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Separation of Acetone/Isopropyl Ether/Water Ternary Mixture via Hybrid Azeotropic-Extractive Distillation

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An industrial ternary mixture of acetone/isopropyl ether/water forms two binary azeotropes and a ternary azeotrope at atmospheric pressure and conventional distillation processes cannot achieve efficient separation. A hybrid azeotropic-extractive distillation (HAED) process is proposed for the separation of acetone/isopropyl ether/water. In this process, isopropyl ether is chosen as a light entrainer aimed to form a binary azeotrope with acetone, so that water can be separated from the bottom of azeotropic distillation column (ADC) at 0.3 atm. Then extractive distillation for the separation of binary azeotrope of acetone/isopropyl ether is investigated with ethylene glycol (EG) as an entrainer. On the basis of minimum total annual cost (TAC), a sequential iterative optimization procedure is performed to optimize the economy of two processes with two different feeding ways of light entrainer. The results demonstrate that both processes can make three products purities achieve 99.5 wt% with three columns. Additionally, compared with the process in which light entrainer is blend with the fresh feed, the process in which the light entrainer is added in the upper of the ADC can reduce the TAC and energy consumption by 9.3 % and 9.21 %.

1. Introduction

As important industrial materials, acetone (C_3H_6O), and isopropyl ether ($C_6H_{14}O$) are commonly used as organic solvents in chemical and pharmaceutical industries. The ternary system which contains 64 mol% of acetone, 17 mol% of isopropyl ether and 19 mol% of water needs to be separated and recycled in the industrial production process and the recovery task of acetone and isopropyl ether is to achieve 99.5 wt%. However, due to the complexity of this ternary mixture which includes a ternary azeotrope and two binary azeotropes formed under atmospheric pressure, conventional distillation methods cannot achieve separation efficiently.

In recent years, the separation of ternary mixture has aroused wide attention from scholars. Luyben (2015) discussed the conventional ternary extractive process and the thermally coupled process in which dimethyl formamide (DMF) was used as a heavy solvent to achieve the separation of benzene/cyclohexane/toluene and the dynamic controllability of extractive distillation process was explored. Timoshenko et al (2015) studied the applicability of all types of the VLE diagrams of ternary azeotropic systems via extractive distillation flowsheets with the partially thermally coupled distillation column. Li et al (2016) put forward a new separation process to separate butanol/butyl acetate/methyl isobutyl ketone system. In the separation process, methyl isobutyl ketone was added as a light entrainer in the first column and the top product which was butanol/MIBK was separated by the double effect pressure-swing distillation.

Compared with ternary mixture, there are a large number of literatures covering the separation of binary azeotropic mixture by extractive distillation (Zhu et al., 2016; You et al., 2015), pressure-swing distillation (Liang et al., 2016), and azeotropic distillation (Wu et al., 2014). Extractive distillation is a separation method in which a third component called extractive agent is added to change the relative volatility of the original components, and it has advantages of low energy consumption, easy recycling of the entrainer. A comprehensive methodology including entrainer selection, thermodynamic modelling and process optimization was investigated for the dehydration of TBA and it demonstrated good economy of extractive distillation using ionic liquids as entrainers (Aniya et al., 2016). Qin (2013) established extractive distillation process for

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benzene/cyclohexane separation system with sulfolane as entrainer and two control structures were studied. Munoz et al. (2006) explored the separation of isobutyl alcohol/isobutyl acetate and optimization procedure was conducted to obtain a minimal TAC. Like extractive distillation, azeotropic distillation also needs to introduce a third component which can form a new azeotrope with original components to achieve separation efficiently. Pressure-swing distillation is an efficient method to separate pressure-sensitive azeotropic systems whose azeotropic composition can alter significantly as operating pressure change.
For this paper, a novel method to separate acetone/isopropyl ether/water by hybrid azeotropic-extractive distillation is investigated through the analysis of the theoretical basis.

2. Basis of design and analysis

2.1 Property method

A suitable property method is selected to describe the phase behaviours of acetone/isopropyl ether/water system. NRTL property method is chosen and based on the vapor-liquid equilibrium, the interaction parameters regressed using the “Data Regression” function was regressed to calculate the thermodynamic property of acetone/isopropyl ether system accurately. All NRTL parameters used in this paper are listed in Table 1.

Table 1: NRTL model parameters of acetone/ isopropyl ether/water/ ethylene glycol system

Comp	Acetone/ isopropyl ether	Acetone/ water	Isopropyl ether/ water	Acetone/ ethylene glycol	Isopropyl ether/ ethylene glycol	Water/ ethylene glycol
aij	0	0	0.035	0	0	0.348
aji	0	0	8.021	0	0	-0.057
bij	190.17	317.554	422.978	386.734	1134.980	34.823
bji	123.08	602.558	-766.417	236.848	1022.293	-147.137
cij	0.300	0.534	0.200	0.300	0.300	0.300

2.2 The analysis to residue curve maps

The residue curve maps of acetone/isopropyl ether/water at atmospheric pressure are obtained through Aspen Plus as shown in Figure 1. There is a distillation boundary that divided this system into two distillation regions. In two regions, both isopropyl ether and water are the stable nodes while acetone is the saddle. Besides, there are two binary azeotropes and a ternary azeotrope, and the ternary azeotrope is a minimum azeotropic point. In Figure 1(a), point A is a ternary azeotropic point which formed at 324.16 K with 63.07 mol% of acetone, 30.03 mol% of isopropyl ether, and 6.9 mol% of water. Point B and C are two binary azeotropic points formed at 327.4 K with 73.81 mol% of acetone, 26.19 mol% of isopropyl ether and at 335.36 K with 22.01 mol% of isopropyl ether and 77.99 mol% of water.
The minimum ternary azeotrope could be obtained in the top of the first column while a binary mixture which formed by acetone and water could be obtained in the bottom of the column by using material-balance lines. The existence of the minimum ternary azeotrope could increase difficulty of the separation as well as the recovery. According to changing of operating pressure, we find that the ternary azeotropic point disappeared when the operating pressure is less than 0.5 atm. Generally, the azeotropic temperature will decrease as the operating pressure decreases and it can achieve the purpose of reducing energy consumption. When the operating pressure is set to 0.3 atm, the azeotropic temperature of the acetone/isopropyl ether is 296.14 K and cold water (278 K) still can be used. Two binary azeotropes still exist and there is a negligible change on the azeotropic composition while setting the pressure at 0.3 atm. Thus, pressure-swing distillation is not feasible to separate this ternary system and a third component need to be added to this system aimed at varying the relative volatilities between the components or forming a new azeotrope with original components.
A novel hybrid azeotropic-extractive distillation (HAED) process is investigated to separate this system. Isopropyl ether is added as a light entrainer to form binary azeotrope with acetone. The appropriate dosage of isopropyl ether should be added so that the final feed composition can get to one point of the material balance line BE in Figure 1(b). From that point, the binary azeotrope with 73.81 mol% of acetone, and 26.19 mol% of isopropyl ether could be obtained from the top of the first column. In this way, only the binary azeotrope acetone/isopropyl ether need to be further separated in the next process which can simplify the separation process. Then, the following thing is to determine the proper amount of isopropyl ether added to the first column. According to the analysis of residue curve, point F2 is the final feed composition with 60.58 mol% of acetone, 21.45 mol% of isopropyl ether and 17.97 mol% of water as shown in Figure 1(b).

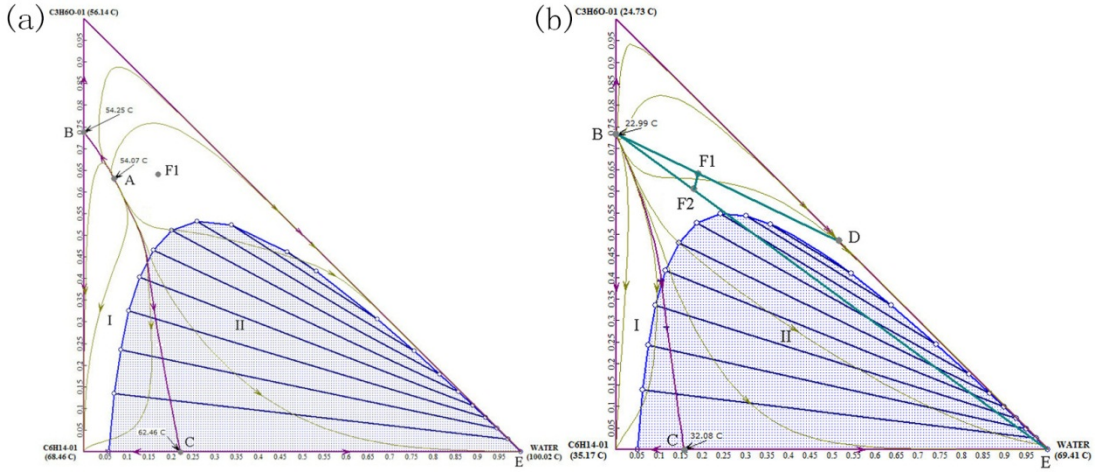

Figure 1: The residue curve maps of acetone/isopropyl ether/water at 1atm (a) and 0.3 atm (b)

3. The design and optimization of HAED process

3.1 The separation of acetone/isopropyl ether

As mentioned above, the next work is to separate the minimum binary azeotrope of acetone/ isopropyl ether after the first column. The azeotropic composition of acetone/ isopropyl ether changes not apparently with pressure, so pressure-swing distillation is not feasible. Extractive distillation is chosen in this paper to separate acetone/isopropyl ether.

The selection of an effective extraction solvent is one of the most critical parts for extractive distillation. In this paper, a preliminary screening is performed and two favourable entrainers including ethylene glycol (EG), propylene glycol are chosen. The effects of EG and propylene glycol on the vapor-liquid equilibrium of the acetone/isopropyl ether system are investigated using the Flash 2 model in Aspen Plus. Figure 2 shows the y-x diagram for acetone/isopropyl ether system when the ratio of the entrainer/(acetone+ isopropyl ether)(E/F) is 1.5: 1. It is observed that two y-x diagrams both are whole above the diagonal and it means both them can break the azeotropic phenomenon of acetone/isopropyl ether. The y-x diagram deviates more from the diagonal when the entrainer is EG which means EG is more effective as compared to propylene glycol for this system. Thus, EG is chosen as the entrainer to separate acetone/isopropyl ether in this paper.

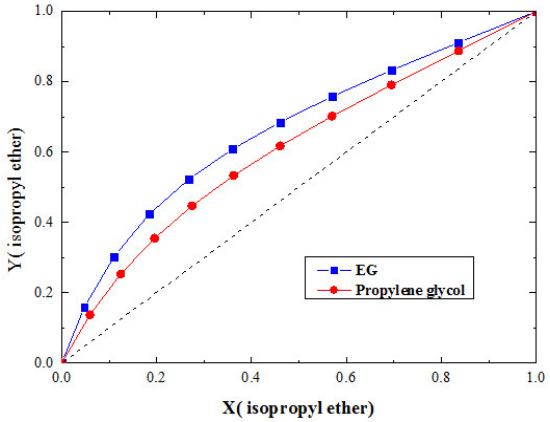

Figure 2: Influence on vapor-liquid equilibrium with different solvents at fixed E/F of 1.5.

3.2 The establishment of process

The conceptual analysis is mentioned in previous section and a specific flowsheet will be proposed in this part. The original design flowsheet includes an azeotropic distillation column (ADC), an extractive distillation column (EDC) and a solvent recovery column (SRC). Two external coolers are set up to keep the light entrainer and EG operating 5 - 15 K below the top temperature of the ADC and EDC. ADC is the low-pressure preliminary separating column with fresh feed which is at 2,000 kg/h with 64 mol% of acetone, 17 mol% of isopropyl ether,

and 19 mol% of water feeding into this column. The appropriate amount of isopropyl ether which is at 200 kg/h is added to form a binary azeotrope of acetone/isopropyl ether. The operating pressure is set at 0.3 atm so that cold water (278 K) can be used. After the preliminary separating of the ADC, high-purity water is obtained from the bottom of the ADC and the binary azeotrope with 73.81 mol% of acetone, 26.19mol% of isopropyl ether is then sent into the EDC in the middle of the EDC while EG is added at the top of the EDC. High-purity isopropyl ether can be obtained from the top of the EDC and acetone goes down with EG. A certain amount of isopropyl ether which is at 200 kg/h distillated from the top of the EDC is recycled to the ADC. Finally, acetone and EG can achieve separation easily in the SRC and this bottom stream which is EG can be recycled back to the EDC. For ADC, EDC and SRC, the initial variables including the number of stages (N_{T1}, N_{T2}, N_{T3}), the feed location (N_{F1}, N_{FE1} N_{F2}, N_{FE2}, N_{F3}), the dosage of entrainer (S) are set up. The “Design Spec/Vary” function is used to achieve the product purity of 99.5 wt% by adjusting the reflux ratio for ADC, EDC and the bottom rate for SRC.

For ADC, the feed location of light entrainer which is isopropyl ether still needs to be explored. As one of the composition of the ternary mixture, it can be blend with the fresh feed to make sure the final feed composition is located at point F1 in figure 1(b). In addition, isopropyl ether can also be added to the ADC at different location with fresh feed. The HAED processes with diverse feed location of isopropyl ether are studied in this work, and the initial flowsheet is shown in Figure 3. The process with two feed locations (1) and (2) are named HAED1 and HAED2, respectively. All of the final design variables mentioned above are determined in the next “Optimization” section with the minimum total annual cost (TAC) as the objective function.

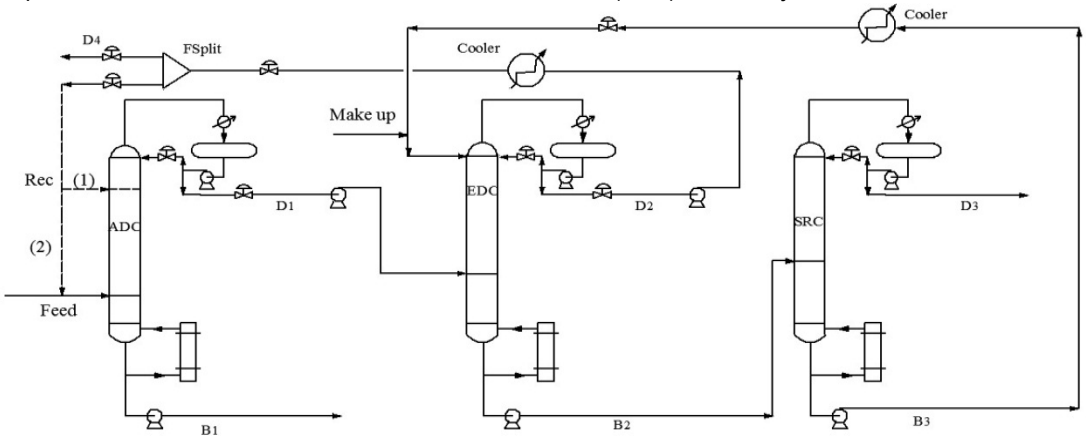

Figure 3: The initial flowsheet of the HAED process with two different feed locations of light entrainer.

3.3 Optimization

In the related studies about the economic optimization of distillation process, TAC was used as the objective function to determine the optimal design (Douglas, 1988). The TAC of the overall system includes annualized capital cost and operating cost. The cost of three columns, three reboilers, three condensers, and two coolers are included in the annualized capital cost. The operating costs consist of the steam for three reboiler, cooling water for three condensers and two coolers. The cost of the vacuum system including the annualized capital cost of a liquid-ring vacuum pump and a mixer can be neglected compared with other main cost in the TAC. The related formula of the annualized capital cost and the operating cost are adopted from Cao (2016). In this paper, a sequential iterative optimization procedure is performed to minimize the TAC of the hybrid azeotropic-extractive distillation processes with two different feed locations of light entrainer. The design variables include: N_{F1}, N_{FE1}, N_{T1}, S, N_{F2}, N_{FE2}, N_{T2}, N_{F3}, N_{T3}. For SRC, it can achieve the separation of the EG-acetone with few total stages and little reflux ratio, thus the N_{T3}, N_{F3} and the reflux ratio are fixed with10, 5, and 0.15. Under fixed triple column pressure, the other design variables of ADC and EDC are determined in turn. The design variables for the other column should remain fixed while optimizing the ADC and the sequential iterative optimization procedures are performed.

At fixed pressure, the overall TAC including all design variables of ADC and EDC in HAED1 process and HAED2 process can be seen in Figures 4. The optimal dosage of entrainer both are 2950 kg/h in two processes. The optimal flowsheets of the HAED process are given in Figure 5 and 6.

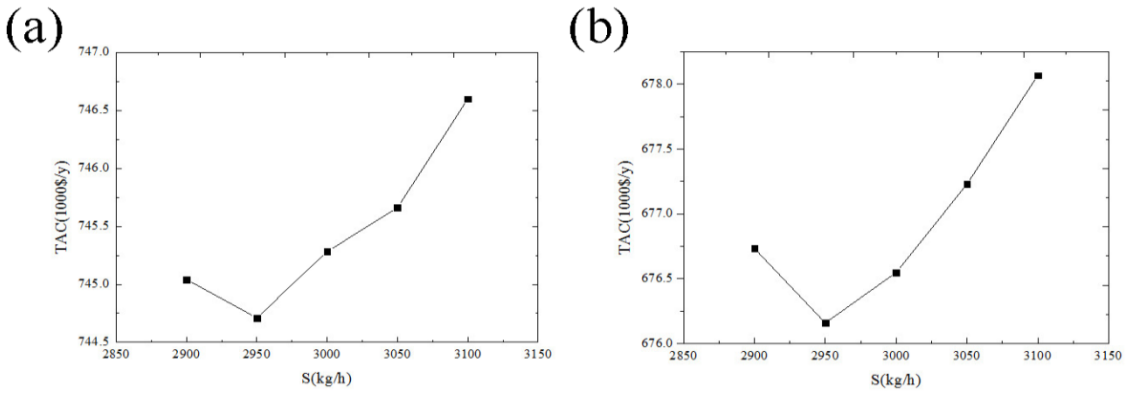

Figure 4: The overall TAC of HAED1 process (a) and HAED2 (b) process.

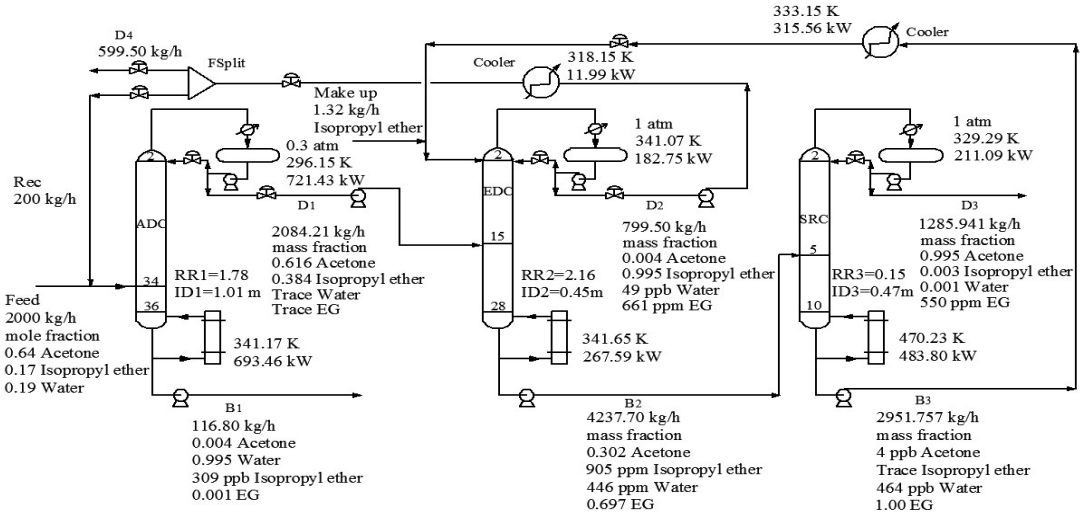

Figure 5: The optimal flowsheet of the HAED 1 process.

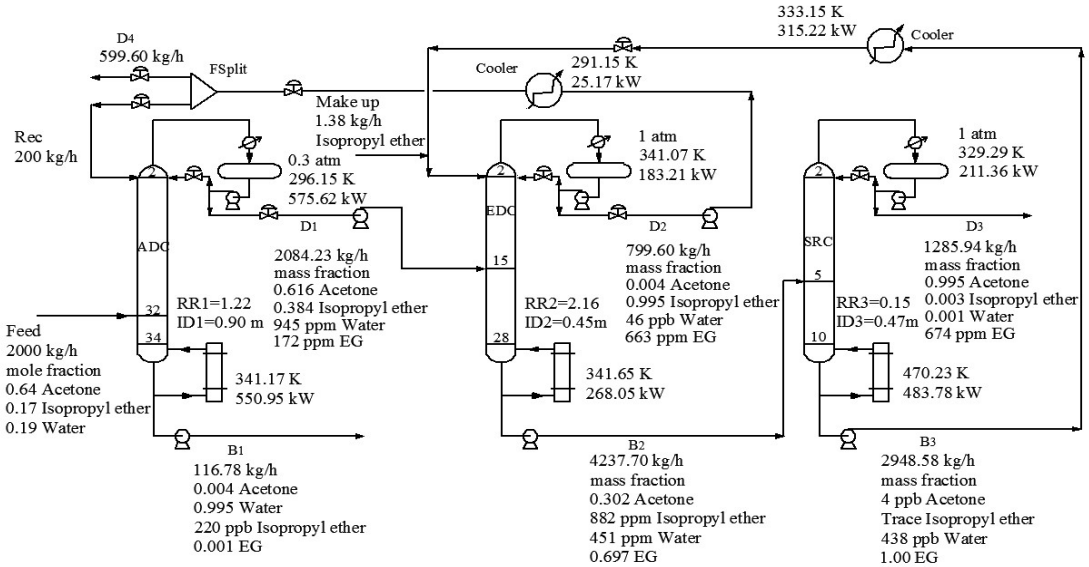

Figure 6: The optimal flowsheet of the HAED2 process.

4. Conclusion

In this paper, a hybrid azeotropic-extractive distillation (HAED) method is investigated to separate a ternary mixture of acetone/isopropyl ether/water. Isopropyl ether is chosen as light entrainer to form a binary minimum azeotrope with acetone. In this way, a ternary mixture can be transformed into a binary mixture of acetone/isopropyl ether only after the azeotropic distillation column (ADC). Then, the acetone/isopropyl ether is separated by extractive distillation and ethylene glycol (EG) is selected as the entrainer. Based on the minimum total annual cost (TAC), operating parameters of ADC and extractive distillation column (EDC) are optimized by adjusting the reflux ratio in the condition of product purity requirement. The process that isopropyl ether is added in the upper of the ADC is more energy-saving and the TAC and energy consumption can reduce 9.3% and 9.21% compared with the process which isopropyl ether is mixed with the fresh feed. A hybrid distillation method is worth considering for the separation of complex ternary mixture.

Acknowledgments

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