On the Nonlinear Wave Theory for Dynamics of Binary Distillation Columns

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A previous article (Hwang, 1991) presented a nonlinear wave theory for predicting the dynamic behavior of binary distillation columns, in particular the severely nonlinear behavior of columns producing high-purity products. Viewing the movement of the composition and temperature profiles in a distillation column as the propagation of nonlinear waves, this theory provides a simple mathematical model for distillation dynamics and a cause-and-effect analysis of nonlinear phenomena such as high steady-state gains, large response lags, strong dependence on disturbances, and asymmetric dynamics. The article also elucidated the inevitable link between nonlinear behavior and high-purity operation with a discussion of two mechanisms for the material transport from one column end to the other. Recently, Han and Park (1993) applied the nonlinear wave model to a profile-position control strategy for a high-purity column and showed very encouraging results.

This article fortifies the nonlinear wave theory by including the dissipation effect at column ends as well as feed and product side streams and nonuniform flow rates. In particular, the column-end effect is so critical to countercurrent processes as emphasized earlier (Hwang, 1987; Hwang and Helfferich, 1988) that a model of practical usefulness must take it into account at least qualitatively, and preferably quantitatively. Such an effect was illustrated previously with numerical simulations for both packed and tray columns, but was discussed only qualitatively in the nonlinear wave model (Hwang and Helfferich, 1988; Hwang, 1991). With the inclusion of this effect in a quantitative way, the generalized model can be used to approximate, with fairly simple mathematics, the very slow return to a balanced steady state in the asymmetric dynamics discussed previously. In addition to the column-end effect, the inclusion of feed and product side streams supplies a more precise criterion for a balanced (optimal) operating condition of a fractionating column. This provides a stepping-stone for the expansion of the nonlinear wave theory to multicomponent distillation. The generalized wave model also accounts for the variation of molar flow rates, which may result from significantly different heats of vaporization of the components or from a considerable heat loss from the column.

Generalized Wave Velocity

The nonlinear wave theory for distillation and for countercurrent processes in general originated from that for fixed-bed sorption (chromatography), of which the central theme was developed based on the assumption of phase equilibrium. For a self-sharpening wave, which is the primary wave type in distillation columns (Hwang, 1991), the wave velocity equations given in that article and by others (Marquardt, 1988) imply vapor-liquid equilibrium (pinched) conditions on both sides of the wave. Such a condition at a column end is necessary for a high-purity product from that end, but the pinch will break if the wave, in response to a disturbance, approaches the column end. This leads to the nonequilibrium column-end effect, which is the major difference between the wave propagation phenomena in countercurrent columns and in fixed beds (Hwang, 1987; Hwang and Helfferich, 1988).

In the nonlinear wave theory, the equilibrium relation is essential in determining the sharpening behavior of waves. Once a wave has been proved to be self-sharpening and ultimately becomes a shock wave (constant pattern), however, its velocity can be calculated without the information of the equilibrium relationship, provided that the compositions of both phases on both sides of the wave are available. After all, the wave velocity represents only a dynamic material balance, and bears no direct connection to the equilibrium between the two phases. Focusing on self-sharpening waves exclusively, the following discussion will consider wave propagation in a general sense of dynamic material (or energy) balance without the assumptions of phase equilibrium as well as uniform flow rates.

Consider a binary distillation column section with entering and leaving streams as illustrated by Figure 1a (the feed F may represent a lumped stream of multiple feeds; so may the side product M). With the liquid and vapor molar holdups per unit length or per tray denoted by W and U, respectively, one can write an integral material balance of the light component around the column section as follows:

$$\frac{d}{dt}\int_{s'}^{s''}(Wx+Uy)ds=(V'y'+L''x''+Fz_F)-(L'x'+V''y''+Mz_M) \quad (1)$$

This equation is applicable to both packed and tray columns; for the latter, the integral becomes a summation with s in terms

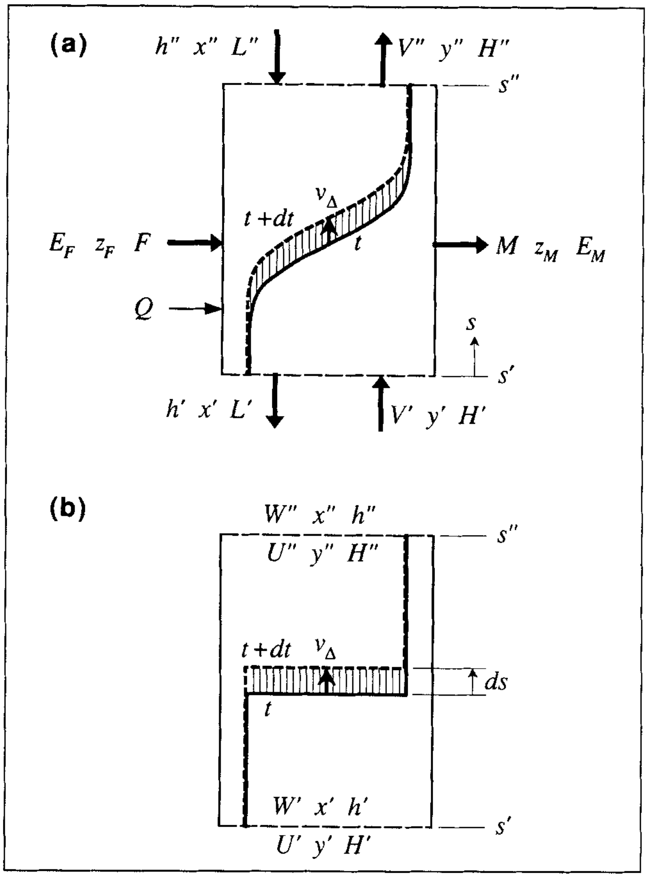

Figure 1. Column section of binary distillation.

(a) Streams and an example shock wave; (b) travel of a step at shock wave velocity.

of number of trays. Note that this material balance is independent of the shape, location, and sharpening character of the profile (wave). For a shock wave as Figure 1a exemplifies (the same shape for the waves in terms of x, y, W, and U—though there may be lags between liquid and vapor properties), its velocity can be determined by viewing the wave as a discontinuous step, as illustrated by Figure 1b. To maintain the integral material balance (Eq. 1), one requires that the step sweeps an equal area (shaded) under the profile of $Wx + Uy$ as the shock wave does in a given time interval dt. This leads to:

$$\frac{d}{dt}\int_{s'}^{s''}(Wx+Uy)ds = \frac{[(W'x'+U'y')-(W''x''+U''y'')]ds}{dt} \tag{2}$$

Combining Eqs. 1 and 2 results in the following shock wave velocity:

$$v_{\Delta x}=v_{\Delta y}=\frac{ds}{dt} = \frac{(V''y''-L''x'')-(V'y'-L'x')-Fz_F+Mz_M}{(W''x''+U''y'')-(W'x'+U'y')} \tag{3}$$

where $\Delta x \equiv x'' - x'$ and similarly for y. Note that Δx and Δy traveling at the same wave velocity does not imply an equilibrium relation between x and y. Considering the movement of a step, Gilles and Retzbach (1980, 1983) gave a similar equation without discussing the wave sharpening character to justify the step approximation.

Strictly speaking, Eq. 3 is exact only for a wave with a strictly constant shape in a column section with no side streams and with both ends pinched. When the wave approaches a column end and leads to a nonequilibrium condition, the shaded area in Figure 1a will lose a small portion (out of the column); besides, the wave shape will usually be slightly distorted. Nevertheless, the wave velocity given by Eq. 3 is still a reasonable representation of the movement of the profile, and a zero velocity corresponds exactly to a standing wave of a steady state. Under the premise of constant molar flows and holdups, Marquardt and Amrhein (1994) recently presented a rigorous wave velocity equation with a term to account for the wave shape distortion. They confirmed that such a term is typically negligible for long columns. For a fractionating column with feeds and possibly side products, the entire profile consists of several waves, each in a section divided by side streams. Applied to the whole column, Eq. 3 represents an average movement of the entire profile, instead of a velocity of any individual wave. Such a holistic view is also valid for nonsharpening waves since an average movement is independent of wave shape.

The nonlinear wave theory for distillation dynamics has been built on the propagation of each shock wave in each column section divided by side streams (Hwang, 1991). For such a wave, if we allow for slight distortion of its shape, we can generally use Eq. 3 to calculate its wave velocity even when it approaches a column end, provided that we can obtain the information of the leaving streams. In practice, such information may be measured; for prediction, this can usually be estimated under certain assumptions. As in the previous treatment, the assumption of uniform flow rates supplied the leaving flow rates L' and V'' in Figure 1a. In addition, when a shock wave in a high-purity column is away from both ends, the assumption of pinched ends enabled us to calculate x' and y'' from y' and x'', respectively, using the vapor-liquid equilibrium relation. When the wave approaches a column end and leads to a nonequilibrium condition, we need a different approximation for the leaving stream composition at that end, which will be discussed below.

Column-End Effect and Asymmetric Dynamics

For a shock wave near a column end, we may say that Eq. 3 provides a generalized wave velocity representing the rate of an overall convective transport, which is a superposition of the column-end dissipation over the net equilibrium convective transport. For demonstrating the application of this generalized wave velocity to account for the column-end effect, the example illustrated by Figure 9 in Hwang (1991) for a rectifying column with uniform molar flow rates will be employed. In such a case, Eq. 3 can be reduced to Eq. 6 of Hwang (1991) in a normalized form (note the slight change of notation). For a shock wave near the top end at which a nonequilibrium condition ($x'' = y'' < 1$) occurs, its velocity at a specific instance can still be calculated with Eq. 3 if y'' is available. Unfortunately, y'' cannot be given *a priori* because it depends

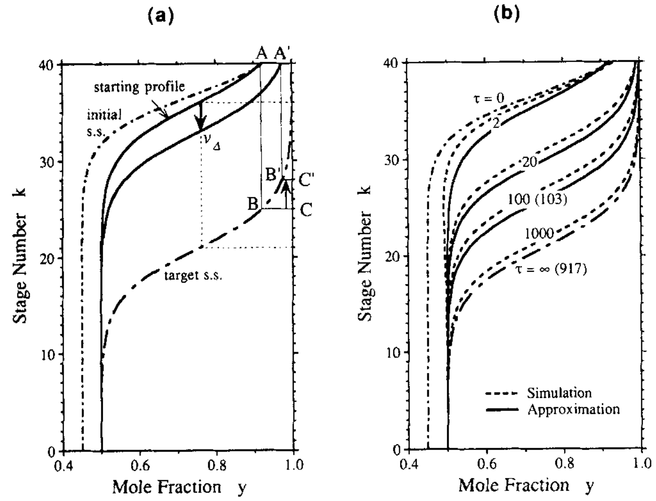

Figure 2. Constant-pattern wave approximation of slow return to balanced steady state.

(a) Computation procedure; (b) transient profiles vs. dynamic simulation (number in parentheses indicating τ for the approximation when it is not the same as for the simulation).

on the propagation of the wave. From a physical point of view, the wave propagation behavior near a column end involves both equilibrium convection and nonequilibrium dissipation mechanisms. Thus, the velocity computation needs to incorporate the latter in some way. The primary behavior resulting from the dissipation mechanism is the wave shape; the shock wave would be a step if there were no dissipation. Accordingly, a simple approximation may be developed by keeping the wave shape constant even when the wave is near a column end. Now, one can obtain y'' at a specific instance from the wave shape near the top end of the column. Since the slow return to the balanced steady state in the phenomenon of asymmetric dynamics is of more interest, such an approximation will be demonstrated quantitatively for the example shown by Figure 9b in Hwang (1991).

For that case, when the feed composition is changed back to that for the balanced steady state, the step change generates a disturbance wave, whose velocity can be calculated using the equilibrium information. It takes about two units in terms of normalized time τ for the disturbance to reach the top end and merge with the initial standing wave (steady-state profile) there. Assuming the resulting shock wave from that moment on retains a strictly constant shape during its travel, its shape should be the same as that of the target balanced standing wave, whose location may be designated by a representative composition (such as that on stage 21), as shown in Hwang (1991). Thus, if the balanced steady-state profile is available, for example, from a steady-state simulation or an operating record, the starting profile can be obtained by shifting the balanced profile upward (from stage 21 to 36) until the top-end composition matches that of the initial steady state, as illustrated by point A in Figure 2a. Using that composition in Eq. 3, the initial wave velocity at which the wave begins to travel downward can be estimated. With this initial condition, one can obtain the transient profiles by an integration of Eq. 3 with the constant wave shape, which can be expressed in terms of a table or a curve-fitting function. The top-end composition A on the starting profile corresponds to composition B (at stage 25 approximately) on the balanced steady-state profile. Point A moves toward unity as the wave travels downward. This is equivalent to the movement of point B along the balanced steady-state profile as one moves line BC upward at the same velocity, as illustrated by point A′ and line B′C′. Accordingly, moving line BC upward stage by stage (from 25 to 40) establishes a sequence of top-end composition. Using these compositions in Eq. 3 results in a sequence of wave velocities for the wave at a sequence of locations (from stage 36 to 21) with one stage apart. Assuming the wave travels between two adjacent locations at the average of the two instantaneous velocities, the wave propagation behavior can be easily constructed with a spreadsheet software, as shown by Table 1. Figure 2b shows the resulting transient profiles from this simple approximation along with those from the dynamic simulation

Table 1. Constant-Pattern Wave Approximation for Slow Return to Balanced Steady State in Rectifying Column

Target Steady State		Wave Velocity** (Normalized) u_Δ	Time Interval† $\tau_{k-1} - \tau_k$	Elapsed Time (Normalized) τ	Wave Location Stage k
Stage	y_{ss}				
25	0.9216*	−0.0331		2.0	36
26	0.9449	−0.0224	0.9	2.9	35
27	0.9619	−0.0151	1.3	4.2	34
28	0.9739	−0.0101	2.0	6.2	33
29	0.9823	−0.0068	3.0	9.2	32
30	0.9881	−0.0045	4.4	13.6	31
31	0.9920	−0.0030	6.7	20.3	30
32	0.9947	−0.0020	10.0	30.3	29
33	0.9965	−0.0013	15.1	45.4	28
34	0.9977	−0.0009	22.8	68.2	27
35	0.9985	−0.0006	34.6	102.8	26
36	0.9990	−0.0004	52.8	155.6	25
37	0.9994	−0.0002	81.5	237.0	24
38	0.9996	−0.0002	127.7	364.8	23
39	0.9998	−0.0001	205.6	570.4	22
40	0.9999	−0.0001	346.1	916.5	21

*The vapor mole fraction on tray 25 at the target balanced steady state is the closest to that at the top end (y'' = 0.9290) at the initial steady state.
**Normalized wave velocity u_Δ is calculated with $x'' = y'' = y_{ss}$ mapped to the top end and x' in equilibrium with y' (feed composition) at the bottom end.
†Time interval needed for the wave to travel one stage down is calculated assuming that the wave travels at a velocity $[(u_\Delta)_k + (u_\Delta)_{k-l}]/2$.

in Hwang (1991). With such a low computation cost, this constant-pattern wave approximation turns out to be fairly good for this example.

Balanced Steady State

As discussed previously (Hwang and Helfferich, 1988; Hwang, 1991), a high-purity column operated at a balanced steady state is optimally utilized with respect to the separation purpose. The term "balanced" means a balance between the *equilibrium* convective transport rates in the two opposite directions. This means that a balanced steady state of a high-purity distillation column is one with both ends pinched. A steady state with only one end pinched is unbalanced and usually has some room for optimization.

On the basis of the generalized wave velocity, any steady state has a zero wave velocity (standing wave). However, the zero wave velocity of a balanced steady state requires equilibrium conditions at both column ends. This clarifies the use of a zero wave velocity to determine the operating condition of a balanced steady state. With the inclusion of feed and product side streams, Eq. 3 can be applied to fractionating columns possibly with side products for determining a balanced operating condition. Although this seems to be trivial for binary fractionation because of the trivial dual-pinch criterion, it is anticipated to be useful for multicomponent distillation.

Column with Nonuniform Flow Rates

The molar flow rates in a distillation column may vary with location if the heats of vaporization of the components are significantly different, or if the column has a considerable heat loss or some intermediate heat inputs. For such a situation, the generalized wave velocity equation 3 is still applicable, but the leaving stream flow rates L' and V'' need to be either measured or calculated. For calculation, the integral overall material balance and energy balance result in the following two shock wave velocities:

$$v_{\Delta W}=v_{\Delta U}=\frac{(V''-L'')-(V'-L')-F+M}{(W''+U'')-(W'+U')} \qquad (4)$$

$$v_{\Delta h}=v_{\Delta H}=\frac{(V''H''-L''h'')-(V'H'-L'h')-FE_F+ME_M-Q}{(W''h''+U''H'')-(W'h'+U'H')} \qquad (5)$$

Consider a column section with no side streams and with given information of the entering streams and the heat input (or loss) Q. In addition, assume that enthalpies and molar holdups are functions of compositions; in other words, h, H, W, and U can be algebraically determined from x and y at any instance at any location. An example case is if one assumes that enthalpies depend only on compositions and temperature, and volume holdups of both liquid and vapor are constant. Under such premises, there exists a unique wave traveling with a velocity which can be tracked in terms of either composition, enthalpy, or holdup:

$$v_{\Delta x}=v_{\Delta h}=v_{\Delta W} \qquad (6)$$

For a shock wave away from the two ends which are pinched, the equilibrium conditions at the two ends supply the leaving stream compositions x' and y''. These in term give h', W', H'' and U''. For a shock wave near a column end with a nonequilibrium condition, these values can be obtained by applying the constant-pattern wave approximation discussed above. Then, the only two unknowns L' and V'' can be solved using Eqs. 6, and the wave velocity can be calculated with Eq. 3, 4 or 5.

For a column for which the above assumptions cannot be justified, it has not yet been clear so far whether multiple waves may exist or not. Without any mathematical proof, an intuitive conjecture suggests that this is unlikely to occur for the reason that compositions, enthalpies, and holdups are physically related to one another, even though the dependency may require the information not only at a specific location but also in its neighborhood. Accordingly, these variables (x, h, and W) are not truly independent like the set of compositions in a multicomponent system (except the summation relation). However, an extensive discussion is beyond the scope of this article.

Notation

E = two-phase overall molar enthalpy
F = molar flow rate of feed stream
h = liquid molar enthalpy
H = vapor molar enthalpy
k = index of equilibrium stage counted from bottom
L = molar flow rate of liquid stream
M = molar flow rate of side product stream
Q = net heat input rate (negative value for heat loss)
s = distance from bottom in terms of length or number of trays
t = time
u_Δ = normalized shock wave velocity (Hwang, 1991)
U = molar vapor holdup per unit length or per tray
v_Δ = shock wave velocity
V = molar flow rate of vapor stream
W = molar liquid holdup per unit length or per tray
x, y = liquid and vapor mole fraction of light component, respectively
z = two-phase overall mole fractions of light component

Greek letters

Δ = prefix for difference between two sides of shock wave
τ = normalized time (Hwang, 1991)

Subscripts and Superscripts

F = feed
M = side product
$'$ = superscript denoting bottom end of column section
$''$ = superscript denoting top end of column section

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