Optimal Operation of Multivessel Batch Distillation Columns

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Increased interest in unconventional batch distillation column configurations offers new opportunities for increasing the flexibility and energy efficiency of batch distillation. One configuration of particular interest is the multivessel column, which can be viewed as a generalization of all previously studied batch column configurations. A detailed dynamic model was used for comparing various optimal operating policies for a batch distillation column with two intermediate vessels. A wide variety of degrees of freedom including reflux ratios, product withdrawal rates, heat input to the reboiler, and initial feed distribution were considered. A mixture consisting of methanol, ethanol, n-propanol and n-butanol was studied using an objective function relating to the economics of the column operation. Optimizing the initial distribution of the feed among the vessels improved column performance significantly. For some separations, withdrawing product from the vessels into accumulators was better than total reflux operation in terms of energy consumption. Open-loop optimal operation was also compared to a recently proposed feedback control strategy where the controller parameters are optimized. The energy consumption of a regular column was about twice that of a multivessel column having the same number of stages.

Introduction

Batch distillation is widely used in the fine and speciality chemical industries for the separation of liquid mixtures. In a regular batch distillation column (shown in Figure 1a), the feed is charged to a pot at the base of the column and the products are withdrawn one at a time from the top in order of decreasing volatility. Alternatives to this column configuration include the *inverted column* and the *middle vessel column*, both of which were initially proposed by Robinson and Gilliland (1950). In the case of an inverted column, the feed is charged to the reflux drum at the top of the column and the products are withdrawn from the reboiler; this column is therefore also referred to as a batch stripping column. This configuration was later studied by Chiotti and Iribarren (1991), Hasebe et al. (1992), Mujtaba and Macchietto (1992), Sørensen and Skogestad (1996), Sørensen (1997), and Lotter and Diwekar (1997). In a middle vessel column (also called a "complex" column), the feed is charged to a vessel located between two column sections. The column thus has a rectifying and a stripping section similar to a continuous distillation column. Studies on this configuration have been reported by Hasebe et al. (1992), Mujtaba and Macchietto (1992, 1994), Davidyan et al. (1994), Meski and Morari (1995), Safrit et al. (1995), Hasebe et al. (1996), Barolo et al. (1996), Lotter and Diwekar (1997), Safrit and Westerberg (1997a,b), and Hilmen et al. (1997).

Hasebe et al. (1995) extended the concepts of the inverted and middle vessel columns to what they called the multi-effect column which is also referred to as the *multivessel column* (see Figure 1b). The latter comprises two or more column sections; liquid feed and products can be charged to, and be withdrawn from, vessels located between these sections. Hasebe et al. (1995) found that, for some separations,

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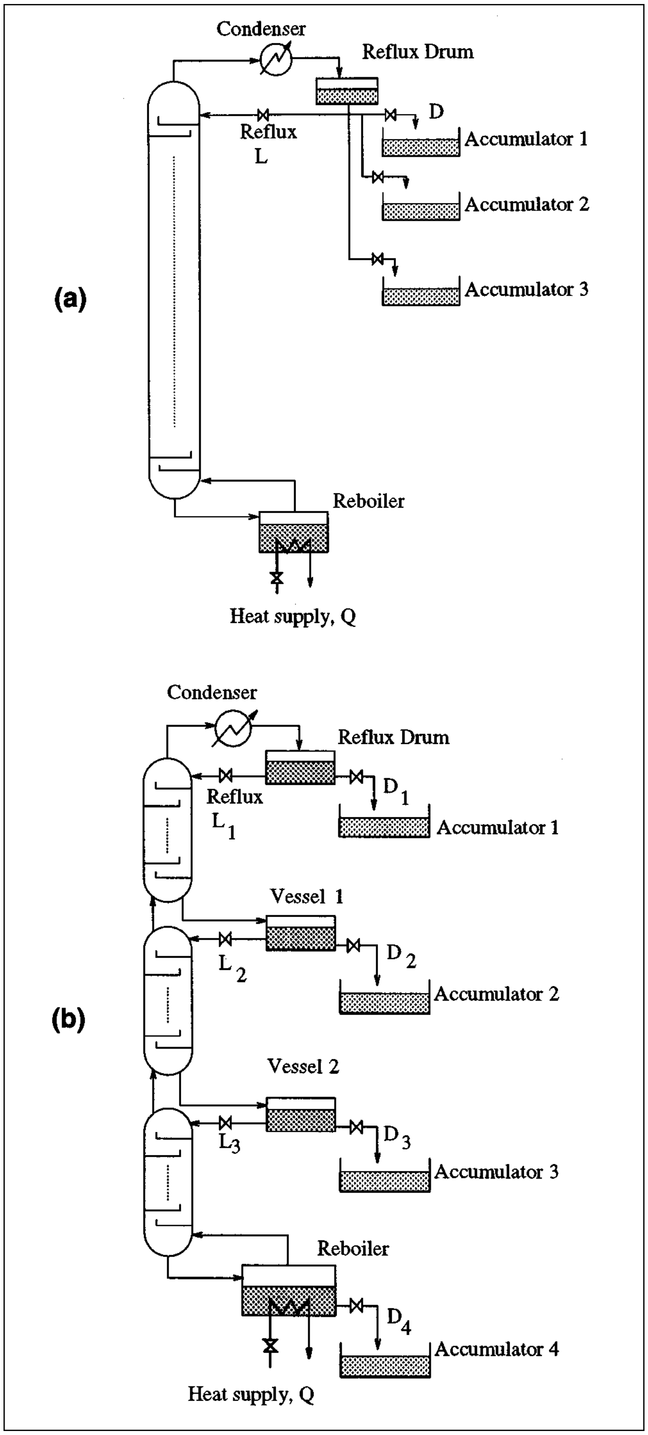

Figure 1. Different batch distillation column configurations.

(a) Regular batch distillation column; (b) multivessel batch distillation column.

the multivessel column consumed less energy than the regular column; in fact, it approached the energy efficiency of a sequence of continuous distillation columns.

Hasebe et al. (1995) considered the total reflux operation of the multivessel column in which the feed was initially distributed among all the vessels. The holdup in each vessel was calculated in advance, based on the quantity and the composition of the feed. A control scheme using level controllers in order to maintain constant holdup in the side vessels was proposed. The disadvantage of this control strategy is that the feed composition must be known *a priori*. In contrast, Wittgens et al. (1996) and Skogestad et al. (1997) used temperature controllers to adjust the holdup in each vessel. The liquid flow rate out of each vessel was manipulated based on a measurement of the temperature within the column section below each vessel. The temperature set points were taken as the average of the boiling points of the key and heavier than key components in each column section. Wittgens and Skogestad (1997) also considered the case where the feed is charged to the reboiler instead of being distributed among the vessels. They showed, via both simulations and experiments, that this control strategy is robust and achieves high purities independent of the feed composition. However, none of these earlier works attempted any optimization of the multivessel column operation.

Hasebe et al. (1997) considered the separation of a ternary mixture using a middle vessel column implementing a *variable* holdup policy. The whole feed was charged initially to the reboiler, and the holdups in the reflux drum and side vessel were allowed to increase by adjusting the reflux. The maximum holdups in the reflux drum and the side vessel, as well as the minimum holdup in the reboiler, were calculated from the feed composition and quantity, just as in the constant holdup policy (Hasebe et al., 1995). The liquid flow rates were optimized so as to minimize the operating time. The variable holdup policy with the feed initially charged to the reboiler yielded between 11% and 43% more distillate per batch time than the total reflux policy in which the feed was distributed among the vessels. The optimal reflux ratio policy of a regular batch distillation column having the same number of trays as the multivessel column was also determined. Four production steps, comprising three main cuts and two off-cuts (which were not recycled), were used and the optimal constant reflux ratio for each production step was determined. The multivessel column proved to be able to produce up to 114% more distillate per unit batch time than the regular column.

Batch distillation is inherently dynamic, and its simulation and optimization can be computationally expensive. In the past, this has led to the introduction of several modeling assumptions; in fact, all of the studies mentioned earlier make use of greatly simplified mathematical models. For instance, constant relative volatility is invariably assumed and energy dynamics are neglected. All of these factors cast certain doubts on the validity of any "optimal" solutions obtained, or indeed on any comparisons made between competing batch distillation configurations.

In general, comparisons between different column configurations are valid only if all of them are operated optimally with respect to *all* the degrees of freedom that are available to them. In this context, we note that the multivessel column offers a much wider range of degrees of freedom than has so far been considered in the literature. For instance, none of the studies mentioned here has considered product removal from the intermediate vessels, the reflux drum, or the reboiler. In fact, the product withdrawal rates can be treated as additional decision variables. Also, the accumulation of liquid in the intermediate vessels makes it possible for reflux flow rates to exceed flow rates into the vessels. In the case of the feedback control strategies described earlier, optimization of the controller parameters has not been undertaken. Furthermore, the performance of the column may be improved if the reboiler heat duty and the initial feed distribution are also optimized.

This article is concerned with the determination of optimal operating policies for multivessel batch distillation columns. In contrast to earlier work, a detailed model of distillation column dynamics is used and the additional degrees of freedom mentioned above are exploited. In the next section, we outline the key features of our model before introducing the optimization objective function and constraints. We then describe a case study involving the separation of a quaternary mixture in a multivessel column with two intermediate vessels. The optimal performance of the column is also compared with that of a regular batch distillation column.

Mathematical Model

As stated earlier, mostly simplified models have been used for studying multivessel batch distillation columns. In Appendix A, the effect of model simplifications on the optimal solution obtained for a regular column is investigated. Our results, as well as the recent findings of Tomazi (1997), show that certain common modeling assumptions (such as negligible tray holdup and constant molar overflow) have a significant impact on the optimal solutions obtained. More specifically, it is demonstrated that optimal solutions obtained using simplified models often violate important constraints; moreover, terms which are often assumed to be negligible or constant throughout the operation are neither.

Hence, our approach is to utilize a more detailed model than has so far been used. In particular, we have disposed of assumptions, the removal of which does not result in excessive increase in mathematical complexity or the need for obtaining values for quantities which cannot readily be estimated or measured. Thus, our tray model:

- incorporates a dynamic energy balance equation instead of relying on the usual assumption of constant molar overflow
- takes account of both liquid and vapor tray holdups, with their combined value being a function of the prevailing pressure and the intertray spacing
- allows the tray liquid holdup to vary, the liquid flow rate from the tray being determined by a modified Francis weir formula
- employs a detailed pressure drop equation that takes account of both ''dry'' and ''wet'' head losses on each tray, thereby determining the vapor flows
- replaces the usual constant relative volatility assumption by the use of liquid and vapor fugacities which may generally depend on temperature, pressure, and composition.

Dynamic material and energy balances are also used to model the accumulator, reflux drum, vessels, and reboiler. In each of these, both liquid and vapor holdups are taken into account. The condenser model assumes total condensation with no subcooling.

The resulting mathematical model, a summary of which is given in Appendix B, takes the form of a set of differential and algebraic equations. The total number of variables for the configurations studied in this article is approximately 3,000, which is well within the range of problems that can be simulated and optimized using currently available computing hardware and software.

Optimization Objective and Constraints

In general, the operation of batch distillation columns must satisfy constraints on the purity and the mean production rate for each of their products. Within these limitations, it is reasonable to seek to maximize the economic performance of the process taking account of the value of its product(s) and the cost of its raw materials and utilities. If it is assumed that the size of the market for the product(s) is determined by existing demands, then there is no benefit to be derived from producing above the minimum throughputs specified. Moreover, for high product purities, the product throughputs essentially determine the mean rate of consumption of the raw material. In such cases, the only remaining factor to be considered is the consumption of utilities and, in particular, energy in the reboiler.

If the processing time t_f per batch is allowed to vary for the purposes of optimization, then we need to consider the mean rate of energy consumption rather than the total energy consumed per batch. Hence, we are led to the objective function

$$\min \frac{\int_0^{t_f} Q_R(t)\,dt}{t_f + t_s} \tag{1}$$

where $Q_R(t)$ is the instantaneous rate of energy consumption in the reboiler and t_s is the time (s) required for charging the column at the start of each batch and emptying it at the end (the batch setup time). The latter is usually assumed to be constant.

As has already been mentioned, the above minimization may have to satisfy purity and production rate constraints for one or more of its products $i = 1, \ldots, N_V + 2$. (Here, N_V is the number of intermediate vessels. The products i are numbered from the top, with $i = 1$ and $i = N_V + 2$ corresponding to the reflux drum and the reboiler respectively.) These can be written as

$$x_{A,i}^{\max} \geq x_{A,i}(t_f) \geq x_{A,i}^{\min}, \qquad \forall i = 1, \ldots, N_V + 2 \tag{2}$$

$$\frac{H_{A,i}}{t_f + t_s} \geq \Pi_i^{\min}, \qquad \forall i = 1, \ldots, N_V + 2 \tag{3}$$

Here, $x_{A,i}$ refers to the vector of compositions in the ith product (mol/mol) and $H_{A,i}$ is the amount of product collected per batch (mol). For the intermediate vessels and the reboiler ($i = 2, \ldots, N_V + 2$), it is assumed that $H_{A,i}$ comprises both the liquid collected in the corresponding product vessel and accumulator by the end of the batch, as well as the liquid that remains in the column section immediately above it. As in the case of the energy consumption, each $H_{A,i}$ is divided by the sum of the processing and setup times to yield the corresponding mean production rate, Π_i.

Solution Methodology

The determination of optimal operating policies for batch distillation columns is a dynamic optimization problem. We solve it using a control vector parameterization approach (Vassiliadis et al., 1994a,b) as implemented within the *gPROMS* process modeling tool (Process Systems Enterprise Ltd., 1998).

A major requirement for the successful use of the control vector parameterization approach is that the underlying DAE system must have a solution for *any* set of values or time variations that the decision variables may take. Ensuring this is not always a trivial task. Consider, for instance, one of the intermediate vessels in a multivessel batch distillation column, as shown in Figure 2. Typically, the flow rates $L(t)$ and $D(t)$ of the streams (mol/s), respectively, returned to the column and were withdrawn as products treated as control variables. On the other hand, the flow rate $L^{\text{in}}(t)$ of the stream entering the vessel is determined by the solution of the column equations.

The overall mass balance equation for the above vessel is simply

$$\frac{dM}{dt} = L^{\text{in}} - L - D \tag{4}$$

The difficulty arises because it is possible to select time profiles for the controls $L(t)$ and $D(t)$ that will drive the holdup $M(t)$ (mol) to negative values, which is physically unrealistic. One way of avoiding this problem is to modify the above mass balance to

$$\frac{dM}{dt} = L^{\text{in}} - L' - D' \tag{5}$$

where L' and D' are *effective* reflux and product flow rates related to the control variables L and D, respectively, via equations of the form

$$L' = L(1 - e^{-M/M^*}) \tag{6}$$

$$D' = D(1 - e^{-M/M^*}) \tag{7}$$

where M^* is a small (constant) holdup value. We can see that for $M(t) \gg M^*$ the above equations reduce to $L' \approx L$ and $D' \approx D$. On the other hand, as $M \to 0$, both L and D also tend to zero. Hence, assuming that the initial holdup value $M(0)$ is positive, $M(t)$ can never become negative for any nonnegative control profiles $L(t)$ and $D(t)$.

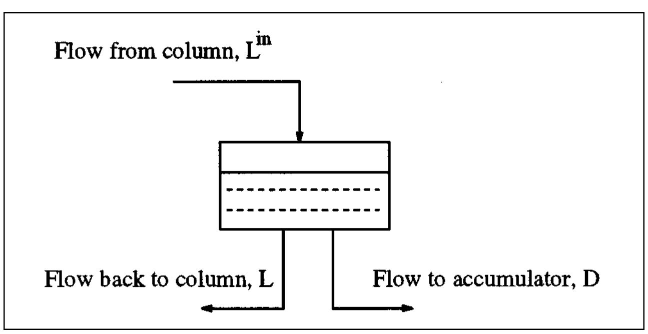

Figure 2. Intermediate vessel in a multivessel batch distillation column.

Table 1. Column Dimensions and Other Characteristics

Accumulator	Volume	1.1×10^{-2} m^3
Reflux drum*	Cross-sectional area	7.9×10^{-4} m^2
	Volume	7.9×10^{-5} m^3
Reflux drum** and Side vessels**	Cross-sectional area	3.0×10^{-2} m^2
	Volume	1.2×10^{-2} m^3
Trays	Weir height	6 mm
	Weir length	18 mm
	Plate diameter	50 mm
	Hole diameter	1.5 mm
	Number of holes	127
	Volume between trays	1.2×10^{-4} m^3
	Total number of trays	30
	Aeration factor	0.6
	Wall correction factor	0.98
Reboiler	Cross-sectional area	3.0×10^{-2} m^2
	Volume	1.2×10^{-2} m^3

*For regular column only.
**For multivessel column only.

The above technique was applied to the cases where the reflux and product withdrawal flow rates from the reflux drum, side vessels, and reboiler were optimized.

Description of Case Study: Separation of Alcohol Mixture

Separation problem

We consider the separation of 100 mol of an equimolar quaternary mixture of methanol, ethanol, *n*-propanol, and *n*-butanol into four high-purity products. We carry this out in a column with two intermediate vessels (cf. Figure 1b).

The column dimensions and other characteristics are in Table 1. The tray dimensions correspond to the laboratory distillation column at the Dept. of Chemical Engineering in University College London. The condenser pressure is atmospheric.

For comparison, we also consider the same separation in a regular column producing three main distillate cuts (see Figure 1a). An off-cut is also allowed to be taken after each main cut; however, no recycling of these off-cuts is undertaken. In order to make the comparison as fair as possible, the same total number of trays is used in both configurations.

For the purposes of modeling, this particular mixture was assumed to be ideal with Wagner's equation being used for calculating vapor pressure. The validity of this assumption was checked by performing an equilibrium flash calculation on the feed mixture at 1 atm and 362 K (that is, the average of the bubble point and dew point temperatures). A comparison of the results with those obtained using the Soave-Redlich-Kwong equation of state indicates that the ideal model provides a reasonable approximation for this mixture.

Liquid and vapor enthalpies were calculated from the specific heat capacities of the individual species and the latent heat of vaporization was determined by the Clausius-

Clapeyron equation. Pure component physical property data were obtained from Walas (1985).

Initial Conditions

Simulations of the startup period of operation, with the trays initially being dry, is not performed. Instead, we have assumed that the initial composition throughout the column is that of the feed and the liquid is at its boiling point (356 K for the equimolar mixture considered). The initial liquid holdup on each tray (taken to be 0.12 mol) was chosen such that the liquid level is above the weir height so that there is liquid flow down the column from the very start of the operation. Sadotomo and Miyahara (1983) have shown that these conditions provide a reasonably accurate approximation.

Operating Policies

In the case of the multivessel column, four modes of operation are considered, differing in the initial distribution of the feed (other than the 3.6 mol initially distributed on the trays) and the allowable control manipulations:

Policy 1: Constant holdup

The feed is distributed equally among the reboiler, vessels, and reflux drum (24.1 mol each). All these holdups are kept constant throughout the operation which takes place under total reflux.

Policy 2: Optimal holdup

The holdups in the reboiler, side vessels, and reflux drum are allowed to vary. The feed either is charged entirely to the reboiler or is optimally located. The column is operated under total reflux and the reflux flow rates (mol/s) ($L_i(t)$, $i=1, \ldots, 3$) are control variables to be manipulated by the optimization.

Policy 3: Optimal product withdrawal

Unlike the two previous policies, material is allowed to be withdrawn into accumulators during the operation. It should be noted that Policies 1 and 2 are special cases of Policy 3 since the latter also allows the option of holding the vessel holdups constant and/or having no product withdrawal. At the end of the operation, the contents of the reflux drum, two side vessels, and reboiler are mixed with those of accumulators 1, 2, 3 and 4, respectively, to give the final products. The holdups in the vessels are allowed to vary by manipulating the total outlet flow rate $L_i + D_i$ from each vessel i, $i=1, \ldots, 4$. (Note that $i=4$ corresponds to the reboiler and, therefore, $L_4 = 0$.) The fraction of this flow rate which is returned to the column, and, hence, the fraction which is withdrawn into accumulators, is given by R_i [$= L_i/(L_i + D_i)$], $i = 1, \ldots, 3$. Both $L_i + D_i$, $i=1, \ldots, 4$, and R_i, $i=1, \ldots, 3$ are treated as control variables to be manipulated for the purposes of the optimization. Three different feed locations are considered: (a) feed equally distributed among the reflux drum, vessels and reboiler; (b) feed placed in the reboiler; (c) feed optimally distributed.

Table 2. Optimization Policies Considered

Column Configuration	Policy	Type of Operating Policy	Optimization Decision Variables
Multivessel	1	Constant holdup[a]	$Q_R(t)$
	2	Optimal holdup variation[b,c]	$L_i(t)$, $Q_R(t)$
	3	Optimal product withdrawal[a,b,c]	$L_i(t) + D_i(t)$, $R_i(t)$, $Q_R(t)$
	4	Feedback control[a,b,c]	$k_i(t)$, $Q_R(t)$, T_i^{set}
Regular		Optimal reflux ratio[b]	$R(t)$, $Q_R(t)$

[a] Feed equally distributed.
[b] Feed charged to the reboiler.
[c] Feed optimally distributed.

Policy 4: Feedback control

A feedback control strategy proposed by Wittgens et al. (1996) is employed, whereby the reflux flow rates ($L_i(t)$, $i=1, \ldots, 3$) are adjusted based on temperature (K) measurements (T_i, $i=1, \ldots, 3$) on the middle tray of each column section (in this case, on the fifth tray from the top of each column section). Proportional controllers of the form

$$L_i = \beta_i - k_i\left(T_i^{\text{set}} - T_i\right), \qquad \forall i = 1, \ldots, 3 \tag{8}$$

are used. The controller gains (mol/sK) ($k_i(t)$, $i=1, \ldots, 3$) and the temperature (K) set points (T_i^{set}, $i=1, \ldots, 3$) are optimized. The biases (β_i, $i=1, \ldots, 3$) are always maintained equal to the flow rate of the liquid entering the vessel at any particular point in time, so that constant holdup operation occurs when $T_i = T_i^{\text{set}}$. Three different cases are considered where the feed is (a) equally distributed, (b) charged to the reboiler, and (c) optimally located.

For the case of the regular column operation, the main optimization decision variables are the reflux ratio $R(t)$ and the durations of each of the cuts, including both main cuts and off-cuts.

In all of the cases considered, the reboiler heat duty is allowed to vary during the operation and is also optimized. The batch processing time t_f (s) is also treated as an optimization decision variable, while the setup time t_s is fixed at 30 min. The different optimization cases considered are summarized in Table 2.

Minimum purity requirements are imposed on a different component in each product. These specifications are shown in Table 3 together with the corresponding minimum mean production rates. The values used were obtained from a dynamic simulation of the multivessel column using Policy 1

Table 3. Minimum Purity and Production Rate Specifications

Product Location	Key Component	Minimum Purity (mol %)	Minimum Mean Production Rate (mol/min)
Reflux drum/Accumulator 1	methanol	92.8	0.209
Vessel 1/Accumulator 2	ethanol	85.4	0.225
Vessel 2/Accumulator 3	*n*-propanol	91.4	0.221
Reboiler/Accumulator 4	*n*-butanol	97.0	0.213

Table 4. Optimal Results for Different Operating Policies

Configuration	Multivessel									Regular
Policy	1	2		3			4			
Feed location	*a*	*b*	*c*	*a*	*b*	*c*	*a*	*b*	*c*	*b*
Mean rate of energy consumption (W)	1,478	1,253	1,224	1,394	1,252	**1,220**	1,334	1,270	1,264	2,692
Batch processing time, t_f (min)	85	85	85	85	85	85	85	85	85	82
Figures	3	4, 5	6, 7	8, 9	10, 11	12, 13	14, 15	16, 17	18, 19	21

a Equally distributed.
b Reboiler.
c Optimal.

(constant vessel holdup under total reflux) with a constant heat input of 2 kW and a batch time of 85 min. The latter corresponds to the time taken for the slope of all four distillate composition curves (that is, the mole fraction of the key component in each vessel) to drop to a value of 0.1 mol % per min, which is considered to be sufficiently close to steady state. It should be noted that the values of the minimum mean production rates of the four products, computed in the above manner, are different. This is due to the different amounts of liquid left in each of the three column sections at the end of the batch.

Bounds on Decision Variables

A piecewise constant representation of the control variables listed in the last column of Table 2 is employed. The decision variables are generally subject to the following bounds

$$Q_R(t) \in [0.75, 5.5]\ \text{kW} \tag{9}$$

$$L_i(t) + D_i(t) \in [0.48, 4.2]\ \text{mol/min} \tag{10}$$

$$k_i(t) \in [0.0048, 0.0960]\ \text{mol/min}\cdot\text{K} \tag{11}$$

$$H_{D,i}(0) \in [0.6, 94.6]\ \text{mol} \tag{12}$$

$$T_1^{\text{set}} \in [337.7, 351.4]\ \text{K} \tag{13}$$

$$T_2^{\text{set}} \in [351.4, 370.3]\ \text{K} \tag{14}$$

$$T_3^{\text{set}} \in [370.3, 390.9]\ \text{K} \tag{15}$$

$$R_i(t) \in [0.5, 1.0] \tag{16}$$

In all the multivessel column cases, the upper bound on the processing time (t_f) is taken as 85 min and the time horizon is divided into six control intervals of variable duration bounded between 0.2 min and 50 min. For Policy 4, the temperature set point of each controller is bounded by the boiling points of the two main components being separated in the respective column section (bounds 13, 14 and 15).

In the regular column case, the upper bound on the processing time (t_f) is also taken as 85 min and the time horizon is divided into seven control intervals of variable duration. During the first interval, the duration of which is fixed at 5 min, the column is operated under total reflux [$R(t) = 1$] and a reboiler heat duty of 2 kW. Each subsequent control interval corresponds to a different cut (main or off-cut).

Comparison of Operating Policies for an Equimolar Alcohol Mixture

The results for the different cases considered are summarized in Tables 4 and 5. The main characteristics of the column behavior for each policy are shown in Figures 3 to 21.

Feed equally distributed

First, consider the cases where the feed is distributed equally among the vessels, that is, the constant holdup policy (Policy 1a), the product withdrawal policy (Policy 3a), and the feedback control policy (Policy 4a). An examination of the optimal solution for Policy 3a indicates that no product withdrawal actually takes place (that is, the reflux ratios are maintained at a constant value of 1). Thus, the lower energy consumption achieved by Policies 3a and 4a in comparison

Table 5. Product Purities and Production Rates Obtained with Different Operating Policies

Configuration	Policy	Product Purity, mol %	Production Rate, mol/min
Multivessel	1a	[92.9*, 85.4, 91.4, 97.0]	[0.209, 0.225, 0.221, 0.213]
	2b	[92.8, 85.4, 91.4, 97.0]	[0.209, 0.225, 0.221, 0.213]
	2c	[92.8, 85.4, 91.4, 97.0]	[0.210*, 0.225, 0.221, 0.214*]
	3a	[92.8, 85.4, 91.4, 97.0]	[0.210*, 0.225, 0.221, 0.213]
	3b	[92.8, 85.4, 91.4, 97.0]	[0.209, 0.225, 0.221, 0.213]
	3c	[92.8, 85.4, 91.4, 97.0]	[0.210*, 0.225, 0.221, 0.214*]
	4a	[92.8, 85.4, 91.4, 97.0]	[0.210*, 0.226*, 0.221, 0.213]
	4b	[92.8, 85.4, 91.4, 97.0]	[0.210*, 0.225, 0.222*, 0.214*]
	4c	[92.8, 85.4, 91.4, 97.0]	[0.216*, 0.226*, 0.221, 0.213]
Regular		[94.6*, 85.4, 91.4, 97.0]	[0.209, 0.225, 0.221, 0.213]

*Above specification.

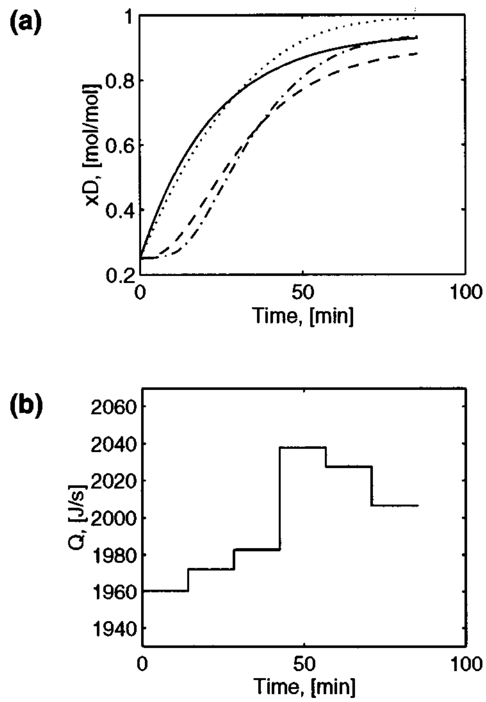

Figure 3. Optimal results for Policy 1a.

(a) Mole fraction of main component in each vessel (—— reflux drum, – – – vessel 1; –·–·– vessel 2, · · · · reboiler); (b) reboiler heat duty.

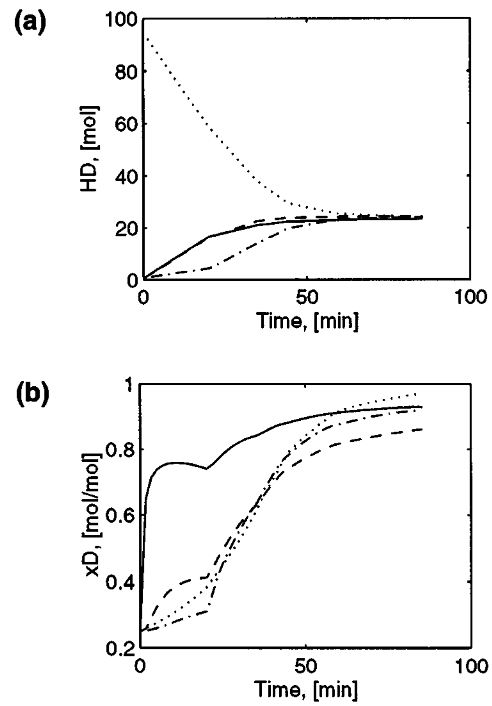

Figure 4. Optimal results for Policy 2b.

(a) Vessel holdups; (b) mole fraction of main component in each vessel (—— reflux drum; – – – vessel 1; –·–·– vessel 2; · · · · reboiler).

with Policy 1a is primarily the result of allowing the holdups to vary (see Figures 8a and 14a).

The superiority of the feedback control policy 4a over the product withdrawal policy 3a in this case may be attributed to the fact that the latter is restricted to a piecewise-constant profile for the control variable $L_i(t)$ (Figure 9a), whereas Policy 4a makes use of a more complex temporal variation (Figure 14c). In order to verify this, we repeated the calculation, this time doubling the number of control intervals used by Policy 3a, thus allowing it more freedom in varying $L_i(t)$. This resulted in an 8% reduction in the mean rate of energy consumption over the feedback control strategy.

Feed charged to the reboiler

Next, consider the cases where the entire feed is charged to the reboiler, that is, the optimal holdup policy (Policy 2b), optimal product withdrawal policy (Policy 3b), and the feedback control policy (Policy 4b). Of these, the optimal product withdrawal policy required the least energy consumption. The vessel holdup, distillate composition, and control variable profiles are shown in Figures 4 and 5 for the optimal holdup policy, and in Figures 10 and 11 for the product withdrawal policy. It is interesting to note that the optimal control variable profiles and energy consumption for the product withdrawal policy are almost identical to those of the optimal holdup policy. In fact, an inspection of the optimal reflux ratio variation for Policy 3b indicates that $R(t) = 1$ throughout the batch. Hence, no product withdrawal actually takes place.

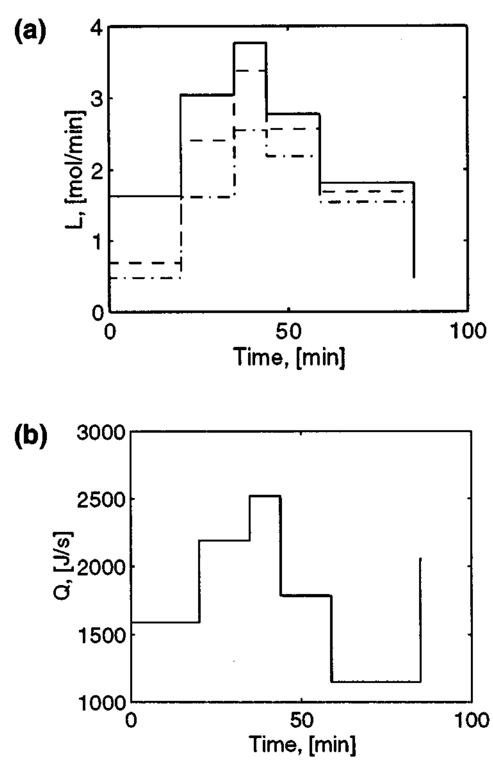

Figure 5. Optimal control variable profiles for Policy 2b.

(a) Reflux flow rates (—— L_1, – – – L_2, –·–·– L_3); (b) reboiler heat duty.

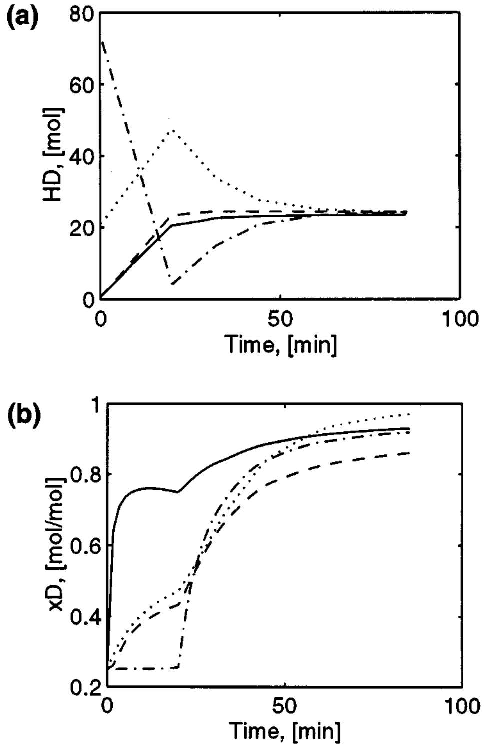

Figure 6. Optimal results for Policy 2c.

(a) Vessel holdups; (b) mole fraction of main component in each vessel (—— reflux drum; – – – vessel 1; –·–·– vessel 2; · · · · reboiler).

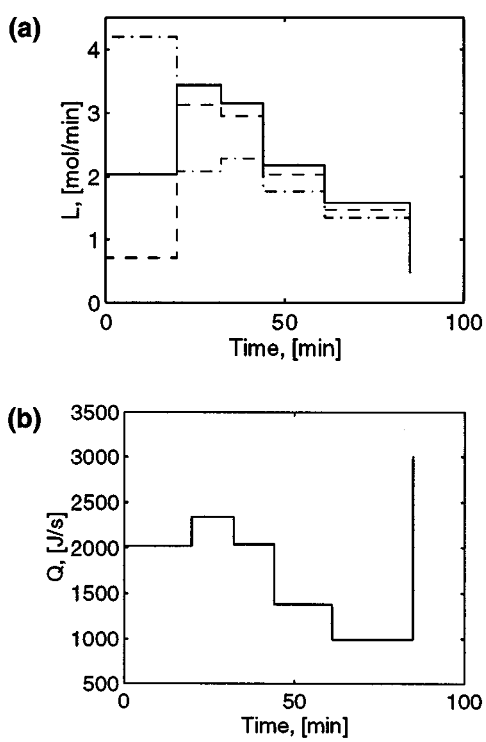

Figure 7. Optimal control variable profiles for Policy 2c.

(a) Reflux flow rates (—— L_1, – – – L_2, –·–·– L_3); (b) reboiler heat duty.

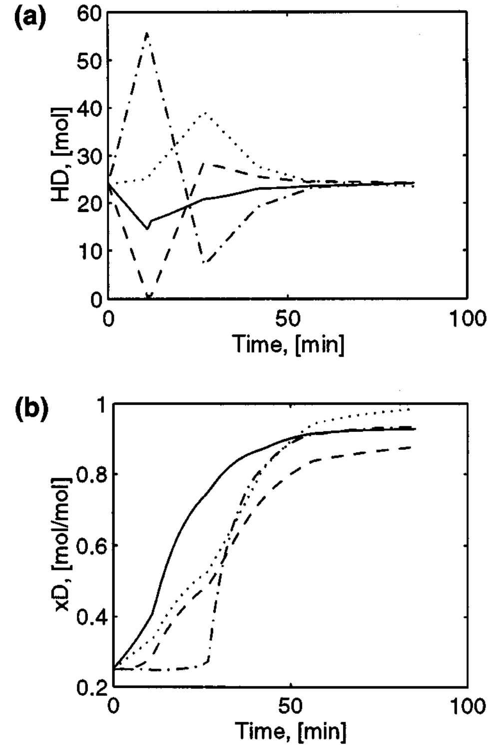

Figure 8. Optimal results for Policy 3a.

(a) Vessel holdups; (b) mole fraction of main component in each vessel (—— reflux drum; – – – vessel 1; –·–·– vessel 2, · · · · reboiler).

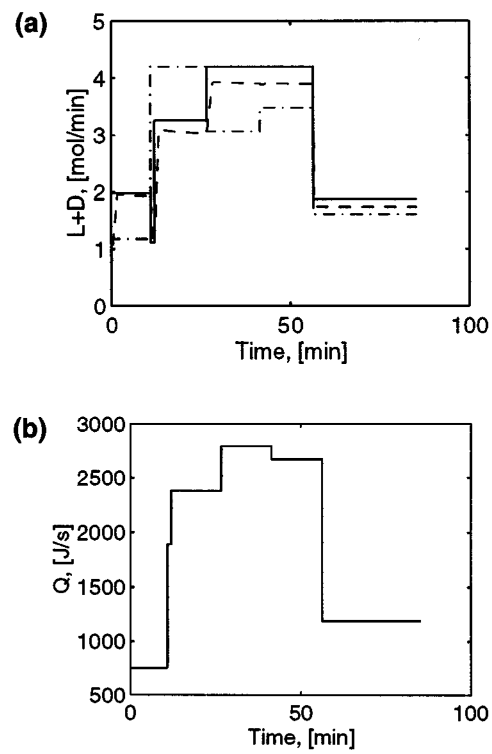

Figure 9. Optimal control variable profiles for Policy 3a.

(a) Vessel outlet flow rates (—— $L_1 + D_1$, – – – $L_2 + D_2$, –·–·– $L_3 + D_3$); (b) reboiler heat duty.

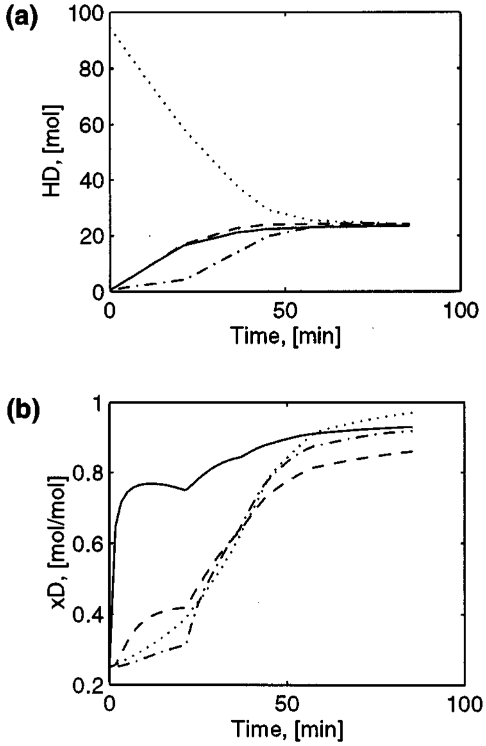

Figure 10. Optimal results for Policy 3b.

(a) Vessel holdups; (b) mole fraction of main component in each vessel (—— reflux drum; – – – vessel 1; – · – · – vessel 2; · · · · reboiler).

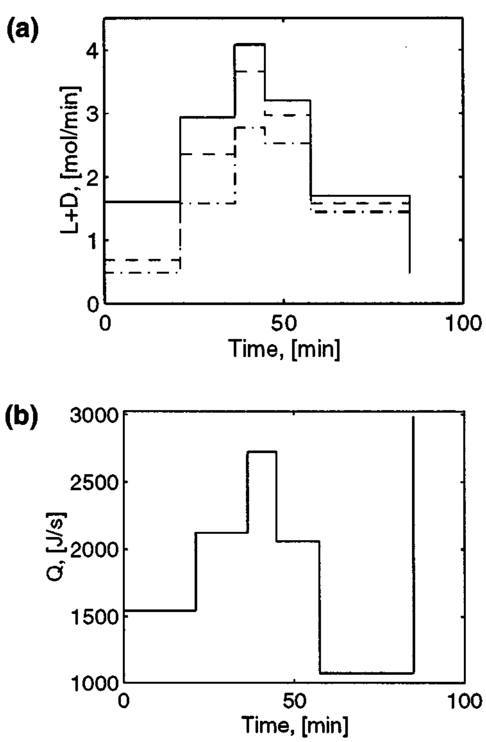

Figure 11. Optimal control variable profiles for Policy 3b.

(a) Vessel outlet flow rates (—— $L_1 + D_1$, – – – $L_2 + D_2$, – · – · – $L_3 + D_3$); (b) reboiler heat duty.

As a result, there appears to be little incentive for the additional complexity introduced by product withdrawal in this particular case.

Optimal initial feed distribution

Now, consider the effect of optimizing the initial feed distribution. First, compare the constant holdup policy (Policy 1a) where the feed is equally distributed, and the optimal holdup policy where the feed is charged to the reboiler (Policy 2b). In both cases, the operation takes place under total reflux; however, the energy consumption is about 15% less when the feed is charged to the reboiler. Similarly, in the case of the product withdrawal and feedback control policies respectively, a 10% and 5% reduction in energy consumption is achieved when the feed is charged to the reboiler over the case where it is equally distributed. Therefore, the initial feed distribution can have a significant impact on the energy efficiency of the operation.

Optimizing the feed distribution results in only a slight reduction in energy consumption (2%, 3%, and 1% for the optimal holdup, product withdrawal, and feedback control policies, respectively) compared to the cases where all of the feed is charged to the reboiler. For the optimal holdup and product withdrawal policies (2c and 3c, respectively), the optimal feed location involved most of the feed (74% and 91%, respectively) being charged to vessel 2 with the rest being placed in the reboiler. In contrast, most of the feed (84%) was placed in the reboiler in the case of the feedback control policy (Policy 4c).

From a practical point of view, it is certainly easier simply to introduce all the feed into the reboiler than to split it up among the vessels, particularly if there is no great economic incentive for doing so. However, there may be other cases where optimizing the feed location does result in significant energy savings; this is investigated later on.

Optimal product location

In contrast to the other policies considered, the product withdrawal policy (Policy 3) also optimizes the product location. In all the cases considered here, little or no withdrawal of product into the accumulators occurred during the batch; consequently, the products were left in the reflux drum, side vessels, and reboiler as shown in Figures 8a, 10a, and 12a. Simulations performed by Hasebe et al. (1995) also suggested that total reflux operation is better than the product withdrawal policy. However, as will be shown later on, this may not always be the case in general.

Feedback control policy

We now consider the feedback control policy (Policy 4) in more detail. In the case where the feed is equally distributed (Policy 4a), the action of the controllers causes the holdups to vary as shown in Figure 14a. The optimal temperature set points were 351.3 K, 359.6 K, and 374.9 K, which differ from the average boiling points of 344.6 K, 360.9 K, and 380.6 K, respectively, used by Wittgens et al. (1996). The reflux flow rate profiles, shown in Figure 14c, follow the same trend as the reboiler heat duty profile, shown in Figure 15c. Optimiz-

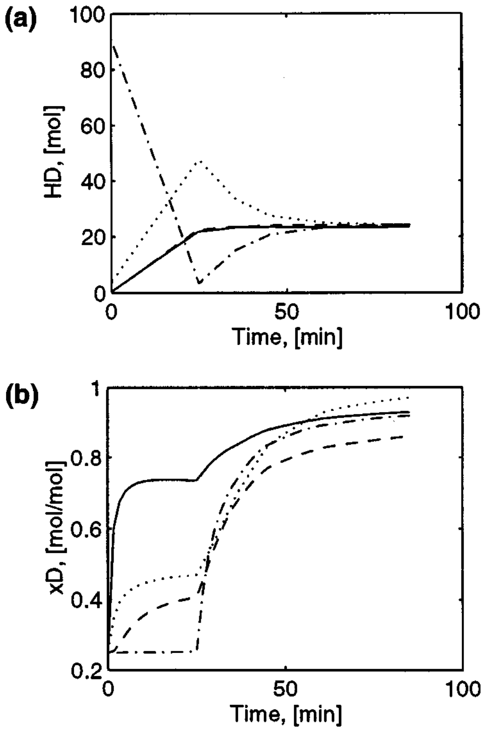

Figure 12. Optimal results for Policy 3c.

(a) Vessel holdups; (b) mole fraction of main component in each vessel (—— reflux drum; – – – vessel 1; – · – · – vessel 2; · · · · reboiler).

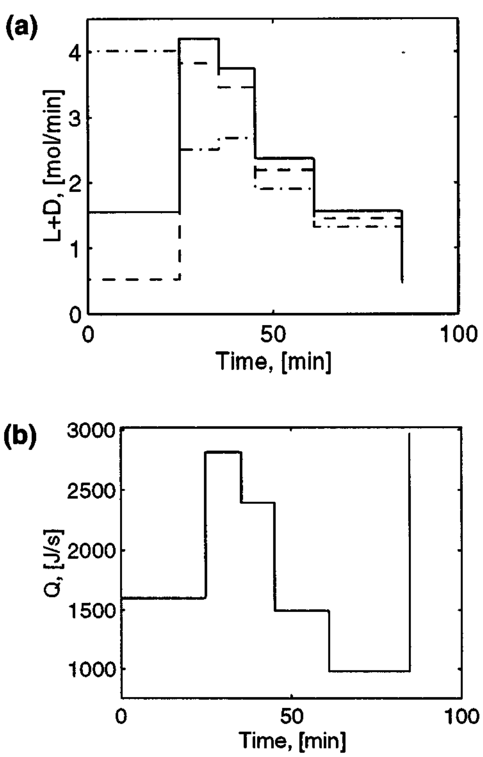

Figure 13. Optimal control variable profiles for Policy 3c.

(a) Vessel outlet flow rates (—— $L_1 + D_1$, – – – $L_2 + D_2$, – · – · – $L_3 + D_3$); (b) reboiler heat duty.

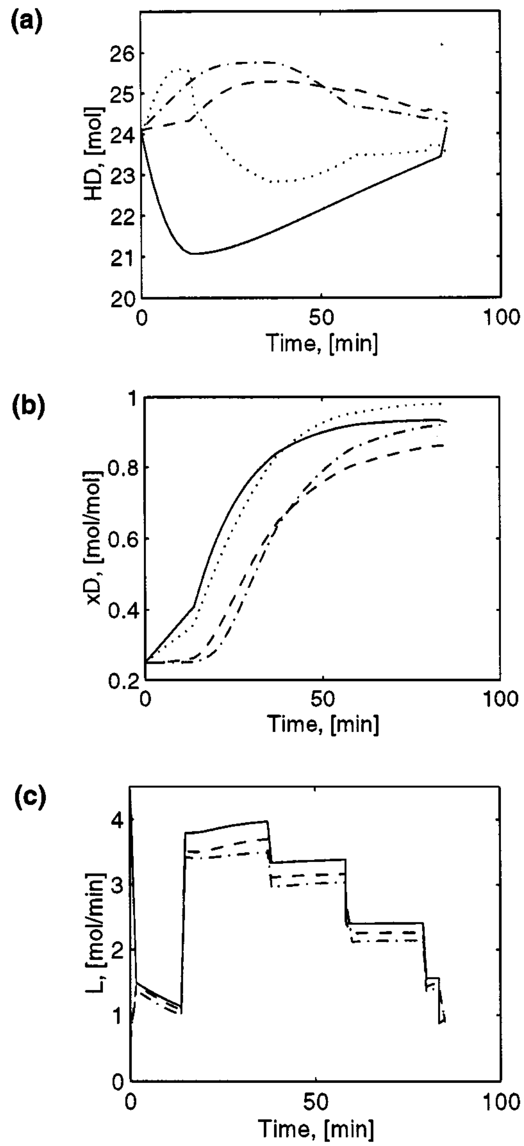

Figure 14. Optimal results for Policy 4a.

(a) Vessel holdups; (b) mole fraction of main component in each vessel; (c) reflux flow rates (—— reflux drum; – – – vessel 1; – · – · – vessel 2; · · · · reboiler).

ing the controller set points and gains (shown in Figure 15a), is essentially an indirect method of optimizing the reflux flow rates.

At the start of the operation, the initial temperature on the trays is the boiling point of the feed. As the operation proceeds, the lightest components (that is, methanol and ethanol) rise up the column, and, consequently, the temperature on tray five of the top column section (that is, the tray on which temperature measurements are taken for controller 1) drops to a value between the boiling points of these components (337.7 K and 351.4 K), as shown in Figure 15b. Before the measured temperature crosses the temperature set point of controller 1 (351.3 K), the holdup in the reflux drum decreases because the reflux flow rate L_1 is greater than the inlet flow rate (according to Eq. 8). The holdup in the reflux

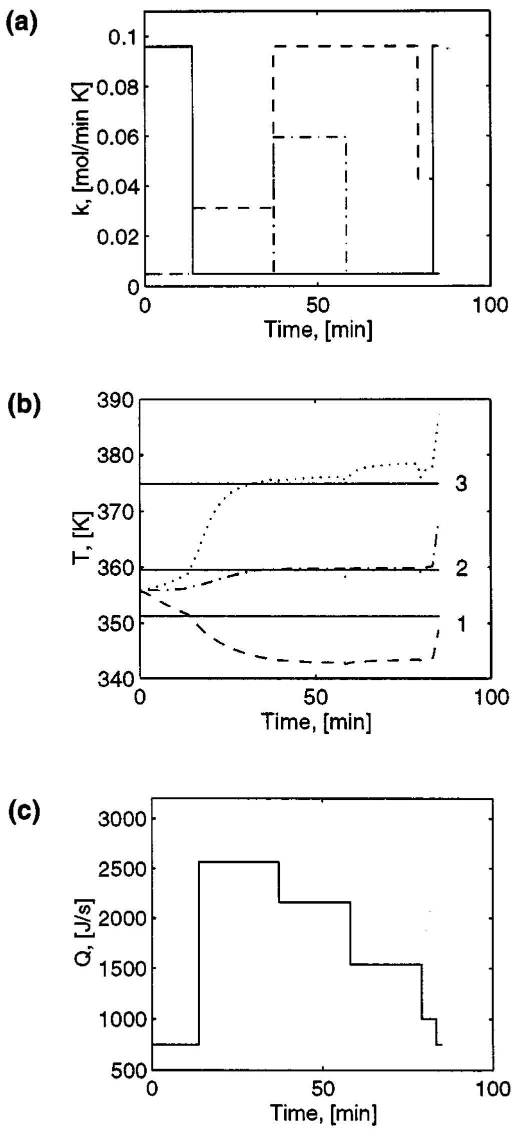

Figure 15. Optimal decision variables for Policy 4a.

(a) Controller gains (— — — k_1, —·—·— k_2, · · · · k_3); (b) controller set points (——) and temperature measurements (— — — T_1, —·—·— T_2, · · · · T_3); (c) reboiler heat duty.

drum begins to increase only when the temperature drops below the set point. The same analysis may be used to explain the variation of the holdup in the other vessels. At the very end of the operation, the measured temperature in each column section increases suddenly as the heavier than key component in each section rises up the column due to the removal of almost all of the key component into the respective vessel.

When the feed is charged to the reboiler (Policy 4b), the temperature set points for controllers 1 and 2 are almost the same as for the previous case (351.4 K and 360.4 K, respectively), but markedly different for controller 3 (390.0 K), as shown in Figure 17b. Since the products accumulate in the vessels, the holdup increases in the reflux drum and side vessels, and decreases in the reboiler. The temperature set points must, therefore, be selected so that the initial liquid flow rates out of the reflux drum and side vessels are less than the inlet flow rates; thus, initially, the set points must be above the measured temperatures, as can be seen from Figure 17b.

When the initial feed distribution is optimized (Policy 4c), almost all of the feed is charged to the reboiler, as shown in Figure 18a; this is in contrast to the optimal holdup and product withdrawal policies where the optimal feed location was in vessel 2. The control variable profiles and temperature set points, shown in Figures 18 and 19, are very similar to those for the case where the feed is charged to the reboiler (Policy 4b). The similarity in the optimal solutions of Policies 4b and 4c is reflected in the marginal difference in the optimal values of the mean rate of energy consumption (less than 1%).

It is interesting to note from the results of Figures 15b, 17b, and 19b that the quality of temperature control achieved

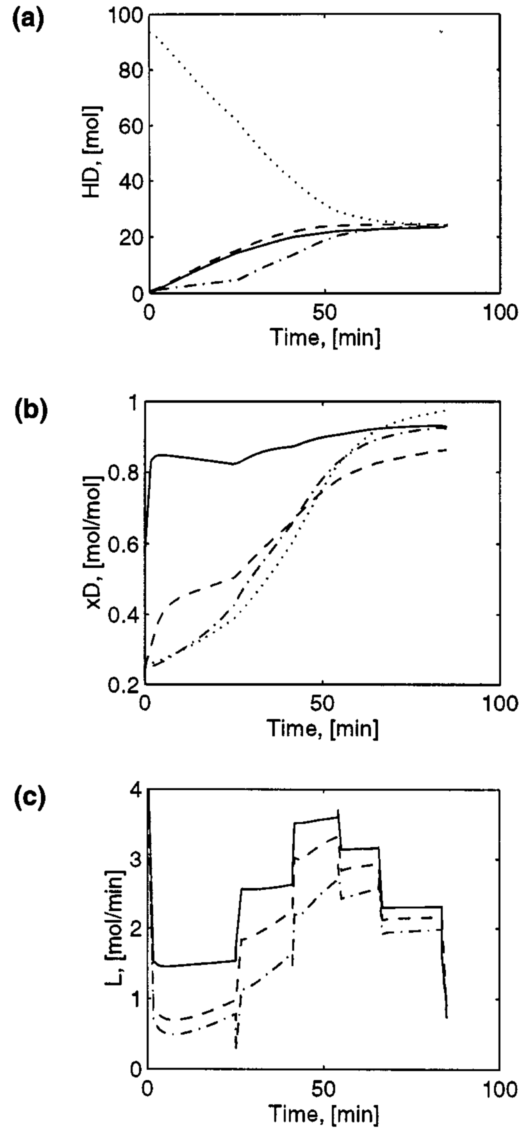

Figure 16. Optimal results for Policy 4b.

(a) Vessel holdups; (b) mole fraction of main component in each vessel; (c) reflux flow rates (—— reflux drum; — — — vessel 1, —·—·— vessel 2, · · · · reboiler).

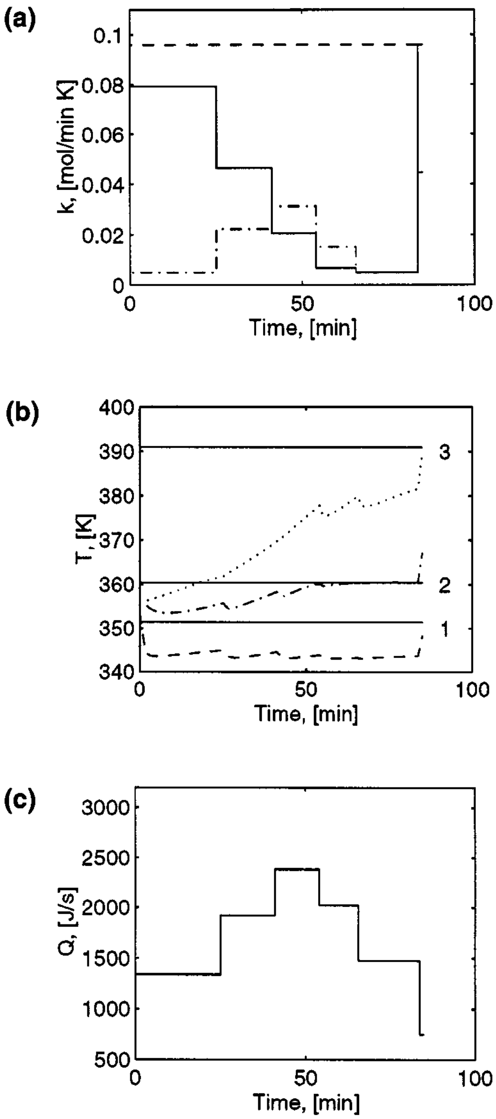

Figure 17. Optimal decision variables for Policy 4b.

(a) Controller gains (– – – k_1, – · – · – k_2, · · · · k_3); (b) controller set points (——) and temperature measurements (– – – T_1, – · – · – T_2, · · · · T_3); (c) reboiler heat duty.

by the optimal choice of values for the controller parameters is actually rather poor. An investigation was performed to determine the effect of tightening the degree of control on the mean rate of energy consumption. A function I_i, which is a measure of the deviation of the measured temperature T_i from the set point T_i^{set}, is defined as

$$I_i \equiv \int_0^{t_f} \left(\frac{T_i^{\text{set}} - T_i}{T_i^{\text{set}}} \right)^2 dt, \qquad \forall i = 1, \ldots, 3 \tag{17}$$

Thus, the smaller the values of I_i, the tighter the control. The base case is taken to be Policy 4a; using the results of Figure 15b, we can compute the corresponding values of I_i (denoted as I_i^{4a}) to be 1.91, 0.14, and 2.4 for $i = 1$, 2 and 3, respectively. We now repeat the optimization imposing the additional constraint

$$I_i \le \alpha_i I_i^{4a}, \qquad \forall i = 1, \ldots, 3 \tag{18}$$

where $\alpha_i \in (0, 1]$ is a fixed value representing the degree of tightening of the control. The resulting temperature measurement profiles and set points for different levels of control (that is, $I_i = I_i^{4a}$, $I_i = 0.5 I_i^{4a}$, and $I_i = 0.2 I_i^{4a}$) are shown in Figure 20. It can be seen that the deviation of the measured temperatures from their respective set points decreases as the level of control is tightened. However, the ''improved'' control results in increased energy consumption, as shown in Table 6.

As a final check on the trade-off between quality of control and energy consumption, we perform an optimization in which the quantity $\sum_{i=1}^{3} I_i$ is minimized by varying the coeffi-

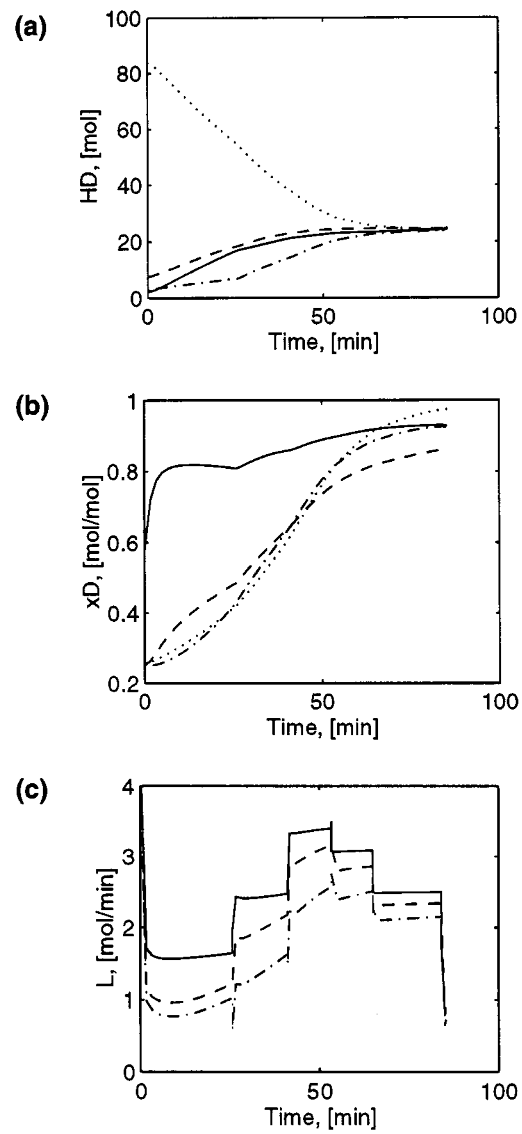

Figure 18. Optimal results for Policy 4c.

(a) Vessel holdups; (b) mole fraction of main component in each vessel; (c) reflux flow rates (—— reflux drum; – – – vessel 1; – · – · – vessel 2, · · · · reboiler).

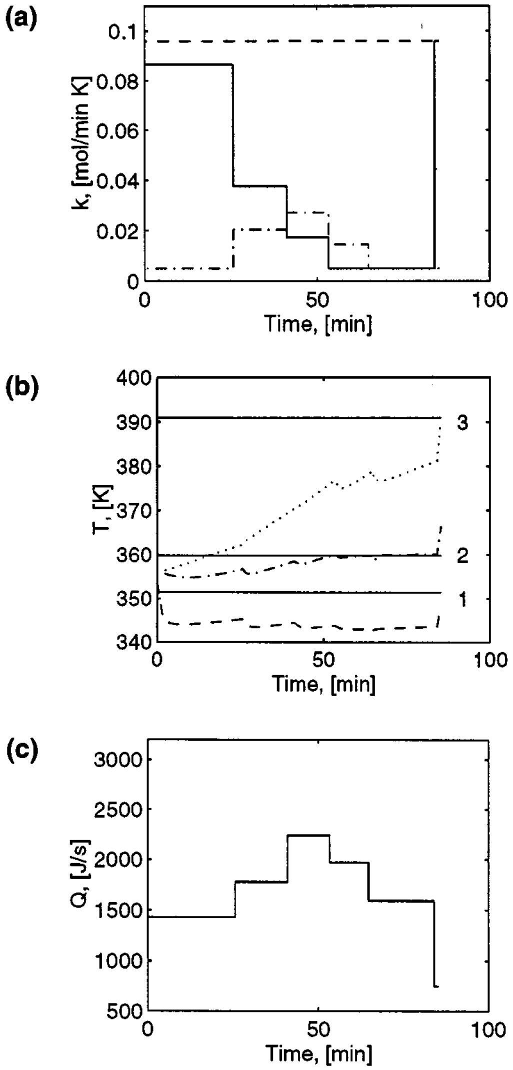

Figure 19. Optimal decision variables for Policy 4c.

(a) Controller gains (— — — k_1, –·–·– k_2, ···· k_3); (b) controller set points (——) and temperature measurements — — — T_1, –·–·– T_2, ···· T_3); (c) reboiler heat duty.

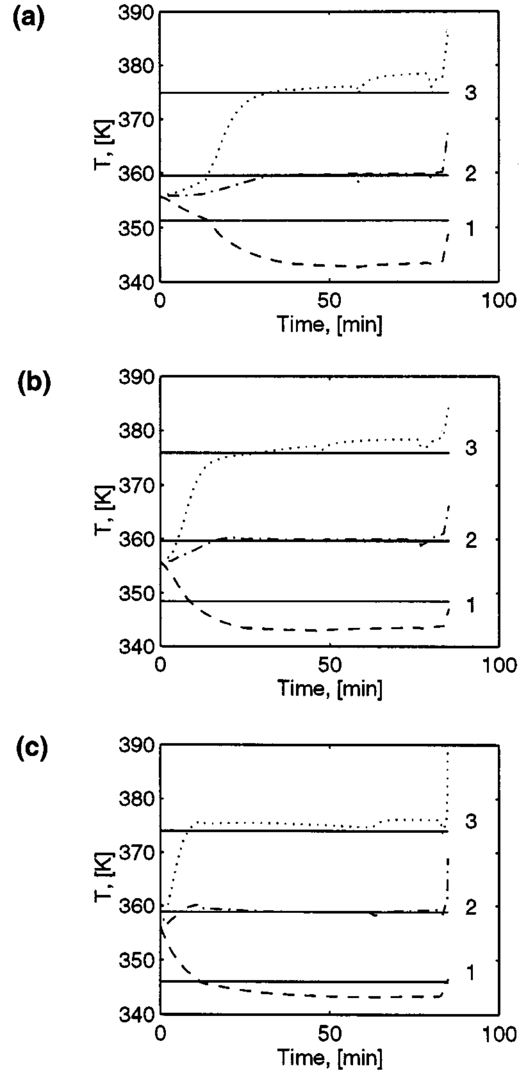

Figure 20. Temperature measurements and set points for different degrees of control.

(a) $I_i = I_i^{4a}$; (b) $I_i \leq 0.5 I_i^{4a}$; (c) $I_i \leq 0.2 I_i^{4a}$ (— — — T_1; –·–·– T_2; ···· T_3; —— T^{set}).

cients α_i, as well as all the other decision variables associated with Policy 4a. The results of this calculation are summarized in the last row of Table 6. We note that the mean rate of energy consumption increases significantly to almost 24% over the base case.

Multivessel column vs. regular column

If the same multivessel column is operated without the use of the side vessels, the configuration effectively switches to that of a regular column. In this case, products are withdrawn sequentially from the top of the column. As a result, the mean rate of energy consumption for the regular column is much higher (by between 82% and 121%) than the multivessel column. The reboiler heat duty profile (shown in Figure 21) is generally higher (between 1.3 and 5.3 kW) than that of the multivessel column (between 0.75 and 2.9 kW). This result highlights the significantly higher energy efficiency of the multivessel column over the regular column.

However, it should be noted that, even though the number of trays are the same in both column configurations, the capital cost of the multivessel column is expected to be higher. In order for a valid comparison to be made, a more detailed

Table 6. Effect of Tightening Temperature Control

Deg. of Tightness of Feedback Control (α_i)	Mean Rate of Energy Consumption (W)
(1.00, 1.00, 1.00)	1,334
(0.50, 0.50, 0.50)	1,352
(0.20, 0.20, 0.20)	1,376
(0.13, 0.14, 0.20)	1,651

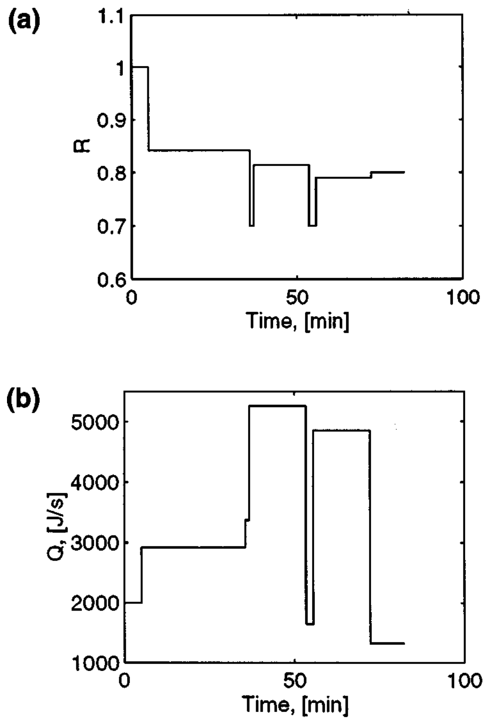

Figure 21. Optimal control variable profiles for the regular column.

(a) Reflux ratio; (b) reboiler heat duty.

objective function (such as annualized profit) should be used, taking account of all the capital and operating costs involved. Also, the possibility of withdrawing off-cuts and more than one main cut from each vessel in the operation of the multivessel column was not considered; this may well improve its performance.

Summary of results

The main findings arising from the results presented in this section are summarized below:

- As can be seen in Table 4, in all cases studied, the optimal multivessel column operation involves the processing time attaining its maximum value of 85 min. This results in the minimum energy consumption while still allowing the minimum production requirement constraints (Eq. 3) to be satisfied. If the production requirements are increased, then solutions with lower processing times (but higher mean energy consumption) will be obtained.
- The simple policy of maintaining constant holdup in all vessels with the feed being equally distributed (Policy 1a) required by far the largest mean rate of energy consumption (up to 21% more than other policies), whereas the optimal product withdrawal policy where the feed location is optimized (Policy 3c) required the smallest mean rate of energy consumption. Clearly, this is due to the greater number of degrees of freedom utilized by the optimal product withdrawal policy (such as the reflux and product withdrawal flow rates).
- Charging the feed to the reboiler was found to be better than distributing the feed according to its composition among the vessels.
- Optimizing the initial distribution of the feed was found to reduce energy consumption by as much as 17%; however, placing all the feed in the reboiler yields a performance that is only marginally worse than this.
- Product withdrawal did not offer significant advantage over the simpler total reflux operation.
- The mean rate of energy consumption of the feedback control strategy studied was found to be greater than open-loop operation. Furthermore, the mean rate of energy consumption was found to increase as the degree of control was tightened. Overall, there appears to be little direct relation between the feedback control objective of controlling the temperature in each column section and the economic objective of minimizing energy consumption. However, as stated earlier, a potential advantage of the feedback control strategy is that it is capable of handling uncertainty in the feed composition, which is an important consideration in many practical applications.
- The optimal operation of a regular column having the same number of trays as the multivessel column was found to require about twice the mean rate energy consumption of the multivessel column for the same separation.

It should be noted that these conclusions are for the equimolar quaternary mixture studied and are by no means general. In the next section, other feed compositions are considered.

Effect of Feed Composition on Optimal Initial Feed Distribution and Product Locations

As shown in the previous section, for the case of an equimolar alcohol mixture, charging the feed to the reboiler and operating under total reflux was close to optimal operation. In this section, we investigate whether this is generally the case, or if there are situations where optimizing the initial distribution of the feed and withdrawing product(s) are significantly advantageous.

The optimal product withdrawal policy where the feed location is optimized (Policy 3c) is used. Four different feeds, each rich (70 mol %) in one of the four components of a methanol, ethanol, *n*-propanol, and *n*-butanol mixture, are considered. As for the cases involving the equimolar feed, the product purity and production rate specifications were obtained from a simulation of Policy 1 (that is, the constant holdup policy where the feed is distributed according to its composition) with the processing time corresponding to the time taken for the slope of the distillate composition curves to drop to a value of 0.1 mol % per min.

The optimal feed and product locations for each case are shown in Table 7. We recall that, for the equimolar feed, the optimal feed location was found to be mainly in vessel 2 and that no product withdrawal to the accumulators actually took place. For ease of comparison, these results are also shown in Table 7.

When the feed is rich in methanol, most of the feed is placed in vessel 1. All of the products are collected in the

Table 7. Optimal Initial Feed Distribution and Product Locations for Different Feed Compositions (Figures are in mol)

Feed Composition, mol %		(25, 25, 25, 25)	(70, 10, 10, 10)	(10, 70, 10, 10)	(10, 10, 70, 10)	(10, 10, 10, 70)
Reflux drum	Feed	0.6	0.6	0.6	41.9	0.6
	Product	24.2	70.2	9.8	5.2	9.3
Vessel 1	Feed	0.6	89.2	8.1	29.7	3.8
	Product	24.4	8.4	68.9	9.9	0.2
Vessel 2	Feed	91.0	3.2	70.2	0.6	34.0
	Product	24.4	8.8	9.0	64.5	0.2
Reboiler	Feed	4.2	1.0	17.6	24.2	58.0
	Product	23.6	8.9	8.6	7.8	68.2
Accumulator 1	Product	—	—	—	5.0	—
Accumulator 2	Product	—	—	—	—	8.9
Accumulator 3	Product	—	—	—	4.0	9.4
Accumulator 4	Product	—	0.3	—	—	—

vessels, except for the *n*-butanol-rich product, a small quantity of which is withdrawn into an accumulator.

For an ethanol-rich feed, most of the feed is placed in vessel 2 with the rest being placed mainly in vessel 1 and the reboiler. All the products are collected in the vessels themselves rather than being withdrawn into accumulators.

When the feed is rich in *n*-propanol, the feed is distributed mainly among the reflux drum, vessel 1, and the reboiler. About half of the methanol-rich product and a small amount of the propanol-rich product is withdrawn into accumulators 1 and 3, respectively, with the rest remaining in the reflux drum and vessel 2, respectively. The other two vessels are operated under total reflux, so that the products are collected in the respective vessels.

For the case where the feed is rich in *n*-butanol, just over half of the total quantity of feed is placed in the reboiler with the rest being placed mainly in vessel 2. The two intermediate products are withdrawn into accumulators 2 and 3, whereas the top and bottom products are collected in the reflux drum and reboiler, respectively.

For the equimolar feed, it was found that Policy 2b, where the feed is placed in the reboiler and the column is operated under total reflux, is both close to optimal (that is, yielding an objective function value that is only marginally worse than that of Policy 3c, where both the feed and product locations are optimized) and practical due to its simplicity. However, the advantage to be gained by optimizing the feed and product locations may be significant in other cases. To illustrate this point, we also performed optimizations using Policy 2b for each of the four mixtures considered in this section. We found that Policy 3c achieves reductions of 47%, 32%, 11%, and 5% in the mean rate of energy consumption over the simpler Policy 2b for feeds rich in methanol, ethanol, *n*-propanol, and *n*-butanol, respectively. Hence, the optimal product withdrawal policy where the feed location is optimized may be justified in some cases depending on the separation to be performed.

For the case where the feed is rich in *n*-propanol, we have also determined the optimal product withdrawal policy with the feed charged entirely to the reboiler (Policy 3b). For this policy, most of the three lightest products are withdrawn into accumulators, whereas the heaviest product is collected in the reboiler (see Table 8). In contrast, for Policy 3c where the initial feed distribution is optimized, only some of the methanol-rich and *n*-propanol-rich products are withdrawn into accumulators. Hence, the optimal product location is influenced by the initial distribution of the feed. Moreover, if we compare the two cases where the feed is charged entirely to the reboiler (Policies 2b and 3b), we notice that an 8% reduction in the mean rate of energy consumption is achieved by optimizing the product location over total reflux operation. However, if we consider the cases where the product locations are optimized (Policies 3b and 3c), only a 3% reduction in the mean rate of energy consumption is achieved by optimizing the initial feed distribution over the case where the feed is charged entirely to the reboiler. Hence, optimizing the product location contributes more to energy savings than optimizing the initial feed distribution in this case. Conversely, optimizing the initial feed distribution proved to be more significant for the cases involving the equimolar feed.

It conclusion, it is clear from the results shown in Table 7 that the feed composition has a major effect of the optimal feed and product locations. There certainly appear to be cases for which distributing the initial feed charge among the vessels and/or withdrawing products to external accumulators offers significant advantages over simpler policies. However, there seems to be no general relationship that can be ex-

Table 8. Different Policies for *n*-Propanol-Rich Feed

Policy		2b	3b	3c
Reflux drum	Feed, mol	0.6	0.6	41.9
	Product, mol	10.0	0.1	5.2
Vessel 1	Feed, mol	0.6	0.6	29.7
	Product, mol	9.1	0.1	9.9
Vessel 2	Feed, mol	0.6	0.6	0.6
	Product, mol	68.8	0.1	64.5
Reboiler	Feed, mol	94.6	94.6	24.2
	Product, mol	8.8	7.8	7.8
Accumulator 1	Product, mol	—	10.0	5.0
Accumulator 2	Product, mol	—	8.9	—
Accumulator 3	Product, mol	—	68.6	4.0
Accumulator 4	Product, mol	—	—	—
Mean Rate of Energy Consumption, W		1,513	1,386	1,342

pressed in terms of a set of simple rules for deducing the optimal policy to be used for a given mixture.

Conclusions

This article has considered the derivation of optimal operating policies for multivessel batch distillation columns using a formal dynamic optimization approach. The optimization objective was to minimize the rate of energy consumption required for producing products of specified purities and mean throughput rates.

Various operating policies differing in the degrees of freedom utilized have been considered. The results have shown that exploiting more degrees of freedom can lead to significant energy savings. For the equimolar quaternary mixture considered, optimizing the initial feed distribution was found to reduce energy consumption by as much as 17%. For all modes of operation, charging the feed to the reboiler was found to be better than distributing the feed according to its composition among the vessels. For this particular separation, product withdrawal from the reflux drum and intermediate vessels to separate accumulator tanks was not found to offer significant advantage over the simpler total reflux operation.

A feedback control strategy recently proposed by Wittgens et al. (1996) was also examined using dynamic optimization to determine optimal settings for the controller gains and set points. However, our results seem to indicate that there is a tradeoff between the quality of temperature control achieved and the primary objective of minimum energy consumption. On the other hand, it should be emphasized that the main benefit of feedback control strategies over open-loop ones is the robustness of the former in the face of uncertainties such as those introduced by variability in the feed composition.

The selection of the optimal feed distribution and product location was found to be strongly dependent on the feed composition. It has also been shown that product withdrawal can be significantly better than total reflux operation for some separations. However, it appears to be difficult to formulate general guidelines as to the most appropriate policy for any particular problem. It may, therefore, be best for rigorous optimization to be carried out for each problem at hand.

A central characteristic of our approach has been the use of detailed dynamic models of the column and its ancillary equipment that remove many of the assumptions present in earlier work in this area. The results presented in this article, as well as the recent work of Tomazi (1997), provide substantial evidence that the use of simplified models can lead to solutions which are far from optimal, and which often violate one or more important constraints that they are supposed to satisfy. This is not entirely surprising since an examination of various assumptions typically employed by simplified models reveals that several of them are not actually supported by the results of more detailed models.

It is worth emphasizing that, in the past, simplified models have played a useful role in exposing general trends in relatively simple column configurations. However, as we begin to consider more complex columns with many more interacting degrees of freedom but also constraints, a more detailed characterization of the physics of the problem appears to be necessary. In particular, it is important to be able to determine the influence of the various control manipulations on the purities of the various products. Without fulfilling this condition, it is difficult to justify the use of sophisticated mathematical tools such as dynamic optimization as these often simply determine ‘‘optimal’’ solutions that actually violate key purity constraints. Fortunately, the process modeling tools and mathematical solution techniques that are currently available permit the direct application of dynamic optimization to detailed batch distillation models.

The batch distillation column model used in this work does not take into account entrainment and weeping effects. Consequently, some of the control profiles obtained may lead to operational problems, especially when the reboiler heat load is set at very low values, or when very large step changes are imposed on it. Additional constraints may be imposed on the solution in order to avoid such difficulties. This is a straightforward task within the overall framework presented in this article.

Finally, it should be stressed that all solutions presented in this article have been obtained using local optimization techniques. Although the detailed models used indicate that all these solutions do satisfy the key purity and throughput constraints, no guarantee can be offered that even better (globally optimal) solutions do not exist.

Notation

A = cross-sectional area of tray, m^2
A_{holes} = total area of holes on a tray, m^2
D_i = product withdrawal flow rate from vessel i, mol/s
F_W = wall correction factor for flow over weir
h = specific enthalpy, J/mol
h^{vap} = specific latent heat of vaporization, J/mol
h_{weir} = weir height, m
h'_{weir} = height of liquid above weir, m
H = liquid level, m
$H_{D,i}$ = holdup in vessel i, mol
l_{weir} = weir length, m
N_C = number of components
P = pressure, Pa
q = volumetric flow rate over weir, m^3/s
Q = rate of heat transfer, W
s = vapor velocity, m/s
T_i = temperature on middle tray in column section i, K
U = internal energy, J
v = volume, m^3
V = vapor flow rate, mol/s
x = liquid composition, mol/mol
$x_{D,i}$ = mole fraction of main component in vessel i, mol/mol
y = vapor composition, mol/mol
$\bar{y}$ = incipient vapor composition, mol/mol
β = aeration factor
β_i = bias of controller i, mol/s
Γ = dry pressure drop coefficient
Π_i = production rate of product i, mol/s
ρ = molar density, mol/m^3
$\bar{\rho}$ = mass density, kg/m^3
ϕ = fugacity coefficient

Subscripts

A = accumulator
C = condenser
R = reboiler
RD = reflux drum
j = component
k = tray

Superscripts

in = inlet stream
L = liquid
out = outlet stream
V = vapor

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Appendix A: Effect of Model Simplifications

Most of the earlier studies on the optimization of batch distillation columns employed relatively simple models. Here, we investigate the effect of model simplifications on the optimal solutions obtained. Models of varying degree of detail are considered.

Different models considered

Three different models are considered. For ease of reference, we call them *simplified*, *semi-detailed* and *detailed*, respectively. The main characteristics of each model are listed below.

(1) *Simplified Model:*

- negligible liquid holdup on the trays
- constant molar overflow
- constant vapor flow rate throughout the column
- negligible vapor holdup

(2) *Semi-Detailed Model:*

- constant liquid holdup on the trays
- algebraic energy balances
- constant vapor flow rate into the condenser
- negligible vapor holdup

(3) *Detailed Model:*

- tray hydraulics (modeled via the modified Francis weir formula)
- dynamic energy balances
- vapor flow rates given by pressure drop relationships
- variable liquid and vapor holdup

A description of the detailed model is given in Appendix B. The others are derived from it by deleting various terms or entire equations.

Problem definition

The three models described in the previous section are used to determine the optimal reflux ratio policy for the separation of an equimolar mixture of methanol, ethanol, *n*-propanol, and *n*-butanol into four main cuts. An off-cut is allowed to be withdrawn between each main cut; however, the recycling of these off-cuts is not considered. A regular column with 30 trays is used for the separation. The column dimensions are the same as those given in Table 1.

The batch size is taken to be 100 mol. For the detailed model, the initial holdup on each tray is taken to be 0.12 mol. In the case of the semi-detailed model, the holdups on each tray and in the reflux drum are kept constant at 0.2 mol and 1.2 mol, respectively; these are the average holdups over the batch time obtained from a dynamic simulation of the detailed model. The vapor flow rates in the detailed model were calculated from pressure drop relationships; a constant reboiler heat duty of 3 kW was used. The vapor flow rate throughout the column for the simplified model and the vapor flow rate into the condenser for the semi-detailed model were kept constant at 4.7 mol/min; again, this was the average vapor flow rate obtained from a dynamic simulation of the detailed model.

The objective function used in the optimization was the minimum processing time per batch (t_f). For each of the four products, the minimum purity specification is taken to be 98 mol % and the minimum quantity to be collected is set at 23 mol.

The reflux ratio $R(t)$ is manipulated by the optimization and is allowed to vary within the bounds

$$R(t) \in [0.5, 1.0] \qquad \text{(A1)}$$

Piecewise constant control is used. The time horizon is divided into seven control intervals of variable duration. During the first interval, the duration of which is fixed at 5 min, the column is operated under total reflux $[R(t) = 1]$. Each subsequent control interval corresponds to a different cut (main or off-cut).

Results

The "optimal" processing times determined using each of the three models, as well as the corresponding computational times (on an IBM RISC System/6000 43P-140 computer), are given in Table A1. The computational times for the semi-detailed model and the detailed model, respectively, were 32% and 58% more than that for the simplified model. Overall, the increased modeling detail has a significant, but far from prohibitive, effect on the computational burden. We note that the processing times for the simplified and semi-detailed models, respectively, are 8% lower and 6% higher than that for the detailed model. More seriously, when the optimal reflux ratio profiles for the simplified and semi-detailed models are implemented in the detailed model, it is found that most of the product specifications are not achieved, as shown in Table A2.

As can be seen from Figure A1, the optimal reflux ratio profiles for the three models differ significantly. The differences can be attributed to the various assumptions which are made in each model. For instance, the reflux drum and tray holdups which are assumed to be negligible in the simplified model and constant at 1.2 mol and 0.2 mol, respectively, in the semi-detailed model, actually vary throughout the operation in the case of the detailed model, as shown in Figure A2. Mujtaba and Macchietto (1991) and Tomazi (1997) have demonstrated the effect of holdup on distillate composition and, hence, on the optimal reflux ratio. In particular, a higher holdup results in a low distillate composition profile and this encourages the use of a high reflux ratio. In the simplified model, negligible holdup was assumed and this contributes to the generally lower reflux ratios and, hence, shorter processing time. In contrast, the reflux drum and tray holdups (1.2 mol and 0.2 mol, respectively) in the semi-detailed model are slightly higher than the average holdups (1.11 mol and 0.18 mol, respectively) in the detailed model. Again, this contributes to the generally higher reflux ratios and longer processing time obtained with the semi-detailed model. It is important, therefore, to characterize as accurately as possible the variation of liquid holdup in the reflux drum and on the trays. In practice, this requires the introduction of a realistic relationship between liquid holdup and liquid flow rate, such as the Francis weir formula (see Eq. B15, Appendix B).

Table A1. Optimal Processing Times Determined by Different Column Models and Corresponding Computational Times

Model	Minimum Processing Time (min)	Computational Time (CPU h)
Simplified	249	24.1
Semidetailed	285	31.8
Detailed	270	38.0

Table A2. Product Characteristics for Different Policies Predicted Using Detailed Model Simulations

Model Used for "Optimal" Solution	Product	1	2	3	4
Simplified	Purity, mol %	98.8	97.4*	94.4*	92.9*
	Amount, mol	23.6	21.8*	20.6*	26.5
Semidetailed	Purity, mol %	98.9	98.5	96.9*	88.6*
	Amount, mol	24.0	21.8*	20.6*	27.9
Detailed	Purity, mol %	99.2	98.0	98.0	98.0
	Amount, mol	23.6	23.0	23.0	23.5

*Constraint violation.

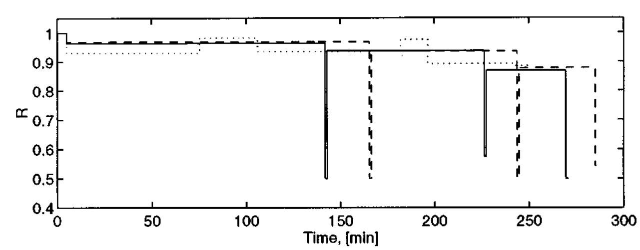

Figure A1. Optimal reflux ratio profile for different models.

(···· Simplified; – – – semi-detailed; —— detailed.)

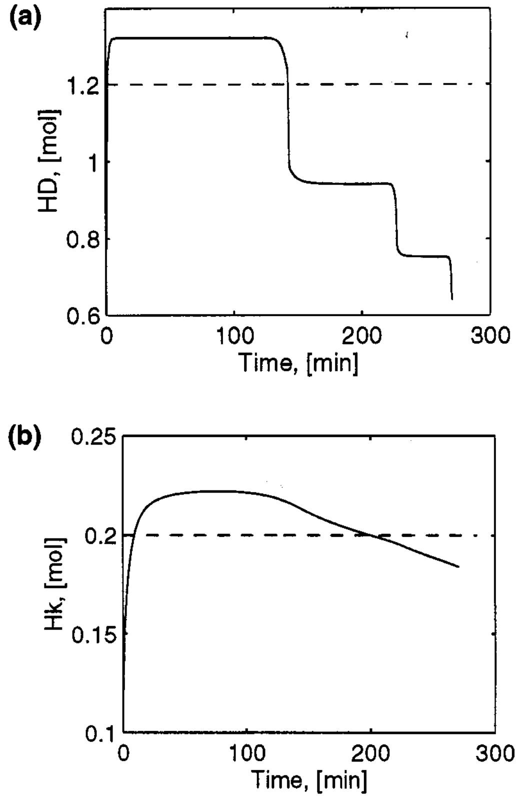

Figure A2. Variation of holdup.

(a) Reflux drum; (b) tray (– – – semi-detailed model; —— detailed model).

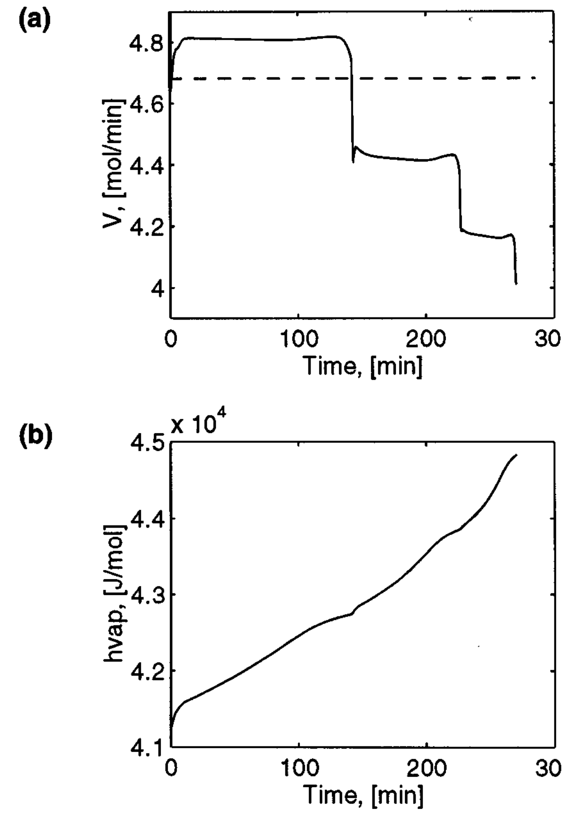

Figure A3. Variation of vapor flow rate into the condenser, and specific latent heat of vaporization of the mixture in the reboiler.

(a) Vapor flowrate into the condenser; (b) specific latent heat of vaporization of the mixture in the reboiler (– – – simplified and semi-detailed models; —— detailed model).

Also, both the vapor flow rate throughout the column in the simplified model, and the vapor flow rate entering the condenser in the semi-detailed model were assumed to be constant. However, the vapor flow rate for the detailed model actually changes appreciably during the operation, as shown in Figure A3a. The boilup rate from the reboiler is a function of the specific latent heat of vaporization of the mixture in the reboiler which is, in turn, a function of the composition. As the mixture becomes heavier due to the removal of the light components, the specific latent heat of vaporization increases (as shown in Figure A3b), thereby causing the vapor flow rate to drop for a constant rate of heating. The energy balance equation also affects the vapor flow dynamics. In view of the significant variations in both tray holdup and tray temperature over the batch, the use of a steady-state energy balance does not appear to be justified. We, therefore, prefer to use a dynamic energy balance coupled with a pressure drop relation (see Eqs. B9 and B19 in Appendix B).

Finally vapor holdup was assumed to be negligible in the simplified and semi-detailed models. The average vapor holdup in the reflux drum, on the trays, and in the reboiler for the detailed model was found to be 0.001, 0.004, and 0.232 mol, respectively. Thus, the vapor holdup is not as substantial as liquid holdup and is not expected to have had a significant influence on the optimal solution obtained. On the other hand, at higher pressures, vapor holdup will be larger and worth taking into account. In any case, it should be noted that very little additional complexity is introduced by taking account of vapor holdup in the detailed model (see Eqs. B10, B11, and B12 in Appendix B).

Appendix B: Mathematical Model for Batch Distillation

Here, the mathematical model used for each of the units comprising a batch distillation column is presented. These include the reboiler, tray, condenser, reflux drum/vessel, and accumulator. The main features of the model used were summarized in the section entitled "Mathematical Model." First, the modeling assumptions made are stated here; then, the model for each of the units is given.

Modeling assumptions

In order to contain the size of model that can be solved within reasonable computational time and to avoid having to obtain values for quantities which cannot readily be estimated, certain assumptions have been retained at present. These are listed below:

(1) *No entrainment and weeping effects.* Correlations, such as those proposed by Kister and Haas (1988), for estimating these effects may be inserted in the model if necessary.

(2) *Total condensation with no-subcooling in the condenser.* This assumption can be removed by modeling the rate of heat transfer in the condenser, and partitioning the latter into distinct condensation and subcooling zones.

(3) *No downcomer dynamics.* In principle, this assumption is easily removed by including the downcomer mass and energy balances. Of course, this will result in increased problem size.

(4) *Adiabatic operation.* The main difficulty in removing this assumption is in determining the effective coefficient for heat transfer from the external surface of the column to the environment.

(5) *Phase equilibrium.* This assumption may be removed in one of two possible ways:

- correlations for the Murphree stage efficiency may be introduced (Zuiderweg, 1982); however, this method is particularly inaccurate for multicomponent systems.
- a nonequilibrium model may be used instead, where the finite mass- and heat-transfer rates are calculated (Kooijman and Taylor, 1995); this would require extensive data and a significantly larger model.

(6) *Perfect mixing.* The degree of mixing between the vapor bubbles and the liquid on the tray may affect the mass transfer and, therefore, the achieved separation. However, removing the assumption of perfect mixing requires a significant increase in the model complexity and size (Wesselingh, 1997).

Model equations

Reboiler.

Molar balance on component j

$$\frac{dM_{j,R}}{dt} = x_{j,R}^{\text{in}} L_R^{\text{in}} - y_{j,R}^{\text{out}} V_R, \qquad j = 1, \ldots, N_C \tag{B1}$$

Energy balance

$$\frac{dU_R}{dt} = h_R^{\text{in}} L_R^{\text{in}} - h_R^V V_R + Q_R \tag{B2}$$

Liquid and vapor contributions to component holdup

$$M_{j,R} = x_{j,R} M_R^L + y_{j,R}^{\text{out}} M_R^V, \qquad j = 1, \ldots, N_C \tag{B3}$$

Liquid and vapor contributions to total internal energy

$$U_R = h_R^L M_R^L + h_R^V M_R^V - P_R{}^{V}{}_{R} \tag{B4}$$

Total volume constraint

$$\upsilon_R = \frac{M_R^L}{\rho_R^L} + \frac{M_R^V}{\rho_R^V} \tag{B5}$$

Equilibrium relationship

$$\phi_{j,R}^V y_{j,R}^{\text{out}} = \phi_{j,R}^L x_{j,R}, \qquad j = 1, \ldots, N_C \tag{B6}$$

Normalization equations

$$\sum_{j=1}^{N_C} x_{j,R} = \sum_{j=1}^{N_C} y_{j,R}^{\text{out}} = 1 \tag{B7}$$

Tray. Here, we consider the kth tray, counting downwards from the top of the column.

Molar balance on component j

$$\frac{dM_{j,k}}{dt} = x_{j,k-1} L_{k-1} + y_{j,k+1} V_{k+1} - x_{j,k} L_k - y_{j,k} V_k, \qquad j = 1, \ldots, N_C \tag{B8}$$

Energy balance

$$\frac{dU_k}{dt} = h_{k-1}^L L_{k-1} + h_{k+1}^V V_{k+1} - h_k^L L_k - h_k^V V_k \tag{B9}$$

Liquid and vapor contributions to component holdup

$$M_{j,k} = x_{j,k} M_k^L + y_{j,k} M_k^V, \qquad j = 1, \ldots, N_C \tag{B10}$$

Liquid and vapor contributions to total internal energy

$$U_k = h_k^L M_k^L + h_k^V M_k^V - P_k{}^{V}{}_{k} \tag{B11}$$

Total volume constraint

$$\upsilon_k = \frac{M_k^L}{\rho_k^L} + \frac{M_k^V}{\rho_k^V} \tag{B12}$$

Equilibrium relationship

$$\phi_{j,k}^V y_{j,k} = \phi_{j,k}^L x_{j,k}, \qquad j = 1, \ldots, N_C \tag{B13}$$

Normalization equations

$$\sum_{j=1}^{N_C} x_{j,k} = \sum_{j=1}^{N_C} y_{j,k} = 1 \tag{B14}$$

Liquid-flow relationship (Francis weir formula, Perry and Green, 1984)

$$h'_{\text{weir},k} = 0.68175 F_W \left(\frac{q_k}{l_{\text{weir}}} \right)^{2/3} \tag{B15}$$

Definition of volumetric liquid flow rate

$$q_k = \frac{L_k}{\rho_k^L} \tag{B16}$$

Liquid holdup on the tray

$$M_k^L = \rho_k^L A_k \left(h_{\text{weir}} + h'_{\text{weir},k} \right) \tag{B17}$$

Relationship between vapor flow rate and vapor velocity

$$V_{k+1} = \rho_{k+1}^V A_{\text{holes}} s_{k+1} \tag{B18}$$

Pressure drop across the tray

$$P_{k+1} - P_k = \Gamma s_{k+1}^2 \bar{\rho}_{k+1}^V + \beta \bar{\rho}_k^L g(h_{\text{weir}} + h'_{\text{weir},k}) \tag{B19}$$

Total Condenser.

Molar balance on component j

$$y_{j,C} = x_{j,C}, \qquad j = 1, \ldots, N_C \tag{B20}$$

Molar balance

$$V_C^{\text{in}} = L_C^{\text{out}} \tag{B21}$$

Vapor flow relation

$$V_C^{\text{in}} = C_v \sqrt{(P_1 - P_C)} \tag{B22}$$

The value of the coefficient for vapor flow between the top tray and condenser C_v (mol/s·Pa$^{1/2}$) was taken as 0.04 mol/s·Pa$^{1/2}$.

Energy balance

$$h_1^V V_C^{\text{in}} = h_C^L L_C^{\text{out}} + Q_C \tag{B23}$$

Equilibrium relationship

$$\phi_{j,C}^V \bar{y}_{j,C} = \phi_{j,C}^L x_{j,C}, \qquad j = 1, \ldots, N_C \tag{B24}$$

Normalization equation

$$\sum_{j=1}^{N_C} \bar{y}_{j,C} = 1 \tag{B25}$$

Vessel/Reflux Drum.

Molar balance on component j

$$\frac{dM_j}{dt} = x_j^{\text{in}} L^{\text{in}} - x_j^{\text{out}} L^{\text{out}}, \qquad j = 1, \ldots, N_C \tag{B26}$$

Energy balance

$$\frac{dU}{dt} = h^{\text{in}} L^{\text{in}} - h^{\text{out}} L^{\text{out}} \tag{B27}$$

Liquid and vapor contributions to component holdup

$$M_j = x_j^{\text{out}} M^L + y_j M^V, \qquad j = 1, \ldots, N_C \tag{B28}$$

Liquid and vapor contributions to internal energy

$$U = h^L M^L + h^V M^V - PV \tag{B29}$$

Total volume constraint

$$v = \frac{M^L}{\rho^L} + \frac{M^V}{\rho^V} \tag{B30}$$

Height of liquid in the vessel

$$H = \frac{M^L}{\rho^L A} \tag{B31}$$

Equilibrium relationship

$$\phi_j^V y_j = \phi_j^L x_j^{\text{out}}, \qquad j = 1, \ldots, N_C \tag{B32}$$

Normalization equations

$$\sum_{j=1}^{N_C} x_j^{\text{out}} = \sum_{j=1}^{N_C} y_j = 1 \tag{B33}$$

In the case of the multivessel column, L^{out} is treated as a control variable; however, for the regular column, we assume that the reflux flow rate L^{out} is driven by gravity and is given by a relationship of the form

$$L^{\text{out}} = C_l \rho^L (P + \bar{\rho}^L g H - P_1) \tag{B34}$$

The value of the coefficient for liquid flow out of the vessel C_l (m^3/s·Pa) was taken as 1.5×10^{-13} m^3/s·Pa.

Accumulator. The model of the accumulator is similar to that of the reflux drum/vessel, except that there is no liquid flow leaving the unit, that is, $L^{\text{out}} = 0$.

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