

R&D NOTE

Separation of p-Xylene from the Multicomponent Xylene System by Stripping Crystallization

Lie-Ding Shiau, Chun-Ching Wen, and Bo-Shiue Lin

Dept. of Chemical and Materials Engineering, Chang Gung University, Taoyuan, Taiwan, R.O.C

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Introduction

Xylenes have very broad applications in chemical industries. The mixed xylenes produced mainly consist of p-xylene (PX), m-xylene (MX), o-xylene (OX), and ethylbenzene (EB). Due to their similar molecular structures, these isomers have close boiling points (e.g., $PX = 138.37^\circ\text{C}$, $MX = 139.12^\circ\text{C}$, $OX = 144.41^\circ\text{C}$ and $EB = 136.19^\circ\text{C}$). Therefore, it is very difficult to separate them from each other by distillation. The cost for separation and purification of these isomers is generally very high.¹

Stripping crystallization (SC), previously known as distillative freezing (DF), is first introduced by Cheng and Cheng² to separate the mixture of the volatile compounds at a reduced pressure. In principle, the SC process is operated at a triple point condition, in which the liquid mixture is simultaneously vaporized and crystallized due to the three-phase equilibrium. Thus, SC combines stripping and crystallization to produce pure crystals. By lowering temperature and reducing pressure during the operation, the SC process results in the formation of pure solid, and liquid phase and vapor phase of mixtures. In essence, the SC process is continued until the liquid phase is completely eliminated, and only the pure solid crystals remain in the feed. The low-pressure vapor formed in the process is condensed and removed.

Shiau et al.^{3,4} described the basic principle of SC in detail and successfully applied the SC process to produce PX crystals from the binary xylene mixture (PX+MX and PX+OX, respectively). The objective of this work is to investigate the feasibility of the SC process in producing pure PX crystals from the mixed xylene system of PX, MX, EB and OX.

Simulation of the SC process for the multicomponent system

The SC process is simulated in a series of N equilibrium stage operation in Figure 1 to produce component-1 crystals

from the multicomponent system of j components ($j \geq 3$). Each stage is operated under an adiabatic condition at a three-phase equilibrium state of solid phase of pure component-1, and liquid phase and vapor phase of the mixture. The temperature of each stage is chosen to meet $T_n - T_{n-1} = \Delta T$ for $n = 2, \dots, N$. The vapor formed in each stage is condensed to the liquid and removed while the solid and the liquid formed in each stage enter the next stage. The whole process starts from the liquid feed and continues until the liquid phase is completely eliminated.

The total material balance in stage n can be described by

$$S_{n-1} + L_{n-1} = S_n + L_n + V_n \quad (1)$$

It is noted that in each stage solid phase comprises only pure solid of component-1, while the liquid and the vapor phase consists of the mixture containing j components. Thus, the material balance of component- i in stage n can be described by

$$L_{n-1}(X_i)_{n-1} = L_n(X_i)_n + V_n(Y_i)_n \quad \text{for } i = 2 \sim j \quad (2)$$

As each stage is operated under an adiabatic condition, the latent heat released in forming component-1 crystals is removed by vaporizing portions of the liquid mixture. Thus, the energy balance in stage n can be described by

$$S_{n-1}(H_S)_{n-1} + L_{n-1}(H_L)_{n-1} = S_n(H_S)_n + L_n(H_L)_n + V_n(H_V)_n \quad (3)$$

The solid-liquid equilibrium of component-1 in stage can be described by^{5,6}

$$\ln[(X_1)_n(\gamma_1)_n] = \frac{\Delta H_{m,1}}{R} \left(\frac{1}{T_{m,1}} - \frac{1}{T_n} \right) + \frac{\Delta C_{P,1}}{R} \left(\frac{T_{m,1} - T_n}{T_n} \right) - \frac{\Delta C_{P,1}}{R} \ln \left(\frac{T_{m,1}}{T_n} \right) \quad (4)$$

Usually at pressures of 1 atm and less, the assumption of ideal gases introduces little error. As SC is operated at low-

Correspondence concerning this article should be addressed to L-D. Shiau at shiau@mail.cgu.edu.tw.

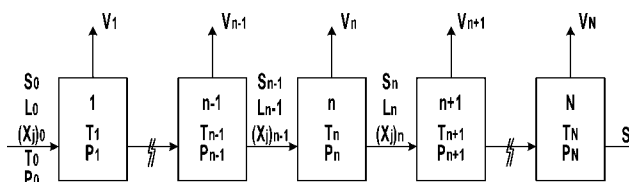


Figure 1. The simulated SC operation where each stage is operated under an adiabatic condition at a three-phase equilibrium state.

pressures ($<1 \text{ atm}$), the vapor-liquid equilibrium of component- i in stage n can be described by^{5,6}

$$P_n(Y_i)_n = (P_i^{\text{sat}})(X_i)_n(\gamma_i)_n \quad \text{for } i = 1 \sim j \quad (5)$$

Besides, we have

$$\sum_{i=1}^j (X_i)_n = 1 \quad (6)$$

$$\sum_{i=1}^j (Y_i)_n = 1 \quad (7)$$

Due to the structure similarity, it is assumed that these isomers form an ideal liquid solution, i.e., $\gamma_i \rightarrow 1$ (for $i = 1 \sim j$). In the SC simulation T_0 is chosen as the triple point temperature of component-1 at the initial composition of the feed. As T_0 is chosen, T_n (for $n = 1 \sim N$) is determined by assigning the value of ΔT . When T_n is specified, Eqs.(1–7) constitute a set of $2j + 4$ equations, that can be simultaneously solved for $2j + 4$ unknowns - S_n , L_n , V_n , P_n , $(X_i)_n$ and $(Y_i)_n$ for $i = 1 \sim j$ in stage n .

Experimental

The experimental method is described in detail by Shiau et al.³

Results and Discussions

The SC experiments are performed to produce PX crystals from the multicomponent system of PX (component-1), MX (component-2), EB (component-3), and OX (component-4). Some physical properties of these isomers are listed on Table 1.¹ Other physical properties needed in the SC simulation are taken from NIST standard reference database 11.⁷

Separation of PX from the ternary mixture of PX, MX and EB ($j = 3$)

As OX is the easiest to distill from the xylene mixture because of the 5°C difference in the boiling point between it

and the next closest boiling isomer (MX), separation of PX from the ternary mixture of PX, MX and EB is investigated first.

The compositions and the temperatures of the eutectic points for the ternary mixture of PX, MX and EB are listed on Table 2. The diagram of the eutectic points is shown in Figure 2. The area within $Apex_1E_{12}E_{123}E_{13}$ represents the ternary compositions of the liquid mixture in which the SC process can be operated to produce pure crystals of PX (component-1). The liquid compositions during the SC operation cannot touch line $E_{12}E_{123}$ or line $E_{123}E_{23}$; otherwise, MX (component-2) or EB (component-3) will also crystallize out with PX. At the eutectic point E_{123} , all the three components crystallize from the liquid mixture.

Similarly, the area within $Apex_2E_{12}E_{123}E_{23}$ represents the ternary compositions of the liquid mixture in which the SC process can be operated to produce pure crystals of MX (component-2). The area within $Apex_3E_{13}E_{123}E_{23}$ represents the ternary compositions of the liquid mixture in which the SC process can be operated to produce pure crystals of EB (component-3).

The simulation results calculated by Eqs. (1–7) are listed in Tables 3–7 for the ternary mixture with various compositions. The simulated liquid compositions in each step are plotted in Figure 3 for various feed conditions. It shows that the simulated liquid compositions in each run all fall within the area $Apex_1E_{12}E_{123}E_{13}$. Therefore, pure PX crystals can be produced without the formation of eutectic crystals in the SC experiments.

Table 3 indicates the simulated results of the SC operation in a series of seven equilibrium stages with $\Delta T = 5^\circ\text{C}$ for a 200g liquid feed of 90% PX, 5% MX and 5% EB. T_0 is chosen as 9.11°C , which is the triple point temperature of PX at the initial feed composition. Based on the simulation, when the temperature and pressure reduces to $T = -24.7^\circ\text{C}$, and $P = 29.7 \text{ Pa}$, the liquid feed completely disappears, and a final PX product of 130.8g is obtained. Similarly, Table 4 indicates that, for a 200g liquid feed of 90% PX, 7% MX and 3% EB, a final PX product of 130.4g is obtained when the final SC is operated at and. Table 5 indicates that, for a 200g liquid feed of 90% PX, 3% MX and 7% EB, a final PX product of 131.1g is obtained when the final SC is operated at $T = -23.4^\circ\text{C}$ and $P = 39.0 \text{ Pa}$. It is seen in Tables 3–5 that the proposed SC operations are slightly different for a liquid feed of 90% PX with various compositions of impurities.

Table 6 indicates that, for a 200g liquid feed of 87% PX, 8% MX and 5% EB, a final PX product of 127.1g is obtained when the final SC is operated at $T = -39.0^\circ\text{C}$ and $P = 8.6 \text{ Pa}$. Table 7 indicates that, for a 200g liquid feed 83% PX, 5% MX, and 12% EB, a final PX product of 123.3g is obtained when the final SC is operated at T

Table 1. Some Physical Properties for PX, MX, EB and OX^{1,7}

Property	PX	MX	EB	OX
Molecular weight	106.167	106.167	106.167	106.167
Boiling point, $^\circ\text{C}$	138.37	139.12	136.19	144.41
Freezing point, $^\circ\text{C}$	13.26	-47.85	-94.95	-25.17
Triple point	581.55	3.21	4.01×10^{-3}	21.97
Triple point pressure, Pa (N/m^2) heat of melting, J/mol	1.711×10^4	1.157×10^4	9.184×10^3	1.360×10^4

Table 2. Compositions and the Temperatures of the Eutectic Points for the Ternary System of PX(component 1), MX(component 2) and EB(component 3)

	X ₁	X ₂	X ₃	T (°C)
E ₁₂₃	0.0125	0.182	0.805	-101.06
E ₁₂	0.128	0.872	0	-52.77
E ₂₃	0	0.187	0.813	-100.79
E ₁₃	0.017	0	0.983	-95.45

= -54.1°C, and $P = 1.7 \text{ Pa}$. The simulation shows that lower temperatures and lower pressures are required in the SC process to obtain a final PX product for a liquid feed with lower composition of PX.

The comparison of the experimental and simulation results for various ternary xylene systems of PX, MX and EB is listed in Table 8. It indicates that, when the experiments are performed based on the SC simulation, the purity of PX crystals can reach 99.5%–99.9% for the ternary xylene mixture with PX ranging from 83 to 90%. The actual recovery rates range from 51.7% to 67.9%, which are lower than the simulated recovery rates ranging from 72.4% to 74.3%.

Separation of PX from the quaternary mixture of PX, MX, EB and OX ($j = 4$)

As the diagram of the eutectic points for the quaternary mixture is complicated, it is not shown here. The eutectic point, at which all the four components crystallize from the quaternary mixture, is determined as $X_1 = 0.0083$, $X_2 = 0.146$, $X_3 = 0.792$, and $X_4 = 0.054$ at $T = -101.4^\circ\text{C}$.

The simulation results calculated by Eqs. 1–7 for the quaternary mixture with various compositions are listed in Tables 9–11. The simulated liquid compositions in each step for the three feed conditions should not touch the eutectic

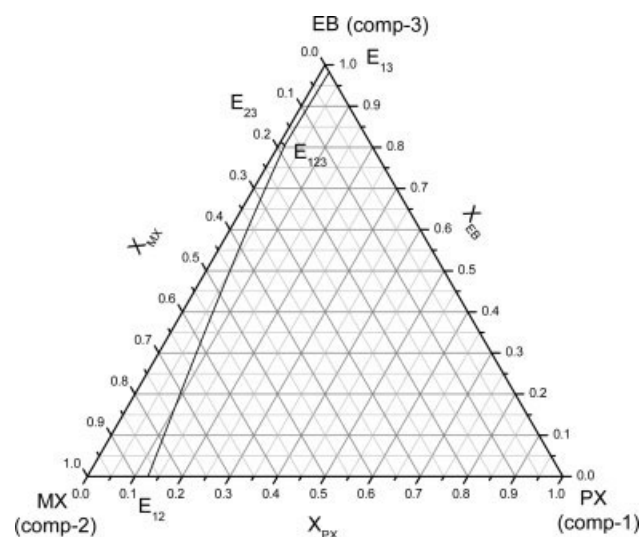


Figure 2. Eutectic points for the ternary system of PX, MX and EB.

Table 3. Simulation Results of Adiabatic SC Operation for the Ternary System of 90% PX, 5% MX and 5% EB ($\Delta T = 5^\circ\text{C}$)

n	T (°C)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}
0	9.11		200	0	0.9	0.05	0.05
1	4.11	320.2	49.1	105.4	0.790	0.109	0.101
2	-0.89	226.2	24.2	121.2	0.691	0.163	0.146
3	-5.89	157.4	14.1	126.7	0.601	0.214	0.185
4	-10.9	107.6	8.41	129.1	0.521	0.262	0.217
5	-15.9	72.3	4.61	130.2	0.449	0.309	0.242
6	-20.9	47.6	1.76	130.6	0.386	0.358	0.256
7	-24.7	34.0	0	130.8	0.342	0.408	0.250

Table 4. Simulation Results of Adiabatic SC Operation for the Ternary System of 90% PX, 7% MX and 3% EB ($\Delta T = 5^\circ\text{C}$)

n	T (°C)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}
0	9.11		200	0	0.9	0.07	0.03
1	4.11	318.2	50.6	104.3	0.790	0.150	0.060
2	-0.89	224.2	25.4	120.5	0.691	0.223	0.086
3	-5.89	155.5	15.0	126.1	0.601	0.291	0.108
4	-10.9	106.1	9.17	128.6	0.521	0.353	0.126
5	-15.9	71.1	5.28	129.8	0.450	0.411	0.139
6	-20.9	46.8	2.38	130.3	0.386	0.468	0.146
7	-25.9	30.1	0.05	130.4	0.330	0.528	0.142
8	-26.0	29.7	0	130.4	0.329	0.545	0.126

Table 5. Simulation Results of Adiabatic SC Operation for the Ternary System of 90% PX, 3% MX and 7% EB ($\Delta T = 5^\circ\text{C}$)

n	T (°C)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}
0	9.11		200	0	0.9	0.03	0.07
1	4.11	322.1	47.6	106.4	0.790	0.066	0.144
2	-0.89	228.3	23.1	121.9	0.691	0.100	0.209
3	-5.89	159.3	13.2	127.2	0.601	0.132	0.266
4	-10.9	109.2	7.66	129.5	0.521	0.164	0.315
5	-15.9	73.5	3.93	130.6	0.450	0.196	0.354
6	-20.9	48.5	1.14	131.0	0.386	0.233	0.380
7	-23.4	39.0	0	131.1	0.357	0.268	0.374

Table 6. Simulation Results of Adiabatic SC Operation for the Ternary System of 87% PX, 8% MX and 5% EB ($\Delta T = 5^\circ\text{C}$)

n	T (°C)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}
0	7.80		200	0	0.87	0.08	0.05
1	2.80	291.6	70.2	90.3	0.763	0.149	0.088
2	-2.20	204.9	39.0	111.1	0.666	0.212	0.121
3	-7.20	141.7	25.2	118.9	0.579	0.270	0.151
4	-12.2	96.4	17.4	123.2	0.502	0.323	0.175
5	-17.2	64.4	12.1	124.8	0.432	0.372	0.196
6	-22.2	42.2	8.28	125.7	0.371	0.418	0.212
7	-27.2	27.1	5.27	126.5	0.316	0.461	0.222
8	-32.2	17.0	2.78	126.8	0.268	0.505	0.227
9	-37.2	10.4	0.65	127.0	0.226	0.554	0.220
10	-39.0	8.6	0	127.1	0.212	0.590	0.198

Table 7. Simulation Results of Adiabatic SC Operation for the Ternary System of 83% PX, 5% MX and 12% EB ($\Delta T = 5^\circ\text{C}$)

n	T ($^\circ\text{C}$)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}
0	5.99		200	0	0.83	0.05	0.12
1	0.99	260.5	89.2	76.7	0.727	0.083	0.191
2	-4.01	182.9	54.1	99.7	0.634	0.113	0.253
3	-9.01	126.3	37.2	110.1	0.550	0.141	0.309
4	-14.0	85.7	27.2	115.2	0.476	0.167	0.358
5	-19.0	57.1	20.5	117.8	0.409	0.191	0.400
6	-24.0	37.3	15.6	120.1	0.350	0.213	0.437
7	-29.0	23.8	11.8	121.7	0.298	0.235	0.467
8	-34.0	14.9	8.73	122.3	0.252	0.256	0.492
9	-39.0	9.1	6.12	122.7	0.212	0.277	0.511
10	-44.0	5.4	3.85	123.0	0.178	0.299	0.523
11	-49.0	3.1	1.84	123.2	0.147	0.327	0.526
12	-54.1	1.7	0	123.3	0.121	0.372	0.507

points; otherwise, pure PX crystals can not be obtained due to the formation of eutectic crystals.

Table 9 indicates that, for a 200g liquid feed of 90% PX, 3.3% MX, 3.4% EB and 3.3% OX, a final PX product of 129.2g is obtained, based on the simulation when the final SC is operated at $T = -30.7^\circ\text{C}$ and $P = 16.7\text{ Pa}$. Table 10 indicates that, for a 200g liquid feed of 88.5% PX, 2% MX, 7% EB, and 2.5% OX 7% MX, a final PX product of 128.2g is obtained when the final SC is operated at $T = -34.1^\circ\text{C}$ and $P = 12.4\text{ Pa}$. Table 11 indicates that, for a 200g liquid feed of 87% PX, 7% MX, 2% EB and 4% OX, a final PX product of 125.5g is obtained when the final SC is operated at $T = -45.5^\circ\text{C}$, and $P = 3.5\text{ Pa}$. The simulation shows that lower temperatures and lower pressures are required in the SC process to obtain a final PX product for a liquid feed with lower composition of PX.

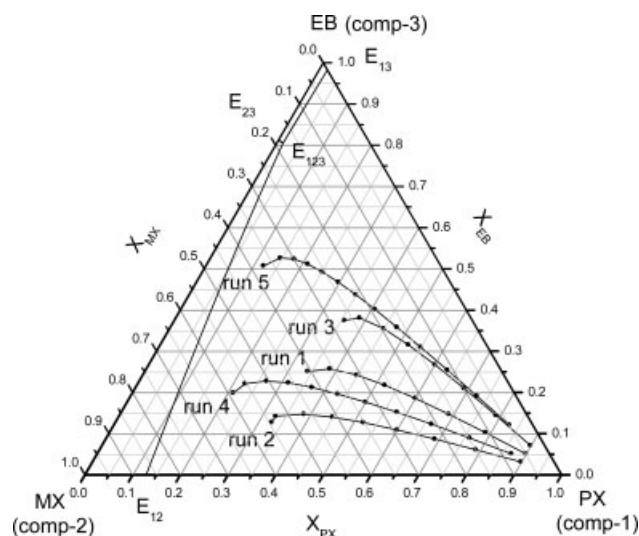


Figure 3. Simulated liquid compositions for the ternary mixture of PX, MX and EB during the SC process (run 1:90% PX, 5% MX and 5% EB; run 2:90% PX, 3% MX and 7% EB; run 3:90% PX, 7% MX and 3% EB; run 4:87% PX, 8% MX and 5% EB; run 5:83% PX, 5% MX and 12% EB).

Table 8. Comparison of the Experimental and Simulation Results for the Ternary System of PX, MX and EB

	Purity of final solid crystal (PX)	Weight of final solid crystal (PX)	Actual recovery rate of PX	Simulated recovery rate of PX
200g feed				
90% PX	99.5% PX	101.2 g	55.9%	72.7%
5% MX				
5% EB				
200g feed				
90% PX	99.9% PX	93.2 g	51.7%	72.4%
7% MX				
3% EB				
200g feed				
90% PX	99.8% PX	97.9 g	54.3%	72.8%
3% MX				
7% EB				
200g feed				
87% PX	99.9% PX	100.1 g	57.5%	73.0%
8% MX				
5% EB				
200g feed				
83% PX	99.6% PX	113.2 g	67.9%	74.3%
5% MX				
12% EB				

$$\text{Actual recovery rate} = \frac{\text{final PX crystals by the DF experiments}}{\text{PX in the feed}}$$

$$\text{Simulated recovery rate} = \frac{\text{final PX crystals by the DF simulation}}{\text{PX in the feed}}$$

The comparison of the experimental and simulation results for various quaternary systems of PX, MX, EB and OX is listed in Table 12. It indicates that, when the experiments are performed based on the SC simulation, the purity of PX crystals can reach 99.7%~99.9% for the quaternary xylene mixture with PX ranging from 87 to 90%. The actual recovery rates range from 48.3% to 51.4%, which are lower than the simulated recovery rates ranging from 71.8% to 72.4%.

Conclusions

In this work, the SC experiments are successfully applied to produce PX crystals from the multicomponent xylene mixture of PX, MX, EB and OX. The SC process is continued until the liquid phase is completely eliminated, and only the pure PX crystals remain in the feed. The low-pressure vapor formed in the process is condensed and collected. The model equations have been derived to simulate the adiabatic SC

Table 9. Simulation Results of Adiabatic SC Operation for the quaternary system of 90% PX, 3.3% MX, 3.4% EB and 3.3% OX ($\Delta T = 5^\circ\text{C}$)

n	T ($^\circ\text{C}$)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}	X _{OX}
0	9.11		200	0	0.9	0.033	0.033	0.034
1	4.11	312.9	54.63	101.5	0.790	0.068	0.064	0.078
2	-0.89	218.1	28.38	118.4	0.691	0.099	0.089	0.121
3	-5.89	149.6	17.47	124.4	0.601	0.126	0.110	0.163
4	-10.9	100.8	11.35	127.1	0.521	0.149	0.125	0.205
5	-15.9	66.7	7.282	128.4	0.449	0.168	0.135	0.248
6	-20.9	43.1	4.278	129.0	0.386	0.180	0.137	0.297
7	-25.9	27.1	1.896	129.2	0.329	0.184	0.127	0.359
8	-30.7	16.7	0	129.2	0.282	0.165	0.098	0.456

Table 10. Simulation Results of Adiabatic SC Operation for the Quaternary System of 88.5% PX, 2% MX, 7% EB and 2.5% OX ($\Delta T = 5^\circ\text{C}$)

n	T ($^\circ\text{C}$)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}	X _{OX}
0	8.46		200	0	0.885	0.02	0.07	0.025
1	3.46	303.6	61.74	96.35	0.776	0.039	0.130	0.054
2	-1.54	212.8	33.01	114.9	0.678	0.057	0.182	0.083
3	-6.54	146.8	20.73	121.9	0.590	0.073	0.225	0.112
4	-11.5	99.3	13.79	125.2	0.511	0.087	0.260	0.141
5	-16.5	66.0	9.189	126.8	0.441	0.100	0.287	0.172
6	-21.5	42.9	5.812	127.7	0.378	0.110	0.303	0.208
7	-26.5	27.2	3.153	128.1	0.323	0.119	0.304	0.254
8	-31.5	16.5	0.960	128.2	0.274	0.122	0.278	0.326
9	-34.1	12.4	0	128.2	0.252	0.110	0.216	0.422

Table 11. Simulation Results of Adiabatic SC Operation for the Quaternary System of 87% PX, 7% MX, 2% EB and 4% OX ($\Delta T = 5^\circ\text{C}$)

n	T ($^\circ\text{C}$)	P (Pa)	L (g)	S (g)	X _{PX}	X _{MX}	X _{EB}	X _{OX}
0	7.80		200	0	0.87	0.07	0.02	0.04
1	2.80	284.2	74.76	87.16	0.763	0.125	0.034	0.078
2	-2.20	197.2	42.99	107.9	0.666	0.174	0.046	0.114
3	-7.20	134.7	28.66	116.4	0.579	0.217	0.056	0.148
4	-12.2	90.4	20.41	120.6	0.502	0.254	0.063	0.181
5	-17.2	59.6	14.93	122.8	0.432	0.285	0.069	0.213
6	-22.2	38.5	10.93	124.1	0.371	0.310	0.073	0.246
7	-27.2	24.4	7.805	124.9	0.316	0.329	0.074	0.280
8	-32.2	15.1	5.238	125.3	0.268	0.340	0.073	0.319
9	-37.2	9.1	3.052	125.4	0.226	0.340	0.068	0.366
10	-42.2	5.2	1.137	125.5	0.190	0.320	0.057	0.433
11	-45.5	3.5	0	125.5	0.168	0.264	0.040	0.528

process, which can provide essential information in the actual SC operation.

In essence, SC is a combination of stripping and crystallization. There are several unique features of the SC operation: (1) SC is conducted under an adiabatic condition at a three-phase equilibrium, where the latent heat released in crystallization is removed by vaporization. Thus, it is an energy-conserving separation method compared to distillation; (2) the addition of solvent is not needed in SC compared to azeotropic or extractive distillation in the separation of the close-boiling mixture. Thus, it is a clean separation technique; (3) SC is continued until the liquid phase is completely eliminated, and only the pure crystals remain in the feed. Compared to crystallization, subsequent filtration or centrifugation is not needed since no mother liquor is present with the pure crystals, and (4) crystal washing is not required since only the

pure crystals remain in the feed and no impurities are adhered on the crystal surfaces at the end of the SC operation.

In addition to the unique features in SC, there is one obvious disadvantage to be overcome: the low-experimental recovery rate as compared with the simulated recovery rate. It can be attributed to the following two reasons: (1) It is difficult to perform the SC experiments under an absolutely adiabatic condition. (2) As SC is operated at low-temperatures and low-pressures, some physical properties of the xylenes in the SC simulation are extrapolated from NIST standard reference database 11,⁷ which leads to the deviations in the calculation of the three-phase equilibrium conditions. Subsequently, the true three-phase equilibrium might not be always attained in the actual SC experiments, resulting in the lower-experimental recovery rate, as compared with the simulated recovery rate. As the improvement of the experimental prod-

Table 12. Comparison of the Experimental and Simulation Results for the Quaternary System of PX, MX, EB and OX

	Purity of final solid crystal (PX)	Weight of final solid crystal (PX)	Actual recovery rate of PX	Simulated recovery rate of PX
200g feed 90% PX 3.3% MX 3.3% EB 3.4% OX	99.7% PX	97.5 g	54.0%	71.8%
200g feed 88.5% PX 2% MX 7% EB 2.5% OX	99.9% PX	91.0 g	51.4%	72.4%
200g feed 87% PX 7% MX 2% EB 4% OX	99.9% PX	84.2 g	48.3%	72.1%

uct recovery is crucial for the SC technique in industrial application, it is the current research task in our laboratory.

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Notation

- $\Delta C_{P,j}$ = change in the heat capacity from solid phase to liquid phase for the i-component
 $\Delta H_{m,j}$ = change in the enthalpy from solid phase to liquid phase for the i-component
 $(H_L)_n$ = enthalpy of the liquid phase in stage n
 $(H_S)_n$ = enthalpy of the solid phase in stage n
 $(H_V)_n$ = enthalpy of the vapor phase in stage n
 L_n = mass flow rate of the liquid phase out of stage n
 P_n = pressure in stage n
 $(P_i^{sat})_n$ = saturated pressure of liquid of the i-component in stage n
 R = ideal gas constant
 S_n = mass flow rate of the solid phase out of stage
 $T_{m,i}$ = melting temperature of the i-component
 T_n = temperature in stage n
 V_n = mass flow rate of the vapor phase out of stage
 $(X_i)_n$ = composition of the i-component of the liquid phase in stage n
 $(Y_i)_n$ = composition of the i-component of the vapor phase in stage n

Greek letters

- $(\gamma_i)_n$ = activity coefficient of the i-component in the liquid phase in stage n

Subscript

- 0 = in the feed
 n = in stage n

Literature Cited

1. Kirk RE, Other DF, Ed. *Encyclopedia of chemical technology*. Vol. 4, New York: John Wiley & Sons, Inc.; 1991.
2. Cheng CY, Cheng SW. Distillative freezing process for separating volatile mixtures and apparatuses for use therein. U.S. Patent, 4218893; 1980.
3. Shiau LD, Wen CC, Lin BS. Separation and purification of p-xylene from the mixture of m-xylene and p-xylene by distillative freezing. *Ind Eng Chem Res*. 2005;44:2258–2265.
4. Shiau LD, Wen CC, Lin BS. Application of distillative freezing in the separation of o-xylene and p-xylene. *AIChE J*. 2006;52:1962–1967.
5. Prausnitz JM, Lichtenthaler RN, de Azevedo EG. *Molecular thermodynamics of Fluid-phase equilibria*. New Jersey: Prentice-Hall, Inc.; 1999.
6. Smith JM, Van Ness HC, Abbott MM. *Introduction to Chemical Engineering Thermodynamics*. Singapore: McGraw-Hill Book Co.; 2001.
7. “NIST standard reference database 11:DIPPR data compilation of pure compound properties.” Version 5.0, sponsored by The Design Institute for Physical Property Data (DIPPR) of the American Institute of Chemical Engineers, copyright by The American Institute of Chemical Engineers; 1985.

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