



M Ű E G Y E T E M 1 7 8 2

BUDAPEST UNIVERSITY OF TECHNOLOGY AND ECONOMICS
DOCTORAL COUNCIL OF THE FACULTY OF MECHANICAL ENGINEERING
DEPARTMENT OF BUILDING SERVICES AND PROCESS ENGINEERING

László Hégyel

**IMPROVEMENT OF BATCH DISTILLATION SEPARATION
OF AZEOTROPIC MIXTURES**

Doctoral Thesis Booklet

Supervisors:

Prof. Péter Láng DSc

BME Department of Building Services and Process Engineering

Dr. Vincent Gerbaud HDR

INP Toulouse – ENSIACET – “Laboratoire de Génie Chimique”

Budapest

2013

Introduction

I started my research work concerning batch distillation as an undergraduate student in 2006 at the Department of Building Services and Process Engineering under the supervision of Prof. Péter Láng. After preparing my diploma work and two scientific student's works in this subject, I began my PhD studies in 2009. Since 2011 my studies has been continued under joint supervision of Dr. Vincent Gerbaud from the university of INP Toulouse – ENSIACET, department of “Laboratoire Génie Chimique”. Between 2011 and 2013, I spent five months in each year at ENSIACET thanks to the scholarship provided by the French Government.

Distillation is the most frequently applied method for the separation of liquid mixtures, which is based on the difference in the volatilities of the components. As distillation comprises successive partial vaporisation and condensation steps, its energy demand is very high. Therefore, the optimal design and operation of distillation processes is very important, both economically and environmentally.

If the mixture is highly non-ideal, azeotropes may occur. In the azeotropic point, the composition of the vapour and that of the liquid phase is identical, which means that an azeotropic mixture can not be separated by conventional distillation process. If the relative volatility of the components is very low, the separation is feasible in theory, but the high reflux ratio and high number of plates render conventional distillation uneconomical.

For the separation of azeotropic and close-boiling mixtures, special distillation methods have to be applied. These methods exploit the eventual pressure-sensibility of the azeotropic composition (pressure-swing distillation), or the favourable influence of a mass separating agent (entrainer). The effect of the entrainer is different depending on the separation technique. By homoazeotropic distillation an azeotropic behaviour induced by the entrainer is exploited. By extractive distillation the entrainer, fed continuously into the column, changes the relative volatilities favourably. By heteroazeotropic distillation, the entrainer forms two liquid phases with one of the original component, and this liquid-liquid split is exploited (by decantation) to separate the original mixture.

In certain industries, e.g. pharmaceutical industry, solvent recovery, fine chemical industry, the production of spirits and dyes, distillation is performed in batch mode. The advantage of batch distillation is that it can be applied for the separation of mixtures with varying (often small) quantity and composition. In this way it is suitable for separating mixtures only seasonally produced. In the above industries, multicomponent azeotropic mixtures are

frequently encountered. In the case of regeneration of waste solvent mixtures, usually a main component is to be recovered, which is advantageous both environmentally (a lower amount of mixture has to be incinerated) and economically. The existence of azeotropes might limit the recovery, or even make the separation infeasible, unless a special distillation method is applied.

The possibility of energy-saving and the stricter environmental regulations caused an increased interest in the research of batch distillation in the recent decades, with an emphasis on special distillation methods and non-conventional column configurations.

The aim of my research is to improve the batch distillation separation of azeotropic mixtures, in particular to

- propose a new algorithm for the determination of the sequence of products and their maximum amounts for the batch distillation of multicomponent azeotropic mixtures,
- compare different closed operational modes of batch column configurations (batch rectifier, middle-vessel column, multivessel column),
- study the batch extractive separation of pharmaceutical azeotropic waste solvent mixtures by laboratory experiments, industrial-size pilot productions and rigorous simulation, and to investigate a new operational policy for batch extractive distillation,
- investigate the effect of off-cut recycle for the traditional batch distillation and batch extractive distillation separation of a pharmaceutical waste solvent mixture,
- construct a general model for batch heteroazeotropic distillation with variable decanter hold-up, and
- extend the above model for batch heterogeneous extractive distillation.

Methods of investigation

The following methods were applied in my research work:

- feasibility analysis with simplified models,
- rigorous simulation,
- laboratory experiments,
- industrial-size pilot plant experiments.

In the course of *feasibility analysis*, a general model of batch heteroazeotropic distillation was proposed, and this model was also extended to batch heterogeneous extractive distillation.

The following simplifying assumptions were applied:

- the composition of the condensate is constant and equals to that of the heteroazeotrope,
- the overall liquid composition in the decanter equals that of the stream entering the decanter,
- the vapour and liquid hold-ups of the column and of the condenser are negligible,
- constant molar overflow,
- the entrainer is boiling point liquid.

Rigorous simulation was performed to compare different open and closed operation modes of non-conventional column configurations, to model traditional batch distillation and batch extractive distillation separations of waste solvent mixtures (including posterior simulation of laboratory experiments and pilot productions), and validate the results of the feasibility analysis.

In the rigorous simulations the following simplifying assumptions were applied:

- theoretical trays,
- constant volumetric liquid holdup on the trays and in the decanter,
- negligible vapour holdup,

The model equations to be solved are well-known:

- non-linear ordinary differential equations (material balances, heat balances),
- algebraic equations (vapour-liquid and liquid-liquid equilibrium equations, summation equations and physical property models).

The phase equilibria are described by the SRK equation of state for hydrocarbons, and the NRTL and UNIQUAC activity coefficient models for all the other systems. The rigorous calculations were performed by the dynamic module (CC-DCOLUMN) of the professional flowsheet simulator ChemCAD. For the off-cut recycle calculations, a program written in Visual Basic for Applications under Microsoft Excel performed the mass balance calculations and called ChemCAD for the rigorous simulation.

Laboratory experiments for the separation of pharmaceutical waste solvent mixtures by traditional batch distillation, batch extractive distillation and hybrid (absorption+distillation) process were carried out in a glass distillation column (*Fig. 1*). The experiments were performed in order to prove that the application of a special distillation method increases the recovery of the main component (or even makes the separation feasible). All experiments were performed at atmospheric pressure. The column (diameter: 5 cm) was filled with PROPAK packing of 0.16 in (0.41 cm) diameter in a height of 153 cm. The still – a round bottom flask of 2 dm³ – can be heated with a heating basket (nominal heat duty: 400 W). Temperatures were measured at four different column heights, at the top and in the still. In the batch extractive distillation experiments, one of the thermometers was removed during water feeding, and the stub was used as the water inlet point. The analysis of the organic compounds was performed by gas chromatography (CHROMPAK with FFAB CB25 mx column, or Varian CP3800 with CP-Sil-5CB column). The water content was determined by Karl-Fischer method.

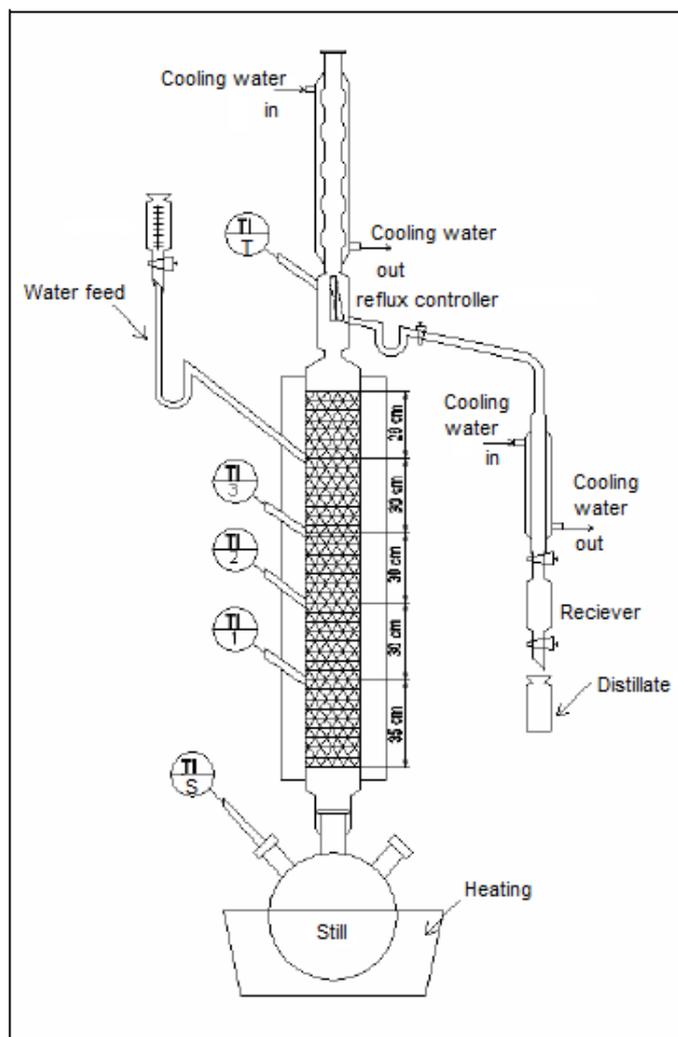


Fig. 1. The sketch of the laboratory distillation column (BED and HP).

After the favourable results of the batch extractive distillation laboratory experiments for the recovery of methanol from a waste solvent mixture, two *industrial-size pilot productions* were performed, as well. The first production was by traditional batch distillation, the second one by a new operational policy of batch extractive distillation. The industrial column had 50 bubble cap plates, its internal diameter was 1.25 m. The volume of the reboiler was 25 m³, its surface was 50 m². The total condenser, operated with cooling water, had a surface of 100 m². The reboiler was heated with steam of 5 bar. The estimated hold-up of the condenser is 0.5 m³, that of the column: 1.5 m³. Based on preliminary calculations and the possibilities available, water as entrainer (without preheating) was fed into the reflux stream. The analysis of the organic compounds was performed by gas chromatography (Varian CP3800 with CP-Sil-5CB column). The water content was determined by Karl-Fischer method.

New scientific results

The new scientific results are briefly presented in the following theses. The theses are related to different aspects of batch distillation: batch homoazeotropic distillation (Theses 1 and 3), non-conventional column configurations (Thesis 2), batch extractive distillation (Theses 3 and 4), batch heteroazeotropic distillation (Thesis 5) and batch heterogeneous extractive distillation (Thesis 6).

Thesis 1

Related publications: [1], [5], [10]

I developed a new algorithm for the determination of product sequences of batch distillation. This new algorithm requires the knowledge of only the boiling points of pure components and azeotropes, and the azeotropic compositions, and can handle any number of components. It is also suitable for treating pressure change and the existence of heteroazeotropes, as well. The algorithms published previously either required the use of a vapour-liquid equilibria (VLE) model or could be applied only for ternary mixtures.

In multicomponent azeotropic mixtures, the composition and amount of the cuts obtained by conventional batch distillation highly depend on the charge composition. With the assumption of maximal separation (very high reflux ratio, infinite number of trays), the sequence of cuts and their relative amount can be determined.

The new algorithm does not require the use of a VLE model, thanks to the assumption that the Serafimov topological class of all the ternary submixtures occur in practice. The steps of the algorithm (*Fig. 2*) are the following ones. First, the stability of all the stationary points of the residue curve map is determined in the ternary submixtures, based on the knowledge of topologically possible residue curve map structures. Then the stabilities are updated as the ternary submixtures are unified into quaternary ones, the quaternary ones into quinary ones, etc. On the basis of the stabilities, the adjacency matrix is completed, and the batch distillation regions (possible product sequences) are determined. Finally, the product sequence and relative amount of cuts are determined for the actual charge composition.

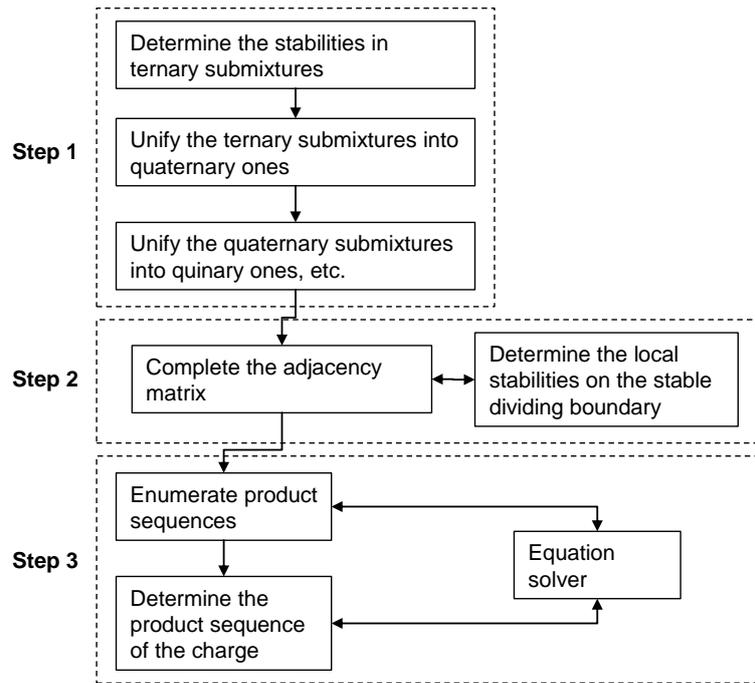


Figure 2. The overall structure of the algorithm

I tested the new algorithm by comparing the results for a five-component mixture with the ones obtained by Ahmad et al. [21]. The stabilities and the set of product sequences (Fig. 3a) calculated by the new algorithm agree with obtained by Ahmad et al. [21]. I also calculated the product sequences for this mixture at a higher pressure (Fig. 3b). With the knowledge of the possible sequences at both pressures, I could determine the product sequence for a pressure-swing distillation production. On the example of a second mixture, I demonstrated that the new algorithm is able to handle heteroazeotropes as well. These results verify that the new algorithm is suitable for the determination of the sequence of the cuts without using a VLE model.

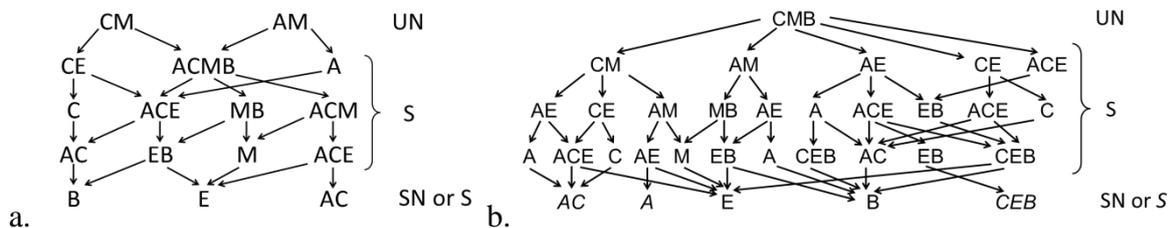


Figure 3. The possible product sequences of the mixture acetone (A) – chloroform (C) – methanol (M) – ethanol (E) – benzene (B) a. 1.013 bar, b. 10 bar. (UN: unstable node, S: saddle, SN: stable node)

Thesis 2

Related publications: [4], [9], [14], [16]

I studied the open and different closed modes (which differ from each other in the operation of the vessels) of batch rectifier and middle-vessel column by rigorous dynamic simulation for a binary and a ternary mixture. I also investigated four different closed modes of the multivessel column. I compared the recoveries under constant product quality and energy consumption.

I suggested a new definition for the reflux ratio, which can be applied for closed operation modes, as well. As no distillate is withdrawn, R is always infinite according to the conventional definition. In the new definition the distillate flow rate is replaced with the difference between the vapour and liquid flow rates:

$$R = \frac{L}{V - L}$$

This definition makes it possible to take the hold-up change in the top vessel (or in the condenser) into account, as well. R is only infinite, when the vapour and liquid flow rates are equal, that is, the hold-up of the top vessel is constant. If accumulation takes place in the vessel, R is a finite, positive number.

I compared the recoveries of the open and six different closed operation modes of batch rectifier and middle-vessel column by rigorous dynamic simulation under constant product quality and energy consumption. I concluded that the closed operation of the batch rectifier provided higher recoveries in case of negligible liquid hold-up (*Fig. 4*). Level control with initially filled up top vessel (Mode 2b) and temperature control with initially empty top vessel (Mode 4a) proved to be the best closed modes

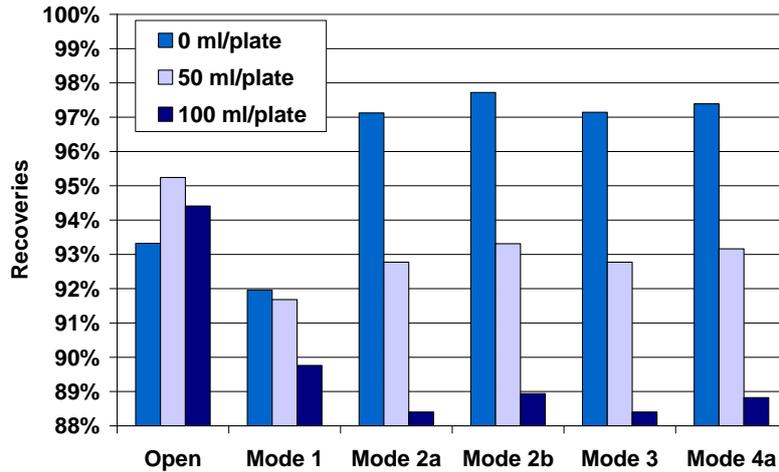


Figure 4. The recoveries of *n*-hexane by batch rectifier at different levels of hold-up.

I concluded that for the middle-vessel column the open operation mode proved to be better than the closed ones in every case (Fig. 5). It was not possible to choose the best closed mode, as the order of the closed modes (with respect to the recoveries) depends on the product and on the hold-up. For negligible liquid hold-up, temperature control (Mode 4a), for higher hold-ups level control with initially empty top and bottom vessels (Mode 2a) can be recommended.

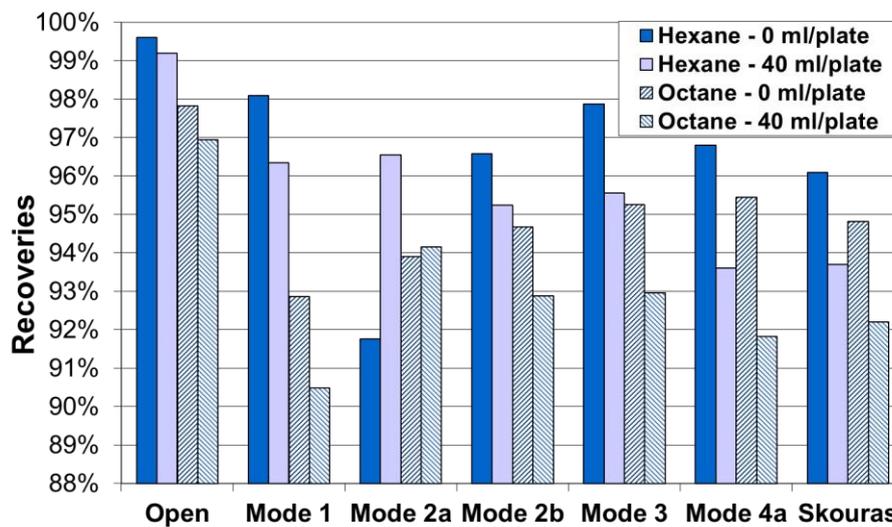


Figure 5. The recoveries of the different middle-vessel column operation modes.

I compared the recoveries and energy consumption of four closed operation modes of the multivessel column for a quaternary mixture (Table 1). The differences in the recoveries are small. The energy consumption of level control with initially empty vessels (Mode 2a) was the lowest. I stated that the division of the charge between the vessels (instead of filling the whole charge into the bottom vessel only) had adverse effect on energy consumption.

	Δt_{total} (min)		A	B	C	D	Average
Mode 2a	16.55	Recovery (%)	94.77	80.57	82.60	97.62	88.89
		Purity (%)	96.01	96.01	96.00	96.17	96.05
Mode 2b	25.85	Recovery (%)	95.38	80.16	82.60	98.63	89.19
		Purity (%)	96.07	96.06	96.00	96.07	96.05
Mode 4a	18.25	Recovery (%)	95.38	79.76	83.82	96.19	88.79
		Purity (%)	96.01	96.01	96.00	96.98	96.25
Mode 4b	22.75	Recovery (%)	95.59	79.55	84.02	97.21	89.09
		Purity (%)	96.01	96.00	96.00	97.13	96.29

Table 1. Calculated results for the multivessel column

(Modes 2a, 4a: charge in the reboiler only, Modes 2b, 4b: distributed charge).

Thesis 3

Related publications: [7], [17]

I studied the influence of recycling off-cuts for the batch homoazeotropic distillation (BD) and batch extractive distillation (BED) regeneration of a four component (methanol - THF - water - toluene) waste solvent mixture of a pharmaceutical plant by rigorous dynamic simulation. I investigated a batch rectification process of 6 consecutive productions, where the first fore-cut was incinerated, whilst the second fore-cut, the after-cut and the column hold-up were recycled. In the BED process, water as entrainer was fed into the top of the column during the heating-up period.

I created a program for the calculation of the production process with off-cut recycle. The program, written in Visual Basic for Applications in Microsoft Excel, performs the material balance calculations of the recycle and calls ChemCAD for the rigorous dynamic simulation of the productions. I determined the optimal volume of the first fore-cut with respect to the profit of the regeneration process, and stated that its value is slightly lower for BED (*Table 2*). I concluded that a minimum volume of first fore-cut must be taken to avoid the accumulation of organic pollutants (THF and toluene) in the charge, which would render the 6-batch process infeasible. This volume is higher for BED. I also found that the optimal BED process gave significantly higher profit than the optimal BD.

Case	$V_{D,1a}=0$ m^3	$V_{D,1a}=1$ m^3	$V_{D,1a}=2$ m^3	Base (BD: 3.3-3.7 m^3 , BED: 2.7-3 m^3)	$V_{D,1a}=3$ m^3	$V_{D,1a}=4$ m^3	No recycle
BD	-153	141	1722	2475	-	2112	295
BED	7	330	1142	2869	2921	1743	911

Table 2. The influence of the amount of fore-cut incinerated on the profit (in \$, optimum in bold)

Thesis 4

Related publications: [6], [8], [11], [15]

The application of BED and that of the hybrid process (HP) was investigated for two pharmaceutical waste solvent mixtures, whose separation is prevented (Mixture 1: methanol, THF, acetonitrile, water, pyridine) or limited by azeotropes (Mixture 2: acetone, methanol, tetrahydrofuran, n-hexane, ethanol, water and toluene). The BED operational policies investigated include a new policy suggested by us.

I studied the performance of the basic and a modified operational policy of BED and HP for the recovery of THF from Mixture 1. I concluded that both water and pyridine are suitable as entrainers, but water is more practical to use. I evaluated experiments performed on a laboratory packed column by rigorous simulations (*Fig. 6*). The prescribed separation task was not feasible with BD, but it was possible to produce THF of desired quality by BED and HP. The highest recovery and production rate were achieved with the HP, while the least efficient process was the basic policy of BED.

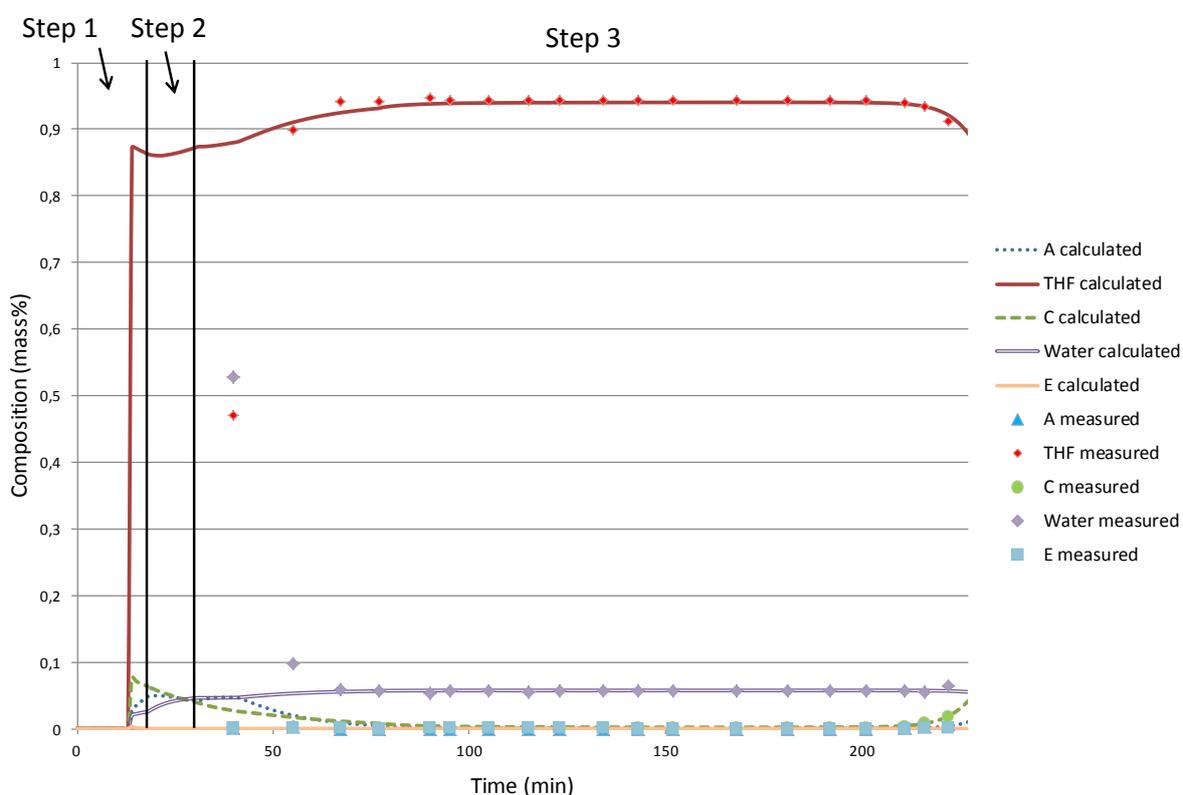


Figure 6. Evolution of the measured and calculated distillate composition (modified operational policy of BED)

I also investigated the effect of operational parameters. I concluded that by BED the higher the feeding location, the greater is the recovery, while the reflux ratio has an optimal value. By HP, the increase of water flow rate slightly increases the recovery but at the price of higher specific energy and water consumption.

In the case of Mixture 2 several azeotropes limit the recovery of methanol by BD by causing a significant loss of methanol. We suggested a new BED operational policy [22], where water (entrainer) feeding was applied only during the heating-up of the column (BED1). At the end of the heating-up, the concentration of organic pollutants increased (compared to BD) and methanol concentration decreased significantly at the top of the column. I stated that water feeding can be continued during the fore-cut (BED2), but this increases the amount of fore-cut and dilutes the mixture from which methanol is recovered.

I performed preliminary simulation, and on its basis, laboratory experiments to compare the BD and the two BED operational policies. I concluded that the highest recovery was obtained by BED1, the lowest one by BD. I also performed the posterior rigorous dynamic simulation of the experiments (*Fig. 7*), which gave the same order of separation methods with respect to recovery. Industrial-size pilot productions of BD and BED1 were performed in a 50 bubble cap tray column. By BED1 the recovery increased considerably, which is explained by the significant decrease of methanol concentration in the distillate at the end of the heating-up period by BED, and thus lower methanol loss with the fore-cut. I investigated the pilot productions also by posterior rigorous simulation, and I stated that the specific energy consumption of the BED production was considerably lower than that of BD.

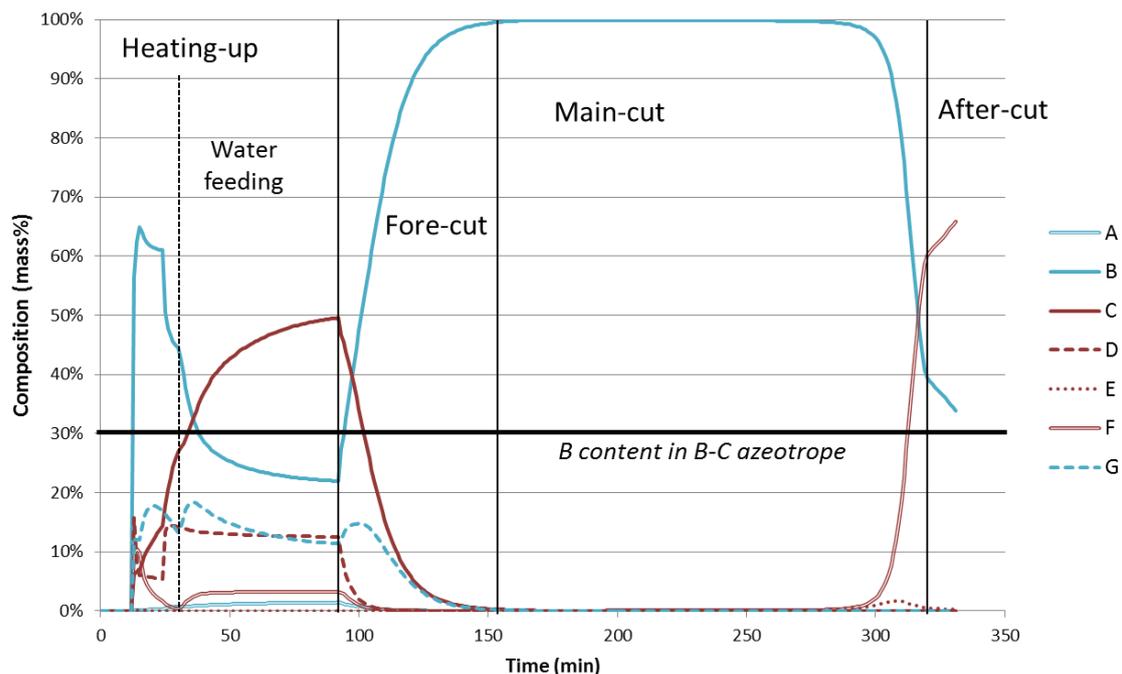


Figure 7. The evolution of calculated top vapour compositions (BED1 laboratory experiment).

Thesis 5

Related publications: [2], [12], [13]

I proposed a general model of batch heteroazeotropic distillation, where both liquid phases can be refluxed or withdrawn as distillate. Their hold-up in the decanter can be increased, decreased or kept constant, as well. I suggested two new operational parameters (r_R and r_W) defining the ratio of the flow rates of the entrainer(E)-rich and E-lean phase refluxed and condensed, respectively.

By assuming maximal separation, I derived the still path equation describing the variation of the still composition in time:

$$\frac{dx_s}{dt} = \frac{V}{H_s} \left[(1 - r_R) \eta_R (x_s - x_{1,R}) + (1 - r_W) (1 - \eta_R) (x_s - x_{1,W}) \right]$$

Depending on the values of r_R and r_W , I distinguished 16 possible operational policies, several of which were proposed by me for the first time. I stated that the still path direction can be located in eight different zones (*Fig. 8*) depending on the value of the operational parameters. These zones cover all the possible directions, that is, the still composition can be changed in any desired direction. I also concluded that it is possible to recover a pure component in the still by choosing the appropriate operational policies, thus eliminating the need for a further separation step.

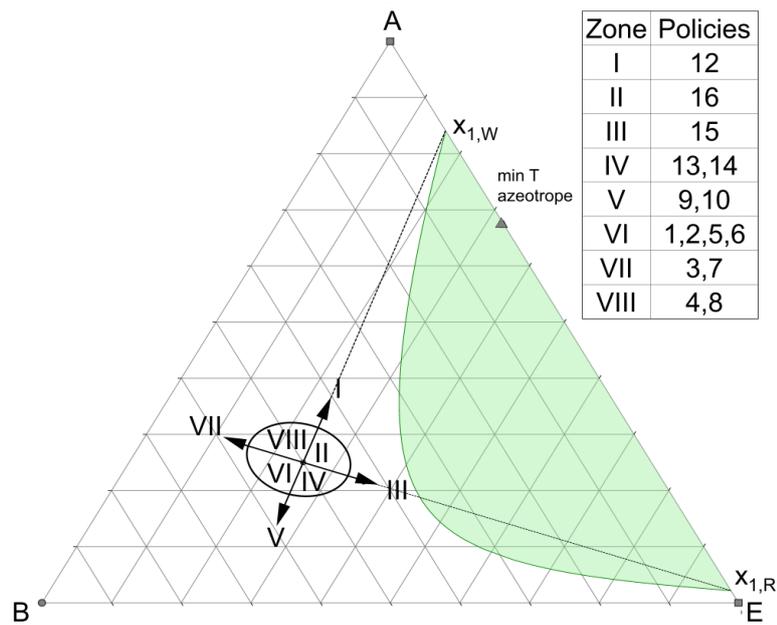


Figure 8. The direction of the still path for the different operational policies.

I validated the still path directions for three new operational policies by rigorous simulation for the mixture water – formic acid – propyl formate. I demonstrated the benefit of using a non-traditional operational policy with hold-up reduction in the decanter, as well.

Thesis 6

Related publications: [3]

I extended the model of batch heteroazeotropic distillation for batch heterogeneous extractive distillation by taking into consideration the continuous entrainer feeding. I distinguished two different entrainer feeding locations: in Case 1 the entrainer is fed into the column, in Case 2, it is added to the decanter.

I derived the equation describing the evolution of the still composition for both cases.

$$\text{Case 1: } \quad \frac{dx_S}{dt} = \frac{V}{H_S} \left[(1-r_R)\eta_R(x_S - x_{1,R}) + (1-r_W)(1-\eta_R)(x_S - x_{1,W}) \right] - \frac{F_E}{H_S}(x_S - x_E)$$

$$\text{Case 2: } \quad \frac{dx_S}{dt} = \frac{V + F_E}{H_S} \left[(1-r_R)\eta'_R(x_S - x_{1,R}) + (1-r_W)(1-\eta'_R)(x_S - x_{1,W}) \right] - \frac{F_E}{H_S}(x_S - x_E)$$

Compared to batch heteroazeotropic distillation, a new term, related to the continuous entrainer feeding appeared, and by Case 2, the influence of the existing terms, related to the operation of the decanter was increased.

I discussed the practical applicability of the possible operational policies and studied the effect of the continuous entrainer feeding on the still path. I concluded that the eight original still path zones of batch heteroazeotropic distillation are modified (*Fig. 9*): some of them disappear, and the remaining zones overlap each other. I also stated that, similarly to batch heteroazeotropic distillation, it is possible to direct the still composition into any direction, however, the influence of the entrainer feeding is large in practice, and it is difficult to move the still path away from the direction of entrainer composition. For the same reason, the variation of the hold-up of the phases in the decanter only has small effect on the still path.

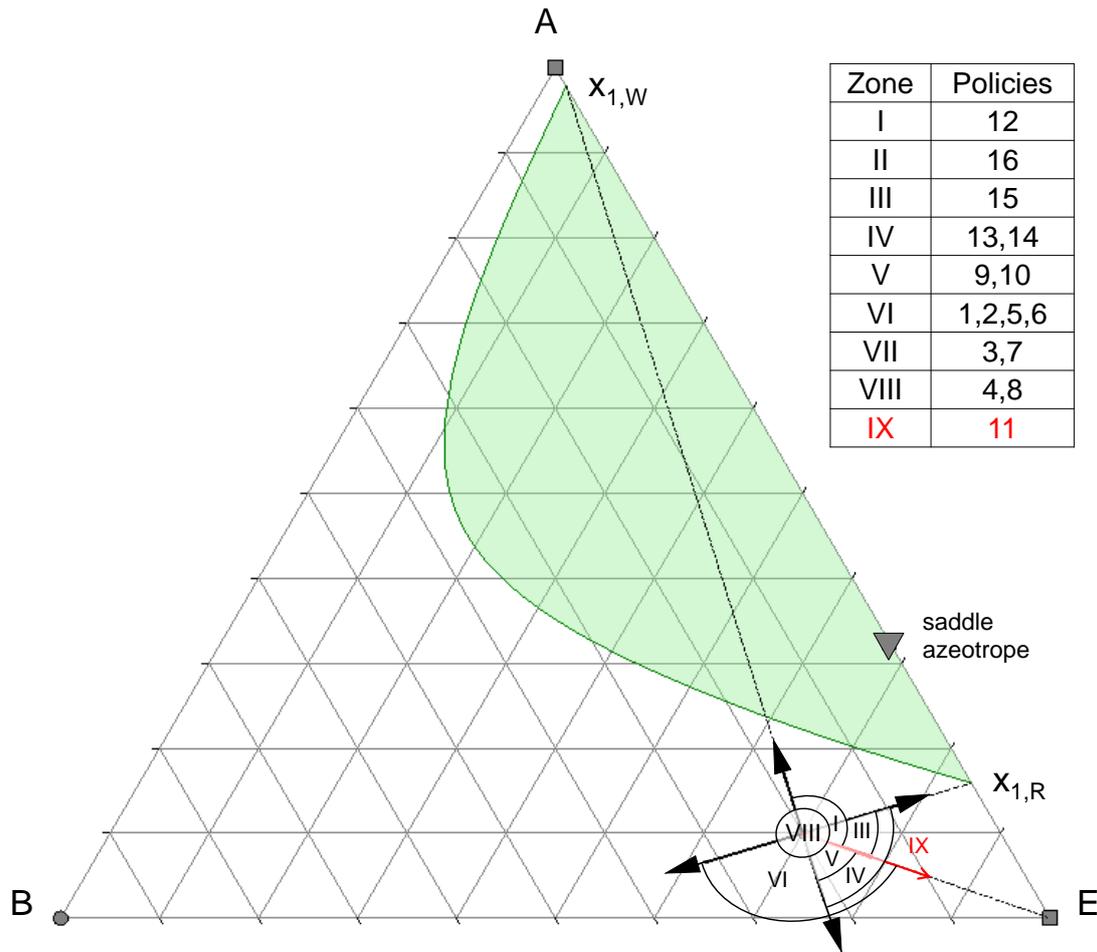


Figure 9. The direction of the still path for the different operational policies (batch heterogeneous extractive distillation).

I validated the still path directions by rigorous simulation of the dehydration of the mixture water – ethanol using n-butanol as entrainer. I stated that by using the new Policy 5 (partial reflux of E-rich phase only), it was possible to reduce the water content of the still residue mainly containing ethanol and butanol.

Application possibilities of the results

The algorithm for the determination of product sequences of batch homoazeotropic distillation, unlike others previously published, requires the knowledge of the boiling points of pure components and azeotropes and the azeotropic compositions only. This means that it can be used more practically in the industry for a rapid assessment of the regeneration of a waste solvent mixture to be separated. The algorithm was coded in Visual Basic for Application under Microsoft Excel, and the program is readily usable.

The comparison of the open and different closed operation modes of batch rectifier showed that the closed modes can provide higher recoveries (lower energy consumption) in the case of low column hold-up. Therefore, existing packed columns could be operated in closed mode, using an available product tank as top vessel, only the piping has to be modified. Moreover, operating the column in a closed way is easier as the product purities increase monotonously.

The new operational policy proposed for batch extractive distillation can be used to increase the concentration of organic pollutants beyond their azeotropic composition in the top of the column, thus reducing the loss of the main component in the fore-cut. This provides an increased recovery and decreased specific energy consumption. A process applying this policy for the recovery of methanol from a multicomponent azeotropic mixture has already been patented [22], and a successful industrial-size pilot production was performed.

Based on my general models of batch heteroazeotropic and heterogeneous extractive distillation, it is possible to obtain one of the original components as product in the still, making a next separation step unnecessary, by choosing the right operational policies, or a combination of them. With the non-conventional operational policies (e.g. variation of decanter hold-up) infeasible separation tasks can become feasible, but the operation becomes more complicated, as well.

Notations

Latin letters

F	feed flow rate
H	hold-up
L	liquid flow rate
r	ratio of the flow rates refluxed and condensed
R	reflux ratio
t	time
V	vapour flow rate
	cut volume (Thesis 3)
x	liquid composition

Greek letters

η	phase split ratio
--------	-------------------

Subscripts

1	reflux stream
1a	first fore-cut
D	distillate
E	entrainer
R	entrainer-rich phase
S	still
W	entrainer-lean phase

Superscripts

'	Case 2 (Thesis 6)
---	-------------------

Publications related to the theses

a. Article in international journal of IF

- [1] L. Hegely, P. Lang, A New Algorithm for the Determination of Product Sequences in Azeotropic Batch Distillation, *Ind. Eng. Chem. Res.*, **50** (22), 12757-12766, (2011).
IF=2.07
- [2] L. Hegely, V. Gerbaud, P. Lang, Generalised model for heteroazeotropic batch distillation with variable decanter hold-up, *Sep. Pur. Tech.* **115**, 9–19, (2013).
IF=2.894
- [3] L. Hegely, V. Gerbaud, P. Lang, General model for studying the feasibility of heterogeneous extractive batch distillation, *Sep. Pur. Tech.* (submitted).

b. Article in international journal

- [4] L. Hégely, P. Láng, Comparison of Closed and Open Operation Modes of Batch Distillation, *Chemical Engineering Transactions*, **25**, 695-700, (2011).
- [5] L. Hegely, P. Láng, New algorithm for the determination of product sequences of special batch distillation methods, *Chemical Engineering Transactions*, **25**, 261-266, (2011).
- [6] L. Hegely, P. Láng, G. Kovács. A New Batch Extractive Distillation Operational Policy for Methanol Recovery, *Chemical Engineering Transactions*, (submitted).
- [7] L. Hegely, P. Láng, Off-cut Recycle for Batch and Batch Extractive Distillation Separation of a Multicomponent Azeotropic Mixture, *Chemical Engineering Transactions*, (submitted).

c. Article in international conference proceedings

- [8] P. Lang, L. Hegely, Gy. Kovacs, J. Gaal-Szilagyi, B. Kotai, Solvent recovery from a multicomponent mixture by batch extractive distillation and hybrid process, *Distillation & Absorption 2010*, Eindhoven, 12-15 September 2010.
- [9] L. Hégely, P. Láng: Study of Closed Operation Modes of Batch Distillation Columns, *Computer Aided Chemical Engineering*, **29**, 1050-1054; Sithonia, 29 May – 1 June 2011, ISBN: 978-0-444-53895-6

- [10] L. Hégyely, P. Láng: New algorithm for the determination of product sequences in azeotropic batch distillation, *Computer Aided Chemical Engineering*, **29**, 281-285; Sithonia, 29 May – 1 June 2011, ISBN: 978-0-444-53895-6
- [11] P. Lang, L. Hegely, V. Gerbaud, Récupération de solvant d'un mélange à multiconstituant par distillation extractive discontinue et procédé hybride, *CIFQ-11*, 46-51, Chicoutimi, 20-22 June 2011, ISBN:978-2-9812621-0-3
- [12] L. Hegely, V. Gerbaud, P. Lang: Batch heteroazeotropic distillation with variable decanter hold-up: Feasibility studies, *22th European Symposium on Computer Aided Process Engineering (ESCAPE-22)*, 527-531, London, 17-20 June 2012, ISBN: 978-0-444-59431-0
- [13] L. Hegely, V. Gerbaud, P. Lang: Heterogeneous batch distillation with variable decanter hold-up, *11th International Symposium on Process System Engineering (PSE2012)*, 1607-1611, Singapore, 17-19 July 2012, ISBN: 978-0-444-59505-8

d. Article in Hungarian conference proceedings

- [14] L. Hegely, P. Lang, M. Lang Lazi: Investigation of Non-conventional Closed Batch Distillation Column Operation Modes, *Gépészet 2010*, 565-570, Budapest, 25-26 May 2010, ISBN 978-963-313-007-0
- [15] L. Hegely, P. Lang,: Simulation of batch extractive distillation laboratory experiments, *Gépészet 2010*, 484-489, Budapest, 25-26 May 2010, ISBN 978-963-313-007-0
- [16] Hégyely L., Láng P.: Szakaszos rektifikálás nem-hagyományos, zárt üzemeltetési módjainak vizsgálata, *Conference of Chemical Engineering '10*, 113-118, Veszprém, 27-29 April 2010, ISBN 978-963-9696-93-8
- [17] P. Lang, L. Hegely, Gy. Kovacs: The Influence of Recycle on the Solvent Recovery with Distillation, *Gépészet 2012*, 282-287, 24-25 May 2012.

Publications not related to the theses

- [18] L. Hegely, T. Balazs: Instrumentation and pH control of a neutralization vessel, *Gépészet 2010*, 466-471, Budapest, 25-26 May 2010, ISBN 978-963-313-007-0

- [19] G. Modla, L. Hégyel: Double Column Batch Stripper in Closed Mode for Pressure Swing Batch Distillation: I. Feasibility Studies, *Gépészet 2010*, 472-475, Budapest, 25-26 May 2010, ISBN 978-963-313-007-0
- [20] Modla G., Hégyel L.: Nyomásváltó szakaszos desztilláció dupla oszlopos rektifikálóban zárt üzemmódban: I. Megvalósíthatósági vizsgálatok, *Conference of Chemical Engineering '10*, 126-131, Veszprém, 27-29 April 2010, ISBN 978-963-9696-93-8

References

- [21] B.S. Ahmad, Y. Zhang, P.I. Barton, Product Sequences in Azeotropic Batch Distillation, *AIChE Journal* **44** (5), 1051-1070, (1998).
- [22] P. Lang, L. Hegely, G. Kovacs, Method for the recovery of methanol from multicomponent solvent mixtures, P 1200245, Hungarian Patent, (2012).