Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

## The Extent of Separation: Applications to Multistage Systems

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**To cite this Article** Rony, Peter R.(1970) 'The Extent of Separation: Applications to Multistage Systems', Separation Science and Technology, 5: 1, 1 — 10

**To link to this Article: DOI:** 10.1080/00372367008057943

**URL:** http://dx.doi.org/10.1080/00372367008057943

SEPARATION SCIENCE, 5(1), pp. 1–10, February, 1970

# The Extent of Separation: Applications to Multistage Systems

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## Summary

The fundamental equations for calculating the extent of separation are given for countercurrent multistage systems operated at either zero reflux, partial reflux, or total reflux. With the component distribution coefficients assumed constant, simple expressions for the maximum extent of separation are derived. The calculations clearly show how multistaging and reflux influence component separation.

## INTRODUCTION

It was shown in the first paper in this series (*1*) that the maximum extent of separation that could be attained in a single equilibrium stage is

$$\xi_{\max} = \text{abs}\left[\frac{\alpha^{1/2} - 1}{\alpha^{1/2} + 1}\right] \tag{1}$$

where $\alpha$ is the quotient of the distribution coefficients, $K_i$, for two components distributing between two regions,

$$\alpha = \frac{K_2}{K_1} \tag{2}$$

Table 1 lists $\xi_{\max}$ for values of $\alpha - 1$ ranging from $10^{-5}$ to $10^2$. Even when $\alpha$ is a large number, it is quite clear that a single equilibrium stage is a relatively ineffective separation method. The problem becomes particularly acute for mixtures of components among which the values of $\alpha$ are close to unity.

To circumvent this inherent limitation of single-stage methods, chemical engineers employ the ingenious principle of *countercurrent multistaging,* whereby the small separation achievable in a single stage is multiplied until a large separation is produced. This paper will demonstrate the application of the extent of separation to such processes. The calculations will clearly show how multistaging influences component separation and why an enchancement in separation occurs.

**TABLE 1**

The Maximum Extent of Separation for a Multistage Column Operated at Zero Reflux and with Equal Washing and Extraction Sections ($n = m$)

| | $\xi_{max}$ | | | |
|---|---|---|---|---|
| $\alpha - 1$ | $n = 1$ | $n = 2$ | $n = 5$ | $n = 10$ |
| $10^{-5}$ | $2.50 \times 10^{-6}$ | $5.00 \times 10^{-6}$ | $1.25 \times 10^{-5}$ | $2.50 \times 10^{-5}$ |
| $10^{-4}$ | $2.50 \times 10^{-5}$ | $5.00 \times 10^{-5}$ | $1.25 \times 10^{-4}$ | $2.50 \times 10^{-4}$ |
| $10^{-3}$ | $2.50 \times 10^{-4}$ | $5.00 \times 10^{-4}$ | $1.25 \times 10^{-3}$ | $2.50 \times 10^{-3}$ |
| $10^{-2}$ | $2.49 \times 10^{-3}$ | $4.98 \times 10^{-3}$ | $1.24 \times 10^{-2}$ | $2.49 \times 10^{-2}$ |
| $10^{-1}$ | $2.38 \times 10^{-2}$ | $4.76 \times 10^{-2}$ | 0.119 | 0.234 |
| $10^{0}$ | 0.172 | 0.333 | 0.700 | 0.939 |
| $10^{1}$ | 0.537 | 0.833 | 0.995 | 0.999+ |
| $10^{2}$ | 0.819 | 0.980 | 0.999+ | 0.999+ |

In order to develop a tractable solution, it will be assumed that the distribution coefficients in the multistage column are constant. This assumption has been previously made by many other investigators. The reader is referred to the excellent paper by Klinkenberg (*2*) and to the corresponding treatments of Kremser (*3*), Fenske (*4*), Souders and Brown (*5*), Stene (*6*), Tiller and Tour (*7*), Tiller (*8*), Bartels and Kleiman (*9*), Rometsch (*10*), and Scheibel (*11*).

## DISTRIBUTION COEFFICIENTS

Steady-state countercurrent multistage systems are qualitatively different from the *closed* single-stage systems previously considered (*1*). In the latter, the distribution coefficient, $K_i$, is governed by the partition coefficient, $\kappa_{i2}$,

$$\kappa_{i2} = \frac{c_{i2}}{c_{i1}} \tag{3}$$

and the ratio of the cross-sectional areas of the two phases, $A_{\perp 2}/A_{\perp 1}$ (assuming a uniform column). Thus,

$$K_i \equiv \frac{A_{\perp 2}c_{i2}}{A_{\perp 1}c_{i1}} = \kappa_{i2}\frac{A_{\perp 2}}{A_{\perp 1}} \tag{4}$$

In contrast to the above, countercurrent multistage systems are *open* systems. The ratio of the molar velocities of the two phases must therefore be incorporated into the definition of the distribution coefficient, $\dot{K}_i$,

$$\dot{K}_i \equiv \frac{A_{\perp 2}v_2c_{i2}}{A_{\perp 1}v_1c_{i1}} = \kappa_{i2}\frac{A_{\perp 2}v_2}{A_{\perp 1}v_1} = K_i\frac{v_2}{v_1} \tag{5}$$

The above distinction between $K_i$ and $\dot{K}_i$ is rarely stated. In the present paper, $\kappa_{i2}$, $K_i$, and $\dot{K}_i$ are all assumed to be constant.

## CONSERVATION-OF-MASS EQUATIONS FOR A COUNTERCURRENT MULTISTAGE SYSTEM

For a countercurrent multistage system with constant distribution coefficients, $\dot{K}_i$, Klinkenberg (*2*) has provided a procedure for obtaining the conservation-of-mass equations. This procedure so simplifies the theoretical treatment of such systems that is is worth repeating:

"1. Number the stage in the direction of the flow of that phase whose solute flow is in the definition of $\dot{K}_i$ in the denominator (i.e., the residue phase).

2. Assign a positive sign to solute flow rates entering, and a negative sign to flows leaving the extraction system.

3. Multiply each solute flow rate by a power of $\dot{K}_i$ the exponent of which is the number of the stage in which such flow *enters*.

4. Equate the sum of the flows found under 2 to zero (= overall material balance).

5. Equate the sum of the products found under 3 to zero (= extraction equation).

6. Reduce the two equations to a single one by eliminating one of the unknowns."

If $n_{ij}(k)$ is the molar flow rate of component $i$ in phase $j$ emanating from stage $k$, the equations governing the system shown in Fig. 1 are

$$-\dot{n}_{i2}(1) + \dot{n}_{i1}(0) + \dot{n}_i^F + \dot{n}_{i2}(n+m) - \dot{n}_{i1}(n+m-1) = 0 \tag{6}$$

$$-\dot{n}_{i2}(1) + \dot{K}_i\dot{n}_{i1}(0) + \dot{K}_i^m\dot{n}_i^F + \dot{K}_i^{n+m-1}\dot{n}_{i2}(n+m) - \dot{K}_i^{n+m}\dot{n}_{i1}(n+m-1) = 0 \tag{7}$$

The distribution coefficient, $\dot{K}_i$, is defined as

$$\dot{K}_i \equiv \frac{\dot{n}_{i2}(k)}{\dot{n}_{i1}(k)} = K_i\frac{v_2}{v_1} \tag{8}$$

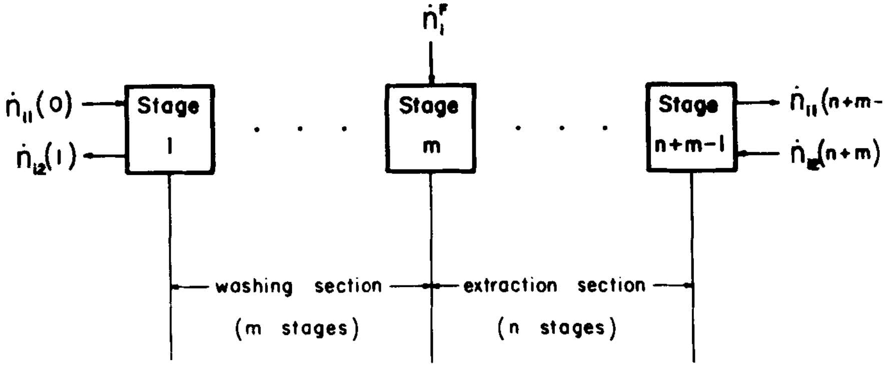

FIG. 1. Definitions of terms for a countercurrent multistage system.

Equations (6) and (7) will now be solved for three different cases: (1) no reflux, (2) total reflux, and (3) partial reflux. The reader is referred to Klinkenberg's articles (*2*, *12*, *13*) for further discussions of the characteristics of these systems.

## EXTENT OF SEPARATION

The extent of separation, $\xi$, is defined by the equation

$$\xi = \text{abs}[\dot{Y}_{11} - \dot{Y}_{21}] = \text{abs}[\dot{Y}_{22} - \dot{Y}_{12}] \tag{9}$$

For the multistage system shown in Fig. 1, the segregation fractions, $\dot{Y}_{ij}$, are

$$\dot{Y}_{i1} \equiv \frac{\dot{n}_{i1}(n + m - 1)}{\dot{n}_{i1}(n + m - 1) + \dot{n}_{i2}(1)} \tag{10}$$

$$\dot{Y}_{i2} \equiv \frac{\dot{n}_{i2}(1)}{\dot{n}_{i1}(n + m - 1) + \dot{n}_{i2}(1)} \tag{11}$$

when the column is operated at either zero or total reflux.

## NO REFLUX

If the entering streams $\dot{n}_{i1}(0)$ and $\dot{n}_{i2}\,(n + m)$ do not contain $i$,

$$\dot{n}_{i1}(0) = 0 \tag{12}$$

$$\dot{n}_{i2}(n + m) = 0 \tag{13}$$

and the segregation fraction, $\dot{Y}_{i1}$, and extent of separation become, respectively,

$$\dot{Y}_{i1} = \frac{\dot{K}_i^m - 1}{\dot{K}_i^{n+m} - 1} \tag{14}$$

$$\xi = \mathrm{abs}\left[\frac{\dot{K}_1^m - 1}{\dot{K}_1^{n+m} - 1} - \frac{\dot{K}_2^m - 1}{\dot{K}_2^{n+m} - 1}\right] \tag{15}$$

Special cases discussed by Klinkenberg (*2*) include (a) $\dot{K}_i = 1$, (b) $\dot{K}_i \gg 1$, (c) $\dot{K}_i \ll 1$, (d) $n = m$, (e) $m = 1$ (extraction with a pure solvent), (f) $n = 1$ (a washing process), and (g) extraction with an impure solvent.

For case (d),

$$\dot{Y}_{i1} = \frac{1}{1 + \dot{K}_i^n} \qquad (n = m) \tag{16}$$

and

$$\xi = \mathrm{abs}\left[\frac{1}{1 + \dot{K}_1^n} - \frac{1}{1 + \dot{K}_2^n}\right] \qquad (n = m) \tag{17}$$

If we define the quotient of the distribution coefficients, $\alpha$, as

$$\alpha = \frac{\dot{K}_2}{\dot{K}_1} \tag{18}$$

substitute Eq. (18) into Eq. (17), differentiate the result with respect to $\dot{K}_1$, and set the derivative equal to zero, we can calculate the optimum value of $\dot{K}_1$,

$$\dot{K}_{1\mathrm{opt}} = \alpha^{-1/2} \qquad (n = m) \tag{19}$$

the optimum value of $\dot{K}_2$,

$$\dot{K}_{2\mathrm{opt}} = \alpha^{1/2} \qquad (n = m) \tag{20}$$

and the maximum value of the extent of separation, $\xi_{\max}$,

$$\xi_{\max} = \mathrm{abs}\left[\frac{\alpha^{n/2} - 1}{\alpha^{n/2} + 1}\right] \qquad (n = m) \tag{21}$$

Thus, the optimum conditions that apply for a single equilibrium stage (*1*) and the Craig countercurrent apparatus (*14*) also apply to a countercurrent multistage column operated without reflux and with equal extraction and washing sections. The effect of multistaging can be clearly seen in Eq. (17): the value of the distribution coefficient, $\dot{K}_i$, in each term is raised to a power of $n$, half the total the number of stages. If the multistage system is operated under the optimum set of conditions, the quotient of the distribution coefficients, $\alpha$, is also raised to a power of $n$ [Eq. (21)]. Table 1 lists $\xi_{\max}$ for values of $\alpha - 1$ ranging from $10^{-5}$ to $10^2$ for $n = 1, 2, 5$, and 10.

## TOTAL REFLUX

The condition of total reflux occurs when solute leaving each end of the multistage column shown in Fig. 1 is re-introduced at the same end, i.e.,

$$\dot{n}_{i1}(0) = \dot{n}_{i2}(1) \tag{22}$$

$$\dot{n}_{i1}(n + m - 1) = \dot{n}_{i2}(n + m) \tag{23}$$

$$\dot{n}_i^F = 0 \tag{24}$$

Accordingly, the segregation fraction, $\dot{Y}_{i1}$, and the extent of separation, $\xi$, become,

$$\dot{Y}_{i1} = \frac{1}{1 + \dot{K}_i^{n+m-1}} \tag{25}$$

$$\xi = \text{abs}\left[\frac{1}{1 + \dot{K}_1^{n+m-1}} - \frac{1}{1 + \dot{K}_2^{n+m-1}}\right] \tag{26}$$

If we substitute Eq. (18) into Eq. (26), differentiate the result with respect to $\dot{K}_1$, and set the derivative equal to zero, we obtain optimum values for $\dot{K}_1$ and $\dot{K}_2$ that are identical to Eqs. (19) and (20). The maximum extent of separation becomes

$$\xi_{\max} = \text{abs}\left[\frac{\alpha^{(n+m-1)/2} - 1}{\alpha^{(n+m-1)/2} + 1}\right] \tag{27}$$

If $n = m$, Eqs. (25) through (27) respectively reduce to

$$Y_{i1} = \frac{1}{1 + \dot{K}_i^{2n-1}} \qquad (n = m) \tag{28}$$

$$\xi = \text{abs}\left[\frac{1}{1 + \dot{K}_1^{2n-1}} - \frac{1}{1 + \dot{K}_2^{2n-1}}\right] \qquad (n = m) \tag{29}$$

$$\xi_{\max} = \text{abs}\left[\frac{\alpha^{n-1/2} - 1}{\alpha^{n-1/2} + 1}\right] \qquad (n = m) \tag{30}$$

Thus, for equal extraction and washing sections, *the condition of total reflux is equivalent to the addition n — 1 extra stages to a column that is operated at no reflux.*

## PARTIAL REFLUX

If only a fraction of the solute exiting from either end of the column is re-introduced into the column, Eqs. (6) and (7) must be solved for the condition of finite reflux ratios, $r_e$ and $r_w$. These ratios are defined by the equations,

$$\frac{\dot{n}_{i1}(n+m-1)}{\dot{n}_{i2}(n+m)} = \frac{r_e+1}{r_e} \qquad \text{(extraction section)} \tag{31}$$

$$\frac{\dot{n}_{i2}(1)}{\dot{n}_{i1}(0)} = \frac{r_w+1}{r_w} \qquad \text{(washing section)} \tag{32}$$

The segregation fraction, $\dot{Y}_{i1}$, can be obtained by solving Eqs. (6) and (7) subject to the restrictions given in Eqs. (31) and (32),

$$\begin{aligned} \dot{Y}_{i1} &= \frac{\dot{n}_{i2}(n+m)}{\dot{n}_{i2}(n+m) + \dfrac{r_e}{r_w}\,\dot{n}_{i1}(0)} \\ &= \frac{\dot{n}_{i1}(n+m-1)}{\dot{n}_{i1}(n+m-1) + \left(\dfrac{r_e+1}{r_w+1}\right)\dot{n}_{i2}(1)} \\ &= \frac{r_w + \dfrac{\dot{K}_i^m - 1}{\dot{K}_i - 1}}{r_w + r_e\dot{K}_i^{n+m-1} + \dfrac{\dot{K}_i^{n+m} - 1}{\dot{K}_i - 1}} \end{aligned} \tag{33}$$

The extent of separation is computed according to the procedure given in Eq. (9),

$$\xi = \operatorname{abs}\left[\frac{r_w + \dfrac{\dot{K}_1^m - 1}{\dot{K}_1 - 1}}{r_w + r_e\dot{K}_1^{n+m-1} + \dfrac{\dot{K}_1^{n+m} - 1}{\dot{K}_1 - 1}} - \frac{r_w + \dfrac{\dot{K}_2^m - 1}{\dot{K}_2 - 1}}{r_w + r_e\dot{K}_2^{n+m-1} + \dfrac{\dot{K}_2^{n+m} - 1}{\dot{K}_2 - 1}}\right] \tag{34}$$

At total reflux,

$$r_e = r_w = \infty \qquad \text{(total reflux)} \tag{35}$$

Equation (34) reduces to Eq. (26), and with no reflux,

$$r_e = r_w = 0 \qquad \text{(no reflux)} \tag{36}$$

Equation (34) reduces to Eq. (15). If either $r_e = \infty$ or $r_w = \infty$, the extent of separation becomes equal to zero,

$$\xi = 0 \qquad (r_e = \infty \text{ or } r_w = \infty) \tag{37}$$

## EXAMPLE

The maximum extent of separation given by Eq. (21) has been plotted as a function of the quantity $\alpha - 1$ for $n = 1$, 10, and 100 (Fig. 2). The shape of the curves are similar to those characterizing

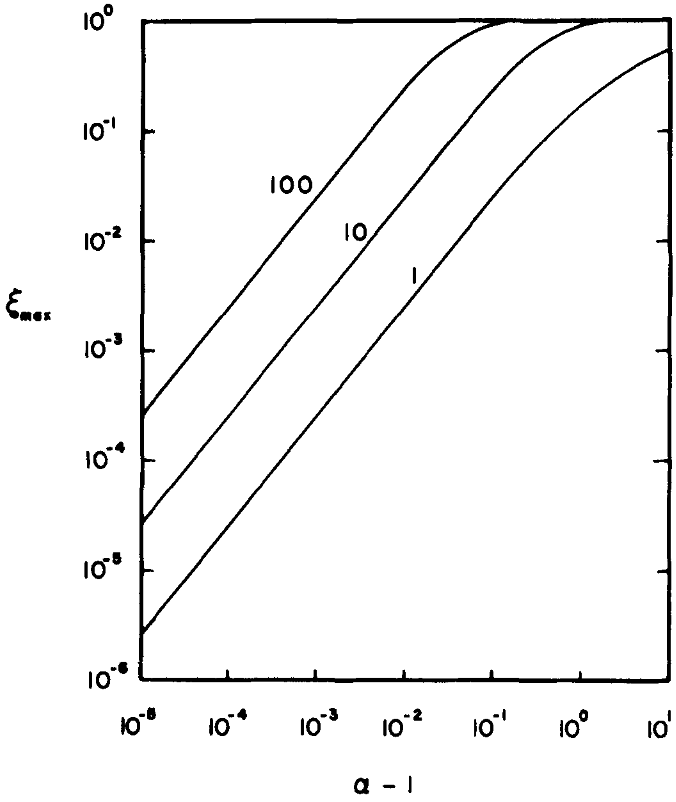

**FIG. 2.** The maximum extent of separation for a multistage column operated at zero reflux and with equal washing and extraction sections ($n = m$). Curves are shown for three different values of $n$.

multicontact separation systems (*14*). If we let $\alpha$ approach unity, we observe that the maximum extent of separation in Eq. (21) approaches a value of

$$\lim_{\alpha \to 1} \xi_{\text{max}} = \frac{n}{4}(\alpha - 1) \tag{38}$$

or

$$\lim_{\alpha \to 1} \frac{\partial \xi_{\text{max}}}{\partial(\alpha - 1)} = \frac{n}{4} \tag{39}$$

Thus, for compounds that differ only slightly from each other, the maximum extent of separation is directly proportional to the number of stages [for multistage systems operated at total reflux, $n$ must be replaced by $n + m - 1$ in Eqs. (38) and (39)]. In contrast to these equations, the maximum extent of separation for multicontact devices (such as the Craig countercurrent apparatus) is proportional to the square root of the number of stages.

In a subsequent paper, we will demonstrate how Eq. (38) can be used to correctly define the "number of theoretical plates" in an elution chromatographic system (*15*).

## List of Symbols

| | |
|---|---|
| $A$ | cross-sectional area (cm$^2$) |
| $c$ | concentration (moles/cm$^3$) |
| $\dot{K}$ | distribution coefficient for an open system (moles/sec: moles/sec) |
| $K$ | distribution coefficient for a closed system (moles/moles) |
| $m$ | number of stages in the "washing" section |
| $n$ | number of stages in the "extraction" section |
| $\dot{n}^F$ | molar flow rate of feed (moles/sec) |
| $\dot{n}_{ij}(k)$ | molar flow rate of component $i$ in phase $j$ emanating from stage $k$ |
| $r$ | reflux ratio |
| $v$ | molar velocity (cm/sec) |
| $\dot{Y}$ | segregation fraction for an open system |

## Greek Letters

| | |
|---|---|
| $\alpha$ | quotient of the distribution coefficients |
| $\kappa$ | partition coefficient (moles/cm$^3$:moles/cm$^3$) |
| $\xi$ | extent of separation |

## Subscripts

| | |
|---|---|
| $e$ | extraction section |
| $i$ | component $i$ |
| $ij$ | component $i$ in phase or region $j$ |
| max | maximum value |
| opt | optimum value |
| $w$ | washing section |
| $i1, i2, 11, 22$, etc. | specific component–region combinations |

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*Received by editor August 15, 1969*