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Integrated system of progressive freeze-concentration combined with partial ice-melting for yield improvement

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ABSTRACT

A 25L-scale tubular ice system for progressive freeze-concentration was constructed and combined with a 75L-scale partial ice-melting system for yield improvement. From the measurement of freezing point of various fruits juices, nine standard operation programs were prepared in cooling and circulation flow rate for the tubular ice system. The standard programs were successfully tested for progressive freeze-concentration of sucrose solutions with concentration varied from 3 to 30%. By choosing one of the standard program, apple juice was effectively concentrated from 12.8 to 21.0 °Brix with 79.0% yield, which was improved to 90% by recovering the 30% of the initially-melted fractions by using the partial ice-melting system. The tubular ice system can be easily scaled-up more, if necessary, simply by increasing the number of tubes.

1. Introduction

Freeze-concentration has been known as the best concentration method in terms of preserving original quality in the concentrated product (Deshpande et al., 1982) among the methods for concentration including evaporation and membrane technique. However, the conventional method for freeze concentration, known as suspension crystallization (SC) (Huige and Thijssen, 1972), requires a complicated system and a high capital cost so that its practical use is still limited for concentration of liquid food. Moreover, this system is applicable only to a large system because its operation mode is limited only to continuous mode with a long residence time.

Progressive freeze-concentration (PFC) is an alternative freeze concentration method where only a single ice crystal is formed on a cooling plate (Matthews and Coggeshall, 1959; Shapiro, 1961). This makes the system much simpler than SC so that the operation mode and the production scale are much flexible as compared with SC. We started from a cylindrical test apparatus for PFC to show its effectiveness for high quality concentration (Liu et al., 1997, 1999) and established theoretical basis for PFC based on the concentration polarization model (Miyawaki et al., 1998).

As for a scale-up for PFC, a falling film reactor has developed (Flesland, 1995) and has applied for concentration of variety of liquid food (Hernandez et al., 2009, 2010; Sanchez et al., 2010; Sanchez et al., 2011). In this system, the ice crystal grows on a vertically placed cooling plate on which the solution to be concentrated flows as a falling film. Although this system is simple, the limited liquid flow rate on the cooling surface may result in limited mass transfer between the ice and liquid phases to limit the separation efficiency (Miyawaki et al., 1998). In addition, this reactor has an open air surface which could lead to the loss of volatile flavor components. Therefore, a closed tubular ice system with a circulating flow was developed for the scale-up of PFC, which provides a good mass transfer and a controlled heat transfer in a closed system (Miyawaki et al., 2005). This system is expected to give a high separation efficiency and a high-quality for the concentrated product especially in the retention of volatile flavors (Miyawaki et al., 2012, 2016; Gunathilake et al., 2014a).

The major drawback in PFC system is the decrease in yield with

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an increase in the concentration, or osmotic pressure, of sample because of the solute inclusion into the ice phase (Gu et al., 2005). To overcome this problem, we proposed partial ice-melting technique (Miyawaki et al., 2012), where the principle in the classical freezing–thawing technique was effectively applied.

In the freezing–thawing process, the initially melted fractions after freezing contains the higher concentration of solute than the latterly melted fractions. This principle has been applied, as a block-ice technique, in desalination from sea water (Johnson, 1993; Khawaji et al., 2008; Mandri et al., 2011; Rich et al., 2012) and various applications in industrial purpose have been explored (Yee et al., 2003, 2004; Nakagawa et al., 2009, 2010a,b; Moreno et al., 2013, 2014a,b). Only by freezing–thawing, however, concentration efficiency is limited so that multiple operations are inevitable (Aider and Ounis, 2012) to obtain a highly concentrated product, which requires a high energy consumption in the process.

To improve the efficiencies in separation and energy, a combination of freezing–thawing technique with PFC will be an attractive option. In the previous paper, we successfully combined the partial ice-melting (PIM) technique with PFC to improve the yield (Miyawaki et al., 2012). Then, we made a test apparatus for the melting of ice under the controlled conditions in temperature and stirring speed and applied it to improve the yield in PFC by the tubular ice system (Gunathilake et al., 2014b). In the present paper, we report on the integrated PFC system combined with PIM system for improvement in yield.

2. Experimental method

2.1. Materials

Apple fruits (*Malus domestica* Borkh. 'Senshu', and 'Shusei'), grape fruits (*Vitis Labruscana* Bailey 'Kyoho', and 'Ruby roman'), and melon (*Cucumis melo* L. 'Earl's Favorite') were grown and harvested in Ishikawa Agriculture and Forestry Research Center. After harvesting, the fruits were crushed with addition of ascorbic acid at 0.2%, if necessary, strained by pulper finisher (Sun Food Machinery, Tokyo), treated with 0.4% pectin-degrading enzyme (Sucrase N, Mitsubishi Chemical Foods, Tokyo) for 2 h at 40 °C, and filtrated for clarification by a filter press (M200, Makino, Aichi). After the filtration, the fruits juice was packed and heated for 10 min in boiling water for inactivation of the added enzyme and sterilization. Pineapple juice (variety unknown) and pear juice (*Pyrus Communis* L. 'La France') were kindly gifted by Kakoh Fruits and Flavors, Tokyo.

2.2. Measurement of freezing point of fruits juices

Freezing point of fruits juices were measured according to our previous method (Miyawaki et al., 1997) with a little modification. The sample (3 mL) in a plastic tube (15 mm in diameter), equipped inside with a thermistor (0.01 °C in accuracy: D641, Takara Thermister, Yokohama), was frozen completely at −20 °C in a cooling bath (NCB-3200, Eyela, Tokyo) and then warmed to melt in the atmosphere at room temperature with stirring by a vortex mixer. The change in the temperature was recorded by a recorder (PRR-5021, Toa DKK, Tokyo). Freezing point depression was determined from the melting curve. Freezing point of the sucrose solution was obtained from the literature (Weast, 1974).

2.3. Progressive freeze-concentration

A vertically placed tubular ice system (Miyawaki et al., 2005) with circulating flow, newly constructed by us, was used for PFC. This system is composed of two straight jacketed pipes (2.3 m long, 72.3 mm in diameter), a bent pipe to combine straight pipes, a pump for circulation (E2H, Toste, Tokyo), and a feed tank. The apparent total capacity of the tubular reactor was 25.0 L.

Sucrose solutions and apple juice (*Malus domestica* Borkh. 'Senshu') were used as samples to be concentrated. A sample, introduced into the PFC system, was circulated and cooled, from the jacket-side at the straight pipe, by a coolant, the temperature of which was controlled by a controller and a refrigerator (SCS-5.5HW-M, Step-Science, Tokyo) to form ice crystal on the inner surface of the pipe. The jacket temperature (0 to −25 °C) and the circulation flow rate (7–35 t/h) were controlled by a program with computer. The flow rate at 29.6 t/h corresponds to the linear velocity at 2 m/s in the tubular ice system when no ice exists inside.

Concentration process was controlled by a program for the coolant temperature and the circulation flow rate measured by an electromagnetic flow meter (Promag 50H, Endress Hauser Japan, Tokyo). Concentration process was monitored by a thermister placed inside the tube, which measured the change in freezing point depression along with the concentration process. The ice crystal volume formed in the system was estimated from the effluent liquid volume from the system because of the inflation in volume (8.5%) accompanied with the phase change from water to ice.

For the PFC-concentration of high-concentration sucrose solution and apple juice, which has a relatively high osmotic pressure, the seed ice lining step was necessary to prevent the initial supercooling for obtaining the higher yield (Liu et al., 1998; Miyawaki et al., 2005). For this purpose, pure water was firstly introduced into the tubular system to form a small amount of seed ice on the cooling plate. Then the water was discharged and the precooled sample was introduced into the system to start PFC-concentration. After the concentration process, the concentrated solution was taken out from the bottom of the system. Then, the coolant temperature was raised up to 20 °C to melt the ice surface formed in the system. The ice slid out gravimetrically from the bottom of the vertically placed tube.

The concentration in Brix was analyzed for the original solution, the concentrate after PFC-concentration, and the melted ice by a refractometer (APAL-1, As One, Osaka). The yield based on Brix was obtained by the following equation.

$$\text{Yield} = C_{conc}V_{conc}/(C_{conc}V_{conc} + C_{ice}V_{ice}) \quad (1)$$

where C_{conc} and C_{ice}, are concentrations in Brix in concentrated solution phase and ice phase, respectively, and V_{conc} and V_{ice} are volumes in those phases after melting.

2.4. Partial ice-melting system

After PFC-concentration, the ice, slidden from the bottom of the tube, was broken in pieces and transferred by a conveyer to the partial ice-melting (PIM) system (75 L in volume equipped with a stirrer inside) with a jacket and the ice was melted with a gentle stirring at a controlled temperature in the jacket side by a circulator (LTC-1200A, As One, Osaka).

Fig. 1 shows the whole picture of the integrated system of the PFC system by the tubular ice system (①) and the PIM system (②) for yield improvement. These two systems are combined with a conveyer (③) to transfer ice.

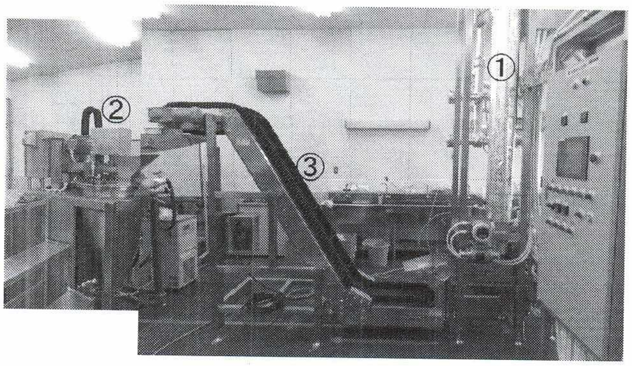

Fig. 1. Integrated system for progressive freeze-concentration by tubular ice system (①) combined with partial ice-melting sytem (②) for yield improvement. The two systems are combined with a conveyer (③) to transfer ice.

3. Results and discussion

3.1. Freezing point of fruits juices and determination of program for tubular ice system

Table 1 shows the concentrations in Brix and freezing points of various fruits juices tested here. Concentration varied from 9.5 to 19.5 °Brix and freezing point changed accordingly from −0.85 to −2.46 °C. Fig. 2 shows the freezing point of fruits juices and sucrose solutions and its relationship with concentration in Brix. Good linear correlations were observed for fruits juices and sucrose solutions, respectively, although the correlations are little different between the two. Fruits juices showed a little higher freezing point depression than pure sucrose solutions probably because of the effect of components other than sugars, such as organic acids (Miyawaki et al., 2016).

In PFC, the freezing point is the ultimately important parameter to determine the cooling program. From the data in Fig. 2, three modes in cooling program were determined as shown in Fig. 3 which corresponds to 3%, 10%, and 30% sucrose solutions, respectively. As for the circulation program, three modes were also determined as 'Slow', 'Standard', and 'Fast' as shown in Fig. 4. According to the concentration polarization model (Miyawaki et al., 1998; Gu et al., 2008), the following equation has been theoretically obtained for the effective partition coefficient of solute, K, ($=C_i/C_L$; C_i, concentration in ice phase; C_L, concentration in solution phase).

$$K = K_0/[K_0 + (1 - K_0)\exp(-u/a/v^{0.8})] \qquad (2)$$

where u is ice crystal growth rate, v is flow velocity at the ice-liquid interface, a is an experimental constant, and K_0 is the limiting partition coefficient, which corresponds to the K at $v \rightarrow \infty$ and/or $u \rightarrow 0$. Eq. (2) shows that the higher v gives the lower K. This means that the higher circulation speed is preferable for the higher yield, if it is allowed by the physical properties like viscosity, of the sample.

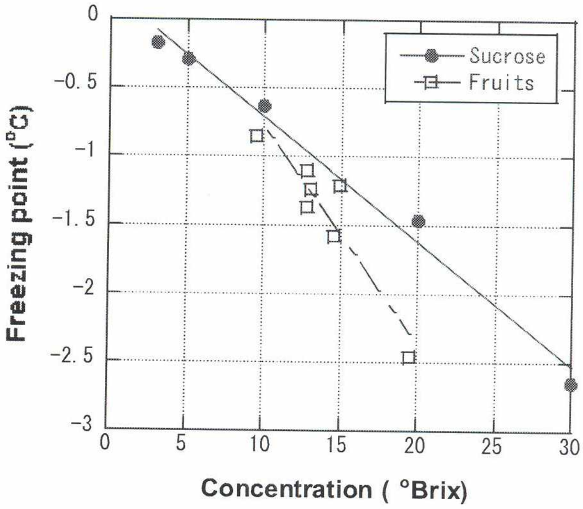

Fig. 2. Freezing point of various fruits juices and sucrose solutions and its relationship with concentration in Brix.

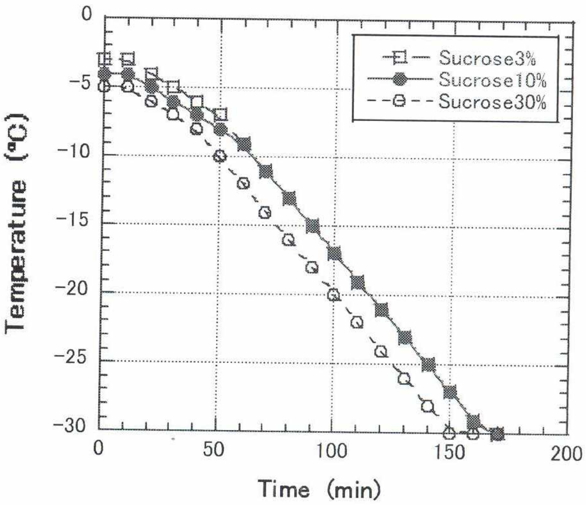

Fig. 3. Three modes in cooling program for progressive freeze-concentration by tubular ice system.

From the above results, the nine standard programs are available by combining three modes in cooling program and three modes in circulation flow rate. In practice, the most appropriate program is employed from these standard programs depending on the physical properties of samples.

Table 1
Freezing point of fruits juices.

Fruits	Variety	Concentration (°Brix)	Freezing point (°C)
Apple	*Malus domestica* Borkh. 'Senshu'	12.8	−1.37
Apple	*Malus domestica* Borkh. 'Shusei'	12.8	−1.11
Grape	*Vitis Labruscana* Bailey 'Kyoho'	19.5	−2.46
Grape	*Vitis Labruscana* Bailey 'Ruby Roman'	14.6	−1.58
Pineapple	Variety unknown	13.0	−1.23
Melon	*Cucumis melo* L. 'Earl's Favorite'	9.5	−0.85
Pear	*Pyrus Communis* L. 'La France'	14.9	−1.21

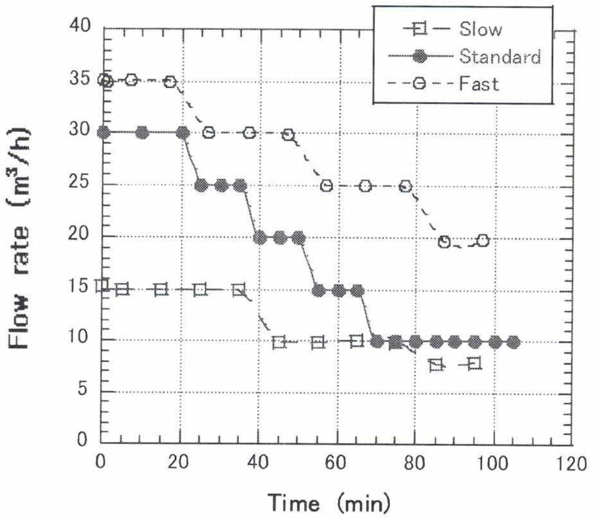

Fig. 4. Three modes in circulation flow program for progressive freeze-concentration by tubular ice system.

3.2. *Progressive freeze-concentration of sucrose solutions*

Sucrose solution at 3.3% was PFC-concentrated by the cooling program for '3% sucrose' with circulation program varied. Results are shown in Table 2, which shows that the faster circulation speed gives the higher yield as was expected by Eq. (2). In the present experimental design, all the experiments were carried put single because differences in yield were clear among the different operating conditions and the difference observed has enough reason predicted theoretically by the concentration polarization model expressed by Eq. (2). Therefore, statistical analysis was not applied by repeating experiments at the present stage. It will be necessary in the next stage of the routine production.

In the case with 3.3% sucrose, freezing point depression was not so high, which corresponds to the relatively lower osmotic pressure (Gu et al., 2005), so that the seed ice may not be necessary (Miyawaki et al., 2005). In Table 2, however, the effect of seed ice was observed. In the standard circulation mode, the yield was slightly improved from 89.7% to 92.9% by applying the seed ice. Effect of seed ice exists in the prevention of initial supercooling in PFC (Liu et al., 1998).

Table 3 shows the results for PFC-concentration of sucrose solutions at various concentrations. In this case, the circulation mode was fixed at 'Fast' in all the case. With an increase in sucrose concentration, the yield decreased drastically because of the increase in the osmotic pressure of the sample (Gu et al., 2005). In this case, a substantial effect of seed ice was observed in the case with 30% sucrose, showing the necessity of seed ice for a sample with a high osmotic pressure.

3.3. *Progressive freeze-concentration of apple juice*

PFC system was applied to concentrate apple juice (*Malus domestica* Borkh. 'Senshu'). In this case, a small amount of seed ice layer was formed inside the tubular reactor and the apple juice sample, precooled down to the freezing point, was introduced into the system. The freezing point of the apple juice was −1.37 °C. The cooling program for 'Sucrose 10%' in Fig. 3 and the circulation program 'Standard' in Fig. 4 were employed. As for the circulation program, 'Fast' program may cause the heat generation because of the high pump speed so that 'Standard' program was employed in this case.

Under the program described above, the ice volume increased in the reactor and the temperature of the sample decreased as shown in Fig. 5. The sample temperature corresponds to the freezing point, which decreased along with the progress in the concentration process. By this system, 23.6 L of the apple juice was concentrated, in about 2 h operation time, to 10.0 L with concentration increased from 12.8 to 21.0 °Brix with a yield of 79.0% as shown in Table 4.

3.4. *Yield improvement by partial ice-melting system*

In the present case of PFC-concentration of apple juice, the yield may not be satisfactory but this could be improved to the necessary level by applying the PIM technique (Miyawaki et al., 2012; Gunathilake et al., 2014b). Therefore, the ice formed was transferred to the PIM system operated at 5 °C with a gentle stirring (~ 5 rpm). The result is shown in Fig. 6. The concentration of the initially melted fractions are much higher than those in the latter fractions so that the yield improvement was expected by recovering the initial fractions (Miyawaki et al., 2012; Gunathilake et al., 2014b). In the present case, the yield could be improved from 79.0% to 90% by recovering the 30% of initially melted fractions. The PIM system is expected to be a breakthrough for the practical application of PFC to overcome the inherent drawback of PFC, in which the incorporation of solute into the ice phase has caused the reduction in yield for highly concentrated samples (Gu et al., 2005). Recently, Moreno et al. (2014a,b) also proposed to apply fractional thawing, or partial ice-melting, to improve the yield in the falling film freeze concentration.

4. Conclusion

A 25 L-scale tubular ice system for PFC-concentration combined with PIM-system was newly constructed. From the measurement of freezing point of various fruits juices, nine standard operation programs were prepared in cooling and circulation flow rate. The standard programs were successfully tested for PFC-concentration of sucrose solutions with concentration varied from 3 to 30%.

Table 2
Progressive freeze-concentration of 3.3% sucrose solution by tubular ice system with cooling mode for '3% sucrose'.

Circulation mode	Seed ice	Classification	Volume(L)	Conc.(°Brix)	Yield
Slow	No	Original solution	24.7	3.3	–
		Concentrate	9.6	6.7	75.3
		Ice	15.1	1.4	
Standard	No	Concentrate	10.5	5.9	89.7
		Ice	14.2	0.5	
Fast	No	Concentrate	11.9	6.1	93.4
		Ice	12.8	0.4	
Standard	Yes	Concentrate	10.7	5.1	92.9
		Ice	13.95	0.3	

Table 3
Progressive freeze-concentration of sucrose solution at various concentrations by tubular ice system.

Cooling mode	Circulation mode	Seed ice	Classification	Volume(L)	Conc.(°Brix)	Yield
3% sucrose	Fast	No	Original solution	24.7	3.3	–
			Concentrate	11.9	6.1	93.4
			Ice	12.8	0.4	
10% sucrose	Fast	No	Original solution	24.7	10.0	
			Concentrate	9.1	19.9	75.8
			Ice	15.6	3.7	
30% sucrose	Fast	No	Original solution	24.7	30.6	
			Concentrate	8.32	31.3	41.7
			Ice	12.7	28.7	
30% sucrose	Fast	Yes	Original solution	24.7	30.6	
			Concentrate	12.0	37.9	72.7
			Ice	13.2	12.9	

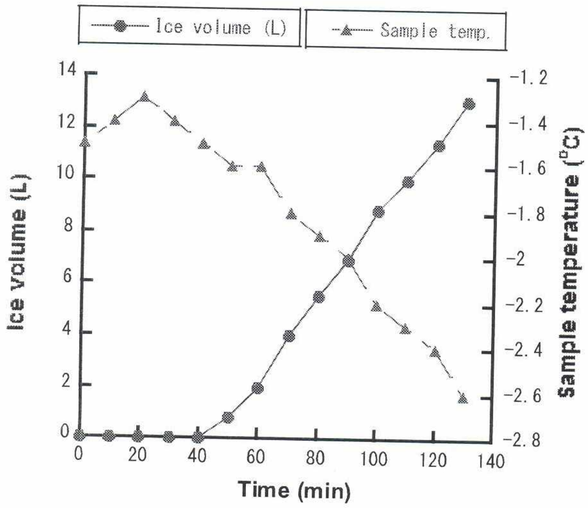

Fig. 5. Changes in ice volume formed and sample temperature during the concentration process of apple juice by progressive freeze-concentration.

Table 4
Progressive freeze-concentration of apple juice by tubular ice system.

Classification	Volume(L)	Conc.(°Brix)	Yield
Original solution	23.6	12.8	–
Concentrate	10.0	21.0	79.0
Ice	13.6	4.1	
Ratio	2.36	1.64	–

Effectiveness of the seed ice was also tested especially for the high concentration sample.

Apple juice was PFC-concentrated from 12.8 to 21.0 °Brix with 79.0% yield, and then the yield was effectively improved to 90% by recovering the 30% of the initially-melted fractions by using PIM-system. As for the general concentration technology with a high quality (Miyawaki et al., 2012, 2016; Gunathilake et al., 2014a), the present system is expected to be applicable not only to fruits juices but also to various liquid food and products in biomedical field by overcoming the inherent drawback of PFC by PIM-system. The present tubular ice system can be easily scaled-up, if necessary, simply by increasing the number of tubes. Now, we are planning to increase the number of tubes from 2 to 4 to increase the scale of apparatus from 25 to 50 L.

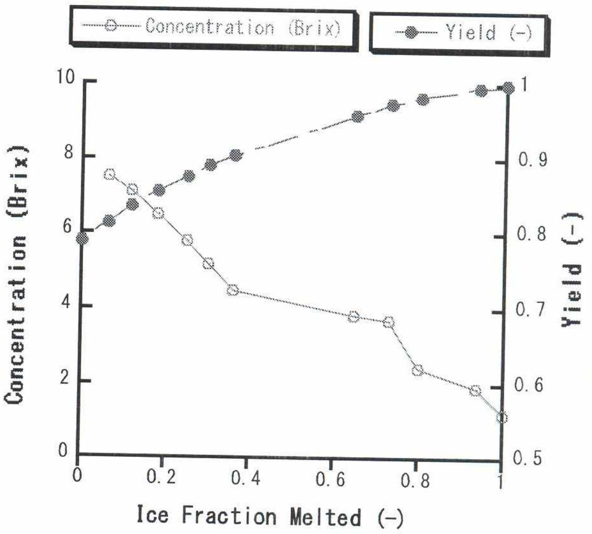

Fig. 6. Yield improvement by partial ice-melting system for PFC-concentrated apple juice operated at 5 °C with a gentle strring (~5 rpm).

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