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Investigating the processing capacity of batch distillation by applying a second, smaller column

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A R T I C L E I N F O

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A B S T R A C T

Increasing the processing capacity is important, especially in the pharmaceutical industry, where a given amount of waste solvent must be processed in a specific time period, else the remaining amount of waste solvent must be incinerated, which results in organic solvent loss and considerable environmental impact. In this paper, the possibility of increasing the processing capacity of batch distillation by using a second, already available, smaller batch column in addition to the original one is studied. The size of the columns is not changed during the calculations performed. Two case studies are presented. In the first one (Mixture 1), acetone, in the second one (Mixture 2), acetic acid must be recovered in high purity from their aqueous mixtures by operating either the larger column only or both columns simultaneously. The smaller column serves for preliminary separation, and its main-cut (Mixture 1) or residue (Mixture 2) is further processed in the larger one. The goal of this work is to maximise the processing capacity of the two-column process with the same recovery as in the single-column process. Both processes are modelled with a professional flow-sheet simulator. For the single-column process, the only independent variable is the reflux ratio. For the two-column process, the independent variables are the reflux ratios of both columns and the amount of intermediate product (distillate or residue) of the first column transferred to the second one. The ranges of the independent variables are determined by sensitivity studies for different charge compositions (20–50-80 mass% acetone or acetic acid), and each case is optimised by a genetic algorithm coupled to the flow-sheet simulator. It is also shown in this work that the processing capacity maximum of the two-column process is not always at the equality of the processing times of the two columns.

1. Introduction

Batch distillation is frequently applied in the fine chemical and pharmaceutical industry since it can be used for treating mixtures of variable quantity and composition [1]. The pharmaceutical plant can produce several final products, which generates different waste solvent mixtures. The organic components of these waste solvent mixtures shall be regenerated. If the processing capacity (Cap) of the regeneration process is insufficient (and the plant starts to produce another final product), the remaining amount of waste solvent must be incinerated. Incineration shall be avoided as it contributes to greenhouse gas emission. Meyer et al. [2] investigated three waste solvent treating alternatives (batch distillation coupled with two different incineration methods and with pervaporation) for isopropanol-water mixture, and showed that conventional waste solvent incineration has the highest environmental impact. Hence, increasing the processing capacity is crucial to make the whole process cleaner.

Several authors investigated the possibilities of increasing the capacity (or annual profit) of batch columns, which is equivalent to the minimum time problem as defined by Mujtaba [1] when a given amount of product shall be recovered with a specified purity. However, their main focus is usually the optimisation of the operation of a single existing column. Fidkowski [3] increased the capacity of an existing batch distillation process in which an industrial multicomponent mixture was separated by maximising the production rate. Because of the loose product specifications, it was possible to increase the capacity by changing the main pollutants of main and off-cuts. Yang and Tjia [4] studied the possibilities of increasing the capacity of an active pharmaceutical ingredient (API) purification batch column with much stricter purity specifications than those of Fidkowski [3]. The capacity of production was significantly increased by optimising the operation of the column (adjusting the reflux ratio, the heating jacket and condenser temperatures and the time of the total reflux period). Aqar et al. [5]

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Notations

Symbols

Cap	Processing capacity
OF	Objective function
R	Reflux ratio
SPF	Specific product flow rate
Δt	Total distillation time including the dead time
Δt_{dead}	Dead time including heating up, filling in and draining
$\overline{x}$	Average mole fraction
η	Recovery

Superscripts

1	Column 1
2	Column 2
S	Single column

Subscripts

A	More volatile component
B	Less volatile component
ch	Feed
D	Distillate
W	Residue

Abbreviations

GA	Genetic algorithm
VBA	Visual Basic for Applications

minimised the processing time of a batch reactive distillation process producing methyl esters. The optimised results were used to compare a novel reflux policy with the conventional batch reactive distillation process. The novel reflux policy, when part of the reflux is fed to the reboiler, proved to be better considering the ester yields.

Typically, the separation is performed in a single batch column, and only a few authors investigated batch distillation processes performed in two batch distillation columns. However, Cap can be increased by operating an additional batch column available in the plant. The main cut or still residue of the first column can be processed in the second one. During the operation of the second column, new batch(es) can be processed in the first one, and the two columns can be operated simultaneously. If the operation of the first column is fast enough, several batches can be processed, and the main cuts or residues can be united into a single charge of the second column. It is also worth uniting these cuts if their amount is low and the still volume of the second column is large.

In the previous work of the authors [6], the two-column process described above was already investigated for a ternary mixture (dichloromethane-acetone–water) based on an industrial separation problem. The main objective was to minimise the specific energy demand of the process. The processing capacity significantly increased compared to the single-column process, so the present work focuses on maximising the capacity of batch distillation processes by using a two-column process. (The recovery of acetone (η) from the ternary mixture was not kept constant in the work of Nemeth et al. [6].) When the capacity of an existing column is already optimised, the simplest way of increasing the capacity is to construct a similar column. However, this solution requires considerable capital investment. Another option investigated in this work is to use another smaller column already available in the plant for preliminary separation. Pre-fractionation is commonly used in continuous distillation processes, especially for multicomponent mixtures, which later also gave the idea of the dividing-wall columns [7].

Aqar et al. [8] introduced a reactive semi-batch distillation process for producing methyl lactate consisting of two columns. The reaction of methanol (continuously fed to the column) and lactic acid takes place in the first column, whose operating time is minimised (for a desired amount of product and purity). The second column is used for recovering methanol from the distillate of the first column. The amount of the methanol recovered under a specified time (which equals to the optimal time of the first column) is maximised. If the methanol recovery is considered as an essential part of the methyl lactate synthesis process, using an individual column for that can indirectly increase the production capacity of the synthesis through operating the two columns parallel. However, the second column serves only for reactant recovery and not for increasing the processed amount of feed. In our work, both columns are necessary for the treatment of feed and serve for increasing the processing capacity: the first column is handled as a pre-fractionator and the second one as the main separator.

Lara-Montaño et al. [9] investigated a two-column process for the purification of bioethanol by simulation and experiment. A continuous pre-fractionation column was applied before the final purification by batch extractive distillation. The authors noted that the final product contained several impurities, which might be removed by a conventional batch distillation step prior to the extractive distillation. In this process, different columns are used for each distillation step, but the processing capacity was not studied.

Zhao et al. [10] used two batch distillation columns for the separation of ethanol – 1-propanol. Both columns had a top and a bottom vessel. The two columns were connected by liquid exchange between these vessels. The authors performed calculations and experiments with different feed compositions. The vessel in which the feed must be charged is the one whose composition is similar to that of the feed. When the product specification is fulfilled in either vessel, it is drained, liquid exchange is performed, and a new charge is fed to the empty vessel. The goal of this work was not to increase the processing capacity, but to study a new operational strategy: the liquid exchange between the two columns. The authors noted that significant time saving and better separation efficiency can be reached with this new operational strategy compared to the conventional closed operation mode. As the recovery increased and the total batch time decreased, the capacity of the process increased.

Zhao et al. [11] also investigated a double-column batch distillation process where the columns are interconnected for the separation of n-hexane/ethanol/butanone including two distillation steps. Three different column configurations were studied by minimising the total annual cost and by calculating the global warming potential for each case. For the first ditillation step a single-column distillation was also investigated. Only the operating pressures of the columns were considered optimisation variables in their work. The double-column configuration resulted in the lowest total annual cost compared to the other cases investigated, including the single-column process for the first distillation step.

Adi and Chang [12] studied the scheduling of a two-column batch heteroazeotropic distillation system. Two mixtures (acetone-chloroform and acetone-chloroform-benzene) were studied, the entrainer was ethanol, and the final product was acetone in both cases. The distillation process was not modelled rigorously but by using only simple material balance equations. Several case studies were investigated for optimal scheduling. The objective for the scheduling optimisation was to maximise the overall profit of a production campaign (e.g. total revenue minus the sum of raw-material costs). The authors concluded that equipment sharing for distillation tasks causes a significant decrease in the overall profit.

In the studies summarised above, the two distillation columns are only connected through liquid transfer: the product of the first column is charged to the still of the second column. Another possibility is to physically connect the two columns by applying a common reboiler or a reflux drum, thereby obtaining double-column system rectifiers or strippers. Denes et al. [13] studied the separation of isopropanol-water by heteroazeotropic distillation using cyclohexane and n-hexane as entrainer in a double-column batch stripper, where the two columns were connected by a common reflux drum (decanter). In their study, two configurations of the double-column system were optimised and compared. The objective was to minimise the duration of the process, which also means the maximisation of processing capacity. The performance of the two entrainers was also compared.

Modla and Lang [14] studied the feasibility of single- and double-column batch pressure-swing distillation processes for a minimum- (ethanol-toluene) and a maximum-boiling (water-ethylenediamine) azeotrope. Both for the single- and double-column processes, batch rectifier and stripper configurations were also studied. For the double-column configurations, the two columns were connected through a common reflux drum (for the stripper configuration) and a common reboiler (for the rectifier configuration). The double-column systems performed better than the single column considering the specific energy demand and product purity. The double-column batch rectifier and stripper systems were further studied by Modla [15] in open and closed operation mode for the separation of binary mixtures: n-pentane-acetone (forming a minimum-boiling azeotrope) and water-ethylenediamine (forming a maximum-boiling azeotrope). The closed operation mode proved to be economically better for both mixtures because of the lower specific energy demand. Li et al. [16] separated a mixture of methanol, tetrahydrofuran and water by pressure-swing distillation, as well. Two-column configurations were investigated in their work: a double-column batch stripper and a triple-column process. In both processes, the high- and low-pressure columns were connected by a common reflux drum. In the triple-column process, tetrahydrofuran was obtained as the bottom product of the high-pressure column, while water was obtained as the bottom product of both the low-pressure and the third column. The bottom product of the low-pressure column was sent to the third column for further processing when its methanol concentration increased. By adding a third column to the process, the purity of the methanol product increased to 99.9 mol%, which was a higher grade product and required no more purification. Wang et al. [17] separated n-heptane and isobutanol by applying a batch rectifier and batch stripper, operating at different pressures. The two columns were connected through a common vessel, which was the reboiler of the high-pressure column and the reflux drum of the low-pressure column. The authors performed multiobjective optimisation to minimise the total annual cost and the CO_2 emission. Both columns were newly designed, so the numbers of theoretical plates were included as optimisation variables.

Kim et al. [18] performed laboratory experiments in which two batch reactive distillation columns were used for the recovery of high purity lactic acid. The esterification of the lactic acid was performed in the reboiler of the first column, while the hydrolysis of the methyl lactate was performed in the second column. The distillate (methanol) of both columns was recycled to the reboiler of the esterification column. The application of the second column improved the yield of the whole process because a higher amount of methanol was recycled from the second column to the first one.

In this work, the two-column batch distillation system, which is the same as in our previous work [6] is studied further from the aspect of increasing the processing capacity. The separation of two mixtures is studied and modelled rigorously: acetone (A)-water (B) (Mixture 1) and water (A)-acetic acid (B) (Mixture 2) are separated at atmospheric pressure by using either a single batch column or two batch columns of different sizes simultaneously. Both acetone and acetic acid must be recovered in a purity of 99.7 mass%. The goal is to investigate by simulation and optimisation whether higher Cap can be reached with the two-column process than with the single-column one keeping the same recovery (η). The increase of Cap (or equivalently the minimising of distillation time) of batch distillation was not yet studied in the literature for existing, already operating two-column distillation systems. Furthermore, the effects of the operating parameters and charge composition on the processing capacity were not yet investigated for a two-column batch system. The effects of the reflux ratios (R^1 and R^2) of the columns and the stopping criterion of the first, smaller column, which is the average distillate composition ($\overline{x}^1_{D,A}$ for Mixture 1 and $\overline{x}^1_{D,B}$ for Mixture 2), are investigated on the whole process for different charge compositions (x_{Ch} = 20 – 50 – 80 mass% acetone or acetic acid) by sensitivity studies. The Cap of the two-column process is maximised for all charge compositions by using a Genetic Algorithm (GA) coupled to the professional flow-sheet simulator ChemCAD.

2. Vapour-liquid equilibrium

Water forms a tangent azeotrope with acetone at high acetone concentrations (Fig. 1a) and with acetic acid at high water concentrations (Fig. 1b).

For both mixtures the specified product (organic component) purity is 99.7 mass%, which equals 99.0 mol% for Mixture 1 and 99.9 mol% for Mixture 2. The UNIQUAC model was used for describing the vapour-liquid equilibrium of Mixture 1. The binary interaction parameters (Table 1) for the acetone–water pair were selected [19] to obtain the best agreement between the measured and calculated data [6]. For Mixture 2, the NRTL model was used to describe the vapour-liquid equilibrium (Table 1).

3. Calculation methods and process data

Section 3.1 describes the separation process in detail for the single- and two-column processes. The sensitivity studies performed with ChemCAD and the method of optimisation are described in Section 3.2.

3.1. Description of the separation processes

First, the separation is performed in a single column, which provides 20 theoretical stages (excluding the reboiler and the total condenser). The reboiler heat duty is kept constant: 720 MJ/h, and it is assumed that the capacity of the condenser and that of the column are always sufficient. The pressure drop of the column is 0.4 kPa, the condenser hold-up is 5 dm^3, and the column hold-up is 3 dm^3/plate; the volume of the charge is 10 m^3. The desired product (acetone or acetic acid) in high purity (99.7 mass%) is obtained as distillate for Mixture 1 and as still residue for Mixture 2, while water remains in the still for Mixture 1, or it is obtained in the accumulator for Mixture 2. Only one distillation step is performed for the single-column process.

The use of two columns is investigated to increase Cap: two batch distillation columns are operated sequentially from the point of view of feeding. The first column (Column 1) is the smaller one (providing 3 theoretical stages (without the reboiler and total condenser)), and the second (Column 2) is the larger one (also used in the single-column case). The volume of the charge of Column 1 is 5 m^3 (25 °C), and the maximum allowable amount of liquid in the reboiler of Column 2 is 10 m^3 (which is equal to the charge volume for the single-column process). The reboiler heat duty of Column 1 is fixed at 360 MJ/h, and again, it is assumed that the capacity of the condenser and that of the column are always sufficient for both columns. The pressure drop of Column 1 is 0.3 kPa, the condenser hold-up is 5 dm^3, and the column hold-up is 3 dm^3/plate. For both processes, a constraint was considered for the amount of still residue in the larger column, which should be minimum 1 m^3 (Volumetric Constraint). The State-Task Network for both mixtures is shown in Fig. 2.

For Mixture 1, the distillate of Column 1 is the charge of Column 2, and the desired product is obtained as the distillate of Column 2. For Mixture 2, the still residue of Column 1 is the charge of Column 2, and the product is obtained as the still residue of Column 2. The distillate (for Mixture 1) or the residue (for Mixture 2) of Column 1 is always off-specification; further purification is required in Column 2. In both columns, only one distillation step is performed.

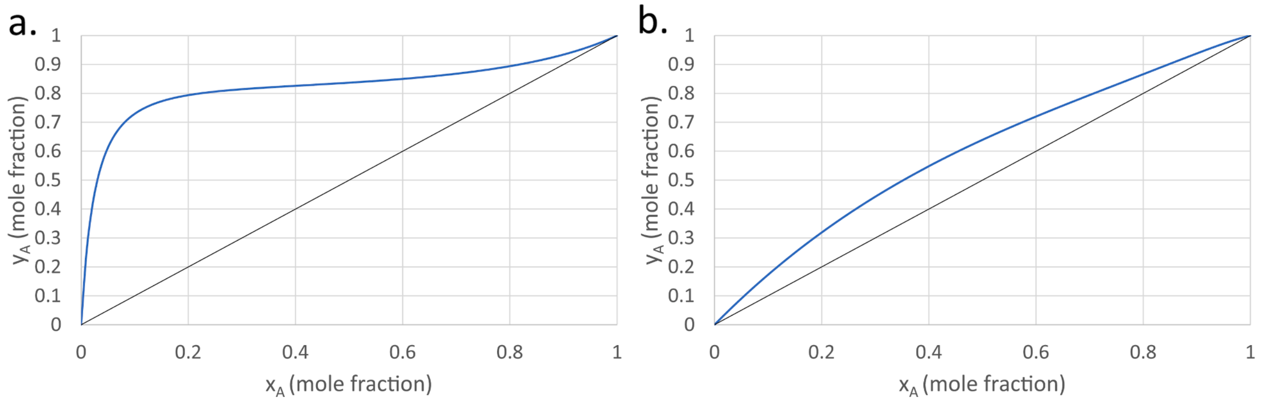

Fig. 1. Calculated vapour-liquid equilibrium curves: a. for acetone–water (Mixture 1); b. for water-acetic acid (Mixture 2).

Table 1
Binary interaction parameters for the UNIQUAC (Mixture 1) and NRTL (Mixture 2) models.

Component i	Component j	U_{ij}-U_{jj}, cal/mol	U_{ji}-U_{ii}, cal/mol		Source
Acetone	Water	704.698	−110.5286		Brunjes and Bogart [7]
		B_{ij}, K	B_{ji}, K	α	
Water	Acetic acid	424.019	−110.568	0.2997	ChemCAD built-in

When the acetone or acetic acid concentration is low in the feed, the distillate or residue of Column 1 is collected from several (n) batches until it is worth starting the larger column (Column 2). In a campaign mode, the two columns can operate simultaneously, which increases the processing capacity. However, a storage tank is required as a buffer: the distillate or residue of Column 1 is collected from several batches, while Column 2 processes its previous feed. Fig. 2 shows the case when two charges (Feed 1/a and 1/b) are processed in series in Column 1.

3.2. Calculation method

The CHEMCAD model of the two-column process is presented in Fig. 3. Unit 1 is the smaller and 5 is the larger column (BATCH COLUMN module of ChemCAD, using equilibrium tray model). Units 2 and 6 are the distillate tanks (accumulators). Units 3, 4 and 7 are "SREF" units. SREF 4 and 7 create reference streams from the bottom of the smaller and larger columns, respectively, to track the composition of the residue. SREF 3 transfers the content of tank 2 to the larger column in the case of

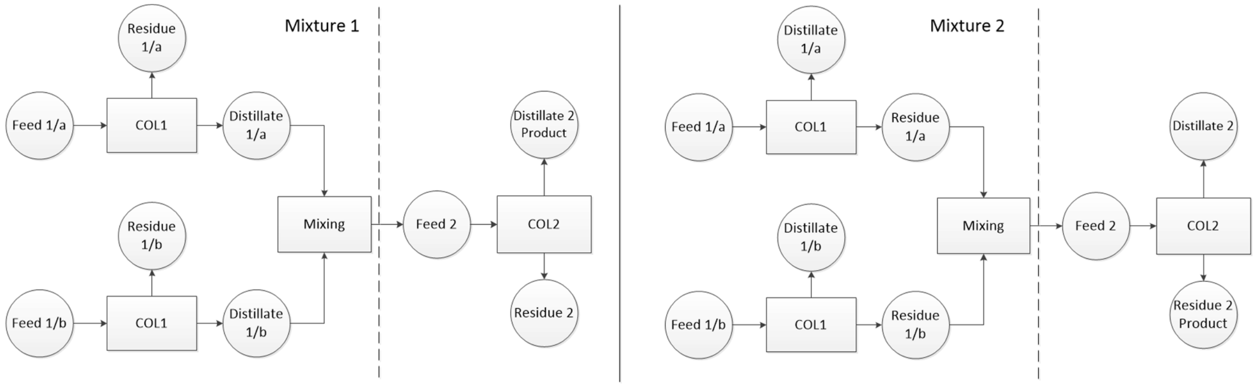

Fig. 2. State-Task Network for both mixtures, when n = 2 (Feed 'a' and 'b' processed in series in Column 1).

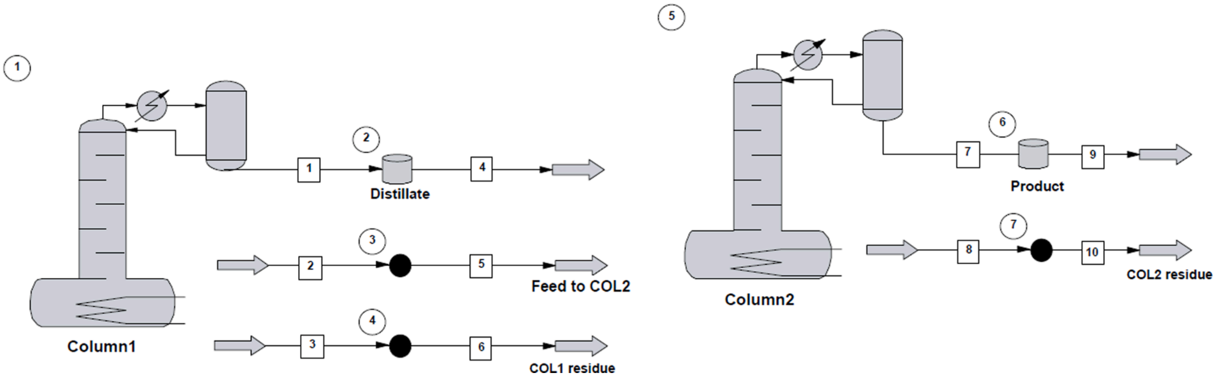

Fig. 3. ChemCAD flow-sheet of the process.

Mixture 1 and the residue of Column 1 in the case of Mixture 2. This unit makes it possible to simulate the two batch columns together.

The reflux ratios of both columns (R^1 for Column 1 and R^2 for Column 2) and for two columns, the stopping criterion of Column 1, which is the average distillate composition ($\overline{x}^1_{D,A}$ is the acetone, $\overline{x}^1_{D,B}$ is the acetic acid concentration in the distillate tank) are independent variables. For the single-column process, the value of the single independent variable (reflux ratio: R^S) is determined by considering the recovery (η) and specific product flow rate (SPF). SPF is the amount of acetone or acetic acid in the product divided by the total process time: Δt, which is the sum of the distillation time and the dead time (Δt_{dead}: including filling-in, heating-up and discharge). The Cap, which is the mass of feed processed per unit time, is calculated for each charge composition.

For the two-column process, an additional constraint is given: η must be equal (or higher) than for the single-column separation (Recovery Constraint). Here, Cap is calculated by Eq. (1):

$$\mathrm{Cap} = \frac{n \bullet m_0}{\max(n \bullet \Delta t^1; \Delta t^2)} \tag{1}$$

where n denotes the number of charges processed in Column 1; for Mixture 1: n = 6 for $x_{Ch} = 20$ mass%, n = 3 for $x_{Ch} = 50\%$ and n = 2 for $x_{Ch} = 80\%$ and for Mixture 2: n = 2 for all investigated charge compositions. m_0 is the mass of charge of Column 1, Δt^1 and Δt^2 are the total distillation times of Column 1 and 2, respectively. The dead times: ${\Delta t^1}_{dead} = 3.5$ h for Column 1 and ${\Delta t^2}_{dead} = 1.6$ h for Column 2 (data are taken from an industrial example).

From Eq. (1), it can be assumed that the maximum of Cap will be where $\Delta t^2 = n \cdot \Delta t^1$. This will be referred to as the time equality hypothesis. The underlying assumption is that decreasing the higher one of the two distillation times increases the other one, therefore in the optimum, these times must be equal.

First, sensitivity studies are performed for all cases and for both mixtures to study the effects of the independent variables on the η and Cap. The values of the variables giving the highest Cap with both constraints fulfilled are determined. The lower and upper bounds of each operation parameter are also determined for optimisation (Table 2). Stopping criterion means $\overline{x}^1_{D,A}$ for Mixture 1 and $\overline{x}^1_{D,B}$ for Mixture 2 except for Case 3, where it refers to $\overline{x}^1_{W,B}$.

Optimisation of the two-column process is performed by a real-coded, elitist GA, written in VBA under Excel. For the evaluation of the fitness of each individual (which is the objective function: OF), first, the values of the optimisation parameters are sent to ChemCAD, then the simulation by using the flow-sheet shown in Fig. 3 is performed. Finally, the results are sent back to Excel, where the value of the OF is calculated. The parameters of the GA: mutation probability: 5 %, crossover probability: 70 %, population size: 30, number of generations: 100. The OF maximised is the Cap (Eq. (1)) with both the Volumetric and Recovery Constraints, which are taken into account by a penalty function (the value of OF will be 0 t/day when any of the constraints are not fulfilled).

4. Simulation results

First, sensitivity studies were performed both for Mixtures 1 and 2. Three different feed compositions were investigated: $x_{Ch} = 20$, 50 and 80 mass% acetone or acetic acid. In the case of the two-column process for Mixture 1, simple batch distillation was also studied using Column 1 ($R^1 = 0$) because the calculated R^1 values to reach the highest Cap were considerably low ($R^1 = 0.01$).

4.1. Single-column process

First, the separation was performed using only the larger column (Column 2). Both η of the desired component and the SPF were calculated at different R^S values for all investigated charge compositions. The main results of the single-column process are summarised in Table 3.

Mixture 1

For Mixture 1, there is a sharp increase in η for all charge compositions around $R^S = 4$ (Fig. 4a). $R^S = 5$ was chosen for all studied compositions considering both SPF and η. The higher x_{Ch}, the lower is the R^S necessary for the separation. With increasing R^S, SPF is decreasing because the effect of the increase in Δt is higher than that in η. However, with increasing x_{Ch}, slightly higher SPF can be reached. Based on these single-column results, the values of the Recovery Constraint were specified for the two-column system. The following constraints were given for $x_{Ch} = 20$, 50 and 80 mass%: η = 94.50% (1,804.7 kg acetone in product), η = 98.08% (4,412.5 kg) and η = 99.92% (6,671.2 kg). On the increase of x_{Ch}, Cap decreased significantly: the more acetone must be distilled, the longer time it takes. Besides, there is a slight effect of the density: that of the mixture of 20% is the highest, which means that the highest amount (in mass) of mixture is processed.

Mixture 2

Contrary to Mixture 1, there is no sharp change in either η or SPF (Fig. 4b) as the function of R^S. SPF has a maximum at lower R^S (between 3.5 and 4) for all compositions studied, but η is very low. η does not change significantly with the charge composition. The charge composition has a significant impact on Cap, which diminishes on the decrease of acetic acid concentration. Based on these results for Mixture 2, the following values of the Recovery Constraint were specified for the two-column process for $x_{Ch} = 20$, 50 and 80 mass%: η = 93.00% (1,879.2 kg acetic acid in product), η = 93.73% (4,810.1 kg) and η = 94.17% (8,757.0 kg).

4.2. Two-column process

In the following sections, Case 1: $x_{Ch} = 20$ mass%, Case 2: $x_{Ch} = 50\%$ and Case 3: $x_{Ch} = 80\%$ acetone or acetic acid were studied for the two-column process. As the minimal volume of the still residue of Column 2 was specified, several batches must be processed in Column 1 to fulfil

Table 3
Main results of the single-column process.

	Mixture 1			Mixture 2		
	x_{Ch}, mass%			x_{Ch}, mass%		
	20	50	80	20	50	80
R^S	5	5	5	10	12	13
Δt^S, h	10.4	21.5	30.75	281.71	213.01	96.05
η, %	94.50	99.08	99.92	93.00	93.73	94.17
Cap, t/day	21.99	9.91	6.50	0.86	1.16	2.60
SPF, t/day	4.16	4.91	5.19	0.16	0.54	1.96

Table 2
Lower and upper bounds for each optimisation variable.

	Mixture 1			Mixture 2		
	Case 1	Case 2	Case 3	Case 1	Case 2	Case 3
R^1	0.01–1.00	0.0008–0.05	0.01–0.10	7–13	5–12	13–20
Stopping criterion of Column 1	0.70–0.85	0.82–0.89	0.84–0.87	0.01–0.05	0.10–0.20	0.80–0.82
R^2	3–5	3–5	3–5	10–17	10–15	11–18

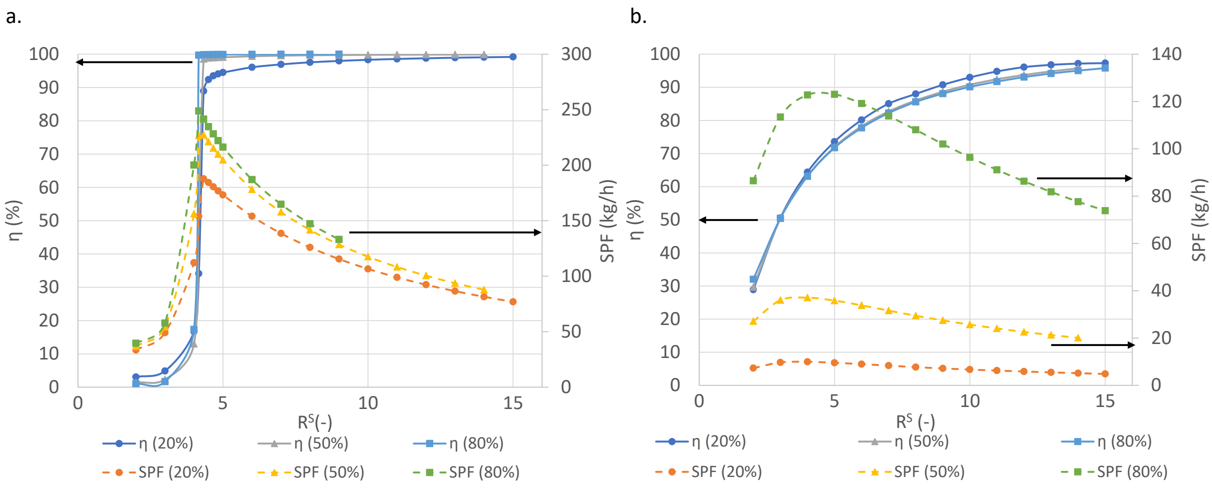

Fig. 4. η and SPF as the function of R for different charge compositions (single-column process); a. Mixture 1; b. Mixture 2.

the Volumetric Constraint. The number of batches processed in Column 1 is the value by which the volume of distillate (for Mixture 1) or residue (for Mixture 2) is the closest to the maximum allowable liquid volume in the reboiler of Column 2 (10 m^3) but does not exceed it. Since η is kept constant, SPF is proportional to Cap.

Mixture 1
Case 1

For the lowest acetone content of the feed, Cap is much lower than for the single-column process because of the Volumetric Constraint. The amount of the distillate of Column 1 is low with low water content, so the Volumetric Constraint cannot be fulfilled without high acetone loss in the residue of Column 2. Hence, at least 6 charges must be processed in Column 1. The dead time of the 6 charges in Column 1 (21 h) is too high to reach a higher Cap than by the single-column process. If fewer charges were processed in Column 1, the Recovery Constraint would not be fulfilled because too much acetone remains in the 1 m^3 residue of Column 2. The calculation was repeated by considering no dead time in both columns to check the influence of this parameter. With no dead time, the dominating time determining Cap is still the distillation time of Column 1, and the Cap of the two-column process is still lower than that of the single-column one. As the results considering Cap are much worse than that of the single-column process, the detailed results are not given for this case, but the main results are summarised in Table 4. However, by 4–5 % higher η can be reached with the two-column process than with the single-column one at $R^2 = R^S = 5$. This higher η can only be reached with the single-column process at $R^S \geq 10$, which makes the single-column process much slower. In these circumstances, the Cap of the two-column process would be by 5–8 % higher than that of the single-column one.

Case 2

In Fig. 5a, the processing capacities are shown as the function of $\overline{x}^1_{D,A}$ at several R^1 values. Three batches were processed in Column 1, while one in Column 2. On the increase of R^1, Cap decreases, but on the increase of $\overline{x}^1_{D,A}$, Cap becomes significantly higher. However, η is insufficient at higher $\overline{x}^1_{D,A}$ values. The “Recovery Constraint” curve shows those points above which the Recovery Constraint is not fulfilled (Fig. 5a). The horizontal line shows Cap of the single-column process. This means that the two-column process is better than the single-column one in the shaded area between the horizontal line and the “Recovery Constraint” curve. At higher R^1 values, neither η nor Cap is sufficient. At lower R^1 values with higher $\overline{x}^1_{D,A}$, the Recovery Constraint cannot be fulfilled – an insufficient amount of water (with acetone in high concentration) is transferred to the reboiler of Column 2 from Column 1, so the Volumetric Constraint can only be fulfilled with too much acetone remaining in the still of Column 2. The highest Cap is obtained at $R^1 = 0.01$ and $\overline{x}^1_{D,A} = 86$ mass%. Close to this point, the equality of the distillation times ($3 \bullet \Delta t^1 = \Delta t^2$) can be reached. At this point, Cap (10.50 t/day) is by ca. 6 % higher than for the single-column process. However, Δt^2, which determines Cap, can be slightly decreased by decreasing R^2.

In Fig. 5b, η is shown as the function of $\overline{x}^1_{D,A}$ for different R^1 values. The horizontal line shows η of the single-column process. On the decrease of R^1, Δt^1 diminishes at fixed $\overline{x}^1_{D,A}$, but so does the amount of acetone sent to Column 2 (the acetone content of the distillate decreases fast to a low value), which leads to a low η. The latter can be increased by decreasing $\overline{x}^1_{D,A}$, thereby transferring more acetone to Column 2. However, the water content of the distillate becomes high very fast, so it takes too much time to collect enough acetone in the accumulator of

Table 4
Main results for the two-column process.

	Mixture 1			Mixture 2		
	Case 1	Case 2	Case 3	Case 1	Case 2	Case 3
Operation parameters	$R^1 = 0.01$ $\overline{x}^1_{D,A} = 0.73$ $R^2 = 5.0$	$R^1 = 0.01$ $\overline{x}^1_{D,A} = 0.86$ $R^2 = 4.1$	$R^1 = 0.01$ $\overline{x}^1_{D,A} = 0.86$ $R^2 = 4.3$	$R^1 = 10$ $\overline{x}^1_{D,B} = 0.03$ $R^2 = 14$	$R^1 = 9$ $\overline{x}^1_{D,B} = 0.139$ $R^2 = 12.3$	$R^1 = 18$ $\overline{x}^1_{W,B} = 0.809$ $R^2 = 14.2$
n	6	3	2	2	2	2
$n \bullet \Delta t^1 / \Delta t^2$, h	48.42 / 25.53	29.67 / 29.65	24.24 / 27.43	193.00 / 255.21	10.10 / 214.90	23.20 / 96.90
η, %	95.10 (100.6 %)	99.21 (100.1 %)	99.92 (100 %)	93.03 (100 %)	93.76 (100 %)	94.19 (100 %)
Cap, t/day	14.16 (64.4 %)	10.77 (108.6 %)	7.29 (112.1 %)	0.96 (111.7 %)	1.14 (98.8 %)	2.59 (99.4 %)

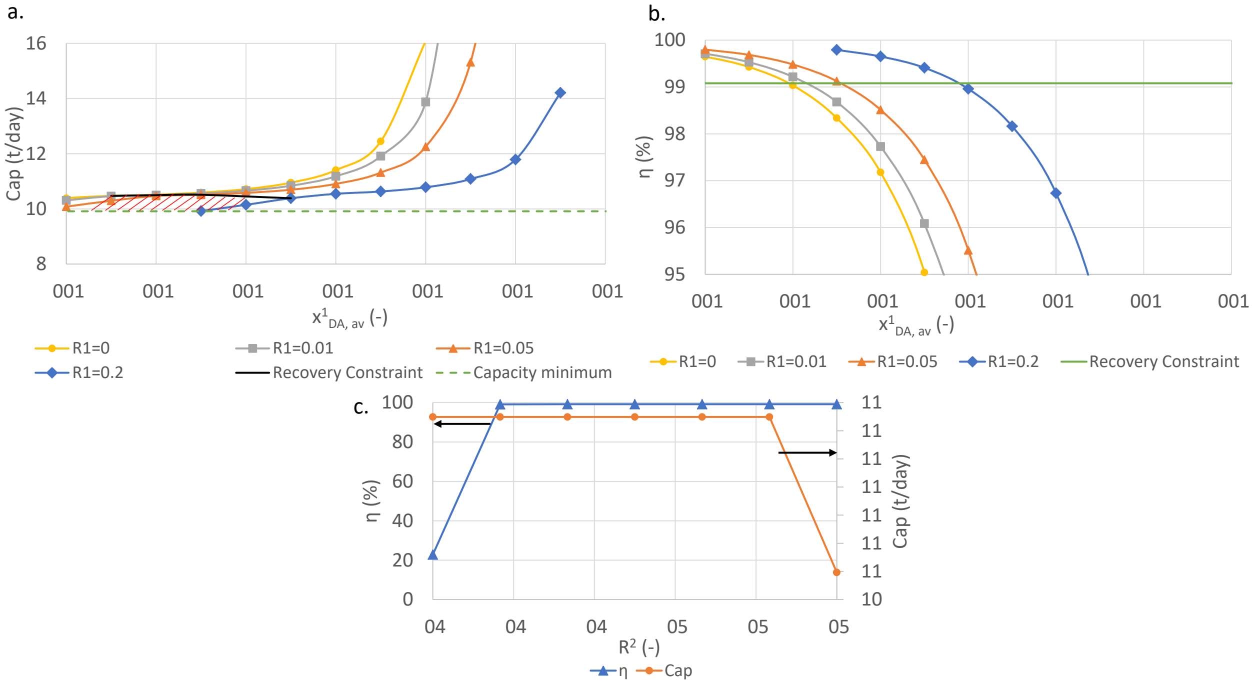

Fig. 5. Results for x_{Ch} = 50 mass%. a. Cap as the function of $\bar{x}^1_{D,A}$ for different R^1 values ($R^2 = 5$); b. η as the function of $\bar{x}^1_{D,A}$ at different R^1 values ($R^2 = 5$); c. η and Cap as the function of R^2 ($R^1 = 0.01$ and$\bar{x}^1_{D,A} = 0.86$).

Column 1 to reach the specified η in Column 2.

In Fig. 5c, η and Cap are shown as the function of R^2. The time equality is fulfilled at $R^2 = 4.83$. Cap is constant below $R^2 = 4.83$, where Column 1 determines Cap. Below $R^2 = 4.17$, η is too low. At this point and below it, Cap increased to 10.77 t/day, which is by 8.6 % higher than for the single-column process. However, R^2 can be further decreased to 4.17 (in order to decrease the energy demand) as the Recovery Constraint is still satisfied at this reflux ratio.

By using simple batch distillation instead of Column 1, if the Recovery Constraint is fulfilled, almost the same Cap (Fig. 5a), but lower η (Fig. 5b) is reached than with the smaller column with low R^1 (0.01).

Case 3

The processing capacities are shown as the function of $\bar{x}^1_{D,A}$ for several R^1 values in Fig. 6a. Two batches were processed in Column 1,

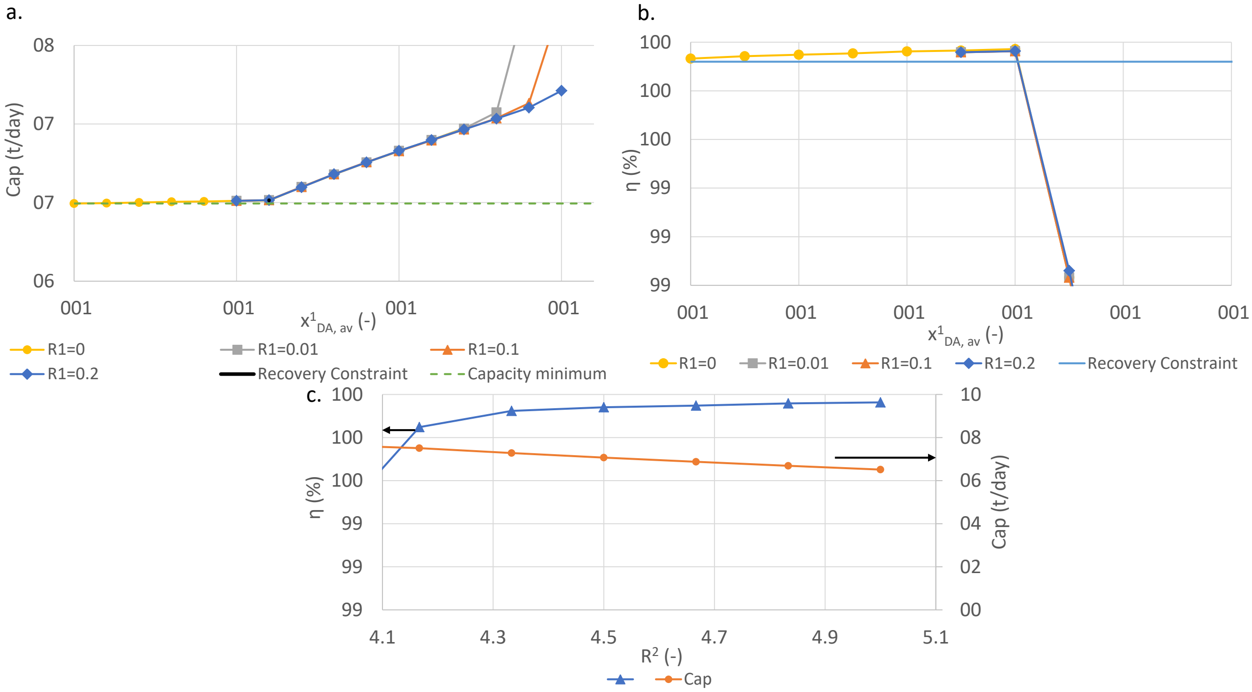

Fig. 6. Results for x_{Ch} = 80 mass%. a. Cap as the function of $\bar{x}^1_{D,A}$ for different R^1 ($R^2 = 5$); b. η as the function of $\bar{x}^1_{D,A}$ for different R^1 ($R^2 = 5$); c. η and Cap as the function of R^2 ($\bar{x}^1_{D,A} = 0.86$).

while one in Column 2. Cap is independent of R^1 below $\overline{x}^1_{D,A} = 0.92$ as it is Δt^2 which determines Cap. The time equality was not fulfilled at the conditions investigated, but it might be fulfilled with higher R^1 values at the cost of higher energy demand.

Below $\overline{x}^1_{D,A} = 0.86$, the Recovery Constraint is fulfilled (Fig. 6a, the "Recovery Constraint" curve is very short). Above this point, Cap starts to increase, but at the same time, η starts to decrease drastically because of the Volumetric Constraint. At higher $\overline{x}^1_{D,A}$ values, there is not enough water in the distillate of Column 1 to satisfy the Recovery Constraint, and too much acetone remains in the residue of Column 2, which causes a significant decrease in η. The highest Cap (6.51 t/day) was reached for $R^1 = 0.01$ at $\overline{x}^1_{D,A} = 0.86$. At this point, Cap is by about 0.3 % higher than for the single-column process. However, η is also higher (99.96 % compared to 99.92 %), so R^2 can be decreased, leading to an increase in Cap as Δt^2 determines Cap.

In Fig. 6c, η and Cap are shown as the function of R^2. For determining R^2, the stopping criterion of Column 2 was changed to 1 m^3 of still residue and the product purity was treated as a constraint. On the increase of R^2, Cap decreases and η increases. The time equality would be fulfilled at $R^2 = 3.6$, but in this case, the purity of the product would be too low (99.5–99.6 mass%). At $R^2 = 4.3$, the specified purity is reached. At this point, Cap (7.29 t/day) is by 12.1 % higher than for the single-column process. At $R^2 < 3.6$, Cap is constant as Δt^1 is the dominating one.

In this case, by using simple batch distillation, the same η and Cap (Fig. 6a-b) are obtained as by using Column 1 with low reflux ratios.

Mixture 2

The same sensitivity analysis was performed as for Mixture 1. In all cases, two charges are processed in Column 1, while only one in Column 2. The Volumetric Constraint is always fulfilled as the product is obtained in the still.

Case 1

On the increase of R^1, η decreases rapidly at $\overline{x}^1_{D,B} > 0.06$, while Cap increases (Fig. 7a). At $\overline{x}^1_{D,B} = 0.03$ the Recovery Constraint is not satisfied for any R^1 studied at $R^2 = 10$, although Cap has a maximum at $R^1 = 10$. By increasing R^2, η can be increased, while Cap is decreasing (Fig. 7b). The points above both minimum lines in Fig. 7b are better than the single-column process. These points are above $R^1 = 9$ and $R^2 = 13$. The best results are obtained at $R^1 = 10$, $R^2 = 14$ and $\overline{x}^1_{D,B} = 0.03$: Cap (0.96 t/day) is by 11.7% higher than for the single-column process. The time equality could be fulfilled at higher R^1 values, but Cap would not increase as Δt^2 determines Cap. This will apply to Cases 2 and 3, too.

Case 2

Considering R^1 and $\overline{x}^1_{D,B}$, the same tendencies can be observed as in Case 1, but the recoveries are much lower than that of the single-column process (Fig. 8a). R^2 has the highest effect both on η and Cap: η increases and Cap decreases on the increase of R^2 (Fig. 8b.). With the sensitivity study, it was not possible to reach a higher Cap than that of the single-column process: either η or Cap was lower than the specified minimum. (However, a better solution is expected between $R^2 = 12$–13. This will be studied by optimisation in Section 4.3.) The highest Cap (1.14 t/day) reached with the specified minimum η is by 1.2 % lower compared to that of the single-column process.

Case 3

For the highest acetic acid concentration in the feed, the stopping criterion of Column 1 ($\overline{x}^1_{D,B}$) was changed to the acetic acid concentration in the residue $\left(\overline{x}^1_{W,B}\right)$ because of practical reasons. R^1 has no significant effect either on η or on Cap (Fig. 9a). The reason for this is that the number of theoretical stages is low in Column 1. By increasing R^2, the minimum η can be reached, but Cap decreases to a lower value than that of the single-column process (Fig. 9b). The highest Cap reached with the specified minimum η is by 0.6 % lower than that of the single-column process.

Summary of the main results of sensitivity studies

By using two columns for the separation of Mixture 1, equal or higher η can be reached for all x_{Ch} studied for the same Cap. Moreover, for Cases 2 and 3, Cap is also higher for the same η. For Mixture 2, a higher Cap for the same η can only be reached in Case 1. The main results are summarised in Table 4, where the results of the single-column process are taken as 100 %.

4.3. *Results of optimisation*

Mixture 1

Case 1 was not optimised as Cap is much lower (by the results of the sensitivity study) than for the single-column process because of the high number of batches to be processed. Case 2 was optimised by GA and Case

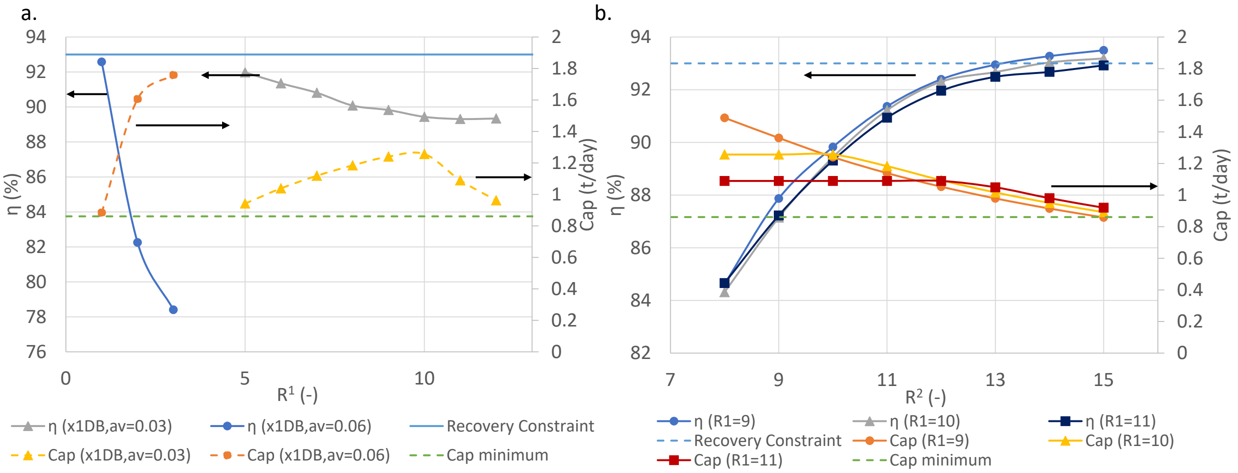

Fig. 7. Results for $x_{Ch} = 20$ mass%. a. Effect of R^1 for different $\overline{x}^1_{D,B}$ values ($R^2 = 10$). b. Effect of R^2 for different R^1 values ($\overline{x}^1_{D,B} = 0.03$).

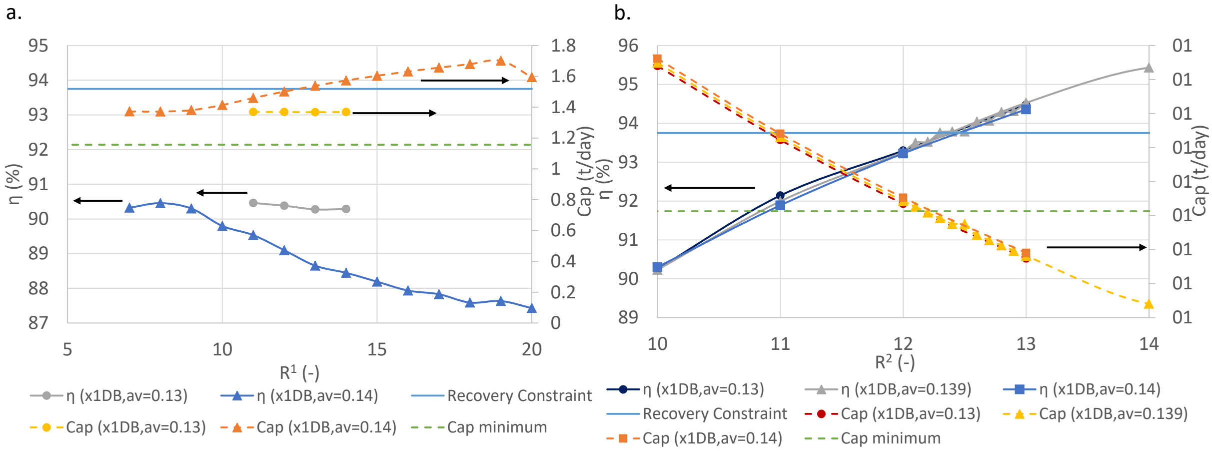

Fig. 8. Results for $x_{Ch} = 50$ mass%. a. Effect of R^1 for different $\overline{x}^1_{D,B}$ ($R^2 = 10$). b. Effect of R^2 for different $\overline{x}^1_{D,B}$ ($R^1 = 9$).

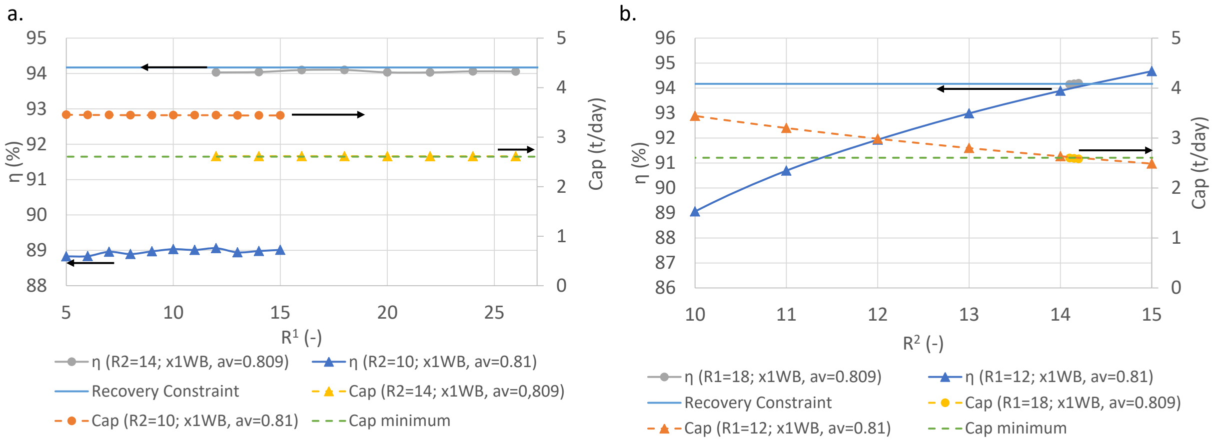

Fig. 9. Results for $x_{Ch} = 80$ mass%. a. Effect of R^1 for different $\overline{x}^1_{W,B}$. b. Effect of R^2 for different R^1.

3 by performing sensitivity studies (Table 5) with much smaller step sizes (0.001 for $\overline{x}^1_{D,A}$; 0.05 for R^1 and R^2) than previously due to convergence difficulties.

By optimisation, in Case 2, Cap slightly increases, while η decreases to its minimum value. This Cap increase is mainly due to the slightly decreased R^1. R^2 and $\overline{x}^1_{D,A}$ slightly increase. In Case 3, the results change similarly to Case 2. This time, however, the small Cap increase is caused by the increased $\overline{x}^1_{D,A}$: somewhat less acetone is transferred to Column 2; hence Δt^2 is shorter. In Case 3, the results are insensitive to the changes of R^1 in the range of 0.0001–0.1.

Mixture 2

For Mixture 2, all three cases were optimised by GA (Table 6).

In Case 1, R^1 increased, and R^2 decreased by optimisation. $\overline{x}^1_{D,B}$ changed only slightly compared to the results of the sensitivity study (Table 4). The decrease in R^2 caused shorter Δt^2; hence Cap considerably increased. With the decrease of R^2, R^1 increased to maintain η at its specified minimum value.

Table 5
Results of optimisation for Mixture 1.

	Case 2	Case 3
Operation parameters	$R^1 = 0.007$ $\overline{x}^1_{D,A} = 0.861$ $R^2 = 4.6$	$R^1 = 0.01$ $\overline{x}^1_{D,A} = 0.864$ $R^2 = 4.3$
$n{\bullet}\Delta t^1 / \Delta t^2$, h	29.64 / 28.52	23.94 / 27.26
η, %	99.08 (100 %)	99.92 (100 %)
Cap, t/day	10.81 (109.1 %)	7.33 (112.8 %)

Table 6
Results of optimisation for Mixture 2.

	Case 1	Case 2	Case 3
Operation parameters	$R^1 = 10.9$ $\overline{x}^1_{D,B} = 0.029$ $R^2 = 12.2$	$R^1 = 17.7$ $\overline{x}^1_{D,B} = 0.128$ $R^2 = 12.2$	$R^1 = 17.9$ $\overline{x}^1_{W,B} = 0.81$ $R^2 = 14.3$
$n{\bullet}\Delta t^1 / \Delta t^2$, h	187.50 / 236.71	10.0 / 213.80	25.90 / 96.10
η, %	93.01 (100 %)	93.73 (100 %)	94.17 (100 %)
Cap, t/day	1.03 (119.4 %)	1.16 (99.7 %)	2.60 (99.9 %)

In Case 2, R^1 increased by 97%, while $\overline{x}^1_{D,B}$ and R^2 decreased by 8% and by 1%, respectively. Δt^2 slightly decreased, resulting in slightly higher Cap, but still by 0.3 % lower than for the single-column process. Δt^1 is remarkably low compared to that of Case 1. Because of the low number of theoretical stages of Column 1, it is difficult to fulfil the Recovery Constraint. The acetic acid concentration in the distillate is high, which causes a considerable loss of acetic acid; hence Column 1 can be operated only for a short time.

In Case 3, the operating parameters hardly changed by optimisation. Cap slightly increased because of the finer adjustment of parameters. For the same reason as in Case 2, Column 1 operates for a relatively short time.

5. Investigation of the time equality hypothesis

Based on the previous results, the operating parameters resulting in the maximum Cap were different from those where the distillation times of the two columns were equal. Although, for Mixture 1, the optimum found by GA is close to the point of time equality (Table 5), but this is not the case for Mixture 2 (Table 6). These results contradict our hypothesis that the maximum Cap is found at time equality. Hence, the location of the time equality point is further investigated.

Further calculations were performed for both mixtures for Cases 1 and 2. R^2 was calculated for different R^1 and $\overline{x}^1_{D,A}$ (for Mixture 1) and $\overline{x}^1_{D,B}$ (for Mixture 2) values by sensitivity studies considering two different aspects:

- the value of R^2 necessary to reach the specified η (denoted as R^2_η in the following figures),
- the value of R^2 at the time equality ($n\bullet\Delta t^1 = \Delta t^2$) of the two columns (denoted as R^2_t in the following figures).

5.1. *Mixture 1*

Fig. 10 shows R^2 as the function of R^1 and $\overline{x}^1_{D,A}$ for Case 1. The points where $n\bullet\Delta t^1 = \Delta t^2$ and η is the specified one, would be located where the two surfaces intersect. Such points were not found in Case 1 because R^2_η changes rapidly by slightly changing $\overline{x}^1_{D,A}$ between 0.73 and 0.74, where the intersection possibly exist. If the time equality hypothesis is valid, the optimum would be the point of the intersection with the lowest R^2 value. The point with the highest Cap calculated by the sensitivity analysis (Table 4) is shown by a black dot in Fig. 10. This point is close to the intersection of the two surfaces regarding R^1 and $\overline{x}^1_{D,A}$. R^2 is much lower than it would be at the intersection line because of the above reasons (R^2_η changes rapidly by changing $\overline{x}^1_{D,A}$).

Fig. 11 shows R^2 as the function of R^1 and $\overline{x}^1_{D,A}$ for Case 2. In this case, the intersection of the two surfaces is visible, because the change in R^2_η was not so rapid by changing $\overline{x}^1_{D,A}$ as in Case 1. The optimum found by the genetic algorithm is close to the intersection of the two surfaces (marked by a red dot). In this case, the optimum might be truly at $n\bullet\Delta t^1 = \Delta t^2$, but this is still difficult to verify exactly due to the steep change in R^2_η in response to extremely small changes in $\overline{x}^1_{D,A}$ (dark blue surface).

In most of the calculations, the dominating time for calculating Cap was Δt^1. Therefore, Cap can be increased by decreasing Δt^1 with the increase of $\overline{x}^1_{D,A}$ or the decrease of R^1. This means that less acetone is transferred to Column 2, hence R^2_η must be increased, thus Δt^2 increases (Fig. 12) to maintain the specified η. The two process durations thus varies in the opposite directions (Δt^2 increases if Δt^1 decreases), conforming to the time equality hypothesis. These results suggest that the real optimum is indeed at the time equality.

5.2. *Mixture 2*

Fig. 13 shows R^2 as the function of R^1 and $\overline{x}^1_{D,B}$ for Case 1. For the recovery of acetic acid with the specified purity at lower R^1 values, a much higher R^2 is required than for fulfilling the time equality (the R^2_η surface is above the R^2_t surface at lower R^1 values in Fig. 13). It must be noted that for Mixture 2, changing the stopping criterion of Column 1 has a much stronger effect on R^2_t than for Mixture 1: the R^2_t surface is steeper in this case. Moreover, R^2_η does not change so rapidly with changing $\overline{x}^1_{D,B}$ than for Mixture 1, hence the intersection line can be more easily calculated.

The optimum found by GA (marked by a black dot) is remarkably far from the intersection of the two surfaces (marked with a yellow curve in Fig. 13). The reason for this lies in that Δt^1 and Δt^2 changes differently with changing the operating parameters of Column 1 ($\overline{x}^1_{D,B}$, R^1). Changing any of these operating parameters affects Δt^2 opposingly via two mechanisms; hence Δt^2 has an optimum in most cases. These mechanisms are detailed below.

By increasing $\overline{x}^1_{D,B}$ or R^1, more acetic acid is lost with the distillate of Column 1 (D^1). As less acetic acid is transferred to Column 2, R^2 must be increased to maintain the specified η, thus Δt^2 increases. However, the increase of $\overline{x}^1_{D,B}$ or R^1 also means that more water is withdrawn with D^1; thus less water is transferred to Column 2 with the residue. Hence, a lower amount of impurity must be removed in Column 2 and D^2 and Δt^2 decrease. At different fixed R^1 values, the above two mechanisms result in a minimum in Δt^2, which means a maximum in Cap as Δt^2 determines Cap in most of the calculations (where the R^2_η surface is above the R^2_t one in Fig. 13). At higher fixed $\overline{x}^1_{D,B}$ values, Δt^2 has a minimum in the range of $R^1 = 4$–8 as a result of these mechanisms (Fig. 14). This also means a maximum in Cap.

The two opposite effects of R^1 and $\overline{x}^1_{D,B}$ are caused by the fact that it is the residue, which is processed further in Column 2: when the distillate is processed further in Column 2, the effect of the amount of impurity

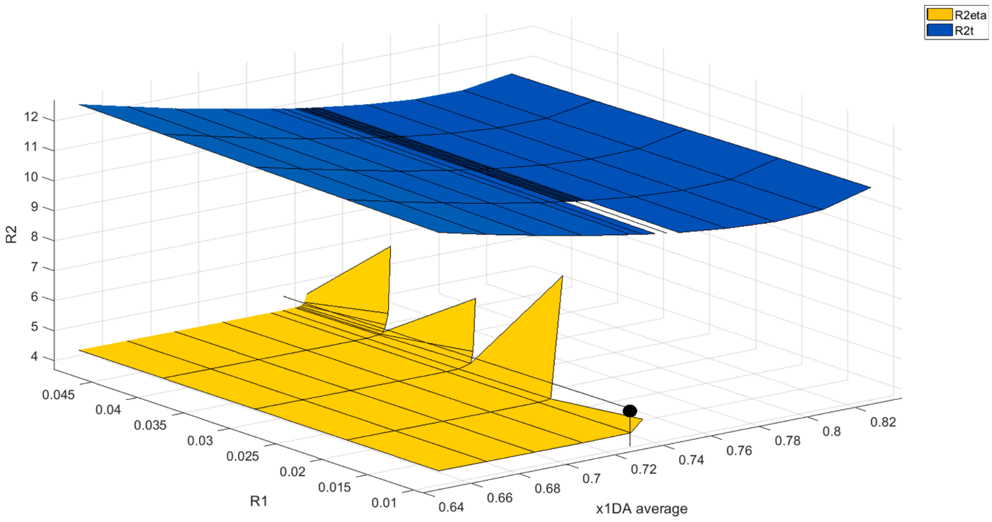

Fig. 10. R^2 as the function of R^1 and $\overline{x}^1_{D,A}$ for Mixture 1 – Case 1.

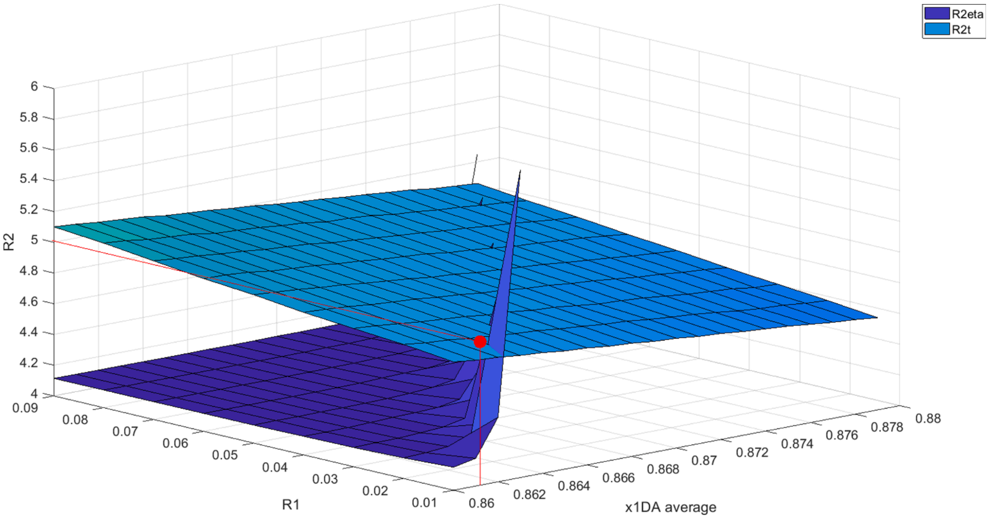

Fig. 11. R^2 as the function of R^1 and $\bar{x}^1_{D,A}$ for Mixture 1 – Case 2.

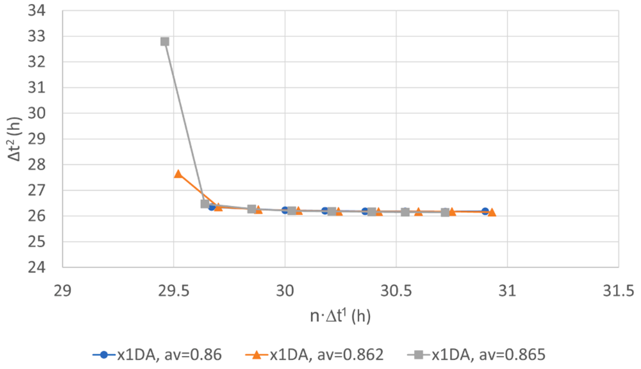

Fig. 12. Δt^2 as the function of $n\bullet\Delta t^1$ for Mixture 1 – Case 2.

transferred on Δt^2 is not significant.

In Case 2, the intersection of the two R^2 surfaces is at higher R^1 and lower $\bar{x}^1_{D,B}$ values (similarly to Case 1), but R^2_η does not change so rapidly as in Case 1. Moreover, there is no such a minimum in Δt^2 as in Case 1 (Fig. 15). The maximum Cap is reached at lower Δt^1 (lower R^1 and higher $\bar{x}^1_{D,B}$), however, it is still lower than that of the single-column process. In this case, there is no significant change in the amount of water transferred to Column 2 when decreasing R^1 or increasing $\bar{x}^1_{D,B}$. Hence, the process duration determining Cap is only influenced by the increasing R^2, which results in decreasing Cap. To avoid the high acetic acid loss in Column 1, Δt^1 shall be decreased (by decreasing R^1 or $\bar{x}^1_{D,B}$) which increases R^2 and consequently Δt^2.

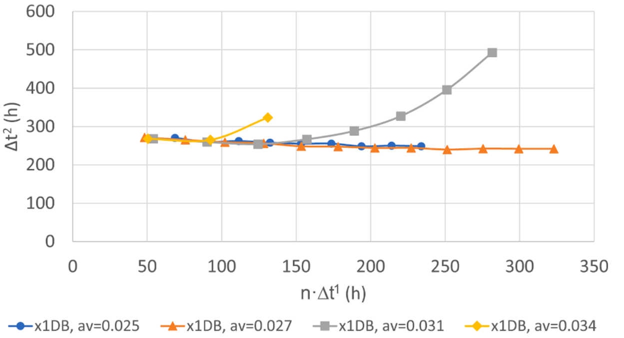

Fig. 14. Δt^2 as the function of $n\bullet\Delta t^1$ for Mixture 2 – Case 1 (R^1 = 4–18).

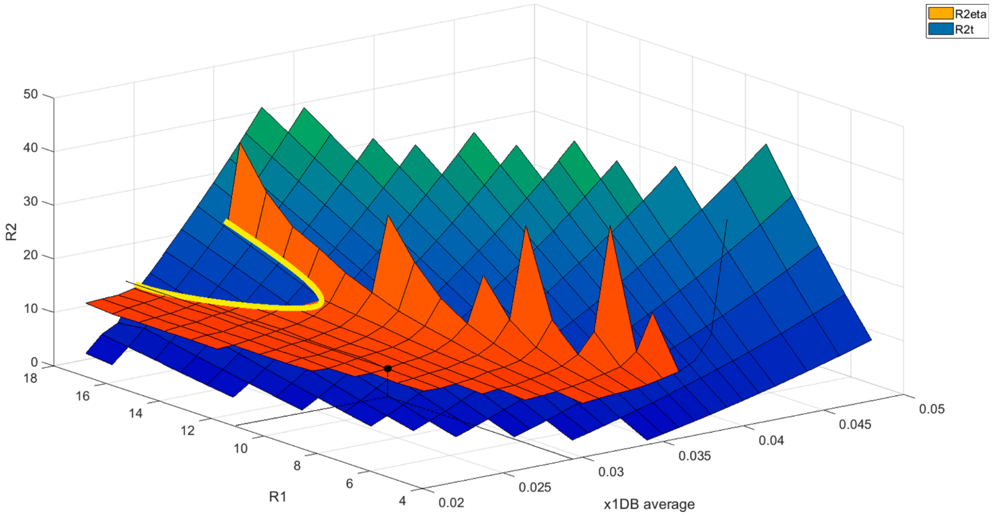

Fig. 13. R^2 as the function of R^1 and $\bar{x}^1_{D,B}$ for Mixture 2 – Case 1.

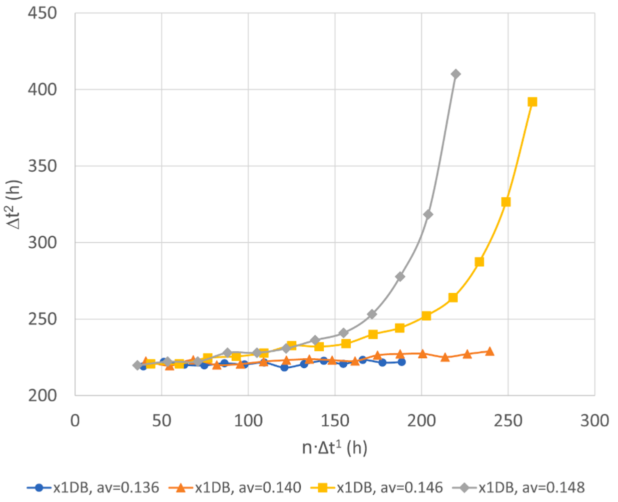

Fig. 15. Δt^2 as the function of $n \bullet \Delta t^1$ for Mixture 2 – Case 2 ($R^1 = 11–23$).

6. Conclusions

Pharmaceutical plants commonly produce several products; hence a given amount of waste solvent must be regenerated in a specific time period, else the remaining amount of waste solvent untreated must be incinerated. The waste solvent regeneration is usually performed by batch distillation. To avoid incineration, the processing capacity (Cap) of the regeneration process must be increased.

In our work, two-column batch distillation processes were studied to increase Cap for the mixtures acetone (A)-water (B) (Mixture 1) and water (A)-acetic acid (B) (Mixture 2). For both mixtures, three different charge compositions were considered: 20 (Case 1), 50 (Case 2) and 80 mass% (Case 3) of organic component. For both mixtures, the organic component must be recovered with a purity of 99.7 mass%. The Cap of the two-column process was maximised while respecting the following two constraints:

1. The minimum liquid volume of the reboiler of Column 2 at the end of the separation process was specified.
2. The recovery (η) must be equal to or higher than that of the corresponding single-column process.

For Mixture 1, by the two-column process, higher Cap was not reached in Case 1 than by the single-column one because both constraints can not be fulfilled at the same time if less than six charges are processed in Column 1. However, when the feed had higher acetone concentration, higher Cap was reached than for the single-column process: in Case 2 by 9.1 % and in Case 3 by 12.8 %.

For Mixture 2, Cap increased by 19.4 % in Case 1, while for the other two cases, Cap decreased by ca. 0.1–0.3 % compared to the single-column process.

For both mixtures, there was a certain range of charge composition where it was possible to reach higher Cap (with the same η and purity) by applying two batch distillation columns sequentially than by the single-column process. When the product was the heavier component (obtained in the still), the two-column process was better at lower concentrations of the heavier (organic) component. When the product was the lighter component (obtained as distillate), the two-column process was better at higher concentrations of the lighter (organic) component.

Since Cap was calculated as the mass of feed divided by the maximum operating time ($n \bullet \Delta t^1$ or Δt^2), its maximum was expected, where $n \bullet \Delta t^1 = \Delta t^2$ (n is the number of charges processed in Column 1). However, this was only valid for Mixture 1, where the pre-fractionation in Column 1 was sufficient, and a large amount of impurity (water) was removed in Column 1 in the still residue.

For Mixture 2, a large amount of impurity can not be removed in Column 1 without high acetic acid loss. That is why at certain combinations of operating parameters increasing Δt^1 also increased Δt^2 at lower R^1 and higher $\overline{x}^1_{D,B}$ values. However, at lower $\overline{x}^1_{D,B}$ values, Δt^2 decreased on the increase of Δt^1 resulting in a minimum in Δt^2 far from the time equality point. The minimal Δt^2 depended on the initial charge composition: with increasing $x_{ch,B}$, better pre-fractionation was reached in Column 1 (more water was removed with the distillate), and the minimum in Δt^2 disappeared.

CRediT authorship contribution statement

Bence Nemeth: Methodology, Investigation, Visualization, Formal analysis, Writing – original draft. **Laszlo Hegely:** Conceptualization, Methodology, Formal analysis, Writing – review & editing, Supervision. **Peter Lang:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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