Bifurcation Analysis in Heterogeneous Azeotropic Distillation

Parameterization with respect to the aqueous reflux ratio has been carried out for the steady-state analysis of a tower to dehydrate sec-butanol using disecondary butyl ether as the entrainer. With an equilibrium-stage model, singularities are observed as the second liquid phase is introduced on the trays and regions of steady-state multiplicity are demonstrated. Furthermore, a critical aqueous reflux ratio, in the vicinity of which two liquid phases are introduced on most of the trays, varies only slightly with the number of trays in the tower. The model predicts that the product purities and recoveries remain high using one-third of the trays in the industrial SBA-II tower. Bifurcation analysis for a single stage does not explain these observations, and the effect of recycle is suspected.

Soemantri Widagdo
Chemistry & Chemical Engineering
Department
Stevens Institute of Technology
Hoboken, NJ 07030

Warren D. Seider
Department of Chemical Engineering
University of Pennsylvania
Philadelphia, PA 19104

Donald H. Sebastian
Chemistry & Chemical Engineering
Department
Stevens Institute of Technology
Hoboken, NJ 07030

Introduction

Many entrainers used in azeotropic distillation systems are partially miscible with one of the species in the mixture. This selection of entrainer gives a better column performance, but results in an overhead stream that condenses into two liquid phases. Normally, the heavy phase is decanted (withdrawn as product), and the lighter entrainer phase is returned to the tower as reflux. Small amounts of the heavy phase, often water, are also added to the reflux; and under certain operating conditions (e.g., at high aqueous reflux ratios), two liquid phases may extend into the rectifying and even the stripping sections (Kovach and Seider, 1987a).

The behavior of a distillation system under these conditions has not been well studied, although potential problems have been suspected for some time. A few experimental studies report that the second liquid phase has a negative effect on the column efficiency (Goodliffe, 1934), while others report no adverse effects (Schoenborn, 1941). In addition, the possibility of unusual tray hydraulics and problems in control has led chemical engineers to overdesign these columns and settle for suboptimal designs.

In this paper, new results are presented for an equilibrium-stage model that simulates an industrial heterogeneous azeotropic distillation tower to dehydrate secondary butyl alcohol (SBA) with disecondary butyl ether (DSBE) as the entrainer. First, a discussion of prior work is presented with emphasis on the computational aspects and the inconclusive experimental results concerning the impact of the second liquid phase on the column efficiency and tray hydraulics.

Prior Work

Experimental and theoretical results

The dehydration of SBA was studied both experimentally and theoretically by Kovach and Seider (1987a), with emphasis on the ARCO SBA-II tower. The SBA-II tower, which is shown schematically in Figure 1, has 40 valve trays and a 1.981-m ID, and is 25.1-m-tall. The tower has a large decanter and a thermosiphon reboiler. In operation, the tower is controlled manually by adjusting the aqueous reflux rate to maintain a temperature difference of 7 K between trays 31 and 36. Under normal operation, the tower has a high recovery of SBA in purities exceeding 98 wt. %. Over a two-week period, during which measurements were recorded twice daily (at 5:00 a.m. and 5:00 p.m.), however, the bottoms product contained approximately 4.5 wt. % water on two occasions, and 9.4 wt. % DSBE was present on one occasion. Unlike many azeotropic towers, the entrainer is present in the feed.

The experimental results show the sensitive coupling of the

Correspondence concerning this paper should be addressed to W. D. Seider.

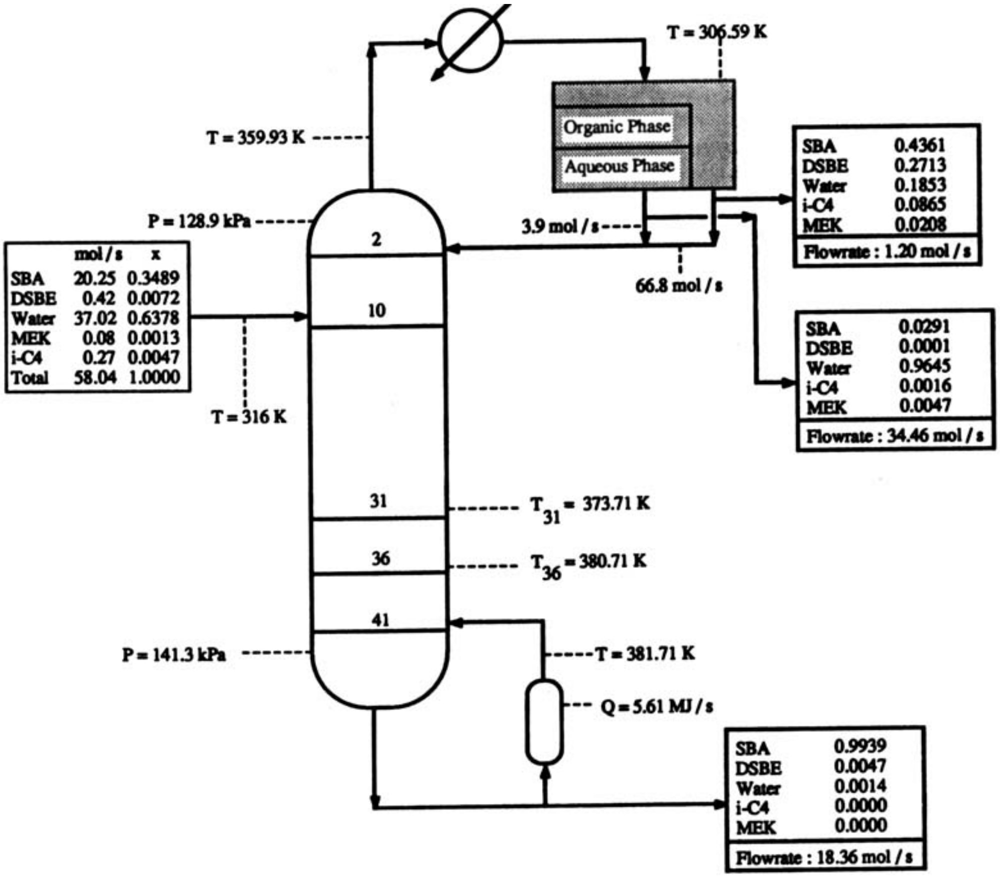

Figure 1. Experimental operating conditions for the ARCO SBA-II Tower.

aqueous reflux ratio and the temperature and composition fronts in the stripping section. This coupling is reproduced in the solution of the mass balance, equilibrium, summation of the molar flow rates, and the heat balance (MESH) equations in the steady state. This solution shows that the temperature and composition fronts correspond to the interface between trays having one and two liquid phases. Furthermore, as illustrated in the profiles of liquid concentration in Figure 2, only small concentration changes occur in much of the stripping section. In this region, the effect of mass transfer and tray hydraulics may cause more gradual changes in the concentrations and temperature as the fronts are approached; yet, the equilibrium-stage model can be expected to track the coupling between the fronts and the aqueous reflux ratio. Indeed, the agreement between the theoretical and experimental results (Kovach and Seider, 1987a) is good, although the movement of the fronts in response to changes in the aqueous reflux ratio may be more sensitive in the theoretical results. Unfortunately, the precision of the experimental measurements is not sufficient to permit a quantitative verification.

Computational aspects

The nonideality of the azeotropic liquid phases produces the steep fronts in the temperature and concentrations, the steepness of which reflect their high sensitivity to small changes in the operating conditions (e.g., reflux ratios) and in the values of the iteration variables. Furthermore, the azeotropic mixtures lead to a narrow region of feasible specifications that permit a realistic solution of the MESH equations (Prokopakis and Seider, 1983).

The extreme sensitivity of the temperature and concentration fronts with respect to changes in the iteration variables is likely to contribute most to the failures of the Newton-based

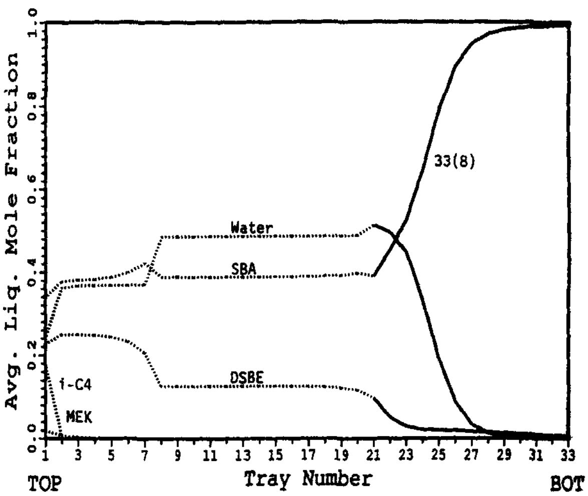

Figure 2. Liquid concentration profiles at an aqueous reflux ratio of 0.1776, computed by solution of the MESH equations.

Two liquid phases exist in the decanter and on trays 2 to 21 (dotted line).

algorithms. As a second liquid phase appears on a tray, the difficulty of solving the MESH equations is further increased. Consequently, improved algorithms are necessary to solve the equations that model these complex distillation systems. Kovach and Seider (1987b) successfully use the homotopy-continuation methods, and Swartz and Stewart (1987) are successful with collocation-based methods.

Experiments on three-phase distillation

Very few experiments have been conducted on three-phase distillation systems. Most of the experiments are performed in packed and tray columns, on a laboratory scale, and concentrate on the effect of the second liquid phase on the column efficiencies (Goodliffe, 1934; Schoenborn, 1941). These studies result in contradictory conclusions. Schoenborn concludes that the presence of the second liquid phase has no detrimental effect on the column efficiency, while Goodliffe observes an adverse effect.

Recently, Herfurth et al. (1987) conducted an experiment on the dehydration of ethanol with cyclohexane as an entrainer, and reported complete temperature and composition profiles. Although not confirmed, several of their liquid-phase compositions appear to be in the binodal region.

Tray hydraulics

Another experimental study was recently completed on the tray hydraulics in three-phase distillation systems. Herron et al. (1988) studied the hydrodynamics and mass transfer in several oil-water mixtures using three sieve trays in a column simulator. The column has a square cross section of 0.0929 m^2 and is 1.27-m-tall. The front panel is made of plexiglass to permit visual observation. The gas-liquid contact takes place on the two bottom trays, and the top tray is used to collect liquid entrained in the vapor phase. Water and mineral spirits are fed separately onto tray 2, and air is introduced at the bottom of the column. The froth height, tray pressure drop, liquid hold-up, downcomer level, and entrainment are measured as a function of the vapor rate, liquid rate, liquid composition, and diameter of the tray perforations. Their study reveals that the hydraulic parameters above can be predicted from two-phase correlations with no or minor modifications; hence, the second liquid phase has no significant effect on the tray hydraulics and efficiency.

Phase equilibrium and mass transfer

Herron et al. (1988) also confirmed, with concentration measurements, that acetone distributes between the two liquid phases at equilibrium. With this observation for the mineral oil-water-air-acetone system on sieve trays, it seems reasonable to expect liquid-liquid equilibrium for the SBA-DSBE-water system on valve trays. Although the resistance to mass transfer is least in the vapor phase, the hydrodynamics of the vapor phase are more complex [mixing is slower, the bubbles are larger than droplets (providing less contact area) and form foams (which entrain liquid), etc.]. Hence, vapor-liquid-liquid equilibrium cannot be assumed and an overall column efficiency is selected. It will be shown that the coupling between the aqueous reflux ratio and the position of the concentration and temperature fronts in the simulation results is insensitive to the choice of this efficiency.

Parametric Study

On the basis of the prior work, both experimental and theoretical, the effects of mass transfer and tray hydraulics are inconclusive. First, the experimental studies of the effect of the second liquid phase on the column efficiency and the tray hydraulics show no clear relationship. Second, the equilibrium-stage model represents the coupling between the aqueous reflux ratio and the temperature and concentration fronts at steady state, but the position of the fronts may be more sensitive to the aqueous reflux ratio than is observed experimentally, as noted previously.

This section presents the results of a parametric study using the equilibrium-stage model, although it may be necessary to model the mass transfer and tray hydraulics to more accurately track this sensitivity. Such models are subject to many assumptions concerning the interface between two and three phases and are difficult to solve. The results presented herein are somewhat unexpected and, although mass transfer and tray hydraulic effects are neglected, help to clarify the unusual steady-state operation of the SBA-II tower. Knowledge of the solution structure (i.e., the position of the temperature and concentration fronts as the reflux ratios vary) helps to locate regions of stable operating conditions and aids in the design of control strategies in the vicinity of stable attractors. Of special concern are regions that contain multiple steady-state solutions that may result in hysteresis behavior. At limit points, an exchange of stability usually occurs: i.e., stable steady states become unstable, and unstable steady states become stable. In these regions, small disturbances can cause the system to oscillate between two steady-state solutions.

Steady-state model

An equilibrium-stage model using the Naphtali-Sandholm (1971) formulation was implemented by Kovach (1986). In his implementation, the MESH equations are extended to model liquid-liquid equilibrium when two liquid phases exist on the trays. The MESH equations are solved using a Newton homotopy, as described by Kovach and Seider (1987b) and implemented in the FORTRAN program, HOMDIS. HOMDIS was used to compute the results reported below.

Results

The parametric studies reported herein begin with 33 *ideal* trays, including a condenser/decanter and a reboiler. This assumes that the 41 tray tower (including the condenser/decanter) has an overall efficiency of 80%. The feed contains secondary butyl alcohol, disecondary butyl ether, water, methyl ethyl ketone and iso-butene, and is fed to tray 8 in the simulation (tray 10 in the SBA-II tower). The feed is subcooled to 316.48 K at 0.122 MPa and comprises two liquid phases. The vapor phase is assumed to be ideal, while the liquid phase is modeled using the UNIQUAC equation (Kovach and Seider, 1988). The interaction coefficients of this equation are in Table 1. Throughout the study, the boilup rate is 5.61 MJ/s, the condenser is subcooled by 1 K, and the organic reflux ratio is 55. The number of trays, the location of the feed tray, and the aqueous reflux ratio are the primary bifurcation parameters.

Most azeotropic distillation towers have been oversized to account for design uncertainty and the difficulties in controlling the purity of the bottoms product. Hence, for these towers, it

Table 1. Parameters for the UNIQUAC Equation

SBA–DSBE–WATER
(Ref., Kovach and Seider, 1988)
Size/shape parameters:

	SBA	DSBE	Water
R	3.9235	6.0909	0.9200
Q	3.6640	5.1680	1.4000
Q′	4.0643	5.7409	1.6741

Interaction coefficients, $a_{ij}/R\{K\}$:

	SBA	DSBE	Water
SBA	0.0000	−97.2021	213.3998
DSBE	209.2880	0.0000	158.6873
Water	52.2446	1974.0559	0.0000

EtOH–Benzene–Water
Size/shape parameters:

	EtOH	Benzene	Water
R	2.1100	3.1900	0.9200
Q	1.9700	2.4000	1.4000
Q′	0.9200	2.4000	1.0000

Interaction coefficients, $a_{ij}/R\{K\}$
(Ref., Bancroft et al., 1942)

	EtOH	Benzene	Water
EtOH	0.00000	−296.73858	−325.31164
Benzene	2247.55531	0.00000	4088.09354
Water	1139.76327	228.78331	0.00000

(Ref., Gmehling and Onken, 1977)

	EtOH	Benzene	Water
EtOH	0.00000	−311.12838	218.61077
Benzene	1690.32170	0.00000	2131.83756
Water	397.54531	851.41553	0.00000

should be possible to reduce the number of trays without significantly reducing the bottoms purity. To test this, the number of trays in the SBA-II tower was decreased until the bottoms purity fell below 99 mol %. The feed tray was adjusted to maintain a constant ratio of the number of trays in the rectifying and stripping sections. It was found, however, that at least five or six trays are necessary in the rectifying section. Furthermore, SBA purities in excess of 98 mol % can be achieved with as few as one-third of the trays currently in use, as shown in Figure 3. The SBA bottoms purity is relatively insensitive to changes in the aqueous reflux ratio below 0.182. For aqueous reflux ratios greater than 0.182, the SBA bottoms purity drops off as the aqueous reflux ratio increases. The recovery of SBA as a function of the aqueous reflux ratio, number of trays, and feed tray location is shown in Figure 4. This figure shows that the recovery of SBA is insensitive to changes in the aqueous reflux ratio (even in excess of 0.182), the number of trays, and the feed locations.

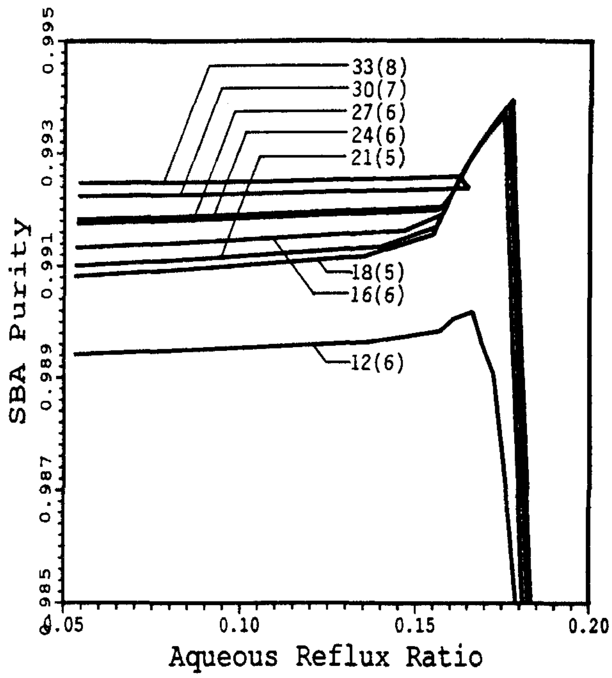

Figure 3. SBA purity as a function of the aqueous reflux ratio.

$N_T(N_F)$ are annotated.

When the movement of the two-liquid-phase front is examined, a critical aqueous reflux ratio appears, which is insensitive to the number of stages, as shown in Figure 5. This sharp variation in trays with two liquid phases is further illustrated in Figure 6. Furthermore, the insensitivity of the critical aqueous reflux ratio to the number of equilibrium stages suggests that the liquid composition that crosses the binodal curve is independent of the number of stages. This is confirmed in Figure 7 which shows that the profiles at constant composition are independent of the number of trays. This occurs only if the temperature at this transition is also independent of the number of trays, as shown in Figure 8. Since the critical aqueous reflux ratio and the liquid composition and temperature at which two liquid phases first appear on the trays do not vary with the number of

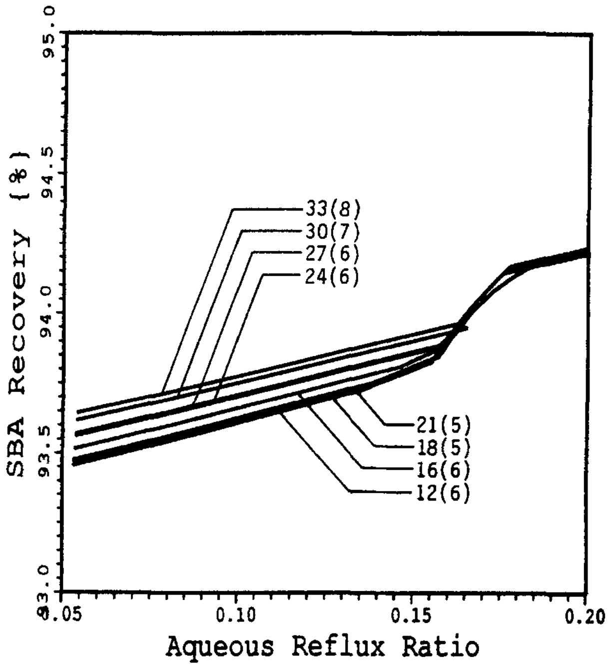

Figure 4. SBA recovery as a function of the aqueous reflux ratio.

$N_T(N_F)$ are annotated.

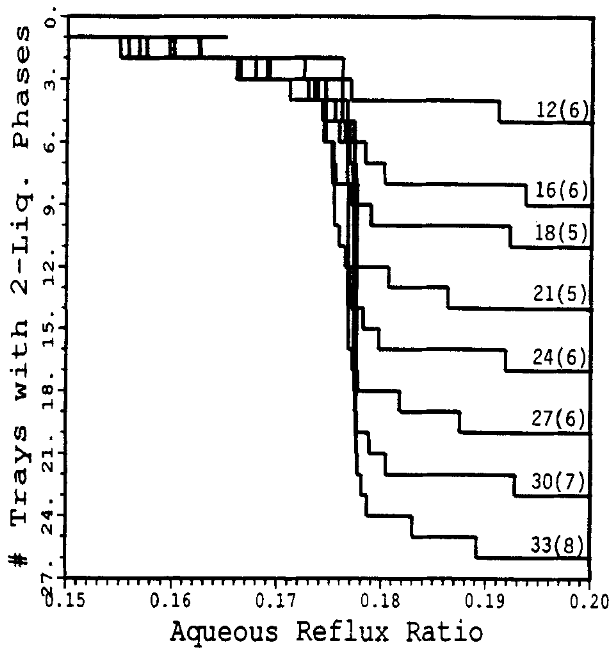

Figure 5. Trays with two liquid phases (above the solid lines) as a function of the aqueous reflux ratio.
$N_T(N_F)$ are annotated.

trays, similarly they are insensitive to the overall column efficiency.

These results clearly show that the bottoms concentration is relatively insensitive in the vicinity of the critical aqueous reflux ratio. Very small changes in the aqueous reflux ratio, on the order of 0.01%, cause the interface between the trays having two and one liquid phase(s) to be displaced by several trays. The associated temperature front is displaced as well, but little change is observed in the bottoms composition. This is confirmed in the experimental observations of Kovach and Seider (1987a); however, the movement of the temperature front was not measured with adequate precision.

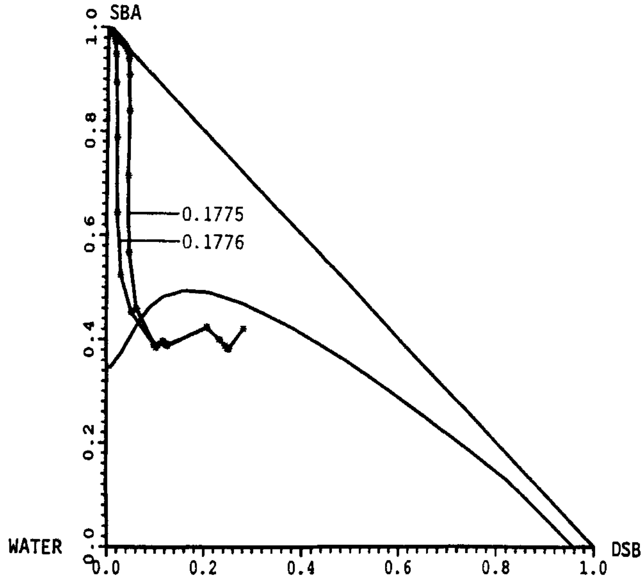

Figure 6. Liquid concentrations for the 33 tray tower with the feed on tray 8 at two aqueous reflux ratios near the critical aqueous reflux ratio.

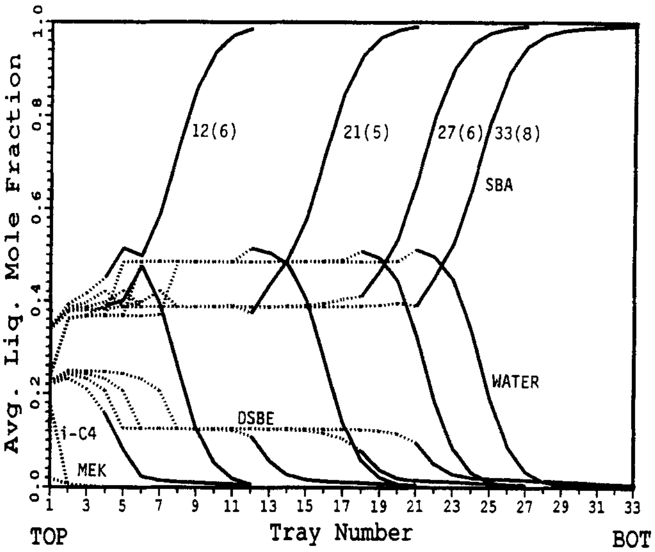

Figure 7. Liquid concentration profiles.
$N_T(N_F)$ are annotated.

Steady-state multiplicity

A sizable region of steady-state multiplicity was found for a 30-tray column with its feed stream located on tray 7, but not for the other configurations examined. This multiplicity occurs over a narrow range of the aqueous reflux ratio between 0.162 and 0.165, well below the critical aqueous reflux ratio (0.1776), and is accompanied by the addition of a second liquid phase on tray 2. The liquid concentration profiles in this region are shown in Figure 9. Note that two liquid phases exist in the decanter (tray 1) and on tray 2 *only* and that the concentrations on the remaining trays are typical of a tower having just one liquid phase on *all* of its trays. The temperature profiles in Figure 10 exhibit Z-shaped folds for trays 3 and lower, where only a single liquid phase is present. On tray 2, where the phase transition occurs,

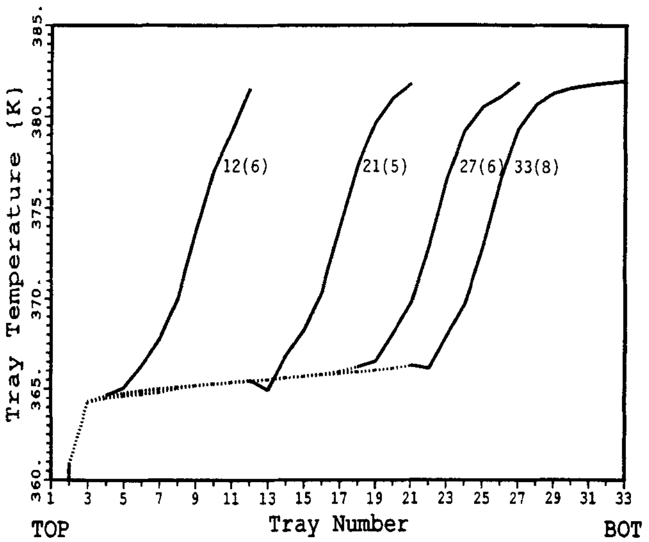

Figure 8. Temperature profiles.
$N_T(N_F)$ are annotated.

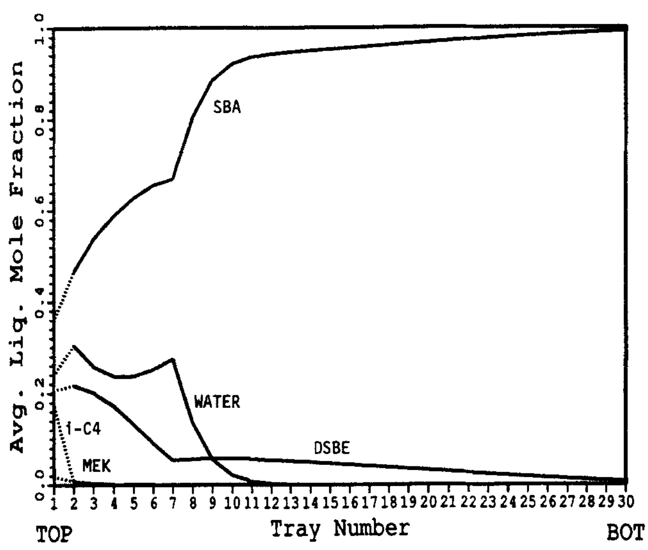

Figure 9. Liquid concentration profiles at an aqueous reflux ratio of 0.162.

Two liquid phases exist in the decanter and on tray 2.

however, a negligible change in temperature is computed. The solid lines denote trays with a single liquid phase, while the dotted lines denote trays with two liquid phases. Figure 10 also shows that the range of the temperature folds diminishes from tray to tray down the tower.

Unfortunately, the temperatures and concentrations were not measured in the rectifying section, although the computed results show a negligible variation in the bottoms composition, which has been confirmed experimentally. Although unconfirmed experimentally, the occurrence of the multiplicity has two important consequences.

1. In the design of control systems, caution should be exercised not to select the temperatures on the trays in the rectifying section as the variables to be measured and controlled. If the computed results are correct, hysteresis would cause oscillation of the aqueous reflux in an attempt to achieve the set-point temperatures.

2. The multiplicity makes it very difficult to obtain convergence when solving the MESH equations with the Newton-Raphson method. In fact, the multiplicity is observed as the second liquid phase is introduced or removed from *all* of the trays. In most cases, however, the aqueous reflux ratio varies in the fifth significant figure. These swings may or may not occur experimentally, yet they introduce the need for special convergence procedures to avoid the singularities at the turning points.

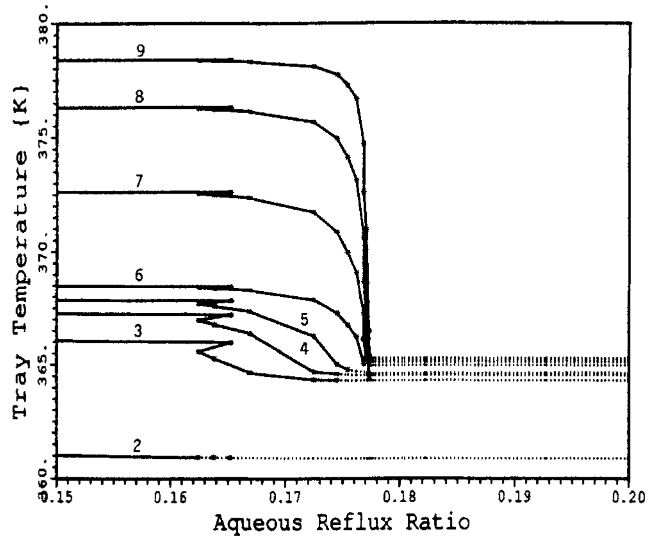

Figure 10. Temperature as a function of the aqueous reflux ratio for $N_T = 30$.

The tray numbers are annotated. Dotted curves denote two liquid phases; solid curves denote a single liquid phase.

Single-Stage Analysis

Steady-state multiplicity is commonly found in situations where competing effects occur, at least one of which varies nonlinearly with a system parameter. Examples include nonisothermal CSTR's (continuous stirred tank reactors) with exothermic reactions, where the heat released by the reactions is balanced by the heat removed through cooling, and predator-prey models.

In distillation, competing effects are not immediately apparent. Van Dongen et al. (1983) have shown that thermodynamic models such as UNIQUAC may introduce nonlinearities that result in steady-state multiplicity. Kingsley and Lucia (1987) performed a single-stage analysis (isothermal flash) for the ethanol-water-benzene system. In their study, the vapor flow rate was selected as the bifurcation parameter, and two- and three-phase envelopes were traced. For a feed composition located in the vapor-liquid-liquid (VLL) region, an improper vapor-liquid (VL) flash calculation produces two distinct branches (AC and BD in Figure 11), with two limit points. While the proper VLL flash calculation produces a single branch (HI) with no turning point, as shown in Figure 11. The improper VL flash calculation produces two dew points (C and D). The stable dew point (B) bounds the BI branch, along which the proper VL flash calculation is performed. Point I denotes the transition between VL and VLL systems at equilibrium (a point on the binodal curve). The proper continuation proceeds from the bubble point (H), along the VLL branch (3), to point I, and along the VL branch (IB). Yet, the other branches exist and can be expected to influence the convergence of the MESH equations for two or more trays, depending upon the initialization, when the phase distribution is uncertain.

These results have been reproduced using two sets of UNIQUAC parameters. For Figure 11, the parameters were reduced from the liquid-liquid equilibrium data of Bancroft (1942), as used by Kingsley and Lucia (see Table 1); and for Figure 12, the parameters were given by Gmehling and Onken (1977), reduced from vapor-liquid equilibrium data. The results are similar; however, the branches are connected differently.

A further bifurcation study has been conducted using AUTO, a software package developed by Doedel (1986). Among other capabilities, AUTO generates one- and two-parameter bifurcation diagrams for systems of nonlinear equations in the steady state.

To model the dynamics of two- and three-phase flash vessels, a system of differential-algebraic equations can be derived:

$$\underline{\mathcal{F}}\{\underline{X}', \underline{X}, \underline{Y}\} = 0 \tag{1}$$

$$\underline{\mathcal{G}}\{\underline{X}, \underline{Y}\} = 0 \tag{2}$$

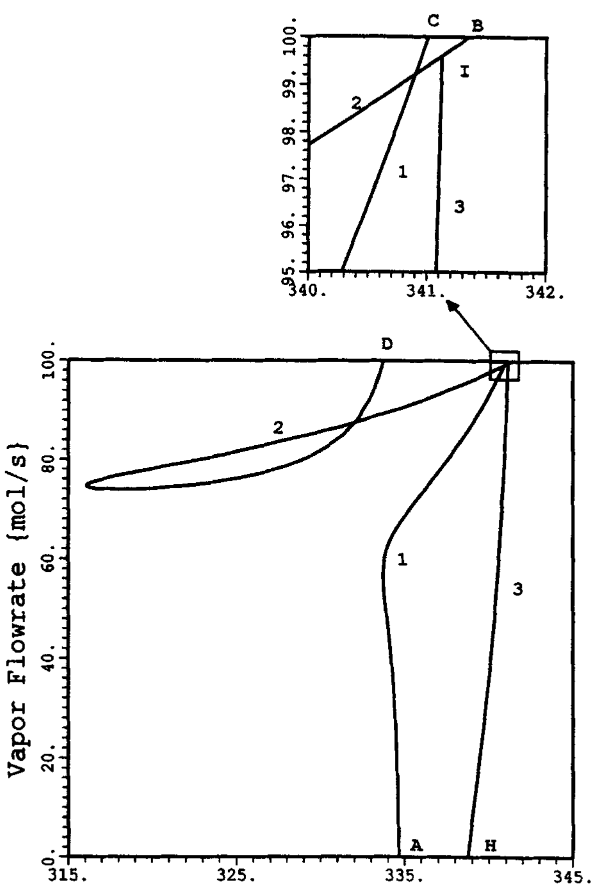

Figure 11. VL and VLL branches for the ethanol-benzene-water system.
UNIQUAC parameters reduced from liquid-liquid equilibrium data.

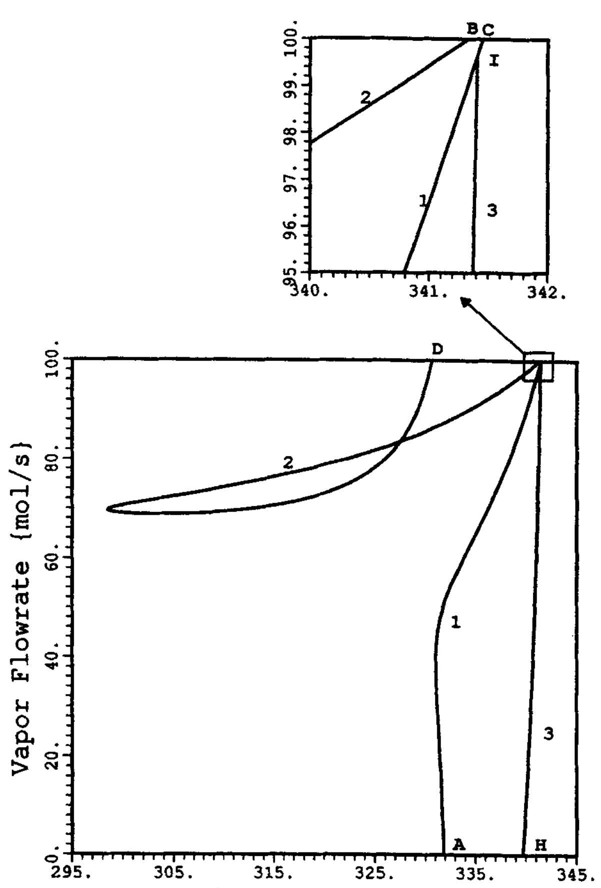

Figure 12. VL and VLL branches for the ethanol-benzene-water system.
UNIQUAC parameters reduced from vapor-liquid equilibrium data.

where $\underline{X}$ is the vector of state variables (often liquid flow rates of the species) and $\underline{Y}$ is a vector of dependent variables (solved for using Eq. 2, given $\underline{X}$). AUTO requires the Jacobian matrix for Eqs. 1 and 2 to determine the stability of a solution. Unlike a system of ordinary differential equations, the Jacobian matrix for the bifurcation analysis is:

$$\underline{\underline{J}} = \left(\frac{\partial \underline{X}'}{\partial \underline{X}}\right)_{\underline{\mathcal{G}}=0} \tag{3}$$

Applying the implicit function theorem gives:

$$\underline{\underline{J}} = -\left(\frac{\partial \underline{\mathcal{F}}}{\partial \underline{X}'}\right)^{-1}\left[\left(\frac{\partial \underline{\mathcal{F}}}{\partial \underline{X}}\right) - \left(\frac{\partial \underline{\mathcal{F}}}{\partial \underline{Y}}\right)\left(\frac{\partial \underline{\mathcal{G}}}{\partial \underline{Y}}\right)^{-1}\left(\frac{\partial \underline{\mathcal{G}}}{\partial \underline{X}}\right)\right] \tag{4}$$

The results from the AUTO software package are shown in Figure 13. Note that the turning points are reproduced, but no new bifurcation points are discovered.

A similar analysis was attempted for the SBA-II mixture using the UNIQUAC equation and the interaction coefficients in Table 1, which were reduced from VLL equilibrium data (Kovach and Seider, 1988). To imitate the changes in the aqueous reflux ratio, the water flow rate in the feed to the flash vessel is selected as the bifurcation parameter. A mixture with the composition of tray 2 in a 30-tray tower (feed on tray 7 and

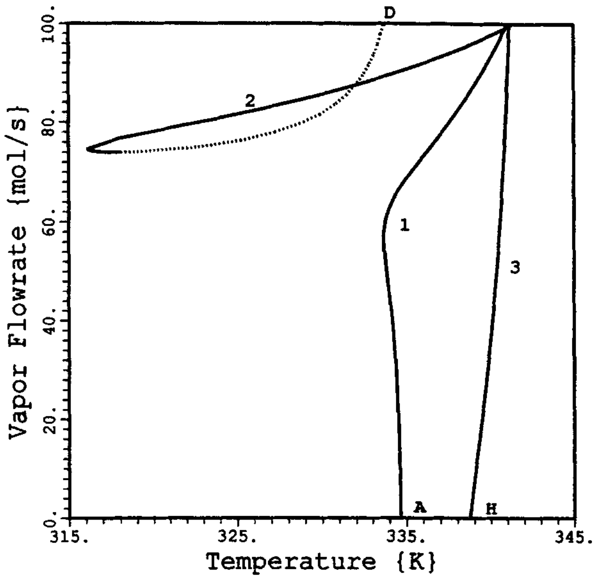

Figure 13. VL and VLL branches of the ethanol-benzene-water system, computed using AUTO.
UNIQUAC parameters reduced from liquid-liquid equilibrium data. Solid lines denote stable solutions and dotted lines denote unstable solutions.

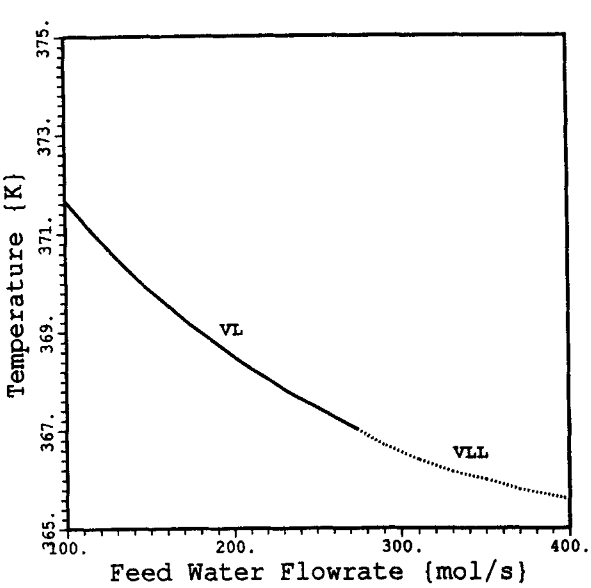

Figure 14. VL and VLL branches of the SBA-DSBE-water system computed using AUTO.

UNIQUAC parameters reduced from VLL equilibrium data.

aqueous reflux ratio, 0.1066), containing vapor and liquid phases, is introduced to the single stage and a VL flash calculation is carried out. When the second liquid phase appears, a VLL flash calculation is performed. The VL and VLL branches are shown in Figure 14. Note that steady-state multiplicity does not occur, as observed in the tower analysis. This suggests that factors beyond those that characterize a single stage, e.g., the impact of recycle, may be responsible for the multiplicity observed.

Conclusions

1. The equilibrium-stage model shows that a high concentration and recovery of SBA can be obtained with as few as one-third of the trays in the SBA-II tower.

2. A critical aqueous reflux ratio has been identified which, in addition to the product composition and recovery, is insensitive to the number of trays and the overall column efficiency. This is probably related to the small variation of the temperature and concentrations above their fronts in the stripping section.

3. Singularities are observed as the second liquid phase is introduced on the trays of the SBA-II tower with variations in the aqueous reflux. Regions with multiple solutions are identified and it appears that the multiplicity does not occur in a single stage. Other effects, such as recycle, may be responsible. When the multiplicity occurs, the composition and flow rate of the distillate and bottoms product are not significantly altered as the aqueous reflux varies. Yet, mathematical convergence procedures, such as the Newton-Raphson method, are adversely affected and more robust methods are required. Furthermore, potential control problems are introduced.

Literature Cited

Bancroft, W. D., and S. S. Hubard, "A New Method for Determining Dineric Distribution," *J. Amer. Chem. Soc.*, **64**, 347 (1942).

Byrne, G. D., and L. A. Baird, "Distillation Calculations Using a Locally Parameterized Continuation Method," *Comp. Chem. Eng.*, **9**, 593 (1985).

Doedel, E. J. "AUTO: Software for Continuation and Bifurcation Problems in Ordinary Differential Equations," Comp. Sci. Dept., Concordia Univ., Montreal, Canada (1986).

Goodliffe, A. H., "The Practical Testing of a Continuous Petroleum Still," *Trans Inst. Chem. Engrs.*, **12**, 107 (1934).

Gmehling, J., and U. Onken, *Vapor-Liquid Equilibrium Data Collection*, DECHEMA, Vol. 1, Pt. 1, 642 (1977).

Herfurth, H., A. Meirelles, S. Weiss, and H. Schubert, "Azeotropdestillation von Ethanol-Wasser mit Cyclohexan als Schleppmittel," *Chem. Tech.*, **39**, 331 (1987).

Herron, Jr., C. C., B. K. Kruelski, and J. R. Fair, "Hydrodynamics and Mass Transfer on Three-Phase Distillation Trays," *AIChE J.*, **34**, 1267 (Aug., 1988).

Kingsley, J. P., and A. Lucia, "Simulation and Optimization of Three-Phase Distillation Processes," AIChE Meeting, Houston (1987).

Kovach III, J. W., "Heterogeneous Azeotropic Distillation—An Experimental and Theoretical Study," PhD Thesis, Univ. of Pennsylvania (1986).

Kovach III, J. W., and W. D. Seider, "Heterogeneous Azeotropic Distillation: Experiment and Simulation Results," *AIChE J.*, **33**, 1300 (Aug., 1987a).

———, "Heterogeneous Azeotropic Distillation—Homotopy Continuation Methods," *Comp. Chem. Eng.*, **11**, 593 (1987b).

———, "Vapor-Liquid and Liquid-Liquid Equilibria for the System sec-Butyl Alcohol-Di-sec-butyl-Ether–Water," *J. Chem. Eng. Data*, **32**, 16 (1988).

Naphtali, L. M., and D. P. Sandholm, "Multicomponent Separation Calculations by Linearization," *AIChE J.*, **17**, 148 (Jan., 1971).

Prokopakis, G. J., and W. D. Seider, "Feasible Specifications in Azeotropic Distillation," *AIChE J.*, **29**, 49 (Jan., 1983).

Ross, B. A., and W. D. Seider, "Simulation of Three-Phase Distillation Towers," *Comp. and Chem. Eng.*, **5**, 7 (1980).

Shoenborn, E. M., J. H. Koffolt, and J. R. Withrow, "Rectification in the Presence of an Insoluble Component," *Trans. AIChE*, **37**, 997 (1941).

Swartz, C. L. E., and W. E. Stewart, "Finite-Element Steady State Simulation of Multiphase Distillation," *AIChE J.*, **33**, 1977 (Dec., 1987).

Van Dongen, D. B., M. F. Doherty, and J. R. Haight, "Material Stability of Multicomponent Mixtures and the Multiplicity of Solutions to Phase-equilibrium Equations: 1. Nonreacting Mixtures," *Ind. Eng. Chem. Fund.*, **22**, 472 (1983).

Vickery, D. J., and R. Taylor, "Path-following Approaches to the Solution of Multicomponent, Multistage Separation Process Problems," *AIChE J.*, **32**, 547 (Apr., 1986).

Manuscript received Jan. 23, 1989, and revision received May 26, 1989.