

An Efficient Design Method for Binary, Azeotropic, Batch Distillation Columns

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Using rigorous models to simulate batch distillation involves the solution of many stiff differential equations. The dimensionality of the problem also increases with an increase in the number of plates and components, which imposes limitations on the use of rigorous models. Furthermore, problems in design, optimization, synthesis, and control involve iterative procedures, and considerable computational efforts would be required to solve them rigorously. As an alternative, the short-cut method (Diwekar and Madhavan, 1991a) provided reasonably accurate solutions without much computational effort. This method in the present form, however, cannot be applied to azeotropic systems because of the impassable barrier of the azeotropic point and needs to be modified. The azeotropic approximation proposed by Anderson and Doherty (1984) was used to modify the short-cut method for binary azeotropic systems. The modified algorithm presented here was tested extensively with a number of binary azeotropic systems for both constant reflux and variable reflux modes of operation, and the results were shown to compare well with the rigorous models. The method requires significantly less computational efforts and is very useful in analyzing feasible region of operation.

Introduction

Sudden increase in the production of high-value-added, low-volume specialty chemicals and biochemicals in recent years has generated a renewed interest in batch processing technology. However, most of the research in batch processing is focused on developing methodologies for synthesizing and sizing equipment networks, where the performance characteristics of the process units are assumed to be given. This is because the time-dependent nature of batch processes and units makes the models to be complex. Another major issue in batch process design or synthesis is operational flexibility, which gives rise to a large number of alternative evaluations. Use of rigorous models becomes prohibitive because of large computational efforts involved and extensive data and memory requirements. Since batch distillation is an important unit operation in the batch processing industry and is most widely used, a fast and accurate model for this unit operation would be very useful. The short-cut method developed in an earlier work (Diwekar and Madhavan, 1991a) provides such an alternative for non-

azeotropic systems. The method is a modification of the short-cut method for continuous columns and is based on the assumption of constant relative volatility throughout the column, updated over each time step.

The essential drawback of azeotropic distillation is that the relative volatility of the mixture is not constant. This is due to the impassable barriers of the azeotropic points. Anderson and Doherty (1984) presented a model for handling this problem for binary, azeotropic, continuous distillation columns. In their work they divided the vapor-liquid equilibrium curve for the binary azeotropic systems into two portions separated by the azeotropic points. A coordinate transformation enabled them to represent the curves by constant relative volatility equations.

In this article the short-cut method (Diwekar and Madhavan, 1991a) was modified for azeotropic distillation using this approach. The results of several binary systems used to test the new algorithm compare very well for both constant reflux and variable reflux operating modes. This approach results in significant computational savings and can be extended to multicomponent systems.

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Apart from the computational efficiency, lower memory requirement, and the noninteger algebraic-equation-oriented form, the short-cut method offers the advantage of analyzing a problem for global properties. This property is extremely handy in identifying proper bounds for feasible operation in design, optimization or control of batch distillation columns and is of great practical significance. This article also briefly discusses the qualitative properties of the short-cut method.

Previous Work

Literature on batch distillation goes back to 1902, when Lord Rayleigh produced the first mathematical treatment of batch distillation. The early models up to 1950 were limited to graphical integration applied to batch distillation using the Rayleigh equation for binary systems. Smoker and Rose (1940) presented an analysis of the constant reflux case, while Bogart (1937) analyzed the case of constant overhead composition. These models were developed based on McCabe-Thiele's graphical method for continuous distillation and were applicable to binary systems.

The empirical short-cut models for binary systems preceded the graphical models. These models had limited applicability and were based on the concept of "Standard Separation Curve" (Rose and Welshans, 1940) and the idea of a "Pole Height" proposed by Zuiderweg (1953).

A rigorous approach to batch distillation involves the solution of the material and energy balance equations in the differential form. Huckaba and Danly (1960) proposed the first model involving material and energy balance calculations for binary systems. Meadows (1963) provided the first comprehensive model for multicomponent systems. In 1968, Distefano analyzed the applicability of different numerical integration techniques for solving the rigorous dynamic model for multicomponent batch distillation in detail and reported the cause of numerical instabilities. The Distefano model is the most cited rigorous model for batch distillation, and most of the studies and simulation models, which have appeared since 1968 (Boston et al., 1981), are based on this model.

Development of simplified models for batch distillation and their use in optimization and optimal control seem to be the recent trend in batch distillation studies (Farhat et al., 1990; Chiotti and Iribarren, 1991; Al-Tuwaim and Luyben, 1991). These models are either confined to binary (Chiotti and Iribarren, 1991) or ternary systems (Al-Tuwaim and Luyben, 1991), or use the semirigorous plate to plate calculation method for the simulation (Farhat et al., 1990).

The BATCH-DIST, a comprehensive package for multicomponent, multifraction batch distillation columns (Diwekar and Madhavan, 1991b), provides a hierarchy of models for simulation, ranging from the most simplified model to the most rigorous model based on Distefano's analysis. The most simplified model in the BATCH-DIST uses the short-cut method (Diwekar and Madhavan, 1991a) and is applicable to nearly ideal systems and columns where the holdup effect is insignificant. Their work also analyzes the effect of holdup and characterizes it with a single parameter τ , which is the time constant of the column. The next in the hierarchy is the modified short-cut method model which can be used when the holdup effect is significant (large τ). The model rigor, data requirement, and computational effort increase as one climbs higher in the hierarchy.

The short-cut-method-based models in the BATCH-DIST or other simplified models in the literature are applicable only to nonazeotropic systems.

Azeotropic distillation is an important and widely used separation technique. Despite their importance, azeotropic distillation techniques remain poorly understood from the standpoint of design. This is because of the complex thermodynamic behavior of the system. Theoretical studies on azeotropic distillation have mainly centered around methods for predicting the vapor-liquid equilibrium data from liquid solution models and their application to distillation design (VanDongen and Doherty, 1985a). However, only during the past decade has there been a concerted effort to understand the nature of the composition region boundaries. Most of the literature in this area pertain to Doherty and his research group. Their work focuses on the use of simple distillation curve maps in analyzing the continuous column design and synthesis. Although, the series of articles by Doherty and his group seek to develop design and synthesis tools for azeotropic continuous distillation, their work on azeotropic batch distillation is restricted to columns with large number of plates, operating at high reflux ratios (VanDongen and Doherty, 1985b; Barnot et al., 1990).

Characteristics of the Short-Cut Method

The two well-known methods of operating batch columns are the variable reflux and the constant reflux. The optimal control policy is essentially a trade-off between the two methods and is based on the concept of optimizing a given performance index.

The rigorous modeling of batch distillation operation involves solutions for a large number of stiff differential equations. The computational intensity and memory requirement of the problem increase with an increase in the number of plates and components. For an n -component system and a column with N number of plates, the simulation of constant reflux or variable reflux operation requires simultaneous solution of $n \times N$ material balance differential equations, N energy balance differential or difference equations, and the vapor-liquid equilibrium calculation associated with each differential material balance equation. The vapor-liquid equilibria calculations are iterative in nature and become more complicated for azeotropic systems. The computational complexity associated with rigorous models does not allow us to derive global properties, such as feasible regions of operation which are critical for optimization and optimal control problems. Even if such information were available, the computational cost of optimization or optimal control with rigorous models is prohibitive.

One approach to deal with the problems associated with rigorous models is to develop simplified models. These simplified models are abstractions of rigorous models, and their accuracy depends on the simplifying assumptions. The process of abstraction can be viewed as the trade-off between simplicity and fidelity. The usefulness of abstracted models depends on the ease with which it can be analyzed for global behaviors, without compromising fidelity. Moreover, abstracted models are expected to be computationally simpler to analyze.

The schematic of the process of abstraction for the batch distillation column model is shown in Figure 1. The abstract

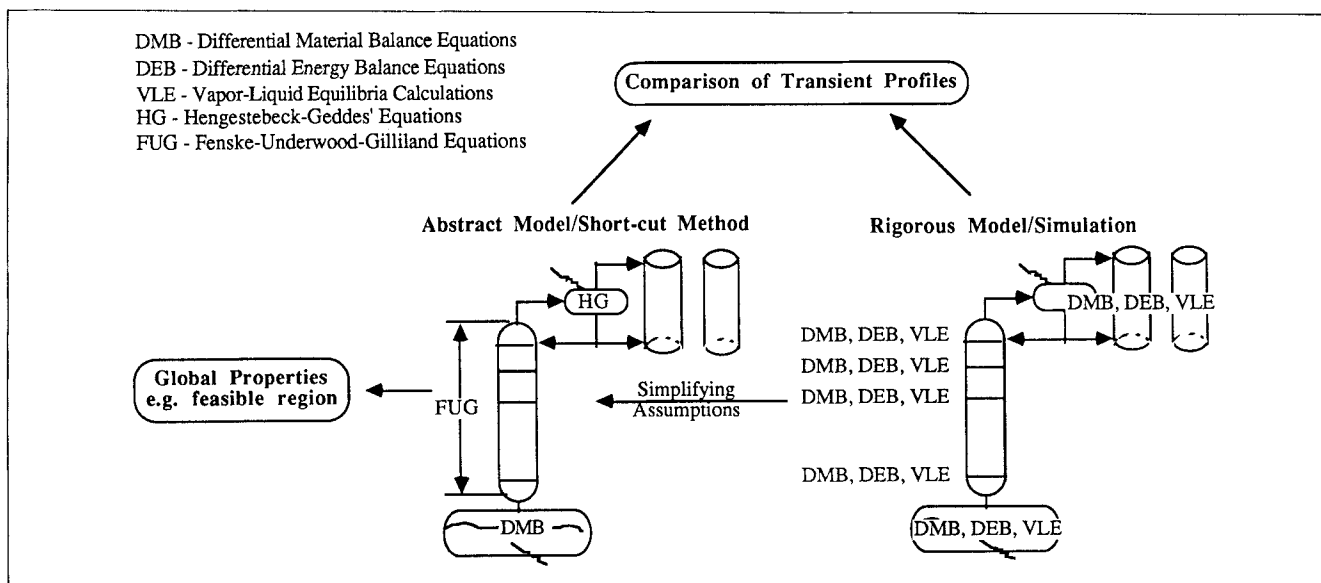


Figure 1. Abstraction process.

model is derived from the rigorous model by making a set of simplifying assumptions. The short-cut method is used on the abstract model to simulate the distillation process. These results are compared with those from simulating the rigorous model. The abstract model can also be used to derive feasible regions of operation.

The short-cut method for batch distillation is based on the assumption that the batch distillation column can be considered as a continuous distillation column with changing feed at any time instant (Figure 2). In other words, the bottom product of one time step forms the feed for the next time step. This is equivalent to having bottom plate as the feed plate and the feed at its boiling point. Since continuous distillation theory

is well-developed and tested, the short-cut method [Fenske-Underwood-Gilliland (FUG) method] for continuous distillation could be modified for batch distillation. The batch time is implicit and can be calculated if the vaporization rate V of the reboiler is known. Because of the algebraic-equation-oriented form of the short-cut method, it is possible to adapt the model for optimal control calculations very easily. Although the development of the short-cut method for different operating modes was discussed in detail elsewhere (Diwekar and Madhavan, 1991a; Diwekar et al., 1987), a unified short-cut method approach to the different operating modes is presented in this work. Table 1 shows the time implicit model for the three operating modes. It can be seen that the same equations with different form can be used to simulate such different operating modes as variable reflux, constant reflux, and optimal reflux.

At each time instant, there is a change in the still composition of the key component, resulting in changes in the still composition of all other components calculated by the differential material balance equations (Table 1). Hengestebeck-Geddes' equation relates the distillate compositions to the new still compositions in terms of the constant C_1 . The constant C_1 in Hengestebeck-Geddes' equation is equivalent to the minimum number of plates, N_{\min} in Fenske's equation. At this stage, the variable reflux operating mode has C_1 and R , constant reflux has $x_D^{(i)}$ and C_1 , and optimal reflux has $x_D^{(i)}$, C_1 , and R as unknowns. Summation of distillate compositions can be used to obtain C_1 for variable reflux and $x_D^{(i)}$ for both constant reflux and optimal reflux operation, and the FUG equations to obtain R for variable reflux and C_1 for both constant reflux and optimal reflux operations. The optimal reflux model of operation has an additional unknown, R , which can be calculated using the concept of optimizing the Hamiltonian, formulated using Pontryagin's Maximum Principle.

Global properties are represented by interval values of parameters and variables rather than numeric values. With the short-cut method it is possible to extract global properties of the batch column in terms of feasible region or inequality

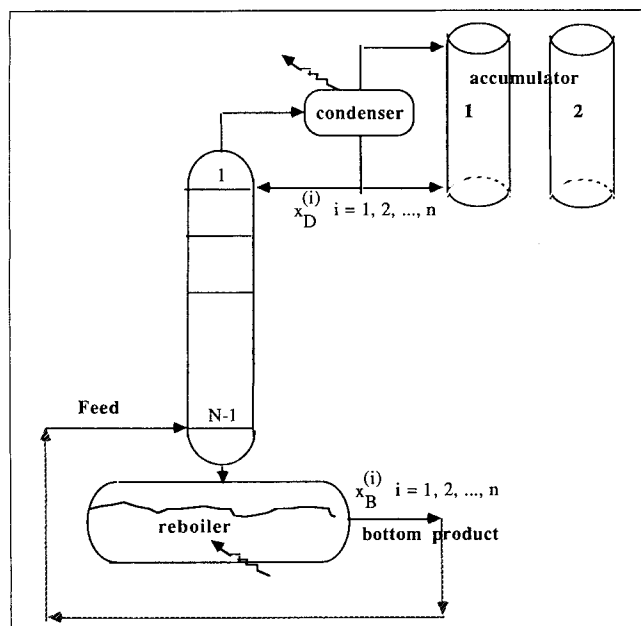


Figure 2. Short-cut method.

Table 1. Time Implicit Model Equations for the Short-Cut Method

Variable Reflux	Constant Reflux	Optimal Reflux
<u>Differential Material Balance Equation</u>		
$x_{B_{new}}^{(i)} = x_{B_{old}}^{(i)} + \frac{\Delta x_B^{(i)} (x_D^{(i)} - x_B^{(i)})_{old}}{(x_D^{(i)} - x_B^{(i)})_{old}}, i = 1, 2, \dots, n$		
<u>Hengestebeck-Geddes' Equation</u>		
$x_D^{(i)} = \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_D^{(1)}}{x_B^{(1)}} x_B^{(i)}, i = 2, 3, \dots, n$		
	Unknowns	
R, C_1	$C_1, x_D^{(1)}$	$R, C_1, x_D^{(1)}$
<u>Summation of Fractions</u>		
	$\sum_{i=1}^n x_D^{(i)} = 1$	
C_1 estimation	$x_D^{(1)}$ estimation	
$\sum_{i=1}^n \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_D^{(1)}}{x_B^{(1)}} x_B^{(i)} = 1$	$x_D^{(1)} = \frac{1}{\sum_{i=1}^n \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_B^{(i)}}{x_B^{(1)}}}$	
<u>Fenske Equation</u>		
	$N_{min} \approx C_1$	
<u>Underwood Equations</u>		
$\sum_{i=1}^n \frac{\alpha_i x_B^{(i)}}{\alpha_i - \phi} = 0; R_{min} u + 1 = \sum_{i=1}^n \frac{\alpha_i x_D^{(i)}}{\alpha_i - \phi}$		
<u>Gilliland Correlation</u>		
R estimation	C_1 estimation	
$R = F(N, N_{min}, R_{min} u)$	$R_{min} g = F(N, N_{min}, R)$	
	$\frac{R_{min} g}{R} - \frac{R_{min} u}{R} = 0$	
	R estimation	
	$R = F(\text{Minimum Hamiltonian})$	

constraints, especially for design variables like number of plates N and reflux ratio R . This is a very useful property of the short-cut method.

Identification of bounds on design parameters is very handy in design, optimization, or control problems. Table 2 shows the feasible region of operation or the identified bounds obtained using the short-cut method. We have used these bounds in our earlier work on optimization and optimal control of short-cut method (Diwekar et al., 1987, 1989; Logsdon et al., 1990).

Modification of the Short-Cut Method for Azeotropic Systems

The short-cut method is based on the assumption of constant relative volatility throughout the column, updated at each time instant, and uses the following relationship for obtaining vapor-liquid equilibria:

$$y_i = \frac{\alpha_i x_i}{\sum_{k=1}^n \alpha_k x_k}; \quad i = 1, 2, \dots, n$$

In the case of azeotropic systems, however, this relation is no longer valid because of the azeotropic points (where the relative volatility becomes unity). Anderson and Doherty (1984) trans-

Table 2. Feasible Region for Multicomponent Batch Distillation Columns

Variable Reflux	Constant Reflux	Optimal Reflux
<u>Final Still Composition</u>		
$0 \leq x_{B_{final}}^{(1)} \leq x_D^{(1)}$		
<u>Distillate Composition</u>		
$x_B^{(1)} \leq x_D^{(1)} \leq 1$		
<u>Reflux Ratio</u>		
$1 \leq \frac{R_{initial}}{R_{min}} \leq \frac{R_{max}^*}{R_{min}}$	$1 \leq \frac{R}{R_{MIN}} \leq \infty^{**}$	
<u>Number of Plates</u>		
$N_{min f} \leq N^*$	$N_{min} \leq N^{**}$	

*The lower limit of $R_{initial}$ is governed by the value of R_{min} calculated at the initial conditions ($x_B^{(1)} = x_B^{(i)}$), and the upper limit by N_{min} , that is, $N_{min f}$ calculated at the final value of $x_B^{(1)}$ (since N_{min} increases and $x_B^{(1)}$ decreases). To obtain the value of R_{max} , N_{min} is calculated at the final value of $x_B^{(1)}$, and this is taken as the limiting value of N . The value of R corresponding to this N calculated at the initial conditions of $x_B^{(1)}$ is taken as R_{max} .

** R_{MIN} is the value of R (not R_{min}) required to obtain the initial distillate composition of the component 1 equal to the specified average composition for the specific value of N . N_{min} is Fenske's value of minimum number of plates calculated at the initial condition assuming the distillate composition of component 1 equal to the average distillate composition.

formed the variables for binary vapor-liquid equilibria calculations and used it for continuous distillation columns. In their approach, the equilibrium curve is split into two parts as shown in Figure 3. The variables are transformed using the azeotropic compositions, as seen in Figure 3.

In Figure 3, the bottom curve represents

$$x'_1 = \frac{x_1}{x_1^{az}}, \quad x'_2 = 1 - x'_1$$

$$y'_1 = \frac{y_1}{y_1^{az}}, \quad y'_2 = 1 - y'_1$$

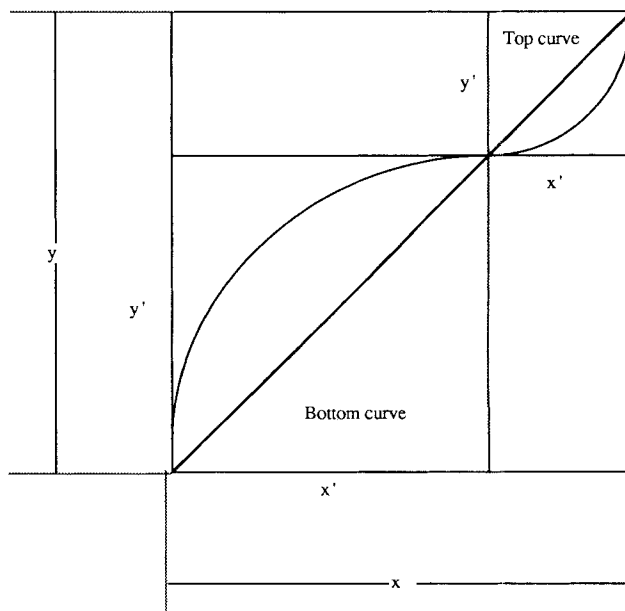


Figure 3. Variable transformation for binary azeotropic systems.

Table 3. Modified Equations for Azeotropic Systems

Variable Reflux	Constant Reflux	Optimal Reflux
Differential Material Balance Equation		
$x_{B_{new}}^{(i)} = x_{B_{old}}^{(i)} + \frac{\Delta x_B^{(i)}(x_D^{(i)} - x_B^{(i)})_{old}}{(x_D^{(i)} - x_B^{(i)})_{old}}, i = 1, 2, \dots, n$		
Variable Transformation		
Top Column		
$x_B^{(i)} = \frac{x_B^{(i)}}{x_i^{az}}, i = 1, 2, \dots, n-1$		
Bottom Column		
$x_B^{(i)} = \frac{x_B^{(i)} - x_i^{az}}{1 - x_i^{az}}, i = 1, 2, \dots, n-1$		
Hengestebeck-Geddes' Equation		
$x_D^{(i)} = \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_D^{(1)}}{x_B^{(1)}} x_B^{(i)}, i = 2, 3, \dots, n$		
Unknowns		
R, C_1	$C_1, x_D^{(1)}$	$R, C_1, x_D^{(1)}$
Summation of Fractions		
$\Sigma_{i=1}^n x_D^{(i)} = 1$	$\Sigma_{i=1}^n x_D^{(i)} = 1$	$\Sigma_{i=1}^n x_D^{(i)} = 1$
C_1 estimation	$x_D^{(1)}$ estimation	
$\Sigma_{i=1}^n \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_D^{(1)}}{x_B^{(1)}} x_B^{(i)} = 1$		
$x_D^{(1)} = \frac{1}{\Sigma_{i=1}^n \left(\frac{\alpha_i}{\alpha_1} \right)^{C_1} \frac{x_B^{(i)}}{x_B^{(1)}}}$		
Fenske Equation		
$N_{min} \approx C_1$		
Underwood Equations		
$\Sigma_{i=1}^n \frac{\alpha_i x_B^{(i)}}{\alpha_i - \phi} = 0; R_{min} + 1 = \Sigma_{i=1}^n \frac{\alpha_i x_D^{(i)}}{\alpha_i - \phi}$		
Gilliland Correlation		
R estimation	C_1 estimation	
$R = F(N, N_{min}, R_{min})$	$R_{min} = F(N, N_{min}, R)$	
	$\frac{R_{min,g}}{R} - \frac{R_{min,u}}{R} = 0$	
	R estimation	
	$R = F(\text{Minimum Hamiltonian})$	
Variable Retransformation		
Top Column		
$x_B^{(i)} = x_B^{(i)} x_i^{az}, i = 1, 2, \dots, n-1$		
$x_D^{(i)} = x_D^{(i)} x_i^{az}, i = 1, 2, \dots, n-1$		
Bottom Column		
$x_B^{(i)} = x_B^{(i)} - x_i^{az}(1 - x_B^{(i)}), i = 1, 2, \dots, n$		
$x_D^{(i)} = x_D^{(i)} - x_i^{az}(1 - x_D^{(i)}), i = 1, 2, \dots, n$		

and the top curve is given by

$$x_1' = \frac{x_1 - x_1^{az}}{1 - x_1^{az}}; \quad x_2' = 1 - x_1'$$

$$y_1' = \frac{y_1 - y_1^{az}}{1 - y_1^{az}}; \quad y_2' = 1 - y_1'$$

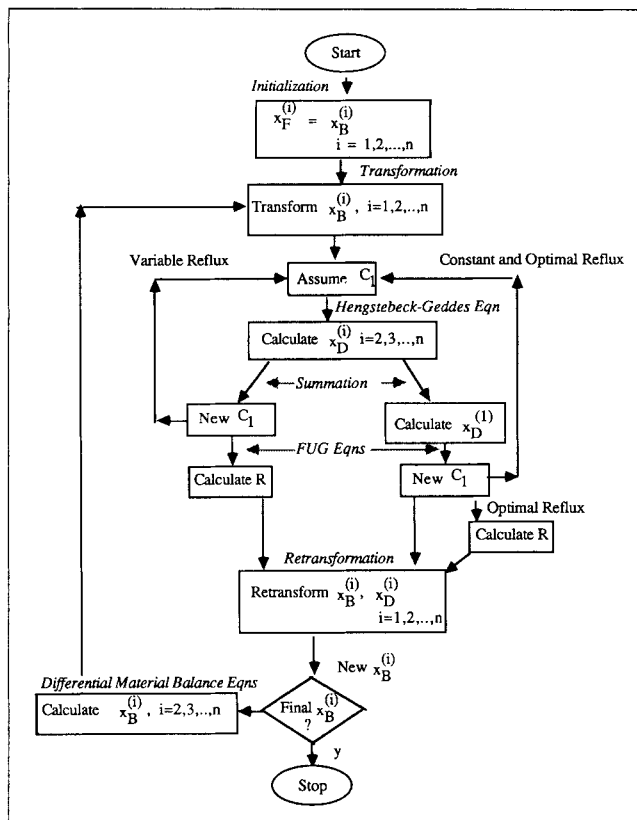


Figure 4. Flowchart for extended short-cut method for binary azeotropic systems.

and the equilibrium relationship for the binary system in terms of the transformed variables is represented by

$$y_1' = \frac{\alpha_1 x_1'}{1 + (\alpha_1 - 1)x_1'}$$

This approach for vapor-liquid equilibria calculations is used to extend the short-cut method model of Diwekar and Madhavan (1991a) for nonazeotropic systems to azeotropic systems. As in the case of short-cut method model for nonazeotropic system (Table 1), a unified model is presented for the three operating modes of variable reflux, constant reflux and optimal reflux. The model equations are given in Table 3 and the flowchart for the unified algorithm is presented in Figure 4.

Table 4. Input Data for the Test Problems

Constant Reflux						
Problem	System	N	R	V	Feed	Comp.
1	Ethyl Acetate/Ethanol	8	2.73	12	0.2, 0.8	
2	Chloroform/Methanol	8	5.0	20	0.2, 0.8	
3	Methanol/Ethyl Acetate	8	2.73	12	0.4, 0.6	
4	Toluene/n-Butanol	8	2.73	12	0.2, 0.8	
5	Tetrahydrofuran/Methanol	8	2.73	12	0.2, 0.8	
6	Water/Pyridine	8	2.73	12	0.2, 0.8	
Variable Reflux						
7	Methanol/Ethyl Acetate	5	0.65	12	0.2, 0.8	
8	Tetrahydrofuran/Methanol	5	0.6	12	0.2, 0.8	

After obtaining the still compositions of all the components as before (differential material balance equations, Table 3), the still compositions, $x_B^{(i)}$, $i = 1, 2, \dots, n$ are transformed in terms of azeotropic points using the above-mentioned transformation formulae (Anderson and Doherty, 1984). For the variable reflux operation, the distillate composition of the key component, $x_D^{(1)}$ is also transformed. These transformed variables are then used in the calculation of distillate compositions of all the components, $x_D^{(i)}$, $i = 1, 2, \dots, n$ Hengestebeck-Geddes' constant C_1 , and the reflux ratio R (for variable reflux mode). The initial variable transformation essentially results in transformed distillate compositions, which needs to be re-transformed to obtain the actual values.

Although the short-cut method for azeotropic systems, presented in Table 3, applies to binary systems, it can also be extended to ternary systems if one has the knowledge of distillation boundaries. VanDongen and Doherty (1985a) have shown that the topology of the simple distillation residue curve maps can be used to identify distillation regions separated by the separatrices. They use the simple distillation differential model (autonomous differential equations) to generate a simple distillation residue curve map. Kalagnanam (1991) in his thesis on "Qualitative Analysis of System Behavior" integrates the use of symbolic and numeric methods to identify equilibrium points, separatrices, etc. from the structure of the autonomous differential equations based on the partial tests from the qualitative theory of differential equations. Such a qualitative analysis approach along with the extended short-cut method for azeotropic systems can provide a very good tool for the analysis of azeotropic batch distillation columns and will be investigated in our future work.

Results and Discussion

The extension of short-cut algorithm proposed here for the azeotropic systems is tested extensively using a large number of binary azeotropic systems for both the constant reflux and the variable reflux operating mode. In each case, the results obtained using the new algorithm are compared with the results from the rigorous model analysis using BATCHSIM (Simulation Sciences Inc.) for constant reflux operating mode and the BATCH-DIST for the variable reflux operating mode. The liquid-phase nonidealities are represented using the NRTL equation.

It has been found that the short-cut model compares very well with the rigorous models and offers significant computational savings over the rigorous models (for some cases, the short-cut model is found to be 200–300 times faster than the rigorous models) without compromising much on the accuracy.

A few of the cases studied are reported here. Table 4 gives the input data and operating conditions. Figure 5 presents the comparison of the composition profiles (distillate and still compositions vs. time) obtained using the short-cut model with the rigorous BATCHSIM model for the constant reflux mode. [The equilibration time or the initial total reflux time is not included in the calculation. However, correlations for calculation of equilibrium time as a function of relative volatility, holdup, and reflux are available in the literature (Ellerbe, 1979) and could be easily incorporated in the short-cut model.] In Figure 5, the circles represent simulation results from rigorous model and the lines show results from the short-cut method model. The top curve in each case represents the transient

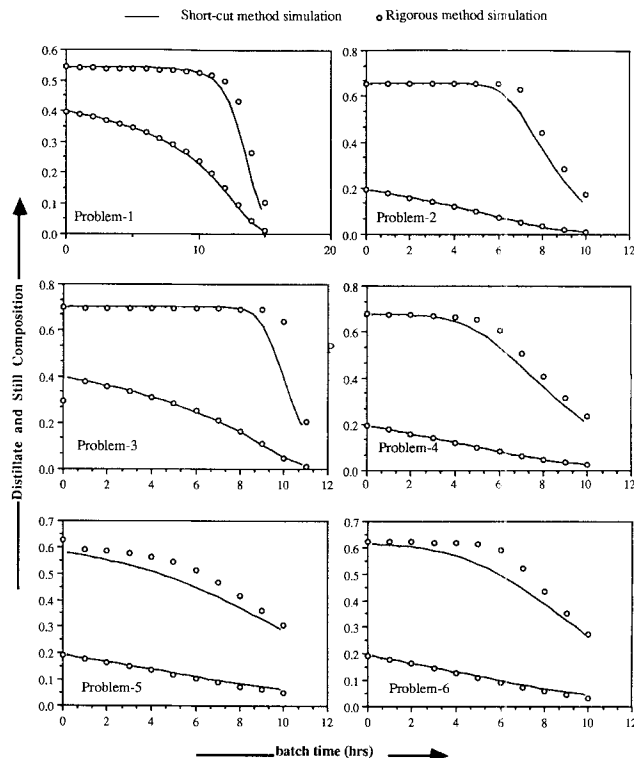


Figure 5. Constant reflux operating mode: short-cut vs. rigorous method.

distillate composition, and the bottom curve shows the transient still composition. As can be seen from Figure 5, the agreement between the short-cut model and the rigorous model for the constant reflux operating mode is excellent (most parts of the curves agree within 1%).

Figure 6 compares the short-cut model, reflux and still composition profiles, and the profiles predicted by the semirigorous (rigorous with zero holdup) model of BATCH-DIST for the variable reflux operating mode. Once again, the circles rep-

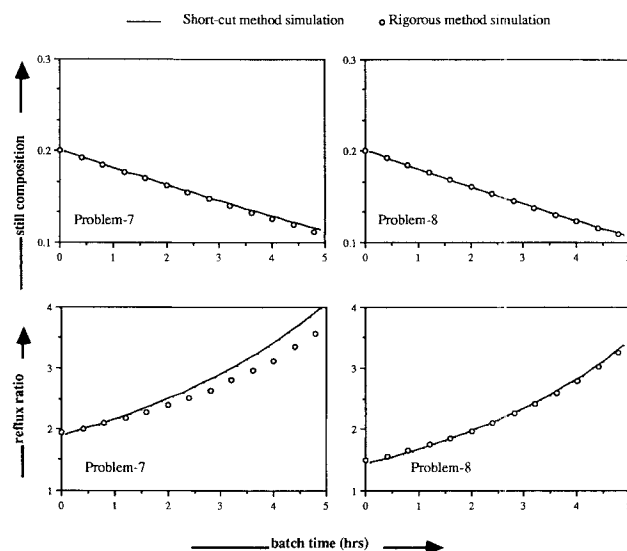


Figure 6. Variable reflux operating mode: short-cut vs. rigorous method.

resent the results from the rigorous model and the lines are the results from the short-cut method model. Even for the variable reflux operating mode, the short-cut method model seemed to provide a very good approximation to the rigorous model.

The binary azeotropic systems considered here are mostly ideal azeotropic systems. For nonideal azeotropic systems like ethanol-water, this extended short-cut method can still be used as an efficient preliminary design tool.

The holdup effect, characterized by the parameter τ = holdup/(reflux \times distillate rate) appeared to be insignificant for the cases presented (Figures 5 and 6). For large τ , the initial composition profiles predicted by the short-cut method departs from the rigorous model, as the reflux is changed from total reflux to a very small value (Diwekar and Madhavan, 1991b). However, the new algorithm proposed here can be easily incorporated in the modified short-cut method model (Diwekar and Madhavan, 1991a) and can be used when holdup effect is predominant. In the modified short-cut model for holdup consideration, the distillation column is partitioned into two portions. The top portion with the condenser and the first plate is used to simulate the holdup effect, and the bottom portion is represented by the short-cut method model. The short-cut method so modified seems to be very effective in including the dynamic effects of holdup (Diwekar and Madhavan, 1991a).

Although the extended short-cut method model is proposed for all the three operating modes of batch distillation, due to the complexity of calculations and the large computational requirement, it was not possible to use the rigorous model for the optimal reflux mode of operation. Nevertheless, the favorable comparison of the constant reflux and the variable reflux operating mode guarantees the success for the optimal reflux mode of operation, because optimal reflux mode is essentially a trade-off between the two operating modes.

Conclusions

The computationally efficient short-cut method was extended for handling azeotropic systems. The qualitative nature of the short-cut method was analyzed. A large number of examples were used for model testing and validation. The problems were rigorously simulated using BATCHSIM and BATCH-DIST, and the results of the proposed algorithm were compared to these results. Comparisons were made by plotting the instantaneous distillate and still compositions for the constant reflux mode, and the instantaneous still composition and reflux ratio for the variable reflux mode. The short-cut method results and rigorous simulation results were shown to agree very well.

The extended short-cut method model represents a higher level of abstraction of the rigorous model. It provides a very powerful and computationally fast tool for the analysis and will be extended to ternary azeotropic system in the future.

Acknowledgment

Many thanks to Jayant Kalagnanam, Department of Engineering and Public Policy, for introducing the author to the subject of qualitative analysis and reasoning and for offering numerous useful discussions related to this article. The author is also grateful to Kevin Simonsen, Manager, Applications and Support, Simulation Sciences Inc., for providing the software and encouraging the research efforts.

Notation

C_1	= constant in Hengstebeck-Geddes' equation
n	= number of components
N	= number of plates
N_{\min}	= minimum number of plates
$N_{\min f}$	= minimum number of plates at the terminal condition
R	= reflux ratio
R_{\max}	= maximum initial reflux ratio for variable reflux mode
R_{\min}	= minimum reflux ratio
$R_{\min g}$	= minimum reflux ratio given by Gilliland correlation
$R_{\min u}$	= minimum reflux ratio given by Underwood equations
V	= vapor boil-up rate, mol/h
x_i	= liquid composition for component i
x'_i	= transformed liquid composition for component i
x_i^{az}	= azeotropic liquid composition for component i
$x_B^{(i)}$	= still composition for component i
$x_D^{(i)}$	= distillate composition for component i
$x_F^{(i)}$	= feed composition for component i
y_i	= vapor composition for component i
y'_i	= transformed vapor composition for component i
y_i^{az}	= azeotropic vapor composition for component i

Greek letters

α_i	= relative volatility of component i
ϕ_i	= Underwood's constant

Literature Cited

- Al-Tuwaim, M. S., and W. L. Luyben, "Multicomponent Batch Distillation: 3. Short-Cut Design of Batch Distillation Columns," *I&EC Res.*, **30**, 507 (1991).
- Anderson, N. J., and M. F. Doherty, "An Approximate Model for Binary Azeotropic Distillation Design," *Chem. Eng. Sci.*, **39**, 11 (1984).
- Bernot, C., M. F. Doherty, and M. F. Malone, "Patterns of Composition Changes in Multicomponent Batch Distillation," *Chem. Eng. Sci.*, **45**, 1207 (1990).
- Bogart, B. J. P., "The Design Equipment for Fractional Batch Distillation," *AIChE Trans.*, **33**, 139 (1937).
- Boston, J. F., H. I. Britt, S. Jirpophpan, and V. B. Shah, *An Advanced System for Simulation of Batch Distillation Operation*, FO-CAPD, Vol. 2, R. S. H. Mah and W. Seider, eds., Engineering Foundations, New York, 203 (1981).
- Chiotti, O. J., and O. A. Iribarren, "Simplified Models for Binary Batch Distillation," *Comput. Chem. Eng.*, **15**, 1 (1991).
- Distefano, G. P., "Mathematical Modeling and Numerical Integration of Multicomponent Batch Distillation Equations," *AIChE J.*, **14**, 946 (1968).
- Diwekar, U. M., and K. P. Madhavan, "Multicomponent Batch Distillation Column Design," *I&EC Res.*, **30**, 713 (1991a).
- Diwekar, U. M., and K. P. Madhavan, BATCH-DIST: "Comprehensive Package for Simulation, Design, Optimization, and Optimal Control of Multicomponent, Multifraction Batch Distillation Columns," *Comput. and Chem. Eng.*, in press (1991b).
- Diwekar, U. M., K. P. Madhavan, and R. E. Swaney, "Optimization of Multicomponent Batch Distillation Column," *I&EC Res.*, **28**, 1011 (1989).
- Diwekar, U. M., K. P. Madhavan, and R. K. Malik, "Optimal Reflux Rate Policy Determination for Multicomponent Batch Distillation Columns," *Comput. Chem. Eng.*, **11**, 629 (1987).
- Ellerbe, R. W., *Batch Distillation: Handbook of Separation Techniques for Chemical Engineers*, p. 1, P. A. Schweitzer, ed., McGraw Hill, New York (1979).
- Farhat, S., M. Czernicki, L. Pibouleau, and S. Domench, "Optimization of Multiple-Fraction Batch Distillation by Nonlinear Programming," *AIChE J.*, **36**, 1349 (1990).
- Huckaba, C. E., and D. E. Danly, "Calculation Procedures for Binary Batch Rectification," *AIChE J.*, **6**, 335 (1960).
- Kalagnanam, J. R., "Qualitative Analysis of System Behavior," PhD Thesis, Dept. of Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, (1991).
- Logsdon, J. S., U. M. Diwekar, and L. T. Biegler, "On the Simul-

- taneous Optimal Design and Operation of Batch Distillation Columns," *Chem. Eng. Des. Res.*, **68**, 434 (1990).
- Lord Rayleigh, "On the Distillation of Binary Mixtures," *Phil. Mag.*, **4**, 527 (1902).
- Meadows, E. L., "Multicomponent Batch-Distillation Calculations on a Digital Computer," *Chem. Eng. Prog. Symp. Ser. Proc. Syst. Eng.*, **59**(46), 48 (1963).
- Smoker, E. H., and A. Rose, "Graphical Determination of Batch Distillation Curves for Binary Mixtures," *Trans. AIChE*, **36**, 285 (1940).
- Rose, A., and L. M. Welshans, "Sharpness of Separation in Batch Fractionation," *Ind. Eng. Chem.*, **32**, 668 (1940).
- VanDongen, D. B., and M. F. Doherty, "Design and Synthesis of Homogeneous Azeotropic Distillations: I. Problem Formulation for a Single Column," *Ind. Eng. Chem. Fundam.*, **24** 454 (1985a).
- VanDongen, D. B., and M. F. Doherty, "On the Dynamics of Distillation Processes: VI. Batch Distillation," *Chem. Eng. Sci.*, **40**, 2087 (1985b).
- Zuiderweg, F. J., "Absatzweise Destillation: Einflub der Bodenzahl, des Rucklaufverhaltnisses und des Holdup auf die Trennscharfe," *Chemie Ingenieur Technik*, **25**, 297 (1953).

Manuscript received July 2, 1991, and revision received Aug. 27, 1991.