V = dispersed phase superficial flow rate in extraction column, cu.ft./(sq.ft.)(hr.)
v = drop velocity, cm./sec.
x = solute concentration in continuous phase of extraction column, lb.-moles/cu.ft.
y = solute concentration in dispersed phase of extraction column, lb.-moles/cu.ft.
$\Delta\rho$ = density difference between phases, g./ml.
μ_c = viscosity of continuous phase, centipoises [g./(cm.)(sec.) in Reynolds number]

Subscripts

a = CCP sample
b = combined sample of CCP and wake
d = diluted DCP sample
i = wake just after drop formation or to DCP
o = reference solution
1 = continuous phase inlet end of extraction column
2 = continuous phase exit end of extraction column

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Continuous Flow in Packed Thermal Diffusion Columns

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A parallel-plate thermal diffusion column packed with glass wool was operated in continuous flow. The standard equation used for continuous flow through nonpacked columns satisfactorily correlates separation and flow rate. The packed-column theory correctly gives the effects on the regression coefficients of plate spacing and packing permeability, the two variables unique in the packed column. The theory gives poor approximations of the absolute magnitudes of the coefficients, however, which is the same situation found in results from nonpacked columns.

The ordinary open (or nonpacked) thermal diffusion column is difficult to construct and very difficult to use in theoretical research because the wall spacing must be very small, on the order of 0.02 in. or less. However, the same mass separations may be obtained with wall spacings as large as ¼ in. if some packing material fills the working space. Debye and Bueche (*1*) first reported this advantage in 1948, and some work on the operating variables was conducted by Sullivan et al. in 1957 (*2*).

Lorenz and Emery (*3*) developed the theory for packed thermal diffusion columns, and showed that in batch operation this theory is as successful with packed columns as the classical theory is with open columns (*4*). The

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object of the work reported here was to test the theory of the packed column in continuous flow.

EXPERIMENTAL WORK

The thermal diffusion column and technique used were about the same as those used by Lorenz (*4*), modified as necessary for continuous flow. The column, shown in Figure 1, consisted of parallel plates, with heat supplied by steam and removed by cold water. The plates were spaced by a ⅛-in. steel spacer and two thin Teflon gaskets. The 50 mole % cumene in cetane feed was introduced through three ports distributed along the midpoint of the column, and the product streams were withdrawn at about equal rates from sets of three ports at the extreme ends of the column. The column was equipped with two pressure taps in the central portion to determine the permeability of the packing.

Two series of runs were made with coarse glass wool of about 30-μ diameter as packing material, and two series with fine glass wool of about 10-μ diameter. The glass wool was prepared outside the column by placing it between plates spaced the same distance as in the column, applying plaster to the edge of the packing to confine the fluid and avoid edge problems in the column, and filing the straggling fibers off. The packing was then placed inside the spacer and the column was bolted together.

The flow through the column was produced by a constant-head tank, and controlled by two banks of capillary tubes, the path through which could be varied by valves. Flow was maintained constant until the compositions of the product streams remained invariant, which sometimes took as long as 9 hr. at low flow rates. Analyses were made by a precision refractometer.

After a series of runs at varying flow rates, the column was cleaned with a solvent, the solvent was evaporated, and the permeability was determined by measuring the pressure drop associated with the flow of pure cumene through the column under isothermal conditions. Finally, the column was opened and the quantity of glass wool in the packing determined.

Some preliminary runs verified Lorenz' observation that it is very difficult to arrange for a reproducible packing. Merely by reversing the packing (rotating around the vertical axis), we changed values of the concentration separation Δ from 2 to 5% of the concentration difference. This suggests that handling the packing changes the configuration of the fibers and thus the operating characteristics. In the determination of permeability, it was possible to obtain values of permeability which varied from each other by 7% by changing the flow rate of cumene. Theoretically, this should not have changed, and the fact that it did suggests that the configuration of the packing is sensitive to the flow rate through it.

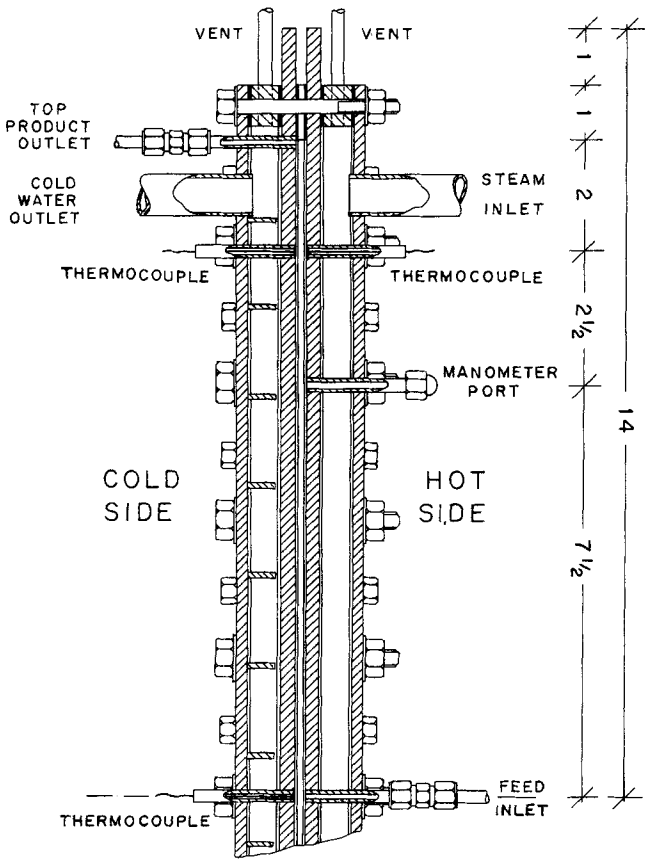

Fig. 1. Parallel-plate thermal diffusion column, top half only. The section below the feed inlet is essentially a mirror image of the top. Horizontal obstructions in the cold water jacket are baffles.

RESULTS

Figure 2 shows separation Δ as a function of feed rate for one of the four series of runs. When the product of the two concentrations of the binary system is assumed to be constant, theory gives the separation for the top part of the column as

$$\Delta_e = \frac{\Delta_o K}{L \sigma_e}\left[1 - \exp\left(-\frac{\sigma_e L_e}{K}\right)\right] \qquad (1)$$

The constant Δ_o, which may be treated as a phenomenological constant, is given by the theory by the expression

$$\Delta_o = \frac{5L}{8g} P_1 V_1 \qquad (2)$$

in which P_1 is a function of the properties of the fluid

$$P_1 = \frac{\alpha}{\beta}\left(\frac{D\mu}{T}\right) \qquad (3)$$

and V_1 is a function only of the variables plate spacing and permeability,

$$V_1 = \frac{H(Y)}{w^4 K(Y)} \qquad (4)$$

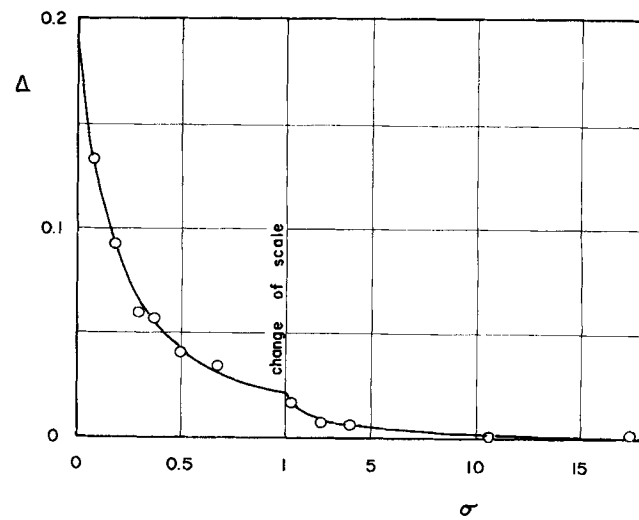

Fig. 2. Representative results showing the variation of separation Δ as a function of feed rate σ for one of the four series of runs. The line is given by Equation (1) and the least-squares values of the phenomenological constants Δ_o and K.

$$H(Y) = \left[1 - \frac{3}{Y^2}(Y \operatorname{ctnh} Y - 1)\right] \Big/ Y^2 \qquad (5)$$

$$K(Y) = \left[1 - \frac{5}{Y}\operatorname{ctnh} Y - \frac{15}{Y^4} - \frac{15}{4Y^2} + \frac{15}{4Y^3}\operatorname{ctnh} Y + \frac{45}{4Y^2}\operatorname{ctnh}^2 Y\right] \Big/ Y^4 \qquad (6)$$

$$Y = w/\sqrt{k} \qquad (7)$$

The constant K, which may also be treated as a phenomenological constant, is given by the theory as

$$K = \frac{g^2 B\,(\Delta T)^2}{15} P_2 V_2 \qquad (8)$$

in which P_2 is a function only of the properties of the fluid

$$P_2 = \beta^2 \rho / \mu^2 D \qquad (9)$$

and V_2 is a function of the variables plate spacing and permeability

$$V_2 = \epsilon w^7 K(Y) \qquad (10)$$

Equation (1) is the same expression as that for the open column, except that the theoretical expressions for Δ_o and K are different. For the packed column, a new parameter, the permeability of the packing, must be included. A similar expression holds for the bottom part of the column, with $(1\text{-}c)$ substituted for c.

The data for each series were subjected to regression on Equation (1) and the best values of Δ_o and K were determined, along with their standard errors. For continuous work, H and K are a more natural pair of constants, but we wanted to compare the results with the batch work on packed columns of Lorenz, and in batch work Δ_∞ and K are the obvious constants. Δ_∞ from the batch work is theoretically the same as Δ_o from continuous work. The solid line in Figure 2 is a good representation of the experimental points. Thus Equation (1) may be safely used in continuous flow thermal diffusion work as a phenomenological equation.

Effects of Plate Spacing and Permeability

To compare these results with those of Lorenz, we present Figures 3 and 4. In Figure 3 Δ_∞ from the batch runs and Δ_o from the continuous runs (which should be the same) are plotted against the function V_1, which contains those parameters peculiar to the packed column. It appears that the continuous runs fall a bit below the batch. However, this is not a safe conclusion, since the points from the batch runs show greater than expected deviation from the best straight line. That is, only about two-thirds of the batch points have $2s$ limits which overlap the line, and Lorenz thus concluded that there are sources of random error (most notably the difficulty in obtaining a reproducible packing) which contribute to variations between series of runs. Thus we feel that the continuous runs give about the same result as the batch.

The situation is not as good in the case of K, shown in Figure 4. Here the values from the continuous runs are significantly different from those from the batch,

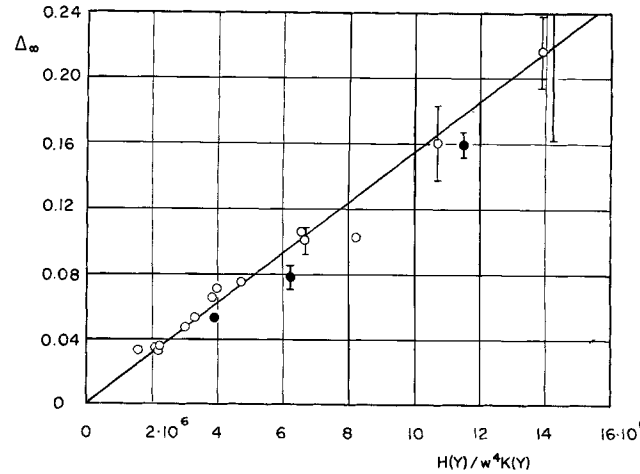

Fig. 3. Variation of the phenomenological constant Δ_∞ (or Δ_o for the continuous work) with V_1, the parameter giving the theoretical effect of plate spacing and packing permeability. Open points, data of Lorenz (4); solid points, present work. Vertical bars are 2s limits, as calculated from the scatter of the data points around the regression curve (such as that in Figure 2); where not shown, the 2s bar is smaller than the point symbol; 2s limits in the abscissa are in all cases smaller than the symbol.

whereas they should be the same. They differ in a very consistent manner, since a good straight line through the origin may be drawn through each set. This difference might be caused by some consistent and reproducible difference in the handling of the packing from Lorenz. Small differences in handling of the packing can make

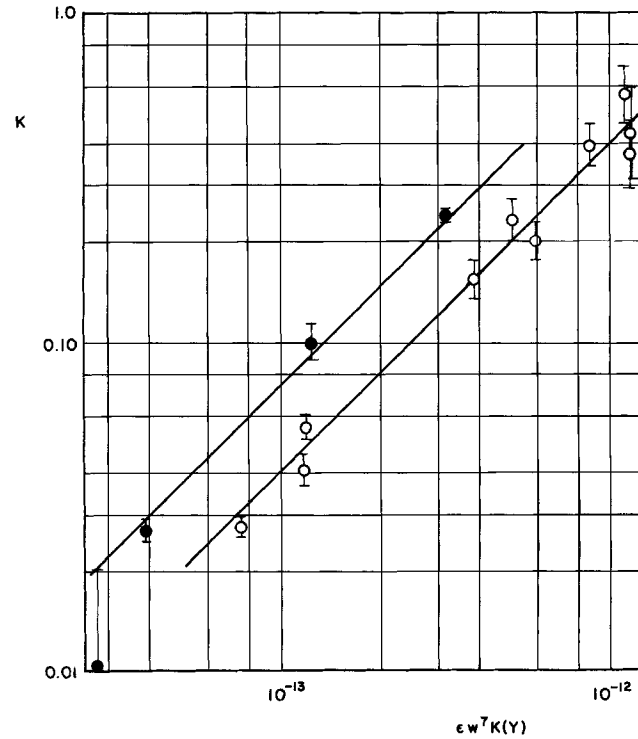

Fig. 4. Variation of the phenomenological constant K with V_2, the parameter giving the theoretical effect of plate spacing and packing permeability. Open points, data of Lorenz (4); solid points, present work.

perceptible differences in the results, as we noted under Experimental Work. However, we feel it unlikely that these errors are this large.

Another possible cause of the difference might be a wall effect, since the throughput shifts the position of maximum velocity toward the center. Emery and Lorenz (*4*) noted that a wall effect is a likely explanation for a deviation in their batch data. However, they found this deviation only at their smallest plate spacing, 1/16 in. and not at plate spacings at or above ⅛ in., and a spacing of ⅛ in. was used in the present work.

The most likely explanation of the discrepancy between batch and continuous results is shifting of the packing. The problem with fibrous packings of this type is that the fibers are fairly free to move, and probably do move with changes in flow rate, as we observed in the permeability measurements. The implication of this for the values of Δ_o and K is that the value of Δ_o is probably relatively unaffected by this shifting, since it is determined by the intercept at zero flow rate, whereas the value of K is probably sensitive to it, since it is determined from the effect of flow rate. If this is to happen, increasing flow rate should increase the permeability consistently. This seems a plausible circumstance; one might expect the packing to shift in a way that would reduce the resistance to flow.

The problems with shifting packing suggest that another kind of packing should be used if reproducibility and theoretical agreement are the objectives. The one type that first comes to mind is granular packing, such as glass beads. Unfortunately, this kind of packing introduces another complication, a very low permeability. Lorenz found that to obtain values of K large enough to give experiments that lasted no longer than a day, the theory indicated that the size of the glass bead would be almost the size of the plate spacing, which is untenable for checks of the theory.

Absolute Values of the Constants

The theoretical equations are supposed to give not only the effects of the variables, but also the absolute values of the constants Δ_o and K. To check this, the values of P_1 and P_2 (and $2s$ limits) calculated from the results are shown in Table 1 along with values predicted from the theory, Equations (3) and (9). Both are collections of properties of the fluid system which have been measured independently.

In only one of these possible comparisons are the values not significantly different, the continuous and theoretical values of P_1. The batch value of P_1 is not far off, but it is significantly different from the other two. In the case of P_2, there is no agreement at all.

Thus the conclusion is the same for continuous work as for batch, that the theory does not predict well the absolute values of the phenomenological constants. Of course this is not much of a condemnation of the packed theory, since the same situation prevails in the case of the open thermal diffusion column, namely, limiting values of Δ_o are not too far off, but experimental values of K do not agree with the theoretical predictions (5, 6).

Table 1. Comparison of Absolute Magnitudes

Group	P_1	P_2
Experimental, batch	$(4.75 \pm 0.06)10^{-7}$	101 ± 4
Experimental, continuous	$(4.1 \pm 0.3)10^{-7}$	180 ± 10
Theoretical	$(3.5 \pm 0.7)10^{-7}$	470 ± 60

CONCLUSIONS

1. The standard equation for continuous operation of open columns, Equation (1), may be safely used as a phenomenological equation for packed columns.
2. The packed-column theory correctly gives the effects of plate spacing and packing permeability, the two variables whose functionality is unique in the packed bed.
3. The theory gives reasonable approximations of limiting values of Δ_o, but poor approximations of K; this is about the same situation as for open columns.

NOTATION

B = width of the column parallel to the plates
D = diffusion coefficient
g = acceleration of gravity
$H(Y)$ = function of Y as given by Equation (5)
k = permeability of the packing
K = coefficient as defined by Equation (8)
$K(Y)$ = function of Y as given by Equation (6)
L = total column height
L_e = height of the top (or enriching) section of column
P_1, P_2 = combinations of properties of the fluid, as defined by Equations (3) and (9)
s = standard error
T = absolute temperature
V_1, V_2 = combinations of apparatus parameters, as defined by Equations (4) and (10)
w = half-width of the column perpendicular to the plates
Y = dimensionless half-width as defined by Equation (7)

Greek Letters

α = thermal diffusion constant
β = $-\partial\rho/\partial T$
Δ = difference in concentration between top and bottom product
Δ_o = Δ at the limit of zero throughput
Δ_e = difference in concentration between top product and feed
Δ_∞ = for the batch column, difference between top and bottom concentration in the column at infinite time
ϵ = void fraction in the packing
μ = viscosity
σ = feed rate to the column
σ_e = top product rate
ρ = density of the fluid

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