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No-Mix and Ideal Separation Cascades

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ABSTRACT

Ideal countercurrent recycle cascades are characterized by two properties: 1) The compositions of heads and tails streams forming the feed stream to individual stages are the same, and 2) the heads separation factor for each stage is constant. When these two criteria are met, the heads and tails separation factors are constant and equal to the square root of the stage separation factor. The mixing of streams with different compositions within a separation cascade obviously constitutes an inefficiency since it is precisely the reverse of this process that is desired, hence Condition 1, which is often referred to as the *no-mix criterion*. Separation cascades for which both criteria are valid are termed *ideal*. However, the ramifications of Condition 2 are not obvious and it is possible to design *no-mix cascades* which do not meet the second condition. Mathematical relationships between ideal and no-mix separation cascades are derived to quantify these differences. It is shown that Condition 2 minimizes the total interstage flow required to make a given separation for any no-mix cascade design, i.e., it is an *ideal cascade* by definition. Finally, the required number of ideal stages necessary to perform a specific separation with no-mix cascades are evaluated and compared to those of the ideal cascade.

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INTRODUCTION AND BACKGROUND

Many separations are carried out in countercurrent recycle cascades such as shown in Fig. 1. Each individual stage in the cascade receives a feed stream with flow rate F_i and composition z_i, and produces a stage heads stream (flow rate M_i, composition y_i) enriched in the desired component, and a tails stream (flow rate N_i, composition x_i) depleted in the desired component. When the amount of separation effected by a single stage is smaller than that required between the waste and product streams, it becomes necessary to connect a number of stages together in series to obtain the desired fractionation. The individual stages operate jointly such that the overall function of the cascade is to separate an external feed stream (flow rate F, composition z_F) into a product stream (flow rate P, composition y_P) and a waste stream (flow rate W, composition x_W).

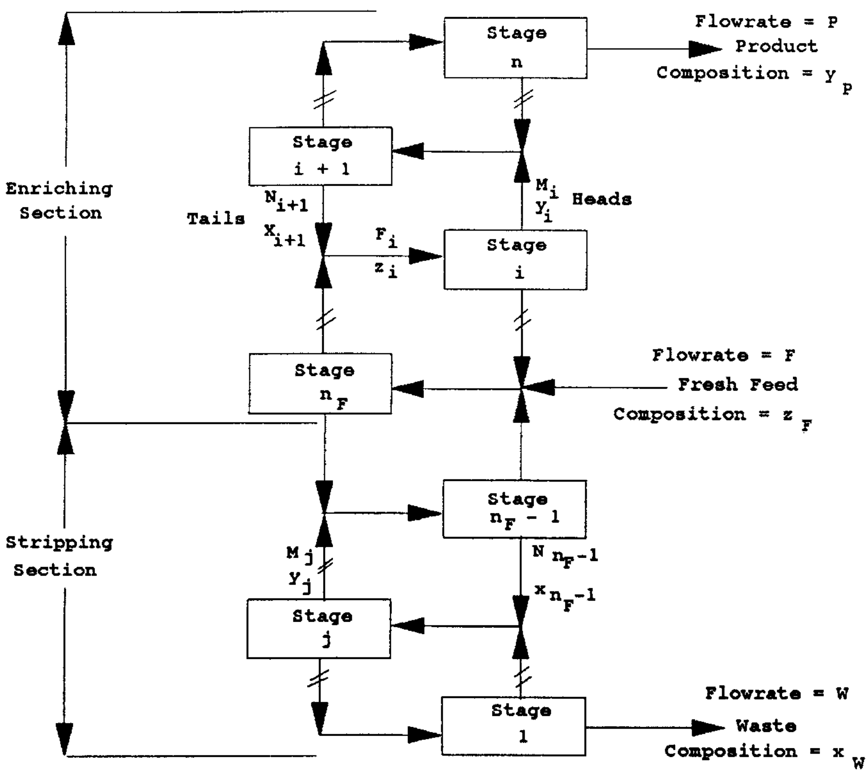

FIG. 1 Flow schematic of a generalized countercurrent recycle cascade.

In the countercurrent recycle cascade of Fig. 1, the heads stream from the next lower stage (M_{i-1}) and the tails stream from the next higher stage (N_{i+1}) are "recycled" or combined to form the feed to stage i. This is essentially the flow pattern which occurs in a number of separation processes including ordinary plate distillation columns and isotope separation cascades. Additionally, a series of permeation stages can be connected in series to form countercurrent recycle cascades for gas separations. This configuration results in significant theoretical advantages with respect to membrane area and compressor duty requirements over "one-compressor" permeators (1–3).

At some stagewise minimum flow rate, separation will cease between adjacent stages, or expressed mathematically, $y_{i+1} = y_i$ (4). Furthermore, this minimum recycle rate increases as the composition departs more from the desired product and waste compositions. Consequently, minimum recycle is greatest at the feed stage, and "tapers" to smaller values as both ends of the cascade are approached (5). It is therefore apparent that the stagewise recycle rates are important design variables. It is convenient to define stagewise recycle as a function of the product (or alternatively, the waste) flow rate in terms of the *recycle ratio*, R_i:

$$R_i \equiv N_{i+1}/P \tag{1}$$

where N_{i+1} is the tails stream flow rate from the next higher stage in the cascade and P is the flow rate of the product stream.

A cascade can be designed to make a desired overall separation provided that the recycle ratio at each stage is above the respective minimum, $(R_i)_{\min}$. As the stagewise $(R_i)_{\min}$ is approached, a great number of stages will be required to transgress the resulting "pinch point," with the number becoming infinite as $R_i \rightarrow (R_i)_{\min}$.

With the restriction that the recycle ratio at each stage in the cascade is above the respective value of $(R_i)_{\min}$, a great number of cascade designs are possible, including constant recycle, tapered, and squared-off cascades. In the case of a constant recycle cascade, the recycle ratio is fixed and constant for every stage. For squared-off cascades, the recycle ratio is constant across consecutive groups (or sections) of stages, but may differ for different sections. In a tapered cascade, the stagewise recycle ratio will vary, but must be above $(R_i)_{\min}$ for each stage. Each design typically requires a different total number of stages. For a given overall separation, the constant recycle cascade will typically require fewer stages while the ideal cascade will require the greatest number of stages. The final design is usually chosen based on economic considerations as related to size and energy requirements.

The total interstage flow through a cascade is the total amount of material, both heads and tails, processed by the collective stages to produce the desired quantity and purity of product. Consequently, the total interstage flow is a measure of the size of the separation plant required to obtain a specified separation. As discussed by Benedict et al. (5), the cascade design that assures minimum total interstage flow is an *ideal cascade* and is characterized by the following conditions:

1. The compositions of heads and tails streams forming the feed to a stage are the same:

$$y_{i-1} = z_i = x_{i+1} \qquad (i = 2, 3, \ldots, N - 1) \tag{2}$$

2. The heads separation factor, β, is constant.

When Conditions 1 and 2 are met, it is easily shown β is equal to the square root of the (constant) stage separation factor (4):

$$\beta = \sqrt{\alpha} \tag{3}$$

Furthermore, when β is constant, the tails separation, γ, will also be equal to $\sqrt{\alpha}$.

The first condition, referred to as the no-mix criterion, is intuitively obvious since the mixing of streams with different compositions constitutes an inefficiency; it is precisely the reverse of this process that is desired and takes place within the stage itself. However, the ramifications of the second requirement are not apparent. It is possible to design a cascade that meets Condition 1 but not Condition 2; that is, Condition 1 is necessary but insufficient to meet the requirements of an ideal cascade.

NO-MIX AND IDEAL CASCADES

For binary systems, certain relationships and calculations for separation cascades are simpler when expressed as mole fractions. The Greek letters η, ξ, and ζ are used to denote these ratios in the stage heads, tails, and feed streams, respectively, as indicated in Fig. 2. The stage separation factors are easily defined in terms of these ratios:

$$\text{Stage separation factor:} \qquad \alpha \equiv \eta/\xi \tag{4}$$

$$\text{Heads separation factor:} \qquad \beta \equiv \eta/\zeta \tag{5}$$

$$\text{Tails separation factor:} \qquad \gamma \equiv \zeta/\xi \tag{6}$$

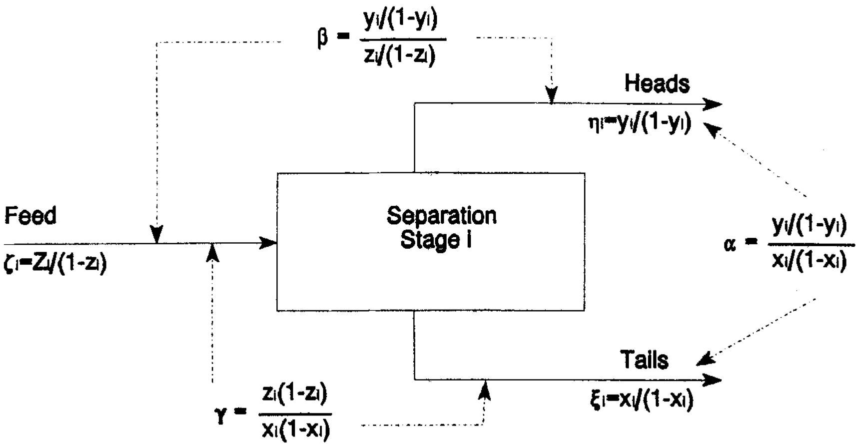

FIG. 2 Relationships between mole ratios in a separation stage.

Consider the three consecutive cascade stages shown in Fig. 3. For a cascade in which the no-mix criterion is met, Eq. (2) may also be expressed in terms of mole ratios:

$$\eta_{i-1} = \zeta_i = \xi_{i+1} \tag{7}$$

Further, consider the relationships between the heads separation factors for *every other* stage in the cascade. From the definition of β, Eq. (5), it follows:

$$\beta_{i+1} = \eta_{i+1}/\zeta_{i+1} \tag{8}$$

$$\beta_{i-1} = \eta_{i-1}/\zeta_{i-1} \tag{9}$$

Dividing Eq. (8) by Eq. (9) and combining with Eq. (7) yields

$$\beta_{i+1}/\beta_{i-1} = (\eta_{i+1}/\xi_{i+1})(\xi_i/\eta_i) = \alpha/\alpha = 1$$

Consequently:

$$\beta_{i+1} = \beta_{i-1} \qquad (i = 2, 3, 4, \ldots, N-1) \tag{10}$$

Hence, one of the requirements for a no-mix cascade is that the heads separation factors for *at least* every other stage in the cascade be equal.

Next, consider the relationship between the heads separation factor for *adjacent* stages shown in the section of the cascade in Fig. 3 (stages $i-1$ and i). It is easily shown by combining equations for β_{i-1} and β_i with the no-mix criterion that

$$\beta_{i-1}\beta_i = \alpha \qquad (i = 2, 3, 4, \ldots, N) \tag{11}$$

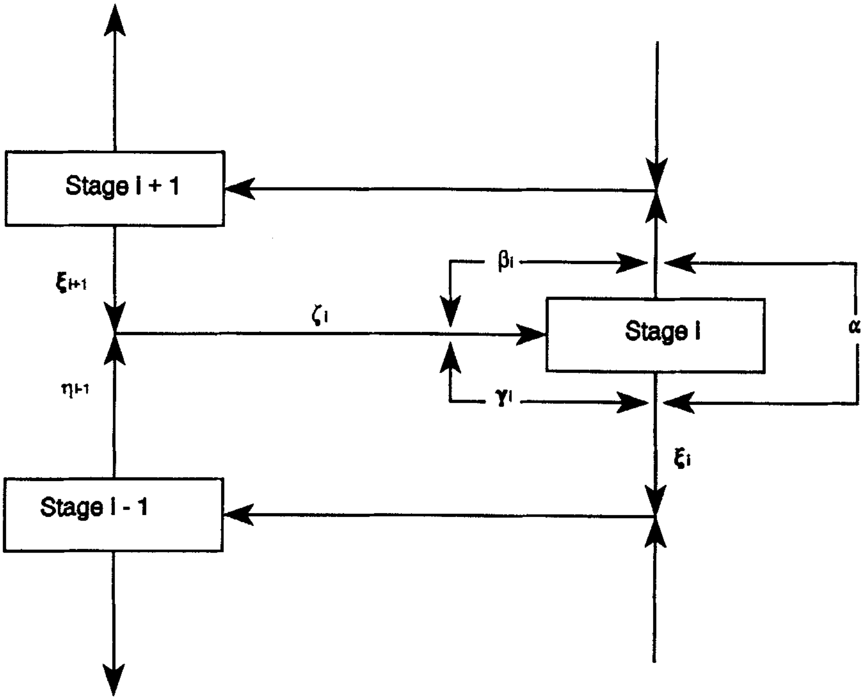

FIG. 3 Three consecutive stages of a countercurrent recycle cascade.

Furthermore, β must lie in the range

$$1 < \beta < \alpha \tag{12}$$

if fractionation is to take place within a stage. Consequently, at least the following conditions must be met in a no-mix cascade:

$$\beta_{i-1} = \beta_{i+1} \tag{13}$$

$$\beta_{i-1}\beta_i = \alpha$$

$$1 < \beta < \alpha$$

Equations (10) and (11) suggest that it may be convenient to relate the heads separation factor, β, to α by the relationships

$$\beta_{i-1} = \alpha^q \qquad \beta_i = \alpha^{1-q}$$

or alternatively

$$\beta_{i-1} = \alpha^{1-q} \qquad \beta_i = \alpha^q \tag{14}$$

where

$$0 < q < 1 \tag{15}$$

The exponential factor, q, must lie within the prescribed range as a result of Eq. (12). Note that the ideal cascade is a special case of the no-mix cascade when $q = 0.5$, for which β is constant and equal to $\sqrt{\alpha}$ at every stage in accord with Eq. (3).

NUMBER OF STAGES

The total number of stages, N, in a no-mix cascade may be evaluated by a procedure similar to that used to derive the familiar Fenske–Underwood equation. For a cascade containing N stages, slightly different equations result depending on whether N is odd or even since the no-mix criterion applies to the heads and tails streams coming from every other stage:

$$\eta_N = \alpha_N \alpha_{N-2} \cdots \alpha_3 \alpha_1 \xi_W \tag{16}$$

For constant α, Eq. (16) reduces to

$$(N + 1)/2 = \frac{\ln(\eta_N/\xi_W)}{\ln \alpha} \qquad (N\ odd) \tag{17}$$

For the case in which the cascade contains an even number of stages:

$$\eta_N = \alpha_N \alpha_{N-2} \cdots \alpha_4 \alpha_2 \xi_2 \tag{18}$$

$$\xi_2 = (\alpha_1/\beta_1)(\xi_W)$$

$$\beta_1 = \alpha_1^q$$

Alternatively,

$$\eta_{N-1} = \alpha_{N-1} \alpha_{N-3} \cdots \alpha_1 \xi_W \tag{19}$$

$$\eta_N = \beta_N \eta_{N-1}$$

$$\beta_N = \alpha_N^{(1-q)}$$

In either case, for an even number of stages and constant α, the equations reduce to

$$N/2 + (1 - q) = \frac{\ln(\eta_N/\xi_W)}{\ln \alpha} \qquad (N\ even) \tag{20}$$

By comparing Eq. (17) and (20) with the Fenske–Underwood equation relating the minimum number of stages at total reflux, N_{TR}, to the feed and product compositions and separation factor, α:

$$N_{TR} = \frac{\ln(\eta_N/\xi_W)}{\ln \alpha} \tag{21}$$

it is apparent that

$$N = 2N_{TR} - 1 \qquad (N\ odd) \tag{22}$$

and

$$N = 2(N_{TR} - 1 + q) \qquad (N\ even) \tag{23}$$

for

$$0 < q < 1$$

Thus, for a no-mix cascade the number of stages, N, will be in the range

$$(2N_{TR} - 1) < N < 2N_{TR} \tag{24}$$

The total number of stages in an ideal cascade, N_{IC}, for which $q = 0.5$ ($\beta = \sqrt{\alpha}$ = constant) is given by

$$N_{IC} = 2N_{TR} - 1 \tag{25}$$

This relation is valid regardless of whether an even or odd number of stages is required in the ideal cascade.

RECYCLE RATIO AND TOTAL INTERSTAGE FLOW

It is of practical interest to examine the effect q has on the relationship between the required recycle ratios as a function of stage number and the required total interstage flow in no-mix cascades.

A plot showing the relationship between stage number and recycle ratio is shown in Fig. 4, derived from a no-mix computer model for the separation of the boron isotopes by isotope exchange reaction between gaseous BF_3 and liquid BF_3·Donor (6). The following external cascade variables were assumed:

$z_F = 0.198$	$F \approx 12$ mol/h
$y_P \geq 0.95$	$W \approx 10$ mol/h
$x_W \leq 0.05$	$\alpha = 1.067$ and 1.2

Although no system is known which gives $\alpha = 1.2$ for the BF_3 isotope exchange reactions, this value was included to indicate trends. Conclusions are not limited to isotope exchange cascades but apply to any system with the indicated, constant values of α. The variables F, W, y_P, and x_W

varied slightly from the prescribed values shown above because the cascade designs were limited to a discrete number of stages. However, in all cases the cascades performed essentially the same overall separation.

It is apparent from Fig. 4 that for $q = 0.5$ the recycle ratio increases monotonically from either end of the cascade up to the feed stage. For $q = 0.4$, the recycle ratio for every other stage increases up to the feed stage with the combined plot appearing more as a band than as a smooth curve. Although not shown in these graphs, as the value of q departs further from the central value of 0.5, the recycle ratios required for adjacent stages will exhibit wider oscillations.

The behavior of the cascades exhibited in Fig. 4 for different values of q can be envisioned as follows. For $q = 0.5$, the amount of material and hence the volume or size of each stage in the cascade decreases smoothly from the feed point toward the waste and product ends of the cascade. Thus, the stage volume in an ideal cascade changes monotonically in *one dimension* for $q = 0.5$. For values of $q \neq 0.5$, the stage volume decreases smoothly for either adjacent or opposite stages in the cascade from the feed point to the ends of the cascade as well as from stage to stage. Consequently, stage volume varies in *two dimensions* for values of q not equal to 0.5. This indicates that when $q \neq 0.5$, part of the cascade, consisting of every other stage, is processing more material than the remaining portion, and the work of performing the desired separation is not equally distributed.

Finally, it is advantageous to discern the effect q has on the total interstage flow rate through a no-mix cascade. Using the same no-mix cascade model mentioned previously, the total interstage flow rate was determined for a variety of q values with the results presented graphically in Fig. 5. The important feature indicated by Fig. 5 is that the required total interstage flow is a minimum at $q = 0.5$. Furthermore, as q approaches the limiting values of 0 and 1, the required total interstage flow becomes infinite. This behavior is due to the fact that very large flow rates are necessary at every other stage (where $\beta_i \rightarrow 1$) in order to satisfy material balances required to meet both the stage equilibrium and the no-mix conditions. The flow requirements at these stages will become infinite when $\beta_i = 1$, i.e., when q or $(1 - q)$ equals zero.

Therefore, it is possible to define a no-mix cascade in which mixing of streams with different compositions is eliminated. In the no-mix cascade, minimum total interstage flow may or may not be approached, depending on the value of the exponential factor, q, defined by Eq. (14) and (15). Furthermore, as a special case of the no-mix cascade, the ideal cascade is one in which no mixing of streams with different compositions occurs ($y_{i-1} = z_i = x_{i+1}$ or alternatively $\eta_{i-1} = \zeta_i = \xi_{i+1}$) *and* the heads separa-

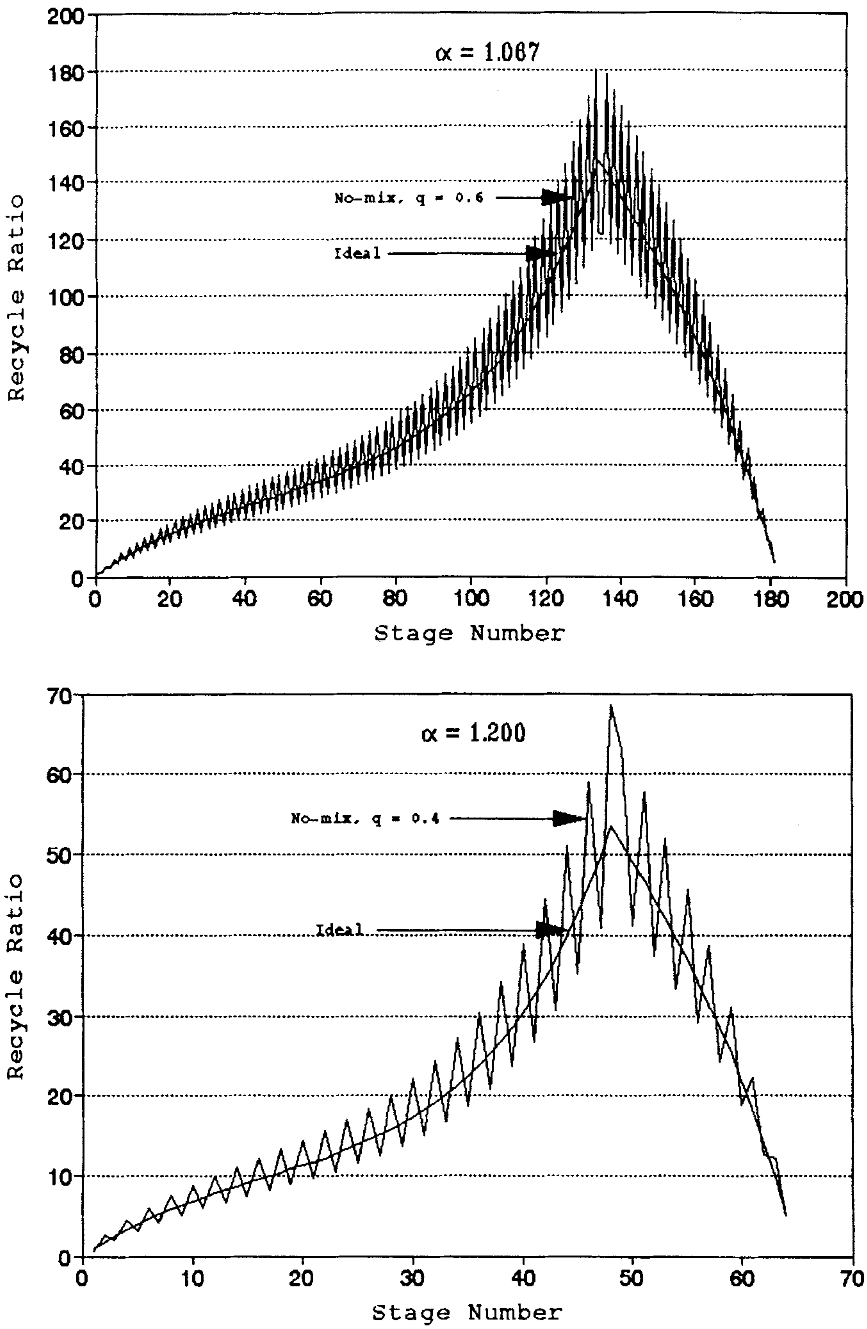

FIG. 4 Recycle ratio as a function of stage number for no-mix and ideal cascades.

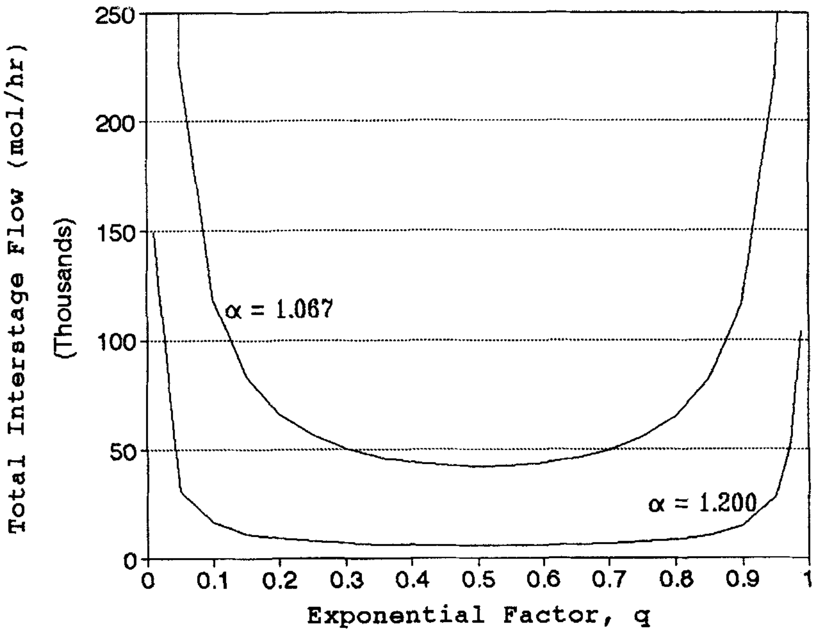

FIG. 5 Total interstage flow as a function of exponential factor q.

tion factor is constant with $\beta = \sqrt{\alpha} = \gamma$. These two conditions insure a cascade with minimum interstage flow, an *ideal cascade* by definition.

NOMENCLATURE

F	flow rate of feed stream
M	flow rate of heads stream
N	flow rate of tails stream, number of stages in a cascade
P	flow rate of enriched product stream
R	stagewise recycle ratio
W	flow rate of depleted product (waste) stream
x	composition of tails or waste stream
y	composition of heads or product stream
z	composition of feed stream

Greek Letters

α	stage separation factor
β	heads separation factor
γ	tails separation factor

ξ	ratio of binary compositions in tails stream
ζ	ratio of binary compositions in feed stream
η	ratio of binary compositions in heads stream

Subscripts

F	feed stage
i	*i*th stage in a cascade
IC	ideal cascade
N	*N*th or product stage in a cascade
NM	no-mix
min	minimum flow rate or recycle ratio
P	product or *N*th stage in a cascade
TR	total reflux
W	waste or 1st stage in a cascade

Superscripts

N	number of stages in a no-mix cascade
N_{TR}	number of stages at total reflux in Fenske–Underwood equation
q	exponential factor

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