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DOCTORAL COUNCIL OF THE FACULTY OF MECHANICAL ENGINEERING

DOCTORAL THESIS BOOKLET

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**NEW DOUBLE-COLUMN SYSTEMS FOR BATCH
HETEROAZEOTROPIC DISTILLATION**

for obtaining the title of PhD

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Introduction and antecedents of the research

I joined in 2005 to the researches pursued in the field of separating techniques at the Department of Building Services and Process Engineering. At that time Dr. Péter Láng, dr. Gábor Modla, and Barnabás Kótai worked on the batch distillation methods. Before the beginning of my PhD studies (2007) I prepared my diploma work and two scientific student's works (a 1st and a 2nd award at the Faculty of Mechanical Engineering) in this topic.

As a 4th year student I took a poster presentation at the world congress Distillation & Absorption 2006. In 2008 I participated at a two-week summer school related to my topic at the Technical University of Berlin. In 2009 I spent 3 months in Toulouse, France at the department "Laboratoire de Génie Chimique" of the university INP – ENSIACET. My stay was financed by the French partner and I performed the first laboratory experiments related to my topic. From 2010 my PhD studies has been continued under joint supervision and I spent three times five months at the above department as a scholar of the French Government.

The efficient and economic separation of the mixtures forming azeotropes and of low relative volatility is a frequent problem in the chemical and food industries. The separation of these mixtures into their components in high purity is not feasible by using traditional distillation methods. The solution is the application of a special distillation method. In my PhD work I dealt primarily with the batch heteroazeotropic distillation. When mixtures of small quantity must be separated and the properties of the feed vary frequently the batch operational mode can be applied more successfully than the continuous one.

If the components of a mixture form a heteroazeotrope or they form a heteroazeotrope with a separating agent (entrainer) the azeotropic point can be crossed by decantation. In the pharmaceutical and fine chemical industries the batch processes are widely applied including also the batch heteroazeotropic distillation (BHD). To our best knowledge in the industry the BHD is applied only in Batch Rectifier equipped with a decanter (BR), in open operational mode (continuous top product withdrawal).

Because of the stricter and stricter environmental regulations and increasing energy prices the optimal design and operation of the distillation processes become more and more important. From the point of view of the environmental protection the efficient regeneration of waste solvent mixtures (whose components frequently form azeotropes) is very important. The recovery of the valuable components can be very advantageous also from economic point of view. The batch distillation is a highly energy-demanding operation, therefore a considerable

energy-saving can be realised by the right choice and optimisation of the geometrical and operational parameters.

The aim of my research is the study of the possibilities of realisation and of the efficiency of the heteroazeotropic distillation in batch mode. The methods applied were simplified and more detailed modelling and laboratory and pilot plant experiments, respectively. I performed studies for two new configurations, whose operation, energy demand and product recovery were compared with those of the batch rectifier (BR).

The new systems studied, unlike BR, contain two simultaneously operating columns and are operated in closed mode, that is, without continuous product withdrawal. The columns of the basic type (double-column system, DCS) have a common total condenser and decanter (*Fig. 1*). In its generalised version (generalised double-column system, GDCS) each column has its own total condenser and the decanter belongs only to one of the columns (*Fig. 2*).

Operation of the DCS (in the case of a binary heteroazeotropic mixture A-B)

One part of the charge is filled into one of the reboilers, the other part is filled into the other one. In the case of heterogeneous charge each phase is fed into separate reboilers: the phase rich in component *A* into the reboiler of Column α (producing *A*), the phase rich in component *B* into the reboiler of Column β (producing *B*).

The top vapours of the two columns (of nearly azeotropic composition) are led into a common condenser. The condensate arrives at the common decanter from where the *A*-rich phase is led into Column α and the *B*-rich one into Column β , respectively.

At the end of the process the product *A* is obtained in Reboiler α and the product *B* is in Reboiler β , respectively. The binary heteroazeotropic mixture is referred henceforth as binary mixture.

Operation of GDCS (in the case of A-B binary homoazeotropic mixture with entrainer E)

The charge is divided between the two reboilers then the entrainer is filled into Reboiler α . The composition of the top vapour of Column α is near to the ternary, that of Column β is near to the *A-B* binary azeotropic one, respectively. The heterogeneous condensate of Column α is led into the decanter. The light (*E*-rich, organic) phase is fed back into Column α , the heavy (*B*-rich, aqueous) one is fed into Column β , respectively. One part of the homogeneous condensate of Column β is refluxed, the other part is fed into the other column. At the end of

the process the product A is obtained in Reboiler α and the product B is in Reboiler β , respectively. The greater part of E remains in the decanter.

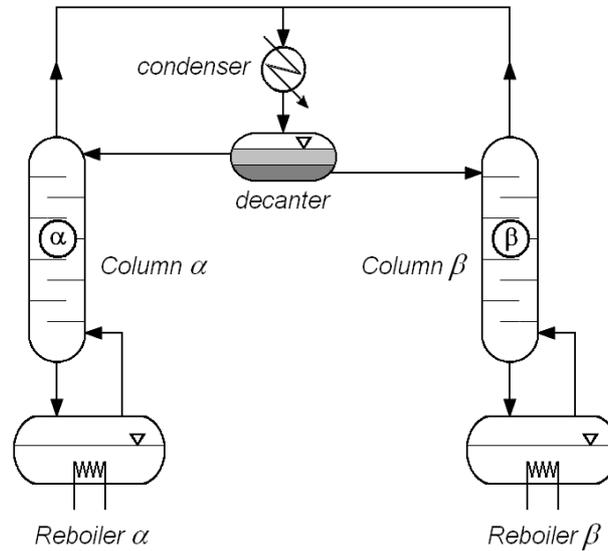


Fig. 1. Scheme of the Double-Column System (basic version)

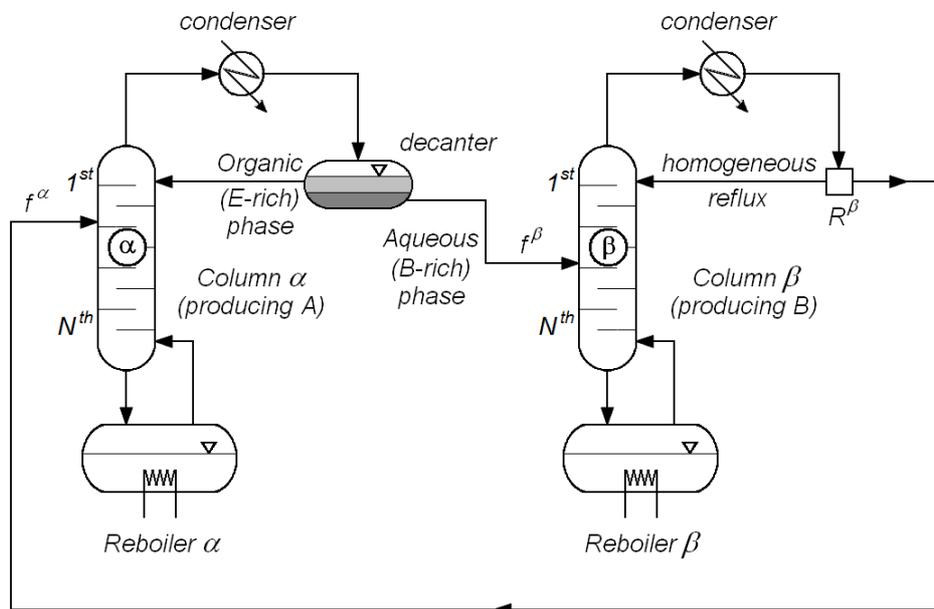


Fig. 2. Scheme of the Generalised Double-Column System

Methods of investigation

The operation of each configuration was studied by the following methods:

- feasibility study (on the basis of a simplified model),
- rigorous simulation,
- laboratory and pilot plant experiments.

The aim of the *feasibility study* is to give estimation for the duration of the process and for the quantity of the products. A simplified model based on the following assumptions was applied:

- maximal separation,
- negligible liquid and vapour holdups on the trays,
- constant liquid holdup in the decanter (overall quantity of components),
- negligible vapour holdup in the decanter,
- constant molar overflow,
- constant reboiler heat duty,
- homogeneous liquid streams leaving the decanter,
- in the case of BR:
 - negligible duration of pumping between the operation steps,
 - negligible loss of entrainer in the case of the binary homoazeotropic charge + entrainer (henceforth ternary mixture).

By the *rigorous simulation* the effects of the most important operation parameters were studied and the minimum duration of each configuration and the quantities of products were determined.

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,
- negligible duration of pumping between the operation steps.

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, holdup and physical property models).

The phase equilibria are described by the NRTL model for the binary mixture and by the UNIQUAC model for the ternary mixture, respectively. For the solution of the equations above the dynamic simulator (CC-DCOLUMN) of the program ChemCAD (versions 5.6-6.4) was used. The ChemCAD model of the DCS is shown in *Fig. 3*.

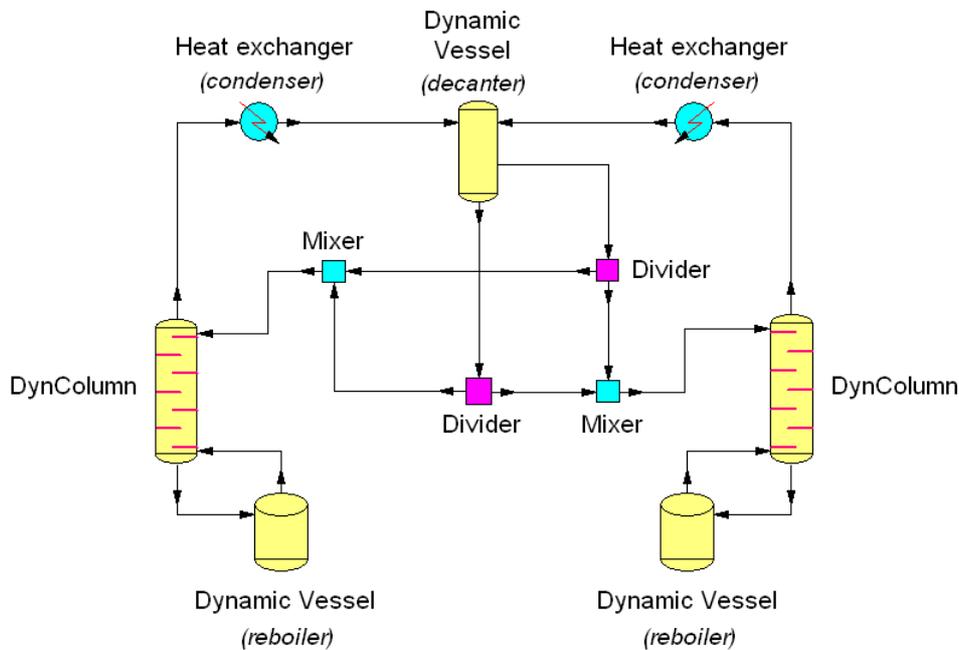


Fig. 3. ChemCAD model of the DCS

The *experimental validation* of the calculation results was done at the ENSIACET university in Toulouse with two different equipments: first on a small size laboratory equipment then in a pilot plant.

The elements of the *laboratory equipment* (*Fig. 4*) are made of glass. The internal diameter of the columns is 30 mm, the height of the packing (Raschig rings of 7 mm) is 1 m. The columns are double-walled and thermally insulated by foamed polymer. The common condenser is a water-cooled spiral cooler. The decanter is thermally insulated where the volumetric liquid holdup is 55 cm³. The volumes of the three-neck flasks used as reboilers are 1000 and 2000 cm³, respectively. The maximum performances of their electric heating mantles are 300 and 450 watts. The liquid temperatures in the reboilers were measured with thermometers. The compositions of the products were determined on the basis of their refractive indices.

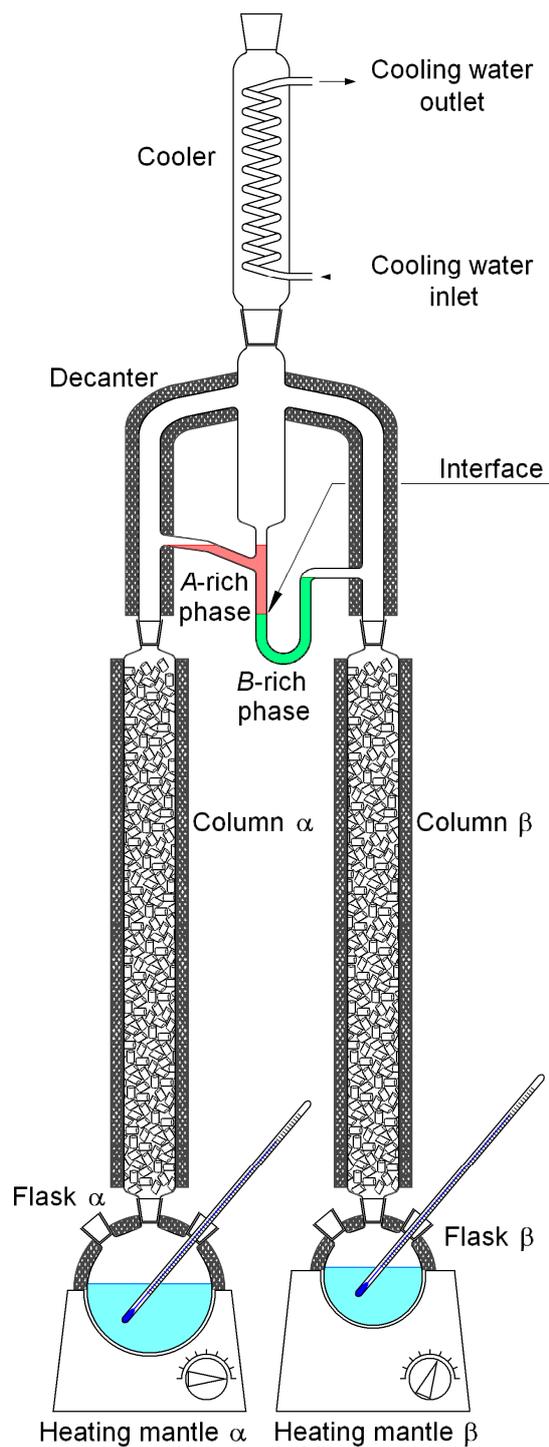


Fig. 4. Scheme of the laboratory distillation equipment

The laboratory equipment served first of all for the experimental verification of the feasibility of the process.

In order to get more information about the operation and for the sake of the more flexible operation we built a *pilot plant equipment* (Fig. 5). Its elements are also made of glass. Both

columns consist of 4 packed sections of height 1 m. The nominal internal diameters of the columns are 80 mm. The types of the metal packings are Sulzer CY (Column α) and Kühni Rombopak (Column β), respectively. Both columns are equipped with a reflux divider. Both columns have a separate 3- (Column α) or 4-pass (Column β) water-cooled spiral cooler and an aftercooler. The volume of the holdup of the decanter is 3.6 dm³. The volumes of the reboilers are 10 (Column α) and 11 dm³ (Column β), respectively. They are heated by double and quaternary glass spirals, in which oil circulated, which is heated by electric heating. The maximum performances of the electric heatings equipments are 6 (Column α) and 4 kW (Column β), respectively. The columns and the reboilers are thermally insulated. The temperature was measured continuously in the reboilers, in all column sections, at the tops of both columns and in the decanter. The measured data were registered by data loggers. The product compositions were determined on the basis of their refractive indices (binary mixture) or by gas chromatography (ternary mixture).

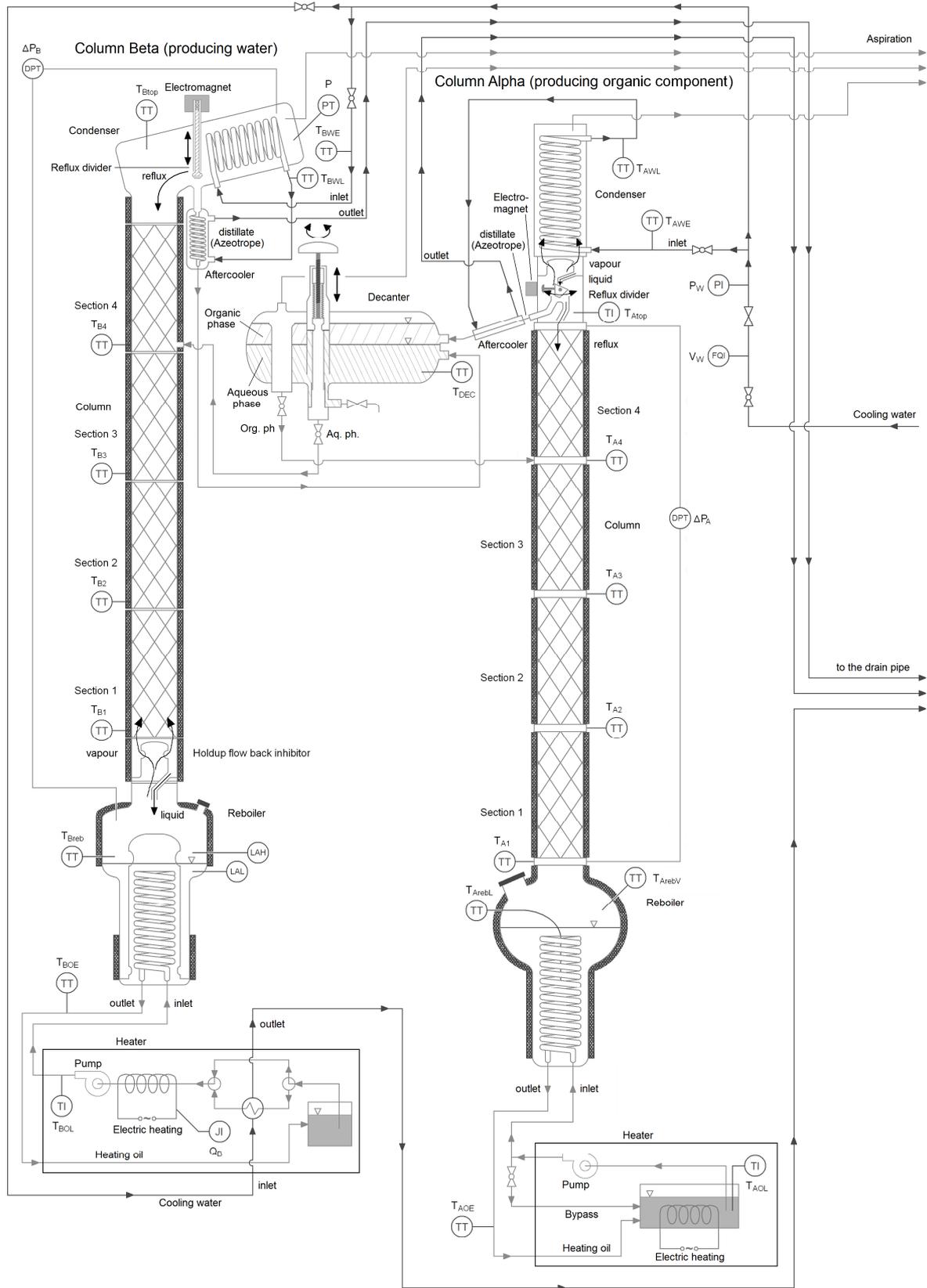


Fig. 5. Scheme of the pilot plant distillation equipment

New scientific results

We developed two new double-column systems for the batch heteroazeotropic distillation of binary heteroazeotropic and homoazeotropic mixtures. For the new configurations and the BR I performed feasibility studies, rigorous computer simulations, and laboratory and pilot plant experiments, respectively. I compared the efficiency of the different configurations. (The test mixtures were 1-butanol – water and isopropanol – water + benzene/cyclohexane/n-hexane entrainer, respectively.)

In addition I deduced an inequality for the determination of the product sequence resulting in less byproduct in the case of the processing of a two-phase binary heteroazeotropic mixture in the BR. I compared the efficiency of the different entrainers used for the separation of the homoazeotropic mixture.

Thesis 1

Related publications: [1], [6], [7], [10], [11]

We suggested a new Double-Column System (DCS) suitable for batch heteroazeotropic distillation. With this configuration components *A* and *B* of a binary heteroazeotropic mixture or a binary homoazeotropic one by adding entrainer can be produced simultaneously.

I developed a simplified model containing component and total material balances, on the basis, of which I made feasibility studies. I solved analytically the material balances for one of the columns and for the decanter. In the case of the DCS I supposed that both products reached the prescribed purity at the same time, that is, the duration of the process is minimal.

The duration of the BR was determined for binary mixtures of different compositions (homogeneous charge rich in *A*, heterogeneous charge, homogeneous charge rich in *B*). For the DCS I determined how the division of the charge between the two reboilers (ratio of the quantity of the liquid charged into the reboiler of Column α and the quantity of the total charge, U_b^α/U_{ch}) influences the division of the total vapour flow rate (ratio of the vapour flow rate of Column α and that of the two columns together, $v^\alpha = V^\alpha/V$, *Fig. 6*), resulting in the minimum duration which was also calculated (τ , *Fig. 7*). For the ternary mixture I determined the duration of the BR and how the division of the total vapour flow rate between the two columns influences the division of charge resulting in the minimum duration.

The new DCS configuration proved to be feasible on the basis of the results of the feasibility studies.

In the laboratory size Double-Column System I realised successfully the separation of the two-phase mixture 1-butanol – water ($x_{ch,A} = 0,256$), thus I proved experimentally the feasibility of the DCS, too.

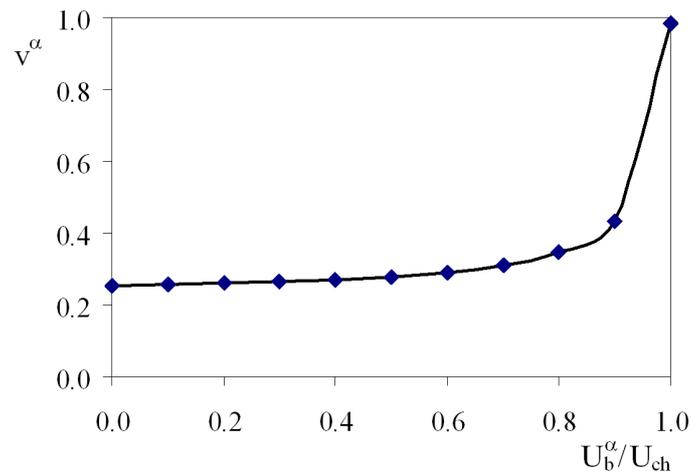


Fig. 6. The variation of the optimal relative vapour flow rate of Column α in the function of the division of charge (charge: 90 mol% of 1-butanol – 10 mol% of water)

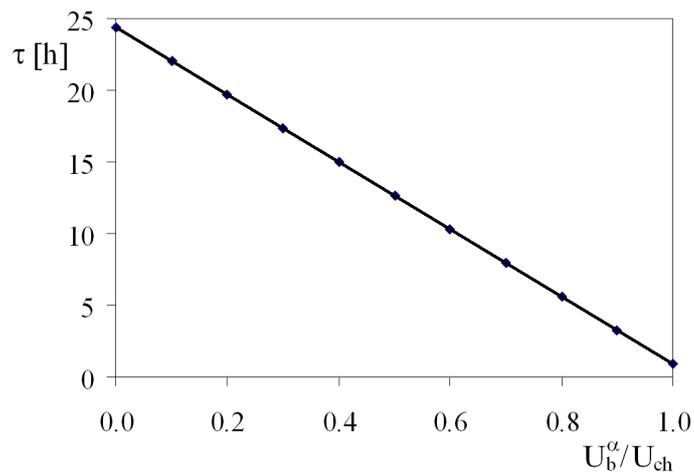


Fig. 7. The duration of the process in the function of the division of charge (charge: 90 mol% of 1-butanol – 10 mol% of water)

Thesis 2

Related publications: [1], [7], [9], [13], [14]

The new Double-Column System was compared with the traditional Batch Rectifier (BR) equipped with a decanter. The comparison was done first on the basis of the results of the feasibility studies, then of those of the rigorous simulation and finally with laboratory experiments, respectively.

During the feasibility studies the total vapour flow rates of the two configurations were equal. I concluded that the Double-Column System was competitive with the traditional Batch Rectifier. During nearly the same time by using the DCS the product recoveries were higher and there was no byproduct (*Table 1*).

		BR			DCS	
		Step 1	Step 2	Total	Column α	Column β
Division of charge	kmol	-	-	-	51.8	48.2
Division of vapour flow rate	-	-	-	-	0.9530	0.0470
Duration	hour	2.006	0.100	2.106	2.141	
Product A	kmol	29.298	0.000	29.298	29.798	0.000
Product B	kmol	0.000	69.823	69.823	0.000	70.202
Byproducts	kmol	-	0.879	0.879	0.000	
Byproduct composition	mol% A	-	56.80	56.80	-	

Table 1. Comparison of the DCS and BR on the basis of the feasibility studies in the case of a heterogeneous binary charge (30 mol% of 1-butanol)

In the rigorous simulation the total heat duties and the total numbers of trays of the two configurations were equal. For the DCS I determined the optimum division of the number of trays, of the charge and of the total heat duty resulting in the minimum duration. For the ternary mixture for both configurations the optimum quantity of the entrainer was also determined. I concluded that similarly to the results of the feasibility studies, the DCS was competitive with the traditional Batch Rectifier. By using the DCS during nearly the same

time the recovery of the organic (more valuable) component was higher in each case (*Table 2*).

		BR			DCS	
		Step 1	Step 2	Total	Column α	Column β
Number of trays	-	10	10	-	5	5
Division of charge	kmol	-	-	-	51.8	48.2
Heat duty	kW	200	200	-	191.2	8.8
Duration	hour	3.15	0.18	3.33	3.50	
Product A	kmol	24.948	0.000	24.948	26.948	0.000
Product B	kmol	0.000	65.851	65.851	0.000	65.449
Byproducts	kmol	6.492	2.709	9.201	7.603	

Table 2. Comparison of the DCS and BR on the basis of the rigorous simulation in the case of a heterogeneous binary charge (30 mol% of 1-butanol)

The separation of the mixture 1-butanol – water of azeotropic composition was also done by operating the laboratory size equipment – beside the DCS – as a traditional Batch Rectifier. By comparing with the BR for the DCS the recoveries of both components were higher (*Table 3*). (Therefore the quantities of the byproducts were lower.)

			BR	DCS
Product A	Duration	min	197	200
	Volume	ml	1110	1230
	Mole fraction of A	-	1.000	1.000
	Recovery	%	87.4	96.9
Product B	Volume	ml	620	710
	Mole fraction of A	-	0.000	0,000
	Recovery	%	84.9	97.3

Table 3. The most important results of the laboratory experiments

Thesis 3

Related publications: [3]

I compared the efficiency of the DCS with that of the BR by rigorous simulation in the case of homoazeotropic mixture, by applying cyclohexane entrainer instead of benzene.

I concluded that if cyclohexane was applied as entrainer instead of benzene during the processing of isopropanol – water mixture then the advantages of the DCS over the BR were even greater (*Table 4*). By using cyclohexane the duration was much shorter for the DCS and the recoveries of the products were much higher. In the columns of *Table 3* concerning the DCS there are more data because several versions of this configuration were studied (e.g. the total number of trays or the number of trays of one column is equal to that of the BR).

		Benzene		Cyclohexane	
		BR	DCS	BR	DCS
Duration	h	36.45	34.0	37.9	28.5
			35.0		28.8
			33.7		
Product A	kmol	55.97	58.70	50.75	59.33
			57.52		58.78
			59.45		
Product B	kmol	23.59	29.32	21.30	25.46
			23.99		26.16
			26.06		

Table 4. Comparison of the different configurations by applying different entrainers

Thesis 4

Related publications: [1]

I deduced an inequality for the determination of the product sequence resulting in less byproduct for the separation of a heterogeneous binary (A - B) heteroazeotropic mixture in the Batch Rectifier. This mathematical formula is deduced from the material balances. It contains the composition of the charge, the mutual solubilities and the prescribed product purities as variables.

If the following inequality is satisfied then the A -rich phase must be processed (and A must be produced) first:

$$x_{ch,A} \geq \frac{\frac{x_A^{ProdA}}{x_A^{Ar}} - \frac{1 - x_B^{ProdB}}{x_A^{Br}}}{\left(x_A^{ProdA} - x_A^{Ar}\right) - \left(1 - x_B^{ProdB} - x_A^{Br}\right)} \cdot x_A^{Ar} \cdot x_A^{Br} = x_{ch,A}^{min} \quad (1)$$

where $x_{ch,A}$ is the mole fraction of component A in the charge,
 x_A^{Ar}, x_A^{Br} is the mole fraction of component A in the A -rich and B -rich phases,
 x_A^{ProdA}, x_B^{ProdB} is the mole fraction of the main component in each product.

It must be noted, that the above rule is independent from the composition of the top vapour, that is, it does not have to be the azeotropic one.

If pure products are assumed ($x_A^{ProdA} = 1, x_B^{ProdB} = 1$) Inequality 1 can be simplified to the following form:

$$x_{ch,A} \geq \frac{1}{1 + \frac{1 - x_A^{Ar}}{x_A^{Br}}} = x_{ch,A}^{min} \quad (2)$$

Thesis 5

Related publications: [2], [4], [5], [8]

We extended the DCS to a Generalised Double-Column System (GDCS, *Fig. 2*) of higher degree of freedom. It can be operated in a more flexible way because the feed tray locations of Columns α and β can be varied and the value of the reflux ratio of Column β is also arbitrary. I extended the feasibility method applied earlier for the GDCS and I performed feasibility studies.

According to the results of this study the configuration DCS also proved to be feasible.

By rigorous simulation I studied the effect of the three above new operational parameters on the duration (the energy demand of the process) for three different charge compositions by using cyclohexane as entrainer.

By rigorous simulation applying the downhill simplex optimisation method I determined the minimum duration and the optimum values of the new operation parameters for three different charge compositions and for two different entrainers (cyclohexane, n-hexane).

I concluded from the studies, that:

- The optimum feed location in Column α is in the top section of the column.
- The optimum feed location in Column β is in the lower section of the column.
- The optimum value of the reflux ratio in Column β is low ($R^\beta < 1$).

The durations for the DCS were also determined for the above six cases. By comparing the DCS and the GDCS I concluded that the durations for the GDCS were always shorter (*Fig. 8*) and in 11 cases of the 12 the specific energy demand of the products were lower for the GDCS (*Fig. 9*).

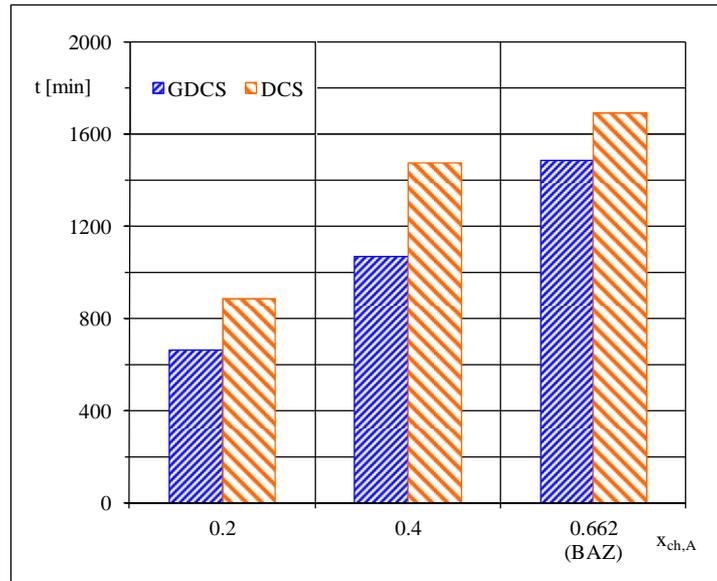


Fig. 8. Minimum duration for each configuration (entrainer: cyclohexane)

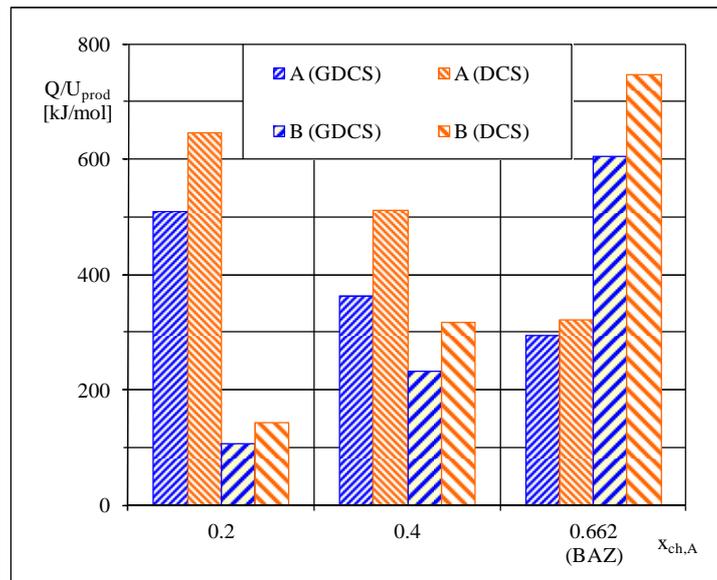


Fig. 9. Specific energy demand for each product and configuration
(A: isopropanol, B: water, E: cyclohexane)

Thesis 6

Related publications: [2]

I compared the possible entrainers for the separation of the mixture isopropanol – water on the basis of different properties determined by the vapour-liquid and liquid-liquid equilibrium relationships and of physical properties (*Table 4*).

If n-hexane is used as entrainer instead of the cyclohexane:

- I. The isopropanol content of the ternary azeotrope (TAZ) is lower.
- II. The tie line passing through the TAZ point is longer.
- III. The division of isopropanol between the organic and aqueous phases is more favourable.
- IV. Higher ratio of the mole fractions of water and isopropanol in the ternary azeotrope

$\left(\frac{x_{TAZ,B}}{x_{TAZ,A}} \right)$ is more favourable. For the more efficient separation it is better if in the

aqueous phase of the ternary azeotrope withdrawn from Column α the mole fraction ratio

of B and A $\left(\frac{x_{TAZ,B}^{AQ}}{x_{TAZ,A}^{AQ}} \right)$ is also the high. By the multiplication of these ratios a selectivity

index can be defined:

$$SI = \frac{x_{TAZ,B}}{x_{TAZ,A}} \cdot \frac{x_{TAZ,B}^{AQ}}{x_{TAZ,A}^{AQ}}$$

The SI-index is much higher for the n-hexane.

- V. Higher $R^\alpha = \frac{n_{TAZ}^{ORG}}{n_{TAZ}^{AQ}}$ can be ensured by refluxing only the organic phase. (R^α is the molar

ratio of the quantities of the organic and aqueous phases of the TAZ.)

- VI. In the case of the DCS the condensate remains in the heterogeneous region at higher ratio of the molar flow rate of the binary to that of the ternary azeotropes (BAZ/TAZ).
- VII. The difference of the densities of the two equilibrium liquid phases ($\Delta\rho$) is much higher.

I concluded that the n-hexane is more advantageous from all aspects than the cyclohexane.

From the most aspects n-hexane is more advantageous than the benzene, as well (*Table 5*).

	I	II	III	IV	V	VI	VII
	$x\%_{TAZ,A}$	$l_{TAZ-line}$	$\frac{n_{TAZ,A}^{ORG}}{n_{TAZ,A}^{AQ}}$	SI	R^α	$max. \frac{n_{BAZ}}{n_{TAZ}}$	$\Delta\rho$
	mol%	mol%	-	-	-	-	kg/m ³
Cyclohexane	24.8	65.8	3.57	2.43	3.79	0.59	83
N-hexane	19.1	73.2	11.58	6.44	6.86	0.77	232
<i>Benzene</i>	23.8	69.4	2.71	8.76	3.43	1.81	70

Table 5. Comparison of the different entrainers on the basis of the phase equilibrium relationships

Application possibilities of the results, further tasks

By the new double-column batch heteroazeotropic distillation configurations (DCS, GDCS) studied by me it becomes possible to separate simultaneously the components of binary liquid mixtures forming an azeotrope. In the chemical and pharmaceutical industry the waste solvent mixtures often are of this type. They can be regenerated by this process and thus the reuse of the valuable components becomes possible.

Comparing with the BR with the new configurations in the most cases the products were produced more quickly. This indicates that significant energy saving can be reached. The duration of the process in the new configuration never exceeded that of the BR.

Columns already existing in a batch plant can be connected easily to a DCS or GDCS, that is, the new configurations do not need any additional investment. For the DCS the columns themselves need not to be modified and for the GDCS only one feeding facility at the lower part of Column β must be ensured. If necessary they can be separated again to BRs without any mounting, just by the switching of some valves. However, the operation of the new configurations is somewhat more complicated compared to that of the BR (division of charge and heat duty).

After the abolishment of the defects of the pilot plant equipment it is worthy to do further experiments for the binary homoazeotropic mixture.

Publications related to the theses

a. Article in international journal of IF

- [1] Denes, F., Lang, P., Modla, G., Joulia, X.: “New double column system for heteroazeotropic batch distillation”, *Computers and Chemical Engineering*, **33**, 1631-1643 (2009).
- [2] Denes, F., Lang, P., Joulia, X.: “Generalised closed double-column system for batch heteroazeotropic distillation”, *Separation and Purification Technology*, **89**, 297-308 (2012).

b. Article in international journal

- [3] Denes, F., Lang, P., Joulia, X.: “Heteroazeotropic batch distillation in a new double-column system”, *Chemical Engineering Transactions*, **18**, 713-718, (2009).
- [4] Denes, F., Lang, P., Joulia, X., “Batch Heteroazeotropic Distillation in a New Generalised Double-Column System”, *Chemical Engineering Transactions*, **25**, 725-730 (2011).

c. Article in international conference proceedings

- [5] Denes, F., Lang, P., Joulia, X., “Nouveaux systèmes de double colonne pour la distillation hétéroazéotrope discontinue”, *CIFQ2011 sur La Thermique des Systèmes*, 40-45 (2011).
- [6] Lang, P., Denes, F., Joulia, X.: “New Configuration for Heteroazeotropic Batch Distillation: I. Feasibility Studies”, *ESCAPE-18, Computer-Aided Chemical Engineering*, **25**, 115-120 (2008).
- [7] Denes, F., Lang, P., Joulia, X., “Experimental validation of a new double-column system for heteroazeotropic batch distillation”, *Distillation & Absorption 2010, IChemE Symposium Series*, 289-294 (2010).
- [8] Denes, F., Lang, P., Joulia, X., “New generalised double-column system for batch heteroazeotropic distillation”, *ESCAPE-21, Computer-Aided Chemical Engineering*, **29**, 366-370 (2011).

- [9] Denes, F., Lang, P., Modla, G., Joulia, X.: “New Configuration for Heteroazeotropic Batch Distillation: II. Rigorous Simulation Results”, *ESCAPE-18, Computer-Aided Chemical Engineering*, **25**, 6 oldal CD-n (2008).

d. Article in hungarian conference proceedings

- [10] Lang, P., Denes, F., Joulia, X.: “Új szakaszos heteroazeotrop rektifikáló rendszer: I. Megvalósíthatósági vizsgálatok”, *Műszaki Kémiai Napok '08*, 77-83 (2008).
- [11] Lang, P., Denes, F., Joulia, X.: “New Configuration for Heteroazeotropic Batch Distillation: I. Feasibility Studies”, *Gépészet 2008*, 12 oldal CD-n (2008).
- [12] Denes, F., Lang, P., Joulia, X.: “Validation of a new double-column system for heteroazeotropic batch distillation”, *Gépészet 2010*, 559-564 (2010).
- [13] Denes, F., Lang, P., Modla, G.: “New Configuration for Heteroazeotropic Batch Distillation: II. Rigorous Simulation Results”, *Gépészet 2008*, 8 oldal CD-n (2008).
- [14] Lang, P., Denes, F., Modla, G.: “Új szakaszos heteroazeotrop rektifikáló rendszer: II. Részletes modellezési eredmények”, *Műszaki Kémiai Napok '08*, p. 312-318 (2008).

