MULTIVESSEL BATCH DISTILLATION [1]

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Submitted for publication in *AIChE Journal*, 21 May 1996

Abstract - The multivessel batch column presented in this paper provides a generalization of previously proposed batch distillation schemes. A simple feedback control strategy for the total reflux operation of a multivessel column is proposed. The feasibility of this strategy is demonstrated by simulations.

1 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 for the conventional batch distillation column (also called the batch rectifier, where the feed is charged to the reboiler and the products are drawn from the top of the column). However, in most cases the difference between an optimal reflux policy and the simple-minded constant reflux policy is small, in practice, other issues are usually more important, such as the recycling of off-spec products, separation of azeotropic mixtures and selection of operation pressure.

More recently, one has started re-examining the operation of batch distillation as a whole. A total reflux strategy, where the final products are collected in the condenser drum and in the reboiler, was suggested independently by Treybal (1970) and Bortolini and Guarise (1971). Sørensen and Skogestad (1994) found the total reflux operation to be better for separations with a small amount of light component.

A generalization of the total reflux strategy is the cyclic operation described by Sørensen and Skogestad (1994). Here, the operation is switched between total reflux operation and

[1] The paper was originally presented at the AIChE Annual Meeting, Nov. 1995, Paper 184i.

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dumping of the product (i.e.: the condenser holdup is introduced as an additional degree of freedom).

Robinson and Gilliland (1950) proposed an inverted batch column, also called the batch stripper, where the feed is charged to the top and the heavy products are drawn from the bottom of the column. Sørensen and Skogestad (1995) found that, also in this case, the inverted column is better than the conventional column for separations with a small amount of light component.

A generalization of the inverted column, is the middle vessel column proposed by Bortolini and Guarise (1971), which has both a rectifying and stripping section. Bortolini and Guarise (1971) proposed to charge a binary feed mixture to the middle vessel and draw products from both the top and the bottom, such that the composition in the middle vessel was approximately constant during the operation. Operation stops when the middle vessel is empty. Hasebe *et.al.* (1992) proposed to charge a ternary mixture to the middle vessel, and let the light and heavy impurities be drawn from the top and the bottom of the column. In this case the operation stops when the intermediate component in the middle vessel has reached its desired purity.

A further generalization is the multivessel column suggested by Hasebe *et.al.* (1995). They proposed a total reflux operation where the products are collected in vessels along the column. Because one may view this column as a stacking of several columns on top of each other, they denote this process a "multi-effect batch distillation system" (MEBAD).

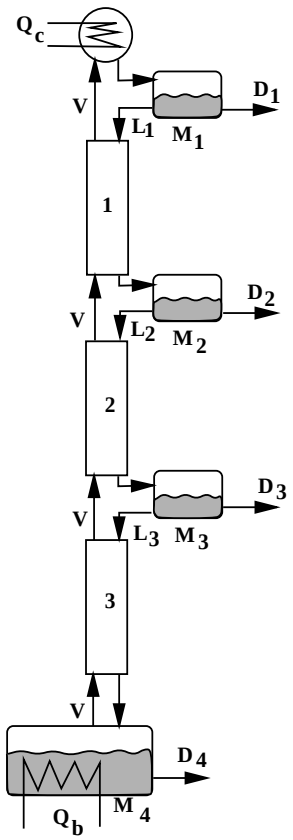

Figure 1: General multivessel batch distillation column for a case with 4 vessels

All the above designs and strategies can be realized in the *multivessel batch distillation column* shown in Figure 1, where both the holdups, $M_i(t)$, and product flows, $D_i(t)$, are degrees of freedom. With N_c vessels along the column and with given pressure and heat input, this column has $2N_c - 1$ degrees of freedom for optimization; namely the $N_c - 1$ holdups $M_i(t)$ (e.g., controlled by the $N_c - 1$ reflux streams) and the N_c product rates $D_i(t)$.

The simplest strategy for operating the multi-vessel column, which is the focus of this paper, is the *total reflux operation* suggested by Hasebe *et.al.* (1995) where the N_c product rates are set to zero ($D_i = 0$). There are at least two advantages with this multivessel column compared to conventional batch distillation where the products are drawn over the top, one at a time. First, the operation is simpler since no product change-overs are required during operation. Second, the energy requirement may be much less due to the multi-effect nature of the operation. In fact, Hasebe *et.al.* (1995) found that for some separations with many components the energy requirement may be similar to that for continuous distillation using $N_c - 1$ columns.

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 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.

The main contribution of our paper is to propose for the total reflux operation of the multi-vessel column, 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, M_i, in each vessel is adjusted indirectly by varying the reflux flow to meet the temperature specifications.

2 Simulation model

All the results in this paper are based on simulations using the dynamic model described in the Appendix. We have made a number of simplifying assumptions, such as constant molar flows, constant relative volatility, linear boiling point curve, constant stage holdup and constant pressure. These assumptions are introduced to simplify the model similar results are obtained when the assumptions are relaxed. The dynamic model is implemented using the SPEEDUP software package (Speedup, 1993).

In the simulations we consider a four-component mixture and a column with three sections and four vessels (including reboiler and condenser). The data for the mixture and the column are summarized in Table 1. The numerical values of the relative volatility are chosen to be close to those of the system methanol-ethanol-propanol-butanol. As mentioned, we assume the mixture temperature , T_k, on stage k to be the molar average of the boiling temperatures of the pure components

$$T_k \;=\; \sum_{j=1}^{N_c} x_j \;\cdot\; T_{b,j} \tag{1}$$

where $T_{b,j} = [64.7, 78.3, 97.2, 117.7]^o C$.

In the simulations we consider two feed mixtures; one equimolar (z_{F1}), and one with smaller amounts of components 2 and 4 (z_{F2}). In all cases the initial (at $t = 0$) vessel holdup is the same ($M_i = 2.5 kmol$) in all four vessels, and the initial composition in all vessels is equal to that of the feed mixture. We start the simulations with a "hot" column, that is, the time needed to heat up the column and the feed mixture is not considered. In all simulations, the vapor holdup is kept constant at $V = 10\ kmol/h$.

Table 1: Summary of column data and initial conditions

Number of components	$N_c = 4$
Relative volatility	$\alpha_j = [10.2, 4.5, 2.3, 1]$
Total number of stages	$N_{tot} = 33$
Number of sections	3
Number of stages per section	$N_i = 11$
Vessel holdup	$M_{i,0} = 2.5\ kmol$
Tray holdup (constant)	$M_k = 0.01\ kmol$
Total initial charge	$M_{tot} = 10.33\ kmol$
Reflux flow	$L_{i,0} = 10\ kmol/h$
Vapor flow (constant)	$V = 10\ kmol/h$

In addition to dynamic simulations, we present the steady-state values which would be achieved if we were to let the batch time approach infinity ($t \to \infty$). Of course, in practice we want the batch time to be as short as possible, and we would terminate the batch when the specifications are met or the improvement in purity is too small. Nevertheless, the steady-state values are interesting because they give the achievable separation for a given case.

3 Total reflux operation with constant vessel holdups

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 predescribed 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.

The simulated composition profiles as a function of time are shown in Figure 2 for the equimolar feed mixture

$$z_{F1} = [0.25, 0.25, 0.25, 0.25] \tag{2}$$

The holdup in each vessel is kept constant at $M_i = 2.5\ kmol$ during the simulation. The purity of the main component in each of the vessels is seen to improve nicely and levels off after about 2 hours. As time goes to infinity the steady-state compositions presented in Table 2 are achieved. The steady state purity of the main component is better than 99% in the top and bottom vessels, and is about 96% in the two intermediate vessels.

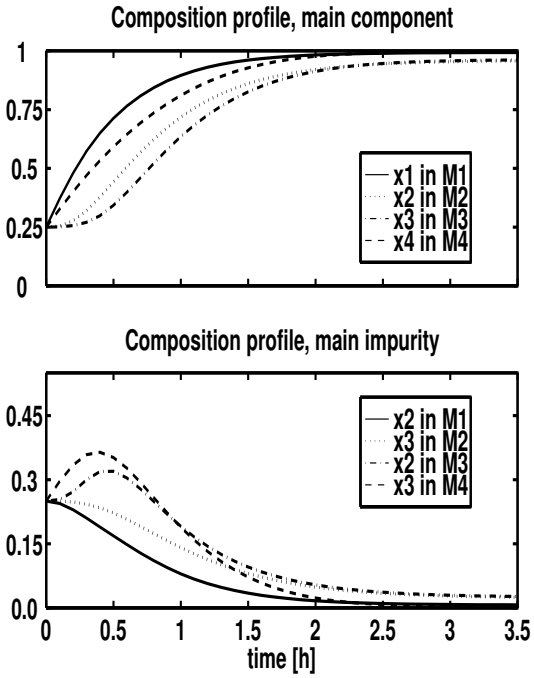

Figure 2: Constant vessel holdup for feed mixture z_{F1}: Composition response in accumulator (1), vessel 2, vessel 3 and reboiler (4)

However, in practice, it may be difficult to keep the vessel holdups constant, and the composition of the feed mixture may be uncertain. The results may be sensitive to holdup errors as is illustrated by considering a case where the actual feed composition is

$$z_{F2} \;=\; [0.30, 0.10, 0.40, 0.20] \tag{3}$$

but the holdup of each vessel is kept constant at $M_i = 2.5\ kmol$, which are the vessel holdups corresponding to the equimolar feed composition, z_{F1}. 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 about 96% to 40%, whereas the purity in vessel 3 is improved from 96% to 99.9%.

Table 2: Steady-state compositions obtained with constant vessel holdups M_i in $[kmol]$ for feed mixture z_{F1}

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
$M_i\ [kmol]$	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 compositions obtained with constant vessel holdups M_i in $[kmol]$ for feed mixture z_{F2}

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
M_i $[kmol]$	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**

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 feedback scheme which is discussed in the next section.

4 Feedback control of multivessel column

A flowsheet of our proposed control structure for the total reflux operation 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 i'th temperature controller (TC_i) controls the temperature (T_i) in the middle of the i'th column section, using as a manipulated input the reflux flow (L_i) out of the vessel above that column section. This enables an indirect control of the holdup (M_i) in that vessel. Note that there is no level controller or level measurement, although some minimum and maximum level sensors may be needed for safety reasons.

The simplest strategy is to let the setpoint for each temperature controller be set as the average boiling temperature of the two components being separated in that column section. This simple strategy is used in the simulations. Alternatively, to reduce the batch time for a specific separation, the setpoints may be obtained from steady-state calculations corresponding to the desired separation, or they may even be optimized as functions of time. However, it is believed that in most cases, except when the number of stages in the column is close to the minimum for the desired separation, the simple strategy will be acceptable.

To demonstrate the feasibility of our proposed control scheme we consider the same column as studied in the previous section. To prove that the scheme is insensitive to the initial feed composition we consider two different initial feed compositions, z_{F1} (Eq. 2) and z_{F2} (Eq. 3).

We use simple proportional temperature controllers to manipulated the reflux flow

$$L_i \ = K_c \ \cdot \ (T_i \ - T_{s,i}) + L_{i,0} \tag{4}$$

with the numerical values given in Table 4. The controller gains were selected such that an offset in the temperature of $\Delta T_i \ = \ 10^o C$ yields a change in the corresponding reflux

flow of L_i of 2.5 *kmol* (25% of the nominal flowrate). The setpoint, $T_{s,i}$, for each section is the average boiling temperature of the components being separated in that section. The temperature sensors are located in the middle of each column section.

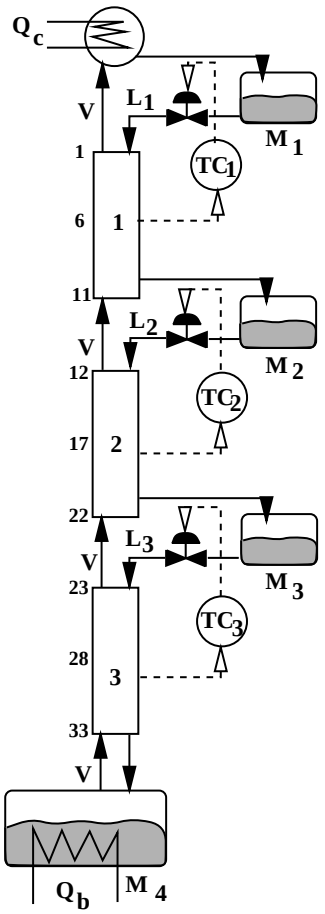

Figure 3: Feedback control structure for multivessel batch distillation column under total reflux

Table 4: Data for temperature controllers

	$T_{s,i}$ $[^oC]$	K_c $[^oC/kmol]$	location *
TC_1	71.5	-0.25	6
TC_2	87.75	-0.25	17
TC_3	107.2	-0.25	28

** stage no. from top of column*

With these temperature controllers, we achieve for *both* feed mixtures the same steady-state compositions ($t \to \infty$) given in Table 5. These steady state compositions are very close to

those found earlier for feed mixture $z_{F,1}$ with constant vessel holdups of $M_i = 2.5\ kmol$; compare Table 2 with 5.

As expected, for feed mixture z_{F1}, the steady-state vessel holdups are close to 2.5 $kmol$; see the first row in Table 6. The composition time responses for feed mixture z_{F1} is shown in Figure 4. The responses are similar to those with constant vessel holdups shown in Figure 2; the difference is that the approach to steady state is faster in vessels 1 and 4 and slower in vessels 2 and 3 for the control structure employing temperature control.

Table 5: Steady-state compositions obtained with temperature control (independent of feed composition)

	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 obtained with temperature control for feed compositions z_{F1} and z_{F2}

	Vessel 1	Vessel 2	Vessel 3	Vessel 4
feed	$M_1\ [kmol]$	$M_2\ [kmol]$	$M_3\ [kmol]$	$M_4\ [kmol]$
z_{F1}	2.506	2.452	2.512	2.530
z_{F2}	3.053	0.788	4.159	2.000

The composition time responses for feed mixture z_{F2} shown in Figure 5 is also similar, although the initial compositions are quite different (see Eq. 2 and Eq. 3). To achieve pure products for feed mixture z_{F2}, the action of the temperature controllers results in very large changes in the vessel holdups; see the second row in Table 6 where the steady state holdups vary from 0.788 kmol in vessel 2 to 4.159 kmol in vessel 3.

In Figures 4 and 5 we also present for feed composition z_{F1} and z_{F2} the time responses for the holdups in the vessels (top), the reflux flows out of the vessels (center), and the controlled temperatures (bottom). The simulations demonstrate how the action of the temperature controllers adjust the reflux flows, which indirectly adjust the vessel holdups such that the final products are of high purity.

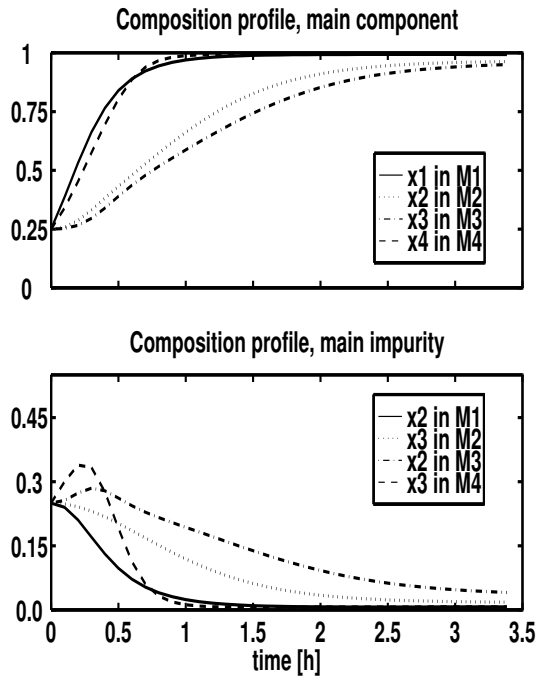

Figure 4: Temperature control for feed mixture z_{F1}: Vessel compositions as a function of time

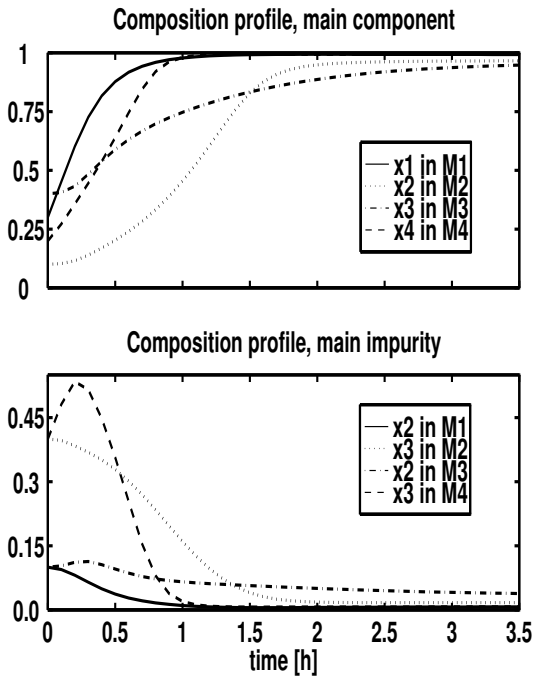

Figure 5: Temperature control for feed mixture z_{F2}: Vessel compositions as a function of time

Three remarks about the results are in order.

1. From Figure 7 we observe that the controlled temperatures reach their setpoint with no offset ($T \rightarrow T_s$ as $t \rightarrow \infty$), even though only proportional controllers are used. The reason is that the model from L_i to T_i contains an integrator, since the system is closed. More specifically, consider the reflux L_i to a column section and the temperature T_i in the that section. We know that we can change the steady-state value of T_i by changing L_i. We also know that a steady-state change in L_i is not allowed, since we must have $L_i \rightarrow V_i$ as $t \rightarrow \infty$ (total reflux operation). Thus the transfer function from L_i to T_i must contain an integrator.

2. With temperature control we achieve the same steady-state compositions in the vessels independent of the initial feed composition (only the 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. This assumes 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.

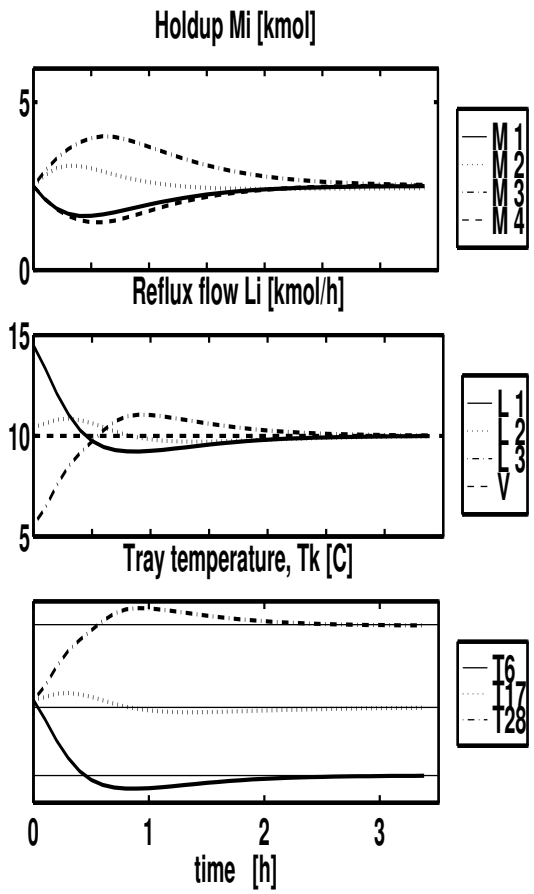

Figure 6: Temperature control for feed mixture z_{F1}: Holdup, reflux flow and tray temperature as a function of time

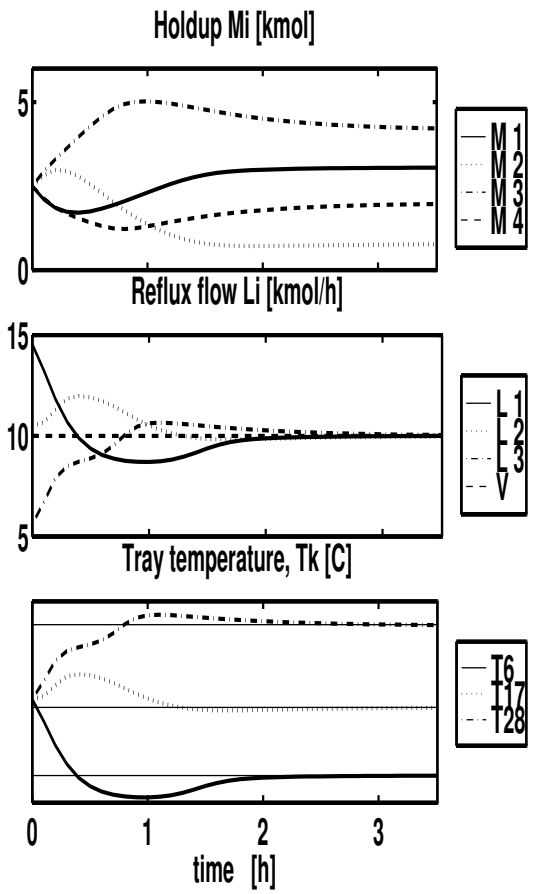

Figure 7: Temperature control for feed mixture z_{F2}: Holdup, reflux flow and tray temperature as a function of time

3. We have also performed some simulations to study the start-up for the case when the entire feed mixture is charged to the reboiler (and not distributed to the vessels). The results indicate that the temperature controllers can be activated immediately after start-up; possibly with some strategy to ensure that the vessels are not emptied. The vessels are then slowly filled up by action of the temperature controllers which reduce the reflux flows for a transient period. The simulations indicate that the required time to reach a desired separation is similar to that found when the feed is initially distributed to the vessels.

5 Achievable separation

The achievable separation is limited by the number of theoretical stages in the column sections. Or, stated in another way, if there are no thermodynamic limitations caused by azeotropes etc., then we can achieve any desired purity in a multivessel column if we have a

sufficient number of stages. This is demonstrated in Table 7 where we present the steady-state product compositions for different numbers of theoretical stages N_i in the three column sections. The total number of stages is $3 \cdot N_i$. We use the same components as before (the feed composition does not matter), and use temperature controllers with the setpoints given in Table 4. With 7 stages in each section we achieve a purity of about 86% in vessels 2 and 3, with 11 stages (as used in the rest of the paper) about 96%, with 15 stages about 99%, and with 25 stages about 99.97%.

Table 7: Temperature control: Steady-state vessel compositions (main component) as a function of number of stages in each section.

N_i	Vessel 1	Vessel 2	Vessel 3	Vessel 4
	x_1	x_2	x_3	x_4
7	0.965	0.864	0.856	0.965
9	0.984	0.932	0.923	0.984
11	0.993	0.967	0.960	0.993
15	0.998	0.992	0.990	0.999
19	0.9997	0.9982	0.9974	0.9997
25	0.9999	0.9998	0.9997	0.9999

6 Discussion

One justification for using multivessel distillation instead of conventional batch distillation is to save energy, or equivalently, for a given heat input the batch time may be significantly shorter. Another advantage is the simple operation of the multivessel column under total reflux. A third advantage is that it may be easier to operate the column close to optimum with the multivessel column. In conventional batch distillation the optimal operation may depend quite strongly on the reflux policy and the use of off-cuts to achieve the desired product composition, and it is difficult to obtain and implement this optimal strategy. On the other hand, in the multivessel batch column there are fewer degrees of freedom and this simplifies the operation considerably; the reflux flow is adjusted with simple temperature controllers such that the desired products are accumulated in the vessels.

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 columns is therefore more closely linked to a specific feed mixture, in particular the relative volatility and the product specifications. Thus, the design process of a multivessel column is similar to the design of a sequence of continuous distillation columns.

For a practical implementation, the simplest configuration is to place the sections and stages on top of each other as indicated in Figure 1. The liquid then flows by the influence of gravity and there is no need for pumps. However, this design is rather inflexible, and it cannot be used if a large number of stages is required. For an industrial multi-purpose separation facility, it is probably better to place the column sections in series with the vessels at ground level as indicated by Hasebe *et.al.* (1995). Reflux pumps are then needed to bring the liquid from the vessels to the column sections. In this case, one can quite easily put several column sections in series to meet the separations requirements for a given feed mixture.

Although the results presented in this paper on the temperature controlled multi-vessel column are most encouraging, a number of questions are open for further research.

1. The simulations need to be verified experimentally. This work is in progress, and the preliminary results show very good agreement with the simulations. These results are presented separately (Wittgens *et.al.*, 1996).

2. The control strategy needs to be studied in more detail, e.g., which controller algorithm to use and the start-up procedures.

3. The effect of optimizing the temperature setpoints as a function of time must be studied.

4. It must be established for what type of mixtures and conditions the new process is most suited.

5. Determine the optimal initial liquid distribution over the column.

6. The multivessel batch distillation column may be suitable for separating azeotropic mixtures by adding an extra component ("entrainer"). The entrainer may be recovered in one of the vessels.

7. Finally, the total reflux operation may be generalized by also allowing withdrawal of products (continuous or discontinuous) from the vessels. In this way the multivessel column forms a "super structure" which has as special cases all the previously proposed batch schemes mentioned in the introduction.

7 Conclusions

A general multivessel batch distillation column is proposed, along with a new control strategy for its total reflux operation. It is shown that the proposed control scheme is easy to implement and operate, even for widely varying feed compositions.

Notation

D	Distillate flow rate $[kmol/hr]$	t	time
K	Controller gain	V	Molar vapor flow
L	Reflux flow rate $[kmol/hr]$	x	Liquid composition
M	Molar holdup $[kmol]$	y	Vapor composition
N_c	Number of components	z	Feed composition
N_i	Number of stages in section i	α	Relative volatility
T	Temperature		

Subscripts

i	section identifier
j	component identifier
k	stage identifier

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Appendix

Mathematical Model of Multibatch Distillation

The model used in the simulations is based on the following assumptions:

- constant relative volatility
- constant molar liquid holdups on the stages (liquid flow dynamics neglect)
- constant molar vapor flows V_i (energy balance neglected)
- constant pressure
- constant tray efficiency (100 %)
- negligible vapor holdup
- perfect mixing on all trays and in all vessels
- total condenser

The distillation column is modeled as a stack of stages (counted from the top). Note that the vapor flow V does not pass through the intermediate vessels so these do not contribute to the number of theoretical stages. The model for stage k in section i consists of a material balance for each component j (M_k is assumed constant)

$$M_k \ \frac{d \ x_{j,k}}{dt} = L_i \ (x_{j,k-1} - x_{j,k}) + V \ (y_{j,k+1} - y_{j,k}) \tag{5}$$

and the vapor liquid equilibrium

$$\alpha_j = \frac{y_{j,k}/x_{j,k}}{y_{H,k}/x_{H,k}} \tag{6}$$

where H denotes the heaviest component in the mixture.

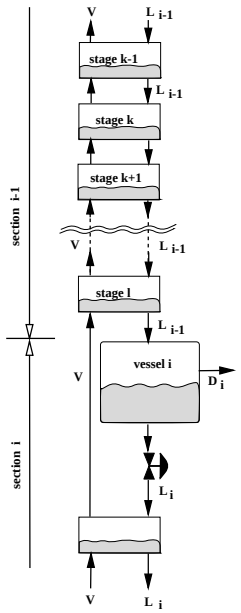

Figure 8: Connection of trays and vessels

The material balance for the condenser ($i = 1$) is

$$\frac{d\ (M_i\ x_{j,i})}{dt} = V\ y_{j+1,i} - L_i\ x_{j,i} \tag{7}$$

and its mass balance

$$\frac{d\ M_i}{dt} = V\ -\ L_i \tag{8}$$

For intermediate vessels (i)

$$\frac{d\ (M_i\ x_{j,i})}{dt} = L_{i-1}\ x_{j-1,i} - L_i\ x_{j,i} \tag{9}$$

with

$$\frac{d\ M_i}{dt} = L_{i-1}\ -\ L_i \tag{10}$$

where x_i is the composition in vessel i and $x_{j-1,i}$ is the liquid composition at the bottom of the section above. The liquid flow L_i leaving vessel i is set by a control valve.

The reboiler ($i = R$)

$$\frac{d\ (M_i\ x_{j,i})}{dt} = L_{i-1}\ x_{j,i} - V\ y_{j,i} \tag{11}$$

where

$$\frac{d\ M_i}{dt} = L_{i-1}\ -\ V \tag{12}$$

where again the vapor liquid equilibrium is described by Equation 6.