CLOSED OPERATION OF MULTIVESSEL BATCH DISTILLATION - EXPERIMENTAL VERIFICATION

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ABSTRACT. The multivessel batch distillation column, as well as conventional batch distillation, may be operated in a closed (total reflux) mode where the products are collected in vessels along the column. We have previously proposed and simulated a feedback control strategy for the closed operation, where the idea is to indirectly adjust the vessel holdups by using the reflux flow out of the vessel to control the temperature at some location in the column section below. The feasibility of this scheme is here demonstrated experimentally on a laboratory scale multivessel column. The experimental column consists of a reboiler, two intermediate vessels and accumulator, where we separate a mixture of methanol-ethanol-propanol-butanol into almost pure components.

The paper presents the first published experimental work on the closed operation of batch distillation, as well as the first published results on the operation of a multivessel column.

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INTRODUCTION

In this paper we study the closed ("total reflux", "redistributive") operation of a multivessel batch distillation column with temperature control. The aim is to confirm experimentally the feasibility of this method of operation which was proposed by Skogestad *et al.* (1997). Some early experimental results were presented in Wittgens *et al.* (1996). Since the multivessel column provides a generalization of a conventional batch distillation column, the results in the paper also demonstrate how a conventional column may be operated in a closed mode.

For conventional bath distillation, the closed operation, where the two final products are collected in the condenser drum (accumulator) and reboiler, was suggested independently by Treybal (1970) and Bortolini and Guarise (1970). Treybal writes that he first learned about the technique from Gustison in 1958, and "has found it most useful" and that it "practically runs the distillation by itself".

The generalization of the closed operation of conventional batch distillation to the case with several vessels along the column (the multivessel column) was proposed by Hasebe *et al.* (1995). With n vessels along the column (including reboiler, condenser and intermediate vessels), it is possible in the multivessel column to obtain n pure products in a single batch, and it was also found that the energy efficiency of this scheme is very good.

Treybal (1970) proposes, as do all other authors except Skogestad *et al.* (1997) that, following the initial startup period, the accumulator holdup (level) should be kept constant during the operation using a level control system. However, this way of operation is sensitive to errors in the feed composition (from which the level setpoint is precomputed) and to errors in the control of the level. To correct this, one may introduce a correction on the level setpoint based on composition measurements (Bortolini, 1970 and Hasebe *et al.*, 1995), but this makes the control system complicated and requires on-line composition measurements. To avoid these problems, Skogestad *et al.* (1997) suggest to *indirectly* adjust the accumulator holdup (level) by using the reflux to control the temperature at some location in the column section below (see Figure 1).

They show through simulations that this simple way of operation works very well, but there has been raised concerns about whether it would work in practice, especially for the multivessel column. The main contribution of this paper is therefore to demonstrate the practicability of the closed operation with indirect level control on a laboratory scale multivessel batch distillation column.

EXPERIMENTAL MULTIVESSEL COLUMN

A laboratory scale multivessel batch distillation unit (see Figure 2) was build to perform the experiments needed to verify the proposed control strategy. The chemical system studied is methanol (boiling point $T_{b1} = 64.7^oC$), ethanol ($T_{b2} = 78.3^oC$), n-propanol ($T_{b3} = 97.1^oC$) and n-butanol ($T_{b4} = 117.7^oC$). This mixture is fairly ideal with a relatively high relative volatility ($\alpha_{i,j} \geq 1.7$).

The objective was to make the apparatus as simple as possible, and to avoid auxiliary equipment such as reflux pumps. Therefore, the column sections and intermediate vessels are placed on top of each other. The unit was built in glass and carefully insulated to reduce heat loss to the surroundings during operation. The apparatus is operated at

atmospheric pressure.

The unit consists of a reboiler vessel (4 l volume), two intermediate vessels (1 l volume), and a condensate accumulator (1 l volume each). The four vessels are connected by three packed column sections of 420 mm length and 30 mm diameter which are filled with double-wound wire mesh rings of 3 x 3 mm made from stainless steel by Normschliff. The normal heat input to the reboiler is about 350 W, which at steady state results in liquid and vapor flows of about 0.5 mol/min [2].

Each column section is equipped with three chromel-alumel-thermocouples placed in the center of the column cross section. Two thermocouples are placed 5 cm from each end and a third in the middle of the column section. The latter temperature measurement was used for control purposes. The reflux into each of the column sections can be adjusted by means of a two-way solenoid (on-off) valve operated by solid-state relays[3]. The reflux is introduced to the center of the column, slightly above the packing material.

The reflux L_i into each section was used to control the temperature T_i in the middle of the section below (as shown in Figure 1). For simplicity the setpoint of the temperature controllers were set to the arithmetic mean of the boiling points of the two components to be separated in that section, $T_{s,i} = [71.5, 87.75, 107.2]^o C$. The temperature controllers are standard PI-controllers,

$$L_i \ = K_c \ \cdot \left[(T_{s,i} \ - T_i) + \tfrac{1}{\tau_I} \int_0^t (T_{s,i} \ - T_i) dt \right]$$

which were tuned to be rather slow to avoid excessive control action during startup and in the presence of disturbances.

Thermocouples are also placed in the liquid phase of the intermediate vessels and in the reboiler for monitoring purposes. A second thermocouple installed in the reboiler measures the surface temperature of the heating element, and the reboiler duty is adjusted by controlling the temperature difference between reboiler holdup and heating mantle. The process is interfaced to a PC-based control system with a sampling frequency of 1 Hertz. Product composition analysis is performed off-line by means of a gas chromatograph. The intermediate vessels are supplied with heating tapes, but after properly insulating the apparatus these were not used from experiment 4 onwards. Start-up of the experimental system is always from a column at room temperature such that the liquid holdup in the reboiler has to be heated up to its boiling point.

After some initial experiments, the following startup procedure was used from experiment 5 and onwards:

- The three temperature controllers (linking column temperature and reflux to a section) are activated as soon as vapor reaches the top of the column and liquid starts condensing.
- The reflux flow at the top of the column (L_1) has a minimum value of $L_{1min} = 5$ ml/min to ensure a minimum degree of separation and to avoid emptying the reboiler.

[2] Because of variations in molecular weight the volumetric liquid flows varies from about 25 ml/min (vessel 1 from top) to 50 ml/min (vessel 3).

[3] The reflux flow is estimated based on the control signal to the solenoid valve. The relation between opening frequency of the valve and liquid flow has been established by calibration.

- For similar reasons we set at any time $L_{2min} = L_{3min} = L_1$ (volumetric flow), i.e. the reflux flow signal from controller TC_1 is passed as a lower bound to the two other temperature controllers.
- The following PI-settings were used for the temperature controllers (for experiments 5 and on): $K_c = -2.88\ \frac{ml}{minK}$, $\tau_{I,1} = 7\ min$, $\tau_{I,2} = 10\ min$ and $\tau_{I,3} = 7\ min$.

EXPERIMENTAL RESULTS

The experimental results verify that the closed operation with temperature control indeed works in practice. A summary of the experiments are given in Table 1. In the table we give the initial feed composition, as well as the mole fraction of the main component in each vessel and the impurity ratio in the intermediate vessels 2 and 3 at the end of the experiment. The impurity ratio gives an indication of in which direction we have to change the temperature setpoint in the section adjacent to a vessel to achieve a certain product quality.

For our mixture with similar relative volatilities, we conjecture that the degree of separation for a component in the intermediate vessel is maximized (*i.e.* x_i for the main component i is maximized) when the impurity ratio x_{i-1}/x_{i+1} is reasonably close to 1.

All experiments were performed with a total liquid feed of approximately 4 liter. Most of the experiments were performed with the liquid initially charged to the reboiler, except for experiments 4, 10, 11 and 14 where approximately 1.5 liter of the initial feed mixture was distributed evenly to the three intermediate vessels.

In Figures 3 to 5 we present experimental results for three selected experiments; no. 12 (feed initally in reboiler), no. 4 (feed initially distributed), and no. 2 (composition measurements). These experiments are discussed in some detail below.

Experiment 12 (feed initially in reboiler)

In Figure 3 we show as a function of time, the temperatures in the vessels (a) and in the column sections (b), the reboiler heat input (c) and the liquid flows to each column section (d) for experiment 12. Note that the time axis is defined such that $t = 0$ when the first liquid starts flowing ($L_1 \geq 0$).

The startup and operation of the column is explained by referring to Figure 3 and is as follows: The feed charge is filled to the reboiler and heated to its boiling point by an electrical heater. The boiling point of the feed mixture is reached at about $t = -0.15\ h$, indicated by the increase in the column temperatures T_i. When liquid starts collecting in the uppermost vessel ($t = 0$) the three temperature controllers TC_i are activated and reflux L_i is recycled to the column sections. Since the implemented overrides on the reflux flow control require $L_3 \geq L_1$ and $L_2 \geq L_1$ (on a volumetric basis), the reflux flows follow each other for approximately 0.5 h. For $t \geq 1h$, the reflux flow controllers manipulate the flows and the column temperatures T_i (Fig. 3 b) approach their setpoints $T_{S,i}$. The control action of the temperature controllers indirectly adjust the level in vessels M_1 to M_3 (not measured). Operation is continued for a pre-specified time (at least 3 h) until the column approaches a steady state [4].

[4] With a holdup of about 500 ml in each vessel and a reflux flow in excess of 15 ml/min, the vessel composition time constant is less than $\tau_c = 500ml/15ml/min = 33min$.

Experiment 4 (feed initially distributed)

Experiment 4, presented in Figure 4, was performed with the feed charge initially distributed over the column; approximately 60 % of the feed charge was fed to the reboiler, the rest was added to accumulator (M_1) and intermediate vessels (M_2 and M_3). The initial (feed) composition was identical in all vessels. Liquid reflux flows were initially set manually to avoid large amounts of subcooled reflux to enter the column and cause flooding. From Figure 4 we see that vessel temperatures (a) and controlled column temperatures (b) level out at about $t \geq 2\ h$. Experiment 4 was performed with PI-control tunings $K_C \simeq -5.2\ \frac{ml}{minK}$ and $\tau_I = 5\ min$; these somewhat agressive tunings are responsible for the oscillatoric reflux flow from $t \geq 2.7\ h$. The experiment was stopped at $t \approx 5\ h$ and samples from the products were taken and analyzed.

Experiment 2: Product composition trajectory

Experiment 2 is included mainly because of available composition measurements. The feed mixture similar to experiment 12 and with the feed charged to the reboiler. The PI-control tunings are the same as in experiment 4. This is an early experiment and the startup was performed manually; this is the reason for the somewehat erratic initial responses. The compositions of the main component in the vessels and the most important impurities are shown in Figure 6. From composition analysis can be seen that the primary purification is finished after approximately 3.5 h for this experiment.

Comparing the trajectories of the main components in the vessels (Figure 6, top) with the simulation of experiment 12 (Figure 9, top), we see that the trajectories are similar in shape for the purification of the main components in accumulator, intermediate vessels and reboiler. Comparable trajectories for the impurities in the vessel holdup are found for accumulator, intermediate vessel 2 and reboiler.

SIMULATION OF EXPERIMENT 12

In this section we present simulation results for a simple equilibrium stage model with conditions similar to those in experiment 12. The data used for the simulations are given in Table 2. The number of theoretical stage was adjusted to match the observed compositions at the end of the experiment.

At startup all liquid is fed to the reboiler and we assume the column is "hot". The initial reflux flow is set to zero. We use PI-temperature controllers with overrides $L_3 \geq L_1$ and $L_2 \geq L_1$ (volumetric basis) as described in the experimental section.

The feed mixture contains methanol, ethanol, n-propanol and n-butanol with boiling points of the pure components of $T_{b,i} = [64.7, 78.3, 97.2, 117.7]^o C$. For simplicity the column temperature is computed to be the average of the boiling temperatures $T = \sum_{i=1}^{N_c} x_i \cdot T_{b,i}$ (this seemingly crude simplification has little effect on the computed temperatures). As described in the experimental section, the setpoints for each temperature controller was set as the mean boiling temperature of the two components being separated in that column section, $T_{s,i} = [71.5, 87.75, 107.2]^o C$, and we use the same PI-settings $K_c = -2.88 \frac{ml}{minK}$ and $\tau_i = [7, 10, 7]\ min$ as in the experiment. The control stages in the simulations are T_6, T_{17} and T_{26}, which correspond to T1, T2 and T3 in the experiment.

The simulated responses for vessel (a) and column (b) temperatures presented in Figure 7 are in good agreement with the experimental data in Figure 3. One major cause for

differences are the neglected heat loss from the intermediate vessels in the simulation. Furthermore we do not compensate for possible subcooling of the reflux flow. The volumetric reflux flows (see Fig. 7 c) show a somewhat different response with respect to the initial increase in reflux flow L_3 compared to the experiment (see Figure 3 d), however the overall trajectories are similar.

With the temperature setpoints given, we achieve for a feed charge of $M_{init} = 55\ mol$ and a composition of $z_F = [0.26, 0.12, 0.18, 0.44]$ the steady-state liquid holdup and compositions ($t \to \infty$) given in Table 3. The achieved product compositions compare well with the experimental result presented in Table 1; the differences in composition are at maximum $x = 0.026$ mole fraction units. Nevertheless, considerable differences between experiment and simulation are found for the impurity ratio x_{i-1}/x_{i+1}. Those differences can be partly explained because we use an integer number of stages in each section in the simulations.

In Figure 9, we present composition time responses of the main components and impurities in the vessels for the simulation of experiment 12.

In Figure 10 we plot simulated composition profiles over the column for the four components for times $t = [0.5, 1, 2, 3, 6]h$. These profiles show nicely how the individual components accumulate along the the column during operation. The simulated temperature profile over the column is presented in Figure 11. A pronounced gradient in temperature is observed close to the center of each column section, which results in a temperature measurement with good sensitivity for control purposes.

DISCUSSION

Main lessons from the experiments

Following our initial proposal for closed operation with indirect level adjustment based on temperature control (Skogestad *et al.* 1997), concerns were raised that this would not work in practice, for example, due to the possibility of non-uniqueness in the specifications or other unforeseen reasons. The aim of this study was therefore to confirm experimentally the feasibility of the proposed method for operation.

The conclusion is that the experiments almost completely verify what was found in the simulations and we find that it is very easy to operate the column in this way. Except for some initial monitoring during startup to make sure thar the reboiler is not emptied, the column essentially "runs itself".

The only modification made compared to the simulation in Skogestad *et al.* (1997) were to include a minimum liquid flows during the startup period, and to add integral action to the controllers. At first we thought integral actiion was not needed because, as mentioned in Skogestad *et al. (1997)*, the process model from liquid flow L_i to twmperature T_i contains an integrator. However, the disturbances, e.g. in boilup V, are also integrating, so integral action in the controller is needed to adjust the bias term for the liquid flows L_i.

Suggestions for controller tunings

PI-controllers were used to manipulate the liquid flow to keep the column temperature in the middle of the section below at its setpoint. The operation depends somewhat on the controller tunings; a higher controller gain may give a somewhat faster response, but

may result in a noisy response and problems with saturation. As a starting point for the controller gain we suggest the value

$$K_c = -\frac{L}{\Delta T_b}$$

where L is the nominal liquid flowrate and ΔT_b is the difference in boiling points of the components to be separated in the section. With this controller gain a change in composition corresponding to a full boiling point difference is needed to make a liquid flow change of 100 %.

For example, for our experimental column we get at the top of the column (vessel 1) $L/\Delta T_b = -25/13.6\frac{ml}{minK} = -1.84\frac{ml}{minK}$, and at the bottom (vessel 3) $L/\Delta T_b = -50/20.5\frac{ml}{minK} = -2.45\frac{ml}{minK}$. In most experiments we used a somewhat higher controller gain of $K_c = -2.88\frac{ml}{minK}$ (in all vessels). Also, recall from experiment 4 that a gain of $K_c = -5.3\frac{ml}{minK}$ was found to be too high as it gave a somewhat oscillatory response.

The integral time used in the experiments was about 5-10 minutes. This is about 1/15 of the time to evaporate the entire feed mixture (internal circulation time) which was about 2 hours in our experiments.

Justification for column temperature control

In the experiments we keep the temperature in the middle of each column section at a given setpoint value by manipulating the liquid reflux into the section. The setpoint value is essentially the cut-point temperature between the components (fractions) to be separated. This control strategy has proven to be work very well, both in simulations and experiments.

At first this may seem somewhat surprising. For example, if we specify $T_{S2} = 87.75^oC$ (middle of the column), then there is clearly an infinite number of possible mixtures of methanol-ethanol-propanol-butanol with this boiling temperature. However, there is only one binary mixture of ethanol-propanol with this boiling temperature, so provided we are able to establish some initial profile in the column, the relationship between temperature and composition is unique.

Thus, there seems to be at least two reasons why the control strategy based on column temperature control works in practice:

1. **Steady-state uniqueness.** Consider a column with $n-1$ sections (and thus with $n-1$ temperature setpoints) separating a mixture of n given components in a column with a fixed number of theoretical stages. We conjecture that there is then a unique steady-state relationship between the temperature setpoints and the vessel compositions. Furthermore, this relationship is independent of the initial feed composition, except for some azeotropic mixtures where there may be several regions (Hilmen *et al.* 1999). The conjecture has been confirmed by simulations and the experiments presented in this paper. It is also confimed by the thermodynamic analysis of Hilmen *et al.* (1999). Where it is pointed out that the steady state temperature profile will be identical to the *distillation lines* (which are closely related to the *residue curves*).

2. **Unique dynamic response (no inverse response behavior).** We conjecture that the dynamic response from the reflux (manipulated input) to the temperature

in the section below (controlled output) has no inverse response behavior which may cause control difficulties. This is based on the assumption that the temperature decreases as we go up the column. An increase in liquid reflux will then result in decrease in temperature in the column section below.

Alternative control variables

We have established that our proposed column temperature control works well. We argue here that some of the alternative schemes *e.g.* based on vessel compositions or vessel temperature, will not work in practice.

Alternative 1: Composition measurement of main component in vessel

This is of course what we really want to control. However, there are two serious problems if composition is used for feeedback control:

First, the composition specifications may not be achievable because there are too few theoretical stages. In another case, the specifications may be "too easy" compared to the number of stages, and we will have difficulty keeping the composition at its "easy" value (as confirmed by simualtions).

Second, the response of the main component in the vessel will depend on the distribution of the impurities: If the impurities are mostly heavy component, then we want to "move" some of the vessel holdup down to the vessel below, and an increase in the reflux flow out of the vessel will *increase* the purity of main component. However, if the impurities are mostly light component, then we want to "move" some of the vessel holdup up to the vessel above, and an increase in reflux will only make the situation worse and will *decrease* the purity of the main component. Thus, the sign of the gain from reflux to composition depends on the operating point, and such a system is almost impossible to control.

These difficulties have been confirmed in simulations (Wittgens, 1999).

Alternative 2: Temperature measurement in vessel

This variable has the same problem with respect to inverse response behavior as just mentioned for the composition of the main component. In addition, we will have the "usual" problem of sensitivity to measurement error and noise which is always encountered when we use temperature as an indicator of composition for a high-purity product.

In conclusion, use of column temperatures as used in this paper is simple and also seems to be the best measurement to use for controlling the multivessel column.

Optimal operation

There are some degrees of freedom for optimizing the operation. These include initial distribution of holdup setpoint temperatures, and controller tunings.

Simulations and experiments have shown that the exact value of the setpoint temperature is not important as long as the column has sufficient number of stages for the desired separation. Thus using the average between the boiling points is a good choice in most cases. Also, note that with a sufficient number of stages we may achieve any desired purity in the intermediate vessels (see Skogestad *et al.* 1997).

The initial holdup distribution has some effect, and it seems from simulations (*e.g.* Fur-

longe *et al.*, 1999 and Hasebe *et al.*, 1999) that in most cases it is best in terms of minimum batch time to charge the feed to the reboiler. In addition we have found experimentally that it is easier to establish a good initial composition profile with light component in the top if we charge the feed to the reboiler.

Closed operation of conventional batch distillation

Our work is for a multivessel column, but it obviously also applies to the special case of closed operation of a conventional batch column with a distillate vessel (see Figure 12).

The resulting closed mode of operation with a single temperature measurement is very simple and requires minimal operator intervention and monitoring. For example, one can leave the column by itself without having to worry about breakthrough of heavy component. Simulations also indicate that it compares well with conventional batch distillation from an energy point of view. It is therefore very surprising that there is no pervious mention of this mode of operation (Figure 12) in the literature, at least to our knowledge.

CONCLUSIONS

The experiments show very good agreement with the simulations, and confirm that the multivessel column can be easily operated with a simple temperature controllers, where the holdups are only controlled indirectly. For a given set of temperature setpoints, we confirm that the final product compositions are independent of the initial feed composition.

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NOTATION

K_c	Controller gain; $\frac{ml}{min\ ^oC}$
L	Reflux flow; $\frac{kmol}{h}$; $\frac{ml}{min}$
M	Holdup; $kmol$, l
N_c	Number of components
N_i	Number of stages in section i
Q_B	reboiler heat duty; $\frac{kJ}{h}$
t	time; h
T	Temperature; ^oC
T_b	Boiling temperature; ^oC
TC_i	Temperature controller
V	Vapor flow; $\frac{kmol}{h}$
x	Liquid composition
y	Vapor composition
z_F	Feed composition
α	Relative volatility

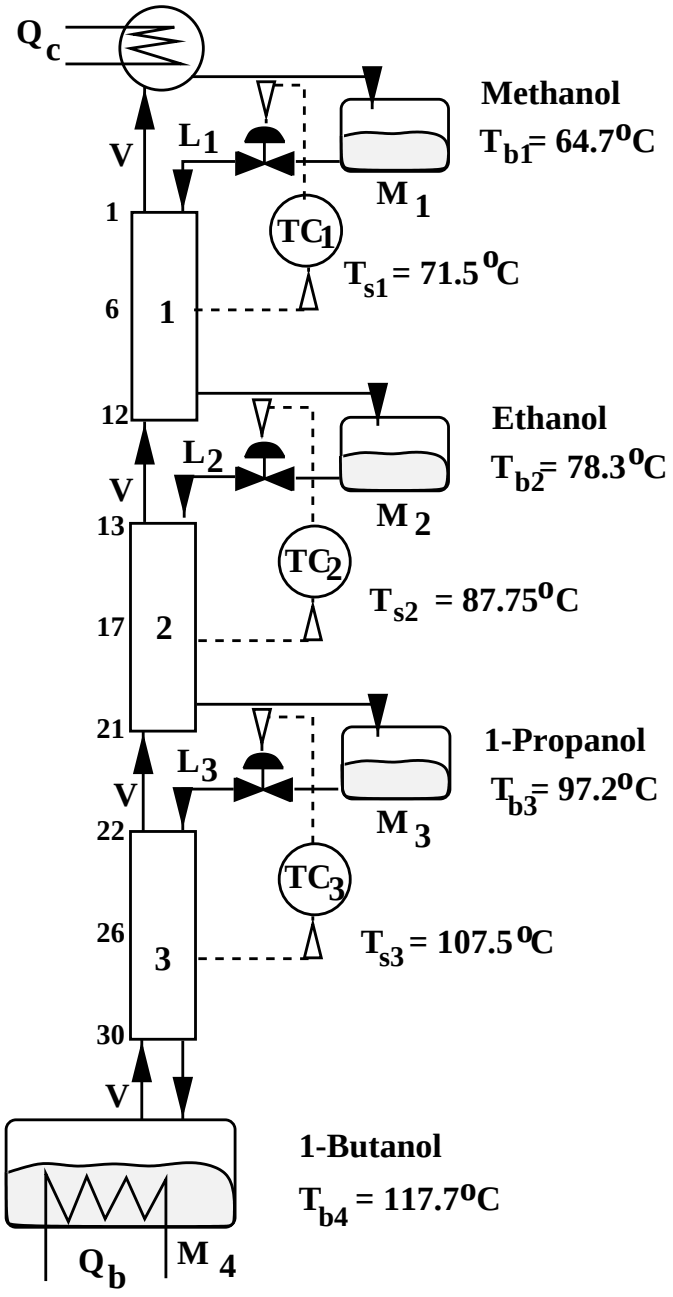

Figure 1: Control scheme for closed operation of multivessel batch distillation column with two intermediate vessels for mixture methanol - ethanol - propanol - butanol.

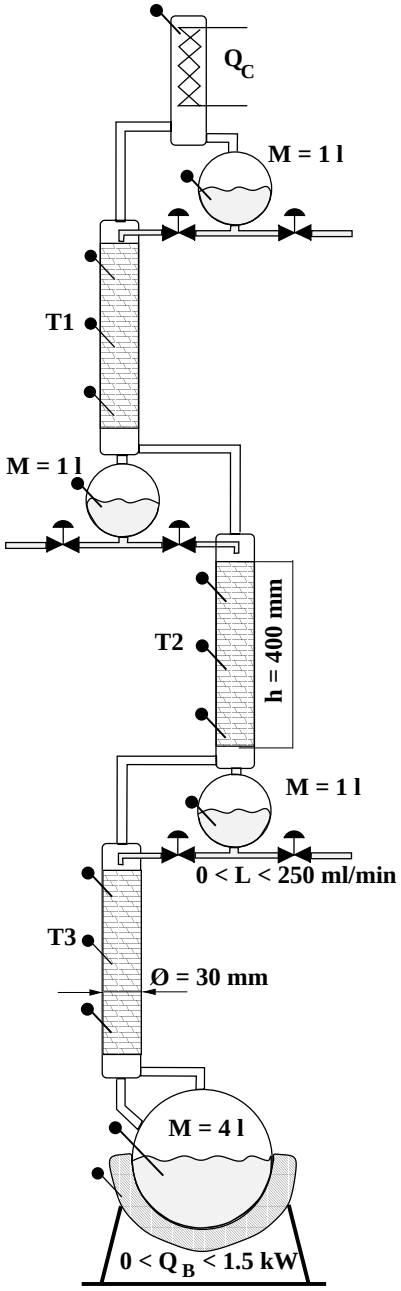

Figure 2: Pilot plant scale distillation column

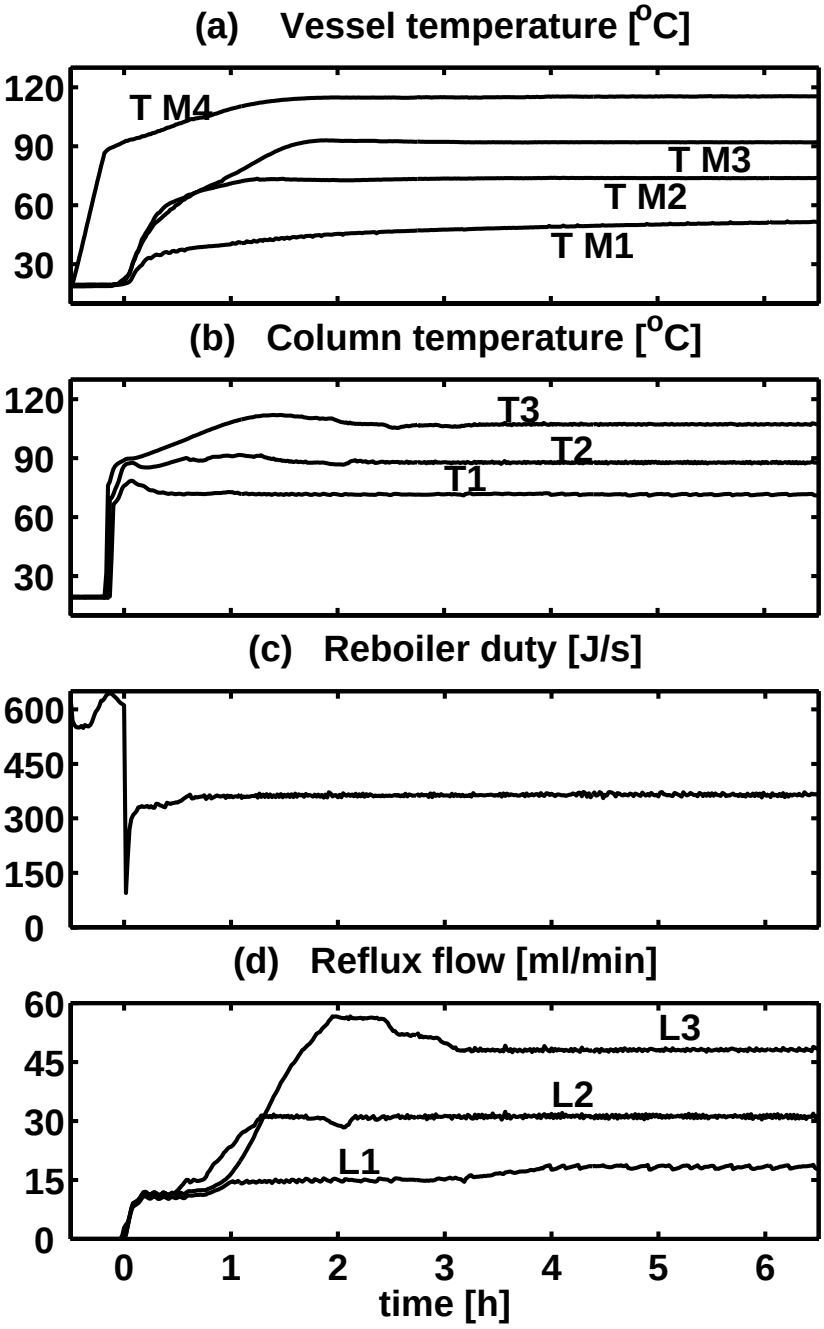

Figure 3: Experiment 12: Temperature responses in vessels (a), in column sections (b), reboiler heat input (c) and volumetric reflux flows as function of time

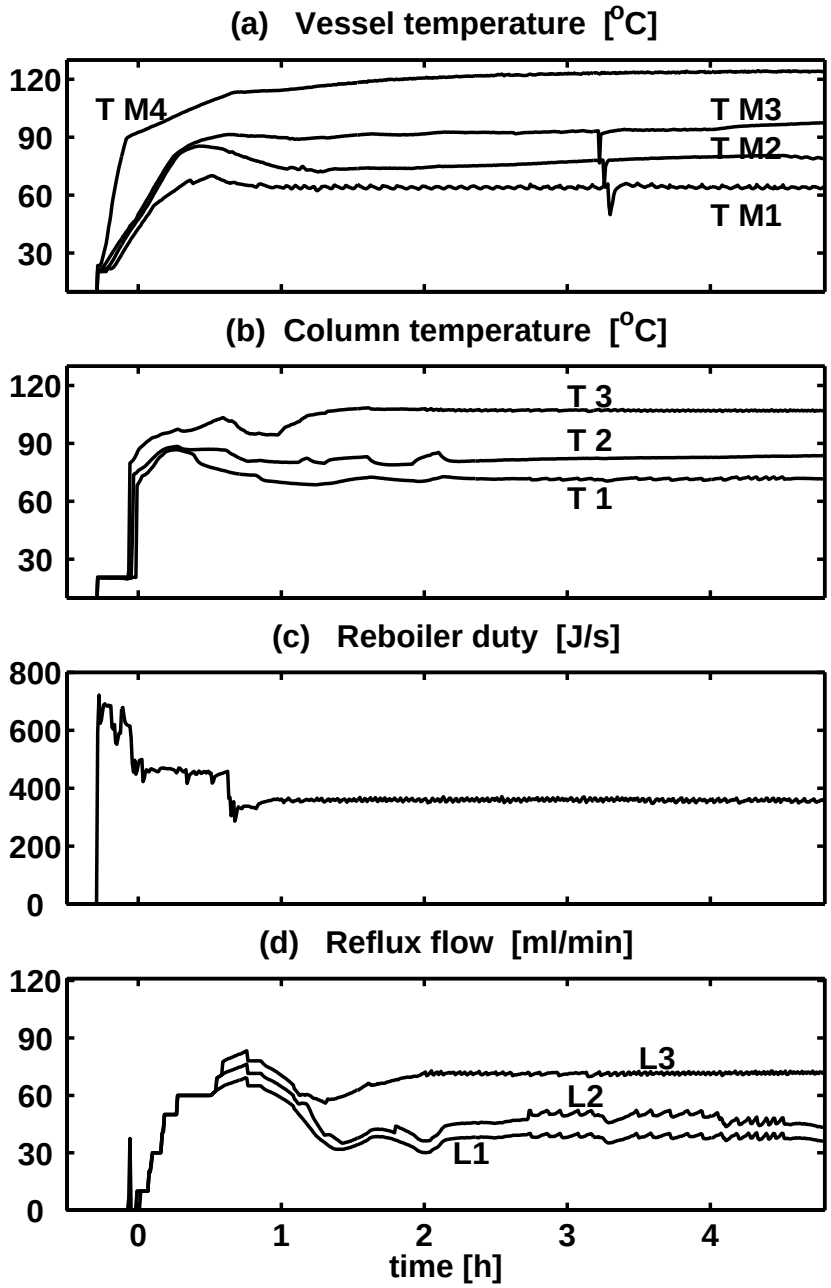

Figure 4: Experiment 4: Temperature responses in vessels (a), in column sections (b), reboiler heat input (c) and volumetric reflux flows as function of time

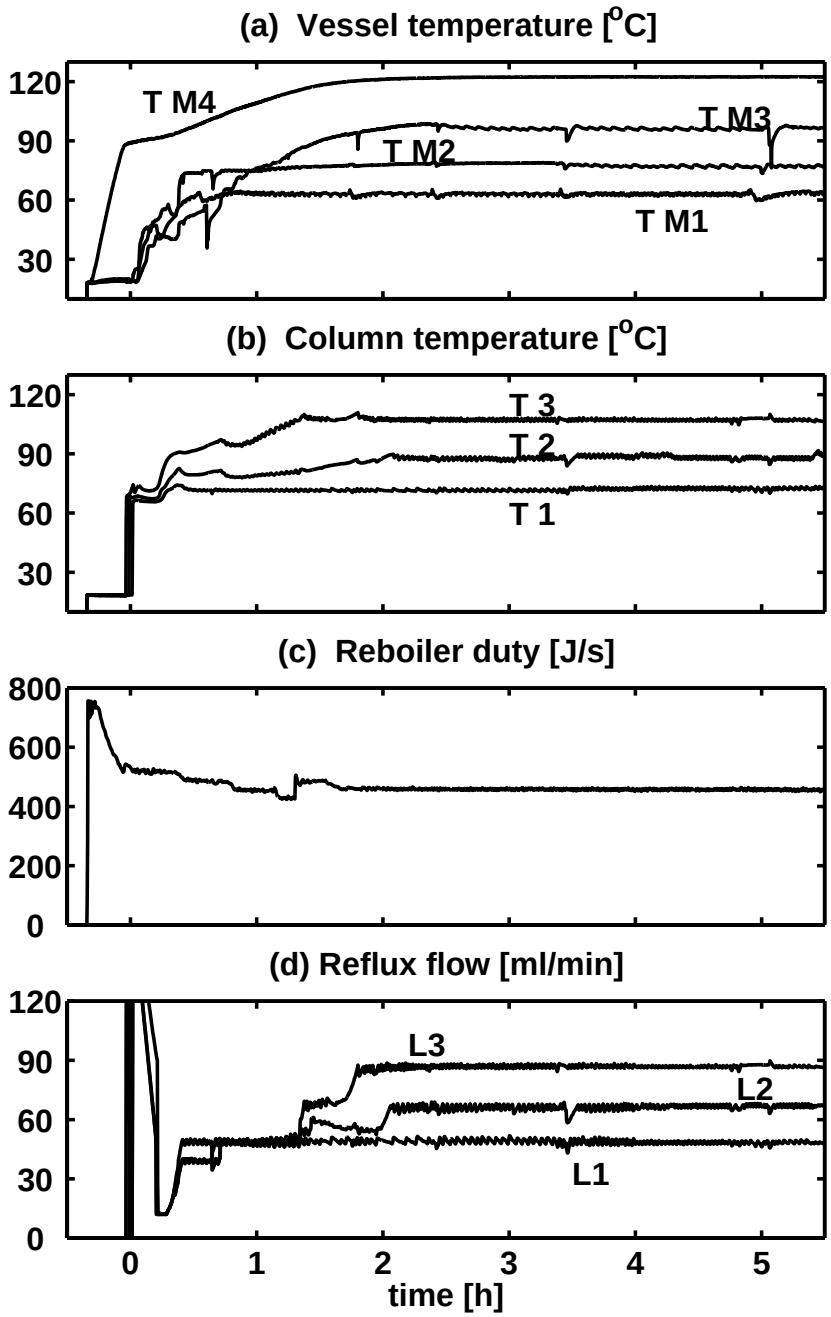

Figure 5: Experiment 2: Temperature responses in vessels (a), in column sections (b), reboiler heat input (c) and volumetric reflux flows as function of time

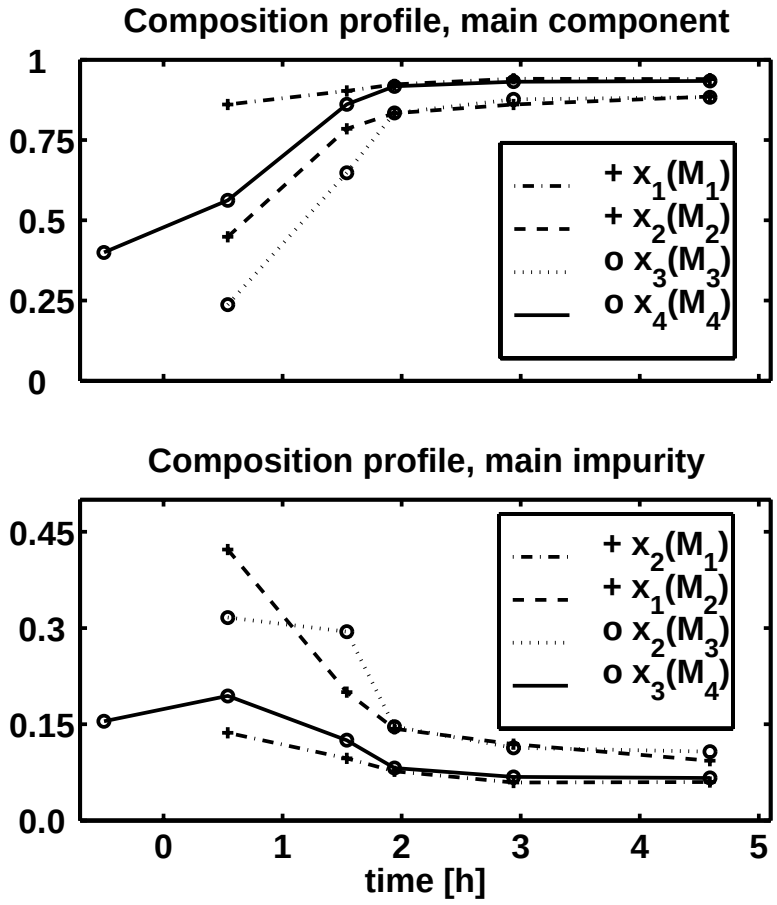

Figure 6: Experiment 2: Compositions of main components (top) and the largest impurity (bottom)

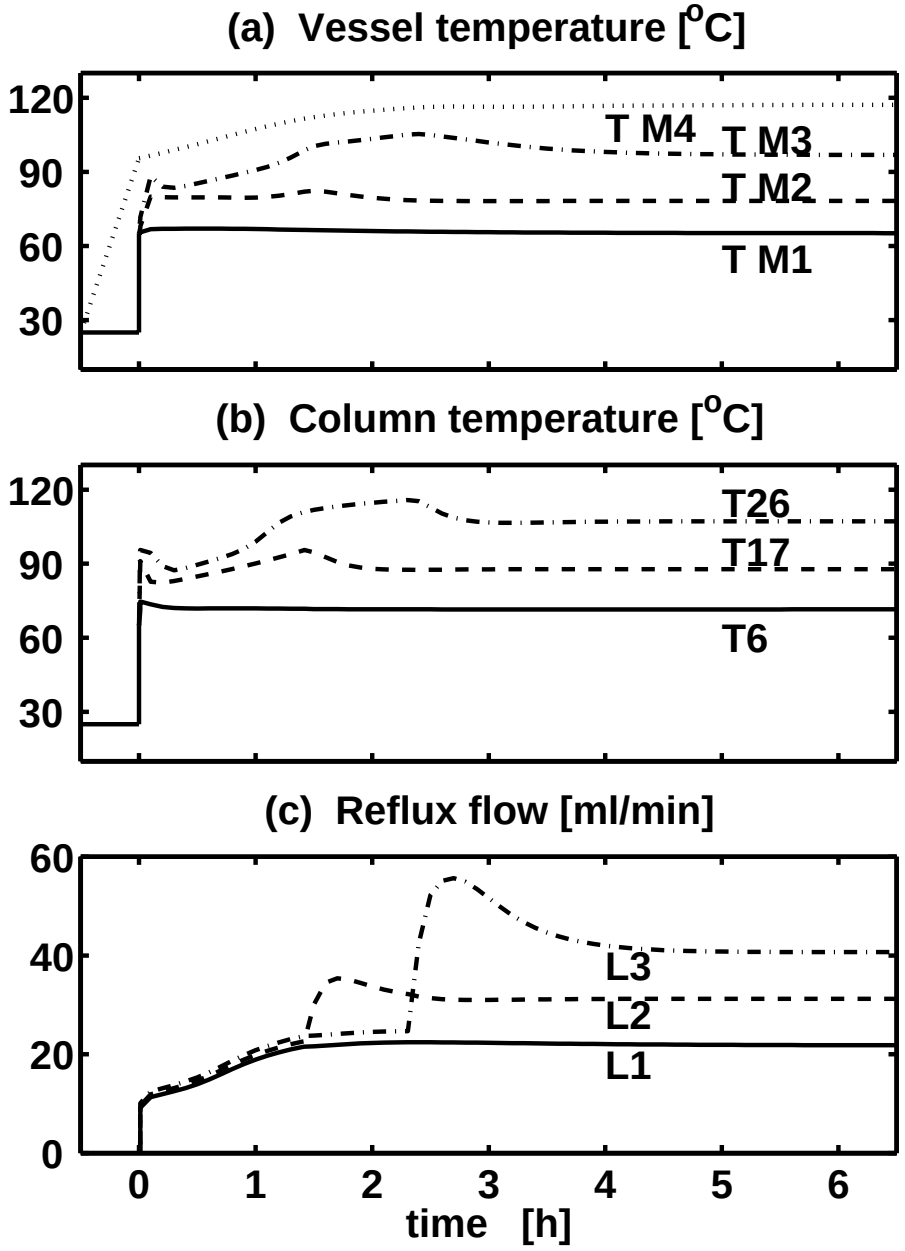

Figure 7: Simulation of experiment 12: Vessel temperature (a) and stages temperature (b) and volumetric reflux flow (c) as a function of time (to be compared with experimental results in Figure 3)

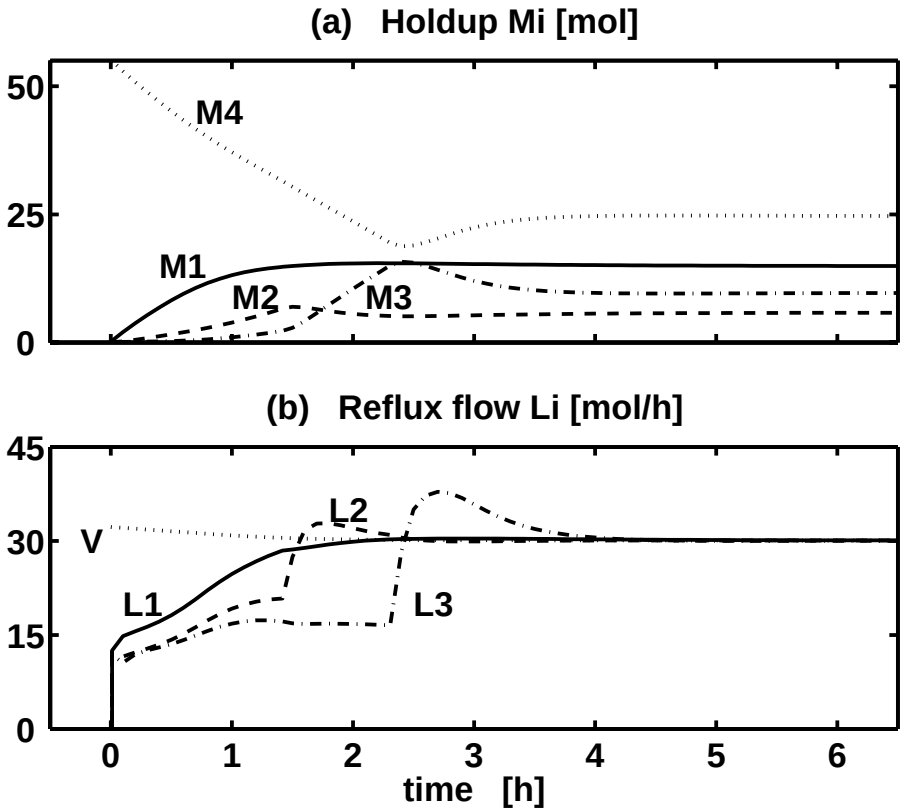

Figure 8: Simulation of experiment 12: molar vessel holdup (a), molar vapor flow and molar reflux flow (b) as a function of time

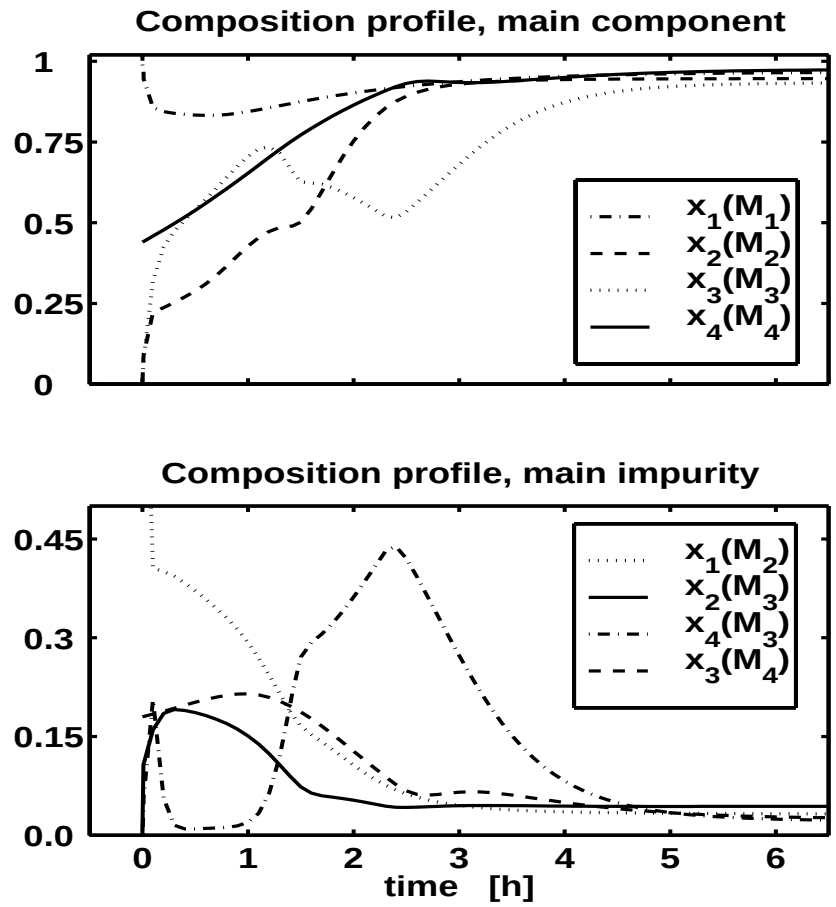

Figure 9: Simulation of experiment 12: vessel compositions (a) and major impurities (b) as a function of time

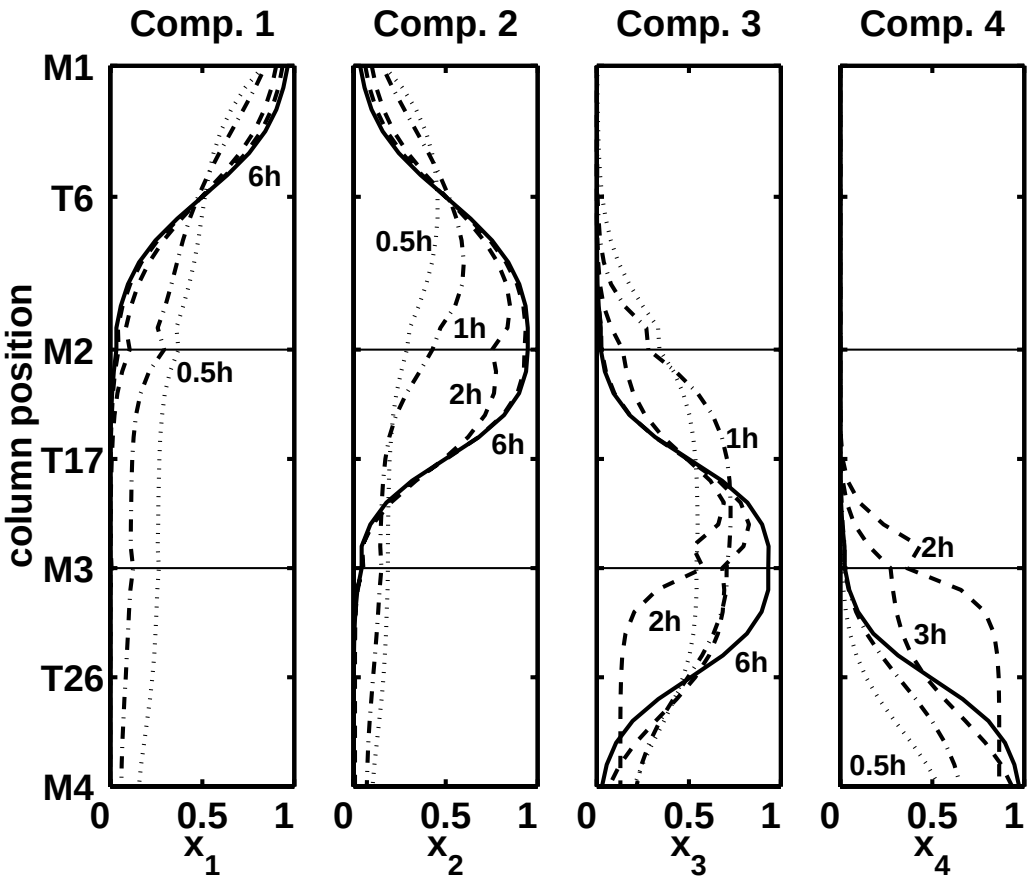

Figure 10: Simulation of experiment 12: Evolution of composition profile over column

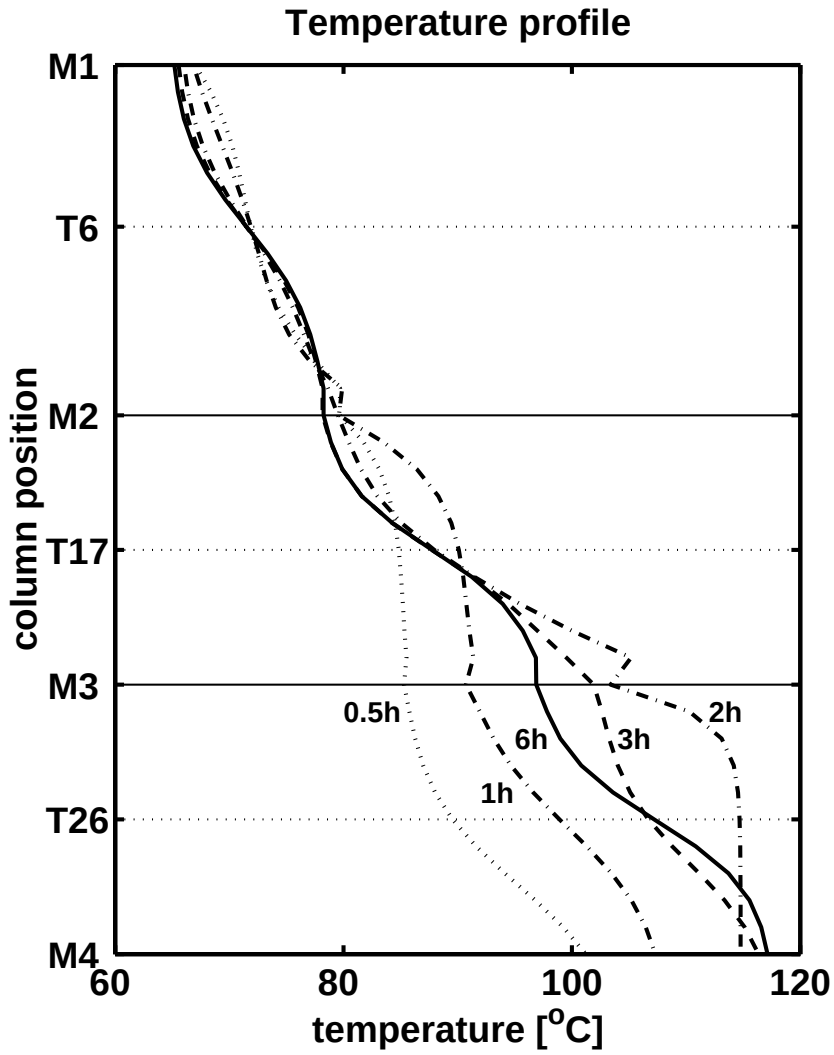

Figure 11: Simulation of experiment 12: Evolution of temperature profile over column

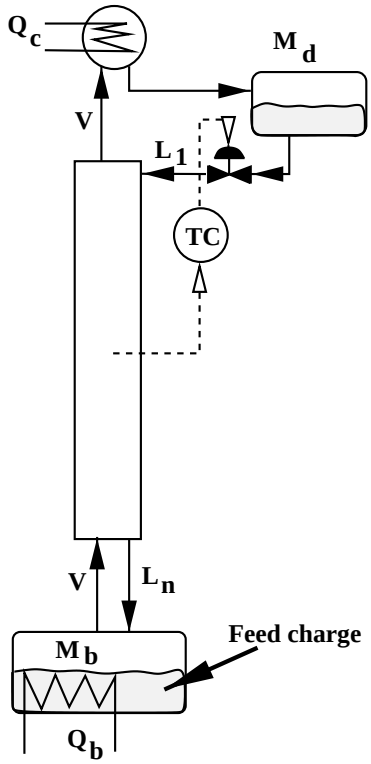

Figure 12: Proposed control scheme for closed operation of conventional batch distillation column (two-vessel column).

Table 1: Summary of experiments

Experiment		reboiler	feed composition	batch	product composition				impurity ratio	
	date	duty $[\frac{J}{s}]$	z_F	time $t_f[h]$	$x_1(M_1)$	$x_2(M_2)$	$x_3(M_3)$	$x_4(M_4)$	$\frac{x_1}{x_3}(M_2)$	$\frac{x_2}{x_4}(M_3)$
0	27.nov.'95	350	[.24, .22, .21, .33]	5.0	0.982	0.960	0.924	0.947	0.57	1.28
1	28.nov.'95	350	[.26, .21, .20, .33]	4.5	0.969	0.547	0.943	0.910	17.45	6.76
2	06.dec.'95	450	[.26, .18, .16, .40]	4.6	0.940	0.886	0.884	0.934	4.39	11.79
3	22.mar.'96	380	[.20, .15, .21, .43]	4.9	0.975	0.915	0.926	0.910	0.890	6.00
4 *	03.apr.'96	390	[.27, .19, .20, .34]	6.8	0.936	0.919	0.907	0.993	13.91	49.50
5	24.sept.'96	375	[.18, .13, .10, .59]	6.9	0.978	0.915	0.962	0.925	7.50	0.52
6	01.oct.'96	385	[.12, .13, .14, .61]	7.1	0.969	0.937	0.950	0.959	3.50	0.67
7	04.oct.'96	370	[.40, .04, .07, .49]	10.8	0.971	0.922	0.945	0.961	5.00	0.35
8	17.oct.'96	380	[.17, .16, .16, .51]	6.0	0.960	0.929	0.941	0.961	4.96	0.67
9	18.oct.'96	375	[.20, .15, .15, .50]	6.2	0.963	0.923	0.941	0.966	3.76	0.74
10*	19.oct.'96	350	[.18, .15, .14, .53]	6.1	0.969	0.914	0.933	0.962	5.71	0.69
11*	20.oct.'96	355	[.18, .15, .14, .53]	6.3	0.970	0.931	0.939	0.958	3.60	0.59
12	07.nov.'96	360	[.26, .12, .18, .44]	8.3	0.971	0.931	0.945	0.949	5.18	0.81
13	18.nov.'96	370	[.18, .16, .16, .52]	6.4	0.963	0.924	0.937	0.957	3.97	1.27
14*	19.nov.'96	360	[.18, .16, .14, .52]	6.5	0.972	0.928	0.933	0.967	3.55	0.73

Note: The liquid was initially charged to the reboiler vessel, except for the experiments marked with $*$ where the feed was initially distributed to all four vessels.
Temperature setpoints in all cases are $T_{s,i} = [71.5, 87.75, 107.2]^o C$

Table 2: Simulation of experiment 12: column data and initial conditions

	Simulation	Experiment
Relative volatility	$\alpha_j = [7.8, 4.5, 2.3, 1]$[1]	
Number of stages per section	$N_i = [12, 9, 9]$[2]	
Initial Vessel holdups ($i = 1, 2, 3$)	$M_{i,0} = 0.01\ mol$	$M_{i,0} = 0\ mol$
Stage holdups	$M_k = 0.01\ mol$	$unknown$
Initial reflux flows	$L_{i,0} = 0\ mol/h$	$L_{i,0} = 0\ mol/h$
Final reflux flow	$L_{i,\infty} = 30\ mol/h$	$L_{1,\infty} \simeq 27 \pm 2\ mol/h$
		$L_{2,\infty} \simeq 28 \pm 2\ mol/h$
		$L_{3,\infty} \simeq 33 \pm 2\ mol/h$
Vapor flow	$V^3_{t=0^+} = 32\ mol/h$	$V_{t=0^+} \simeq 32 \pm 2 mol/h$
	$V_{t\to\infty} \simeq 30\ mol/h$	$V_{t\to\infty} \simeq 30 \pm 2\ mol/h$
Total initial reboiler charge	$M_4 = 55\ mol$	$M_4 \simeq 55\ mol$
Initial reboiler composition	$z_F = [0.26, 0.12, 0.18, 0.44]$	$z_F \simeq [0.26, 0.12, 0.18, 0.44]$ ± 0.01
Final reboiler holdup	$M_4 = 25\ mol$	$M_4 \simeq 24 \pm 2\ mol$

[1] *Approximated data for mixture: methanol - ethanol - n-propanol - n-butanol;*

[2] *Determined from experimental data (rounded to the nearest integer) excluding reboiler (note that the reboiler is a theoretical stage)*

[3] *The steady state vapor flow is computed from* $V \simeq Q_b/\Delta H_{vap,i}$

Table 3: Simulation of experiment 12: Steady-state holdups and compositions

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
M	14.91	5.83	9.69	24.66
x_1	**0.967**	0.032	0.0	0.0
x_2	0.033	**0.947**	0.044	0.0
x_3	0.0	0.021	**0.934**	0.025
x_4	0.0	0.0	0.022	**0.975**
x_{i-1}/x_{i+1}	-	1.52	2.00	-