[Research Note]

Operability of the Modified Petlyuk Process for Separation of Benzene, Toluene, and *p*-Xylene Ternary MixtureTakehiro YAMAKI^{†1)*}, Ryosuke AKIMOTO^{†2)}, Kei TAKANE^{†2)}, and Keigo MATSUDA^{†2), †3)}^{†1)} Research Institute for Chemical Process Technology, National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, JAPAN^{†2)} Dept. of Chemistry and Chemical Engineering, Graduate School of Science and Engineering, Yamagata University, 4-3-16 Jonan, Yonezawa, Yamagata 992-8510, JAPAN^{†3)} Renewable Energy Research Center, National Institute of Advanced Industrial Science and Technology (AIST), 2-2-9 Machiikedai, Koriyama, Fukushima 963-0298, JAPAN

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A modified Petlyuk process with a reboiler at the bottom of the prefractionator eliminates the need for a vapor interconnection between the main column and the prefractionator and thus improves operability. However, the effect of this configuration on the operability has not been clarified. The present study examined the operability of a modified Petlyuk process with a reboiler at the bottom of the prefractionator for the separation of benzene, toluene, and *p*-xylene ternary mixture. The effect of the bottom-out to feed ratio of the prefractionator on the separation characteristics of the modified Petlyuk process was evaluated for different feed flow rates. The operation range of the modified Petlyuk process was clarified by presenting the conditions satisfying the product specifications as a contour plot for the feed flow rate and the bottom-out to feed ratio of the prefractionator. The operability of the modified Petlyuk process was compared with that of the original Petlyuk process. Based on the obtained results, a dynamic simulation verified that the product specifications can be satisfied simply by adjusting the bottom-out to feed ratio of the prefractionator for a disturbance of the feed flow rate.

Keywords

Distillation, Petlyuk process, Modified Petlyuk process, Dynamics, Operability

1. Introduction

Distillation is one of the main separation methods used in the chemical industry, as well as petroleum refining and petroleum chemistry. However, distillation consumes large amounts of energy, so to reduce the environmental load of the process, energy savings are required in the distillation process. Several possible methods of realizing such energy savings have been proposed^{1),2)}.

The Petlyuk process is one such energy-saving distillation process, which features two thermally coupled distillation columns^{3),4)}, a prefractionator and a main column. Products can be recovered from the distillate, side-cut, and bottom-out streams. Therefore, the Petlyuk process can separate a ternary mixture. The prefractionator has no condenser or reboiler, as the liquid and vapor in the prefractionator are fed into the main column. Since the prefractionator and the main column are thermally coupled, overall energy consump-

tion is reduced. A practical application of the Petlyuk process uses a dividing-wall column^{5),6)}, which achieves the same level of performance as the Petlyuk process by dividing the distillation column into two with a partition plate. However, the flow rates of the vapor and liquid into the prefractionator are determined by the position of the partition plate, so a major disadvantage of this design is that the ratio of the vapor and liquid flow rates cannot be adjusted after construction of the column. This issue also affects the Petlyuk process.

A modified Petlyuk process would allow for the adjustment of the vapor or liquid flow rates to the prefractionator. The separation characteristics of a modified Petlyuk process were studied with either a condenser at the top or a reboiler at the bottom of the prefractionator⁷⁾. The condenser at the top of the prefractionator allows liquid reflux operation, so no liquid interconnection between the main column and the prefractionator is necessary. The reboiler at the bottom of the prefractionator supports vapor feed, so no vapor interconnection between the main column and the prefractionator is necessary. Examination of the steady-state condition found that the operability of the modified Petlyuk process was improved by incorporat-

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ing a condenser or reboiler into the prefractionator. Investigation of six types of modified Petlyuk process concluded that the appropriate configuration should be selected according to the relative volatilities and feed composition⁸⁾. The control properties can be improved by reducing the interconnection steam number⁹⁾. Optimization of the liquid split ratio and bottom-out to feed ratio of the prefractionator could maximize the separation performance of a modified Petlyuk process with a reboiler at the bottom¹⁰⁾. Furthermore, the separation characteristics of the modified Petlyuk process were examined by determining the mole fraction of the target component in the side-cut stream, together with the liquid split ratio and the bottom-out to feed ratio, on a contour plot.

Optimization of the design and operating conditions of the modified Petlyuk process has been undertaken, but the operability of the modified Petlyuk process from the viewpoint of process dynamics has not been studied. Product specifications may be maintained in a modified Petlyuk process with a condenser or reboiler in the prefractionator by appropriately adjusting the operation of the condenser or reboiler as a disturbance occurs. Therefore, the operation range of the modified Petlyuk process may be greater than that of the Petlyuk process.

This study focused on the contour plot produced to clarify the operation range¹⁰⁾. The present study adopted the concept of the contour plot but changed the axis variables to the disturbance and operating variables of the prefractionator. The output displayed on the contour plot shows the conditions that satisfy the product specifications. Revision of the diagram should show the operation range of the modified Petlyuk process. The operation range of a modified Petlyuk process with a reboiler at the bottom of the prefractionator, as applied to the separation of a benzene, toluene, and *p*-xylene ternary mixture, was examined as a case study. The bottom-out to feed ratio was employed as an operation parameter of the prefractionator. First, the influence of the feed flow rate and the bottom-out to feed ratio (operating variable of the prefractionator) were evaluated on the separation characteristics of the modified Petlyuk process. Based on these simulation results, a diagram of the operation range of the modified Petlyuk process was created and compared with that of the Petlyuk process. Then, a dynamic simulation was used to examine whether the modified Petlyuk process can attain the conditions needed to satisfy the product specifications by adjusting the bottom-out to feed ratio of the prefractionator as the feed flow rate changes as the result of a disturbance.

2. Simulation Model

The present study investigated the separation of benzene, toluene and *p*-xylene ternary mixture using

Fig. 1 Schematic Diagrams of the a) Petlyuk Process, b) Modified Petlyuk Process

feed flow rate in the base case of 5 mol/s, and mole fractions of benzene, toluene, and *p*-xylene of 0.2, 0.6, and 0.2, respectively¹⁰⁾. The product specifications assumed that benzene, toluene, and *p*-xylene were all 99 mol% or more. The thermodynamic properties of the ternary mixture were estimated using the non-random two-liquid (NRTL) model¹¹⁾.

Figure 1 shows schematic diagrams of both the Petlyuk and modified Petlyuk processes. The Petlyuk process consists of a prefractionator and a main column. The overhead vapor and the bottom-out liquid from the prefractionator are fed to the main column. In the main column, benzene, toluene, and *p*-xylene are recovered from the distillate, side-cut, and bottom-out streams, respectively. Some of the liquid and vapor is fed from the main column to the prefractionator. The modified Petlyuk process consists of a prefractionator with a reboiler at the bottom and a main column. The presence of the reboiler prevents vapor from the main column passing to the prefractionator. In the main column, benzene, toluene, and *p*-xylene are recovered from the distillate, side-cut, and bottom-out streams, respectively.

Table 1 lists the design and operating conditions for both the Petlyuk and modified Petlyuk processes. The total number of stages was set to 64 for both processes. The feed locations for the ternary mixture, overhead vapor, and bottom-out liquid from the prefractionator, the liquid and/or vapor from the main column, and the reflux ratio and the reboiler duty for the main column were as previously specified¹⁰⁾.

Figure 2 shows schematics of both the Petlyuk and modified Petlyuk processes, together with the controllers required for the dynamic simulation. A pressure controller was installed at the top of the main column for both dynamic simulation models. The pressure at the top of the main column was adjusted according to the condenser duty because this was the simplest variable to manipulate. Liquid level controllers were

Table 1 Input Design and Operating Conditions

		Petlyuk process		Modified Petlyuk process	
		Prefractionator	Main	Prefractionator	Main
Design conditions					
Number of stages		20	44	20	44
Feed location	Stage	10th from the bottom	-	10th from the bottom	-
Operating conditions					
Pressure at top	[kPa]	108.5	101.3	108.5	101.3
Pressure drop in stage	[kPa]	0.5	0.6	0.5	0.6
Distillate flow rate	[mol/s]	-	1	-	1
Side-cut flow rate	[mol/s]	-	3	-	3
Bottom-out flow rate	[mol/s]	vary	1	vary	1
Reboiler duty	[kJ/s]	-	308.2	vary	327.5

Fig. 2 Schematic Diagrams of the a) Petlyuk Process, b) Modified Petlyuk Process, Both Controllers

installed in both the reflux drum and the reboiler of the main column. The liquid level in the reflux drum was adjusted according to the distillate flow rate in the main column as this was the simplest variable to manipulate. The liquid level in the reboiler was adjusted according to the bottom-out flow rate of the main column as, again, this was the simplest variable to manipulate.

In the modified Petlyuk process, a liquid level controller was installed in the reboiler of the prefractionator. The liquid level in the reboiler of the prefractionator was adjusted according to the bottom-out flow rate of the prefractionator as this was the simplest variable to manipulate. The gain and integral time of the pressure controller were 20 %/% and 12 min, respectively. The gain and integral time of the liquid level controller were 10 %/% and 60 min, respectively. The gain and integral time used the default values provided by the manufacturer (Aspen Plus Dynamics). The side-cut flow rate was set to 0.6 times the feed flow rate, using a flow rate controller. In the modified Petlyuk column, the bottom-out to feed ratio at the prefractionator was adjusted using a flow rate controller. The gain and integral time of the flow rate controller were 0.5 %/% and 0.3 min, respectively. The conventional flow rate controller tuning was adopted as previously reported¹²⁾.

Fig. 3 Effect of Feed Flow Rate on Product Mole Fraction of the Petlyuk Process

The tray sizes of the prefractionator and the main column were calculated using the manufacturer's functions. The sizes of the reflux drum and the reboiler were determined using the previous method¹²⁾. All steady state and dynamic simulations were performed using Aspen Plus and Aspen Plus Dynamics software (V8.0).

3. Simulation Results and Discussion

3.1. Steady State Performance

To clarify the operation range of the modified Petlyuk process, the separation characteristics of the Petlyuk and modified Petlyuk processes were examined. Here, the reflux ratio and the reboiler duty were simulated as constants. Additionally, for the Petlyuk process, the liquid and vapor split ratios were set to 0.306 and 0.554, respectively, to satisfy the product specifications. The liquid and vapor split ratios were also calculated as constants. On the other hand, for the modified Petlyuk process, the liquid split ratio was set to 0.257, and the bottom-out to feed ratio was variable.

Figure 3 shows the mole fraction of benzene in the

Fig. 4 Operation Range of the Modified Petlyuk Process Required to Satisfy the Product Specifications

distillate stream, toluene in the side-cut stream, and *p*-xylene in the bottom-out stream with the feed flow rate between 4.5 mol/s and 5.5 mol/s in the Petlyuk process. Using a feed flow rate of less than 5 mol/s, the mole fraction of the *p*-xylene in the bottom-out stream was increased, whereas the mole fractions of benzene in the distillate stream and toluene in the side-cut stream were decreased, because the vapor generated by the reboiler in the main column was increased and the boilup ratio was higher. The *p*-xylene reached the side-cut stage, whereas the toluene shifted to the top of the main column. Using a feed flow rate of more than 5 mol/s, the mole fraction of benzene in the distillate stream was increased, whereas the mole fractions of toluene in the side-cut stream and *p*-xylene in the side-cut stream were decreased, as the amount of vapor generated by the reboiler in the main column was decreased and the boilup ratio was lower. The benzene reached the side-cut stage, whereas the toluene shifted to the bottom of the main column. Therefore, the Petlyuk process could not satisfy the product specifications as the feed flow rate changed under the constant reflux ratio, with constant reboiler duty, and constant vapor and liquid-split ratios.

Figure 4 shows the effects of the feed flow rate and the bottom-out to feed ratio of the prefractionator on the mole fraction of the toluene in the side-cut stream. In the modified Petlyuk process, the reboiler of the prefractionator can be adjusted, so the bottom-out to feed ratio can also be varied. Adjusting the bottom-out to feed ratio showed that the product specifications could be satisfied within the region surrounded by the solid line in Fig. 4. Using a feed flow rate of 5.0 mol/s, operation at the bottom-out to feed ratio between 0.58 and 0.59 can satisfy the product specifications. The bottom-out to feed ratio should be increased as the feed

flow rate decreases, and decreased as the feed flow rate increases. The bottom-out to feed ratio is increased as the feed flow rate decreases to prevent a decrease in the liquid flow rate to the main column. On the other hand, the bottom-out to feed ratio is decreased as the feed flow rate increases to compensate for the low vapor flow rate from the reboiler in the main column. In addition, at least 99 mol% of the toluene in the side-cut stream can be recovered within the region surrounded by the dashed line, although the product specifications cannot be satisfied.

The simulation results in Figs. 3 and 4 clearly show that the operation range of the modified Petlyuk process is wider than that of the Petlyuk process. The simulation also showed the modified Petlyuk process can be used to achieve the product specifications by adjusting the bottom-out to feed ratio of the prefractionator with the feed flow rate from 4.75 to 5.5 mol/s.

3.2. Dynamic Performance

The steady-state simulation showed that the modified Petlyuk process can be used to satisfy the product specifications by adjusting the bottom-out to feed ratio of the prefractionator as the feed flow rate changes. Therefore, the modified Petlyuk process can attain a state in which the product specifications can be satisfied, simply by adjusting the bottom-out to feed ratio of the prefractionator using a dynamic simulation.

Figure 5 shows the transient responses of the Petlyuk process that occur with change in the feed flow rate of 5 % or 10 %. The feed flow rate was step changed as a disturbance, after 1 h from the start of the dynamic simulation. As the feed flow rate decreased, the mole fraction of *p*-xylene in the bottom-out stream of the main column increased, whereas the mole fractions of benzene in the distillate stream and toluene in the side-cut stream decreased as shown in Figs. 5a), and 5b). The mole fraction of each stream settled about 8 h after the start of the simulation. As the feed flow rate increased, the mole fraction of benzene in the distillate stream increased, whereas the mole fractions of toluene in the side-cut stream and *p*-xylene in the bottom-out stream of the main column decreased as shown in Figs. 5c), and 5d). The mole fraction of each stream settled about 5 h after the start of the simulation. The mole fraction of each stream, once settled, was almost the same as the value shown in Fig. 3.

Figure 6 shows the transient responses of the modified Petlyuk process to changes in the feed flow rate of 5 % and 10 %. As the feed flow rate changed from 5.0 to 4.5 mol/s, the mole fraction of the *p*-xylene in the bottom-out stream of the main column increased, whereas the mole fractions of benzene in the distillate stream and toluene in the side-cut stream decreased. The same behavior was observed in the modified Petlyuk process, if the bottom-out to feed ratio of the prefractionator remained constant, as in the Petlyuk

Fig. 5 Transient Responses of the Petlyuk Process in the Event of Stepwise Change in Feed Flow Rate: a) -10% , b) -5% , c) $+5\%$, d) $+10\%$

process. The bottom-out to feed ratio in the prefractionator was changed from 0.58 to 0.697 after 1 h from the start of the dynamic simulation to satisfy the product specification. The bottom-out to feed ratio of the prefractionator was set to 0.697, required to attain a toluene mole fraction in the side-cut stream of greater than 99 mol%, as shown in **Fig. 4**. By adjusting the bottom-out to feed ratio of the prefractionator to 0.697, the mole fraction of toluene in the side-cut stream increased to 99 mol%. Then, as the feed flow rate was changed from 5.0 to 4.75 mol/s, the mole fraction of the *p*-xylene in the bottom-out stream of the main column increased, whereas the mole fractions of the benzene in the distillate stream and the toluene in the side-cut stream decreased. With a feed flow rate of 4.75 mol/s, the product specifications could be satisfied by adjusting the bottom-out to feed ratio of the prefractionator to 0.664 as shown in **Fig. 4**. Therefore, the bottom-out to feed ratio of the prefractionator was changed from 0.58 to 0.644, 10 h after the start of the simulation. As a result, the mole fraction of benzene in the distillate stream and toluene in the side-cut stream increased, so that the product specification was satisfied. As the feed flow rate increased from 5.0 to 5.25 mol/s, the mole fraction of benzene in the distillate stream increased, whereas the mole fractions of toluene in the side-cut stream and *p*-xylene in the bottom-out stream in the main column decreased. This behavior is the same as that observed as the feed flow rate increases in the Petlyuk process. For a feed flow rate of 5.25 mol/s, the product specifications could be satisfied by adjusting the bottom-out to feed ratio of the prefractionator to 0.535 as shown in **Fig. 4**. The bottom-out to feed

ratio of the prefractionator changed from 0.58 to 0.535, 10 h after the start of the simulation. The mole fraction of toluene in the side-cut stream and *p*-xylene in the bottom-out stream in the main column increased, so that the product specification was satisfied. Also, as the feed flow rate was increased from 5.0 to 5.5 mol/s, the product specification could be satisfied by adjusting the bottom-out to feed ratio of the prefractionator to 0.485. These simulation results clearly showed that the product specification could also be satisfied in a dynamic simulation by adjusting the bottom-out to feed ratio of the prefractionator according to the disturbance in the feed flow rate. Therefore, presentation of the separation characteristics of the modified Petlyuk process as a contour plot clarified the operation range of the modified Petlyuk process, and appropriate operating conditions for the bottom-out to feed ratio of the prefractionator corresponding to the disturbance could be identified.

4. Conclusion

This study examined the operation range of the modified Petlyuk process with a reboiler at the bottom of the prefractionator, using the separation of a ternary mixture of benzene, toluene, and *p*-xylene. The effects of the feed flow rate (disturbance) and the bottom-out to feed ratio of the prefractionator (operating variable) on the separation performances of the Petlyuk and modified Petlyuk processes were evaluated using steady-state simulation. Under the conditions of constant reflux ratio and reboiler duty in the main column, the Petlyuk process could not satisfy the product specifica-

Fig. 6 Transient Responses of the Modified Petlyuk Process in the Event of Stepwise Change in Feed Flow Rate and Adjustment of Bottom-out to Feed Ratio

tions as the feed flow rate changed, whereas the modified Petlyuk process could satisfy the product specifications with a feed flow rate of between 4.75 mol/s and 5.5 mol/s by adjusting the bottom-out to feed ratio in the prefractionator. The operation range of the modified Petlyuk process was clarified by indicating the range satisfying the product specifications as contour lines for the feed flow rate and the bottom-out to feed ratio of the prefractionator. Dynamic simulation

showed that the modified Petlyuk process reached a state that satisfied the product specifications by adjusting the bottom-out to feed ratio, even if the feed flow rate changed as a result of a disturbance.

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要 旨

ベンゼン／トルエン／*p*-キシレン混合物の分離を対象とした改良型 Petlyuk プロセスの操作性

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予備蒸留塔の塔底にリボイラを備えた改良型 Petlyuk プロセスは、主塔からの予備蒸留塔への蒸気供給が不要となり操作性が向上することが知られている。しかしながら、予備蒸留塔の塔底のリボイラ操作が操作範囲に及ぼす影響は明らかになっていない。本研究では、ベンゼン-トルエン-*p*-キシレンの分離を対象に、予備蒸留塔の塔底にリボイラを備えた改良型 Petlyuk プロセスの操作範囲を明らかにする。外乱を想定しフィード流量が異なる条件において、予備蒸留塔の缶出液とフィードの流

量比が改良型 Petlyuk プロセスの分離特性に及ぼす影響を評価した。フィード流量と予備蒸留塔の缶出液とフィードの流量比に対して、製品規格を満たす範囲を等高線図として示すことで、操作範囲を明らかにした。操作範囲の検討結果に基づき、外乱としてフィード流量が変化した場合、缶出液とフィード流量比の調整のみで製品規格を満たせることを非定常シミュレーションにおいても検証した。

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