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A Modified Kremser Equation for Stagewise Countercurrent Processes

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Summary

Although the Kremser equation has served for many years as a limiting design and analytical criterion for idealized countercurrent absorption and extraction systems, it contains a basic flaw which limits its effectiveness over certain ranges of the operating variables. This defect, which has not been generally recognized by writers in the field, can be remedied by assigning separate ranges to a certain operating parameter while still maintaining the same functional form of the original expression. This note presents a rational derivation, illustrates the fallacy in the old form, and demonstrates the use of the new form.

The amount of solute transferred between two immiscible solvent streams, either gas–liquid or liquid–liquid, in continuous countercurrent contact depends on the relative flow rates of the streams, the equilibrium distribution of solute between solvent phases, and the number of contacting stages provided. A compact analytical relationship among these variables was first developed by Kremser (*1*) and modified by Souders and Brown (*2*) for ideal cases where the equilibrium distribution may be assumed to be linear. Although the Kremser equation has been widely used for analysis of absorption and extraction systems, it has shortcomings which restrict its usefulness over certain ranges of the independent variables. These defects have been identified by Tiller (*3*), but none of the principal textbooks in the field (*4–8*), all of which postdate Tiller's article by considerable

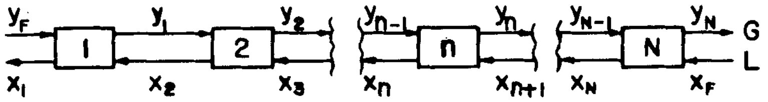

FIG. 1. Schematic diagram and nomenclature for N-stage countercurrent system.

periods, have included his ideas in their treatments. This note features a different presentation of some of Tiller's observations plus a modified form of the Kremser equation.

Figure 1 depicts a set of N isothermal stages, each assumed to effect ideal countercurrent mixing between two immiscible solvent streams whose flow rates are denoted by G and L and whose entering solute concentrations are given by y_F and x_F, respectively. Solute equilibrium is taken to follow the simple distribution law $y = mx$, so that the streams leaving from each ideal stage are related by $y_n = mx_n$, where n is an integer variable. It will be assumed, initially, that solute is being transferred from stream G to stream L, requiring that $y_{n-1} \geqslant mx_n$. A solute balance around stage n is

$$G(y_{n-1} - y_n) = L(x_n - x_{n+1}) \tag{1}$$

which may be rearranged to

$$y_{n-1} - y_n = \frac{L}{mG}(mx_n - mx_{n+1}) \tag{2}$$

Defining the important parameter $L/mG = A$ and rearranging further gives

$$Ay_{n+1} - (A + 1)y_n + y_{n-1} = 0 \tag{3}$$

This second-order, linear, homogeneous difference equation with constant coefficients has the general solution (*9*)

$$y_n = C_1 + \frac{C_2}{A^n} \tag{4}$$

The arbitrary constants C_1 and C_2 may be evaluated using the boundary conditions $y_0 = y_F$ and $y_{n+1} = mx_F$. Insertion of these results in Eq. (4) and letting $n \rightarrow N$ yields

$$y^N = \left(\frac{A^{N+1} - A}{A^{N+1} - 1}\right) mx_F + \left(\frac{A - 1}{A^{N+1}}\right) y_F \tag{5}$$

and additional algebraic manipulation gives

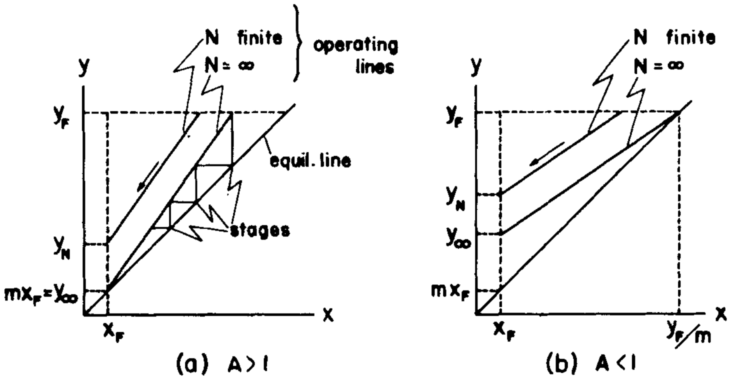

FIG. 2. Schematic graphical representations of operating lines, equilibrium lines, and terminal concentrations for N-stage countercurrent system with $y_{n-1} \geqslant mx_n$ (arrow indicates direction of increasing stage number).

$$\frac{y_F - y_N}{y_F - mx_F} = \frac{A^{N+1} - A}{A^{N+1} - 1} \tag{6}$$

Equation (6) is the celebrated Kremser equation. The derivation above differs somewhat from the originals (*1*, *2*), but it is similar in that the final algebraic steps are not necessarily related in a rational manner to the physical situation. An inconsistency arises when the expression is referred to a limiting condition of infinite stages.

The left-hand side of Eq. (6) may be interpreted as the ratio of solute recovered in N stages to that which would be recovered by an infinite number of stages, or

$$E_\infty(A > 1) = \frac{y_F - y_N}{y_F - y_\infty} = \frac{y_F - y_N}{y_F - mx_F} = \frac{A^{N+1} - A}{A^{N+1} - 1} \tag{7}$$

where $E_\infty(A > 1)$ is defined as the removal efficiency (the $A > 1$ restriction will be explained presently). The validity of this interpretation can be seen from Fig. 2a, a plot of y vs x which shows schematically the relationship among the variables in Eq. (6). The operating line, whose equation is

$$y_{n-1} = \frac{L}{G}x_n + \left(y_N - \frac{L}{G}x_F\right) \tag{8}$$

if obtained from a component balance around the right-hand end of the N stage system (the left-hand end furnishes an equivalent expression which would serve just as well), relates the solute compositions

y_{n-1} and x_n of passing solvent streams at any interstage point.* It has a slope of L/G and terminates on the vertical and horizontal projections of the terminal concentrations x_F and y_F, respectively. Figure 2a shows two operating lines, one for finite N and the other for $N = \infty$, each having the same slope but different positions. Sequential stages are represented by rectangular steps between operating and equilibrium line, the terminal steps coinciding with the ends of the operating line. A few illustrative stages are drawn for the $N = \infty$ line only. This particular line forms a "pinch point" with the equilibrium line at x_F, implying that an infinite number of stages would be required to reach that point. The relationship among the ordinates of Fig. 2 and the ratio of removal factors given by Eq. (7) is readily seen and the equivalence of mx_F and y_∞ is certainly valid.

For the above case the ratio of the slopes of the operating and equilibrium lines, $L/mG = A$, is greater than unity. If this were the only case, Eq. (7) and the interpretation given would be adequate. However, the physical situation for $A < 1$ is quite different, as Fig. 2b shows, and so is the corresponding equation. The "pinch point" for $N = \infty$ is now at the high concentration side of the diagram and y_∞ is no longer equivalent to mx_F. Expressions for $y_F - y_\infty$ and the corresponding $E_\infty(A < 1)$ are easily obtained from the geometry of the diagram. The slope of the $N = \infty$ operating line is

$$\frac{y_F - y_\infty}{(y_F/m) - x_F} = \frac{L}{G} = mA$$

which means $y_F - y_\infty = A(y_F - mx_F)$, so that

$$E_\infty(A < 1) = \frac{y_F - y_N}{y_F - y_\infty} = \frac{y_F - y_N}{A(y_F - mx_F)} = \frac{1 - A^N}{1 - A^{N+1}} \tag{9}$$

The Kremser equation, Eq. (6), has always been applied without restriction on the values of A, a procedure which is inconsistent since for $A > 1$ the efficiency E_∞ is referred to a y_∞ which is asymptotically realizable, whereas for $A < 1$ it is not. Use of Eqs. (7) and (9) for the two ranges of A removes this inconsistency.

As was mentioned above, the algebraic steps transforming Eq. (5) into Eq. (6) are purely ad hoc, and it is not inconceivable that

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* This is a first-order, nonhomogeneous linear difference equation whose solution, with appropriate boundary conditions, also yields Eq. (5) but the treatment is more involved than that based on the second-order homogeneous form, Eq. (1), as given above.

Kremser might have derived Eq. (9) instead of Eq. (7), so that the asymmetry would then have favored the $A < 1$ cases. A more rational derivation follows by applying a limiting process to Eq. (5), or

$$y = \lim_{N \to \infty} y_N = \lim_{N \to \infty} \left[\left(\frac{A^{N+1} - A}{A^{N+1} - 1} \right) m x_F + \left(\frac{A - 1}{A^{N+1} - 1} \right) y_F \right] \tag{10}$$

The result depends on the range of A, hence

$$y_\infty(A > 1) = m x_F \tag{11a}$$

$$y_\infty(A < 1) = A m x_F - (A - 1) y_F \tag{11b}$$

If each of these y_∞'s is substituted separately into $E_\infty = (y_F - y_N)/(y_F - y_\infty)$, also using Eq. (5) for the y_N in each case, the resulting expressions simplify to

$$E_\infty(A > 1) = \frac{A^{N+1} - A}{A^{N+1} - 1} \tag{12a}$$

and

$$E_\infty(A < 1) = \frac{1 - A^N}{1 - A^{N+1}} \tag{12b}$$

which are the same as Eq. (7) and (9). Hence, derivation through a limiting process leads naturally to two equations for the separate ranges of A, without resorting to geometric interpretation, whereas the purely algebraic derivation does not. It should be noted that for $A = 1$, Eqs. (12a) and (12b) are both indeterminate, but application of L'Hopital's rule to either gives

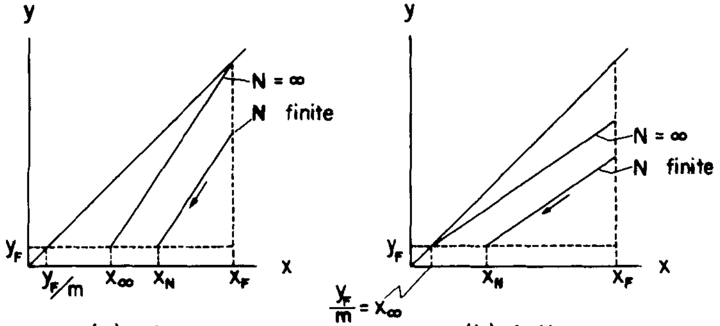
$$E_\infty(A = 1) = \frac{N}{N + 1} \tag{13}$$

Also, Eqs. (12a) and (12b) are interconvertible by exchanging $1/A$ for A. This property suggests combining Eqs. (12a), (12b), and (13) into the single expression

$$E_\infty = \frac{\alpha^{N+1} - \alpha}{\alpha^{N+1} - 1}; \qquad \alpha = \begin{cases} A, & A \geq 1 \\ 1/A, & A \leq 1 \end{cases} \tag{14}$$

which preserves the form of the original Kremser equation but includes the required symmetry of referring all cases to a bona fide y_∞.

An equally important set of conditions occurs when the direction of mass transfer is reversed, so that solute now moves from stream L to stream G, thus requiring that $x_n \geqslant y_{n-1}/m$. The two cases representing



(a) A>1 (b) A<1

FIG. 3. Schematic graphical representations of operating lines, equilibrium lines, and terminal concentrations for N-stage countercurrent system with $x_n \geqslant y_{n-1}/m$ (arrow indicates direction of increasing stage number).

this situation are shown schematically in Figs. 3a and 3b. For developing the analytical expressions, Fig. 1 is altered by exchanging all the x's and y's, as well as the L and G, but leaving stage numbers and composition subscripts in place. The solute balance around stage n is then

$$G(y_n - y_{n+1}) = L(x_{n-1} - x_n) \tag{15}$$

and the resulting difference equation

$$x_{n+1} - (A + 1)x_n + Ax_{n-1} = 0 \tag{16}$$

has the general solution

$$x_n = C_3 + C_4A^n \tag{17}$$

When arbitrary constants C_3 and C_4 are evaluated, using the boundary conditions $x_0 = x_F$ and $x_{N+1} = y_F/m$, the complete solution to Eq. (16) becomes, letting $n \rightarrow N$,

$$x_N = \left(\frac{A^N - 1}{A^{N+1} - 1}\right)\frac{y_F}{m} + \left(\frac{A^{N+1} - A^N}{A^{N+1} - 1}\right)x_F \tag{18}$$

Removal efficiency is analogously defined as $E_\infty = (x_F - x_N)/(x_F - x_\infty)$, and the x_∞ quantities are evaluated by $\lim_{N\to\infty} x_N$, giving

$$x_\infty(A > 1) = \frac{1}{A}\frac{y_F}{m} + \frac{1}{A}(A - 1)x_F \tag{19a}$$

$$x_\infty(A < 1) = \frac{y_F}{m} \tag{19b}$$

TABLE 1

Comparison of Kremser Equation and Eq. (14)

Operating parameters			Relative fraction solute remaining $(1 - E_\infty)$	
A	α	N	Eq. (6)	Eq. (14)
0.5	2.0	2	0.57	0.14
		4	0.52	0.03
		6	0.50	0.007
		10	0.50	0.0005
1.5	1.5	2	0.20	0.20
		4	0.075	0.075
		6	0.03	0.03
		10	0.006	0.006

Inserting these and Eq. (18) into E_∞ again gives Eqs. (12a) and (12b). Therefore Eq. (14) is applicable to all cases, regardless of the direction of solute flow, with the understanding that E_∞ refers to y's when $y_{n-1} \geqslant mx$ and to x's when $x_{n-1} \geqslant y_{n/m}$.

In addition to its elegance and rational derivation, Eq. (14) provides an improved practical basis for design criteria. Table 1 lists some values of $1 - E_\infty$, the fraction of solute remaining relative to what would remain for $N = \infty$, calculated for various A's and N's. As shown, the old Kremser expression, Eq. (6), is inadequate for $A < 1$, making little or no distinction among the various N values, particularly for large N, whereas Eq. (14) weights them correctly. For $A > 1$ both equations are the same and, of course, give the same results.

Graphical solutions of Eq. (6) constructed by previous writers use either log $(1 - E_\infty)$ vs N with A as parameter (*7, 10*) or log $(1 - E_\infty)$ vs A with N as parameter (*4*). Such plots will still serve for Eq. (14), merely by using α instead of A, and, since $\alpha \geqslant 1$, the portions of the graphs containing $A < 1$ may be excluded or ignored.

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