



## TOTAL REFLUX OPERATION OF MULTIVESSEL BATCH DISTILLATION

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**Abstract** - The multivessel batch column presented in this paper provides a generalization of previously proposed batch distillation schemes. A simple feedback control strategy for total reflux operation of a multivessel column is proposed. The feasibility of this strategy is demonstrated by simulations. An experimental column based on the proposed control scheme has been built and the experiments verify the simulations.

### INTRODUCTION

Although batch distillation generally is less energy efficient than continuous distillation, it has received increased attention in the last few years because of its simplicity of operation, flexibility and lower capital cost. For many years academic research on batch distillation was focused primarily on optimizing the reflux policy. However, in most cases the difference to the simple-minded constant reflux policy usually is small.

More recently, one has started re-examining the operation of batch distillation as a whole. The total reflux operation of a conventional batch distillation column was suggested independently by Bortolini and Guarise (1971) and Treybal (1970). A generalization is a cyclic operation where the operation is switched between total reflux operation and dumping the product (i.e., the condenser holdup is introduced as an additional degree of freedom) may be better (Sørensen and Skogestad, 1994). The simplest operation strategy is with only one cycle, that is, the column is operated under total reflux and the final products are collected in the condenser drum and in the reboiler. Another alternative is to "invert" the column by charging the feed to the top and removing the heavy product in the bottom (Robinson and Gilliland, 1950; Sørensen and Skogestad, 1995). It has also been suggested to use a middle vessel where the feed is charged to the middle of the column (Bortolini and Guarise, 1970). Hasebe et al. (1995) extended this idea and proposed a multivessel column with total reflux operation where one can separate more than two components. They denoted this a "multi-effect batch distillation system".

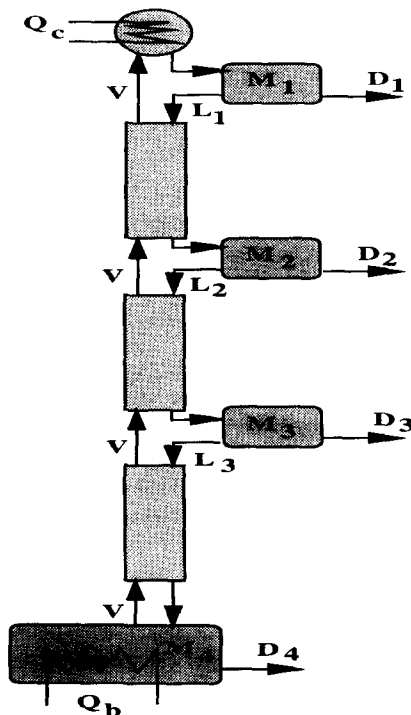


Figure 1: Multivessel batch distillation column

Hasebe et al. (1995) propose to "control" the total reflux multivessel batch distillation column by calculating in advance the final holdup in each vessel and then using a level control system to keep the

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holdup in each vessel constant. For cases where the feed composition is not known exactly they propose to, after a certain time, adjust the holdup in each vessel based on composition measurements. Their scheme, involving the optimization of the vessel holdups and their adjustment based on composition measurement in these vessels, is rather complicated to implement and requires an advanced control structure to implement the control law.

We propose a feedback control structure based on  $N_c - 1$  temperature controllers (see Fig. 3). The idea is to adjust the reflux flow out of each of the upper  $N_c - 1$  vessels by controlling the temperature at some location in the column section below. There is no explicit level control, rather the holdup in each vessel is adjusted indirectly by varying the reflux flow to meet the temperature specifications.

The remainder of this paper is divided into 6 parts. First we present the principle of operation and a simulation example to show the feasibility of the proposed process. The dynamic models are implemented in the SPEEDUP environment (Speedup, 1993). In the second part the proposed implementation of the multivessel column and dynamic simulation results of its operation are given and in part 3 we outline the experimental setup. The procedure to operate the multivessel batch column and experimental results are presented in section 4 and compared to simulations. Finally the discussion and conclusions are given.

## TOTAL REFLUX OPERATION

In this section we follow Hasebe et al. (1995) and present simulations which demonstrate the feasibility of the multivessel batch distillation under total reflux. The holdup of each vessel is calculated in advance by taking into account the amount of feed, feed composition and product specifications. After feeding the prescribed amount of raw material to the vessels, total reflux operation with constant vessel holdup is carried out until the compositions in all vessels satisfy their specifications. Data for the column and feed mixture are given in Table 1.

Table 1: Summary of column data and initial conditions

Number of components	$N_c = 4$	Relative volatility	$\alpha_i = [10.2, 4.5, 2.3, 1]^*$
Total number of stages	$N_{tot} = 33$	Number of sections	$N_s = 3$
Number of stages per section	$N_t = 11$		
Vessel holdup	$M_m = 2.5 \text{ kmol}$	Tray holdup	$M_t = 0.01 \text{ kmol}$
Total initial charge	$M_{tot} = 10.33 \text{ kmol}$	Boilup ratio	$M_{tot}/V = 1.03 \text{ hr}$
Reflux flow	$L = 10 \text{ kmol/hr}$	Vapor flow	$V = 10 \text{ kmol/hr}$

\* The numerical value of ratios of the relative volatilities are chosen to be close to the experimental system (methanol, ethanol, n-propanol, n-butanol) in the pilot plant. Constant molar flows are assumed.

Typical simulated composition profiles as a function of time are shown in Figure 2 for a 4-component mixture with an initial feed composition of

$$z_{F,1} = [0.25, 0.25, 0.25, 0.25] \quad (1)$$

Table 2: Steady state composition for initial feed composition  $z_{F,1}$ ; Constant vessel holdups  $M_i$  [kmol]

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
$M_i$	2.5	2.5	2.5	2.5
$x_1$	0.993	0.017	0.0	0.0
$x_2$	0.007	0.959	0.025	0.0
$x_3$	0.0	0.024	0.963	0.004
$x_4$	0.0	0.0	0.012	0.996

Table 3: Steady state composition for initial feed composition  $z_{F,2}$ ; Constant vessel holdups  $M_i$  [kmol]

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
$M_i$	2.5	2.5	2.5	2.5
$x_1$	0.999	0.203	0.0	0.0
$x_2$	0.001	0.404	0.001	0.0
$x_3$	0.0	0.393	0.999	0.180
$x_4$	0.0	0.0	0.0	0.820

As time goes to infinity the steady state compositions presented in Table 2 are achieved. However, the operation policy of keeping the holdup of the vessels constant may be difficult to achieve in practice and also is very sensitive to errors in the assumed feed composition. The last problem is illustrated by considering a case where the actual feed composition is

$$z_{F,2} = [0.30, 0.10, 0.40, 0.20] \quad (2)$$

but the holdup of each vessel is equal to the example with feed composition  $z_{F,1}$  in Eq. 1. This results in large changes in the final vessel compositions as seen from Table 3. For example, the purity in vessel 2 is reduced from  $x_2 = 0.959$  to  $x_2 = 0.404$ .

To compensate for these feed variations Hasebe et al. (1995) propose a rather complicated algorithm for adjusting the holdup based on measuring the composition in the vessels. We propose a much simpler scheme which is discussed in the next section.

## A NEW FEEDBACK CONTROL STRUCTURE

A flowsheet of our proposed control structure is shown in Figure 3. The separation of a mixture containing

$N_c$  components require  $N_c$  vessels and  $N_c - 1$  temperature controllers. The temperature controller ( e.g.:  $Tc_2$  ) adjust the reflux flow ( e.g.:  $L_2$  ) out of the vessel ( e.g.:  $M_2$  ) above that column section. This enables an indirect control of the holdups in the vessels. Note that there is no level controller or level measurement, although some minimum and maximum level sensors may be needed for safety reasons.

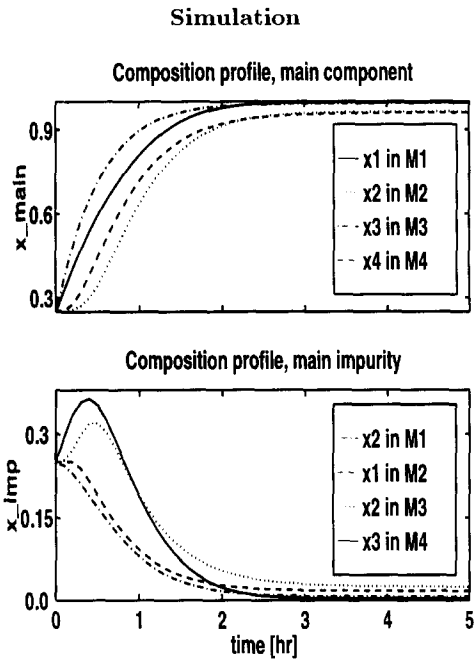


Figure 2: Composition response in accumulator, vessel 2, vessel 3 and reboiler; Feed  $z_{F,1}$ ; Constant vessel holdup

The setpoints for each temperature controller may be in the simplest case be set as the average boiling temperature of the two components being separated in that column section. These setpoints are used in the simulations presented below. Alternatively, they may be obtained by steady-state calculations to get a desired separation, or they may be optimized as a function of time. To demonstrate the feasibility of our proposed control scheme we consider the same example as studied above (see Table 1).

Table 4: Data for temperature controllers in simulations

	$T_{s,i} [^{\circ}C]$	$K_c [ \frac{^{\circ}C}{kmol} ]$	location
$Tc_1$	71.5	-0.25	6
$Tc_2$	87.75	-0.25	17
$Tc_3$	107.2	-0.25	28

Table 5: Steady state compositions obtained by distillation with feedback control

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
$x_1$	0.993	0.016	0.0	0.0
$x_2$	0.007	0.967	0.034	0.0
$x_3$	0.0	0.017	0.960	0.007
$x_4$	0.0	0.0	0.006	0.993

Table 6: Steady state holdup distribution for feed compositions  $z_{F,i}$

feed	Vessel 1	Vessel 2	Vessel 3	Vessel 4
$z_{F,1}$	2.506	2.452	2.512	2.530
$z_{F,2}$	3.053	0.788	4.159	2.000

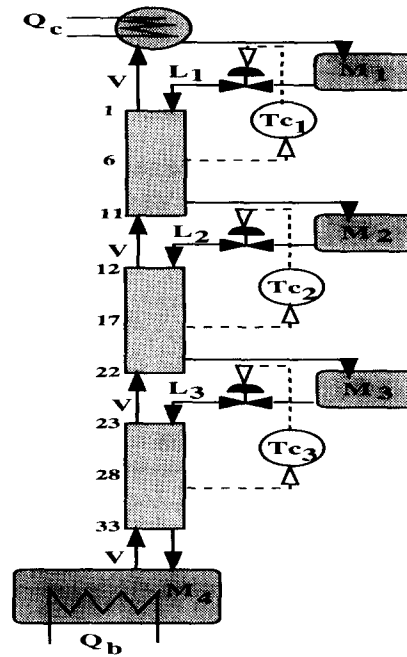


Figure 3: New feedback control structure for multi-vessel batch distillation column under total reflux

The utilized controllers are simple proportional controllers as given in Table 4.

The proportional control algorithm is:

$$L_j = K_c \cdot (T_j - T_{s,j}) \quad (3)$$

For simplicity the column temperature is assumed to be the average of the boiling temperatures

$$T = \sum_{i=1}^{N_c} x_i \cdot T_{b,i} \quad (4)$$

where  $T_{b,i} = [64.7, 78.3, 97.2, 117.7]^{\circ}C$ .

To demonstrate that the proposed control scheme is insensitive to the initial feed composition we use two different initial feed compositions,  $z_{F,1}$  (Eq. 1) and  $z_{F,2}$  (Eq. 2). In both cases the same steady state compositions (see Table 5) are reached as  $t \rightarrow \infty$ . However, the resulting holdups in the vessels are different in each case, as shown in Table 6.

The composition time responses for mixture  $z_{F,1}$  generated with feedback control are not shown. These responses are quite similar to these for the case with constant holdup (no control) shown in Figure 2 (The approach to equilibrium is somewhat faster in vessel 1 and 4 and slower in vessel 2 and 3). On the other hand, with feed composition  $z_{F,2}$  the new policy ensures that the required product qualities are achieved. This is seen from Table 5 and is further demonstrated in Figures 4 and 5, which show the composition profiles in the vessels (Fig. 4) and the holdup in the vessels (Fig. 5, top), flows out of the vessels (Fig. 5, center), and controlled temperatures (Fig. 5, bottom).

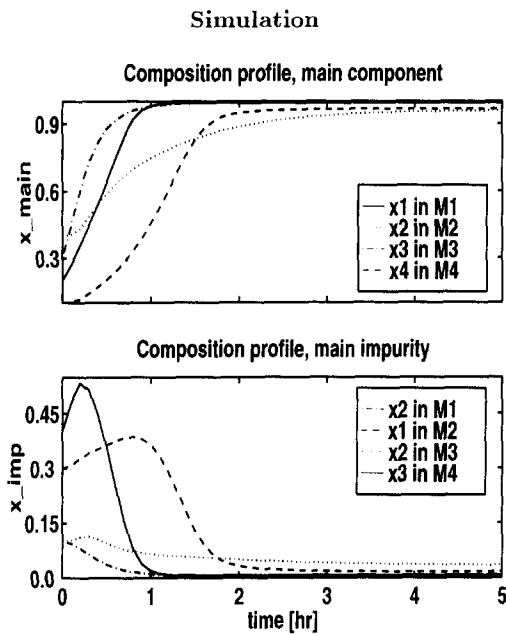


Figure 4: Temperature control: Composition response in vessels 1, 2, 3, and 4 for mixture  $z_{F,2}$

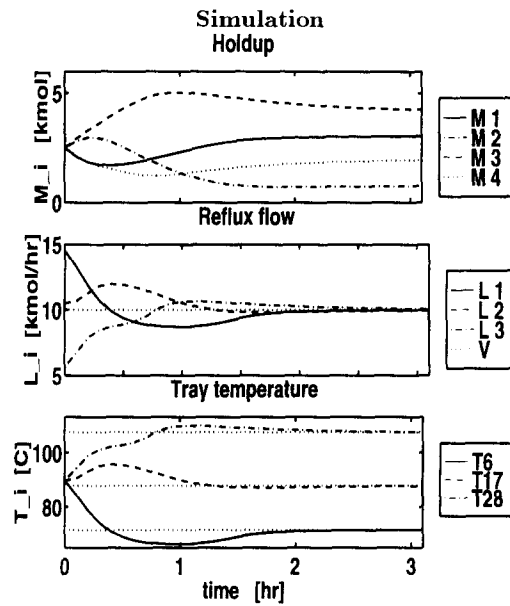


Figure 5: Temperature control: Responses for holdup, reflux flow and tray temperature for mixture  $z_{F,2}$

In Fig. 5 it is observed that the controlled temperatures reach their setpoint  $T \rightarrow T_s$  as  $t \rightarrow \infty$ , even though only proportional controllers are used. The reason why we get no offset is that the model contains an integrator, since the system is closed. More specifically, consider the reflux  $L$  to a column section and the temperature  $T$  in that section. We know that we can change the steady state value of  $T$  by adjusting  $L$ . We also know that a steady state change in  $L$  is not allowed, since we have to have  $L = V$  as  $t \rightarrow \infty$  (total reflux operation). Thus the transfer function from  $L$  to  $T$  must contain an integrator.

With temperature control we achieve the same steady state compositions in the vessels independent of the initial feed composition (only vessel holdups differ at steady state). The reason is that the column has only three degrees of freedom at steady state and if we fix three temperatures at three locations in the column, then the temperature profile over the column at total reflux is determined (if we assume that we do not have multiple steady states). Multiple solutions are not likely when temperatures are specified, but may be encountered if we specify the composition of a given component.

## MULTIVESSEL BATCH DISTILLATION PILOT PLANT

The pilot plant consists of a 4 l reboiler (vessel 4); three packed column sections of 350 mm length and 30 mm diameter, two intermediate vessels (vessel 2 and 3) and accumulator (vessel 1) of 1 l volume each and a total condenser. The column sections are filled with double-wound stainless steel wire mesh rings of 3 x 3 mm. The column sections are stacked on top of each other, so no pumps are needed. A simplified flowsheet of the entire unit is shown in Figure 3. A summary of data for the pilot plant are presented in Table 7. Note that these differ from the simulated multivesel batch column (see Table 1). The number of stages in the columns were estimated by comparing the separation achieved in the experiment with simulations.

Each column section is equipped with a thermocouple placed in the middle of the section. It is used to control the reflux from the vessel above by means of two-way solenoid valves. The setpoint of the temperature controller is set to the average of the boiling points of the components which are expected to accumulate in the two adjacent vessels.

Thermocouples are also placed in the accumulator, the intermediate vessels and in the reboiler. These temperature measurements give an indication of the product qualities. A second thermocouple in the electrical heated reboiler is placed between heating mantle and glass vessel. This arrangement facilitates the

control of the heat input to the reboiler by keeping the temperature difference between heating jacket and reboiler holdup constant.

The unit is build in glass and insulated to reduce heat loss to the surroundings during operation. In addition, intermediate heaters are installed at the intermediate vessels and the accumulator using a temperature controlled heating tape. Inspection openings facilitate the monitoring of liquid distribution over the system. The unit operates at atmospheric pressure.

Table 7: Summary of column data and initial conditions for the experimental system

Total number of stages	$N_{tot} \approx 24$
Number of stages	$N_i \approx 8$
Vessel holdup	$1 \leq M_m \leq 20 \text{ mol}$
Tray holdup	$M_t \approx 0.025 \text{ mol}$
Total initial charge	$M_{tot} \approx 46.8 \text{ mol}$
Heat input reboiler	$Q_b \approx 454 \text{ W}$
Intermediate heater	$Q_m \approx 5 \text{ W}$
Flows	$L \approx V \approx 40 \text{ mol/hr}^*$
Boilup ratio	$M_{tot}/V \approx 1.17 \text{ hr}$

\* Note: The steady state vapor flow is computed from  $V = Q_b/\Delta h_{vap}$  at steady state

## EXPERIMENTAL RESULTS

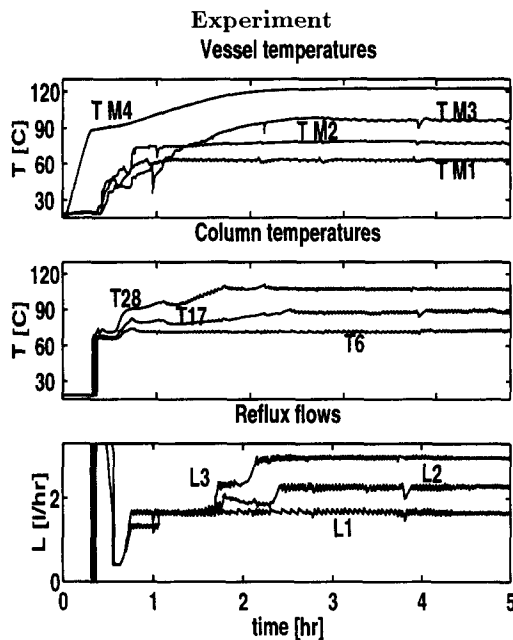


Figure 6: Temperatures in the product vessels, column sections and the reflux flow

The experimental results confirm the results from the simulations. In Figure 6 we present the controlled temperature profiles in the vessels 1 to 4, temperature transients in the column sections and the reflux flow to the column. The start-up and operation of the column is explained by referring to Figure 6 and is as follows: The feed charge is filled to the reboiler and heated to its boiling point ( $t = 0.3 \text{ hr}$ ) by the manually controlled electrical heater. Vapor rising through the column sections is condensed and collected in the accumulator (vessel 1). When liquid starts collecting in the uppermost vessel ( $M_1$ , see Fig. 1), reflux is sent to the column ( $t = 0.35 \text{ hr}$ ). When the temperature  $T_6$  in the upper column section approaches its pre-defined setpoint (at  $t = 0.8 \text{ hr}$ ), the controller  $Tc_1$  for reflux  $L_1$  is activated. The level in vessel 1 floats and is controlled indirectly by the reflux flow controller.

Similarly, after establishing a minimum holdup in vessel 2, reflux is introduced to the second column section. This procedure is repeated for intermediate vessel  $M_3$ , such that liquid flow downwards to the reboiler is established. During start-up (until  $t = 1 \text{ hr}$ ) we set the volumetric flows  $L_2 = L_3 \leq 0.8L_1$  to avoid emptying the reboiler and accumulation of light component in the lower vessels. After establishing

The process is interfaced to a PC-based process control system from INTEC Controls Cooperation (Paragon FS 502, version 2.32). For product composition analysis a Chrompack CP9000 Gas Chromatograph with a Flame Ionization Detector is used. The GC is equipped with an on-column injector and a Chrompack CP-Wax 52 CB fused silica capillary column.

We study a four-component mixture consisting of methanol (1), ethanol (2), n-propanol (3) and butanol (4). This mixture is chosen due to the relative high relative volatility between the components ( $\alpha_{i,j} \geq 2$ ) and their fairly ideal behavior.

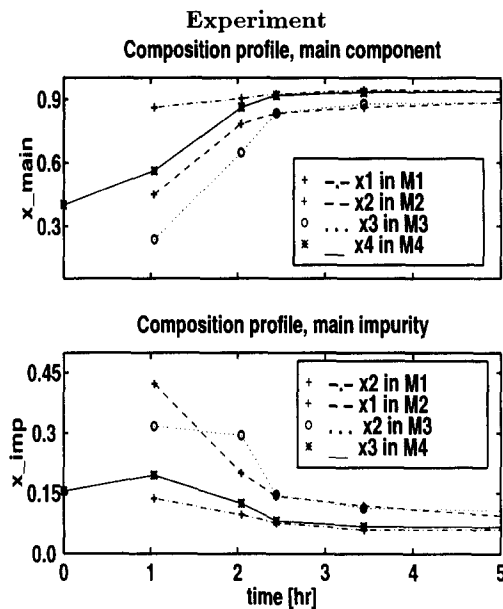


Figure 7: Compositions of main components (top) and the largest impurity (bottom) determined from experiment

stable flows in the column, the controllers for reflux  $L_2$  and  $L_3$  are activated ( $t = 1$  hr) and the setpoints for temperatures  $T_{17}$  in section 2 and  $T_{28}$  in section 3 are approached. Distillation is continued until temperatures are constant for a pre-specified time to achieve the desired product compositions in the vessels.

The compositions of the main component in the vessels and the most important impurity are shown in Figure 7. Comparing the trajectories of the main components in the vessels with the simulation (see Fig. 4) for feed  $z_{F,2}$ , we see that the trajectories are similar in shape, but the purification of the lightest component (1) in vessel 1 and the heaviest component (4) in the reboiler are faster in the experiment, whereas the intermediate components (2 and 3) accumulated in vessel 2 and 3 are purified at a slower rate. This can be explained by the different start-up procedures used for simulation and experiment. The experiment is started from a "cold" column with the entire feed charge in the reboiler at a temperature of  $T = 20$  °C. The simulations are started with a "hot" column with liquid distributed over the columns and vessels. Heating the reboiler charge, introducing vapor to the column and filling the vessels delay the experimental composition trajectories by approximately 0.5 hr. Further delay is caused by the slightly lower boilup ratio of the experiment.

## DISCUSSION

The main reason for using a multivessel column is that it is usually more energy efficient (or equivalently, the batch time for a given heat input is shorter) than a regular batch distillation column. In conventional batch distillation the optimal operation depends quite strongly on the reflux policy and the use of off-cuts to achieve the desired product composition. On the other hand, in multivessel batch distillation there are fewer degrees of freedom and this simplifies the operation considerably. After start-up, the reflux flow is controlled by simple proportional-integral controllers such that the desired products are accumulated in the vessels, after stabilization simple proportional controllers are applied. One disadvantage with the multivessel column compared with the conventional batch distillation is that the column itself is more complicated. Also, whereas in a conventional batch column one only has to make decision on the length of one single column section, one has to decide on the number of sections and their length for a multivessel column. The design of the multivessel column is therefore more closely linked to a specific feed mixture. This problem may be avoided by using an implementation where one column section can be made by interconnecting several shorter ones in series with vessels placed next to each other, similar to that of Hasebe (1995).

Although the work presented here is rather encouraging from a viewpoint of practical implementation many questions remain before this process will be accepted by industry. Further work will include:

1. Study improved methods for practical operation. *e.g.*: controller types and start-up procedures.
2. Study the potential problems with non-uniqueness of temperature as an indicator of composition of a multi-component mixture, especially during start-up.
3. Determine the optimal initial liquid distribution over the column.
4. Study the benefit of constant value or optimizing the temperature setpoints over time.
5. Determine the types of mixtures and conditions which are most suited for the separation in the new process.

## CONCLUSIONS

A new control strategy for the multivessel batch distillation column is proposed. By simulations it has been shown that the proposed control scheme is feasible and easy to implement and operate. Experiments performed on the pilot plant confirm the simulation results.

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