An Underwood-Like Model for a Finite Mass Exchanger

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A mathematical model is presented for a multicomponent, two-stream, countercurrent mass exchanger with a finite number of stages. The model relates the outlet compositions to the inlet compositions and the flow rate ratio of the two streams going through the exchanger, as well as the number of stages in the exchanger. An iterative solution procedure employing this model is proposed for the mass exchanger design problem. The model requires solution of a polynomial equation whose roots may be complex. Bounds to the real roots of the polynomial have also been established. Two examples illustrating how the method is employed are presented. The second example demonstrates that the iterative procedure converges even in the complex domain, and that complex roots exist for a real column.

Introduction

Mass exchange includes most commonly employed countercurrent unit operations for separation: gas absorption, adsorption, liquid–liquid extraction, and distillation. The design of such mass exchange unit operations is often performed using group methods. These are based on equations that relate stream inlets and outlets without requiring tedious and complex stage-by-stage calculations (King, 1981; Lo et al., 1983; Edmister, 1948). These equations provided an *explicit, analytical* model for the entire exchanger by employing simplifying assumptions. Thus, a designer is able to predict the outlets of an exchanger, given information on its inlet streams, flow rate ratio, and number of stages. This feature makes such models particularly suitable for use in network optimization (Bagajewicz and Manousiouthakis, 1992; Gupta and Manousiouthakis, 1994).

A typical simplifying assumption employed in the development of group methods is that the molar flow rate of each of the two phases passing through the exchanger remains constant. This assumption is valid for systems with low concentrations of the transferable components, or with a heat of vaporization independent of composition (McCabe–Thiele assumption for binary distillation), or with constant inert component flow rates. A mass exchanger in which this assumption is violated can be represented as multiple exchangers in series, each of which is modeled in this simplified manner. Another assumption, typically employed in group methods, is that the equilibrium relation among the compositions of the transferable components in each phase can be represented as a polynomial function. Examples of such group methods are the Kremser–Souders–Brown method (Brown et al., 1932; Kremser, 1930) for mass exchangers (constant flow rates, linear independent equilibrium relations for each transferable component) and the Underwood method (Underwood, 1945, 1946) for distillation (constant flow rates, constant relative volatility of each component with respect to any other component).

Martin (1963) presented a group method for the design of a mass exchanger that separates a binary mixture and features:

1. Variable flow rates, for each phase, giving rise to an operating line expressible as $y = \alpha x + \beta xy + \gamma$.
2. An equilibrium relation represented as $y = ax + bxy + c$.

The development of this model leads to a Riccati equation that may have real or complex roots. Once the roots are known, the number of plates of an exchanger required to achieve the desired separation target can be determined.

In this work we develop a group method for a *multicomponent* mass exchanger (two or more transferable components in each phase) that has constant flow rates for each phase, and a composition-dependent equilibrium relation with constant relative volatility. Aside from the importance of such a model for the design of single mass exchange units, it can also be useful for the design of multicomponent separation networks. For instance, Bagajewicz and Manousiouthakis (1992) showed that a distillation network can be represented as a combined mass and heat exchange network, through the state-space approach. The mass exchange network may em-

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ploy a representation consisting of countercurrent vapor–liquid mass exchange units. The success of such a general representation depends on a simplified mass exchanger representation that is able to predict the outlets of an exchanger based on its inlet streams, flow rate ratio, and number of stages. In this way,

- The computational effort associated with stage-by-stage calculations is avoided.
- The size of the associated optimization problem is reduced.
- The extent of problem nonlinearity may be reduced.
- The design variables for each unit operation employed (such as heat exchange area, A, number of stages of a mass exchanger, N) appear explicitly in the mathematical problem formulation.

The article is structured as follows. First, two relevant group methods are briefly reviewed. Then the new group model is presented. Several properties of this group method are investigated and bounds on the real roots of the model polynomial are established. Then the proposed model and its properties are employed in formulating an algorithm for the design and simulation of mass exchange units. Finally, the algorithm is demonstrated through two examples. The second example demonstrates that real columns may feature complex roots for the model polynomial. Nevertheless, the proposed algorithm is shown to converge to a physically meaningful solution.

Group Model Preliminaries

In this section, we briefly review the Kremser–Souders–Brown method (Brown et al., 1932; Kremser, 1930) for mass exchange and the Underwood method (Underwood, 1946, 1945) for distillation.

Consider the mass exchanger shown in Figure 1. Let the molar flow rates through the exchanger, L_{N+1}, L_1, V_N, and V_0, be assumed constant (independent of the stage) and equal to L and V, respectively. This constant molar flow condition is typically employed in the development of group methods. Let also the component equilibria be independent of each other, as defined by the linear relation

$$y_j = m_j x_j + b_j, \qquad j \in \boldsymbol{C}, \tag{1}$$

where m_j and b_j are parameters that determine the slope and intercept of the equilibrium line for component j, and $\boldsymbol{C}$ is the component index set

$$\boldsymbol{C} \triangleq \{j \mid j = 1,\ldots,C\}.$$

Such a countercurrent mass exchanger can be described by the Kremser–Souders–Brown (Brown et al., 1932; Kremser, 1930) equation:

$$\frac{y_{j,0} - y_{j,N}}{y_{j,0} - m_j x_{j,N+1} - b_j} = \frac{(L/m_jV) - (L/m_jV)^{N+1}}{1 - (L/m_jV)^{N+1}}, \qquad \forall j \in \boldsymbol{C}, \tag{2}$$

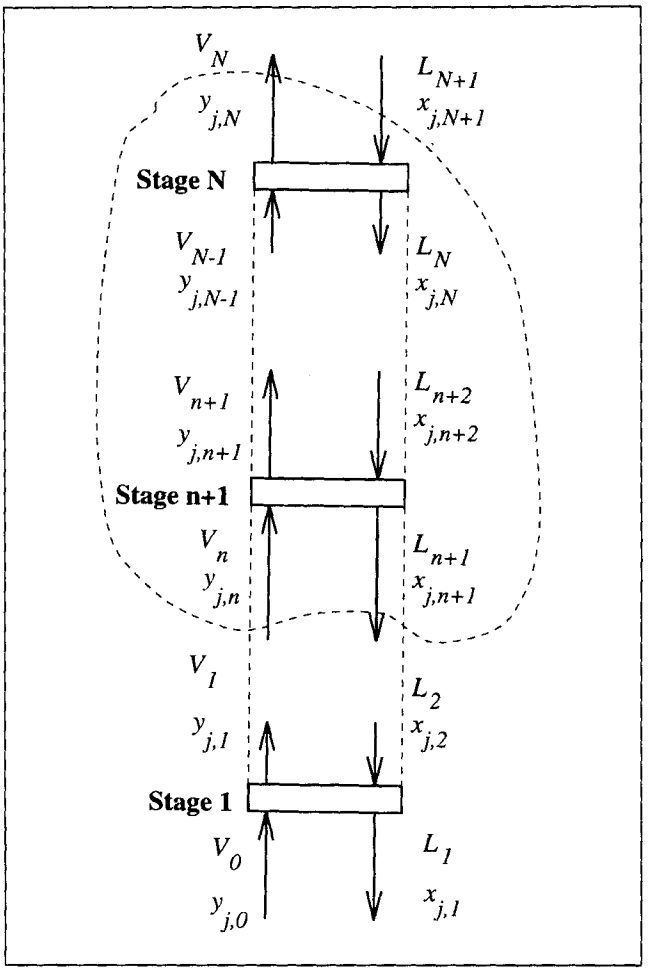

Figure 1. Countercurrent mass exchanger with two streams and N stages.

The loop shows a mass balance around the upper section (to the $n+1$th plate).

where N is the number of stages in the exchanger.

For either design or simulation, the inlet compositions $y_{j,0}$ and $x_{j,N+1}$, L/V, and either a target composition or N is given; the outlet compositions and either N or the target composition can then be determined through Eq. 2 and component mass balances.

Let us now consider a composition-dependent equilibrium relation. Let the equilibrium relation be defined as

$$y_j = K_j x_j, \qquad \forall j \in \boldsymbol{C}, \tag{3}$$

where K_j is the equilibrium ratio, and y_j, x_j indicate mole fractions. To account for temperature or concentration dependence of the equilibrium relation, we will let K_j vary. However, we will require that the relative volatility of each component, with regard to, say, the first component, remain unchanged. In mathematical terms this implies that the relative volatility, α_j, defined through Eq. 4, is constant.

$$\alpha_j \triangleq \frac{K_j}{K_1}, \quad \forall j \in \boldsymbol{C}. \tag{4}$$

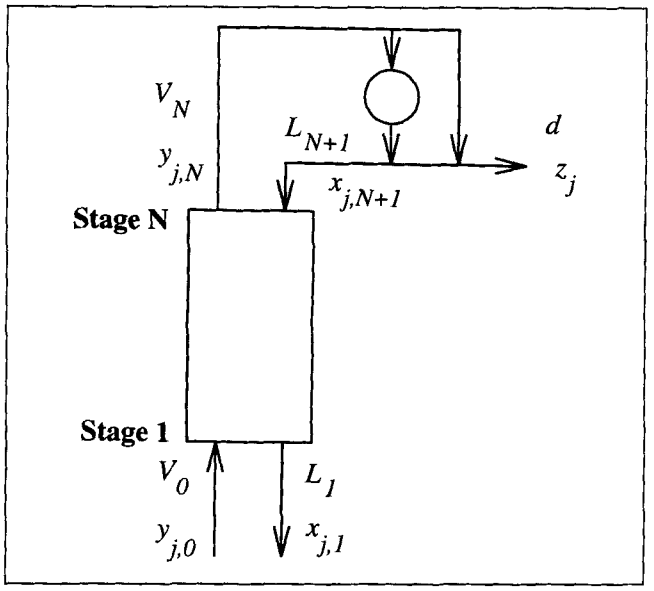

Figure 2. Rectifying section of a distillation column with *N* stages and distillate *d*.

Based on Eq. 3 and since y_j's indicate mole fractions, it also holds:

$$1 = \sum_{j \in C} y_j = \sum_{j \in C} K_j x_j. \tag{5}$$

Then based on Eqs. 4 and 5 the equilibrium relation can be written as

$$y_j = \frac{\alpha_j x_j}{\sum_{k \in C} \alpha_k x_k} \tag{6}$$

For a one-feed, two-product distillation column section, shown in Figure 2, with constant L/V ratio and constant α_j, Underwood (1946, 1945) developed the following group method:

$$\left(\frac{\varphi_q}{\varphi_p}\right)^N = \frac{\sum_{j \in C} \frac{\alpha_j x_{j,1}}{\alpha_j - \varphi_p}}{\sum_{j \in C} \frac{\alpha_j x_{j,1}}{\alpha_j - \varphi_q}}, \tag{7}$$

where, φ_p, φ_q are any of the C real roots of the following equation in φ:

$$\frac{V-L}{V} \sum_{j \in C} \frac{\alpha_j z_j}{\alpha_j - \varphi} = 1 \tag{8}$$

where, $V-L$ is the product molar flow rate d, and z_j is the distillate composition or the net upward flux of component j. These equations relate the compositions in the outlet $x_{j,1}$ to the number of the plates in the column section, N. Such a relation can be written for both a rectifying and a stripping section.

For the design of a column section as in Figure 2, the inlet compositions, $y_{j,0}$, L/V, and a key component target (any one z_j) are given. Then an iterative procedure can be employed to identify the exit compositions z_j and the number of plates. Design of a whole column requires an additional iteration that matches the column feed to the section feeds. Alternative iterative design methods exist in the literature (Hanson and Newman, 1977). They are often initiated by assuming that a zone of constant composition (infinite stages) exists near stage 1. Then estimates of $x_{j,1}$ and $y_{j,0}$ are made through limiting flow equations (King, 1981; Shiras et al., 1950; Edmister, 1948). Note that every root of Eq. 8 is guaranteed to be real and to lie in one of the intervals $(0, \alpha_C)$, (α_{j+1}, α_j) $\forall j = 1,\ldots,C-1$. This point will be elaborated on later in the article.

The preceding discussion demonstrates that simplified representations exist for mass exchangers with independent equilibria and for distillation columns with constant volatility equilibria. It is therefore desirable that such a simplified representation be developed for mass exchangers with constant volatility equilibria. In this work we undertake this task for a mass exchanger with a finite number of stages.

Mathematical Model

We now develop equations to model a mass exchanger with a finite number of stages, N. The mass exchanger is a contact device between two streams, one with molar flow rate, L, the other with molar flow rate, V. These flow rates, following the usual assumptions for group methods, are assumed constant throughout the exchanger. Each stream may have up to C components. The compositions (mole-fractions) of the two phases are indicated by x and y, respectively. The phases that exit each plate are assumed to be in phase equilibrium, and a constant volatility-based equilibrium model is employed (Eq. 6). Let us now define the following sets:

$$C_- \triangleq \{j \mid 1 \le j \le C, z_j < 0\}$$

$$C_+ \triangleq \{j \mid 1 \le j \le C, z_j \ge 0\}.$$

The term z_j employed in the preceding definitions is the net flow-normalized flux of component j in the V direction, and is

$$z_j \triangleq \left(y_{j,N} - \frac{L}{V} x_{j,N+1}\right) = \left(y_{j,0} - \frac{L}{V} x_{j,1}\right), \qquad \forall j \in C. \tag{9}$$

The mass balance around the section shown in Figure 1 is

$$L_{n+1} x_{j,n+1} + V_N y_{j,N} = L_{N+1} x_{j,N+1} + V_n y_{j,n}. \tag{10}$$

As assumed before, L and V are constant over the column. Then,

$$L x_{j,n+1} + V y_{j,N} = L x_{j,N+1} + V y_{j,n}. \tag{11}$$

The streams leaving stage n are in equilibrium with each other. Hence, $y_{j,n}$ is related to $x_{j,n}$ through Eq. 6. Thus,

$$\frac{L}{V}x_{j,n+1}+y_{j,N}-\frac{L}{V}x_{j,N+1}=\frac{\alpha_j x_{j,n}}{\Sigma\alpha_k x_{k,n}}, \tag{12}$$

where the symbol Σ without any index denotes summation over all C components. Multiplying this equation with $\alpha_j/(\alpha_j-\varphi)$, and adding over all components, yields

$$\frac{L}{V}\Sigma\frac{\alpha_j x_{j,n+1}}{\alpha_j-\varphi}+\Sigma\frac{\alpha_j y_{j,N}}{\alpha_j-\varphi}-\frac{L}{V}\Sigma\frac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi}=\frac{\Sigma\dfrac{\alpha_j}{\alpha_j-\varphi}\alpha_j x_{j,n}}{\Sigma\alpha_k x_{k,n}}. \tag{13}$$

Based on Eq. 9, the preceding equation becomes

$$\frac{L}{V}\Sigma\frac{\alpha_j x_{j,n+1}}{\alpha_j-\varphi}+\Sigma\frac{\alpha_j z_j}{\alpha_j-\varphi}=\frac{\Sigma\dfrac{\alpha_j}{\alpha_j-\varphi}\alpha_j x_{j,n}}{\Sigma\alpha_k x_{k,n}}. \tag{14}$$

Now let φ be defined so that

$$f(\varphi)=0, \tag{15}$$

where, the function $f(\varphi)$ is

$$f(\varphi)\triangleq\Sigma\frac{\alpha_j z_j}{\alpha_j-\varphi}-1. \tag{16}$$

This equation has C roots for φ, which are unaffected by plate numbers since α_j is constant. With this definition and this property, Eq. 14 can be manipulated to give

$$\frac{L}{V}\Sigma\frac{\alpha_j x_{j,n+1}}{\alpha_j-\varphi}=\varphi\frac{\Sigma\dfrac{\alpha_j x_{j,n}}{\alpha_j-\varphi}}{\Sigma\alpha_j x_{j,n}}. \tag{17}$$

Equation 17 holds for any of the C roots of Eq. 15. Let φ_p, φ_q be any two roots of Eq. 15, and write Eq. 17 for both of these roots. Dividing the resulting two equations, one obtains,

$$\frac{\Sigma\dfrac{\alpha_j x_{j,n+1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,n+1}}{\alpha_j-\varphi_q}}=\frac{\varphi_p}{\varphi_q}\frac{\Sigma\dfrac{\alpha_j x_{j,n}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,n}}{\alpha_j-\varphi_q}}. \tag{18}$$

This equation relates the composition on plate $n+1$ to those on plate n. Moreover, since φ does not depend on n, this equation can be developed into a geometric progression over the entire column. (The stages are numbered from the bottom to the top.)

$$\frac{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_q}}=\left(\frac{\varphi_p}{\varphi_q}\right)^N\frac{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_q}}. \tag{19}$$

Equation 19 relates the conditions at one end of the column to those at the other end, through a new variable, φ, defined through Eq. 15, which is independent of the number of plates in the column. Note that only $C-1$ Eq. 19s are independent of one another. Thus, when the φ's are real, φ_q can always be selected to be the root closest to α_C, call it φ_C, and φ_p to be any root of Eq. 15 different from φ_C, call them φ_p, $p=1,\ldots,C-1$. Then, Eq. 19 becomes

$$\frac{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_C}}=\left(\frac{\varphi_p}{\varphi_C}\right)^N\frac{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_C}}\qquad p=1,\ldots,C-1. \tag{20}$$

The φ's determined through Eq. 15 are not guaranteed to be real. In this case, one can define

$$\rho_p e^{i\theta_p}\triangleq\frac{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_p}}{\Sigma\dfrac{\alpha_j x_{j,N+1}}{\alpha_j-\varphi_C}}\frac{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_C}}{\Sigma\dfrac{\alpha_j x_{j,1}}{\alpha_j-\varphi_p}} \tag{21}$$

and

$$\sigma_p e^{i\eta_p}\triangleq\frac{\varphi_p}{\varphi_C}, \tag{22}$$

where $i=\sqrt{-1}$, $0\le\theta_p\le 2\pi$, and $0\le\eta_p\le 2\pi$. Equation 20 can now be expressed as

$$\rho_p e^{i\theta_p}=\sigma_p{}^N e^{iN\eta_p}. \tag{23}$$

Equation 23 holds if and only if

$$\rho_p=\sigma_p{}^N \tag{24}$$

and

$$\theta_p=2k\pi+N_{\eta_p}, \tag{25}$$

where $k\in Z$ (k is an integer). Equations 24 and 25 relate, like Eq. 19, the compositions at one end of the mass exchanger to those at the other end.

Model Properties

The parameter φ is defined so that Eq. 15 is satisfied. It is thus desirable that the characteristics of the function $f(\cdot)$ be understood. The following properties hold for this function:

Property 1. *$f(\varphi)$ is infinitely differentiable everywhere except at $\{\alpha_1,\ldots,\alpha_C\}$, where $\alpha_1>\alpha_2>\ldots>\alpha_C>0$. Its first derivative is*

$$f^{(1)}(\varphi) = \Sigma \frac{\alpha_j z_j}{(\alpha_j - \varphi)^2}$$

Property 2. $\lim_{\varphi \to \pm\infty} f(\varphi) = -1$ and $\lim_{\varphi \to \alpha_j^{\pm}} f(\varphi) = \mp \text{sgn}(z_j)\infty$.

Property 3. $\lim_{\varphi \to \alpha_j^{\pm}} f^{(1)}(\varphi) = \text{sgn}(z_j)\infty$.

Property 4. $\lim_{\varphi \to 0} f(\varphi) = \Sigma z_j - 1 = -\frac{L}{V}$.

Property 5. $\lim_{\varphi \to 0} f^{(1)}(\varphi) = \Sigma \frac{1}{\alpha_j} z_j$.

Property 6. The roots of $f(\cdot)$ and $f^{(1)}(\cdot)$ satisfy the following:

- *If $z_j z_{j+1} > 0$, there exists at least one real root of $f(\varphi)$ in the interval (α_{j+1}, α_j).*
- *If $z_j z_{j+1} < 0$, there exists at least one real root of $f^{(1)}(\varphi)$ in the interval (α_{j+1}, α_j).*

Property 7. Given φ_p that solves $f(\varphi_p) = 0$, then

$$\varphi_p \Sigma \frac{z_j}{\alpha_j - \varphi_p} = \frac{L}{V}.$$

The roots of Eq. 16 are not guaranteed to be real. Nevertheless, when real roots exist, the following theorems establish lower and upper bounds on them.

Theorem 1. Let $L/V > 0$. If φ_p is a real-valued solution to $f(\varphi) = 0$ and Eqs. 9, 31–34 also hold, then, $\varphi_p > 0$.

Proof. To contradict this, assume there exists a $\varphi_p \le 0$ that is a real-valued solution to $f(\varphi) = 0$. If $\varphi_p = 0$, we obtain $f(0) = \Sigma z_j - 1 = -L/V \neq 0$. Thus, $\varphi_p = 0$ cannot be a root of $f(\varphi) = 0$.

Now, let $\varphi_p < 0$. Since $\alpha_j > 0$, $\forall j \in \boldsymbol{C}$, $0 < \alpha_j/(\alpha_j - \varphi_p) < 1$. Then, $0 \le \alpha_j z_j/(\alpha_j - \varphi_p) \le z_j$, $\forall j \in \boldsymbol{C}_+$, and $0 > \alpha_j z_j/(\alpha_j - \varphi_p) > z_j$, $\forall j \in \boldsymbol{C}_-$. Summing over all $j \in \boldsymbol{C}_+$ and $j \in \boldsymbol{C}_-$, respectively, we obtain

$$\left.\begin{aligned} 0 \le \sum_{j \in \boldsymbol{C}_+} \frac{\alpha_j z_j}{\alpha_j - \varphi_p} \le \sum_{j \in \boldsymbol{C}_+} z_j, \\ 0 > \sum_{j \in \boldsymbol{C}_-} \frac{\alpha_j z_j}{\alpha_j - \varphi_p} > \sum_{j \in \boldsymbol{C}_-} z_j. \end{aligned}\right\}$$

$$\Rightarrow \sum_{j \in \boldsymbol{C}_-} z_j - 1 < f(\varphi_p) < \sum_{j \in \boldsymbol{C}_+} z_j - 1. \quad (26)$$

Clearly, $\Sigma_{j \in \boldsymbol{C}_-} z_j - 1 < -1 < 0$. Moreover,

$$\Sigma z_j = \sum_{j \in \boldsymbol{C}_-} z_j + \sum_{j \in \boldsymbol{C}_+} z_j = 1 - \frac{L}{V} \Rightarrow$$

$$\sum_{j \in \boldsymbol{C}_+} z_j - 1 = -\frac{L}{V} - \sum_{j \in \boldsymbol{C}_-} z_j \Rightarrow$$

$$\sum_{j \in \boldsymbol{C}_+} z_j - 1 = -\sum_{j \in \boldsymbol{C}_-} y_{j,N} - \frac{L}{V}\left(1 - \sum_{j \in \boldsymbol{C}_-} x_{j,N+1}\right).$$

Since $L/V \neq 0$ and Eqs. 32 and 34 hold, the preceding quantity is strictly negative unless both $\Sigma_{j \in \boldsymbol{C}_-} y_{j,N} = 0$ and $\Sigma_{j \in \boldsymbol{C}_-} x_{j,N+1} = 1$ hold. But $\Sigma_{j \in \boldsymbol{C}_-} y_{j,N} = 0 \Leftrightarrow y_{j,N} = 0$, $\forall j \in \boldsymbol{C}_-$ Eq. 6 $\Leftrightarrow x_{j,N} = 0$, $\forall j \in \boldsymbol{C}_-$. But z_j is constant throughout the column. Thus $z_j = y_{j,N-1} - (L/V)\, x_{j,N}$, $\forall j \in \boldsymbol{C}_-$. Then $x_{j,N} = 0$, $\forall j \in \boldsymbol{C}_- \Rightarrow z_j = y_{j,N-1} \ge 0$, $\forall j \in \boldsymbol{C}_-$. This contradicts the definition of $\boldsymbol{C}_-$ unless $\boldsymbol{C}_-$ is the empty set. If $\boldsymbol{C}_-$ is empty, then $\Sigma_{j \in \boldsymbol{C}_-} x_{j,N+1} = 0 < 1$. Therefore, $\Sigma_{j \in \boldsymbol{C}_+} z_j - 1$ is strictly negative. Hence, φ_p cannot be a root of $f(\cdot)$.

Theorem 2. Let K denote the number of elements of $\boldsymbol{C}-$ and let $\alpha_1 > \alpha_2 > \ldots \alpha_C > 0$. Then

1. *If $K = 0$, $z_j > 0$, $\forall j \in \boldsymbol{C}$, $L/V > 0$, then $f(\varphi)$ has one real root in each of the intervals $(0, \alpha_C)$, (α_{j+1}, α_j) $j = 1, \ldots, C-1$.*
2. *If $K > 0$, then a strict upper bound on all the real roots of $f(\varphi)$ is*

$$\max\left[\max_{j \in \boldsymbol{C}_-} [\alpha_j(1 - Kz_j)], a_1\right].$$

Proof.

1. Since $K = 0$, $z_j > 0$, $\forall j \in \boldsymbol{C}$, Property 6 implies that there exists at least one real root of $f(\varphi)$ in each of the $C-1$ intervals (α_{j+1}, α_j) $j = 1, \ldots C-1$. Furthermore, due to Property 4, $f(0) = -(L/V) < 0$, and due to Property 2, $\lim_{\varphi \to \alpha_C^-} f(\varphi) = +\infty$. Thus, there is also a real root in $(0, \alpha_C)$ and all C roots are accounted for.

2. We prove this through contradiction. Let φ be such that $\varphi > \alpha_j(1 - Kz_j)$, $\forall j \in \boldsymbol{C}_-$, and $\varphi > \alpha_1$. Then,

$$K > 0 \Rightarrow \frac{\varphi}{K} > \frac{\alpha_j}{K} - \alpha_j z_j \Rightarrow \alpha_j z_j > \frac{(\alpha_j - \varphi)}{K}.$$

Since $\varphi > \alpha_1 > \ldots > \alpha_C$, the above implies

$$\frac{\alpha_j z_j}{\alpha_j - \varphi} < \frac{1}{K}, \quad \forall j \in \boldsymbol{C}_-, \Rightarrow \sum_{j \in \boldsymbol{C}_-} \frac{\alpha_j z_j}{\alpha_j - \varphi} < \sum_{j \in \boldsymbol{C}_-} \frac{1}{K} = 1 \Rightarrow$$

$$\sum_{j \in \boldsymbol{C}_-} \frac{\alpha_j z_j}{\alpha_j - \varphi} + \sum_{j \in \boldsymbol{C}_+} \frac{\alpha_j z_j}{\alpha_j - \varphi} < 1 \Rightarrow \sum_{j \in \boldsymbol{C}} \frac{\alpha_j z_j}{\alpha_j - \varphi} - 1 < 0$$

Thus φ cannot be a root of $f(\varphi)$.

Let us now analyze the importance of Theorems 1 and 2. When $z_j > 0$, $\forall j \in \boldsymbol{C}$, it is established in Theorem 2 that all the roots of $f(\varphi)$ are real. This suggests that when the net upward flux, z_j, in the mass exchanger is positive, then the aforementioned roots are always real. This is guaranteed in the case of the original Underwood model for a distillation column section (see Eq. 8), since the net upward flux z_j is the same as the product component flows that are always nonnegative. For a mass exchanger, however, this is not guaranteed, and negative upward fluxes can be commonplace. Nevertheless, in Kolokol'nikov et al. (1986a,b) it is shown that a mass exchanger with infinite plates and a zone of constant composition (ZCC), features only real roots in Eq. 15. These roots were shown to coincide with some of the roots of two

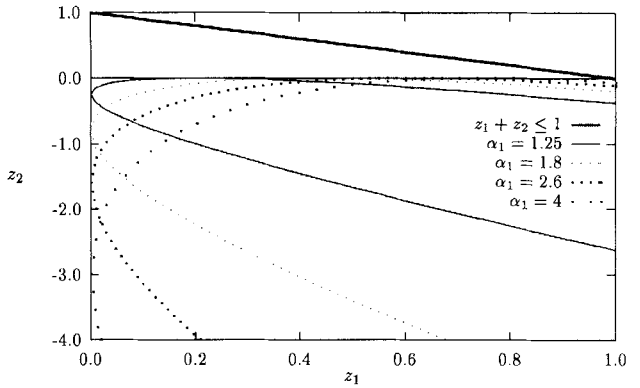

Figure 3. Net component flux region in which $f(\cdot)$ possesses complex roots for a binary mixture.

"feed"-related functions whose roots are guaranteed to be real. Interestingly, as shown below in the examples a real mass exchanger with a *finite* number of plates may feature complex roots in Eq. 15. This characteristic is investigated next, in more detail, for a binary mixture.

Consider a binary mixture whose equilibrium is governed by a constant relative volatility relation:

$$y_1 = \frac{\alpha_1 x_1}{(\alpha_1 - 1)x_1 + 1}, \qquad (\alpha_2 = 1). \tag{27}$$

For this system, Eq. 15 becomes

$$f(\varphi) = \frac{\alpha_1 z_1}{\alpha_1 - \varphi} + \frac{z_2}{1 - \varphi} - 1 = 0. \tag{28}$$

This leads to a quadratic equation in φ:

$$-\varphi^2 + (-\alpha_1 z_1 - z_2 + 1 + \alpha_1)\varphi + \alpha_1 z_1 + \alpha_1 z_2 - \alpha_1 = 0. \tag{29}$$

The roots of this equation are guaranteed to be real if and only if its discriminant is nonnegative. This requirement leads to the condition:

$$\alpha_1^2 z_1^2 + 2\alpha_1 z_1 z_2 + z_2^2 - 2\alpha_1(\alpha_1 - 1)z_1 + 2(\alpha_1 - 1)z_2 + \alpha_1^2 - 2\alpha_1 + 1 \ge 0. \tag{30}$$

The lefthand side of this inequality is of the general quadratic form: $az_1^2 + 2bz_1 z_2 + cz_2^2 + 2dz_1 + 2ez_2 + f$. Since $ac - b^2 = \alpha_1^2 - \alpha_1^2 = 0$, this lefthand side defines a parabolic curve in the z_1-z_2 plane, for all values of $\alpha_1 > 0$ (Yefimov, 1964). A plot of the parabolic relation for several values of α_1 is shown in Figure 3. Complex roots exist for z_1-z_2 values that lie within the parabola. As α_1 increases, the region contained in the parabola increases.

The parabola is bound to the left and above by the $z_1 = 0$, $z_2 = 0$ axes, respectively. The points of intersection with the two axes are $z_2 = 1 - \alpha_1$, and $z_1 = 1 - (1/\alpha_1)$, respectively. Also, from Eq. 35, we have

$$z_1 + z_2 = 1 - \frac{L}{V}.$$

Since $L/V \ge 0$, we have $z_1 + z_2 \le 1$, for all feasible values of z_1 and z_2. This line is also plotted in Figure 3.

For large values of α_1, it is apparent from the plot (Figure 3) that several feasible z_1 and z_2 values will lead to complex roots for real mass exchangers.

Having outlined the proposed model and its mathematical properties, we proceed to propose a design and simulation procedure for a multicomponent mass exchanger.

Mass Exchanger Design and Simulation

First, note the following relations for the inlet or outlet streams of the exchanger:

$$\sum y_{j,0} = 1, \quad y_{j,0} \ge 0, \qquad j \in \boldsymbol{C}, \tag{31}$$

$$\sum y_{j,N} = 1, \quad y_{j,N} \ge 0, \qquad j \in \boldsymbol{C}, \tag{32}$$

$$\sum x_{j,1} = 1, \quad x_{j,1} \ge 0, \qquad j \in \boldsymbol{C}, \tag{33}$$

$$\sum x_{j,N+1} = 1, \quad x_{j,N+1} \ge 0, \quad j \in \boldsymbol{C}. \tag{34}$$

Also, summing Eq. 9 over $\boldsymbol{C}$ and substituting the preceding equations, one obtains the relation

$$\sum z_j = 1 - \frac{L}{V}, \quad \frac{L}{V} \ge 0. \tag{35}$$

The model developed in the section on group model preliminaries can now be used for either design or simulation of mass exchangers. Each mass exchanger is defined by $C - 1$, Eqs. 19; $2C$, Eqs. 9 and Eqs. 32-34. That gives a total of $3C + 2$ equations. The variables are L/V, $4C$ inlet ($x_{j,N+1}$, $y_{j,0}$) and outlet ($x_{j,1}$, $y_{j,N}$) compositions for the two streams, C component fluxes (z_j), and the number of plates, N, a total of $5C + 2$ variables. The solution of either a design or simulation problem, therefore, requires that $5C + 2 - 3C - 2 = 2C$ variables be specified. Any combination of three equations from Eqs. 31-34 can be employed.

In a typical mass exchanger design problem, one is given $2(C - 1)$ inlet ($x_{j,N+1}$, $y_{j,0}$) compositions, the L/V ratio, and a target composition for one component (the "key" component target, say, $y_{C,N}$) in one of the streams. It is then desired to determine the remaining variables (N, $x_{j,1}$, $y_{j,N}$, $x_{C,N+1}$, $y_{C,0}$). The remaining two inlet compositions can be evaluated from Eqs. 31 and 34. Given $y_{C,N}$ and L/V, $x_{C,1}$ and z_C can be determined through Eqs. 9 applied for $j = C$.

We now present an iterative solution algorithm for the mass exchanger design problem. Let us define, in the kth iteration, the following vectors: $\boldsymbol{x}_{N+1}$ and $\boldsymbol{y}_0$ (the iteration-independent inlet compositions), $\boldsymbol{y}_N^k$ (the V-phase outlet compositions), z^k and $\hat{z}^k$ (the guessed and evaluated fluxes), $\boldsymbol{x}_1^k$ and $\hat{\boldsymbol{x}}_1^k$ (two evaluated L-phase outlet compositions), z_*^k and $\hat{z}_*^k$ (vectors consisting of, say, the first $C - 2$ elements of z^k and $\hat{z}^k$, respectively), φ^k (the roots of Eq. 15, where z_j, $j \in \boldsymbol{C}$ are the elements of z^k), and N^k (the solutions of Eq. 20, where

$x_{j,N+1}$, $x_{j,1}$, $j \in C$ are the elements of $\boldsymbol{x}_{N+1}$ and $\boldsymbol{x}_1^k$, respectively). Then the iterative algorithm can be described as follows:

Step 1. Set $k = 0$. Guess z_*^0.

Step 2. This is the kth iteration and z_*^k is given. The following functions are then evaluated (numbers in parentheses refer to equation numbers):

$$g_0 : z_*^k \xrightarrow{z_C,(35),L/V} z^k,$$

where, z_C is the last element of z^k;

$$g_1 : z^{kL/V,y_0,C(9),} \boldsymbol{x}_1^k,$$

Note that the last element of $\boldsymbol{x}_1^k$ is equal to $x_{C,1}$ and Eq. 33 is guaranteed to be satisfied.

$$g_2 : z^{k\,(15),(16)} \boldsymbol{\varphi}^k,$$

When there are at least two real elements of $\boldsymbol{\varphi}^k$ (say, φ_1^k and φ_C^k), the following function is evaluated:

$$g_{3a} : (\boldsymbol{x}_1^k, \boldsymbol{\varphi}^k) \xrightarrow{x_{N+1},1(20)} N^k,$$

else the following function is evaluated:

$$g_{3b} : (\boldsymbol{x}_1^k, \boldsymbol{\varphi}^k) \xrightarrow{x_{N+1},1(24)\text{ or }1(25)} N^k.$$

(Only two elements of $\boldsymbol{\varphi}^k$ have been employed in evaluating g_{3a} or g_{3b}.) The choice of Eq. 24 or Eq. 25 may affect the convergence of the algorithm, though use of either equation guarantees N^k to be real. In either case, upon convergence, both equations are required to hold. The composite function g_3 refers to either g_{3a} or g_{3b} as appropriate.

$$g_4 : (\varphi^k, N^k) \xrightarrow{x_{N+1},x_{C,1},C-2(20),1(33)} \hat{\boldsymbol{x}}_1^k,$$

Note that $x_{C,1}$ is the last element of $\hat{\boldsymbol{x}}_1^k$, and that given $\boldsymbol{x}_{N+1}$, N^k and φ^k, evaluation of $\hat{\boldsymbol{x}}_1^k$ only requires the solution of a linear system of C equations. If some of the elements of φ^k are complex, the $C-2$ Eq. 20s are evaluated in the complex domain, and only the real parts of each lefthand side are equated to the corresponding real parts of the righthand side, thus yielding the required $C-2$ linear equations.

$$g_5 : \hat{\boldsymbol{x}}_1^k \xrightarrow{L/V,y_0,C(9)} \hat{z}^k,$$

The last element of $\hat{z}^k$ is at its known value, z_C, and Eq. 35 is satisfied.

$$g_6 : \hat{z}^k \longrightarrow \hat{z}_*^k = [\boldsymbol{I}_{C-2}\ \mathbf{0}\ \mathbf{0}]\hat{z}^k,$$

Step 3. $\hat{z}^k$ and z^k (or equivalently $\hat{z}_*^k$ and z_*^k) should be equal to each other for the mass exchanger design problem to be solved. Consider the equations:

$$h(z_*) - z_* = \mathbf{0},$$

Table 1. Data for the Three Components of Example 1

Component	Rel. Volatility α_j	Inlet Compositions $x_{j,N+1}$	$y_{j,0}$
1	4	0.7840	0.2750
2	2	0.0440	0.4625
3	1	0.1720	0.2625

where, $h(z_*) \triangleq g_6\ [g_5(g_4\{g_1[g_0(z_*)],\ g_3(g_2[g_0(z_*)]),\ g_1[g_0(z_*)]\})]$. In this step, a Newton iteration is employed on the equations $h(z_*) - z_* = \mathbf{0}$, to generate z_*^{k+1}. Thus,

$$z_*^{k+1} = z_*^k - \left[\frac{\partial h}{\partial \boldsymbol{z}_*} - \boldsymbol{I}_{C-2}\right]\Bigg|_{z_*^k}^{-1} (h(z_*^k) - z_*^k).$$

Step 4. If $|z_*^{k+1} - z_*^k| < \epsilon$ (a prespecified tolerance), then terminate. z is evaluated through z_*^{k+1}, z_C, and Eq. 35. If $|z_*^{k+1} - z_*^k| \geq \epsilon$, set $k = k+1$ and return to step 2 with the new value for z_*^k equal to the just evaluated z_*^{k+1}.

Thus, steps 1–4 describe an algorithm for the design of a multicomponent mass exchanger.

The model given earlier can also be employed to simulate a mass exchanger. The simulation of a mass exchanger requires that, given the inlet compositions, the L/V ratio, and the number of plates, all the outlet compositions be determined. An algorithm similar to the one just outlined can be employed for this purpose.

Examples

Example 1

Consider two three-component streams with compositions and relative volatilities as in Table 1 and an L/V ratio of 0.625. Design a mass exchanger to yield an outlet V-phase composition for component 1 equal to $y_{1,2} = 0.64$.

Clearly, Eqs. 9 imply $x_{1,1} = 0.2$, $z_1 = 0.15$. Thus, the solution of this problem requires a Newton–Raphson iteration over the variable z_2. We start with the initial value $z_2^0 = 0.16$. The derivatives of the composite function defined in the previous section, step 3, $h(z_2^k)$, are calculated numerically. The result is shown in Table 2. The column is found to have two stages.

Example 2

Consider two three-component streams with compositions and relative volatilities as in Table 3 and an L/V ratio of 2.0. Design a mass exchanger to yield an outlet V-phase composition for component 1 equal to $y_{1,2} = 0.60$.

Table 2. Results for the Three Components of Example 1

Comp.	Outlet Compos. $x_{j,1}$	$y_{j,N}$	Flux z_j	Intermed. Compos. $y_{j,1}$	$x_{j,2}$
1	0.2000	0.6400	0.1500	0.4000	0.4000
2	0.4000	0.2400	0.2125	0.4000	0.3000
3	0.4000	0.1200	0.0125	0.2000	0.3000

Table 3. Data for the Three Components of Example 2

Component	Rel. Volatility α_j	Inlet Composition $x_{j,N+1}$	$y_{j,0}$
1	4	0.2500	0.8500
2	2	0.4750	0.1000
3	1	0.2750	0.0500

Equations 9 imply $x_{1,1} = 0.375$ $z_1 = 0.10$. Again, a Newton–Raphson iteration over the variable z_2 is pursued. We guess the initial value of $z_2^0 = -0.75$. Then, from Eq. 35, $z_3^0 = -0.35$. Following the procedure in the previous section, a Newton–Raphson iteration over the variable z_2 is carried out. Numerical derivatives are employed. At the first iteration, two elements of φ^1 are complex and conjugate of each other. Equation 20 is therefore solved in the complex domain, through Eq. 25. Function g_4 is evaluated by equating the real coefficients of Eq. 20. Upon convergence, the resulting mass exchanger is found to have one stage. The outlet compositions and fluxes are summarized in Table 4.

At this solution, the net component fluxes are found to be $z_1 = 0.100$, $z_2 = -0.650$, and $z_3 = -0.450$. By Property 6, there is a real root, of $f(\cdot)$, $\varphi \in (\alpha_3, \alpha_2)$, but no guarantee of the existence of a real root $\varphi \in (\alpha_2, \alpha_1)$ or $\varphi \in (0, \alpha_3)$. A plot of the function $f(\cdot)$ is shown in Figure 4. In fact, the roots of $f(\varphi)$ are $\{1.183642564, 3.583178718 \pm 0.8236653305\, i\}$, that is, *two of the roots are complex.*

This example sheds some light on possible column operating scenarios when complex roots occur. Clearly, for small L/V ratios ($L/V \to 0$), the net component fluxes are guaranteed to be positive, thus ensuring all roots are real. As the L/V ratio increases, some of these fluxes may become negative. For example, this will be the case if the liquid phase inlet is rich in the heavy (less volatile or low α_j) components, and the vapor phase inlet is rich in the most volatile (light) component. Then, for some value of L/V, the net flux of the heavy components will be downward (in the direction of liquid flow) and negative, while the net flux of the light component will be upward (positive). One should note that as L/V becomes large ($L/V \to \infty$), all net component fluxes become negative. Nevertheless, the polynomial roots in this case are again guaranteed to be real.

Conclusions

The simulation, design, and optimization of multicomponent mass exchanger units and networks necessitates the development of simplified mathematical models that relate the outlet compositions of a countercurrent exchanger to its inlet compositions, its flow rate ratio, and its number of plates. In this article such a model was developed for an exchanger with a finite number of stages. The proposed model required solution of a polynomial whose roots are not guaranteed to be real. We established properties to bound the real roots when they exist. With these results, an iterative solution procedure was then proposed for this mass exchanger model. Two examples that illustrate this procedure were presented. The second example demonstrated that a realizable mass exchanger may have complex roots for the aforementioned polynomial, yet the proposed design algorithm can be utilized for its design.

Table 4. Results for the Three Components of Example 2

Component	Outlet Composition $x_{j,1}$	$y_{j,N}$	Flux z_j
1	0.3750	0.6000	0.100
2	0.3750	0.3000	−0.650
3	0.2500	0.1000	−0.450

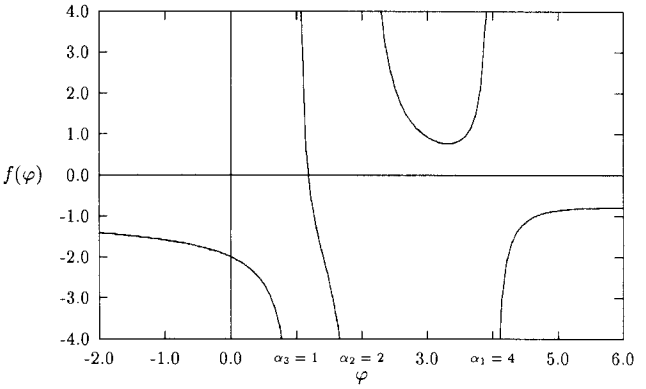

Figure 4. Function $f: \varphi \to f(\varphi) = 0.4/(4 - \varphi) - 1.3/(2 - \varphi) - 0.45/(1 - \varphi) - 1$ (eq. 16) for Example 2.

Notation

i = imaginary axis
k = component index
n = a stage of the mass exchanger
η = angle, polar representation of a complex number
ρ = radius, polar representation of a complex number
σ = radius, polar representation of a complex number
θ = angle, polar representation of a complex number
$*$ = reduced vector of $C-2$ components

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Correction

In the article titled "Breakage and Coalescence Models for Drops in Turbulent Dispersions," by C. Tsouris and L. L. Tavlarides (March 1994, p. 395), the following corrections are made:

- Eqs. 8, 36, 39 and 42 are frequency functions, therefore the term n_d should be removed, since it already exists in the drop rate functions of Eqs. 2 and 3.
- In Eq. 14, $e^{2/3}$ should read $\epsilon^{2/3}$.
- In Eq. 42, ϵ_1 should read ϵ_i.
- Following Eq. 42, $c_1 = 0.48$ instead of 1.3. All computations were performed with $c_1 = 0.48$, which is smaller than unity as expected.