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# Performance Investigation on a New Design for Progressive Freeze Concentration System

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**Abstract:** Freeze concentration freezes or crystallizes water component in a solution out as ice crystals and leaves behind a highly concentrated solution. Conventional Suspension Freeze Concentration (SFC) system has to deal with difficult separation of ice formed in a suspension of the mother liquor, which leads to a high cost of operation. Scraped surface heat exchanger used to form seed ice to ensure production of large ice crystals also hiked up the capital cost involved. Progressive Freeze Concentration (PFC) is believed to be able to reduce the capital and operation cost by eliminating some unit operations used. In PFC, ice crystals were formed progressively as a single block of ice, on the surface where cooling is supplied. In this particular study, a new helical structured copper Crystallization Chamber (CC) was designed and fabricated. Three operating parameters which affect the performance of the system were investigated namely the circulation period, circulation flow rate and coolant temperature. Effective partition constant, K was used as an indication of the system efficiency and to assess its performance, calculated from the volume and concentration of the solid and liquid phase. Circulation period was found to produce a constant value of K at each period tested with the CC. It was also discovered that higher flowrate resulted in a lower K, which translated into high efficiency. The efficiency is the highest at 1000 mL $min^{-1}$. The process also gives the highest efficiency at a coolant temperature of -6°C.

**Key words:** Freeze concentration, progressive freeze concentration, ice crystallization, effective partition constant

## INTRODUCTION

Freeze Concentration (FC) is a process where the water component in a solution is frozen and crystallized as ice so that a more concentrated solution will be left behind. The process could be applied to liquid food and wastewater. In application to liquid food, FC is used for concentration of heat sensitive material or solution that would deform when extensive heat is applied. It is also preferred when concentrating liquid food with aromatic quality that should be detained even after a concentration process. As the process do not involved heating, the aromatic compound is more likely to dwell in the concentrated solution.

In application to wastewater, it could be categorist as a type of treatment as it would produce almost pure water from the thawed ice crystals. The concentrated solution or wastewater left leads to reduction of treatment cost as the wastewater volume is also reduced extensively. The FC has been recorded to be able to concentrate approximately 80% of the dissolved compounds in 25% of the original volume (Maurer *et al*., 2006). The water/ice crystals produced is supposed to be highly pure because the small dimensions of the ice crystal lattice makes the inclusion of any foreign compounds impossible except for fluorohydric acid and ammonia (Lorain *et al*., 2000), thus resulting in a highly effective separation of water components from the solution.

In this study, FC is focused to be applied for the purpose of wastewater treatment, therefore, the emphasis is on producing as pure water achievable and to reduce the volume as far as possible. There are two methods available for freeze concentration, conventional Suspension Freeze Concentration (SFC) and Progressive Freeze Concentration (PFC). The SFC is a process of freeze concentration where the ice crystals are formed in a suspension of the mother liquor and is characterized by the generation of a size distribution of crystal growing isothermally. However, in this conventional method, the

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size of ice crystal is still limited (Gu *et al*., 2005). The small ice crystals formed has to be transferred to a ripening vessel to be enlarged, then to a washing column and separated from the mother solution after washing with water (Widehem and Cochet, 2003). These steps: ice nucleation, ice crystal growth and ice crystal separation make the whole system very expensive, which has made it unfavourable.

In compensating the disadvantages of SFC, a totally different concept of crystallization, PFC has been introduced. In this method, a large single ice crystal instead of a group of small ice crystals suspension is formed. The ice crystal is formed on the surface of the heat conducting material where the cooling is supplied. As only a single crystal is formed, its separation from the mother liquor is much easier to be handled and at a lower cost. However, despite of the easier separation, the productivity of PFC is found to be lower than the conventional SFC.

The design of the apparatus where the crystallization of ice is supposed to occur is an important factor in influencing the system efficiency. The selection of material of constructions and the design shape of the apparatus should be carried out carefully in order to ensure successful operation of freeze concentration. In this particular research a helical copper crystallization chamber was fabricated, where the crystallization of ice should take place. The newly fabricated chamber was then evaluated in terms of its efficiency according to three parameters, which are the circulation period, initial concentration of the initial solution and the circulation flowrate during operation.

## MATERIALS AND METHODS

**Materials:** Glucose solutions at a concentration of 7 mg $mL^{-1}$ were used to represent the simulated wastewater. It is very common that glucose be used in assessing the performance of a wastewater treatment system. Glucose used was 99.9% pure.

**Equipment:** Figure 1 shows the Crystallization Chamber (CC) fabricated using copper as the material. The thickness of the copper tube is 0.8 mm with internal diameter of 1 inch. The chamber has three layers or stages and is also equipped with 6 stainless steel flanges where the chamber could be split into two. This is to enable visualization of the ice layer produced in each experiment. Nine temperature probes (thermocouples type K) were engaged in each stage for temperature profiling purpose, where the solution, copper wall and coolant temperatures are displayed by PicoLog recorder software through a connected computer. This crystallization chamber was then immersed in a refrigerated waterbath at the desired temperatures. The coolant used was ethylene glycol at 50% volume with water.

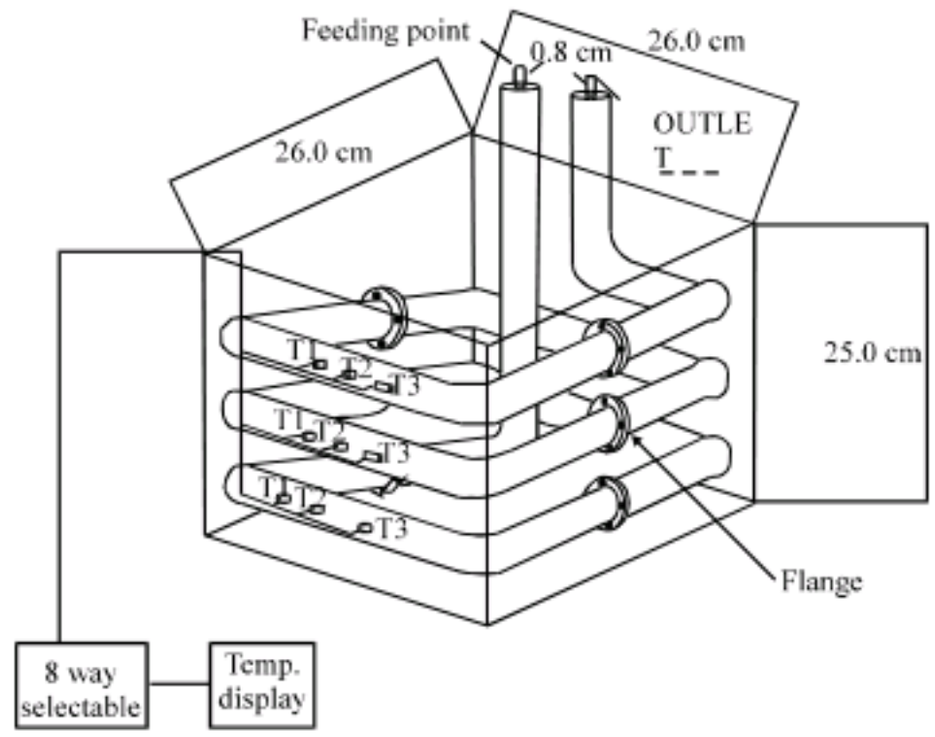


Fig. 1: Diagram of the helical copper Crystallization Chamber (CC) structure

**Experimental procedure:** Glucose solution prepared was first kept in a freezer where the temperature of the solution should be near the freezing temperature of water. The temperature was kept at 3 to 4°C and the solution was mixed with glucose solution ice cubes to maintain the temperature during feeding. The solution was then fed to the chamber using a peristaltic pump through a silicone tube until its full volume was filled. Each end of the silicone tube was then connected.

The filled CC was then immersed in a precooled waterbath at the desired operation temperature, while the pump was run at the desired circulation flowrate. The solution then was left for crystallization to occur for 15 min. After the designated time, the circulation was stopped and the chamber was taken out of the waterbath to be thawed. The concentrated solution in the silicone tube was then collected as the concentrate sample via flushing with the pump. The flanges were unassembled and the whole volume of the concentrated solution was collected. The ice layer thickness at each flange point was measured and a sample of the ice layer produced was collected. Refractive index of each sample was then measured in order to determine its concentration.

## RESULTS AND DISCUSSION

A calibration curve for the concentration of glucose via Refractive Index (RI) was first constructed by making several standard solution of glucose with concentration

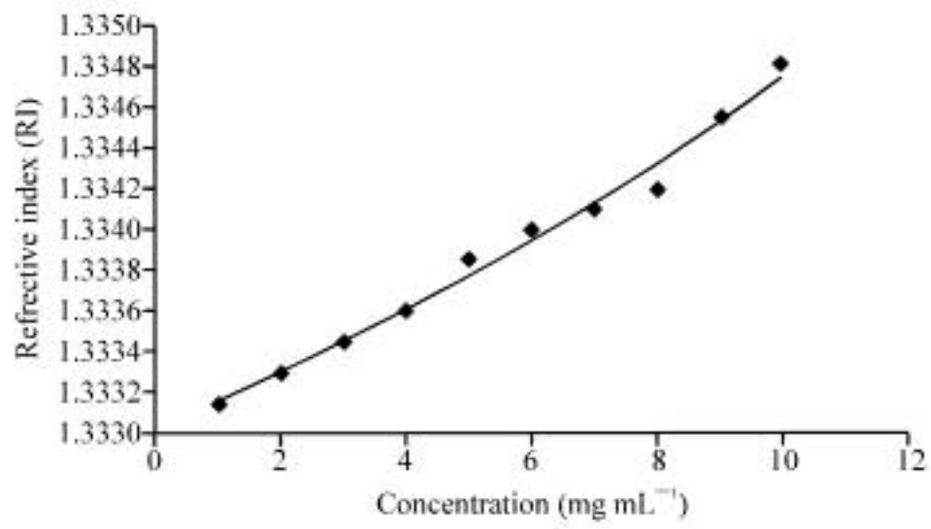


Fig. 2: Calibration curve

Fig. 3: Ice layer formed

Fig. 4: A close-up of the ice layer formed

in the range of 1 to 10 mg $mL^{-1}$. The calibration curve is shown in Fig. 2 which agrees with previous calibration curves produced previously by other researchers (Vaz *et al*., 2000).

During freezing, ice crystals were formed on the inner surface of the copper tube wall. Figure 3 and 4 show the ice layer formed in the CC at the end of the experiments. The thickness of the layer varied with the operating conditions varied throughout the experimental works.

the efficiency of the system is portrayed by the effective partition constant of the system which can be calculated through Eq. 1.

$$K = C_s/C_L \quad (1)$$

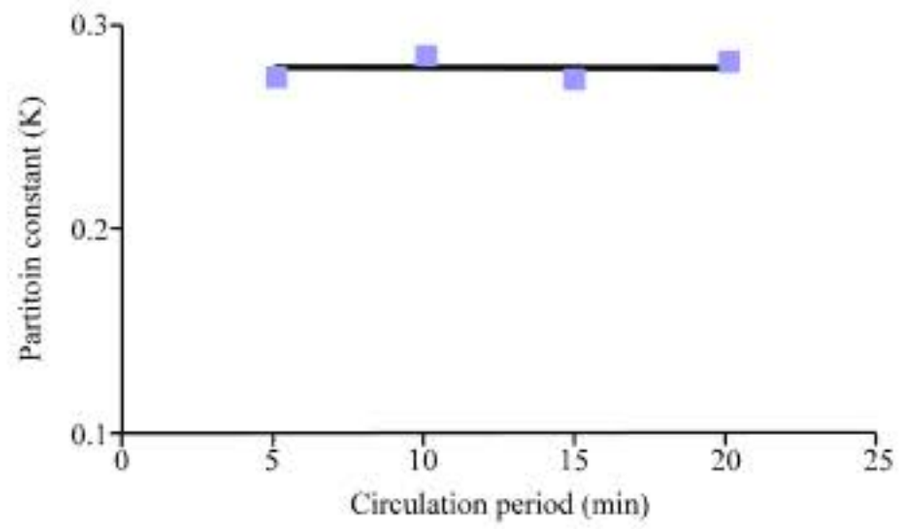


Fig. 5: Graph of K against circulation period

where, $C_s$ is and $C_L$ are solute concentrations in ice and solution phase, respectively (Miyawaki *et al*., 2005).

The experimental value of K is measured by Eq. 2, where, $V_0$ and $C_0$ are the volume and solute concentration at the beginning in the solution phase, respectively. $V_L$ is the volume of concentrate produced:

$$(1-K) \log (V_L/V_0) = \log (C_0/C_L) \quad (2)$$

**Effect of circulation period:** Some screening experiments have revealed that the range of circulation period in the capacity of this CC is 5 to 20 min. Experiments have been carried out in the variation of these circulation periods and the outcome is plotted in a graph shown in Fig. 5. The graph shows that the partition constant K is evidently around 0.28 regardless of the period of the circulation. This agrees with the core concept of the partition constant, which is calculated from $C_s/C_L$ to generate Eq. 2.

A previous research by Miyawaki *et al*. (2005) stated that the method of calculating K is by plotting a graph of $\ln(V_L/V_0)$ against $\ln(C_0/C_L)$, as shown in Fig. 6. K is calculated from the gradient of the lines, which is also a constant for each variation of the operating parameter. However, by revealing that the value of K stays at one value throughout the 20 min of operation, it is assumed safe just to obtain one value of K, i.e., at the end of each experiments, calculated from the concentration of solute in the liquid and solid state. Therefore, K for the experiments to evaluate the effects of flowrate and initial concentration will be calculated according to the findings in the investigation to see the effect of the circulation period.

**Circulation period (min):** Effect of circulation flowrate. The studied range of circulation flowrate for the newly designed PFC system was 400 to 1000 mL $min^{-1}$, which was chosen based on the existing pump capacity. While the circulation flowrate was varied, the other operating

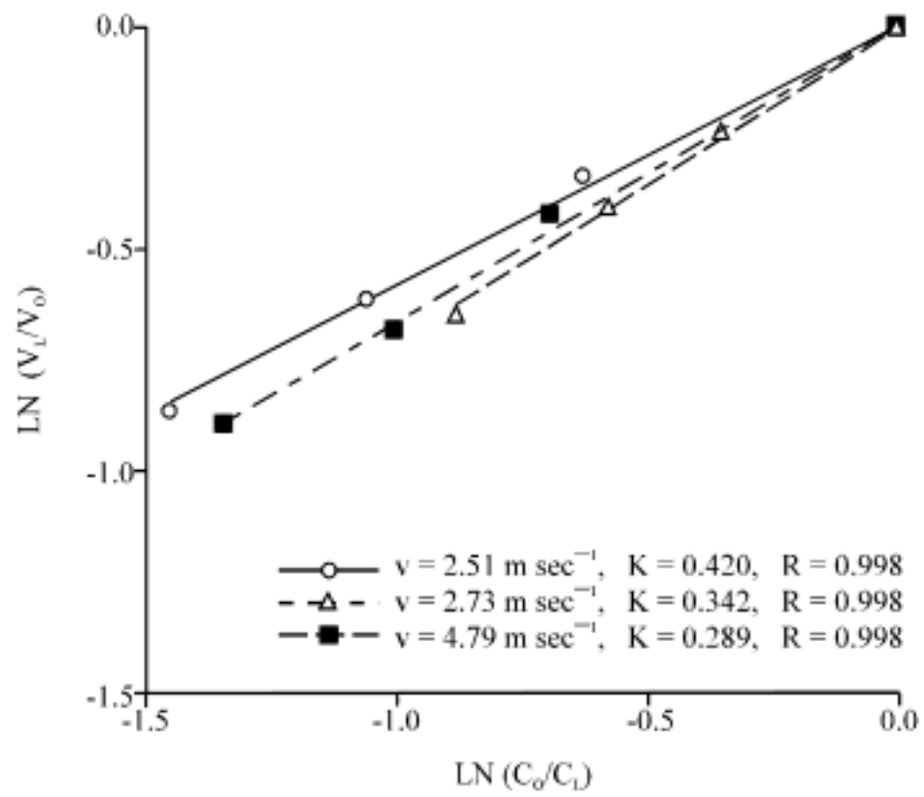


Fig. 6: A graph plotted in Miyawaki *et al.* (2005) to calculate K for various flowrate

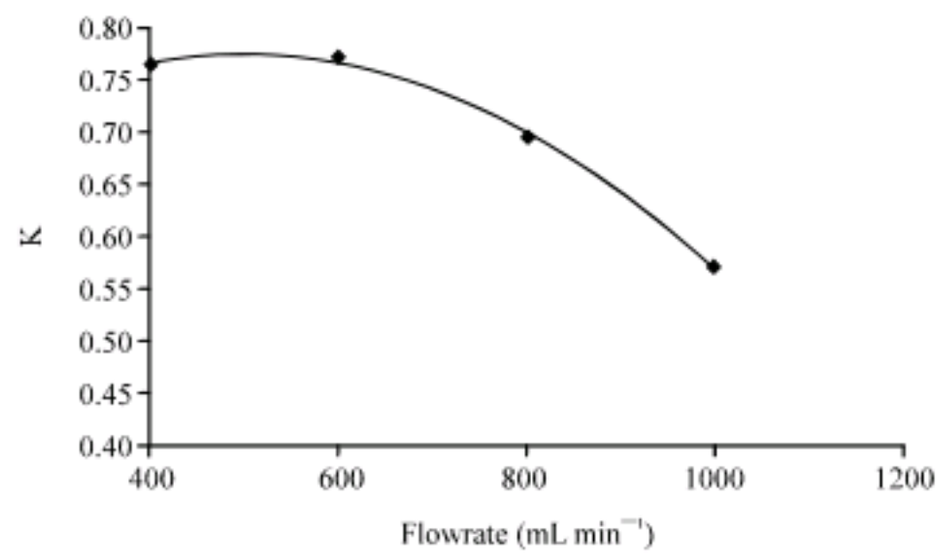


Fig. 7: Effect of circulation flowrate on K

conditions were kept constant. Coolant temperature was kept at -8°C and the circulation period at 15 min. The effect of circulation flowrate on K is shown in Fig. 7.

From the plotted graph, it can be seen that higher flowrate resulted in a lower K, which means better efficiency. This finding agrees with what was discussed earlier by Miyawaki *et al.* (2005) and Ramos *et al.* (2005), higher flowrate will result in a highly pure ice crystal layer.

Increasing the flowrate of solution promotes heat transfer with ice crystals from its tips, hence enhancing the planar ice growth from the cooling wall by keeping contaminants away from the ice-liquid interface (Wakisaka *et al*., 2001). Therefore, higher flowrate will result in ice layer with higher purity.

For this system, the K value is predicted to be lower if the flowrate is further increased. Figure 8 shows the effect of circulation flowrate on ice purity from this study which is very similar to the previous findings.

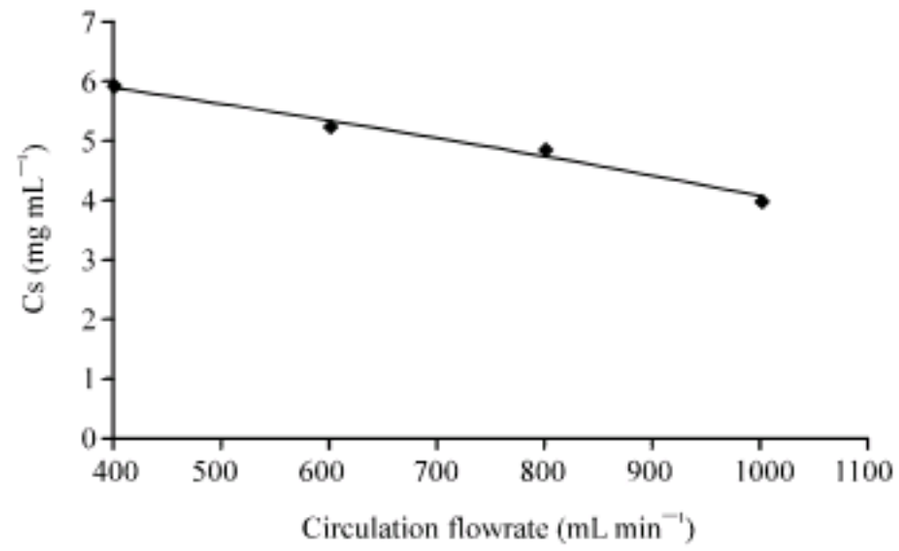


Fig. 8: Effect of circulation flowrate on ice purity

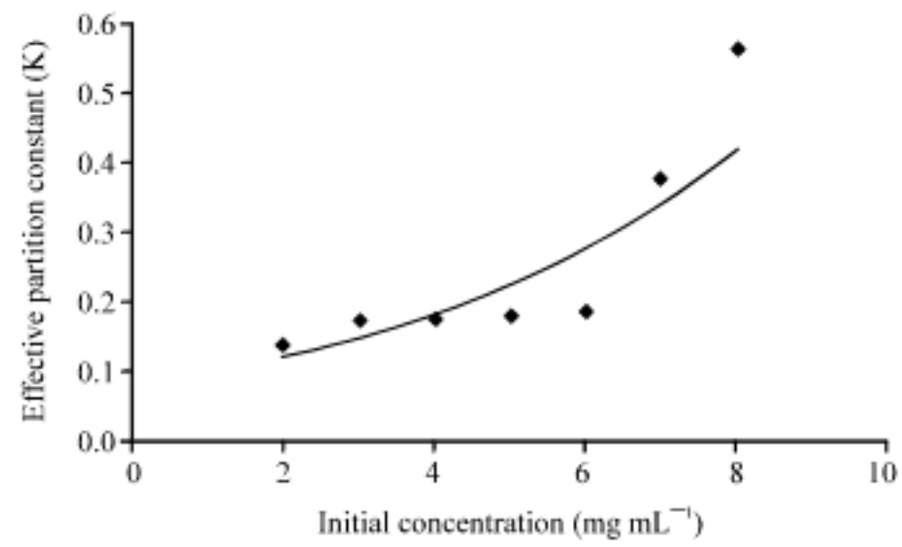


Fig. 9: Effect of initial concentration on K

**Effect of initial concentration:** The same experimental procedure was used in order to investigate the effect of initial concentration on the efficiency of this system. Solute concentration was investigated for the range of 2-8 mg mL$^{-1}$. Other parameter kept constant was the circulation flowrate at 1000 mL min$^{-1}$ and circulated for 15 min for crystallization.

After examining the samples and determination of its concentration, the effect of initial concentration on K is depicted in Fig. 9. It can be observed that higher initial concentration resulted in higher K, which means lower efficiency for the system and vice versa. This also means that the efficiency can be affected by the initial amount of solute in the solution to be concentrated and K is dependent on it. In the solidification process, the solution concentration increases at the ice-liquid interface because the solutes accumulate at this region (Miyawaki *et al*., 2005). This causes constitutional super-cooling, which strongly affects the dendritic structure at the interface. Higher initial concentration means higher amount of solutes in the initial solution, which will cause higher accumulation of solutes at this interface, causing the ice layer concentration to be higher. This causes an increase in the effective partition constant, K. Therefore, it can be concluded that the initial

concentration affects the efficiency of the process through constitutional supercooling, which causes a change in K (Miyawaki *et al*., 1992).

## CONCLUSION

This study has proven that the newly designed crystallization chamber is capable of producing ice crystals with good purity. The efficiency assessed by the value of K for the freeze concentration system shows high competency with existing designs by active researchers. However, the circulation flowrate should be further investigated by applying higher values to see if better separation is possible.

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