Studies on the Thermal Separation Column. V. Thermal Separation Column Having Horizontal Barriers

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The performance of a hot wire type thermal separation column in which many horizontal barriers of Teflon or copper plates are installed has been examined. At the optimum spacing of barrier plates, the quality factor defined by $(2AL)_{barrier}/(2AL)_{open}$ exceeds 3. This means that a barrier column constructed properly has a performance equivalent to that of an open column of three times taller in height. Visualization of a stream line in the barrier assembly indicates that a helical transverse motion takes place in the space partitioned by two adjacent barrier plates.

We have studied the effect of installing flow barriers inside the gas chamber of a thermal separation column.[1–3] It was concluded that the presence of a flow barrier retards the convectional velocity of separating gas mixture and the largest separation is attainable when convectional velocity is minimized. A similar conclusion was also obtained from the study of a tilted column.[4]

Although the use of a vertical barrier has been proved effective to improve the performance of thermal separation, there are some difficulties as regards the construction of the vertical barrier system.

This paper deals with the effect caused by the use of horizontal barriers in place of vertical ones. The structural difficulty in the vertical system can mostly be eliminated by use of the horizontal barrier system for which there is no difficulty in selecting the standard of comparison.[3] Utilization of horizontal barriers was first studied by Treacy and Rich[5] who claimed the merit of the barriers. However, the procedure of the experiment and the data do not seem convincing.

Experimental

Apparatus. An outline of the column is shown in Fig. 1. A straight column employing an iron tube for the outer wall of the gas chamber is fixed vertically as accurately as possible. Inclination of a column seriously affects the degree of equilibrium separation.[4] The upper end of the center wire (diam. 0.23 mm, nichrome) (9) is tightly fixed to the column top which is electrically insulated from the column body. A micalex rod (1) was used for this part. At the lower end of the heater wire, a copper rod (10), 99 g, is connected to assure the verticality and tautness of the heater wire. A mercury pool (11) is provided in contact with the tail end of the weight to make allowance for the thermal elongation of heating wire. The elongation is measured with a travelling microscope in order to estimate the heater temperature. The length of the column as measured by the distance between two sampling ports (5) at top and bottom is 54 cm.

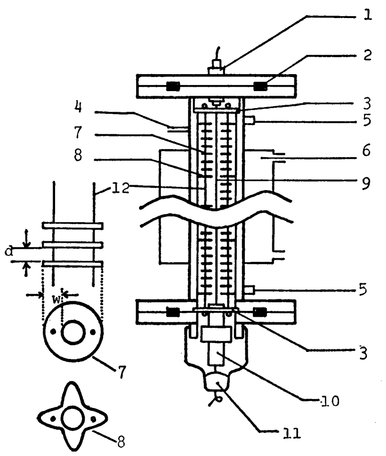

Fig. 1. Illustration of barrier-column.
1: Micalex rod, 2: O-ring seal, 3: terminal plates, 4: gas inlet, 5: sampling port, 6: water jacket, 7: flat doughnut barrier, 8: cruciform barrier, 9: nichrome heater, 10: weight, 11: mercury pool, 12: fine wire.

Barriers. The horizontal barrier assembly consists of many plane parallel plates made from Teflon (0.8 mm thick) or copper sheets (0.55 mm thick). Each barrier plate is in the form of a flat doughnut (7) having outer and inner diameter of 12.0 and 4.7 mm, respectively. The desired number of barrier plates are assembled into one unit by keeping the plates plane parallel with a regular distance. This was done by sewing the plates with two pieces of fine wire (12). At least 89 and at most 260 plates were fitted over the whole length of the barrier assembly (52 cm). Regardless of the number of barrier plates fitted, twelve cruciform barriers (8), slightly larger than the doughnut barriers, were placed every 4.7 cm in place of the doughnut barrier. This was necessary for assuring the installation of the barrier assembly in the proper position inside the column space. When copper was used as barrier material, the contact point between the plate and sewing wire was cemented by epoxy resin, while no cement was necessary for Teflon plates. The barrier assembly built up outside the column was carefully transferred into the column space and fixed to the terminal plates (3) located at the top and bottom of the gas chamber.

Operation of the Column. Operation of the column was more or less the same as described previously.[2] A d.c. power source with stabilized voltage was used for heating the nichrome wire. The column was filled with test gas before heating current was supplied. After steady temperature difference had been set up the column was evacuated and filled with fresh gas in order to start the measurement. The gas pressure was kept at 785±4 Torr throughout the experiment. The test gas was an equimolar mixture of argon and nitrogen.

Results and Discussion

Theoretical Value of q_e for the Open Column. So far we have no reliable means to predict a theoretical value of q_e attainable in a column having a horizontal barrier

TABLE 1. CALCULATED VALUES OF TRANSPORT COEFFICIENTS AND EQUILIBRIUM SEPARATION FACTOR

ΔT (°C)	$h\times10^3$	$k_c\times10^2$	k_d	$H\times10^4$ (g/s)	$K_c\times10^2$ (g cm/s)	$K_d\times10^4$ (g cm/s)	$A^{a)}\times10^3$ (cm^{-1})	$q_e^{b)}$
221	4.10	1.74	0.52	1.60	1.97	6.17	3.93	1.53
310.4	6.30	2.15	0.53	2.46	2.44	6.28	4.91	1.70
419	8.53	2.48	0.54	3.33	2.81	6.47	5.80	1.87
527.7	10.5	2.51	0.56	4.11	2.84	6.66	7.05	2.14

a) $A=\dfrac{H}{2(K_c+K_d)}$. b) $q_e=\exp(2AL)$.

assembly. A theoretical value can be obtained only for the open column.

The basic equation derived by Jones and Furry[6] was used for calculation. The following values were used for basic parameters;

$$\alpha = 0.127-13.6/T,^{7)}\ (D_{12})_1 = 0.20\ \text{cm}^2\ \text{s}^{-1},$$

$$(\bar{\rho})_1 = 1.53\times10^{-3}\ \text{g cm}^{-3},\ (\bar{\eta})_1 = 1.97\times10^{-4}\ \text{pois}.$$

Subscript 1 outside the parentheses refers to the temperature of the cold wall taken to be 289 K. The numerical tables by Saxena and Raman[8] were used for the corrections as regards the cylindricity of the column. This is equivalent to assuming both argon and nitrogen molecules to be rigid spheres. The calculated values of equilibrium separation factor, q_e, as well as the transport coefficients are given in Table 1.

Experimental Results. Measurements were carried out (1) with an open column in which no barrier was installed, (2) with a column having Teflon barriers, and (3) with a column having copper barriers. In the measurements with barrier-columns the effect of barrier distance, *i.e.* vertical distance between two adjacent plates, was also studied. Altogether, seven series of experiments were made, the results of which are summarized in Table 2.

The progress of separation with time for the initial 15 min of each measurement is shown in Fig. 2. The curves are numbered according to the series of experiments and correspond to those in Table 2.

Variations of equilibrium separation, q_e, with temperature difference, ΔT, are shown in Fig. 3. We see that in each series of experiments with the barrier-column, q_e increases linearly with increasing ΔT.

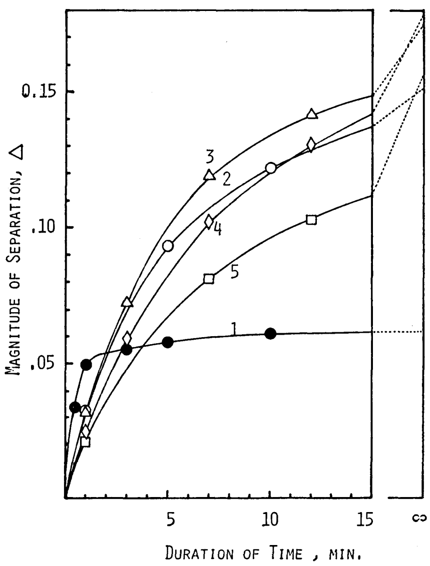

Fig. 2. Progress of separation with time.
The magnitude of separation is the difference in mole fractions of component 1 between top and bottom of the column.

The dotted line connects theoretical values calculated for the open column. The experimental values for q_e for the open column are much lower than those of theoretical prediction. In contrast, the q_e values

TABLE 2. EXPERIMENTAL RESULTS

Series	Barrier	Barrier spacing, d(mm)	ΔT(°C)	$q_e^{a)}$	t_r (min)	f
1	None	—	200±5	1.205	2.1	1
			400±10	1.279	0.6	1
2	Teflon	5.3	200±5	1.463	7.6	2.04
			400±10	1.845	5.4	2.49
3	Teflon	3.5	200±5	1.534	10.6	2.29
			400±10	2.027	6.2	2.87
4	Teflon	2.1	200±5	1.527	13.6	2.27
			400±10	2.059	8.8	2.93
5	Teflon	1.0	200±5	1.460	16.7	2.03
			400±10	1.885	11.1	2.58
6	Copper	5.3	200±5	1.521	12.9	2.25
			400±10	1.963	7.5	2.74
7	Copper	2.8	200±5	1.61	18.3	2.55
			400±10	2.15	10.4	3.11

a) Average of at least three measurements.

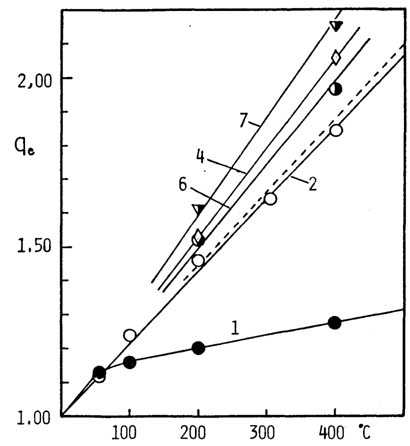

Fig. 3. Equilibrium separation factor, q_e, against the temperature difference applied. Numerical figures attached are serial number of experiments.

observed with barrier-columns are always larger than those with the open column even in comparison with theoretical prediction. The quality factor, f,[2] defined by

$$f = (\ln q_e)_{\text{barrier}}/(\ln q_e)_{\text{open}}$$

takes values greater than two (Table 2). In this equation, q_e is defined by

$$\ln q_e = 2AL,$$

where A is the characteristic constant of each individual column and L the column length. The two fold increase in the quality factor is equivalent to using an open column of two times taller in height. The quality factor seems to be affected largely by spacer distance. This is shown in Fig. 4, where the solid curve stands for Teflon barriers and the dotted curve for copper barriers. An optimum appears at about 2.8 mm.

Effect of Barrier Material. The barrier material affects the equilibrium separation, copper being superior

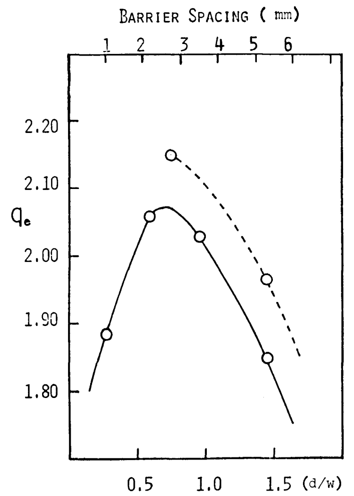

Fig. 4. Change in q_e against the barrier spacing, d, as well as the ratio of d to the barrier width, w. Full line; Teflon plates, Dotted line; copper plates.

to Teflon. Prior to the experiment we surmised that q_e would be reduced by the local disturbance of uniform temperature gradient caused by the metallic heat conductor. The result was contrary to expectation but it is understandable if we take the temperature gradient in a field of forced convection into consideration. The temperature is thought to fall steeply at the surface region of the central heater, never changing linearly. It seems that, in the column studied, most part of the temperature gradient is set up in a restricted region near the heater surface and the space in which barrier plates are located is almost isothermal. If this is the case, the barrier material would have no effect. Actually, however, copper enhances the equilibrium separation. The enhancing effect of metallic barriers can be attributed to the enhancement of net heat conduction through the column. The primary driving force of thermal diffusion is undoubtedly the heat flux. The presence of a heat conductor inside the gas chamber, even in the isothermal region, would increase the net heat flux giving rise to an enhanced separation. This is in accordance with the fact that when the barrier assembly of copper plates was used approximately 10% larger power dissipation was necessary for maintaining a given temperature difference as compared to the case in which Teflon plates were used. Teflon plate is also a better heat conductor when compared with separating gas. As a result, power dissipation always increases when barriers are used.

Effect of Barrier on the Rate of Separation. Use of horizontal barriers resulted in an appreciable increase in q_e (Table 2). This is reflected in the retardation of the rate of separation. If we assume that the separation proceeds by the exponential function[10]

$$\Delta = \Delta_e(1-e^{-t/t_r}),$$

we can estimate the relaxation time, t_r, experimentally. The values of t_r thus determined are given in Table 2. We see that the use of barriers increases the transition time a great deal; the denser the barrier spacing, the greater the transition time (Fig. 5). This is in line with what we found in the case of vartical barrier, that the retardation of convectional velocity is the primary factor for the enhancement of equilibrium separation.[1] In the present case, however, it should be emphasized that too dense installation of barrier plates reduces q_e and an optimum appears in the relation between q_e and barrier spacing (Fig. 4). This suggests that the enhancement in equilibrium separation can not be entirely attributed to the retardation of convectional velocity (Fig. 2). The rate of separation (tangential slope of each curve at $t=0$) varies with curve. However, the slopes have no simple correlation with the relaxation time. It seems that the greater the equilibrium separation the faster the intial rate of separation.

Visualization of Stream Line. Let us consider the question of why the use of the barrier assembly increases the magnitude of q_e. Each barrier plate serves to disturb the stream line of flowing gas making the flow turbulent. A turbulent flow would act to mix rather than to separate the mixed components. Actually, this was not the case and the use of barrier was unexpectedly effective for

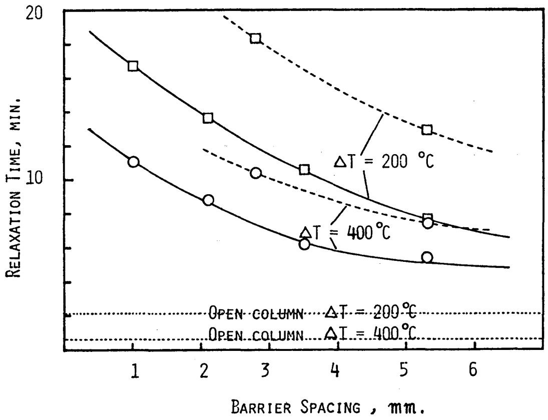

Fig. 5. Relaxation time against the barrier spacing. Full line, Teflon plates, Broken line, copper plates, Dotted straight line, open column.

attaining a larger separation. We thus attempted to visualize the flow pattern inside the barrier-column. Attempts in a gas sample using several kinds of aerosols were unsuccessful. We then carried out the experiment with use of a solid suspension in a liquid. Fine powder of polyethylene suspended in a water–isopropyl alcohol mixture was employed. A model column of a glass tube, in which a barrier assembly and a central heater wire were installed, was filled with the indicator solution. The whole apparatus was immersed vertically in a thermostatted water bath (25 °C). The temperature of the heater wire was not measured but the heating current was so controlled as to make the flow pattern visible.

A typical example of stream line is shown in the photograph attached. It is evident that the flow of fluid occurs in a regular manner. The path of particle movement as observed in a vertical cross section is a circle. It is unlikely that indicator particles move only in a fixed vertical plane. The path of each particle must be continuous in the transverse direction. It is highly

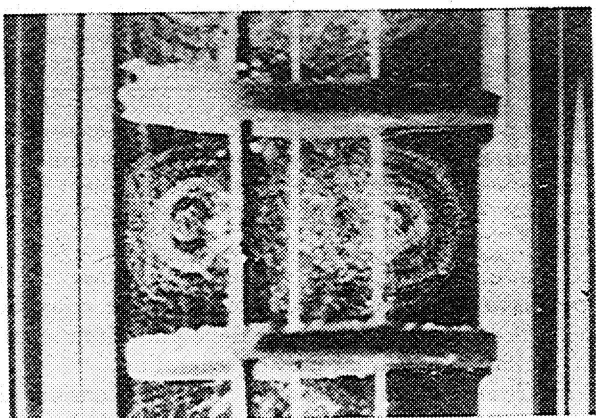

Fig. 6. Photograph of stream line observed in a glass column. Moving particles are polyethylene powder suspended in a water–isopropyl alcohol mixture. Inner diameter of glass tude; 18 mm, Outer and Inner diameters of barrier; 14 and 5 mm, Barrier spacing; 8 mm.

probable that the path of stream line forms a doughnut-shaped circular helical coil surrounding the central heater wire. Although the picture was taken in a dummy column with a liquid sample which might differ from gaseous sample in many respects, a rough concept on the nature of flow pattern can be attained.

The flight length of a particle in upward convection stream should be L, the length of column, in the open column. The flight length should be elongated in the barrier column by involvement in the helical transverse motion. Involvement in the helical transverse motion might be rationalized from the view that the value of the quality factor, f, can be considered as an effective elongation of column length.

Optimum Spacing of Barriers. We see in Fig. 4 that an optimum barrier distance exists for the largest separation. The scale of abscissa is also expressed tentatively in terms of the ratio of spacer distance, d, against the width of barrier plate, w. The optimum lies at about 0.75 of this scale. The optimum distance should be a function of the relative dimension of inner and outer diameters of barrier plate relative to those of column and heater wire.

Conclusion

1. Installation of a horizontal barrier assembly, consisting of many plates with the form of a flat doughnut, remarkably increases the equilibrium separation as compared with the open column. The spacing between two adjacent barrier plates has an appreciable effect on the eqilibrium separation. There is an optimum spacing for the largest separation. Almost three times greater separation can be obtained by suitable choice of horizontal barriers.

2. As the barrier material, copper was found to be superior to Teflon. The presence of the barrier assembly, which is a better heat conductor as compared with gaseous material, increases the effective heat conductivity of the whole apparatus and the increase in heat flux enhances the separation. The enhanced separation would be partly due to this effect. The power dissipation per unit time increases by using barriers.

3. Installation of a barrier assembly reduces the rate of separation and elongates the relaxation time for separation. The relaxation time is a simple function of the barrier spacing, increasing monotonously with decreasing barrier spacing (increasing number of barrier plates). However, the actual rate of separation determined from the tangential slope of separation-time curve has no simple correlation with the relaxation time. The rate so determined varies in line with the statement "the greater the separation, the greater the rate."

4. Attempts were made to visualize the flow pattern in the barrier column with use of a dummy column. The movement of polyethylene powder suspended in a water–isopropyl alcohol mixture in the field of forced convection was recorded in order to show that the stream line forms a helical coil at each space partitioned by barrier plates. It is suggested that the use of horizontal barriers is equivalent to lengthening the open column. The situation is similar to that observed in

a tilted column.[4]

5. In contrast to the vertical barrier, the enhancement of separation in a horizontal barrier system can not be attributed solely to the retardation of convectional velocity. The regular helical motion of molecules would have an important effect.

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