline solution and coolant temperature. Figure 4 shows a snapshot of the temperature profiling obtained from the PicoLog recorder. From the snapshot, it can be seen that the temperature of the solution outlet ($T_{so}$) decreases as the time increases. This shows that the heat is successfully removed from the solution, proving that the crystallization process has occurred. This graph is used to determine the temperature difference when the heat transfer occurred. A slow rate of heat transfer, resulting from small driving forces, will also tend to reduce the energy consumption during the ice making (Shone 1987).

In this study, the overall heat transfer coefficient $U_o$ was calculated for every operating parameter that was manipulated based on the data displayed by the PicoLog recorder. This overall heat transfer coefficient $U_o$ was calculated to determine the ability of heat transfer in the system. It can be measured by the equation of resistances involved as stated in Equation (3) (Geankoplis 2003):

$$ R = \frac{1}{A_i h_i} + \frac{x}{k_i A_m} + \frac{1}{A_o h_o} = \frac{1}{U_o A_m} $$

(3)

where $U_o$ (W/m$^2$ K) represents the overall heat transfer coefficient, $A_i$ is the inside surface area of the tube (m$^2$), $A_o$ is the outside surface area of the tube (m$^2$), $h_i$ is the heat transfer coefficient for saline water (W/m$^2$ K), $h_o$ is the heat transfer coefficient for ethylene glycol 50% (W/m$^2$ K), $x$ is the thickness of the ice layer (m), $k_i$ is the thermal conductivity of ice (W/m) and $A_m$ is logarithmic mean area. In this case, the $A_m$ value can be calculated by considering the area of the ice layer surface, hence producing Equation (4) below

$$ A_m = 2\pi L \left( \frac{x}{\ln (r/(r-x))} \right) $$

(4)

where $r$ and $L$ are the radius of the copper tube and the total length of the crystallizer, respectively.
The graph in Figure 5 shows the trend for initial concentration and coolant temperature against the overall heat transfer coefficient. This trend could be explained by referring to ice thickness. In this case, the ice thickness will be a chief factor affecting the heat transfer process as it provides resistance to heat transfer occurring.

It can be observed that as the initial concentration increased, the value of $U_o$ increased. At low concentration, the ratio of water volume and solute is high which means that the volume of water is significantly bigger compared to the solute. Therefore, the potential for production of pure ice is higher, which would then make the ice thicker. In contrast, at the highest concentration, the volume of pure water is less as compared to a lower concentration solution. Therefore, less ice was produced. Small ice thickness has less resistance, so more heat transfer can occur, resulting in a high value of $U_o$.

Based on the experimental results, it is observed that $U_o$ is the highest at a coolant temperature of $-10^\circ$C. This shows that the heat transfer from the solution to the coolant through the wall of the crystallizer on the acquired surface had occurred at its maximum in this condition. The small temperature difference between the solution and coolant could cause the small thickness of the ice formed as observed at a coolant temperature of $-10^\circ$C. Thus the resistance for heat transfer was low at this point and therefore the heat from the solution could be transferred effectively to the coolant. As the coolant temperature decreased, the temperature difference between the solution and coolant was higher, and caused almost all the solution to solidify, leaving no passage for the solution to be circulated.

**CONCLUSION**

This study has successfully proven that the PFC system with coil crystallizer has a large potential to be used for the desalination process. The system has achieved its best performance at low initial salt concentrations and intermediate coolant temperature. Ice thickness seems to be the chief factor that affects the heat transfer process.
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