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Two-Dimensional Cross-Flow Cascades

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Summary

The two-dimensional cross-flow (2DCF) apparatus is a new cascade design which will separate a multicomponent feed into all its components in a continuous operation. The flow pattern for a 2DCF cascade is presented and discussed. An ideal theory shows that in discontinuous operation with continual feed 2DCF gives the same distributions as batch countercurrent distribution. (CCD). Discontinuous 2DCF could be used for preparatory work for any separations that CCD can be used. A steady-state continuous theory and possible alterations of the basic 2DCF apparatus are also presented.

INTRODUCTION

Many different techniques for separation of multicomponent mixtures have been developed. Although the various chromatographic techniques and countercurrent distribution (CCD) techniques can separate mixtures into all their components, they are not convenient for preparatory work since they are usually operated in a discontinuous fashion and large quantities are difficult to obtain. Some of these techniques can be operated in continual fashion (e.g. counter-double-current distribution with feed at each transfer step), but then they usually have the same limitation as the common continuous processes. When operated continually, the mixture can be separated into at best two pure components plus a mixture.

There are a few processes which can be operated continuously and still separate a multicomponent mixture into all its components. These systems all have the characteristic that the separation occurs in two spatial directions. Examples are electrochromatography (*1*) and continuous paper electrophoresis (*2*). Both systems require a large amount of solvent or carrier gas for the amount of material separated, and they are difficult to use for production of large quantities of materials.

In this paper the theory for a new type of cascade is presented. This cascade system consists of an array of stages in two dimensions, and theoretically will separate a multicomponent mixture into all of its components. These cascades could be applied to many different separation techniques, but they are discussed only in terms of extraction. The simplest such cascade, a two-dimensional cross-flow (2DCF) system, is discussed here. An ideal theory for the 2DCF system is presented for operation with a continual but discontinuous feed and for steady-state operation with a continuous feed. The system is compared with CCD and possible alterations of the basic cascade are discussed.

TWO-DIMENSIONAL CROSS-FLOW CASCADE

The basic 2DCF cascade is arranged as a two-dimensional matrix of stages and is interconnected so that each row and each column is a cross-flow cascade. An example is shown in Fig. 1 where the cascade is arranged as a rectangle. Consider an extraction process where the material to be separated is fed into stage (0,0) and one type of solvent is fed into the bottom of each column and a second type of solvent is fed into the left-hand side of each row. In the particular system shown there would be $N + M + 2$ solvent feeds and $N + M + 2$ product streams. We can consider that the top phase is transferred up a column and the bottom phase is transferred to the right across a row. The 2DCF cascade is the staged analog to the continuous two-dimensional processes listed earlier.

To illustrate the separation that can be expected, consider a three-component mixture fed continuously into stage (0,0). If one component greatly prefers the solvent flowing up the column, very little of this component will be carried to the right by the row solvent. Thus most of this first component will exit from stage (N,0), less from stage (N,1), etc. If the second component greatly prefers the row solvent, very little of this component will be carried up the cascade. Thus most of this

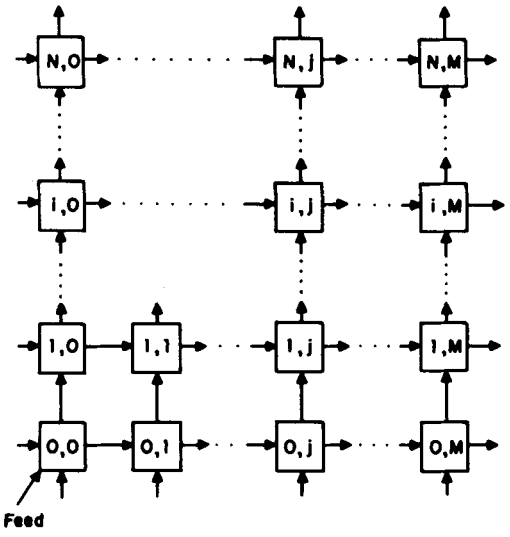

FIG. 1. Two-dimensional cross-flow cascade.

component will exit from stage $(0,M)$, less from stage $(1,M)$, etc. If the third component is distributed roughly equally between the two phases, it will tend to move up and to the right at the same speed. Thus this component will exit in the vicinity of stage (N,M). From this illustration we see that the 2DCF cascade can separate the components of a multicomponent feed if the distribution coefficients of the components differ.

Before proceeding, we note that Fig. 1 is very similar to the diagrams used by Craig and Craig (*3*) to illustrate the operation of the CCD apparatus and to illustrate a method of studying the approach to steady-state of a continuous counter-current apparatus. The one basic difference in these diagrams is that Fig. 1 represents a continuous or continual process in two spatial directions while the diagrams in Craig and Craig represent a discontinuous process with one spatial direction and time.

THEORY

Discontinuous Operation

In the discontinuous mode of operation all of the upper phase and all of the lower phase from each stage would be transferred up and to the

right, respectively. After the transfer step, the two phases would be mixed and then be allowed to equilibrate. At each transfer step solvent would be fed into the left-hand and the bottom stages of the cascade. Discontinuous operation could also be conducted in several other ways, but the method given here is analogous to continuous steady-state operation and leads to a separation which is analogous to CCD.

Consider an ideal 2DCF cascade where the distribution coefficients are constant, the two solvents are completely immiscible, the volume of upper phase V_U and lower phase V_L are also constant. With these simplifying assumptions the analysis is very similiar to that for CCD. If we let f_A = fraction of Component A in the upper phase and $K_A = C_{AU}/C_{AL}$ be the distribution coefficient it can be shown that

$$f_A = \frac{K_A V_U/V_L}{1 + K_A V_U/V_L} \tag{1}$$

Thus f_A is the same for all stages. If $M_{A_{i,j,s}}$ is the mass of Component A in stage (i,j), after transfer step s the following recursion relation can be derived.

$$M_{A_{i,j,s}} = f_A M_{A_{i-1,j,s-1}} + (1 - f_A) M_{A_{i,j-1,s-1}} \tag{2}$$

Equation (2) can be solved with various initial conditions. If the solvent streams entering the cascade are pure solvent then

$$M_{A_{-1,j,s}} = M_{A_{i,-1,s}} = 0 \tag{3}$$

Since the cascade initially contains no solute,

$$M_{A_{i,j,0}} = 0 \qquad \text{for} \quad i > 0, \quad \text{or} \quad j > 0 \tag{4}$$

There are two possible methods of feeding the column. The feed could be as one pulse at the initial time as in CCD. Then

$$M_{A_{0,0,0}} = M_{A_F} \tag{5}$$

Alternatively, we can feed the mixture to be separated at each transfer step. We will call this process continual operation. Then the initial condition is

$$M_{A_{0,0,s}} = M_{A_F} \tag{6}$$

For initial conditions (3), (4), and either (5) or (6), recursion Eq. (2) can be solved on the computer. An analytical solution can also be obtained by a probability analysis. Consider a single pulse of feed.

After s transfers, a solute molecule will be in stage (i,j) if and only if it was in the upper phase i times and the lower phase j times when it was transferred. Since all the material in a stage is transferred, $i + j = s$. The probability that the molecule of A is in the top phase is just f_A. The probability that it makes i jumps in s transfers is the binomial distribution or,

$$\frac{M_{A_{i,j,s}}}{M_{A_F}} = \frac{s!}{i!(s-i)!} f_A{}^i (1 - f_A)^{s-i} \tag{7}$$

Equation (7) is restricted to $i \geq 0, j \geq 0$, and we define $M_{A_{i,j,s}}$ so that Eq. (3) is satisfied.

Replacing s with $i + j$ we have

$$\frac{M_{A_{i,j,s}}}{M_{A_F}} = \frac{(i+j)!}{i!j!} f_A{}^i (1 - f_A)^j \tag{8}$$

If we feed only one pulse, the components to be separated occur only in the stages where $i + j = s$. This will be a diagonal line of stages from $(s,0)$ to stage $(0,s)$ in Fig. 1. Along this diagonal line the separation is given by Eq. (8) and is exactly the distribution obtained for CCD (*3*).

Suppose that a pulse of feed is introduced to stage (0,0) before every equilibration step. The material introduced for any one pulse must satisfy Eq. (8) where s is now the number of transfer steps since that pulse was introduced. This must occur since there is no mixing of materials fed at different times. The material to be separated will arrange itself in a series of diagonal lines of stages with equation $i + j =$ constant, where along each line the distribution is the same as a CCD distribution with $s = i + j$. When $s \geq \max (i + j)$, steady-state will have been obtained. Then with the continual feed and continual withdrawal each stage will have the concentration given by

$$\frac{M_{A_{i,j}}}{M_{A_F}} = \frac{(i+j)!}{i!j!} f_A{}^i (1 - f_A)^j \tag{9}$$

where i and j can take on any values.

For the rectangular array shown in Fig. 1 there will be $N + M + 2$ product streams. For the top products the weight of Material A leaving each stage at each transfer step is

$$f_A M_{A_{N,j}} = M_{A_F} \frac{(N+j)!}{N!j!} f_A{}^{N+1} (1 - f_A)^j \tag{10}$$

while for the side product streams the weight of Material A leaving each stage is

$$(1 - f_A)M_{Ai,M} = M_{AF}\frac{(i + M)!}{i!M!}f_A{}^i(1 - f_A)^{M+1} \tag{11}$$

Thus this method will split a multicomponent feed into its individual components in a continual operation. The number of stages needed to obtain the desired purity can be determined from Eqs. (9), (10), and (11) in the same fashion used for CCD.

Continuous Operation

The 2DCF cascade can also be operated in continuous fashion. If we assume that the distribution coefficients are constant, the flow rate of upper phase V is constant and the flow rate of lower phase L is constant for all stages, an expression for the product concentrations at steady-state can be developed. Define p_A as the ratio of the amount of Solute A transferred in stream V per unit time to the total amount of Solute A transferred per unit time for an individual stage. Then by a straightforward calculation similiar to that used to obtain Eq. (1),

$$p_A = \frac{K_A V/L}{1 + K_A V/L} \tag{12}$$

Also define $Q_{Ai,j}$ as the total amount of Solute A transferred from stage (i,j) per unit time. Then at steady-state without any chemical reactions a solute mass balance around stage (i,j) gives,

$$Q_{Ai,j} = p_A Q_{Ai-1,j} + (1 - p_A)Q_{Ai,j-1} \tag{13}$$

Equation (13) must be solved subject to the boundary conditions

$$Q_{A\,-1,j} = Q_{Ai,-1} = 0 \tag{14}$$

$$Q_{A0,0} = Q_{AF} \tag{15}$$

Equations (13), (14), and (15) are equivalent to Eqs. (2), (3), and (6) when the latter equations are written for steady, continual operation. Thus the binomial distribution found previously must also satisfy the equations for steady-state continuous operation. Thus

$$\frac{Q_{Ai,j}}{Q_{AF}} = \frac{(i + j)!}{i!j!}p_A{}^i(1 - p_A)^j \tag{16}$$

where $i \geq 0$ and $j \geq 0$.

The concentration of Solute A in the product streams leaving the cascade in the top row is given as

$$\frac{p_A Q_{A_{N,j}}}{V} = \frac{(N+j)!}{N!j!V} p_A{}^{N+1}(1-p_A)^j Q_{A_F} \tag{17}$$

while the concentration of Solute A leaving the cascade from the right-hand column is

$$\frac{(1-p_A)Q_{A_{i,M}}}{L} = \frac{(i+M)!}{i!M!L} p_A{}^i(1-p_A)^{M+1} Q_{A_F} \tag{18}$$

Thus in continuous operation the 2DCF cascade will separate a multicomponent feed into its individual components.

DISCUSSION

The 2DCF cascade has exactly the same distribution as the CCD apparatus, but the former apparatus will give this distribution with continual addition of feed. Thus the 2DCF cascade should work well for preparatory separations. Unfortunately, the number of stages required can be quite large so that the 2DCF may be too big. A 2DCF arranged as an isosceles triangle will have $(N+1)(N+2)/2$ total stages to be equivalent to a CCD with $N+1$ total stages. This large number of stages may put a practical limit to the size of N which can be used.

The triangular layout will often not be the most economic. For multicomponent mixtures the total number of stages in a CCD is usually controlled by the most difficult separation while the separation of other components will be more complete than necessary. To a certain extent the shape of the 2DCF apparatus can be adjusted to minimize the number of stages between easy to separate components. Consider a four-component feed where Component A greatly prefers the upper phase, Component B greatly prefers the lower phase, and the other two components, C and D, are roughly equally distributed. Separation of the first two components from the last two is relatively simple. If the triangular 2DCF were used, we could obtain pure A, a mixture of C and D, and then pure B. If many more stages were added, we would have pure A, stages with pure solvent, pure C, pure D, stages with pure solvent, and finally pure B. As an alternative the rectangular cascade

shown in Fig. 1 could be used. Now components C and D are transferred more times than either A or B. This is the desired action since C and D are more difficult to separate. Equations (9), (10), and (11), or (16), (17), and (18) still hold. The shape of the curves generated by the binomial distribution are available elsewhere (*3*) so they will not be presented here.

The use of a rectangular cascade has the further advantages that all solvent streams are used a maximum number of times and most of the components will appear in product streams dissolved in only one solvent. Any solvent streams leaving the cascade can naturally be recycled. Also, it would probably be advantageous to recycle product streams which contain very dilute concentrations of the components to be separated.

The 2DCF cascade has a large number of variables which would allow for many other possible variations. For instance, the flow rates or stage sizes could be varied from row to row or column to column. Different solvents could be used in the different rows or columns, or the temperatures of the stages could be varied. The geometry of the cascade could be varied to obtain an optimum separation. All of these changes would require an adjustment in the basic theory which is presented here.

No experimental work was tried in this study. A discontinuous 2DCF apparatus could probably be constructed using counter-double-current distribution (CDCD) stages (*4*) arranged in a two-dimensional pattern. The entire arrangement could be automated as are CCD and CDCD. The continuous 2DCF apparatus could be set up as a series of mixer-settlers or in an apparatus designed to occupy a single shell. The 2DCF apparatus will probably be most useful for multicomponent separations requiring relatively few stages. This combination utilizes the strength but minimizes the weakness of the 2DCF apparatus.

The theory presented here is an ideal theory which cannot be expected to agree completely with experiment. The agreement between theory and experiment should be quite similiar to the agreement between theory and experiment for CCD. A more exact theory could be constructed and solved on the computer.

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