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# Validation of a New Double-Column System for Heteroazeotropic Batch Distillation by Experiments and Dynamic Simulation

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## Abstract

The separation of a binary heteroazeotropic mixture (1-butanol – water) by batch distillation is studied in a new Double-Column System (DCS) by pilot plant experiments and dynamic simulation performed with a professional flowsheet simulator. The DCS is operated in closed mode. At the end of the process products of high purity were obtained with high recoveries in the reboilers. However the limited facility of setting accurately the ratio of the reboiler heat duties caused difficulties concerning the optimal operation of the new system.

**Keywords:** batch heteroazeotropic distillation, closed system, experiment, dynamic simulation

## 1. Introduction

If the components of a mixture form a heteroazeotrope, or by the addition of an entrainer a heteroazeotrope can be formed, the separation can be performed by heteroazeotropic distillation (HAD). So far the batch heteroazeotropic distillation was applied in the industry only in batch rectifiers (equipped with a decanter, BR) in open operation mode. Different operation modes of the batch HAD and their strategies are presented in the book of Luyben and Chien (2010). The batch HAD is studied experimentally among others by Ooms et al. (2013, ethyl acetate – isooctane by using methanol and acetonitrile as entrainer).

We developed a new batch double-column HAD configuration (double-column system, DCS, Figure 1) operated in closed mode (without continuous product withdrawal), (Denes et al., 2009). The results of feasibility studies and dynamic simulation calculations showed that comparing with the BR significant reduction of the energy demand can be achieved. Then the DCS was experimentally validated for the separation of a binary heteroazeotropic mixture in a simple laboratory glass equipment (Denes et al., 2010). After that the DCS was extended to a generalised double-column system (GDCS, Denes et al., 2012) for the batch HAD separation of binary homoazeotropic mixtures by using entrainer. The GDCS has three further degrees of freedom compared with the DCS (locations of two continuous feedings, reflux ratio in Column  $\beta$ ) and is more advantageous for the separation of binary homoazeotropic mixtures with a heterogeneous entrainer.

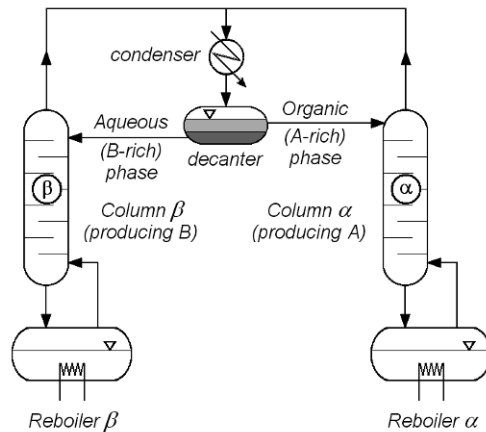


Figure 1. Theoretical scheme of the DCS for the separation of a binary heteroazeotropic mixture

In the next a pilot plant was installed, by which comparing with the laboratory column much more information can be acquired about the process (temperature profiles, pressure drop etc.), and whose operation is more flexible (variable reflux ratio) and appropriate (higher decanter holdup). The goal of this paper is

- to validate the DCS by pilot plant experiments,
- to evaluate the results with dynamic simulation.

## 2. Pilot plant experiment

### 2.1. The pilot plant equipment and its operation

The columns ( $D = 80$  mm,  $H = 4 \times 1$  m) of the pilot plant equipment (Figure 2) are filled with structured packing: Sulzer CY (Column  $\alpha$ ), Kühni Rombopak (Column  $\beta$ ). Both columns have an own condenser, reflux divider and aftercooler. Hence both condensates can be refluxed bypassing the decanter, too. The heating medium (oil) is circulated and heated by electricity (maximal performances:  $Q_\alpha = 6$  kW,  $Q_\beta = 4$  kW). The temperature of the oil entering into the reboiler heating spiral is set.

The charge (of  $18.6$  °C) contains 1-butanol ( $A$ ,  $7.5$  dm<sup>3</sup>) and water ( $B$ ,  $9.5$  dm<sup>3</sup>). Into the reboilers  $8.0$  dm<sup>3</sup> saturated organic and  $9.0$  dm<sup>3</sup> aqueous phases are filled. The decanter is filled with heterogeneous liquid (Figure 2). The two columns are operated simultaneously. There are three operational steps:

1. Boiling-up: until the liquid in the reboiler begins to boil.
2. Heating-up: until the vapour arrives at the top of the column.
3. Purification: until the end of the process.

In the boiling-up and heating-up steps both columns are operated under total reflux. Since Heater  $\alpha$  generates too high vapour flow rate, therefore 60 % of the condensate was directly refluxed in order to avoid the too short residence time in the decanter.

The purification steps of both columns were started at the same time. However, the durations of the boiling-up and heating-up steps were different because of the limited controllability of the reboiler heat duties.

The charge in Reboiler  $\alpha$  would have boiled up much earlier than in the other reboiler therefore its heating was strongly reduced until the heating-up of the other column

approached to the end (for 76 min). Table 1 shows the lengths of the different operation steps. (The optimal lengths if they are different are in parentheses.) The experiment was finished when either reboiler temperature did not significantly increase more.

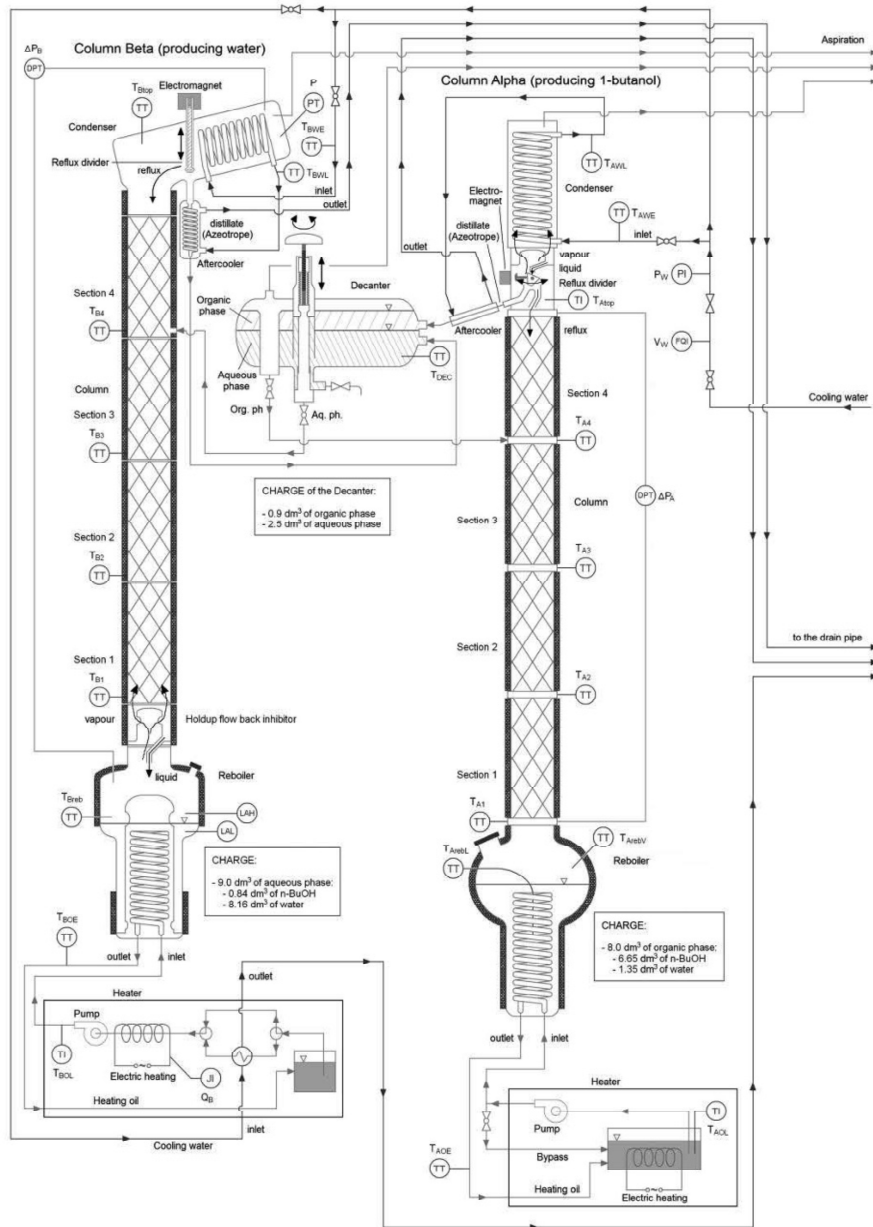


Figure 2. DCS pilot plant equipment for the separation of 1-butanol – water

Table 1. Duration of the different operation steps [min]

Step	Column $\alpha$		Column $\beta$	
	Time at the end	Duration	Time at the end	Duration
Boiling up	148	148 (53)	93	93
Heating-up	175	27	175	82 (63)
Purification	357	182	357	182

## 2.2. Experimental results

The reboiler temperatures (Figure 3) verify that products of very high purity were obtained at the end of the purification step with very high recoveries: 1-butanol: 99.9 mass%, 7.4 dm<sup>3</sup>; water: 99.9 mass%, 9.5 dm<sup>3</sup>. At the end of heating-up both reboiler temperatures are near to the azeotropic one.

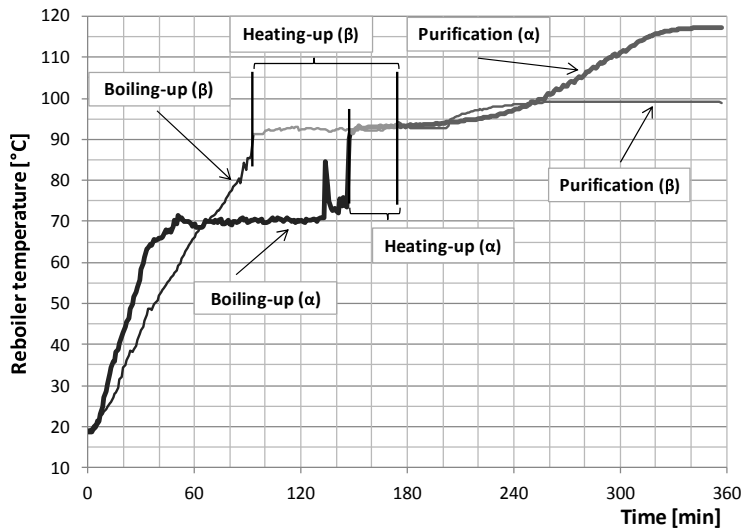


Figure 3. Evolution of the reboiler temperatures (experiment)

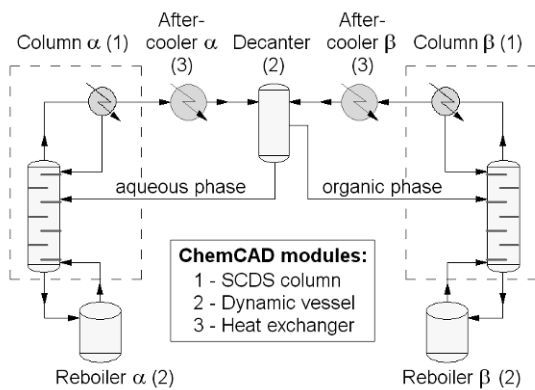


Figure 4. ChemCAD model of the DCS

### 3. Posterior simulation

#### 3.1. Simulation method

We performed rigorous simulation applying the following simplifying assumptions: theoretical trays, constant volumetric liquid holdup on the trays and in the decanter, negligible vapour holdup. The model equations to be solved are non-linear ODEs (material and heat balances) and non-linear algebraic equations (VLE, hold-up and physical property models etc.). The phase equilibrium is described by the NRTL model (BIPs,  $K^1$ : 215.427 and 1468.34;  $\alpha = 0.3634$ ). For the solution of the above equations the dynamic simulator of ChemCAD 6.4 (CCDCOLUMN) is applied. The model of the pilot plant is shown in Figure 4.

#### 3.2. Input data

The number of theoretical stages of the columns (of holdup  $25 \text{ cm}^3/\text{stage}$ ) without condenser, decanter and reboiler is estimated to 20. The levels of the liquid phases (of  $60 \text{ }^\circ\text{C}$ ) are kept constant in the decanter. The net heat duties of the reboilers ( $Q_{reb}^{net}$ ) are so chosen to give the same durations of the operation steps as in the experiment (Figure 5). The calculations are started with dry plates. The boiling-up step of Column  $\alpha$  was started by 76 minutes later in order to avoid the “waiting period” of the experiment (Figure 5, Table 2).

Table 2. Operation steps in the simulation

No.	1	2	3	4	5
Column $\alpha$	No heating	Boiling-up		Heating-up of the column	Purification
Column $\beta$	Boiling-up		Heating-up of the column		

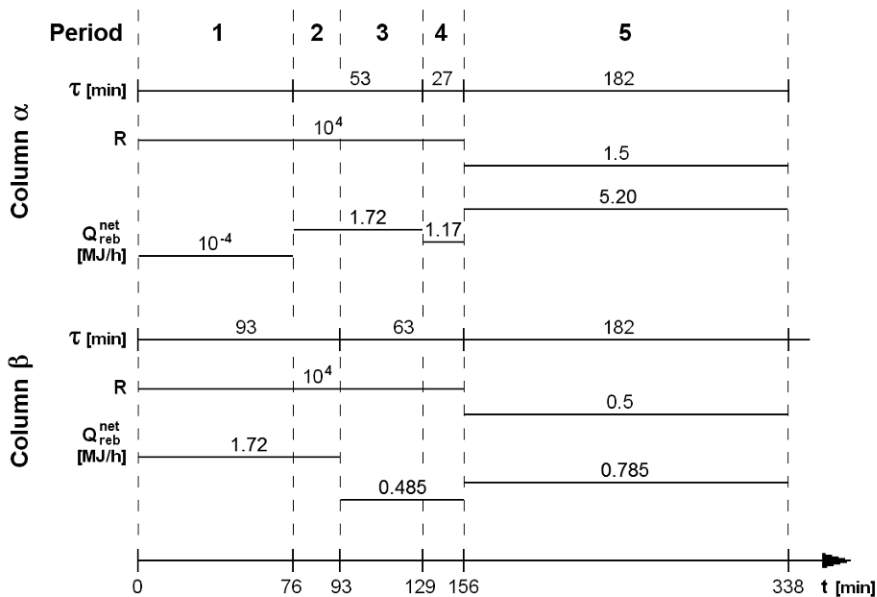


Figure 5. Operational parameters in the different operation steps

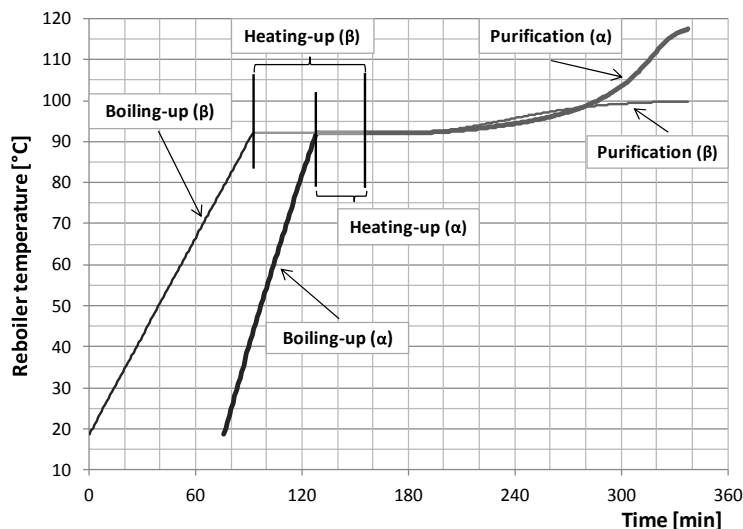


Figure 6. Evolution of the reboiler temperatures (simulation)

### 3.3. Results

The posterior simulation verified the production of high purity products: 1-butanol: 99.9 mass%, 7.72 dm<sup>3</sup>; water: 99.9 mass%, 9.48 dm<sup>3</sup>. The product quantities and qualities are in good agreement with those of the experiment. Figure 6 shows the evolution of the reboiler temperatures.

## 4. Conclusions

The separation of the mixture 1-butanol – water by batch heteroazeotropic distillation is studied in a new closed Double-Column System (DCS) by pilot plant experiments and posterior dynamic simulation with ChemCAD 6.4. At the end of the process products of high purity were obtained in the reboilers with high recoveries. However the experiment highlighted the importance of the accurate control of the reboiler heat duties in order to operate the DCS at optimal conditions and to avoid the loss of time and energy. This is even more important for the GDSCS used for the separation of a binary homoazeotropic mixture with a heterogeneous entrainer where the composition of the condensate may get outside the heterogeneous region if the ratio of the heat duties is not appropriate.

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