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Research Article

Optimal Cyclic Reflux Operational Policy for Ternary Mixtures Separation

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Abstract: The optimal control of a multicomponent batch distillation campaign with a variable reflux ratio, and an analysis of the influence of different parameters, have been presented. At first, the influence of initial mixture composition combined with the influence of the top pressure brought about: 1) a clear 'bang-bang' policy, which is easier to verify and apply in an industrial environment, and 2) time reduction by 14.07%. Furthermore, the influence of vapor boil-up showed that with the increase of vapor boil-up by 2.73 times, there is a significant increase in recovery rate by 27.55%, whilst a significant reduction of overall time horizon by 41.71% for total recovery. Finally, the influence of reboiler volume: with the doubling of reboiler volume, again, there is a significant increase in recovery rate by 32.98%, and, interestingly, the overall time horizon reduction is almost the same as in the previous case, as it gives 41.90% for total recovery. These results give flexibility in terms of process design: one can choose between different variable parameters to achieve the targeted recovery rate and/or final time.

***Keywords*:** multicomponent distillation, optimal control, direct method, Pontraygin's maximum principle

1. Introduction

The purpose of this paper is to extend the research given in an earlier paper,[1] from the binary to the ternary mixture case. From the aspect of the optimal control of the operation, the separation of a ternary mixture of hydrocarbons, namely cyclohexane-heptane-toluene, has been researched in depth by many authors.[2-9] For a binary mixture of cyclohexane-toluene, Mujtaba and Miladi[10] examined the influence of the total number of trays. In general, the increased number of trays reduced the total batch time and reflux ratio. For a specific case, they found that increasing the total number of trays six times decreased the reflux ratio by 13.85% and the total batch time by 45.63%. Aqar et al.[11] combined separation and reaction into a single configuration to show that the optimal condenser vapor flowrate and feed rate should be gradually increased whilst both the reflux ratio and the total batch time are to be reduced to increase the number of batches of product meeting the increasing product demand. Marquez et al.[12] succeeded in applying their findings on batch reactor to the reactive batch column: a linear parameter-varying model was embedded in the model learning predictive controller, with the outcome of three application-dependent strategies to calculate both the disturbances and parameters of the model.

Ferchichi et al.[13] for a ternary mixture of hydrocarbons, compared two different semi-batch modes to the batch mode: 1) direct feeding to the reboiler (Semi-Batch Distillation (SBD_1)) resulted in a 31.93% reduction in time, and

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required 2.14 times less specific energy demand; and 2) namely, SBD_2, feeding into the column at a fixed stage, resulted in the same time reduction as SBD_1 and somewhat better specific energy demand, i.e., required 2.49 times less. Ferchichi et al.[14] introduced the "varied feed location semi-batch mode," namely SBD_3. In contrast, the stages were divided into two sections: namely, feed plates being lowest as $SBD_{3,A}$ brought in, in almost all time intervals, more energy-intensive operation—for example, in the second time interval, even 46.44% compared to $SBD_{3,B}$, namely, feed plates of the middle section. Vibhute and Jogwar[15] first generated the best open-loop policy to use this trajectory in subsequent closed-loop studies, where the extended Kalman filter is used as one of the most favored estimators to provide reliable estimates in the presence of plant-model mismatch and process measurement noise. For the open-loop policy, it was the amount of product collected per unit energy consumption factor that gave significant production maximization, i.e., 83%, along with significant energy savings, i.e., 97%, for all purity specifications. Vibhute and Jogwar[16] reworked the study for the same vapor-recompressed batch column, this time under total reflux to obtain both products in high purity: the open-loop trajectory is subsequently traced by maximizing some of the key indices as previously, in the closed-loop trajectory using a model predictive controller. Interestingly, as in the previous study, the authors did a test with a disturbance after 100 min, consequently, the tray efficiency dropped by 5%, the total time increased by 13.84%, and the overall performance factor dropped by 14.23%.

Radhika et al.[17] presented a vapor recompression scheme for the separation of a ternary zeotropic mixture of alcohols, where the driving force (temperature difference) is maintained by a variable-speed compressor with an estimated compression ratio. For the open-loop operation with a single stage (one compressor), they perceived that throughout 30% of the operation, the reflux flowrate profile remained almost constant at the maximum value; then it dropped almost 67%, afterward instantaneously rose by more than 30%, remained constant for the next one-tenth of the time, and then almost linearly dropped to the same value previously achieved, remaining constant till the end of the process. Also, it requires more than twenty compression ratios to compress the vapor completely, whereas for double-stage operation, the compression ratio is divided equally between both compressors; however, it brings only 1.2% energy savings compared to the single-stage. Rihm et al.[18] used a first-principles model, i.e., a Radfrac column of ten stages with a full set of mass and energy balances, along with a pressure-driven model, to show that if not enough data from the real plant is available, high-accuracy process simulators can be used to parametrize data-based models. Niazi et al.[19] showed that for the dehydration of methyl isobutyl ketone, a simple distillation unit operation with no reflux is better to use than the batch distillation unit with a decanter: if the total return fraction (methyl isobutyl ketone and water) is higher than 0.7, then it is proven to give both better energy efficiency and production rate. On the other hand, Niazi et al.[20] compared an inverted batch distillation unit with a decanter to a conventional batch distillation unit to conclude that it achieves 7%/9%/17% in fractional recovery/time reduction/energy savings, respectively, due to gradual feeding that provides a shorter pre-heating step and top feeding that assumes direct contact between vapor and liquid phases.

Liu et al.[21] introduced an improved white shark optimizer in their study to investigate the four-component batch distillation process: the algorithm searches for prey at random positions, and its movement update behavior towards the prey is described with a specific equation enhanced by parameters describing a stepwise acceleration search method.

In this study, the optimal control for the operation of multicomponent batch distillation for a ternary mixture of hydrocarbons (i.e., cyclohexane-heptane-toluene) was investigated in an open-mode double rectifier configuration. In order to obtain a clear "bang-bang" policy solution that could be verified and possibly applied in industry, the total pressure at the top of the column was reduced by 5%, and the initial mixture composition was set so that the content of the least volatile component was increased. This already led to a reduction in the total number of batch cycles and a time reduction of 14.07% for total recovery. Further, the investigation of the influence of vapor boil-up was done to show that for an increase in vapor boil-up by 2.73, there is a significant reduction in total batch processing time by 27.55%. Moreover, it gives total recovery in the overall time horizon, reduced by 41.71%. Also, it was discovered that there is an influence on the structure of the optimal control pattern, whereas, by rule, with the increase of vapor boil-up, the total number of cycles increases linearly. Secondly, the investigation of the influence of reboiler volume was done: with a doubling of reboiler volume, there is again an increase in recovery rate by 32.98% over a fixed time horizon, whereas the total recovery is achieved with a time reduction of 41.90%. Again, there is an influence on the optimal control pattern, as the total number of cycles doubled as well. Finally, the dual-process optimal control strategy was verified by the ChemCad dynamic simulator: from here, the extracted values for reboiler duty were presented by creating a "discontinuous heating energy curve." For the first stage of the dual process, it seems there exists a characteristic sequence after 75% of the step is completed, which can be described as a "quasi-zero-bang-zero-bang-bang" sequence. For the second part of the process, the definition seems to be even clearer and dominated over all time horizons as a "quasi-zero-bang-bang-zero-bang-bang" sequence.

2. Problem formulation

In this study, it is assumed that distillation is carried out with only one rectifier column with a reboiler of maximum capacity (U_N) and a given volume (10 mol) of a mixture of cyclohexane-(n-heptane)-toluene being treated. The total number of stages chosen is the minimal number sufficient for separation (7 stages), and, like the volume of the reboiler, is not a control variable. The purity specified for all the main cuts is 0.95; therefore, every step of the multicomponent batch distillation is finished when the most volatile component reaches its specified purity in the accumulator and/or reboiler. The campaign contains two distillations in total, i.e., the least volatile component is obtained in the reboiler after the second distillation. In contrast, the first/second, respectively, component is obtained in the first/second accumulator. The policy used is the optimal cyclic reflux policy obtained for a variable reflux ratio in the earlier paper.[1]

The chosen model of multicomponent batch distillation is based on the following assumptions: 1) equimolar overflow, 2) total condensation, 3) vapor hold-up dynamics are neglected, 4) pressure drop along the columns is neglected, and 5) ideal mixing of the vapor and the liquid phase. The detailed mathematical model developed by Stojkovic et al.,[1] containing mass balances on all parts of the batch column (Mathematical Equation System (MES)), was extended from a binary mixture to the case of a ternary mixture, i.e., the total number of components is equal to three. The control variables correspond to those described in an earlier paper by Stojkovic et al.[1] In Figure 1, a scheme for ternary mixture separation is depicted.

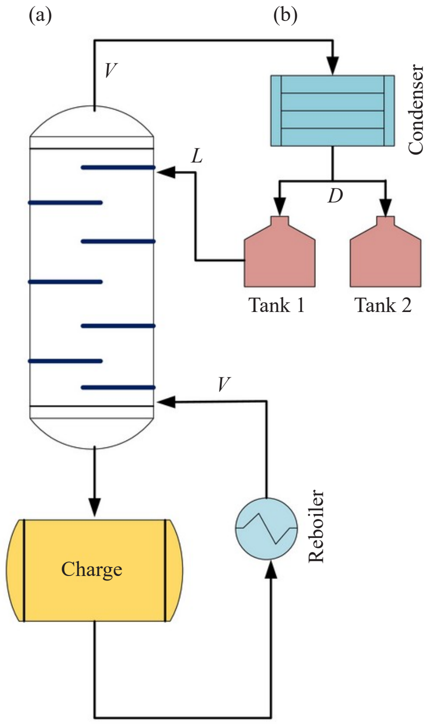

Figure 1. A scheme for ternary mixture separation: batch rectifier (a) left: double batch rectifier, (b) right: double batch inverse column

The retained objective is the distillate rate, defined as the total amount of the main cuts produced during the total time of the (N_{batch} = 2) distillations of the operation. The optimal control problem for N_{batch} distillations is defined as:

$$\max_{R, t_f} \sum_{i=1}^{i=N_{batch}} \sum_{i=1}^{i=N_C} D_{i,j}$$

Where $D_{i,j}$ is the distillation rate for the main cut of the *jth* component at batch *i* and t_f is the total time for batch *i*.

The optimal control problem can be described as follows: for a given multicomponent batch distillation configuration (*N* - total number of trays, *P* - working pressure), batch composition, the distillation task, and overall time horizon (t_f), determine the optimal reflux ratio so as to maximize the distillate at every step, subject to any constraints (model equations, bounds on the optimization variables). The problem above is a nonlinear programming problem, and, it is solved by the Bocop solver using the direct method with a choice of discretization schemes for dynamics discretization, so to convert the problem into a finite-dimensional.[22]

3. Numerical results and discussion

The working conditions and predefined parameters are tabulated in Table 1, with concentrations expressed starting from the most volatile component, cyclohexane, and the separation being done in a double batch rectifier.

Table 1. Working conditions

Parameter	P (atm)	U_N^0 (l)	U_i^0 (l)	V (mol/h)	t_f (h)	x_N^0	y^*
Predefined/ initial value	1	10	0.1	11	1. 0.8 2. 1.6	0.1	0.95

** 1. t_f = 0.8 h, 2. t_f =1.6 h

Firstly, the operation is managed under the standard atmospheric pressure (760 mm Hg), to distill cyclohexane (95% left in the accumulator). To reach the satisfactory recovery rate of 98.30%, a step must be repeated. Still, the recovery rate is almost 10% higher than what was reported by Bonny[9] for a particular configuration and a varying reflux policy. The total duration proposed for the best performance of the first step of the process is t_f = 2.008 (h). The optimal control policies for the zeotropic mixture of cyclohexane-(n-heptane)-toluene are depicted in Figure 2a, whereas the optimal costs and recovery rates are tabulated in Table 2.

Table 2. The optimal costs and the recovery rates

Product recovered	N	U_0 (t_f)	Recovery rate (%)	Discr. scheme, nb. of points
Cyclohexane	7	1.4376	1. 49.19% 2. 98.30%	Gauss, 2,000
n-heptane	24	2.6251	94.92%	Gauss, 1,600

** 1. t_f = 0.8 h, 2. t_f =1.6 h

For the first step, i.e., cyclohexane distilling in a small column, as expected the initial and final arc match with the maximum distillate rate (infinite reflux condition – rectifier). After the initial bang arc, the succession of the first range of chattering arcs is perceived, after which only a singular arc exists between 0.025 and 0.13 (h), it is followed by the second succession of chattering arcs, which exist until 0.16 (h). The optimal control trajectory starts as an innocent-looking problem, with chattering arcs for which the controls switch infinitely often on an arbitrary small interval as the switchings accumulate at the final time. These trajectories are defined as chattering arcs with an infinite number of switchings that accumulate with a geometric progression at the final time t_f = 0.[23]

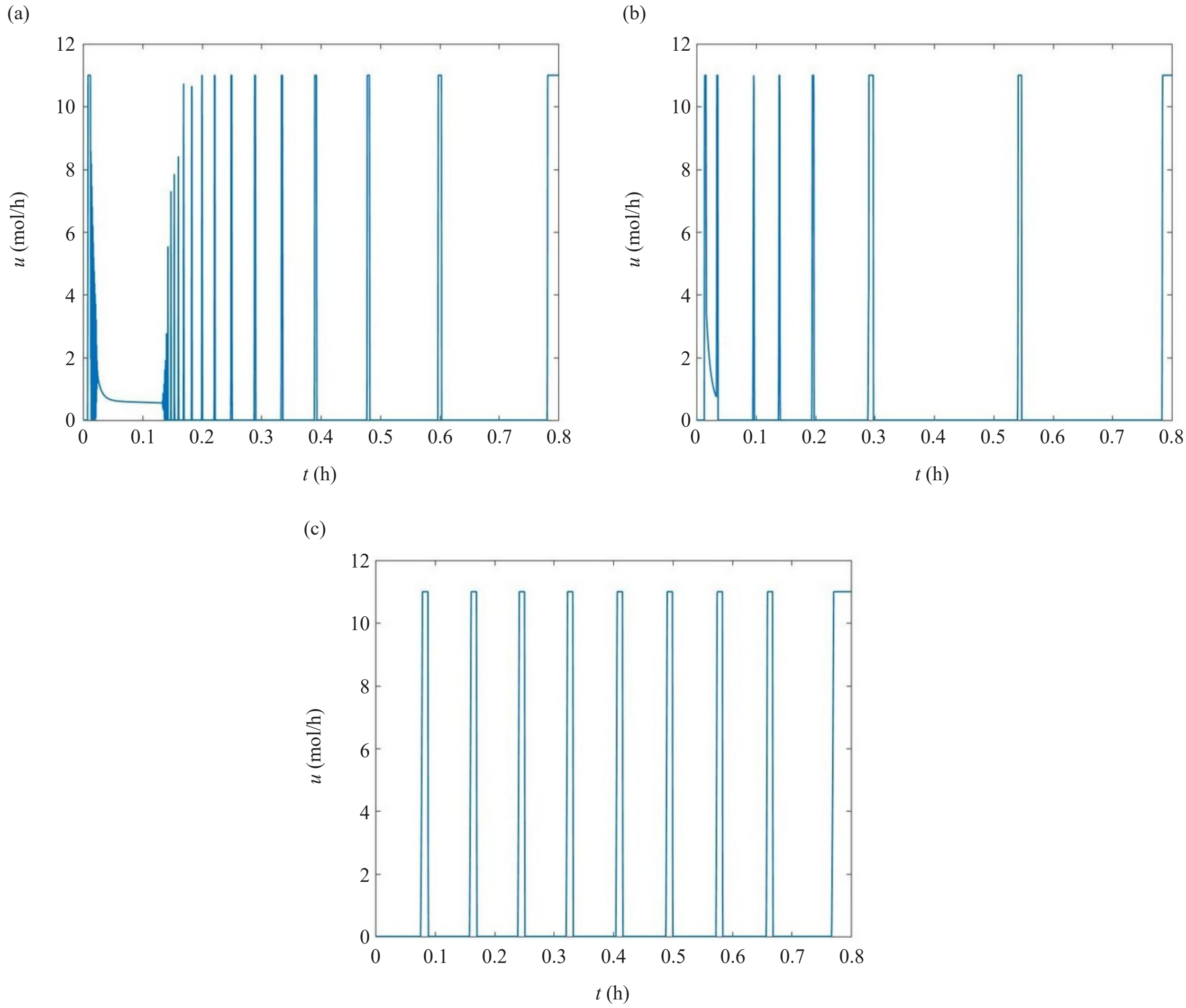

Figure 2. The optimal distillate rate for $u(t)$ for the first step: (a) the main solution, (b) influence of the top pressure, (c) influence of initial mixture composition with the influence of the top pressure

After this specific moment, it is observed that the optimal control trajectory is composed of bang and zero arcs only. There are 18 switches between different types of control. An interesting fact is that with time, zero arcs become longer, whereas the final one becomes the greatest of all existing arcs.

In Table 1, it is noted that the tray hold-up is set at 0.1 mol; however, the influence of this parameter will be examined by changing it to 0.05. Figure 3a presents the optimal control pattern for the column functioning with a twofold decrease in hold-up on all trays. As in the previous case, the optimal trajectory starts with the initial bang arc but is followed by two consecutive singular arcs, concatenated to the second bang arc. It is noted that both previously mentioned bang arcs are of extremely short duration, and that the described part takes 12.5% of the total batch time. One can perceive that the control pattern has changed: there are no more chattering arcs, but in the place of the second succession of chattering arcs, after the zero arc, there exists a succession of “quasi-bang” arcs. The latter are of very short durations and reach not the allowable maximum but 55.55% to 90.90% of it; there are 14 switchings between different types of control. The rest of the optimal control trajectory follows the same pattern as in the previous case, since it is composed of bang and zero arcs only, with eight bang/zero arcs, respectively, both of which tend to last longer over time. Unfortunately, the influence of the twofold decrease in tray hold-up also affects the recovery rate, as it brings 88.02%—i.e., less than 10% lower compared to the previous case, for the same total batch time (3.2 (h)). Moreover, in the work of Farhat,[5] it was found that the total pressure at the top of the rectifier was decreased by 5% from its standard atmospheric pressure value (i.e., 722 mm Hg); for that reason, the influence of this parameter will be examined as well.

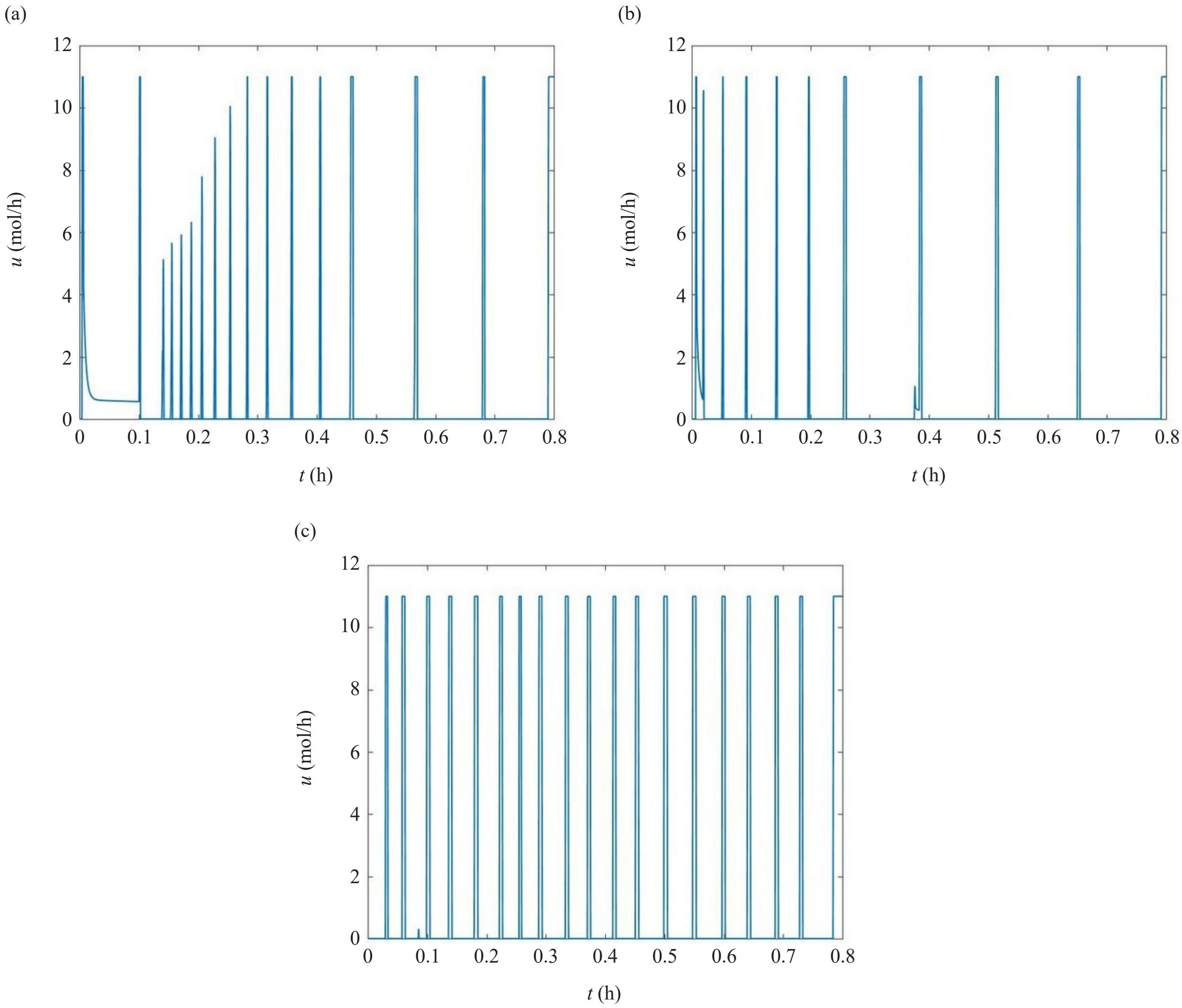

Figure 3. The optimal distillate rate for $u(t)$ for the first step with the influence of tray hold-up: (a) main solution, (b) influence of pressure, (c) influence of initial mixture composition with the influence of the top pressure

Figure 2b presents the optimal control pattern for the column functioning with a 5% reduction in total pressure at the top of the rectifier. As in the previous case, the optimal control pattern starts with the bang arc, followed by a longer singular arc, switching to the second bang arc, whereas both bang arcs are of extremely short duration. The previously described structure takes around 0.025 (h), after which the optimal control trajectory follows a similar pattern as in the previous cases: six zero and six bang arcs exist, and they again tend to be longer with time. In terms of recovery rate, this policy gives almost 20% less over the same time frame compared to the very first case presented in this paper. This means that it would take over 2.035 (h) to reach the acceptable yield. For this case, the influence of tray hold-up can also be embedded. Figure 3b depicts the optimal control pattern where both parameters are varied: total pressure and tray hold-up. The optimal control trajectory likely follows the same trace as explained before, but the first part seems to be of shorter duration, as it takes only around 0.020 (h). Afterwards, the structure is composed of nine bang, nine zero, and two singular arcs; the latter is concatenated to the sixth bang arc on a time interval of less than 0.025 (h). As in the previous case, the decrease in tray hold-up results in a lower recovery rate, giving almost 30% less compared to the very first case examined above.

Moreover, the influence of the initial composition will be investigated. In the very first case above, as noted in Table 1, the initial composition of the ternary mixture was set to (cyclohexane: 0.1; (n-heptane): 0.1; toluene: 0.8); however, the data set will be changed to the case where the first two components are decreased and the third, least volatile, component is increased (cyclohexane: 0.063; (n-heptane): 0.088; toluene: 0.8490). Figure 2c shows the optimal control pattern for the varied initial mixture composition. The optimal control structure is a clear "bang-bang" type, which means that it is composed of bang and zero arcs only, and they are equal in number. There exist 17 switches between different types of control. Compared to the very first case treated in this paper, this policy is much easier to apply; however, it must be noted that it brings 6.80% lower recovery rate. On the other hand, it gives a significant reduction in total batch time, namely 14.07%. Similarly to the previous cases, the influence of two parameters will be investigated as well: initial mixture composition and tray hold-up. Figure 3c shows the optimal control pattern for both parameters varied: initial mixture composition and tray hold-up. Interestingly, the optimal control structure is of the "bang-bang" type too; however, the total number of bang/zero arcs more than doubled, as it contains 19 bang/zero arcs. Consequently, it has 37 switches between different types of control. When it comes to recovery rate, over the same time horizon, it gives somewhat better gains than the previous case—1.35% more.

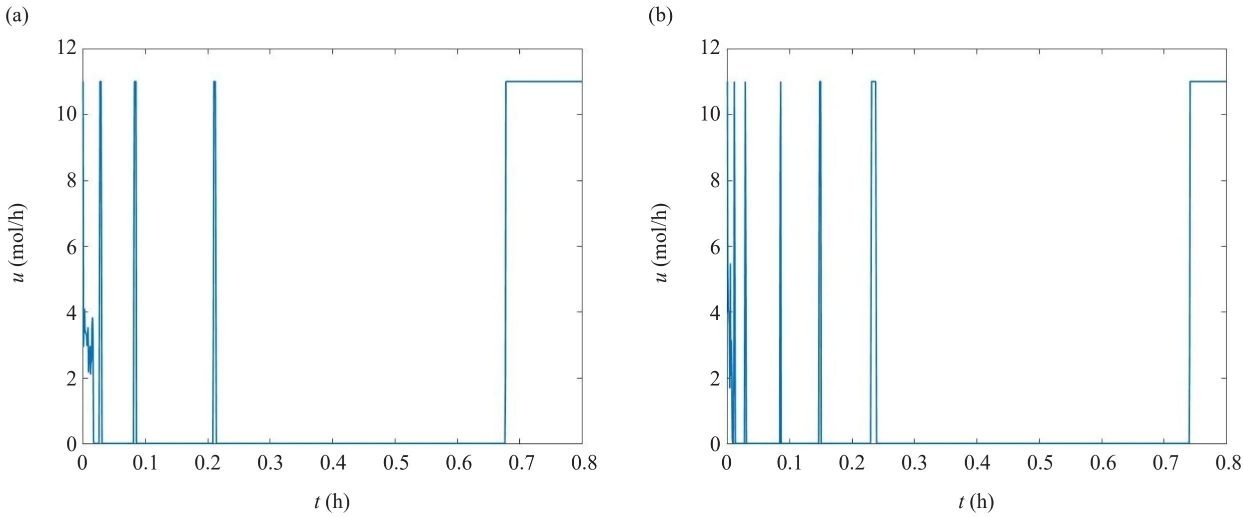

Figure 4. The optimal distillate rate for $u(t)$ for the second step: (a) the main solution, (b) the influence of tray hold-up

For the second step, the binary mixture of (n-heptane)-toluene will be separated in a second rectifier with a total number of 24 trays, with a fixed total batch time of 0.8 (h); from here, the maximum total duration of the overall two-step distillation process is 4.0 (h). Figure 4a shows the optimal control structure for the step of distilling (n-heptane) in an accumulator, with purified toluene left in the reboiler. The initial arc is a singular arc that lasts less than 0.020 (h); next, the "bang-bang" type begins. However, here it has four bang and four zero arcs, with seven switches between different types of control. It is noted that the first three bang arcs are of the same duration, while the last bang arc is much longer than the others, as it takes 0.125 (h) of the total time, which is almost 13 times longer than any other bang arc. For the zero arcs, one should notice that they become longer in duration over time: here, the second one is six times longer than the first, the third is more than twice as long as the second, and the fourth is almost 3.5 times longer than the third. The recovery rate (Table 1) is satisfactory, as it yielded 94.92% of pure n-heptane in the accumulator.

As in the previous cases, the influence of tray hold-up can also be embedded here. Furthermore, the optimal control structure looks familiar compared to the previously described case, but there are significant differences: the optimal control pattern also starts with a singular arc concatenated to a bang arc of extremely short duration, after which the "bang-bang" type of control continues. Here, even five bang and five zero arcs can be perceived. As a consequence, nine switches between different types of control exist. While the very first three bang arcs are of the same duration, the fourth bang arc is twice as long as the former, and the fifth one is even three times longer than the former. Similarly, one can notice for the zero arcs that they again become longer with time: the second one is three times longer than the first, the third is 20% longer than the second, the fourth is almost 13% longer than the third, and the fifth is more than five times longer than the fourth one. Compared to the previous case, the recovery rate is somewhat improved with the decrease of tray hold-up, as it brings an increase of 0.61%. Figure 4b shows the optimal control pattern for the binary distillation case with the influence of tray hold-up.

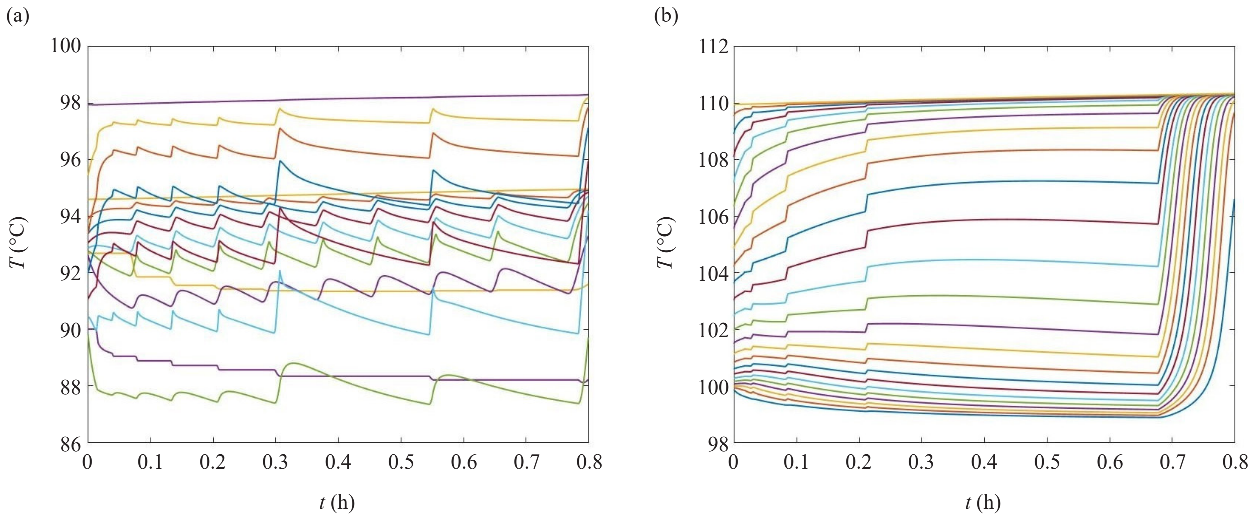

Figure 5. The optimal temperature evolutions for: (a) the first step, (b) the second step

Moreover, Figures 5a and 5b present the output for the temperature evolutions and the optimal solution, which corresponds to the main solutions presented in Figures 2a and 4a, respectively. In Figure 5a, the temperatures on all the stages of the column for the first rectifier are depicted. From here, the temperature decreases along the rectifier, going from the bottom to the top (batch to accumulator), following the optimal trend from a maximal temperature of 98.1578 °C to a minimal one of 88.4921 °C for the first rectifier, and within the temperature interval from a maximal temperature of 110.3222 °C to a minimal temperature of 106.6115 °C for the second rectifier. This confirms the fact that cyclohexane is recovered in the first tank, and n-heptane is recovered in the second product tank. Consequently, it is verified that the ultimate goal of the designed two-step process, i.e., the consecutive production of cyclohexane, n-heptane, and finally toluene, is achieved.

3.1 *The influence of the vapor boil-up*

In previous work, the influence of vapor hold-up was investigated thoroughly and found to be important for minimum-boiling azeotrope mixtures,[24] and this sparked interest in researching in more depth the same parameter variation for the case of a ternary mixture. In Table 3, the investigation of the vapor boil-up is presented; here, the vapor boil-up is a variable parameter. This is intended to challenge the stability of the optimal solution obtained as the clearest “bang-bang” policy (Figure 2c). As previously mentioned, the case of a lower concentration of the most volatile component in the initial mixture showed the best convergence and resulted in the lowest optimal distillate quantity achieved. Observations made are as follows: the total number of cycles does not change with the increase of the vapor boil-up up to 3 (kmol/h), but with further increases, things get more complicated. Initially, it seems that approximately for every additional cycle, one needs to increase the vapor boil-up by a step of 2 (mol/h), which explains the situation for a period of V = 11-25 (mol/h). After this specific point, it will take at least 5 (mol/h) to achieve the same additional number of cycles. All the mentioned facts lead to the idea of using vapor recompression in order to retrofit the column(s) as a possible way to reduce energy requirements.

Table 3. The investigation of the vapor boil-up for lower content of the most volatile component case mixture

Vapor boil-up, V (mol/h)	Distillate quantity, (mol) U_0 (t_f)	Recovery rate (%)/ total batch time (h)	Total number of cycles, n_c	Starting period duration, (h)
11	1.3381	1. 45.75/0.800 2. 99.99/1.748	9	0.080
15	1.5867	1. 54.38/0.800 2. 99.99/1.471	12	0.060
25	1.9964	1. 68.76/0.800 2. 99.99/1.164	14	0.058
30	2.1282	1. 73.30/0.800 2. 99.99/1.019	16	0.052

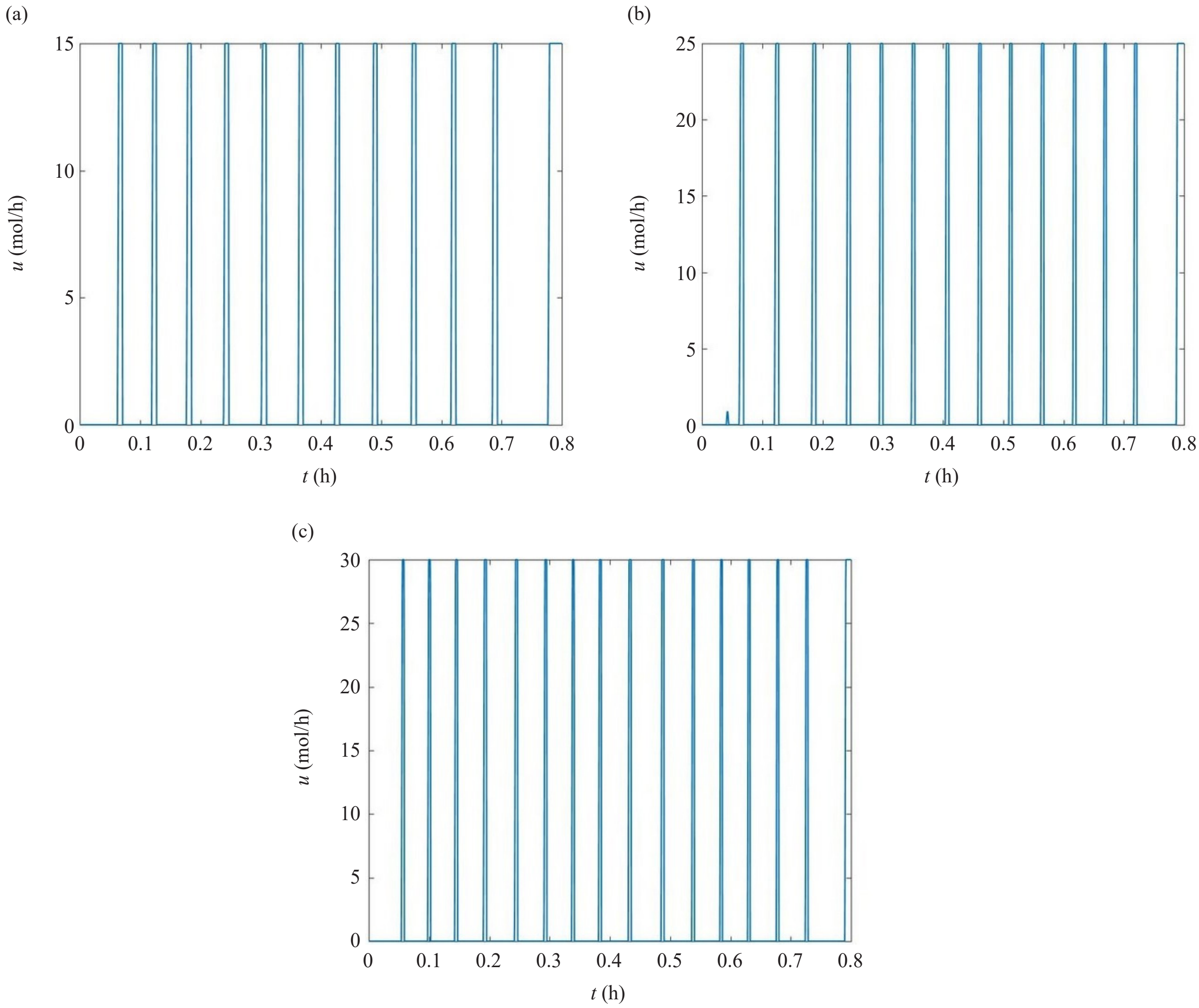

Figure 6. The optimal control pattern for vapor boil-up varied

In Figure 6, the influence of the vapor boil-up on the structure of the optimal control is also observed and depicted: for all values of vapor boil-up applied, the optimal control pattern shows the same clear "bang-bang" policy. As can be seen from Table 3, the starting period duration shows a tendency to shorten with the increase in vapor boil-up; from this observation, one can assume that the start-up heating period should be shortened with every stepwise increase in vapor boil-up. From the recovery rate records, the increase in vapor boil-up is followed by an increase in recovery rate. From the data above, one can perceive that vapor boil-up needs to be increased by 2.73 times to be followed by an increase in recovery rate of 27.55%, i.e., to achieve the maximum recovery over a time horizon reduced by 41.71%.

3.2 *The influence of reboiler volume*

In this chapter, the stability of the solution obtained as the clearest "bang-bang" policy (Figure 2c) is tested by the variation of the reboiler's total volume. Here, if the first step is repeated after its reboiler is filled with an additional charge, an even more complex optimal control pattern is obtained. As one can perceive, the optimal control trajectory is composed almost entirely of bang-bang arcs, with two singular arcs—namely, the second and the 24th—which by their shape resemble "quasi-bang" arcs that failed to reach their allowable maximum, achieving only about 30% and 5%, respectively. There exist 40 switches between different types of optimal control. Interestingly, going from the third to the penultimate zero arc, one can perceive them as sequences of equal duration; however, the final zero arc is shown to be more than 30% longer than the others. In summary, the optimal control has 18 bang arcs, 20 zero arcs, and two singular arcs (Figure 7). The working conditions and optimal costs with recoveries are tabulated in Tables 4 and 5, respectively. Regarding the recovery rate, it is improved by 32.98% compared to the base case presented in Figure 2c; in terms of total recovery, this is achieved in a total batch time reduced by 41.9%. Last but not least, there is an influence on the optimal control pattern, as the total number of cycles also doubled compared to the base case.

Table 4. Working conditions

Parameter	Step	U_N^0 (l)	U_i^0 (l)	V (mol/h)	t_f (h)	x_N^0	y^*
Predefined/ initial value	1	20	0.1	20	1. 0.800 2. 1.016	0.1	0.95

Table 5. The optimal costs and recovery rates

Product recovered	N	$U_0\ (t_f)$	Recovery rate (%)	Discr. scheme, nb. of points
Toluene	7	2.2774	1. 78.73% 2. 99.99%	Gauss, 2,000

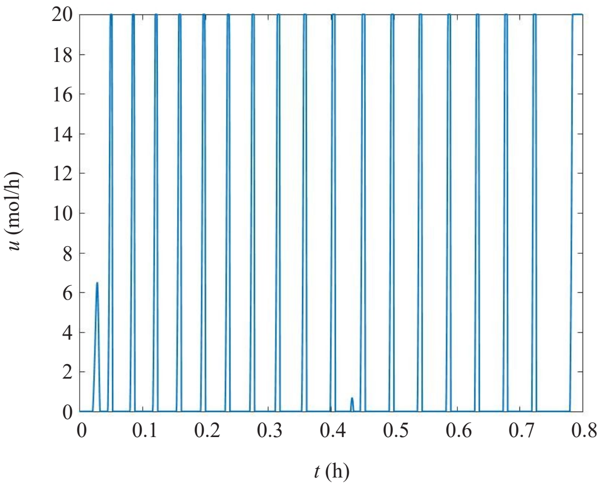

Figure 7. The optimal control pattern for reboiler volume varied

As mentioned in the previous chapter, the mathematical model used in this research is based on the one already developed by Stojkovic et al.;[1] however, the total number of components treated here is increased to three. Therefore, two consecutive separation steps occur: the first step describes the way the least (most) volatile component is separated from the ternary mixture, whereas the second step describes the binary mixture separation. For this reason, all the analyses above are focused on the first step, as it brings novelty to the research. Moreover, a more rigorous thermodynamic model is incorporated (Non-Random Two-Liquid (NRTL)), which consequently assures a better-defined dependence of thermodynamic properties on temperature. To validate the results, ChemCad 6.1, the dynamic simulator, is employed to design the identical cyclic optimal reflux strategy obtained by Bocop over the predefined time horizon. The best way to validate the results is to choose the clearest "bang-bang" case obtained for the lower content of the most volatile component in the mixture. Consequently, from the dynamic simulation, the "discontinuous function of heating energy" is extracted. In Figure 8, the optimal reboiler duty vs time, the extracted results are depicted for each respective step of the process.

In Figure 8a, for the presented reboiler duty function, one can perceive that the strategy applied was as follows: after the initial reboiler duty imposed, in the second step, it needs to be decreased by at least 3.45 times, afterwards it needs to be increased again by at least 6.35 times. After this particular moment, a stepwise gradual increase was imposed of not more than 1.5% until the process reached its midpoint. From this particular moment, a stepwise gradual increase continued, however, with a higher step, being at least 10%. Once the process reaches its penultimate stage, the value should be increased by at least 6.5 times, whereas in the next stage, the value should be drastically decreased by at least 197 times, which gives the impression of a "quasi-zero-bang" sequence, followed by another "quasi-bang" sequence if the process is to continue. From here, the minimum total energy requirement is obtained as 89,750 (kJ/mol).

In Figure 8b, for the second part of the process, the applied heating startegy was as follows: after the same initial reboiler duty imposed as in the previosu case, in the second step, it needs to be incrreased by at least 5.1 times, afterwards, a "plateau" is perceived that lasts for less than 25% of the total batch times, and is formed from reason that, here, a stepwise gradual decrease begins with at least 3%, and continues with much lower values under 0.5%. Once it reaches the penultimate period, the value should be decreased twice in two consecutive steps by almost 4 times and 14.45 times, respectively. Similarly to the previous part of the process, this seems like a "quasi-bang-zero" sequence concatenated with a "quasi-bang-bang" sequence if the process is to continue. Consequently, the minimum heating reboiler duty is found to be 98,750 (kJ/mol).

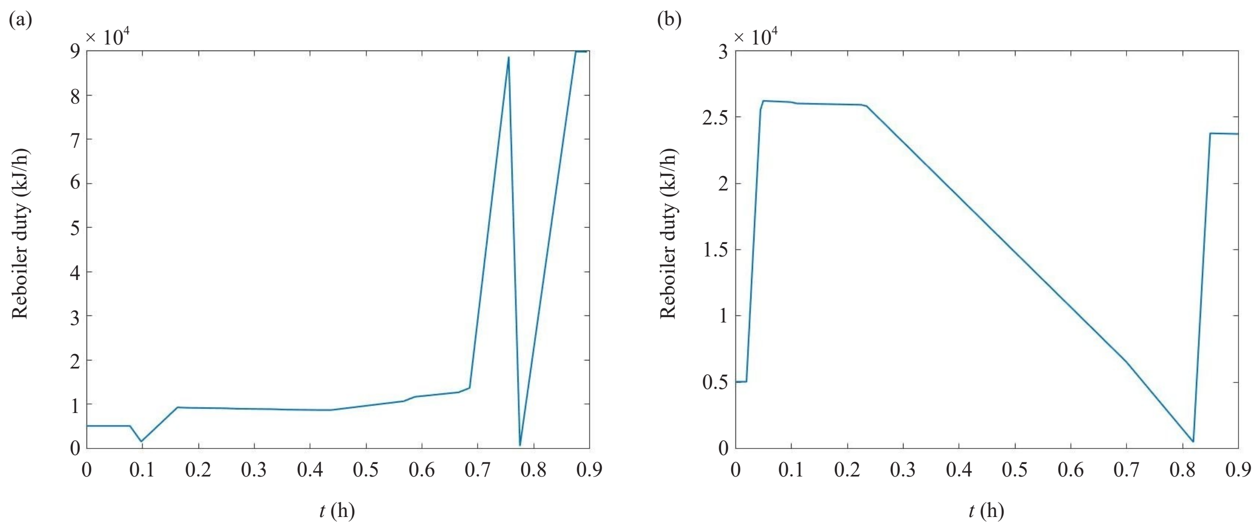

Figure 8. The optimal reboiler duty vs time for: (a) the first step, (b) the second step

4. Conclusions

The optimal control method used solves the problem of optimizing a multicomponent batch distillation campaign using variable reflux ratios. The very first solution, where the initial parameters were still not varied, brought the greatest yield and was shown to be almost 10% better than those reported by Bonny.[9] However, due to the existence of two successive chattering arcs concatenated by a singular arc, this solution would be hard to apply in industry. For this reason, investigations into the influence of parameters such as total pressure at the top of the column, initial mixture composition, and tray hold-up were conducted. As an outcome, clear "cyclic reflux ratio policies" were obtained only after the initial mixture composition was changed to contain less than 15% of cyclohexane and n-heptane together in total, and the total top pressure was reduced by 5% compared to the standard atmospheric pressure. This latter policy requires fewer cycles and less time, with time reduced by 14.07% to achieve the total recovery rate.Moreover, the influence of vapor boil-up and reboiler volume was investigated: the choice should be made between increasing vapor boil-up by 2.73 times and/or reboiler volume by 2 times to achieve very similar total batch processing time reductions, i.e., 41.71%/41.90%, respectively, for total recovery (99.99%). Last but not least, after validation, the data were extracted to depict a "discontinuous heating energy curve," supporting the idea that the dual process can be controlled by the reboiler duty control parameter: "quasi-bang-bang" and "quasi-zero-bang" sequences as a rule covering all parts of the process. This later provides space for further optimization based on energy expenditure minimization, and the approach could be expanded to even more complex multicomponent mixtures, i.e., those containing azeotropes.

Conflict of interest

There are no conflicts of interest for this study.

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