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Research Article

Pressure-Driven Batch Distillation Optimal Control for Acetone-Methanol Separation

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Abstract: Over the last fifteen years, immense progress has been made in the research of pressure-swing batch distillation. The challenge lies in the fact that certain pressure-sensitive azeotropic mixtures cannot be separated in a regular open batch mode with an acceptable outcome. Throughout most of this text, findings contradict previously established facts. The separation of acetone-methanol by pressure-swing batch distillation in a mixed double system, consisting of a regular and inverted column, is the process under investigation. In this work, a comprehensive global solution to the optimal control problem is derived in the form of a sequential synthesis of controlled trajectories. A thorough analysis of the control variables, with and without parametric sensitivity of manipulated variables, on the optimal control pattern with variable reflux is presented. Most authors did not achieve significant results by simultaneously optimizing reflux and liquid division ratios or by using a "variable pressure gap." This study challenges previous findings, emphasizing the importance of simultaneous optimization of all three factors mentioned. During this study, the optimal reflux strategy through cyclic operation was extended to optimize energy expenditure over a fixed time horizon. Moreover, the proposed scheme offers a high level of operational flexibility, allowing units to be operated independently or as a system. Additionally, the facility of connecting additional devices is highlighted by the significant difference in output temperatures and the potential for using evaporated fluids consecutively. Lastly, the research provides a solid basis for future investigations into internal and external heat integration, potentially leading to conclusions about expanding the network using well-known heat cycles.

***Keywords*:** pressure-swing, batch distillation, optimal control, direct method, cyclic distillation

Nomenclature

AIB	Advanced inverted batch with an additional tank at the top
ANN	Artificial Neural Network
BB	Big bottoms
BE	Batch extractive column
BR	Batch rectifier
BS	Batch stripper
DCBR	Double column batch rectifier

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DCBS	Double column batch stripper
DEG	Diethyleneglycol
DHSA	Distillation harmony search algorithm
EAPSD	Entrainer assisted pressure-swing distillation
GA	Genetic algorithm
HHIPSD	Hybrid heat-integrated pressure-swing distillation
MEG	Mono ehtylene glycol
MBDS	Multi Batch Distillation Software
PSOA	Particle swarm optimization algorithm
RB	Regular batch
SIM	Sequential iteration method
SMBO	Surrogate model-based optimization
SQP	Sequential Quadratic Programming
TAC	Total annual cost
TCC	Triple column configuration
THF	Tetrahydrofuran

1. Introduction

Repke and Klein[1] were the first to propose a simple control scheme for a pressure-swing inverted batch double-column system: product concentration was controlled by the reflux rate flows. Indeed, the low-pressure column showed a rapid response, with a jump from third to second decimals. Repke et al.[2] recognized the volumetric ratio against molar-decisive and recommended the inverted configuration in the case of the light component if the initial feed is lower than 0.25. Modla and Lang,[3] however, compared different configurations to conclude: (1) single/double rectifier are only suitable for maximum boiling azeotrope, (2) single/double inverted column are only suitable for minimum boiling point azeotrope, (3) a combination of the rectifier and inverted column is possible to use only if in each step the azeotrope is continuously withdrawn as a product, (4) middle vessel is not suitable for any. Later, Modla and Lang[4] introduced a "speed of pressure change" to end up with conclusions about the products obtained, respectively: (1) for maximum boiling azeotrope, double batch rectifier was the most "recovering" configuration, giving 7.5%/10.6% more concerning the combination of rectifier and stripper, and only 1.5%/6.4% more for a single rectifier, (2) for the minimum boiling azeotrope, double stripper shown to be the best "recovering" configuration giving 3.3%/29.2% more to the combination of rectifier and stripper, respectively, and 19.8%/11.9% to a single stripper. But Modla[5] established as follows: (1) minimum boiling azeotrope separation is feasible in all except closed stripper, (2) maximum boiling azeotrope separation is feasible in closed stripper and open rectifier.

Even though their work involves classical control techniques, according to Modla,[5] Li et al.[6] and Kopacz et al.,[7] it is worth mentioning the findings. Kopacz and coworkers[8] introduced a vapor division ratio which proved to affect the reflux ratio and consequently total time/energy requirement. Modla[5] again highlighted the significance of the influence of vapor division ratio on energy requirement: double-column rectifier in closed mode resulted in 3.2/1.74 times more reduction of energy requirement compared to the open mode/single inverted column, respectively, but double stripper configuration in the open mode required even 1.54 times more energy compared again to the single open stripper. Modla and Lang[9] (DCBS open mode) examined the process with/without heat integration: having pressure gap increased by 9 times, CO_2 emission reduction increased by 19.12%/29.05%, respectively. Modla,[10] for the charge composition inside/outside of the pressure-swing operating region, claims the decrease in energy expenditure by (29.85%/49.56%)/37.42%, for DCBR/TCC against the (BS-DCBR/TCC)/(BS-DCBR-DCBR), respectively. Further, Modla and Lang[11] proved that thermal integration benefits go up to 59.18%/67.13%, for the pressure-swing/extractive processes, respectively. Modla[12] studied a system comprised of the reactive rectifier combined with a non-reactive stripper to discover a "sharp uprisal" in energy demand, for feed plate locations ranging from 35 to 40.

Even with classical control techniques applied, the results of Li et al.[13] and Wang et al.[14] are noteworthy. It is to note that, after having combined a system of double column stripper batch with a continuous rectifier, Li et al.[13] proved a minimum pressure gap of 4.4 atm and succeed in impure alcohol products to their maximum. On the other side, having low/high-pressurized column pressure increase by 39.50%/75%, respectively, Wang et al.[14] gained 12.11%/19.93% in TAC reduction. Compared with the improvement in terms of greenhouse gas emissions, high-pressurized columns saved almost 10% more. Most recently, for a minimum-boiling azeotrope of THF-water, Luyben,[15] using a double continuous mode rectifier with vapor recompression, delimited LP/HP working pressure conditions to a vacuum of (0.6 atm)/above technical (11.3 bar): This outperformed "traditionally researched scheme" (Luyben[16]), by 19.4% in TAC, whereas, by 40.15% with heat integration.

Having considered three cases according to the content of the light component, Li et al.[6] concluded that: (1) if a low-pressurized column operated at atmospheric pressure and the pressure of a high-pressurized column is increased by 27.28%, it influences TAC to decrease by 23.87%, (2) also if low-pressurized column pressure increases only by 0.57%, the same pressure increase as in the first case for the high-pressurized column here invokes reduction of TAC by 34.19%. For a particular case examined, it seems that if the pressure gap is reduced by less than 1%, TAC is, however, reduced by more than 10%. Even with classical control techniques applied, the results of Li et al.[13] and Wang et al.,[14] are noteworthy. It is to note that, after having combined a system of double column stripper batch with a continuous rectifier, Li et al.[13] proved a minimum pressure gap of 4.4. atm and succeed in impure alcohol products to their maximum. On the other side, having low/high-pressurized column pressure increase by 39.50%/75%, respectively, Wang et al.[14] gained 12.11%/19.93% in TAC reduction. Compared with the improvement in terms of greenhouse gas emissions, where, high-pressurized columns saved almost 10% more. Most recently, for a minimum-boiling azeotrope of THF-water, Luyben,[15] using a double continuous mode rectifier with vapor recompression, delimited LP/HP working pressure conditions to a vacuum of (0.6 atm)/above technical (11.3 bar): This outperformed "traditionally researched scheme" (Luyben[16]) by 19.4% in TAC, whereas, 40.15% with heat integration. Afterward, Luyben[17] in particular proved benefits from vacuum distilling: For a specified range of pressure within the technical limits, for an increase of [2-2.5] folds, the reduction in capital cost varies from 20.64%-21.22%. Furthermore, for a ternary mixture of MEG/DEG and water, the author concluded for derived increments on investment, going from higher to lower vacuum pressure, to rise if moving toward the higher decimal value.

Most recently, Kale et al.[18] studied and optimized the batch distillation system of acetone and n-heptane, and found the optimal parameters to be a reflux ratio of 2.2, the total number of plates to be 19, in the total time interval fixed for 1 h, to purify more than 99% of acetone from the azeotropic mixture. Ding et al.,[19] in their first case, separated acetone from methanol in a double-column system, to compare different algorithms in terms of the optimal value for TAC: Distillation harmony search algorithm (DHSA) gave, 0.5%/0.9%/0.6% lower objective than that of sequential iteration method (SIM)/genetic algorithm (GA)/particle swarm optimization algorithm (PSOA), respectively. Hegely et al.[20] separated methanol from a 5-component mixture to discover that the influence of the price of incineration on the objective function, i.e., profit, is significant, because the profit decreases by 78.42% when the price of incineration increases only by 17.72%. Qin et al.[21] used entrainer-assisted pressure-swing batch distillation for separating pressure-insensitive binary azeotrope: The authors showed that "the order" of the different pressure set-up sequences is also important as they proved that heat-integrated high-low pressure sequence has lower TAC and CO_2 emissions, by 7.04%/4.27%, respectively, compared to low-high pressure sequence. Miao et al.[22] introduced a new hybrid heat-integrated pressure-swing distillation process with a heat pump to separate both maximum- and minimum-boiling azeotropes, and, compared with the conventional fully heat-integrated pressure-swing distillation process, to conclude that the first one gives from 28.8% to 51.9% reductions in TAC, depending on the azeotropic system observed.

In this study, the optimal control for the operation of pressure-swing batch distillation for acetone production in an open-mode double-column configuration is researched. The goal of this study is to investigate the optimal control functioning of the combined scheme, composed of both batch rectifier and batch stripper, in "dwelling" between pressures of atmosphere and high pressure allowed the industrial practice. Throughout the manuscript, firstly the balances are introduced for column functioning at elevated pressure, then the optimal control problem is defined, where some important thermodynamic conditions are introduced as an operational constraint since the latter is key for safety operation(s). From here, the optimal control reflux policy as a cyclic operation is obtained. Furthermore, to research more in-depth the influence of different parameters on the optimal control structure, the author extends this study by (1) investigating the influence of liquid ratio, and, the investigation of the influence of vapor boil-up, (2) investigating the influence of reboiler volume, (3) investigating the influence of the pressure gap. Least, but not last, within the investigation of the influence of vapor boil-up and the investigation of the influence of the pressure gap, the optimal reboiler duties are obtained as discontinuous functions over the fixed time horizon for each investigation. This is of the most importance, as it shows that it is possible to achieve the reduced energy requirement.

All in one, for the very first time a cyclic operation as an optimal control reflux policy of long-time researched azeotropic mixture is developed. Table 1, shows a list of studies on the optimal control for homogeneous systems in discontinuous and/or semi-continuous mode for pressure-swing distillation.

Table 1. A list of different studies on the optimal control for homogeneous systems in discontinuous and/or semi-continuous mode for pressure-swing distillation

Authors(s)	year	column (s)	method/control	mixture
Repke et al.[1]	2005	BS	gPROMS	acetonitrile-water
Repke et al.[2]	2006	BR, BS	gPROMS	acetonitrile-water
Modla and Lang[4]	2007	BS, BR; DCBS, DCBR, BS+BR	CCDCOLUMN	Ethanol-toluene water-ethylenediamine
Modla and Lang[9]	2010	BS	ChemCad	Acetone-methanol
Modla and Lang[11]	2011	DCBS BE	ChemCad	Acetone-methanol
Li et al.[13]	2019	DCBS, TCC	SQP/AspenPlus	THF-methanol-water
Zhao et al.[23]	2021	DCBS-series DCBS-parallel BS+DCBS	AspenPlus	hexane/ethanol/butanone
Zhao et al.[24]	2021	DCBS with decanter *heterogeneous	AspenPlus	Cyclohexane-acetonitrile-toluene
Qin et al.[21]	2022	EAPSD	GA	Water-isopropanol
Miao et al.[22]	2022	HHIPSD	GA	Water-ethylenediamine, Methanol-aceetone
Kale et al.[18]	2023	BR	MBDS	Acerone-(n-heptane)
Ding et al.[19]	2023	DBCR, TCC	SIM, GA, PSOA, DHSA	Acetone-methanol, Silicon tetrachloride-trichlorosilane-dichlorosilane
Hegely et al.[20]	2023	BR	ANN, GA, SMBO, SQP	Acetone-methanol-tetrahydrofuran-water-toluene

2. Materials and methods

2.1 *Process dynamics model*

The following assumptions are taken: (1) equimolar overflow, (2) total condensation, (3) vapor hold-up dynamics neglected, (4) pressure drop along the columns neglected, and (5) ideal mixing of the vapor and the liquid phase. The detailed mathematical model developed by Stojkovic et al,[25] which includes mass balances on all the parts of the batch column (MES), is used.

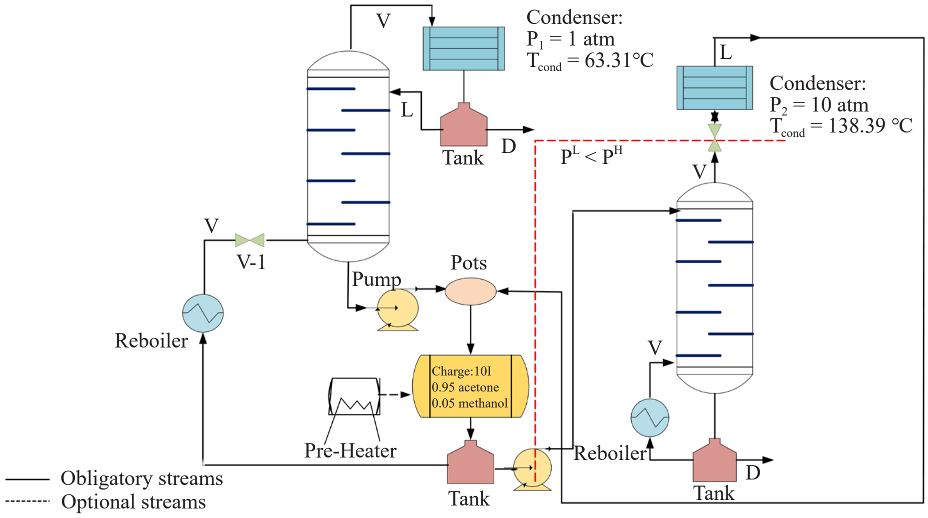

Figure 1. A combined scheme for pressure-swing batch distillation

However, having the second column set at high pressure (P > 3 atm) implies the necessity to take the pressure effects into account. According to Doherty and Malone,[26] for high-pressure distillation, the vapor-liquid equilibrium relation recommended is:

$$Py_i = P\gamma_i x_i \exp\left(\frac{(v_i - B_{ii})(P - P_i)}{RT}\right) \tag{1}$$

Equation (1), P, P_i refers to total/partial pressure, respectively, whereas, x_i, y_i refers to the molar concentrations of the respected component in the liquid/vapor phase, respectively. Furthermore, γ_i, υ_i refers to the coefficient of activity/volume of the pure liquids/second virial coefficients, respectively. The coefficients of activity are described by the NRTL model, subsequently, the parameters are tabulated in Table 2. In Table 3 and Table 4, however, Antoine's parameters are tabulated, for respective components over different intervals of temperature.[27] A second virial coefficient denoted as B_{ii} is calculated from Pitzer-Curl equations for non-polar molecules.[28]

Overpressuring effects should be avoided at first because if the control valve on a feed line falls fully open, it can result in overpressure in a reboiler, accumulator, or even main trays. Consequently, the left side of equation (1) must be constrained to the value of the working pressure during the operation.

Table 2. Thermodynamics data

Binary interaction parameters for NRTL model					
Component i	Component j		A_{ji} [cal/mol]	A_{ji} [cal/mol]	α
Acetone	Methanol		78.317	140.046	0.47
Antoine parameters from DECHEMA					
Acetone					
n°	A	B	C	T_{min}, [C°]	T_{max}, [°C]
1.	7.1327	1,219.97	230.65	-64	70
2.	7.6313	1,566.69	274.42	57	205
Methanol					
n°	A	B	C	T_{min}, [°C]	T_{max}, [C°]
1.	8.0810	1,582.27	239.7	15	100
2.	7.9701	1,521.23	234	65	214

The pressure-swing process scheme presented in Figure 1, assumes "one-pass" through the column at the time [2], in other word, it is assumed that columns work consecutively ("two-step" process). In step (1) LP run (rectifier), and, in step (2), HP run (stripper), whereas the maximum achievable distillate concentration at lower pressure P_1 = 1 atm is 78% of acetone, but, at P_2 = 10 atm only 37% of acetone.

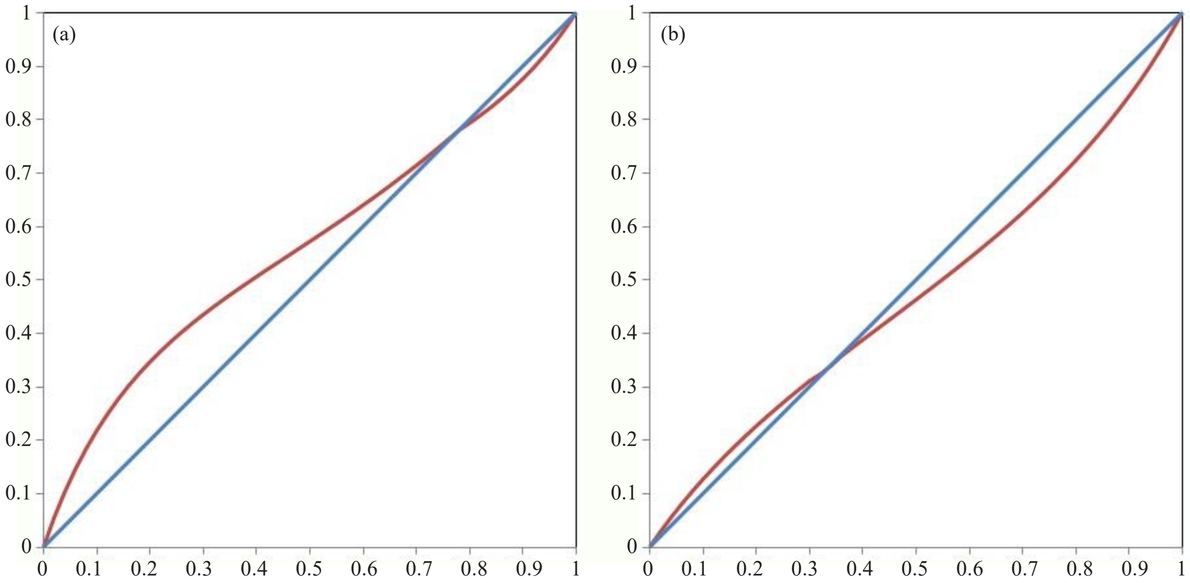

Figure 2. Vapor-liquid equilibrium curves for acetone-methanol at different pressures: (a) P_1 = 1 atm; (b) P_2 = 10 atm

2.2 *Optimal control problem*

In the same Figure 2, the vapor-liquid equilibrium is presented for a mixture of acetone-methanol at different pressures: The data obtained from Matlab Version R2017a simulation are plotted against ChemCad 6.1 professional simulator for the same thermodynamics model, described previously. In Figure 2, on the right/left side plot, respectively, the vapor-liquid equilibria for pressure set at P_1 = 1 atm/P_2 = 10 atm, respectively, show that the azeotropic point coincides with 78%/37%, respectively, containing acetone in the mixture.

Since the set of equations describing the related mathematical model is attributed to the conservation of the mass property, hereby, the "conservation of the total working pressure" through equation (1) is assured as well: The summation of partial pressures of components needs to be equal to the working pressure at any moment, whereas to respect the physical limits imposed by the molar fractions of components in vapor/liquid phase.

Consequently, relations defining physical properties are:

$$K_{j,\,i} = K_{j,\,i}\left(y_{j,\,i}, X_{j,\,i}, T_j, P\right) \tag{2}$$

whereas, j, i denote, plate/component number respectively.

The optimal control problem can be described as follows: For a given pressure-swing 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, $R = LD$, to maximize the distillate, subject to any constraints (model equations, bounds on the optimization variables).

The maximum distillate problem retained as:

$$\min\ q_{N+1}\left(t_f\right) \tag{3}$$

$$\frac{dq}{dt} = f_0(q) + uf_1(q),\ q \in X_1 \tag{4}$$

Hereby, the X_1 denoted as reduced state space and defined as $q_{\mathrm{N}+1}(t_f)$

$$X_1 = \left\{ q = \left(X_1,\ \ldots,\ X_N,\ U_N,\ z\right):\ x_i \in [0,\ 1]\forall_i \right\} \tag{5}$$

In the equation (4), q is composed of X_1 ... X_N, molar concentrations of the light component in the liquid phase, U_N, liquid hold-up in the reboiler, and z, purity deviation.

$$q_0 = q^{\mathrm{ss}},\ q_{\mathrm{N}+2}\left(t_f\right) = 0 \tag{6}$$

where is t_f predetermined and $q^{\mathrm{SS}} \in X_1$ is composed of the steady state values of state variables. Vectors f_0, f_1 previously defined by Stojkovic et al.[25]

As suggested by Modla,[5] the boundary conditions to assure the infinite reflux condition for open mode rectifier:

$$U_{N_\alpha}(0) = U^0_{N_\alpha} = U^0_{\mathrm{ch}};\ U_{i_\alpha}(0) = U^0_i; \tag{7}$$

and to ensure the total reflux condition for the open-mode stripper are:

$$U_{N_\beta}(0) = U^0_{N_\beta};\ U_{i_\beta}(0) = 0; \tag{8}$$

Whereas, the first/second column is denoted with the first/second letter of the Greek alphabet, respectively, and *ch* denotes a respective charge content.

3. Numerical results and discussion

3.1 *High-pressure driven process set-up*

In this study, the investigation is expanded on a homogeneous azeotropic mixture, which thermodynamic properties match very well with the thermodynamic model used (Figure 2). The working conditions and predefined parameters are tabulated in Table 3, whereas concentrations are expressed from acetone, and LP/HP refers to low/high-pressure column, respectively.

Table 3. Working conditions

Parameter	P [atm]	U^0_N [l]	U^0_i [l]	V [mol/h]	t_f [h]	x^0_N	y^*
Predefined/initial value	LP: 1 HP: 10	10	0.1	11 12	1.6	0.1	LP: 0.75 HP: 0.35

At the beginning of the dual process, the reboiler is filled with $U_N^0 = 10$ (l), and liquid hold-up on the trays is set to $U_i^0 = 0.1$(mol) for $i = 0$, ... N. For the boil-up, the constant value for the LP step is set to $V_1 = 11$ (mol/h), however, at V_2 = [11-13] (*mol*/*h*) for the HP step. The low-pressurized column works with the total number of trays N_1 = 23, whereas the high-pressurized column at even N_2 = 13. As stated in the previous section, for the azeotropic kind of mixture, the maximum achievable distillate concentration is limited by the position of the azeotropic point. Therefore, the purities imposed for each step of the process: low/high-pressurized column to achieve, 75%/35%, respectively. Next, the working conditions and predefined parameters are tabulated in Table 3, whereas LP/HP refers to low/high-pressure columns, respectively.

Firstly, the operation is managed under standard atmospheric pressure, to distill acetone, i.e., 75% of left in the accumulator. Consequently, the purified methanol (96.59%) is obtained in the reboiler and withdrawn. Furthermore, the reboiler is heated by an additional auxiliary exchanger (Luyben[17]), before being pumped to the higher pressure and introduced to the second column, this is necessary to increase boil-up. In the stripping step, the mixture near the azeotropic mixture is collected in the condenser, to obtain purified methanol in the second tank. Moreover, the interesting fact is that both, the purity and the recovery rate, tend to improve but at the cost of the total batch time. As a consequence, it is a must to "repeat" a step to purify the next batch up to 95.50% and increase the recovery rate by an additional 3% (Table 4). The total duration proposed for the best performance of the dual process is t_f = 2.4 [h].

Despite the fact that the property of distilling under the pressure change, is exploited at maximum, the recovery rate for the acetone is 72.83%, and, still almost 10% more than what was reported by Modla and Lang[4] for the particular configuration. In conclusion, the proposed operation is recommended to be employed as a two-step process for methanol production and purification. Moreover, it is possible to have a second run for the HP step, where the acceptable purity of 95% is achieved. The recovery rate improved by almost 4%, consequently, after the third run one can expect to reach the maximum purity, but at the cost of the batch time increase, i.e., the total duration of 2.4 [h].

3.2 *Numerical solution by BOCOP solver*

A specific optimal control problem is formulated based on the assumptions of constant boil-up and product specification for the respective step. Numerical solutions are brought by the BOCOP solver, as detailed by Bonnans et al.[29] This solver, in particular, employs the direct method with a choice of discretization schemes for dynamics time discretization, which furthermore conversed the problem into finite-dimensional. In Table 4, the optimal costs and the recovery rates of the respected component recovered in a two-step process are displayed.

Table 4. The optimal costs and the recovery rates

Product recovered	N	U_0 (t_f)	Recovery rate	Discr. scheme, nb. of points
Acetone	23	2.47865	72.83%	Gauss 2,800
Methanol	13	1.37305	91.21% * 94.47% **	Euler 2,800

* t_f = 0.8 h, "single-period", ** t_f = 1.6 h, "double-period"

The optimal control policies for the azeotropic mixture of acetone-methanol are depicted in Figure 3, as the LP step in the batch rectifier/HP step in the batch stripper, respectively.

For the first step, i.e., low-pressurized column (N_1 = 23), as expected the initial and final arc match with maximum distillate rate (infinite reflux condition, rectifier). It is to note that, the instantaneous pique that appeared after 0.47 [h], will be disregarded. The optimal control trajectory is composed of 3 bang arcs of maximal distillate rate, but, even three consecutive singular arcs with the tendency to reach approximately 27.30%/3.64%22.73% from the maximal distillate rate. It is to note that, the latter mentioned, are taking over (more than any particular bang arc), approximately 23% of the total time. In summary, the optimal control has an equal number of arcs of each type: 4 bang arcs, 3 arcs of zero control, and, 3 singular arcs. Finally, there are 8 switchings between different types of optimal control.

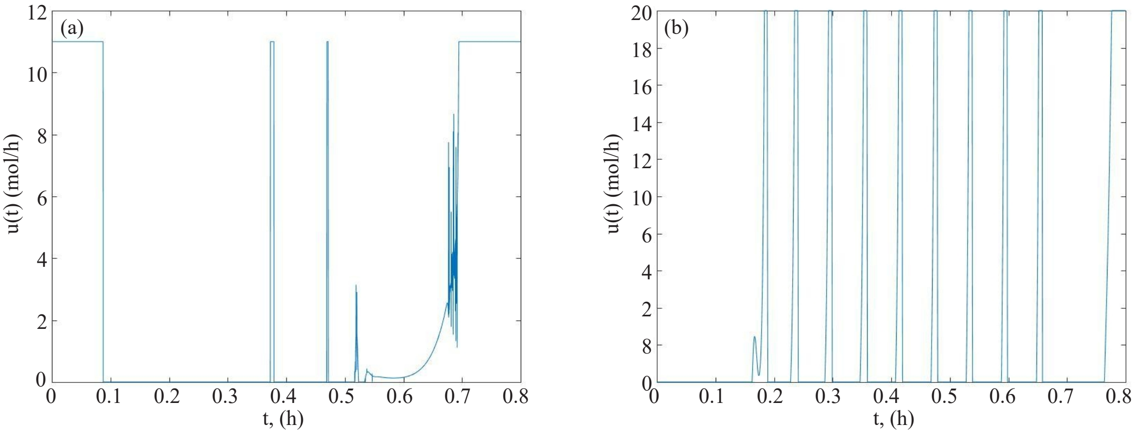

Figure 3. The optimal distillate rate for *u*(*t*) for the: (a) LP step, (b) HP step

In the second step, i.e., high-pressurized process, as supposed the initial arc matches with zero distillate rate (total reflux condition-stripper), but the final with maximal distillate rate. Further, it is observed that the optimal control trajectory is composed of almost all bang-bang arcs, with two consecutive singular arcs preceding the final maximal distillate arc. There are 13 commutations between different types of control, an interesting fact is that the singular arc (s) are 'positioned' again in the penultimate period, whereas here it lasts approximately 6.25% of the total operation time or more than 3 times shorter than in an LP step. In summary, the optimal control has an equal number of bang and zero arcs, 6 and one singular arc.

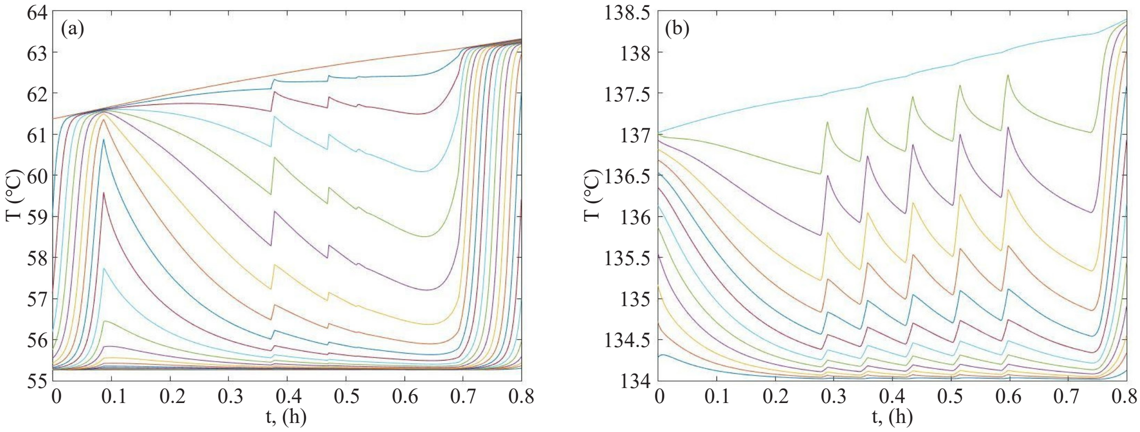

Figure 4. The optimal temperature evolution for the: (a) LP step, (b) HP step

Moreover, in Figure 4, the output for the temperature evolutions along with the optimal solution is presented. Above, the temperatures on all the stages of the column for the rectifier (LP step) are depicted. From here, the interval of temperature increases along with the rectifier, going from the bottom to the top (batch to accumulator), following the optimal trend from the minimal temperature of 55.25 °C to the maximal one at 63.31 °C, at the specified working pressure of P_1 = 1 atm. As previously stated, verify that the acetone is recovered in the product tank. Below, the optimal temperature evolutions are presented for the HP step of the process, i.e., stripper, at the specified working pressure of P_2 = 10 atm, temperature rise, but in an inverse sense, from the top to the bottom, following the trend within the temperature interval from the minimal temperature of 134.27 °C to the maximal temperature of 138.39 °C. Hereby, previously written goes in favor of the fact that methanol is recovered in the inverted column product tank. All previously stated are also following the findings reported by Luyben.[16] As a consequence, it is verified, that the ultimate goal of the designed two-step process, i.e., consecutive production of acetone and methanol, is achieved.

3.2.1 *The influence of liquid ratio*

Focusing on the HP step, a short parametric study, whereas, the setup parameter of boil-up is varied, provided a further study on the influence of the liquid division ratio on energy requirement. Data for the optimal liquid division ratios obtained are tabulated in Table 5. Furthermore, after the validation of each pattern, the optimal reboiler duty "pattern" was shown to be highly sensitive to the liquid division ration change, as if the latter varied by less than 9%, it jumps for the next 19.81%.

Table 5. The liquid division ratio ϕ tabulated for different values of vapor boilup

L	3.182	3.311	3.533	3.543	3.652	3.756
ϕ	0.459	0.487	0.537	0.548	0.575	0.601

Additionally, the influence of vapor boil-up on the structure of the optimal control pattern is observed, as well. In Figure 5, are plotted. Observations made, are as follows: the total number of cycles does not change for the decrease of the vapor boil-up to 2 [mol/h], but further "gradual/stepwise" decrease results in the "equal decrease" of the total number of cycles, since in the next step of the decrease by 4 [mol/h], the disappearance of the one cycle occurs; again up to 6 [mol/h], the disappearance of even three cycles occurred, and up to 8 [mol/h], four cycles are disappeared. The observations regarding the influence of the vapor boil-up on the optimal control pattern enable us to extend the conclusions toward the prediction of the optimal heating strategy for each particular case. Finally, conclusions are drawn about the total energy expenditure for the entire process, and the possibility of internal heat integration is thoroughly examined, as discussed by Li et al.[6]

In Table 6, the investigation of the vapor boil-up is given, with different values of vapor boil-up, the achieved distillate quantities, recovery rates, total number of cycles, and starting period duration.

Table 6. The investigation of the vapor boil-up

Vapor boilup, V [mol/h]	Distillate quantity, [mol] U_0 (t_f)	Recovery rate	Total number of cycles, n_c	Staring period duration, [h]
7	3.2111	95.011%	4	0.4850
8	3.3017	93.427%	6	0.4195
10	3.5322	92.815%	8	0.3350
11	3,6423	92.185%	10	0.3000
12	3,7517	91.510%	12	0.2795
13	3,8556	90.853%	12	0.2595
14	3,9564	90.260%	14	0.2395
15	4,0539	89.660%	18	0.2195
16	4,1486	89.086%	18	0.2050
18	4,3285	87.971%	20	0.1825
19	4,4137	87.478%	22	0.1780
20	4,4960	86.961%	21	0.1600

* calculated per $t_f = 0.8$ h, "single-period"

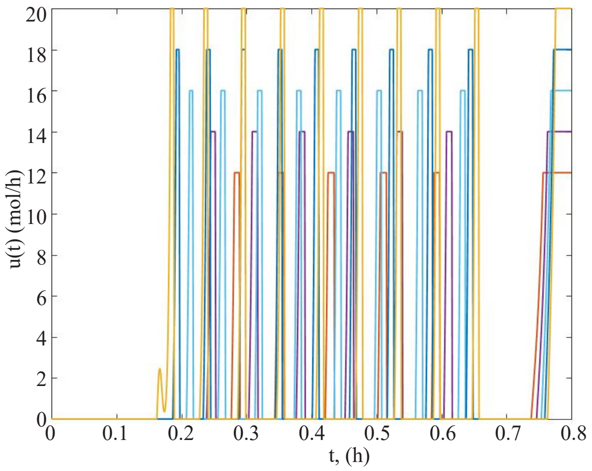

Figure 5. The optimal control pattern for vapor boil-up varied

Compared to the previous cases, the operation should be initialized at higher values, however, the subsequent almost instantaneous decreasing pique appeared again (Figure 6). It is perceived, that both consecutive "energy gaps" are broadened", this is followed by a very flattened decrease till the end of the first half of the total time. For the first operational strategy proposed, the flattened area keeps its average value, but, almost double, compared to the second one. This is followed by a more steep, since greater step, stepwise increase reaching the very last pique before switching to the minimum of energy, followed by the final instantaneous increase of more than 3 times. On the other side, the second proposed strategy, the "last" stepwise increase is instantaneously switched to the even greater pique, even on average three times more compared to the first one. It is also important to mention that, the last pique of the increase reaches more than 30% of the final estimated maximum, reboiler duty.

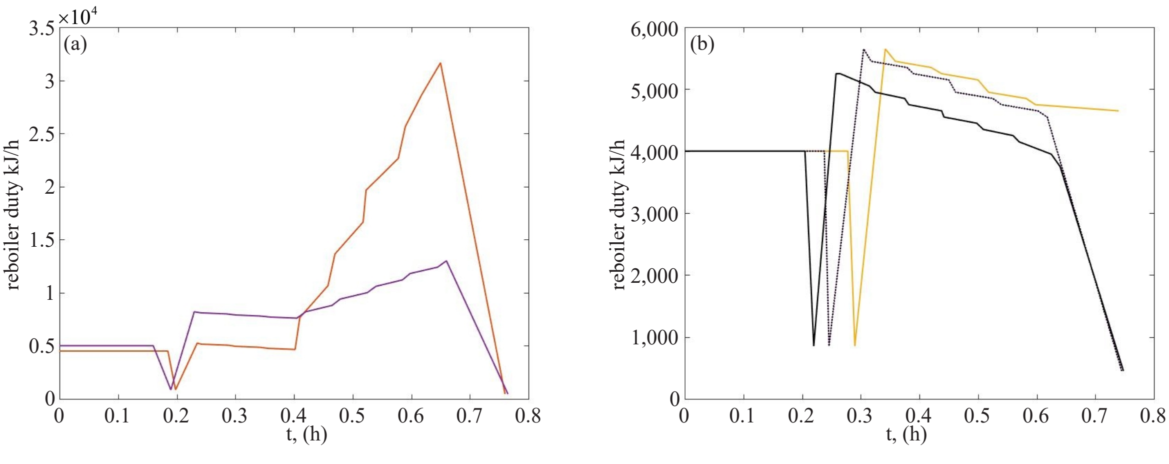

Figure 6. Optimal reboiler duty vs time for vapor boil-up parameter increase as: (a) V = [12, 14, 16] [mol/h], (b) V = [18, 20] [mol/h]

It should be noted, however, that the "optimal reboiler duty" is a discontinuous time trajectory and, due to this, could not be directly compared with the results obtained by Modla and Lang,[9] since they used the continual heating reboiler strategy over the total batch time. Despite this, one can perceive that for the same value of the fixed liquid division ratio, the average energy required was at least 8.25% higher according to Modla and Lang[9] compared to the process proposed in this study.

3.2.2 *The influence of reboiler volume*

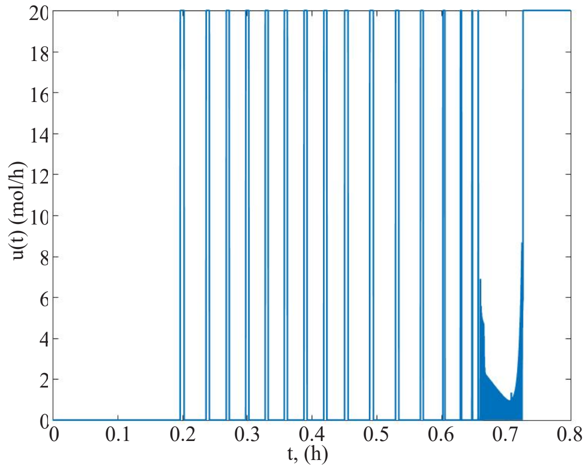

Figure 7. The optimal distillate rate $u(t)$ for the HP step

Table 7. Working conditions

Parameter	P [atm]	U_N^0 [l]	U_i^0 [mol]	V [mol/h]	t_f [h]	x_N^0	y^*
Predefined/initial value	HP:1	17.5271	0.1	20	0.8 1.6	0.1	LP: 0.75

Table 8. The optimal costs and recovery rates for acetone

Product recovered	N	U_0 (t_f)	Recovery rate	Discr. scheme, nb. of points
Acetone (HP)	24	2.9588	88.93% 92.89%	Gauss 3,200

Hereby, if the high-pressurized step, in particular, is "repeated" after its feed tank is filled with the additional charge, a more complex pattern is for the optimal control structure. In the second step, i.e., high-pressurized process, as supposed the initial arc matches with zero distillate rate (total reflux condition-stripper), but the final with maximal distillate rate. Further, it is observed that the optimal control trajectory is composed of almost all bang-bang arcs, with three consecutive singular arcs preceding the final maximal distillate arc. There are 30 commutations between different types of control, since the three instantaneous piques achieving maximum distillate rate, are not taken into consideration due to the extremely short period of duration. An interesting fact is that the singular arc (s) are 'positioned' again in the penultimate period, whereas here it lasts approximately 7% of the total operation time or 8 times shorter than in an LP step. The initial zero arc is of the longest duration, as it takes almost 40% of the operation time. In summary, the optimal control has 15 bang arcs, 15 zero arcs, and three singular arcs (Figure 7). The working conditions, and, optimal costs with recoveries, are tabulated in Table 7 and Table 8, respectively.

3.2.3 *The influence of working pressure*

Together with the influence of the reboiler volume, it is possible to examine the influence of high working pressure, ranging between 6 and 10 [atm], i.e., pressure gap. The observations are tabulated in Table 9, for lower [6, 7 (atm)] and intermediate pressure [8 (atm)], one case study is done, however, even two case studies are provided for the higher pressures [9, 10 (atm)], notably for different charges: (1) 17.5271 [l], (2) 13.95 [l]. Consequently, the standard increase of 20% led to a productivity increase of 4.27%/4.89%, respectively, for the same imposed high pressures of 9/10 [atm], respectively. All the more, keeping the first/second standard pressure constant, and increasing the higher pressure in the second part of the process for not more than 10%, brings an improvement of 16%/17.37%, respectively, in the yield. From the aspect of profit, one should consider the first option as a more profitable one. All optimal control policy patterns are "near-BB" or "BB" (one short and pre-ultimate singular arc). It is to be noted that all the calculations are done for the "single period" duration of the high-pressurized step, i.e., total batch time is set to a "single period" of time.

Table 9. The investigation of the influence of the pressure gap

Working pressure, P_2 [atm]	Distillate quantity, [mol] U_0 (t_f)	Recovery rate	Total number of cycles, n_c	Staring period duration, [h]
6	2.7890	91.08%	13	0.2020
7	2.6706	91.46%	12	0.2300
8	2.4165	92.27%	11	0.2780
9	2.4401	92.20%	11	0.2700
	2.5041	89.87%	12-13	0.2640
10	2.9588	88.93%	15	0.1880
	3.0391	87.74%	17	0.1860

* calculated per $t_f = 0.8$ h, "single-period"

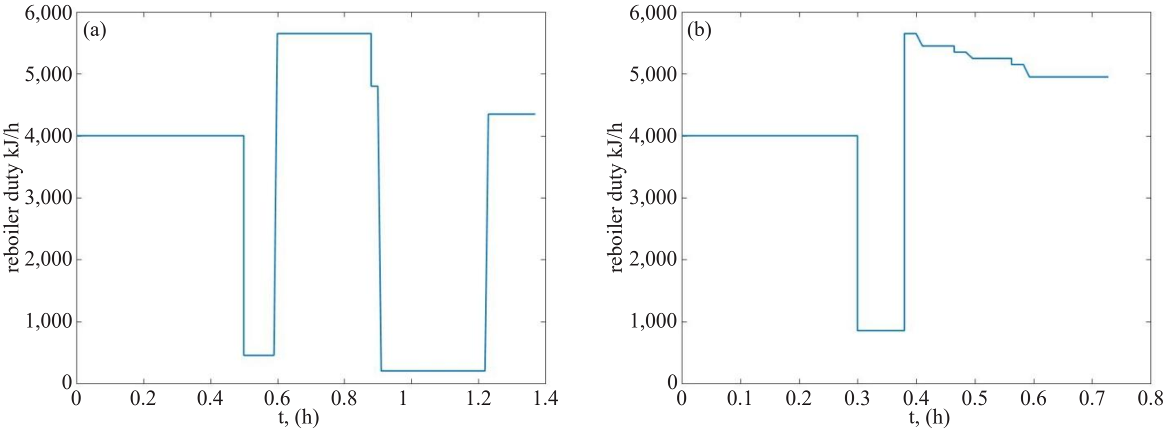

Figure 8. The optimal reboiler duty: (a) for LP step, (b) for HP step, for boil-up set at 11 [mol/h]

In this work, the number of strong assumptions on the fundamental dynamics of the column is reduced compared to those made in the previous studies by Zhao et al.[23] and Zhao et al.[24], as it relies on the well-defined dependence of the thermodynamic properties of the mixture from the temperature. However, to validate the results, ChemCad 6.1, the dynamical simulator is employed, whereas the same applied thermodynamic model. Hereby, without any further optimization, the interesting facts were retrieved for the change of heating energy during the respective process. In Figure 8, the optimal reboiler duty vs time function for all stages was presented for the LP step, however, also depicts the HP step: The extracted values for optimal reboiler duty vs time corresponding to the previously examined cases of boil-up set at 11 [mol/h]. Compared to the energy required to heat the reboiler of the batch rectifier, it takes at least 14.88 times more to start up the batch stripper, from here the minimum total energy requirement is to be calculated for the start-up of the proposed dual process: 60,400 kJ/h.

4. Conclusions

The optimal control of a mixed configuration, comprising the batch rectifier and batch stripper, is determined in terms of minimum distillate rate for a given amount of feed and quality. A rigorous model is built within the optimal control framework in Bocop 2.2.1. Additional conditions on vapor-liquid equilibrium, necessary to accurately describe high-pressure processes, are highlighted. Handled as thermodynamic constraints, some deeper insights into the dependence of the optimal control structure on pressure effects are reported. The patterns of heating energy demand over time during the process are developed: Compared to the previous research by Modla and Lang,[9] in a double-column batch stripper configuration, for the same fixed liquid division ratio, the average energy required was at least 8.25% higher than that found for the process proposed in this study. Additionally, the authors did not discuss productivity, whereas this study provides an overview of it. Moreover, investigating the influence of vapor boil-up on the optimal control pattern showed that for an increase in vapor boil-up by 2.85 times, the total number of cycles increased by more than five times, while the starting period duration decreased by 3 times. Furthermore, investigating the influence of the pressure gap on the optimal control structure showed that for an increase in the pressure gap of 44.45%, the total number of cycles increased by 23.53%, while the starting period duration decreased by 7.9%.

Given this, further research should focus on heat integration between different or the same units to achieve the most acceptable outcomes in terms of production, energy, and profit. Moreover, delving deeper into optimal control theory could extend ideas to newly established processes, such as multiple azeotropes treatment, unconventional configurations/modes, and unified operations. Future research should also explore the introduction of entrainers to investigate extractive column functioning and the recently presented entrainer-assisted pressure-swing distillation. Moreover, none of the heat-integrated configurations is considered here, which should be in the future: thermally integrated columns (DWC), internally integrated dividing-wall single columns (HIDIC), internally thermally coupled dividing wall rectifier (TCDTBR), bottom flashing vapor recompressed batch distillation (BDBF), etc.

Conflict of interest

I declare there is no conflict of interest and the entire responsibility for the published results data is up to the author.

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