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Separation by Thermal Diffusion in a Rotary Column

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Abstract

A rotary thermal diffusion column with the inner cylinder rotating and the outer cylinder static was used to separate *n*-heptane–benzene mixtures at different speeds of rotation. The results show that the column efficiency depends on the speed of rotation. For the optimum speed the increase in efficiency relative to the static column was of the order of 8%. The role of the geometric irregularities in the annulus width on performance of the rotary column is also discussed.

INTRODUCTION

In the separation of components by thermal diffusion, the role played by the natural convection currents existing inside the annular space of a thermogravitational column has been well established. On the one hand its intrinsic cascading effect promotes the development of a vertical concentration profile, but on the other hand it also induces a remixing effect which counteracts the separation process.

Several means have been suggested to control the convective currents;

namely, the modification of the basic design of the conventional column, but the net result has been in most cases either an improvement in the degree of separation at the cost of longer times to reach equilibrium separation or a reduced flexibility in the allowable operating conditions.

The modified designs of the basic thermogravitational column so far proposed are of two main types:

(1) Columns in which the modifications merely deflect the particles streamlines without introducing any additional external driving force, so that the flow remains by natural convection. Examples include the use of packing, barriers, inclining the column, or more sophisticated methods such as the "permeable immiscible layer-column" recently suggested (*1*).
(2) Columns in which some external driving force is superimposed, adding forced convection to the natural convection either in the same or different directions. Examples where the forced convection had the same direction as the natural convection include the "external reflux" column of Von Halle (*2*) and the "moving-walls" column (*3*). The rotary column with one or both cylinders rotating is the example where the natural and forced convection are not in the same direction.

The fact that in a rotary column the forced and natural convection are not in the same direction implies that three spacial coordinates are required to describe the hydrodynamics inside the annular space, in contrast to the other types of columns where two dimensions are sufficient. This 3-dimensional nature of the rotary column, besides making it a "unique" apparatus, is, indeed, the main cause for the difficulties that have been encountered in deriving a phenomenological model for its operation.

The hydrodynamics associated with the rotary column become more complicated at higher speeds of rotation. In fact, it has been well established for these columns with the outer cylinder static that, above a certain speed of rotation—the critical velocity—the flow inside the annulus, though still laminar, becomes unstable and a steady secondary flow formed by pairs of counter-rotating ring-shaped vortices—the so-called Taylor vortices—arises (*4*, *5*).

The identification of the actual flow regime was achieved by Chandrasekhar (*5*) through a dimensionless number called the Taylor number, Ta, after the pioneering work of Taylor (*6*). The Taylor number may be defined as

$$\mathrm{Ta} = \frac{4r_1^2(2\omega)^4}{r_2^2 - r_1^2}\left(\frac{\Omega\rho}{\eta}\right)^2$$

where r_1, r_2 = the cylinder radii
2ω = the annulus width
ρ = the fluid density
η = the fluid viscosity
Ω = the speed of rotation of the inner cylinder

For narrow annuli, the critical value of Ta corresponding to the transition to a "laminar plus Taylor vortices" regime was found to be of the order of 3.0×10^3 (*5, 7*).

So far it would seem that only those rotary columns in which the outer cylinder is static have been used experimentally. The first report was in 1955 by Sullivan et al. (*8*) who, according to their own description of the flow pattern observed, appear to have worked in the "laminar plus Taylor vortices" region.

The results presented by these authors were most promising since they showed that a dramatic increase in the column efficiency was obtained by rotating. Somewhat surprisingly, only 15 years later was the subject reconsidered; this time Romero (*9*), who studied the rotary column performance in some detail, reached quite different conclusions from Sullivan et al. (*8*). In fact, not only did Romero observe a decrease in efficiency when the column was allowed to rotate, but also that the separation decreased as the speed increased. Furthermore, at speeds above the critical velocity the separation observed by Romero was almost negligible.

A slightly different picture was presented by Bott (*10*) in his study of the first period of the transient phase [broadly, the region where the Ruppel and Coull equation applies (*11*)]. This author noted that in this period the separation was enhanced by rotating the column, the results at different speeds of rotation being of the same order of magnitude.

From this brief discussion it becomes apparent that at the present state of knowledge it is not possible to appreciate the true potential of the rotary column, let alone the influence of the speed of rotation on the performance of the apparatus. The objective of the present work is to obtain information on these aspects in order to assess the worthiness of the rotary apparatus relative to other more conventional columns.

The discussion is restricted to batch operation since not only is it the one of greater interest for concentric cylinders but also because it allows an easy extrapolation for continuous operation if necessary.

EXPERIMENTAL

The experimental apparatus used in this work has already been described elsewhere (*10*) so that only the principal characteristics are mentioned. It is comprised of two concentric cylinders made of brass, the height being 102 cm and the annular volume of about 80 cm^3. The inner cylinder may rotate at speeds up to 300 rpm while the outer cylinder is static. The heating and cooling of the walls is made through thermostatic water circuits which flow inside the inner cylinder (hot wall) and through a water jacket surrounding the outer cylinder (cold wall). The wall temperatures are measured by a group of eight thermocouples placed in each wall at four different heights. The column has 11 sampling ports equally spaced along the height.

The test mixture used was *n*-heptane–benzene with an initial composition of $C_0 = 0.560$ (molar fraction of benzene), each component being of high purity (*12*). The analysis of the mixture composition was by refractometry with an accuracy greater than 0.001 of the molar fraction.

The temperature difference between hot and cold walls was $18.5 \pm 0.25°C$ and the average temperature $20 \pm 2°C$. The vertical variation of the temperature was in no case larger than 0.5°C.

The rate of sampling was of 1 sample/run in accordance with the recommendations of Vichare and Powers (*13*), i.e., after a sample was withdrawn, the column was emptied, washed with fresh mixture, emptied again, and finally filled up with fresh mixture for a new run.

In the runs referred to as 1B, the column was operated at full height (102 cm) and in Runs 2B with a height of 91 cm by leaving the upper section empty. This was intended as a means of assessing the influence of a suspected imperfect zone located near the top of the column (between $L = 91$ cm and $L = 102$ cm) on the column efficiency.

The i.d. of the outer cylinder and the o.d. of the inner cylinder were carefully measured (*10*) at 9 different points equally spaced along the height. The mechanically measured (or theoretical) annulus width is obtained by subtracting the cylinder radii at corresponding heights. The average values of these measurements are listed in Table 1 for Runs 1B and 2B. The table shows that not only do Runs 1B and 2B differ in the height of column used, but also on the mean value of the theoretical annulus width, i.e., Runs 1B and 2B represent virtually two geometrically

TABLE 1

Column Dimensions and Operation Velocities

	Height, L (cm)	$(r_2 - r_1)_{av}$ (cm)	Speed of rotation, N (rpm)
Column 1 (Runs 1B)	102	0.0571	0, 1, 3, 6, 12, 28, 48, 88
Column 2 (Runs 2B)	91	0.0565	0, 1, 3, 10, 20, 44

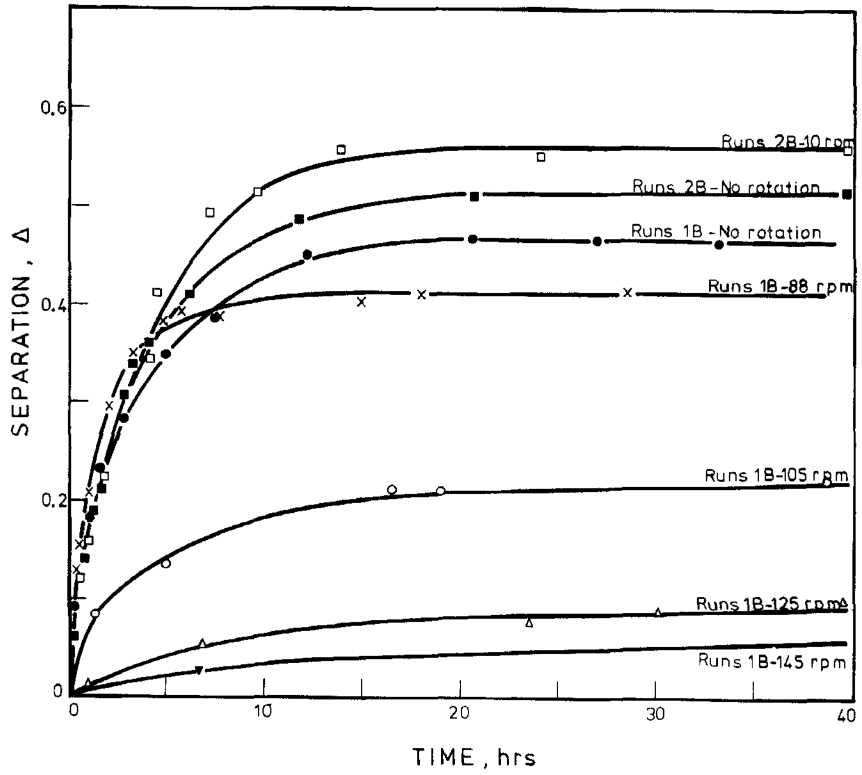

FIG. 1. Typical examples of the experimental separation vs time curves.

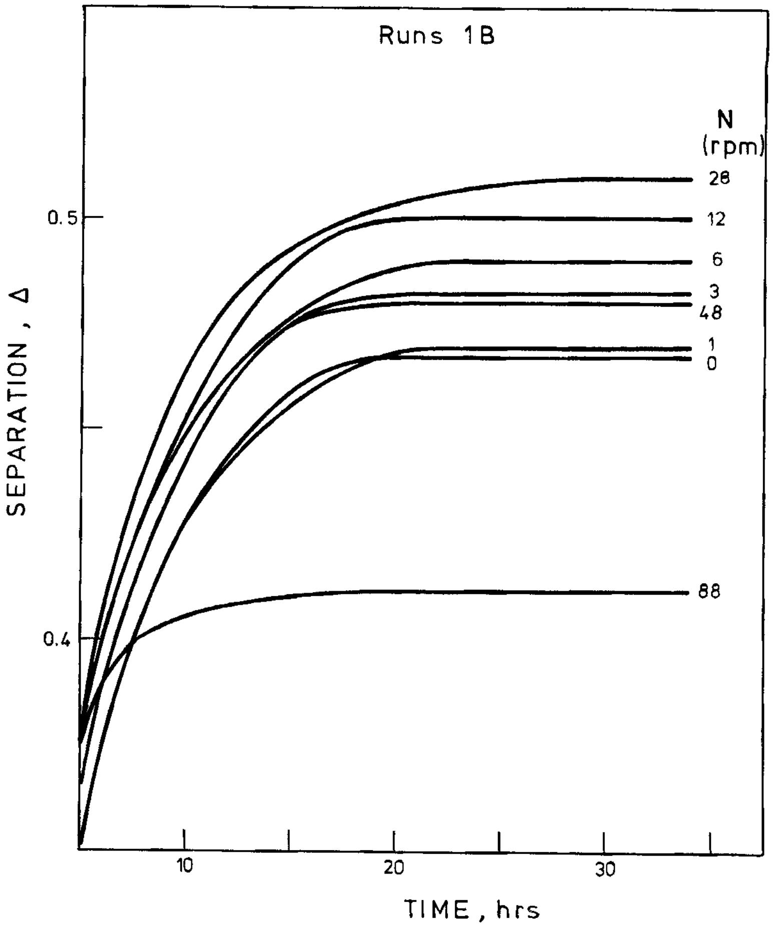

FIG. 2. Experimental separation curves ($t > 5$ hr) for different speeds of rotation in Column 1 (Runs 1B).

different columns which may be simply designated by Columns 1 (Runs 1B) and 2 (Runs 2B). Table 1 also shows the various values of the speed of rotation at which the columns were operated.

For each speed of rotation a curve of the degree of separation, Δ—defined as the difference in molar fraction between the top and bottom of the column—as a function of the time of operation was determined. Typical examples of these curves are shown in Fig. 1. Figures 2 and 3 show the experimental curves obtained in, respectively, Columns 1 and 2 for times greater than 5 hr. For $t < 5$ hr the various curves are virtually superimposed on one another.

DISCUSSION

Separation in the "Laminar Plus Taylor Vortices" Region

For the experimental conditions used in this work, the critical speed of rotation, N^L, above which the Taylor vortices occur is 99 ± 10 rpm. This means that of the speeds used in Column 1, $N = 125$ and 145 rpm, are undoubtedly in the "laminar plus Taylor vortices region" and $N = 105$ rpm, most probably, also has vortex circulation.

From the experimental curves shown in Fig. 1 it is apparent that the column efficiency in the region above the critical velocity is dramatically reduced in comparison with the other cases in the pure laminar region without Taylor vortices. This conclusion substantiates the findings of Romero (*9*) and disagrees strongly with the results of Sullivan et al. (*8*).

It is interesting to note also that the present results agree well with the heat transfer studies on this type of apparatus conducted by Tachibana et al. (*14*). These authors observed that the vortex circulation promoted the homogenizing of the temperature across the annulus. It is reasonable to admit that the same kind of homogenizing effect will affect the mass transfer process in the horizontal direction, which means that the horizontal concentration gradient (caused originally by the Soret effect) will be decreased with the consequent reduction in the separation potential of the column.

In conclusion, it appears that the interest in rotary operation should be directed toward the pure laminar region without Taylor vortices.

Separation in the Pure Laminar Region

Examination of Figs. 2 and 3 shows quite clearly that the performance of the rotary column is affected by the speed of rotation. Moreover, the

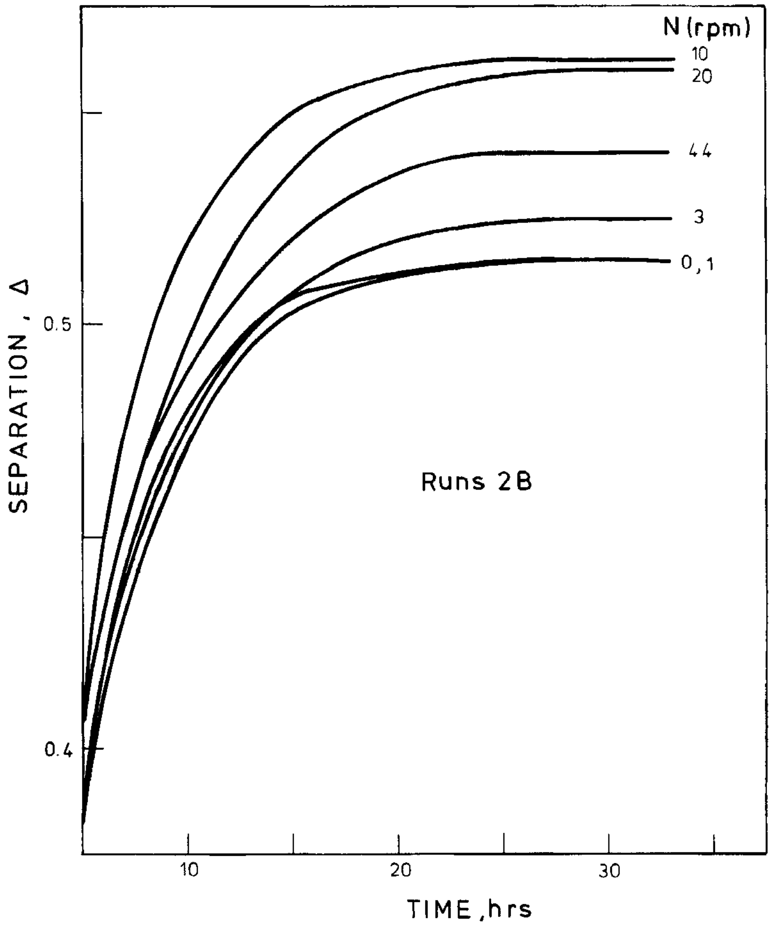

FIG. 3. Experimental separation curves ($t > 5$ hr) for different speeds of rotation in Column 2 (Runs 2B).

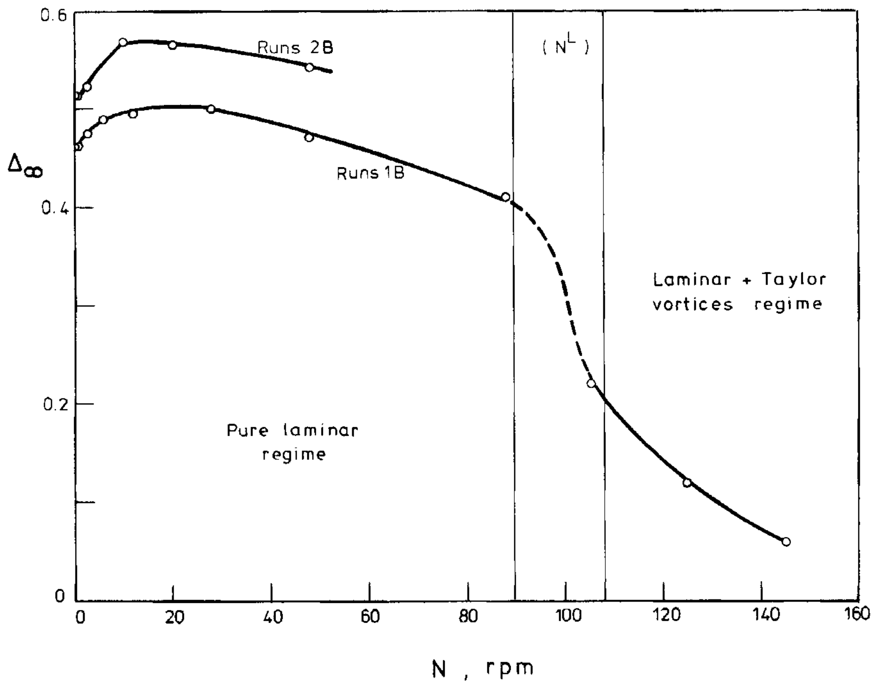

FIG. 4. The experimental variation of the equilibrium separation, Δ_∞, with the speed of rotation of the inner cylinder, N, and theoretical flow regime.

experimental curves also indicate that the column efficiency may be greater in rotary than static operation. This is particularly true for larger values of the time near to the steady-state situation where significantly different degrees of separation are obtained for different speeds of rotation (particularly if one discounts the curve at $N = 88$ rpm due to its proximity to the critical velocity).

The influence of the speed of rotation upon the separation attainable at equilibrium is better emphasized by plotting the steady-state separation, Δ_∞, against the speed of rotation. This is done for both columns in Fig. 4 which, on examination, suggests the following observations:

(1) The rotation increases the separation capability of the column over a wide range of speeds of rotation. The maximum improvement in efficiency occurs in the region of 10 to 20 rpm and is of

the order of 8%. As the speed increases, there is a steady decrease in the efficiency of the column.

(2) Near the critical speed of rotation, N^L, there is a marked drop in the column efficiency, confirming the undesirable nature of the vortex circulation.

These results indicate that the rotary column does not produce the dramatic improvement in the separation claimed by Sullivan et al. (*8*), but rather a moderate improvement much more in the line of the results reported by Romero (*9*) and Bott (*10*).

The Influence of Geometric Irregularities

Since Bott and Romero (*15*) and Korchinsky and Emery (*16*) have pointed out the very serious influence of geometric irregularities (mainly eccentricities) of the annular space on the performance of thermal diffusion columns, it is of importance to discuss, though briefly, the role these irregularities play in the rotary column.

Bott and Romero (*15*) characterized the "degree of imperfection" of a column through the definition of an "equivalent annulus width"—the annulus width of a geometrically perfect column that yields the same separation as the actual imperfect one, the other conditions being constant. In practical terms this is equivalent to use for the value of the annulus width, 2ω, not the mechanically measured radii difference, $(r_2 - r_1)$, but rather

$$2\omega = (r_2 - r_1)v$$

The factor v in this equation is obviously equal to unity for a perfect column and increases as the degree of imperfection increases.

The method of evaluating v for static columns was presented by Romero and Pinheiro (*17*), and its application to Columns 1 and 2 yields, respectively,

$$v_1 = 1.075$$

$$v_2 = 1.020$$

which means that Column 1 is more imperfect than Column 2. The importance of the concept and its evaluation becomes apparent when noting that the introduction of the factor v reverses the relationships between the two columns, i.e., while the (theoretical) ratio $L/(r_2 - r_1)^4$ is larger for Column 1, the new (practical) ratio $L/[(r_2 - r_1)v]^4$ is larger for Column 2.

Therefore, Column 2 must, in practice, yield the larger degree of separation.

The above discussion, valid for the static operation, explains clearly the experimental points for $N = 0$ shown in Fig. 4. Yet, simple observation of the same figure strongly suggests that the rotation does not alter—at least qualitatively—the relative efficiency of the two columns, i.e., the greater values for Δ_∞ are still obtained in Column 2 at any speed of rotation. As a direct consequence, one must therefore admit that it is necessary to consider also a "rotary equivalent annulus width" or, briefly, that this "rotary equivalent annulus width" is not, in principle, strictly equivalent to the static concept of Bott and Romero (*15*), mainly because the hydrodynamical considerations upon which these authors based the concept cannot be applied to the rotary column hydrodynamics.

In the rotary column the effect of an annular eccentricity can be visualized as making the quasi-tangential flow pass alternatively through "enlargements" and "funnels" in a somewhat pulsating pattern. It seems logical that this pulsating flow will grow in importance (frequency) as the speed of rotation increases. Moreover, it is also apparent that the amplitude of this periodical pattern is closely related to the existing eccentricities which, in turn, may be described through the static v factor (*17*). It may, therefore, be concluded that the rotary factor v_{rot} must depend on both v_{static} and N.

In accordance with the above arguments, it is now possible to interpret further the experimental results; namely, the shape of the curves shown in Fig. 4 for speeds below the critical region. At low values of N the predominant factor contributing to v_{rot} is v_{static} and, since for $N = 0$, $v_{\text{rot}} = v_{\text{static}}$, for small values of N, the two factors—v_{rot} and v_{static}—must have close values. But, as the speed of rotation increases, so does v_{rot}, therefore reducing the separation potential of the column. This negative effect is, however, at least in part, compensated by the increase in cascading associated with the rotation which tends to improve the efficiency of the column. This positive effect is greater for higher speeds of rotation.

The experimental curves of Δ_∞ vs N as shown in Fig. 4 represent, therefore, the net effect of the rotation in the sense that they represent the combined result of two basic effects.

From this viewpoint the relative improvement in efficiency brought about by rotation as well as the location of the optimum speed will depend essentially on the type and degree of existing imperfections. In the work under discussion, the less imperfect apparatus (Column 2) showed the greater improvement after rotation, with an optimum speed of about

10 rpm which is lower than for Column 1 (~20 rpm). The present data are not sufficient to allow for a generalization.

It seems that it will be necessary to consider in some detail the role of the geometric irregularities before attempting to compare any experimental results with possible eventual theories or to draw any definitive conclusion on the true worthiness of the rotary apparatus.

CONCLUSIONS

The principal conclusions suggested by the present work may be summarized as follows.

(1) The separation in a rotary column at speeds above the critical Taylor number (i.e., with Taylor vortices) is much smaller than that obtained in the pure laminar region, and therefore of little practical interest.

(2) At speeds below the critical Taylor number, the efficiency of the rotary column used varied with the speed of rotation. For small values of the speed of rotation, the efficiency increased with the speed, reaching a relative maximum at about 10 to 20 rpm, and then decreased monotonically as the speed increased. At equilibrium the maximum improvement in efficiency relative to the static operation was of the order of 8%.

(3) As in the static column, it appears that geometric irregularites in the annulus width will greatly affect the column performance and may conceal the true effects of other variables such as the speed of rotation.

SYMBOLS

L	column length
N	speed of rotation of the inner cylinder (rpm)
N^L	critical speed above which Taylor vortices occur (rpm)
r_2, r_1	cylinder radii
Ta	Taylor number

Greek Letters

Δ	degree of separation = difference in molar fraction between the top and bottom of the column

Δ_∞	equilibrium separation
η	viscosity
ν	equivalent annulus width factor
ρ	density
ω	half distance between the hot and cold walls
Ω	speed of rotation of the inner cylinder (rad/sec)

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REFERENCES

1. J. de D. R. S. Pinheiro and T. R. Bott, *Sep. Sci.*, *11*(2), 193 (1976).
2. E. Von Halle and S. Jury, *AIChE J.*, *13*, 709 (1967).
3. J. H. Ramser, *Ind. Eng. Chem.*, *49*, 155 (1957).
4. J. Kaye and E. C. Elgar, *Trans. ASME*, *80*, 753 (1958).
5. S. Chandrasekhar, *Mathematica*, *1*, 5 (1954).
6. G. I. Taylor, *Philos. Trans. R. Soc. London, Ser. A*, *223*, 289 (1933).
7. R. Donnely, *Proc. R. Soc. London, Ser. A*, *246*, 312 (1958).
8. L. Sullivan, T. C. Ruppel, and C. B. Willingham, *Ind. Eng. Chem.*, *47*, 208 (1955).
9. J. J. B. Romero, *DECHEMA-Monogr.*, *65*, 337 (1971).
10. T. R. Bott, *Chemeca 70*, Butterworths, Australia, 1970, p. 35.
11. T. C. Ruppel and J. Coull, *Ind. Eng. Chem., Fundam.*, *3*, 368 (1964).
12. J. de D. R. S. Pinheiro, M. M. S. Pinheiro, and J. J. B. Romero, *Rev. Fis. Quim. Eng., Ser. A*, *25*, 1 (1973).
13. G. G. Vichare and J. E. Powers, *AIChE J.*, *7*, 650 (1961).
14. F. Tachibana, S. Fukui, and H. Mitsumura, *Bull. JSME*, *3*, 119 (1960).
15. T. R. Bott and J. J. B. Romero, *Trans. Inst. Chem. Eng.*, *47*, 166 (1969).
16. W. J. Korchinsky and A. H. Emery, *AIChE J.*, *13*, 224 (1967).
17. J. J. B. Romero and J. de D. R. S. Pinheiro, *Chem. Eng. Sci.*, *30*, 1459 (1975).

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