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An Analogy between Countercurrent and Two-Dimensional Separation Cascades

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Abstract

The relationships between countercurrent fixed bed and two-dimensional cascades are explored for continuous contact and for staged systems. With moving feed and product ports, fixed beds will simulate countercurrent operation. Two-dimensional cascades will simulate countercurrent operation if slanted feed and product lines are employed. The relationships are also extended to three-phase systems for one-, two-, and three-dimensional cascades. Fixed bed systems simulating countercurrent systems are in commercial operation. The two-dimensional cascade offers an alternative geometry for obtaining the same result. This alternative may be attractive for some separations such as electrophoresis.

INTRODUCTION

The relationships between different types of separation cascades have been sporadically studied since at least 1841 when the Shanks system for simulating countercurrent motion was introduced in England (*1*). Recent interest has included industrial applications of simulating countercurrent motion in fixed beds (*2*, *3*), laboratory simulation of countercurrent motion for gas chromatography (*4*) and for gel permeation chromatography (*5*), and the mathematical relationship between unsteady one-dimensional systems and steady-state two-dimensional cascades (*6*, *7*). Hybrid systems which simulate countercurrent development during feed and utilize chromatographic development during the remainder of the cycle have also been studied (*8*, *9*). Morphological relations have also been employed as one-dimension in a general scheme for classifying and developing separation method (*10*).

In this paper we will extend the existing analogies and develop the conditions for which two-dimensional cascades produce the same separation as countercurrent cascades. This will be done first for continuous-contact systems and then for staged systems. Then the analogy will be extended to three-phase separation systems, and possible ramifications of the analogy will be discussed.

CONTINUOUS CONTACT SYSTEMS

The analogy between one-dimensional and two-dimensional cascades has been mathematically delineated (*7*). This was done for the two-dimensional case where there is vertical fluid flow through a packed annulus and the annulus rotates. That analysis will be briefly repeated here.

The usual form of the solute balance for adsorption or chromatography in a packed column assumes plug flow and ignores radial gradients in velocity, concentration, and temperature. This balance for nonreacting systems is

$$\varepsilon \frac{\partial c}{\partial t} + \varepsilon v \frac{\partial c}{\partial z} + (1 - \varepsilon)\rho_s \frac{\partial q}{\partial t} - D \frac{\partial^2 c}{\partial z^2} = 0 \tag{1}$$

The solute balance on the solid phase is

$$\rho_s(1 - \varepsilon) \frac{\partial q}{\partial t} = k_M a(c - c^\circ) \tag{2}$$

At equilibrium, the solid and fluid concentrations are related by an equilibrium expression of the general form

$$q = q(T, c^\circ) \tag{3}$$

These equations can be compared to the equations for steady-state operation in a two-dimensional rotating annulus system. Radial gradients are again assumed to be negligible, and the resulting solute balance in cylindrical coordinates is

$$\varepsilon w \frac{\partial c}{\partial \theta} + \varepsilon v \frac{\partial c}{\partial z} + (1 - \varepsilon)\rho_s w \frac{\partial q}{\partial \theta} - D \frac{\partial^2 c}{\partial z^2} - D \frac{1}{r^2} \frac{\partial^2 c}{\partial \theta^2} = 0 \tag{4}$$

The rotation of the annulus results in a solid body rotation of both phases. The solute balance on the solid phase for the steady-state rotating system is

$$(1 - \varepsilon)\rho_s w \frac{\partial q}{\partial \theta} = k_M a(c - c^\circ) \tag{5}$$

The equilibrium expression is the same as Eq. (3).

Comparison of Eq. (1) to Eq. (4) shows that there is an extra diffusional term in the latter equation. Under conditions where diffusion is important, the one- and two-dimensional systems are not analogous. However, the diffusional terms can commonly be ignored, at least for a first approximation. Under the conditions of negligible diffusion and dispersion, there is a term-by-term correspondence between Eqs. (1) and (4), and (2) and (5). If the simple transformation

$$t \rightarrow \theta/w \tag{6}$$

is made, then Eqs. (1) and (2), are transformed into Eqs. (4) and (5), respectively, for the case with negligible diffusion. As long as q is not a function of t or θ, this transformation is valid regardless of the equilibrium relationship, Eq. (3), which is used. Equation (3) is unaffected by the transformation.

In order for the systems to be analogous, the boundary conditions must also transform under Eq. (6). The necessary boundary conditions for a two-dimensional system to be equivalent to a countercurrent system will be explored shortly.

Equation (1) was written for a fixed bed. However, the equation will also be valid (with appropriate changes in variables) if countercurrent operation is used and the reference frame moves with the solid. To transform to a fixed (z', t') reference frame, let

$$\begin{aligned} z' &= z - v_s t - nL \\ t' &= t, \qquad n = 0, 1, 2, \ldots, \\ 0 &\leq z' \leq L \end{aligned} \tag{7}$$

where z' is positive in the direction of fluid flow and the solid flows in the negative z' direction with velocity v_s ($v_s > 0$). Utilizing the chain rule, Eq. (1) becomes

$$-v_s \frac{\partial c}{\partial z'} + \frac{\partial c}{\partial t'} + v \frac{\partial c}{\partial z'} + \frac{\rho_s(1 - \varepsilon)}{\varepsilon}\left[- v_s \frac{\partial q}{\partial z'} + \frac{\partial q}{\partial t'}\right] = 0 \tag{8}$$

If we note that $v_F = v - v_s$ is the fluid velocity in our new reference frame and apply steady-state conditions,

$$\frac{\partial c}{\partial t'} = \frac{\partial q}{\partial t'} = 0 \tag{9}$$

Eq. (8) simplifies to

$$\frac{\partial c}{\partial z'} v_F = \frac{\rho_s(1 - \varepsilon)}{\varepsilon} v_s \frac{\partial q}{\partial z'} \tag{10}$$

This is exactly the equation which is derived for a steady state countercurrent process.

This development shows that the correct countercurrent equations are obtained if we first model the countercurrent system as a fixed bed with a moving reference frame and then transform our reference frame. The process can also be reversed. That is, a fixed bed process can simulate a countercurrent process (*2*, *3*) by using a transformation like Eq. (7) *and* by utilizing boundary conditions which will also transform. Since the development from Eqs. (1) to (6) showed that fixed beds and two-dimensional processes can give equivalent separations, it follows from the sequence:

countercurrent → fixed bed → two-dimensional

that there is a two-dimensional arrangement which will give the same separation as a countercurrent separator. What is required is an arrangement which will give boundary conditions which transform properly.

To look at boundary conditions, consider the adsorption system shown in Fig. 1(a) (*2*, *3*). For a countercurrent apparatus separating Components A and B using Desorbent D, the boundary conditions are

$$\begin{aligned} z' &= 0, & q_B &= 0 \text{ (or small specified value)} \\ z' &= z_B', & c_A &= c_{AB-\text{Prod}} \\ z' &= z_A', & c_B &= c_{BA-\text{Prod}} \\ z' &= L, & c_A &= 0 \text{ (or small specified value)} \end{aligned} \tag{11}$$

Also the inlet concentrations are set:

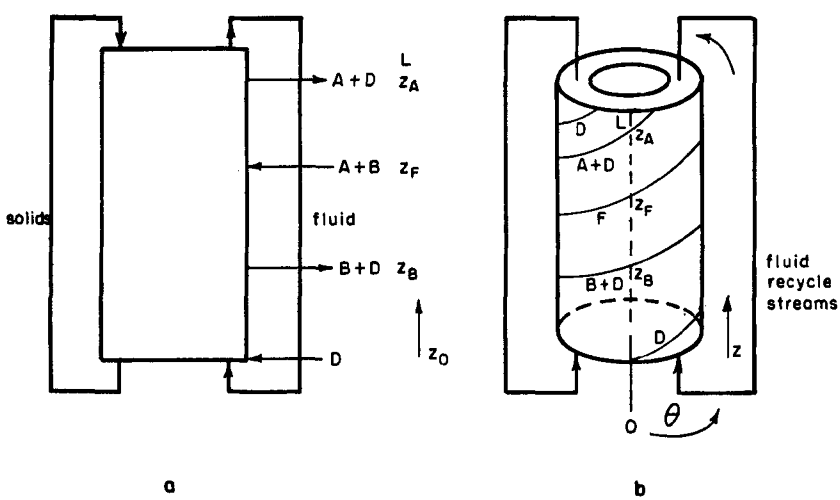

FIG. 1(a). Countercurrent cascade utilizing desorbent (*2*). (b) Equivalent two-dimensional cascade.

$$z' = 0, \qquad c_{\text{IN}} = c_D$$
$$z' = z_F', \qquad c_{\text{IN}} = c_F \tag{12}$$

To obtain the same separation in a fixed bed, we can move the locations of the inlet and outlet ports upward at a switching velocity v_s (*2, 3*). If the fluid velocity is v, the fluid velocity relative to the ports is $v_F = v - v_s$ and the solids velocity relative to the ports is $-v_s$. If we now have our reference frame move with the ports, we have

$$z'' = z' + v_s t - nL \tag{13}$$

where n = integer 0, 1, 2, ..., and $0 \leq z'' \leq L$, $t'' = t$.

The boundary and inlet conditions written in Eqs. (11) and (12) are valid if written in the z'' coordinate system. Equations (13) are essentially the same as Eqs. (7), and the fixed bed mass balances will transform over to the appropriate countercurrent equations. Since the basic equations and the boundary conditions both transform over, the two systems give equivalent separations. This is utilized in the commercial simulated countercurrent process (*2, 3*).

The two-dimensional system shown in Fig. 1(b) will give separations equivalent to the separations obtained in the countercurrent apparatus shown in Fig. 1(a). In Fig. 1(b) the inlet and outlet streams are run along slanted parallel lines in a manner similar to the slanted feed proposal for two-dimensional separators (*9*). The boundary conditions for this two-dimensional apparatus are

$$z' = 0 + S\theta, \qquad q_B \text{ small}$$
$$z' = z_B + S\theta - nL, \qquad c_A = c_{AB-\text{Prod}}$$
$$z' = z_A + S\theta - nL, \qquad c_B = c_{BA-\text{Prod}}$$
$$z' = L + S\theta - nL, \qquad c_A \text{ small} \tag{14}$$

where S is the slope of the feed and withdrawal lines, $0 \leq \theta \leq 360°$, and n is 0 or 1 as required to make $0 \leq z' \leq L$. The inlet conditions are set:

$$z' = 0° + S\theta, \qquad c_{\text{IN}} = c_D$$
$$z' = z_F + S\theta - nL, \qquad c_{\text{IN}} = c_F \tag{15}$$

We will now make the transformation in Eq. (6), $t = \theta/w$, and the transformation

$$z'' = z' - (Sw)t - nL \tag{16}$$

which is the same as Eq. (13) with

$$v_s = Sw \tag{17}$$

This transformation converts the boundary and inlet conditions in Eqs. (14) and (15) to the countercurrent boundary conditions in Eqs. (11) and (12). The solute mass balance, Eq. (4), will first be transformed to the mass balance for a fixed bed, Eq. (1), assuming that dispersion and diffusion can be neglected. The second transformation, along with the steady-state requirements $\partial q/\partial t = \partial c/\partial t = 0$, converts this to

$$\frac{\partial c}{\partial z''} v_F = \frac{\rho_s(1 - \varepsilon)}{\varepsilon} v_s \frac{\partial q}{\partial z''} \tag{18}$$

which is the equation for a steady-state countercurrent system. Since both boundary conditions and the solute mass balance transform into the countercurrent equations, the steady-state, two-dimensional system shown in Fig. 1(b) develops the same separation as a countercurrent process. This analogy is subject to the restriction that dispersion and diffusion are negligible, and refers to the (z'', t) coordinate system. This is also true in the (z'', θ) coordinate system if z'' is defined as

$$z'' = z' - S\theta - nL \tag{19}$$

Another way of saying this is if we use the same values of z_A', z_B', z_F', L, and v_F, and force Eq. (17) to be satisfied, then the apparatus in Fig. 1(b) (assuming it could be built and operated) will give the same separation (same product concentrations) as the countercurrent system shown in Fig. 1(a). The argument presented is not restricted to fluid–solid adsorption, and can be easily extended to other two-phase systems. Also, the argument can be extended to systems with reflux. The generalized definition of reflux is to withdraw one phase, change it to the other phase, and return part of the stream at the same location. This is hard to conceive of in an adsorption system. If the two-dimensional system were distillation with the fluid rotating, the reflux would consist of withdrawing upward flowing vapor, condensing the vapor, and returning part of it to the separator. The only difference between this reflux and countercurrent reflux is that now the refluxed liquid flows horizontally instead of downward.

STAGED SYSTEMS

Two-dimensional staged systems have been studied for continuous multicomponent separations and two-dimensional developments (see Ref. *6* for a review). These two-dimensional staged systems can also produce the same separation as a countercurrent cascade if the feed and product lines are arranged on slanted lines. This is illustrated in Fig. 2 for an extraction system.

Before writing the mass balances for this system, we need to consider the

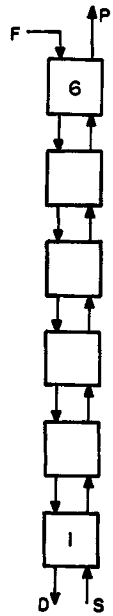

FIG. 2A. Staged countercurrent extraction cascade.

symmetry of the cascade shown in Fig. 2B. At steady-state all stages labeled 1 will have the same concentrations (x_F is constant) and y_P and x_1 will be the same for all stages labeled 1. This is also true for all stages labeled 2, 3, etc. This occurs since all stages labeled 1 have exactly the same environment. This property that all stages on a given diagonal have the same concentration is crucial in developing the mass balances.

If we write a mass balance for one of the solutes around Stage i and solve for y_i we obtain

$$y_i = \frac{L}{V}x_{i+1} + y_{i-1} - \frac{L}{V}x_i \tag{20}$$

This equation is the same equation as one obtains for a countercurrent cascade with y_i and x_{i+1} being the passing streams and y_{i-1} being input from the stage below. Usually the mass balances are written around the top or bottom of the column. For the two-dimensional system this is illustrated in Fig. 2A for the specific case where the material balance envelope is drawn from Stage 3 around the top (Stage 6) of the column. The material balance is drawn as if the stages in Fig. 2 repeat themselves indefinitely. The recycle streams are equivalent to repeating the stages. When the stages are continued, the material balance envelopes A–A, B–B

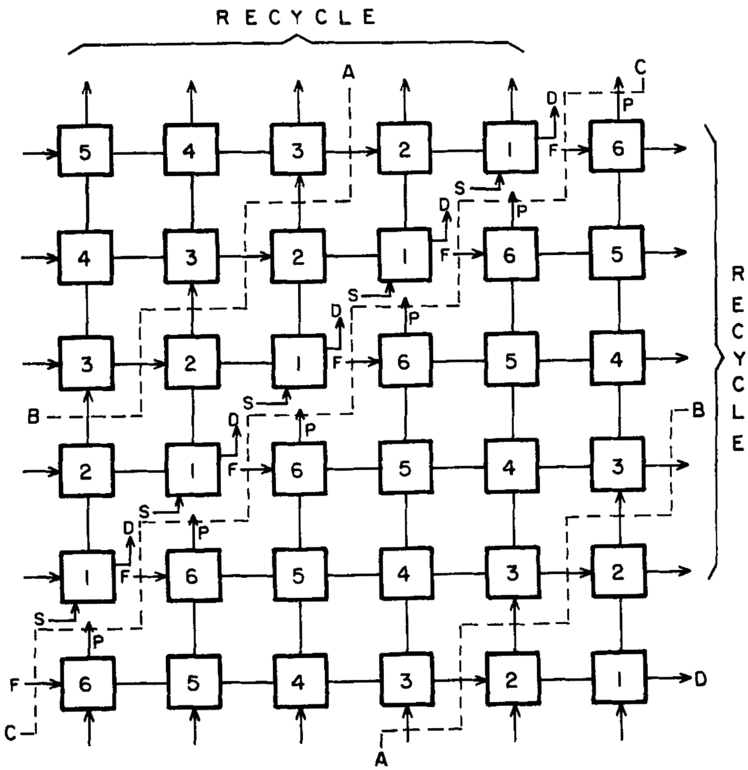

FIG. 2B. Equivalent two-dimensional staged cascade.

and C–C would be connected. The material balance envelope can be closed by going from C to A on the bottom row, from B to C on the right-hand column, and from A around the upper left-hand corner to B. Adding these two regions, the mass balance is

$$V_5y_5 + V_4y_4 + V_3y_3 + \sum_{j=1}^{N} L_3x_3 + \sum_{j=1}^{N} Py_P$$
$$= V_5y_5 + V_4y_4 + V_3y_3 + \sum_{j=1}^{N} Fx_F + \sum_{j=1}^{N} V_2y_2 \quad (21)$$

where the summations sum terms on each diagonal. The recycle streams cancel and each summation becomes N times the term inside the sum ($\sum (L_iX_i) = NL_ix_i$). Upon rearrangement, Eq. (21) becomes

$$y = \frac{L_3}{V_4}x_3 + \frac{P}{V_4}y_P - \frac{F}{V_4}x_F \quad (22)$$

If the flow rates are constant and $F = L$ and $P = V$, then Eq. (22) when generalized is

$$y_{i+1} = \frac{L}{V}x_i + y_P - \frac{F}{V}x_F \tag{23}$$

Equation (23) will also be obtained if a mass balance is written for the countercurrent system shown in Fig. 2A.

Since the mass balances are the same, the cascades shown in Figs. 2A and 2B will produce the same separation. Graphical, analytical, and numerical methods of solving the countercurrent equations (*1*) can be applied to the two-dimensional cascade shown in Fig. 2B. Usually the stages will be treated as being equilibrium stages.

The two-dimensional system with slanted feed and product lines can easily be extended to more complex columns with or without reflux. The two-dimensional cascades are restricted in that they must have the feed and product lines slanted at a 45° angle and the cascades must be square. If a countercurrent cascade required 10 stages, a 10 × 10 two-dimensional cascade or 100 stages would be required. However, for equal size stages the two-dimensional cascade has a capacity 10 times that of the countercurrent cascade. Thus the capacity/stage is the same for the countercurrent and the two-dimensional cascades.

THREE-PHASE STAGED SYSTEMS

Three-phase separations have not been extensively studied, but they do occur. Examples are distillation of organics when water is present, slurry adsorption, and liquid membrane separators. A countercurrent system with two of the phases moving cocurrently is usually employed. This is illustrated in Fig. 3A. The mass balance for Stage i is

$$y_i = \frac{W_{i+1}}{V_i}z_{i+1} + \frac{L_{i+1}}{V_i}x_{i+1} + \frac{V_{i-1}}{V_i}y_{i-1} - \frac{W_i}{V_i}z_i - \frac{L_i}{V_i}x_i \tag{24}$$

The balance around the bottom of the column is

$$y_i = \frac{W}{V}z_{i+1} + \frac{L}{V}x_{i+1} + \left(y_0 - \frac{L}{V}x_1 - \frac{W}{V}z_1\right) \tag{25}$$

where constant flow rates have been assumed. In limiting cases, solutions for the countercurrent three-phase separators are easily developed (*12*).

The same result can be obtained in the two-dimensional cascade shown in Fig. 3B where slanted feed and product lines are used to make every stage on Diagonal i the same. Then the mass balance around Stage i is given by Eq. (24). If the mass balance is done around the stages where L

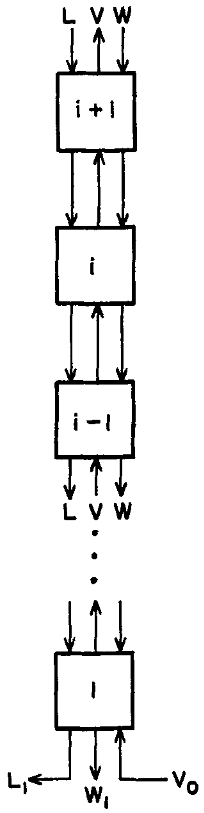

FIG. 3A. Staged "countercurrent" cascades for three-phase systems: One-dimensional.

and W exit the cascade and the procedure used in Eqs. (21) to (23) is followed, Eq. (25) results. Thus the cascades shown in Figs. 3A and 3B give the same results.

Three-phase systems can also be operated in a three-dimensional cascade of stages (*11*) as shown in Fig. 3C. The three-dimensional cascade can give the same separation as a countercurrent cascade if feed is input and product streams are withdrawn in parallel planes. Planes parallel to the plane through Stages A–B–C or through Stages A–B–D can be used for the feed and product planes. The stages in Fig. 3C are labeled for the case where the A–B–D planes are used. With all Stages i being the same, the mass balance around Stage i (either A, B, or D) gives Eq. (24), and a balance around stages with L and W exiting the cascade will give Eq. (25) after suitable manipulation.

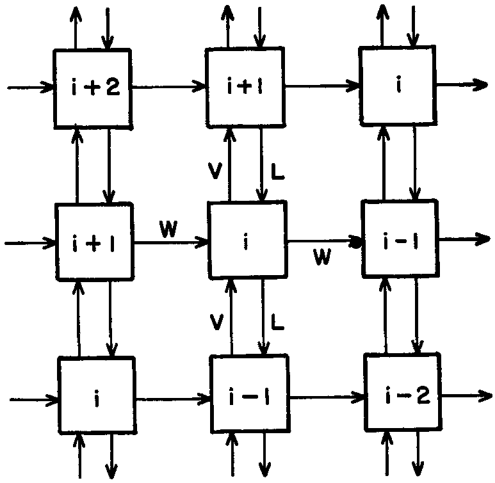

FIG. 3B. Two-dimensional.

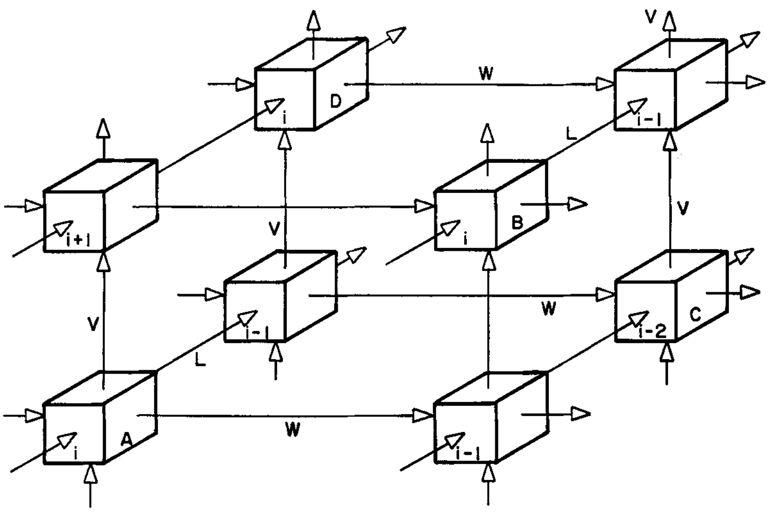

FIG. 3C. Three-dimensional.

More complex three-phase cascades with or without reflux can be employed in the three cascade types shown in Fig. 3. An analogy between one-, two-, and three-dimensional systems also exists when continuous contact systems are considered. The analysis considered in the first part of this paper can be extended to the three-phase systems.

DISCUSSION

The energy balance has not yet been considered. If the energy balance undergoes the transformation in Eq. (6), the appropriate two-dimensional energy balance will transform if diffusion is negligible. The boundary conditions will also transform appropriately.

Practical use of these techniques may be difficult. However, there is some incentive to do this in adsorption and chromatography where countercurrent flow is difficult to obtain. The two-dimensional process offers an alternative to simulated countercurrent operation. The analogy might also be useful in electrophoresis where two-dimensional processes are routinely used but countercurrent flow is difficult. The use of slanted feed and product lines with recycle would give an electrophoretic separation equivalent to a countercurrent separation. However, the separation would now be limited to binary separations.

A steady two-dimensional separator can develop the same multicomponent separations as a one-dimensional, time-dependent chromatograph (*6*, *7*). When parallel feed and product lines are employed, this ability is lost, but the binary separation which is achieved is improved. As noted previously, the throughput per cross-sectional area will be the same as for a countercurrent separator at the same L and V (flooding may be less of a problem). When one goes from a fixed bed, time-dependent column (e.g., chromatograph) to a countercurrent flow, steady-state system, one also loses the ability to fractionate multicomponent mixtures but obtains a better, more efficient binary separation. Thus the advantages and disadvantages of "countercurrent" type operation are the same in both cases. Two-dimensional separators offer an alternative geometry for operating cascades.

One interesting possibility for cascades which has not been extensively explored is hybrid cascades which have both "countercurrent" and "chromatographic" behavior. In a two-dimensional system slanted feed and slanted product lines would be employed, but this would be discontinuous. By proper location of these ports, multicomponent separations could be obtained but some of the efficiency of countercurrent operation would be retained (see Ref. *9* for the slanted feed line argument with a constant product location).

The corresponding simulated countercurrent system would utilize moving feed and withdrawal ports for part of a cycle and chromatographic development (no feed, solvent only) during the remainder of the cycle (see Ref. *8* for the moving feed point argument). Feed and withdrawal ports can move at different velocities and out-of-phase. In a countercurrent system the downward flow would be stopped when chromatographic development was desired.

The purpose of the hybrid processes is to obtain multicomponent separation in a single cascade but with higher efficiency than is obtainable with the usual elution chromatography. Partial use of countercurrent operation should make this possible.

SUMMARY

Two-dimensional cascades can produce the same separation as countercurrent cascades if parallel, slanted feed and product withdrawal lines are used. This is true for both continuous contact and staged systems. For three-phase systems the analogy can also be extended to three-dimensional cascades with feed and product withdrawal planes.

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SYMBOLS

a	interfacial area, cm^2/cm^3
c	concentration of solute in fluid, mol/cm^3
c°	concentration at equilibrium, mol/cm^3
D	diffusion coefficient, cm^2/s
F	feed rate, mol/s
k_M	mass transfer coefficient, $1/s \cdot cm^2$
L	column length, cm
L	liquid flow rate, mol/s
n	integer 0, 1, 2, 3, ...
P	product flow rate, mol/s
q	solute concentration on solid, mol/g
r	radial coordinate, cm
S	slope of feed and withdrawal lines, cm/radian
t, t'	time, s
T	temperature, °C or °K
v	velocity of fluid relative to solid, cm/s
V	flow rate of fluid phase, mol/s
v_F	fluid velocity relative to fixed reference frame, cm/s
v_s	port velocity or solid velocity relative to fixed reference frame, cm/s
w	angular velocity, radian/s
W	flow rate of third phase, mol/s

x_i, y_i, z_i concentration in a fluid phase leaving Stage i, mol/cm^3

z, z', z'' axial coordinate in reference frame moving with solid, stationary reference frame, and reference frame moving with ports, respectively, cm

Greek

θ angular coordinate, radians

ε porosity

ρ_s solid density, g/cm^3

Subscripts

A, B components

D desorbent

F feed

P product

s solid

$0, 1, 2, \ldots, i, i+1$ stage number

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