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## Effect of process parameters on progressive freeze concentration of sucrose solutions

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### Abstract

The progressive freeze concentration of sucrose solutions was tested. The effect of the initial concentration of the solution ( $C_0$ ), the temperature of the refrigerant (T) and the stirring speed ( $\omega$ ) on the final concentration of the solution was determined. The effects were significant on the freeze concentration, for both individual and combined effects. The maximum concentration achieved in the progressive freeze concentration was 53° Brix, when the initial concentration was 35° Brix, at a speed of 800 rpm and a temperature of refrigerant of -20°C. The best values of the concentration index are obtained at low concentrations, high stirring speed and low temperature. The average distribution coefficient increased with the initial concentration of the solution. The average yield parameter at different initial concentrations is around 0.6 kg ice·kg sol<sup>-1</sup>·h<sup>-1</sup>.

**KEYWORDS:** Freeze Concentration, sucrose, concentration, agitation, temperature, crystallization

## INTRODUCTION

The freeze concentration process has been widely studied. This technique involves the removal of water as ice crystals by cooling the fluid to be concentrated at temperatures below the freezing point (Auleda et al., 2011, Gulfo et al., 2014). The concentrations achieved for food juices are between 45 and 55 °Brix. The main objective of this operation is to preserve the properties of the fluid due to the low processing temperatures (Benedetti et al., 2015). The progressive freeze concentration (Halde, 1980; Liu et al., 1997, 1999) consists in the partial freezing of the fluid with a constant agitation. The agitation is performed to decrease the solute occlusion in the ice layer (Sánchez et al., 2011). An ice layer is produced in the tank walls. When a desired concentration is achieved, the operation is finished and the concentrated fluid is separated.

The operational variables can influence the freeze concentration process were studied in the present work. Sucrose solutions were freeze-concentrated by the progressive technique to determine the best operational conditions. Several authors (Liu et al. 1997, Chen et al. 1999, 2000, Jusoh et al., 2013, Miyawaki et al., 2001, 2005, Moreno et al., 2014a) have studied different fluids and proved the potential of the technique. However in our knowledge, the three main operational variables have not been studied simultaneously yet. These variables are: the initial concentration of the solution ( $C_0$ ), the refrigerant temperature ( $T$ ) and the stirring speed ( $\omega$ ).

The first variable, the initial concentration of the solution, affects the freezing point. It is known that solutions of low molecular weight solutes produce a change in the freezing

point; the higher the concentration, the lower the freezing point. This freezing point depression continues until the eutectic point, which is specific for each product. In this eutectic point, the solid and liquid fractions have the same concentration. The great importance of the initial concentration has been stated in several studies for other freeze concentration techniques (Moreno et al., 2014a; Chen and Chen, 2000). Secondly, the refrigerant temperature can affect the cost of the operation of freeze concentration with indirect contact and internal cooling. In addition, the temperature of the refrigerant affects the freezing rate and, consequently, the occlusion of solutes (Caretta et al., 2006; Moreno et al., 2014a; Petzold and Aguilera, 2009). Finally, the stirring speed influences the efficiency of the separation during progressive freeze concentration. The agitation produces a solute removing from the ice front and this particle motion can increase the efficiency of the separation (Miyawaki et al., 2005).

The aim of the present work was to determine the effect of the initial concentration of sucrose, the refrigerant temperature and the stirring speed on the final concentration of the solution, the concentration index and the average distribution coefficient. In addition the concentration limits for the progressive freeze concentration were determined.

## **MATERIALS AND METHODS**

Sucrose solutions were prepared with distilled water and commercial sucrose (Azucarera Española) to obtain 1200 mL of solution for each test. The solutions were stored at 1°C for 24 hours before the tests were performed. The freeze concentration tests were developed in the set-up showed in Figure 1. The device consisted in an internal and indirect cooling

system. The jacketed vessel (6) ID 115 x 230 mm (Trallero and Schlee, Barcelona, Spain) had a volume of 2400 ml. The vessel is isolated with polystyrene foam to avoid heat loss. The vessel was cooled with an ethylene glycol – water mixture (50% w/w) as the refrigerant fluid (9). The refrigerant was provided by a thermostatic bath (1) (Polyscience 9505, USA) allowed a temperature interval from  $-30^{\circ}\text{C}$  to  $150^{\circ}\text{C} \pm 0.5^{\circ}\text{C}$ . provided with a temperature control system (2). The fluid to be concentrated (8) was stirred by an agitator (11) RGL-100 (Heidolph Instruments, Germany) provided with a speed regulation system (7) PCE-DT62 (PCE Deutschland GmbH, Germany) with a precision of 0.05% and a resolution of 0.1 rpm.

In the freeze concentration tests, the concentration of the solution was measured each 10 minutes for 90 minutes according to the results of previously works (Raventos et al., 2007). The soluble solids content was measured using an DBX-55A (ATAGO, Japan) refractometer with a precision of  $\pm 0.1^{\circ}\text{Brix}$  and with a measurement range of  $0\text{--}55^{\circ}\text{Brix}$ .

The temperature of the solution was registered with a digital datalogger Testo 925 (TESTO, Germany) provided with a thermocouple type K with accuracy  $0.1^{\circ}\text{C}$ . The ice layer was produced in the walls and the bottom of the recipient. At the end of each test, the concentrated solution is poured into a vessel to separate it from the ice. As the solution contains a small amount of ice, it is filtered with a mesh of 0.39 mm in diameter. Ice retained in a mesh is added to the formed ice on the walls of the jacketed vessel. The mass of the solution and the ice was measured with a precision scale KB 1200-2N (KERN, Germany).

### ***Experimental Design***

The experiment was performed using a complete factorial design with three factors: the initial concentration of sucrose solutions at three levels (15, 25 and 35 °Brix), the refrigerant temperature at three levels (-10°, -15° and -20°C) and the stirring speed at four levels (0, 500, 800 and 2100 rpm). The tests were developed in triplicate.

### ***Statistical Analysis***

A generalized linear model (GLM) was performed to determine the influence of the three studied factors, namely the initial concentration of the solution ( $C_0$ ), the refrigerant temperature ( $T$ ), and the stirring speed ( $\omega$ ), on the final concentration of the solution ( $C_f$ ). Tukey's method was applied to compare all possible pairs of means. Simple linear regression was applied to study the final concentration kinetics. Additionally, the behavior of the following parameters was studied in a descriptive way: the concentration index (CI), the average distribution coefficient ( $\bar{K}$ ), and yield parameter ( $W_Y$ ). The analysis was developed using the software Minitab 16 for Windows (Minitab Inc. State College, PA, USA). A statistical significance level of  $\alpha = 0.05$  was used in all tests.

### ***Data Analysis***

#### **Concentration Index (Ci)**

CI was defined as the relationship between the solute concentration in the liquid fraction ( $C_f$ ) and the solute concentration in the initial solution ( $C_0$ ) (Moreno et al., 2015), as shown in Eq. 1

$$CI = \frac{C_f}{C_0} \quad (1)$$

### Average Distribution Coefficient $\bar{K}$

The average distribution coefficient is a measure of the amount of solute that is occluded in the ice (Flesland 1995; Chen and Chen 2000; Moreno et al. 2014a). It is defined as the ratio of the solute concentration in the ice ( $C_{ice}$ ) and solute concentration in the freeze concentrated liquid ( $C_f$ ) as shown in Eq. 2.

$$\bar{K} = \frac{C_{ice}}{C_f} \quad (2)$$

### Yield Parameter ( $W_y$ )

Another important parameter to indicate the success of the process is the yield parameter ( $W_y$ ) or dewatering capacity (Ramos et al., 2005; Moreno et al., 2015) defined by Eq. 3. The physical size and operating cost of any freeze concentration plant are largely determined by its ice dewatering capacity (Ramos et al. 2005).

$$W_y = \frac{W}{t} = \frac{C_f - C_0}{C_f - C_{ice}} = \frac{C_f - C_0}{(C_f - C_{ice}) \cdot t} \quad (3)$$

Where  $W_y$  is the yield parameter ( $\text{kg ice} \cdot \text{kg sol}^{-1} \cdot \text{h}^{-1}$ ),  $W$  is the amount of water removed ( $\text{kg ice} \cdot \text{kg initial sol}^{-1}$ ),  $C_0$ ,  $C_f$  and  $C_{ice}$  are the initial solute concentration, the final solute concentration and the ice solute concentration, both expressed in °Brix, and  $t$  is the total process time (h).

## RESULTS AND DISCUSSION

### Effect Of Freeze Concentration On Final Solute Concentration

The final concentrations ( $C_f$ ) obtained in the freeze concentration tests at the different of initial concentrations ( $C_0$ ), refrigerant temperatures ( $T$ ) and stirring speeds ( $\omega$ ) are shown in Table I.

The highest concentrations were obtained for the highest stirring speeds (800 and 2100 rpm), the lowest refrigerant temperatures (-15 and -20°C) and the highest initial concentration (35°Brix). When the stirring speed was decreased the final concentration decreased also. The lowest concentration was obtained at  $\omega = 0$  and was similar to the results reported in progressive freeze concentration (Miyawaki et al., 2005). The higher the stirring speed, the higher the solute elution. This result is explained by the increasing mass transfer rate of the solutes from the ice front to the liquid fraction due to the fluid motion (Liu et al., 1999; Caretta et al., 2006; Moreno et al., 2014a). It is relevant to appoint that the freeze concentration phenomenon was presented at a stirring speed of 0 rpm, equivalent to a partial block freeze concentration (Nakagawa et al., 2010).

GLM was applied to test the effect of the three factors ( $C_0$ ,  $\omega$  and  $T$ ) on the response variable ( $C_f$ ). The main effects for the three factors, as well as the double and triple interaction effects were statistically significant ( $p$ -value  $< 0.05$ ). The effect of  $\omega$  and  $T$  on the final concentration at the initial concentration of 35 °Brix was drastically smaller than the effect at 15 and 25°Brix. The viscosity, which is one of the limiting factors of the kinetics of the process increases rapidly from 30°Brix and close to the freezing point temperatures (Telis et

al., 2007). The increasing bulk solute concentration caused increasing viscosity, thus decreasing solute diffusivity. The mass transfer of the solute near the ice-liquid interface would be retarded, thus there would be a greater tendency of the sucrose molecules to be trapped by the ice. The agitation had a positive effect on the final concentration of the solution. It is known that increasing agitation speed can increase the mass transfer coefficient to help the solute at the ice-solution interface to be transported into the bulk. However, the highest speed (2100 rpm) produced the best result just at the initial concentration of 15°Brix. For 25° and 35°Brix, for each temperature, the increase in agitation from 800 to 2100 rpm does not improve significantly the final concentration obtained. This may be because the higher heat generated at high agitation and the increases in viscosity affect the energy balance in the ice formation. These results are in agreement with those reported by Gu et al. (2008). The analysis of the refrigerant temperature revealed that the best results were obtained at -20°C and with agitation speed. This result is explained by the greater ice formation achieved at the lowest temperature. The average ice growth rate of the tests, calculated according to Chen et al. (1999), is within the interval between 1.89 and 3.93  $\mu\text{m/s}$ . These values coincide as those reported by Flesland (1995), Chen et al, (2000), Moreno et al. (2014b).

A classification of the results was made from the mean separation data obtained with the Tukey's tests. Three groups could be identified. The first group (dark gray in the Table I) corresponded to the combinations in which the highest final concentrations were obtained; this corresponded to the highest stirring speeds (between 800 and 2100 rpm), the lowest temperatures (-20 and -15 °C) and an initial concentration of 35 °Brix. The second group

(gray in the Table I) was made for the lowest final concentrations; this corresponded to the lowest or null stirring speed, the highest temperature (-10°C) and the lowest initial concentration (15 °Brix). Finally, the third group (white in the Table I) corresponded to the middle values of the final concentration, but the highest values of the concentration index.

The highest concentration obtained in the tests was 53°Brix. This result is comparable with the results of a progressive tubular system in which the sucrose solution was concentrated from 41.4 % w/w to 54.8 % w/w (Miyawaki et al., 2005). The limit of the freeze concentration systems is determined by the eutectic point. Crystals of ice and solute are produced simultaneously in a binary solution at the eutectic point. Consequently, the separation is not possible (Deshpande et al., 1984; Flesland, 1995). The eutectic concentrations of sucrose solutions varies from 54 to 62.4 % w/w according to the source (International Critical Tables (1926-1930); Chudotvortsev and Yatsenko, 2007; Chandrasekaran and King, 1971). Consequently, the limiting concentration of the sucrose solution system was reached with the progressive freeze concentration used in the present work.

## **Behaviour Of The Other Response Variables**

### **Concentration Index**

The concentration index (CI) was calculated from the final concentration of the tests. The results of the highest and the lowest CI for each initial concentration are shown in Table II.

The highest CI values were obtained for the lowest initial concentration. The lower the initial concentration, the easier the diffusion of water molecules to the ice crystal surface. This result is explained because the viscosity of the solution increases when the concentration increases, consequently the heat and mass transfer rate decrease. For this reason, the progressive freeze concentration was more efficient at low solute concentrations (Miyawaki et al., 2001; Moreno et al., 2014a). The highest concentration index was obtained at 2100 rpm, -20°C, and an initial concentration of 15 °Brix. In this treatment the final concentration was 46°Brix with a concentration index of 3.06. This result is remarkable compared with other freeze concentration techniques (Raventós et al., 2007). On the other hand, the least CI values were obtained for the null stirring speed and the highest refrigerant temperatures. This result can be explained by the fact that the decrease in stirring speed can decrease the mass transfer, and the higher the temperature of the refrigerant, the heat transfer can be reduced. The combined effect reduces the ice growth rate the final concentration obtained.

#### **Average Distribution Coefficient $\bar{K}$**

The average distribution coefficient  $\bar{K}$  is a measure of the amount of solutes occluded in the ice.

Figure 2 shows the  $\bar{K}$  and eff as a function of the initial concentration values for the freeze concentration tests.  $\bar{K}$  increased and eff decreased linearly with the initial concentration of the solution. These results are according to the expected and the results reported by other authors (Flesland, 1995; Chen and Chen, 2000; Miyawaki et al., 2012). In those studies the

most influent variable was the initial concentration. The increasing of  $\bar{K}$  values can be explained by the low mass transfer rate due to the solute presence, which promotes the retention of the solutes in the growing ice layer. More recent studies of optimization process in progressive freeze concentration with glucose solutions (Jusoh et al., 2013), mentioned that the controlling factors of effective distribution coefficient were in order: the flow rate of the solution, the initial concentration and the refrigerant temperature. In general, the higher the amount of solute retained in the ice, the lower solution concentration is achieved. Similar results were reported in falling film freeze concentration of sucrose solutions (Raventós et al., 2007), and coffee solutions (Moreno et al., 2014a).

#### **Yield Parameter ( $W_y$ )**

The average values of the yield parameter at different initial concentrations are presented in Figure 3. For all initial concentrations, around  $0.6 \text{ kg ice} \cdot \text{kg sol}^{-1} \cdot \text{h}^{-1}$  were separated.

The result was slightly smaller for the initial concentration of 35 °Brix due to the less water availability at high solute concentrations (Auleda et al., 2011; Belen et al., 2012). However, the differences were not significant. The yield for progressive freeze concentration was twice that reported for sucrose and coffee solutions with falling film freeze and block freeze concentration techniques respectively (Raventós et al., 2007 and Moreno et al., 2015).

## Concentration Kinetics

Figure 4 shows the concentration kinetics (Brix degrees) for the freeze concentration tests at a refrigerant temperature of  $-20^{\circ}\text{C}$ . The experimental results of the kinetics fitted a linear relationship, as shown in Table III.

The higher the initial concentration, the lower the concentration kinetics for all the stirring speeds. In addition, the velocity of concentration is hardly increased with the stirring velocity at the highest concentration, as shown in the slopes of the adjusted models in Table III. Considering that the power consumed for the agitation is proportional to the third power of the speed, it could be modified during the operation as a possible strategy to reduce operational costs. Bayindirli et al. (1993) reported on the behaviour of progressive freeze concentration of apple juice. The evolution of the concentration with the time is fitted to a sigmoidal function and the total process time varies from 200 to 500 minutes. However, the first 100 minutes described a linear behavior similarly to the present work.

## CONCLUSIONS

The three studied factors, namely the initial concentration, the stirring speed and the refrigerant temperature had a significant effect on the final concentration obtained by progressive freeze concentration of sucrose solutions. Likewise, the double and triple interactions had significant effects.

The highest concentration obtained was  $53^{\circ}\text{Brix}$  starting with a solution with  $35^{\circ}\text{Brix}$ , a stirring speed of 800 rpm and a refrigerant temperature of  $-20^{\circ}\text{C}$ . This result was close to

the eutectic point of sucrose solutions which is the limit of the freeze concentration technique.

The highest concentration index was obtained at the lowest initial concentration (15°Brix), the highest stirring speed (2100 rpm) and the lowest refrigerant temperature (-20°C).  $\bar{K}$  increased and eff decreased linearly with the initial concentration of the solution. The average yield parameter at different initial concentrations is around 0.6 kg ice·kg sol<sup>-1</sup>·h<sup>-1</sup>.

The concentration kinetics was fitted to a linear function for all the conditions. The concentration velocity was highest at the lowest concentration and the highest stirring speed.

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Table I. Final solute concentration after freeze concentration tests

$\omega$ (rpm)	$C_0 = 15^\circ\text{Brix}$			$C_0 = 25^\circ\text{Brix}$			$C_0 = 35^\circ\text{Brix}$		
	T=-10°C	T = -15°C	T =-20°C	T = -10°C	T = -15°C	T =-20°C	T = -10°C	T = -15°C	T =-20°C
0	17.1±1.4 <sup>p</sup>	27.7±1.1 <sup>o</sup>	27.1±0.9 <sup>o</sup>	26.6±0.4 <sup>o</sup>	37.2±1.1 <sup>lmn</sup>	33.6±1.2 <sup>n</sup>	38.7±1.7 <sup>klm</sup>	40.4±0.3 <sup>ijkl</sup>	42.6±1.1 <sup>ghij</sup>
500	26.6±0.4 <sup>o</sup>	27.8±1.7 <sup>o</sup>	39.5±1.7 <sup>kl</sup>	33.7±0.3 <sup>n</sup>	36.9±1.0 <sup>lmn</sup>	44.7±1.7 <sup>efgh</sup>	41.8±0.2 <sup>ghijk</sup>	45.2±1.4 <sup>defg</sup>	49.5±1.3 <sup>abc</sup>
800	28.4±0.6 <sup>o</sup>	34.9±0.5 <sup>mn</sup>	42.5±1.2 <sup>ghij</sup>	38±0.3 <sup>klm</sup>	42±0.4 <sup>fghijk</sup>	49±2.3 <sup>abcd</sup>	42.3±1.1 <sup>fghij</sup>	49.6±0.9 <sup>abc</sup>	53±0.9 <sup>a</sup>
2100	33.6±0.3 <sup>n</sup>	42.3±0.9 <sup>ghij</sup>	46±0.6 <sup>cdef</sup>	40.8±0.7 <sup>hijkl</sup>	44.4±0.7 <sup>efghi</sup>	47.1±0.8 <sup>bcde</sup>	44.7±0.9 <sup>efgh</sup>	49.6±1.8 <sup>abc</sup>	50.7±0.9 <sup>a,b</sup>

Data are expressed as mean  $\pm$  SD (n=3). Superscript letters show the grouping information for the 3-way interaction, using the Tukey method (95% confidence). Means that do not share a letter are significantly different.

Table II. Concentration index values for the freeze concentration tests

Initial concentration	15°Brix		25°Brix		35°Brix	
	Min	Max	Min	Max	Min	Max
Stirring speed (rpm)	0	2100	0	800	0	800
Temperature (°C)	-10°C	-20°C	-10°C	-20°C	-10°C	-20°C
CI values	1.02	3.06	1.06	2.02	1.06	1.53

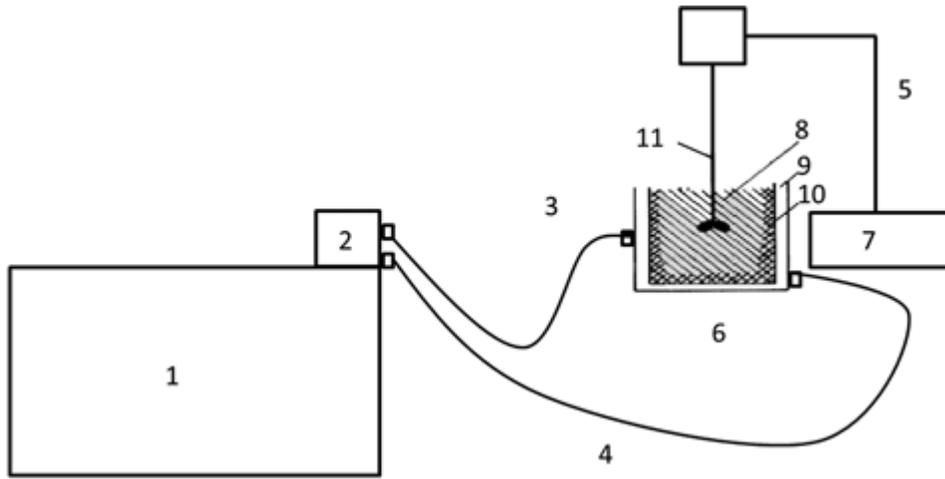
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Table III. Fitted models for the kinetics of concentration at -20°C.

	15 °Brix		25 °Brix		35 °Brix	
rpm	Equation	R <sup>2</sup>	Equation	R <sup>2</sup>	Equation	R <sup>2</sup>
0	$y = 0.15x + 14.15$	0.98	$y = 0.10x + 23.25$	0.83	$y = 0.08x + 33.78$	0.88
500	$y = 0.24x + 13.25$	0.97	$y = 0.22x + 24.28$	0.97	$y = 0.16x + 35.99$	0.98
800	$y = 0.30x + 13.29$	0.91	$y = 0.25x + 27.33$	0.96	$y = 0.17x + 37.76$	0.91
2100	$y = 0.40x + 14.44$	0.99	$y = 0.27x + 25.61$	0.99	$y = 0.194x + 36.45$	0.98

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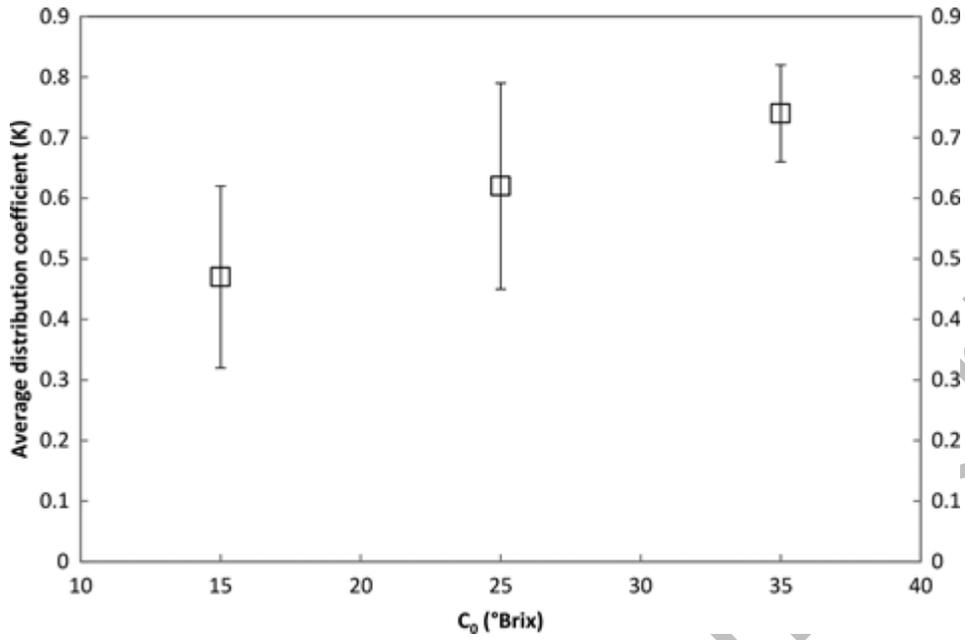
Figure 1. Experimental set-up



- 1 : thermostatic bath
- 2 : temperature control system
- 3,4 : refrigerant inlet and outlet ducts
- 5 : communication between agitator and speed regulation system
- 6 : jacketed vessel
- 7 : speed regulation system
- 8 : sucrose solution to be concentrate
- 9 : refrigerant into jacketed vessel
- 10 : ice formed on wall
- 11 : agitator

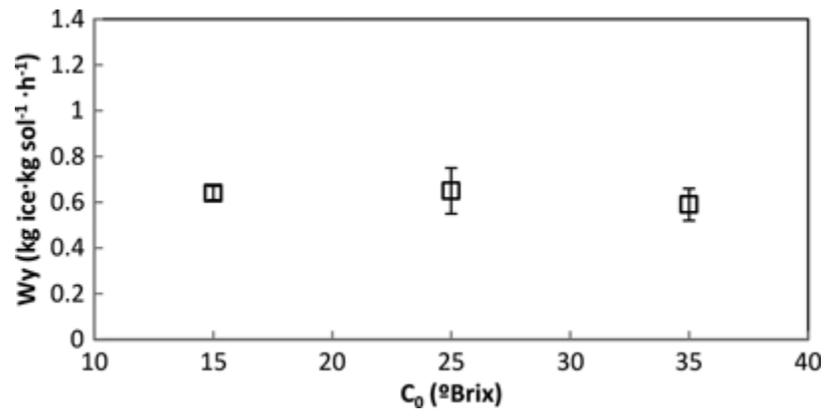
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Figure 2. Average distribution coefficient ( $\square$ ) as a function of the initial concentration



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Figure 3. Average yield parameter ( $W_y$ ) as a function of the initial concentration.



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Figure 4. Kinetics of the concentration process at  $-20^{\circ}\text{C}$ . (○) 0 rpm, (○) 200 rpm, (○) 500 rpm, (○) 2100 rpm. (a): 15 °Brix, (b): 25 °Brix, (c): 35 °Brix.

