Nonlinear Waves and Asymmetric Dynamics of Countercurrent Separation Processes

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Continuous countercurrent separation processes have been widely used in chemical and related industries for a long time. A well-recognized difficulty in controlling such processes is the nonlinearity of their dynamic behavior. Stemming from nonlinearity, an intriguing phenomenon known as "asymmetric dynamics" was observed by several investigators (Rose et al., 1956; Moczek et al., 1965; Mizuno et al., 1972; De Lorenzo et al., 1972; Fuentes and Luyben, 1983; Stathaki et al., 1985) in their numerical simulation of distillation columns. This term refers to the difference in response times either for a steady state perturbed by a pair of step changes of the same magnitude but in opposite directions, or for the forward and reverse transitions between two steady states. The latter type of asymmetry is of more interest for countercurrent processes. In particular, it was found that the transition departing from the optimal steady state (in the sense of maximum separation) is always faster than the corresponding transition returning to it (De Lorenzo et al., 1972; Stathaki et al., 1985). The difference is usually drastic (Rose et al., 1956; Mizuno et al., 1972). This note aims at providing a cause-and-effect analysis of such behavior by using the concept of wave propagation.

Dynamic asymmetry emerges from nonlinearity of a process. Although nonlinearity of countercurrent processes may arise from complex configurations, its major cause is the nonlinearity of equilibrium between the two phases. Only a few studies to date have included the effect of nonlinear equilibrium on countercurrent dynamics. Mohr (1965) examined the effect of curvature of the equilibrium curve on the response time in binary distillation. Fuentes and Luyben (1983) showed severe dynamic nonlinearity in long distillation columns producing high-purity products, but did not identify the cause.

To exploit nonlinear wave theory, we employ a differential rather than stagewise model because it gives a more readily visualized description of wave propagation. Especially for long columns (large number of transfer units), the differential model can provide a close approximation even to plate columns, and with comparable or less mathematical complexity. Specifically, the differential model developed previously (Hwang, 1987; Hwang and Helfferich, 1988 and 1989) is used here to show that asymmetric dynamics in countercurrent operation is a natural consequence of nonlinear wave behavior. In addition, this analysis reveals a monitoring problem and suggests some control strategies for countercurrent processes.

Model

Considered here is a stripping column, which resembles either section of a fractionating column, but without reflux and reboil. For mathematical demonstration, this stripper is described by a simple differential model which assumes a single-solute system (equivalent to binary distillation), negligible axial dispersion, constant flow rates and holdups, and a constant mass-transfer coefficient based on overall resistance across both phases. However, the effect of axial dispersion and the responses to flow disturbances are discussed. Furthermore, this approach can be applied to multicomponent systems by incorporating our earlier work on such systems (Hwang and Helfferich, 1989).

In terms of normalized quantities, the governing equations of the simple model are:

$$\begin{cases}(\partial x/\partial \tau) + (\partial x/\partial \zeta) = -rsN[f(x) - y] \\ (\partial y/\partial \tau) - s(\partial y/\partial \zeta) = sN[f(x) - y]\end{cases} \tag{1}$$

The initial condition is the initial steady-state profile, $x^o(\zeta)$ and $y^o(\zeta)$, with constant entering-stream concentrations: $x^o(0) = a$ and $y^o(1) = b$. For an x-feed disturbance, as considered here, the boundary condition is $x(0,\tau) = a \pm \delta a$ and $y(1,\tau) = b$. The analysis is equally applicable to responses to y-phase disturbances.

As demonstrated previously (Hwang and Helfferich, 1988), the wave behavior of such a stripper can be qualitatively predicted by the following equilibrium approximation:

$$(\partial x/\partial \tau) + v(x)(\partial x/\partial \zeta) = 0 \quad y = f(x) \tag{2}$$

where the "eigenvelocity," v, depends on the equilibrium relation as follows:

$$v(x) \equiv (\partial\zeta/\partial\tau)_x = [1 - rs(df/dx)]/[1 + r(df/dx)] \quad (3)$$

A wave may be "self-sharpening" or "nonsharpening" depending on whether the eigenvelocity decreases or increases with composition in the ζ direction. Under local equilibrium, a self-sharpening wave either remains, or eventually becomes a discontinuous "step" with the following "step velocity":

$$v_\Delta(x'|x'') \equiv (\partial\zeta/\partial\tau)_\Delta = [1 - rs(\Delta y/\Delta x)]/[1 + r(\Delta y/\Delta x)] \quad (4)$$

where $\Delta x \equiv x' - x''$ while x' and x'' are the values on the two sides, respectively, of the step in ζ direction (rather than in the propagating direction).

As discussed in our earlier work, the travel of a wave in a countercurrent column is driven by its intrinsic (equilibrium) propagating tendency characterized by its eigenvelocity or step velocity, as well as by the asymmetric dissipation caused by the column-end effect owing to a finite mass-transfer rate (nonequilibrium). The intrinsic tendency dominates wave propagation, except near column ends, where net dissipation pushes the wave inward, preventing it from leaving the column. The longer the column in terms of transfer units, the less prominent is the dissipative end effect. If axial dispersion is taken into account, its effect is nothing more than superimposing of similar dissipative column-end effect on that resulting from a finite mass-transfer rate, and is usually weaker than the latter.

Nonlinear Wave Propagation

For comparison with the asymmetric dynamics of distillation columns observed by others, our discussion is limited to equilibrium curves concave toward the operating line, as is typical for distillation. Wave propagation behavior with such type of equilibrium can be clearly illustrated by start-up of a stripper, in which the wave is always self-sharpening (as are those in distillation columns). The wave may travel in either direction or tend to stand still, depending on its step velocity, v_Δ. A wave with positive v_Δ tends to travel forward in the x-stream direction (called "x-favored") until this intrinsic tendency is counteracted by the backward (net) dissipation near the x-stream exit. On the other hand, a wave with negative v_Δ tends to propagate in the y-stream direction ("y-favored") and thus not to enter the column, but is pushed into the column for a short distance by dissipation. In contrast, a wave with zero v_Δ has no intrinsic tendency to travel ("balanced" wave); it is driven into the column by the asymmetric dissipation exclusively, and eventually settles in the middle portion where dissipation becomes symmetric.

For illustration, our analysis will be supplemented by an example of a stripping operation with the following equilibrium relation, having the same form as for binary distillation with constant relative volatility:

$$f(x) = \alpha x/[1 + (\alpha - 1)x] \quad (5)$$

In this example, a solute-free gas ($b = 0$) is used to process liquid feeds with various steady-state concentrations, a. For each case, Eq. 1 is solved at both transient and steady states, using the numerical techniques presented previously (Hwang and Helferich, 1988), with the same parameter values: $\alpha = 5$, $r = 0.1$, $s = 3$, and $N = 30$, in all cases.

Steady State and Its Sensitivity

The start-up described above leads to a steady state whose character depends on the operating conditions, as implied by Eq. 4. The best separation requires convective transport of the two countercurrent streams to be balanced so as to utilize the column as completely as possible. Such an optimal steady state results from a "balanced" wave (with zero step velocity).

Around the optimal steady-state condition, however, the column is very sensitive to the feed composition and flow rate. For the optimal steady state, the wave has zero step velocity and so naturally remains stagnant. However, any perturbation of feed composition or flow rate causes an unbalance in convective transport and so leads to a nonzero step velocity, which drives the wave toward one column end until the intrinsic driving force is counteracted by the inward (net) dissipation near that end. In a column with a large number of transfer units, the middle portion with weak dissipative end effect is relatively long; therefore, even a small perturbation will produce a large wave shift. This also explains the high sensitivity of distillation columns to feed composition observed by Stathaki et al. (1985) and Kapoor et al. (1986).

For quantitative illustration, Figure 1a presents steady-state profiles for the stripper, the optimal steady state being denoted by a letter O. These profiles can be viewed as "standing waves," each characterized by a step velocity, $v_\Delta(a|0)$, indicating its intrinsic moving tendency. This velocity is plotted as a function of a in Figure 1b. For convenience, the location where such a wave stands can be represented by a "stagnation point" ζ_s

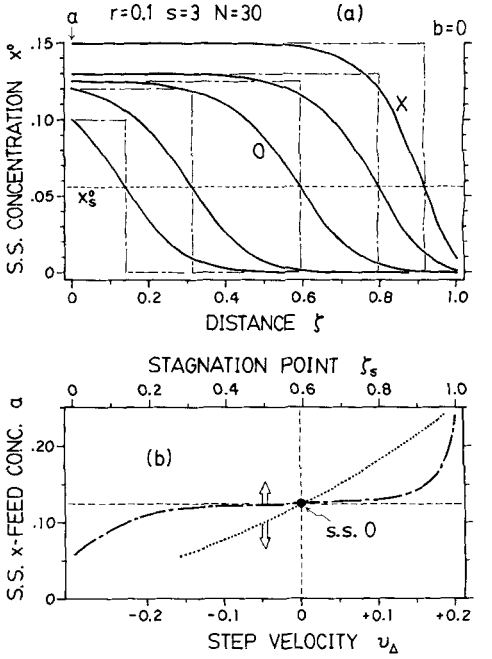

Figure 1. Steady states of stripping column.
a. Liquid concentration profiles
b. Stagnation point and step velocity vs. liquid feed concentration

(Hwang and Helfferich, 1988) defined as:

$$df/dx = 1/rs \quad \text{at } x_s^o \equiv x^o(\zeta_s) \tag{6}$$

Figure 1b includes a relation of ζ_s vs. a to illustrate the high sensitivity around the optimal condition.

Asymmetric Dynamics

With this picture of nonlinear waves, cause and effect of asymmetric dynamics and the dependence on system variables become apparent. Consider the transition departing from the optimal steady state, which is "balanced" and stands in the middle portion of the column. A disturbance in the feed composition (or flow rate) raises a disturbance wave, which soon catches the standing wave and merges into it, upsetting the balance in convective transport. The resulting wave is driven by its nonzero step velocity toward one column end until impeded by the inward (net) dissipation near that end. As can be deduced from our earlier work, the time needed for the disturbance wave to reach the standing wave, and the ensuing interference, amounts to merely a small portion of the entire response; thus, the transition time is essentially governed by the step velocity of the resulting wave corresponding to the eventual steady state.

For the transition returning to the optimal steady state, the restoring disturbance wave merges into the nonoptimal standing wave near the column end and results in the original "balanced" wave with zero step velocity. Without the contribution of the intrinsic driving force, the dissipative column-end effect alone has to move this wave back to its original position in the middle of the column, and this effect fades as the wave is pushed away from the column end. This explains why the return to the optimal steady state is extremely slow. It furthermore clarifies that such dynamic asymmetry will be most pronounced if the number of transfer units is large (dissipative end effect weak in a large portion of the column) and equilibrium is highly nonlinear (strong variation of step velocity with feed concentration).

For quantitative demonstration, consider the transition from the optimal steady state with $a = 0.125$ (O), to one with $a = 0.15$ (X), and the return to the optimal steady state. Figure 2 shows the slow returning transition in terms of the concentration histories at the x-stream exit, and the stagnation points of the two steady states, respectively. To demonstrate the nonlinear wave behavior in more detail, Figure 3 presents transient concentration profiles. The wave pattern in each transition is governed by step velocities (and eigenvelocities) as shown below. For the transition from steady state O to X, illustrated by Figure 3a, the disturbance wave is self-sharpening with a step velocity, $v_\Delta(0.15|0.125) = 0.31$. It soon merges into standing wave, O, at $\zeta_s = 0.59$; this results in an x-favored wave traveling virtually at its step velocity, $v_\Delta(0.15|0) = 0.048$, until impeded by the dissipative end effect, and eventually settles at $\zeta_s = 0.92$. For the returning transition, shown by Figure 3b, the disturbance step leads to a nonsharpening wave traveling with an eigenvelocity ranging from $v(0.125) = 0.27$ to $v(0.15) = 0.35$. This wave merges into standing wave X, at $\zeta_s = 0.92$, resulting in a wave with the same concentration levels as for steady state O. Thus, the resulting wave has zero step velocity, $v_\Delta(0.125|0) = 0$, and is driven back toward its original position ($\zeta_s = 0.59$) by the dissipative end effect alone. As the wave moves away from the column end, dissipation rapidly becomes more symmetric (and thus weaker), and the wave slows down.

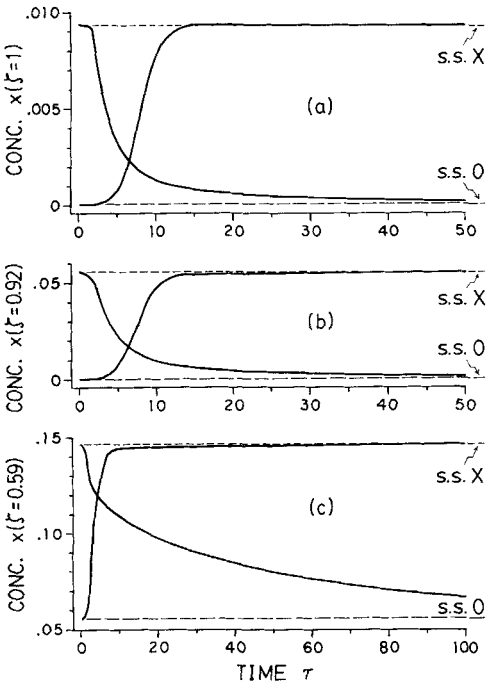

Figure 2. Asymmetric dynamics of stripping column illustrated by liquid concentration histories for transitions between steady states *O* and *X*.
a. At liquid exit
b. At stagnation point of steady state X
c. At stagnation point of steady state O

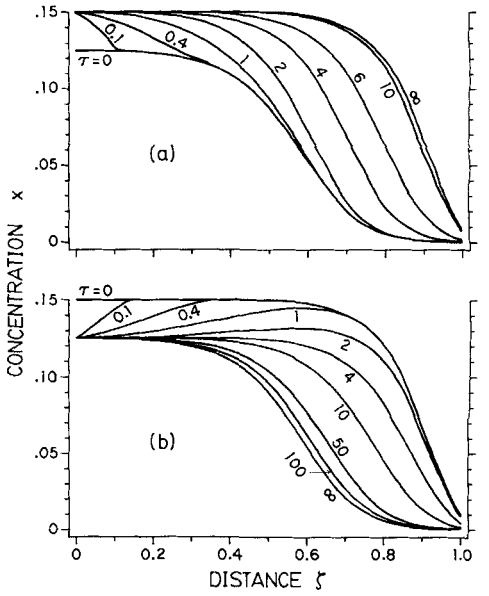

Figure 3. Asymmetric dynamics of stripping column demonstrated by nonlinear wave propagation.
a. Transition departing from optimal steady state
b. Slow returning transition

In addition to asymmetric dynamics, Figures 2 and 3b reveal a so far unrecognized monitoring problem: the effluent composition returns to the optimal steady state more quickly than does the entire composition profile (compare Figures 2a and 2c, noting different time scales). This implies that sensors at the column ends tend to give a premature signal for a return to the optimal steady state (see profiles in Figure 3b), and may therefore lead to erroneous predictions of responses to subsequent disturbances. [A superficially similar problem arises in a countercurrent adsorber-reactor, and was discussed in detail by Viswanathan and Aris (1974) as well as Rhee et al. (1986) who assumed local equilibrium to be maintained. Unfortunately, this is not possible here as the monitoring problem arises from a nonequilibrium effect.]

Propositions for Control

Based on the step velocity, some control strategies can be proposed to combat excursions from the optimal steady state most effectively. For a regulation problem in response to feed composition disturbances, a feedforward controller may adjust the flow ratio to maintain zero step velocity, according to Eq. 4. More interesting is the application to a servo-problem. Supposing the optimal condition has to be restored from a nonoptimal steady state, say, an x-favored one, a better maneuver than adjusting the flow ratio directly to the "balanced" condition is a two-step sequence: first employ a higher flow ratio to generate a y-favored wave in order to speed up the returning transition, and estimate with its step velocity the time needed to reach the target position; then, at that time restore the "balanced" flow ratio.

Notation

a = steady-state x-feed normalized concentration, $a = x^o(0)$
b = steady-state y-feed normalized concentration, $b = y^o(1)$
f = function representing equilibrium relation: $y = f(x)$
N = number of transfer units based on overall-resistance mass-transfer coefficient in terms of y phase
r = mass holdup ratio of y to x phase
s = linear velocity (mass velocity divided by mass holdup) ratio of y to x phase
x, x^o = transient and steady-state normalized concentration of x phase (liquid)
x_s^o = value of x^o at stagnation point, ζ_s
y, y^o = transient and steady-state normalized concentration of y phase (gas)

Greek letters

α = parameter in equilibrium relation, ~ relative volatility
ζ = normalized distance from x-phase entrance
ζ_s = position (ζ value) of stagnation point on steady-state concentration profile
τ = normalized time
v, v_Δ = normalized eigenvelocity and step velocity under local equilibrium

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