

Design and Performance of a Simulated Countercurrent Moving-Bed Separator

Barry B. Fish, Robert W. Carr, and Rutherford Aris

Dept. of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455

The design, construction and isothermal (298 K) operation of a continuous flow simulated countercurrent moving-bed chromatographic separator for the gas-solid separation of binary mixtures is reported. It consists of only four packed columns, with countercurrency simulated by advancing the feedpoint from column to column in the direction of gas flow. The separated components have periodic concentration profiles, with a duty cycle that depends on operating parameters, and concentrations greater than in the feed stream. Test mixtures of propylene and dimethyl ether, which have retention times differing by a factor of 2 on the 60/80 mesh Chromosorb 101 packing, were successfully separated. Optimum feed advancement intervals and maximum product purity were experimentally determined as a function of feed rate. At low feed rates, when the adsorption isotherm is nearly linear, dimethyl ether was nearly completely separated from propylene, but propylene was found to contain varying amounts of dimethyl ether due to the effects of a nonlinear isotherm and heterogeneous adsorption.

Introduction

Chromatography provides a separative technique that requires less energy than such conventional methods as distillation and low temperatures beneficial to substances prone to degrade thermally. The throughput of the traditional batch chromatographic approach can be substantially improved by employing continuous chromatography, for which the primary requirement is relative motion between the feed stream and the granular chromatographic solids.

Although countercurrent systems provide efficient utilization of the solid phase for continuous chromatographic separations (Ruthven, 1984), the difficulties associated with implementation of a moving bed have led to the consideration of simulated countercurrency. Separation effected by countercurrent contact of solid and gas streams is normally accomplished by flowing a granular solid and a counterpropagating carrier fluid past a stationary inlet in a vertical tube. The countercurrent motion can be simulated by employing a fixed bed with several axially aligned inlets and outlets, and moving the feed and product positions sequentially along them in the direction of carrier fluid flow. That is, moving the feed past the bed rather than the bed past the feed. This is essentially the arrangement of the commercial Sorbex processes (Broughton, 1984/85). Alternatively, a configuration in which the fixed

bed is replaced by packed columns in series can be employed. In this article, we report the design and performance of a laboratory-scale columns-in-series configuration of a simulated countercurrent moving-bed chromatographic separator (SCMCS).

In the multiple-column SCMCS, a port which can serve as inlet or outlet is located between each column. Feed inlet advancement in the direction of carrier fluid flow simulates countercurrent motion, although not continuously but in discrete steps at timed intervals. Either binary separations or separation of several components into two groups are possible, since the advancement rate and the carrier flow rate can be adjusted so that the more strongly adsorbed component(s) travels more slowly than the feed position, while the less strongly adsorbed component(s) moves ahead of it.

Barker and Deeble (1973, 1975) pioneered the multiple-column configuration, successfully separating mixtures of 1,1,1-trichloroethane and 1,1,2-trichloro-1,2,2-trifluoroethane in a 12-column apparatus by means of a DC200/50 on Chromosorb P stationary phase with air as the carrier gas. Product purities were in excess of 99.9%. A 12-column configuration was also employed by Szepesy et al. (1975) for recovery of benzene from gasoline and separation of C_{16} - C_{22} fatty esters. A similar system

was used to separate glutathione and glutamic acid (Maki et al., 1987). Hashimoto et al. (1983a,b) employed the same principle in a 16-column configuration with a novel rotary valving system to separate glucose-fructose mixtures. Ching and Ruthven (1985a,b,c, 1986a,b, 1987, 1988) have investigated many aspects of the glucose-fructose separation in a 12-column apparatus, as have Barker and coworkers (Barker and Ganetsos, 1985; Barker and Abusabab, 1985; Barker and Thawait, 1986).

A related process is the moving feed point method (Wankat, 1977), in which the feed is periodically advanced along a chromatographic column or series of columns at a rate intermediate between that at which two components travel. This approach permits greater throughput in preparative chromatography, but presumably less productivity than the SCMCS since product streams are not removed at the intermediate stages, but only after the last feedpoint.

In most of the work to date, the 12-column configuration has been employed. With 12 columns the behavior of "true" countercurrent operation is approached, with model predictions in good accord with those from countercurrent moving beds (Ching and Ruthven, 1985a). It is shown here that, in principle, separations can be obtained in a SCMCS with as few as three columns, and that, as a practical matter, four columns function well. The eluent concentrations, however, are periodic, not stationary as with the countercurrent moving-bed separator. Fewer columns provide improved utilization of adsorbent and require less valving for the feed and product line switching operations than larger numbers of columns. Although the experimental system here is a gas-solid separation, the results, in principle, should apply to liquid-solid separations as well.

Design Considerations

The design of a SCMCS is considered here for separation of two-component mixtures which are sufficiently diluted by carrier fluid that the carrier flow rate is very nearly uniform throughout the separator. It incorporates dispersionless flow, infinite mass-transfer rates, linear adsorption isotherms, and identical packed columns.

It is instructive to investigate first how the axial concentration profile develops for a single adsorbate as it is shifted from column to column at time intervals t_s . Figures 1 and 2 show how the concentrations change for two adsorbates, A and B, with different breakthrough times. The adsorbate, mixed with carrier, enters the first column (arrow position) at $t=0$, and at t_s is advanced to the adjacent column, the carrier position remaining unchanged. Transfer between columns is instantaneous. The concentration profile at $2t_s$, just at advancement to the third column, is shown in row 2. Succeeding rows show the profile at multiples of the switch time. After the last column in line has been the feed column for t_s , the feed position reverts to the beginning and a new cycle through the series of columns begins. The concentration profile that develops in the separator depends on whether t_s is shorter or longer than the breakthrough time of the adsorbate. Figure 1 shows the development of the concentration profile for a single component, initiated by flowing it (species B) into the first column. At the time of the first feed advancement, t_s , B has occupied 75% of the length of the first column, and is at concentration C_B . Upon advancing the feed, the concentration entering the second col-

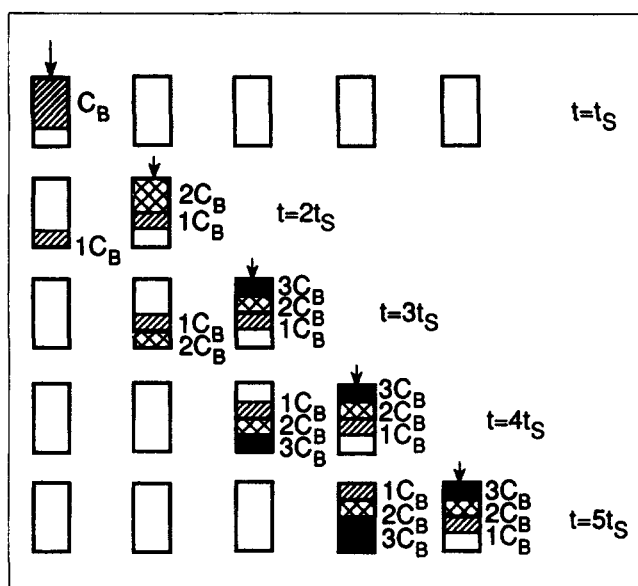


Figure 1. Development of an ideal one-component concentration wave in a multiple-column SCMCS when the switch time, t_s , is $3/4$ of the breakthrough time of B, t_B .

Feed (arrow) enters the first column at $t=0$ and the concentration is shown at t_s , just before switching to the second column. The second row shows the expected concentration profile at $t=2t_s$, just as feed is shifted to the third column. Carrier gas flow is identical in each column.

umn is C_B , and remains so until the front breaks through the first column, when the concentration entering the second becomes $2C_B$. Figure 1 shows the concentrations in both at $2t_s$, and the development of the ideal concentration profile up to $5t_s$. When the advancement rate, defined as the length of a single section of column divided by t_s , is larger than the speed of the adsorbate front, a profile develops in which the concentration rises to a maximum near the feed point and then decreases behind it. It is noteworthy that, at the maximum, the concentration is greater than in the feed.

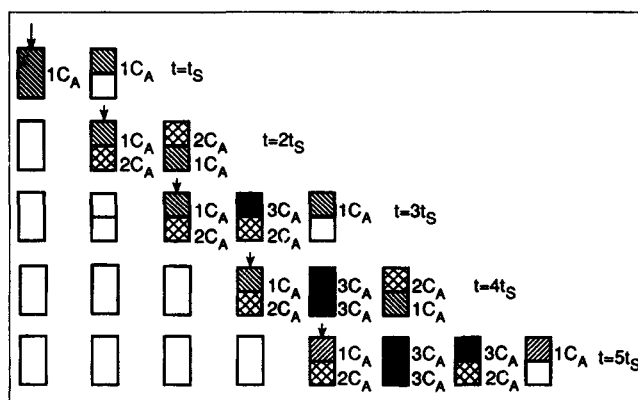


Figure 2. Development of an ideal one-component concentration wave in the SCMCS when $t_s = 1.5 t_A$.

It shows how the front moves in front of the feedpoint (arrow) and concentrates. Description of switch times are similar to Figure 1.

If the advancement rate is slower than the speed of the adsorbate front, a different profile develops. Figure 2 shows the situation when the adsorbate travels 1.5 column lengths during t_s . A maximum also develops, concentrating the adsorbate, but the maximum is downstream from the feed point.

The concentration profiles that develop during the separation of a binary mixture, in which one component (B) has a breakthrough time $t_B = 4/3 t_s$ and the other (A) has $t_A = 2/3 t_s$, will just be a superposition of Figures 1 and 2 when the isotherms are linear and hence independent of each other. This is a particularly easy separation because the breakthrough time ratio $t_B/t_A = 2$, but it serves to illustrate how the separation develops.

It is clear that A (in the carrier, but separated from B) can always be removed from the feed column if $t_B > t_s > t_A$ and if the feed is advanced to the next column before the B front breaks through. It is also apparent that component A is carried ahead of the feed position, while component B falls behind it. From Figures 1 and 2 it is apparent that a three-column configuration is capable of separating a mixture in which the components have breakthrough times differing by a factor of two. Starting the feed at the first column, it is seen that two advancements later, only B is left in column 1, and during the time that 3 is the feed column, B can be removed from 1. At $3t_s$ column 1 is clean and can again be used as the feed column, leading to the conclusion that the minimum number of columns necessary for this separation is three.

Figure 3 shows the temporal development of the A and B product streams, where A is taken from the feed column and B is taken from the second column behind it. The output approaches a periodic steady state, with a duty cycle for the concentrations of A and B resulting from the relationships among t_s , t_A , and t_B . The duty cycle arises from the time elapsed waiting for the fronts to elute.

Next, we consider the selection of t_s . If t_s is too close to t_B , some B will probably contaminate A in a separator with realistically broadened fronts. Similarly, if t_s is close to t_A , then the B product will probably be contaminated with A . A conservative approach would place t_s midway between t_A and t_B :

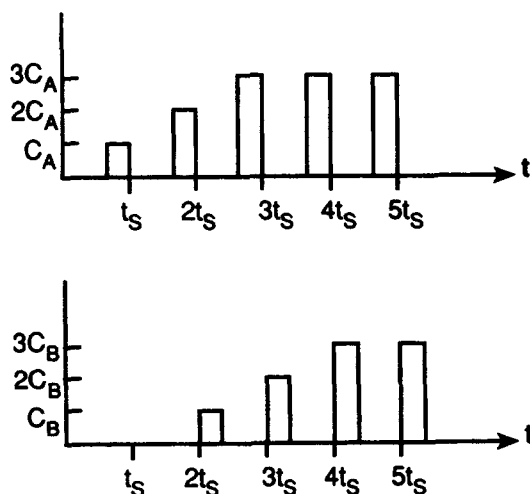


Figure 3. Temporal evolution of ideal concentration waveforms during startup of a SCMCS.

It shows the duty cycle and the increase in concentration at the periodic steady state.

$$t_s = (t_A + t_B)/2 \quad (1)$$

The concentration of the product streams can be calculated from a mole balance:

$$C_i^{\text{in}} F t_s = C_i^{\text{out}} F |t_i - t_s| \quad (2)$$

where F is the total flow rate and C_i^{out} is the average output concentration of species i . Applying the criterion of Eq. 1, the increase of output concentration for component B is given by:

$$C_B^{\text{out}}/C_B^{\text{in}} = (t_A + t_B)/(t_B - t_A) \quad (3)$$

A similar relationship can be obtained for component A . This quantifies the concentration of the feed in Figures 1 and 2. In addition to separation, there is a concentrating effect that may facilitate the subsequent separation of carrier from each component. The time that the output is nonzero, t_{out} , as a fraction of t_s is given by:

$$t_{\text{out}}/t_s = (t_B - t_A)/(t_A + t_B) \quad (4)$$

If a larger duty cycle is desired, it may be achieved by adding a fourth column to the SCMCS, placed between the feed column and the B product take-off column, and forwarding some fraction of the feed column effluent to the new column. See Figure 4. Since, upon advancing the feed at t_s , the new column becomes the feed column, and A already has made some progress in it, less time is consumed waiting for A to break through than is required for the three-column configuration, where it is t_A . Now the steady-state mole balance on A becomes:

$$F C_A^{\text{in}} t_s = F_A C_A^{\text{out}} (t_s - t_d) \quad (5)$$

where F_A is the total flow rate of the A product stream, C_A^{out} is the average concentration of A in the output, and t_d (the dead time) is the interval between the shift of feed to a new column and the breakthrough of A from that column. If $F_A = F$, then $t_d = t_A$, as before. If continuous output is desired, t_d must be reduced to zero, which requires $t_s = t_A$. In this case, $C_A^{\text{out}} F_A = C_A^{\text{in}} F$, and the extent to which A becomes concentrated in the output depends on the split flow ratio (or F_A) selected. Using Eq. 2 to eliminate C_A^{out} reveals that the minimum output flow of A is:

$$F_A^{\text{min}} = \frac{t_B - t_A}{t_B + t_A} \quad (6)$$

A possible simplification providing more efficient use of carrier gas, as well as simplifying the valving, would be to route the carrier from column III to column IV. This, however, changes the problem since F_A is no longer an independent variable. The flow that is routed to III is now the flow that is available to purge B from IV and must be sufficient for that purpose. Calling this F_B and specifying that t_d is equal to the amount of time the concentration of B is zero after being eluted from the purge column, the mass balance is:

$$\begin{aligned} C_A^{\text{out}} (t_s - t_d) F_A &= C_A^{\text{in}} t_s F_T \\ C_B^{\text{out}} (t_s - t_d) F_B &= C_B^{\text{in}} t_s F_T \end{aligned} \quad (7)$$

where $F_T = F_A + F_B$. The ratio $C^{\text{out}}/C^{\text{in}}$ is still given by Eq. 3 since the concentrating effect is still due to the separation occurring in I and II. Equations 7 and 3 combine to yield:

$$F_B/F_A = (t_B - t_s)/(t_s - t_A) \quad (8)$$

and if $t_s = (t_A + t_B)/2$, then

$$F_A = F_B \quad (9)$$

The minimum flow rate to purge B from column IV, F_B^{min} , is:

$$F_B^{\text{min}} C_B^{\text{out}} = F C_B^{\text{in}} \quad (10)$$

and if $t_s = (t_A + t_B)/2$ it can be shown that the maximum flow of A , F_A^{max} , which can be taken from port A and still permit B to be purged from column IV, is given by:

$$F_A^{\text{max}} = F \left[\frac{2}{(t_B/t_A) + 1} \right] \quad (11)$$

Equations 11 and 5 indicate that $F_A^{\text{max}} = F_A^{\text{min}}$ when $t_B/t_A = 3$.

For separations with $t_B/t_A > 3$, a switching speed of $t_s = (t_A + t_B)$ cannot be used, because if half the carrier is removed at port A , the flow to column III will sweep component A into column IV, where it will eventually leave with the B product stream. Furthermore, the carrier flow in column IV will not be sufficient to purge it of B , and the A product stream will become contaminated with B . When $t_B/t_A > 3$, t_s must be set closer to t_B , and more than half the carrier removed at port A .

Although the minimum number of columns is three, a four-column configuration may be better. If heterogeneity of surface sites and isotherm nonlinearity causes some retention of the more strongly adsorbed component in the purge column when it becomes the feed column, the more weakly adsorbed component will displace the retained adsorbate and become contaminated.

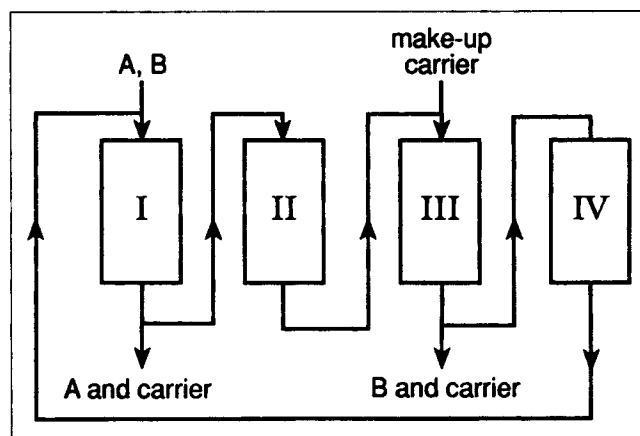


Figure 4. Four-column SCMCS.

lytical gas chromatography, with automatic computer-controlled sampling and computer data acquisition (Fish et al., 1988).

Propylene (P) and dimethyl ether (DME) were selected as a convenient and easily separable pair of model substances with which to test the performance of the SCMCS against the above design considerations. Their relative retention time of 2 ($t_r = t_{DME}/t_P$) on Chromosorb 101 at 298 K corresponds to the above design example, providing an experimental system to test these conclusions. Also, they have absolute retention times that require modest column lengths at room temperature. Adsorption isotherms for both species were determined by frontal analysis (Paryjczak, 1986). The isotherms were non-linear and of the convex type. Care was taken during experimentation to work at low concentration of adsorbates, in the nearly linear region of the isotherms to conform as closely as possible to the linear isotherms of the model. Routine analytical gas chromatography was done on a 1/8-in. (3.2-mm) OD by a 10 ft (3-mm) column packed with Chromosorb 101 and thermostatted at 40°C. Calibration curves that were linear in peak area were obtained. The peak areas were plotted against peak height and fitted to a cubic equation so that peak height measurement could be used for quantitation. This greatly facilitated on-line data acquisition and permitted GC analyses

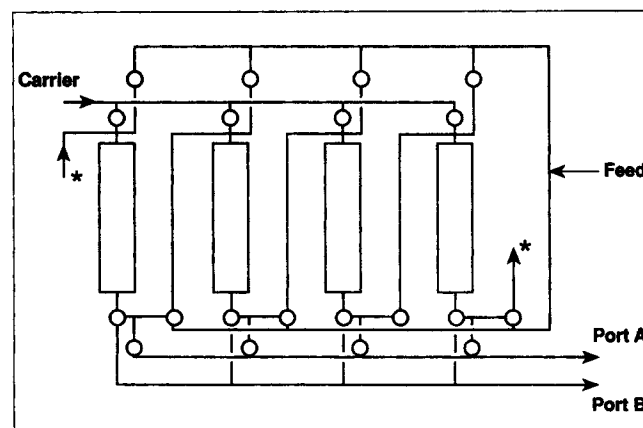


Figure 5. Valving diagram of a four-column SCMCS.

The asterisked lines are connected.

Experimental Studies

A four-column laboratory-scale SCMCS was constructed in the configuration of Figure 4. Each of the sections consists of a 12 in. by 1/2 in. (305 mm by 13 mm) OD tube packed with 60/80 mesh Chromosorb 101. The sections are connected by 1/8-in. (3.2-mm) tubing, and flows are directed with two- and three-way solenoid valves. The valving configuration is shown in Figure 5. At each column exit it is possible to either (1) route the flow to the next column, (2) remove the flow from the reactor, (3) split the flow with a portion directed to the next column and a portion removed, or (4) combine it with the feed stream. The valves are controlled with a microcomputer via solid-state relays. Since groups of valves must be opened or closed simultaneously, all 20 valves are operated by only four relays, by preprogrammed timing and sequencing. The binary feed is continuously injected by a syringe pump and mixed with carrier gas, which is then routed to the proper feed column for a preselected period of time before being advanced to the next column. Analysis of the separator effluent was by ana-

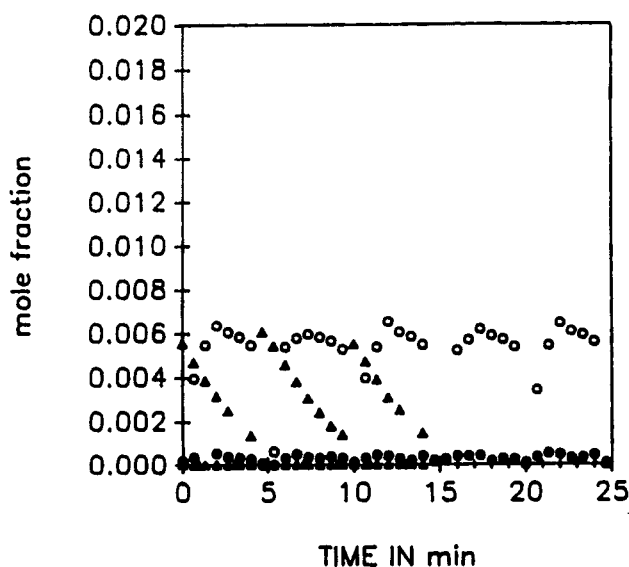


Figure 6. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 5 min.

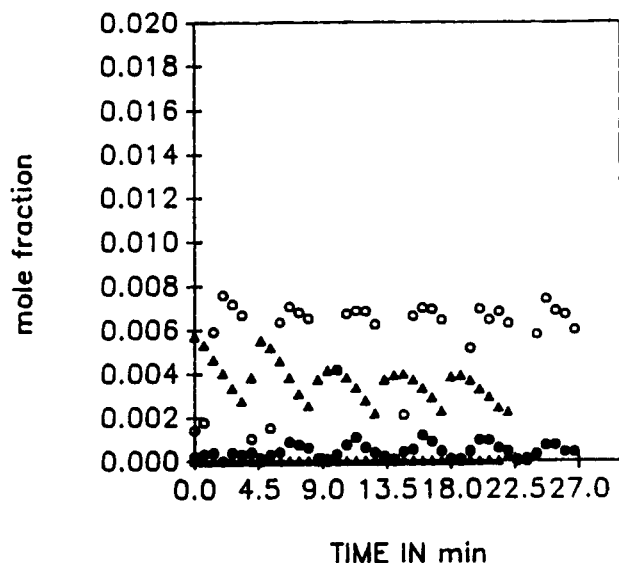


Figure 7. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 4.5 min.

of the separator product streams at 40-s intervals so that periodic concentration waveforms could be discerned.

Results

The total carrier gas flow rate was set at 360 mL/min, for which the pressure drop through each column was only 100 torr. The feed composition in all experiments was about 50 mol % each of *P* and *DME*. According to Eq. 6, the minimum flow that can be removed from part *A* is $0.33 F_T$ when $t_s = 1/2(t_A + t_B)$, and the maximum, according to Eq. 11, is $0.67 F_T$. In these experiments, the split flow ratio at port *A* was conservatively set at 0.5, since F_A^{\min} and F_A^{\max} were calculated by considering linear isotherms, and the experimentally determined isotherms of *P* and *DME* showed them to be non-linear.

Several separations are presented below. The product take-off from the feed column is designated port *A*, and the other is port *B*. With GC analyses, every 40 s and switching periods of 5 min selected according to the criterion of Eq. 1, the concentration waveforms eluting from these ports can be discerned. For example, Figure 6 shows the time dependence of *P* and *DME*. These waveforms are achieved after an initial transient period requiring several cycles and may be described as a periodic steady state. A switching cycle (through all four columns) is represented by the period from 0 to 20 min. During the 20- to 25-min interval, the feedpoint is located on the same column as during the 0- to 5-min period. Very little *P* is present immediately after a switch (0, 5, 10, ... min), but its concentration then rises steeply, revealing the presence of the front as it breaks through the feed column. The concentration of *DME* at port *B*, shown by filled triangles, falls steadily as *DME* elutes by the carrier gas flow. At the end of the 5-min period, it is still nonzero due to tailing (discussed below). Thus, some *DME* remains behind at the switch of feed position and appears at port *A*, contaminating *P*. The average composition

of the condensables at port *A* during this experiment is 93% *P* and 7% *DME*, while that of *DME* at port *B* is 100% *DME*.

Figure 7 shows the effect of a decrease of switching period to 4.5 min, everything else remaining the same. There is now a longer "dead time" before *P* appears at port *A*, and the *DME* profile at port *B* now shows evidence of a maximum. The *P* purity has deteriorated. Figure 8 shows the effect of increasing the switching period to 6 min. There is no dead time for the appearance of *P* at port *A*, but *DME* now appears at fairly high concentration in the last three samples of the 6-min period. The average *P* purity is only 64%. At $t_s = 4.5$ min and $t_s = 6$ min, the *DME* fraction contains only traces of *P*.

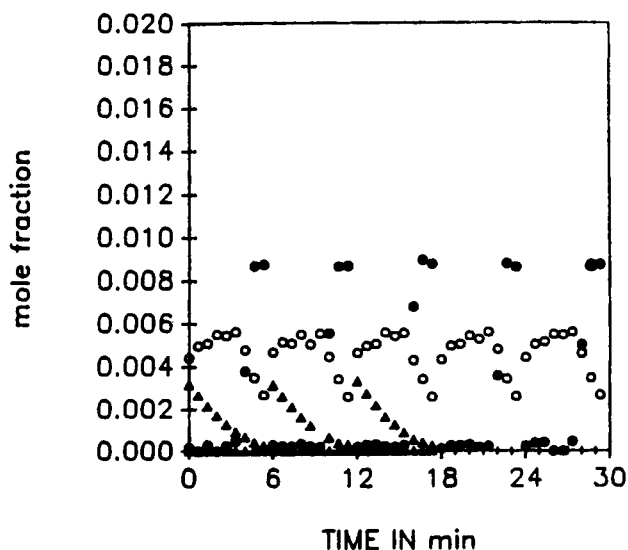


Figure 8. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 6.0 min.

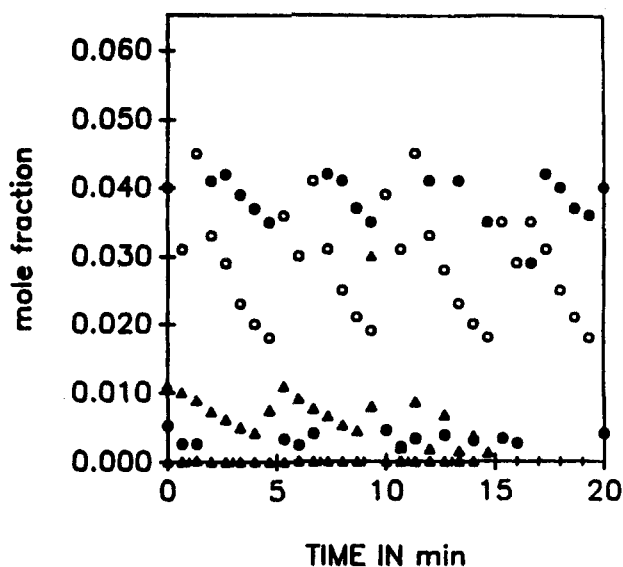


Figure 9. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 5 min.

Figures 9, 10 and 11 show the effects of increased feed concentration on SCMCS performance. In Figure 9, the interval between advancements is 5 min, which gave the best performance at lower concentrations. Here, however, *DME* breaks through at about 2 min, and the effluent at port *A* contains comparable amounts of *P* and *DME*. Furthermore, the *DME* at port *B* is highly contaminated with *P*. The poor performance results from the nonlinearity of the isotherm, which gives faster breakthrough times at higher concentrations.

Reducing the switching period improves the performance. Figure 10 shows that a 3-min period is still not satisfactory, since *DME* still breaks through and appears at port *A* before

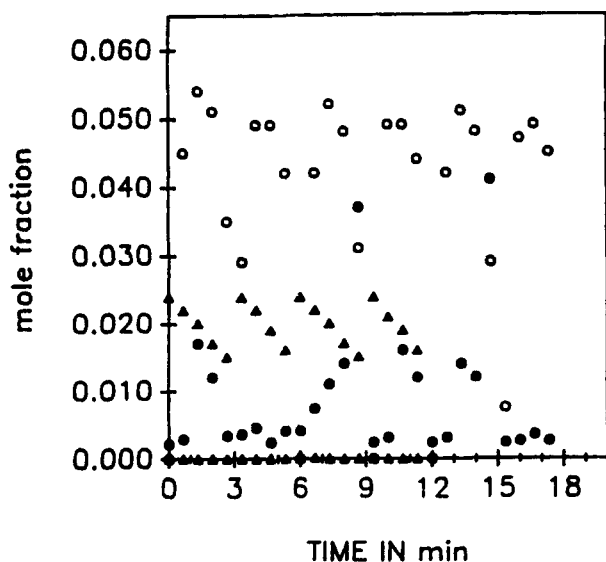


Figure 10. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 3 min.

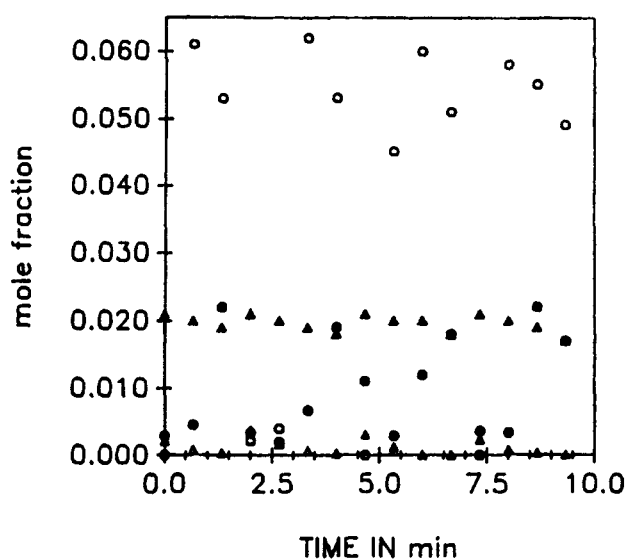


Figure 11. Concentration profiles of *P* (open symbol) and *DME* (filled symbol) at ports *A* (circle) and *B* (triangle) for a switching time of 2.5 min.

the feed can be switched. The *P* purity at port *A* is 80%, while the *DME* at *B* contains only trace amounts of *P*. Reducing the switching interval to 2.5 min does not improve the *P* purity at port *A*, and the *DME* at *B* is now contaminated with about 5% *P*, since the feed switching occurs faster than *P* can be swept along the SCMCS.

Discussion

The experimental results are in good general accord with the predictions from the design criteria for selecting t_s and the split flow ratio at port *A*. Good separations were obtained with $t_s = 1/2(t_A + t_B)$, and $F_A = 1/2 F$, the latter a compromise between the maximum and minimum flow rates prescribed by Eqs. 6 and 11. More quantitative comparisons were not attempted, since the design considerations are currently based on dispersionless flow, and it is clear from the product concentration waveforms that dispersion must be taken into account in any attempt to model them. If a mathematical model of the SCMCS were developed, both dispersion and a nonlinear isotherm would have to be included in the model equations.

The use of a carrier fluid causes dilution of the feed and imposes a further separation. This, however, may not be serious, since for gas-solid operations the carrier can be N_2 or air, which may be separated from a heavier vapor by condensing the latter at low temperature or adsorbing it on a suitable sorbent. Thus, a difficult separation of *A* from *B* can be replaced by two relatively easy separations of *A* and *B* from the carrier. It may be possible to operate with undiluted feed, in which case an undiluted *A* stream may be recovered, but a desorbent would be necessary to purge *B* from the separator.

Isotherm nonlinearity will adversely affect the quality of the separation attainable. With favorable isotherms, higher concentrations cause faster breakthrough with steeper fronts, which is an advantage in the feed column because the switching rate can be increased to just keep ahead of the *DME* front, but a

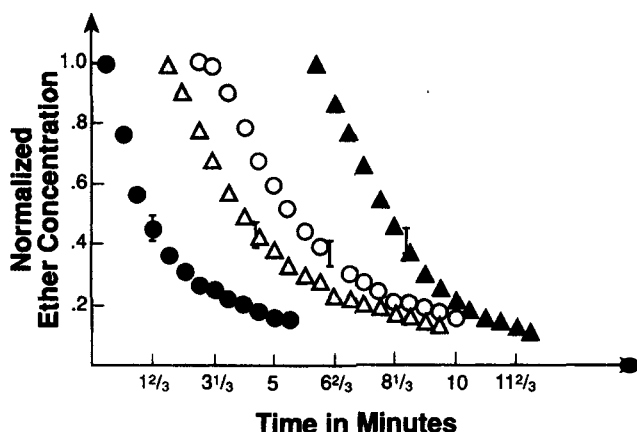


Figure 12. Elution of *DME* from a column packed with Chromosorb 101 saturated with *DME* with inlet mole fractions of: \blacktriangle 0.003; \circ 0.020; \triangle 0.042; \bullet 0.24.

Vertical bar is the measured breakthrough time at the corresponding inlet mole fraction.

problem in the purge column where the isotherm creates a longer tail which may leave some *DME* in the purge column when the feed (and product) streams are advanced. Heterogeneous adsorption also may cause *DME* to be retained in the purge column.

The extent of tailing was measured by saturating one of the separator columns with various concentrations of *DME* and then stopping its flow while maintaining the carrier flow and measuring the *DME* concentration in the effluent. Figure 12 shows the rate of decrease of *DME* at four different inlet mole fractions. Also shown on Figure 6 are the breakthrough times measured at the same inlet mole fraction, showing the effect of the nonlinear isotherm. For the lowest, $y_{DME}=0.003$, the column will be nearly purged of *DME* by about two breakthrough times, whereas for $y_{DME}=0.24$ the effluent concentration has fallen by only 85% after three breakthrough times. This problem can be combatted by isolating the purge column, employing a higher carrier flow rate, operating at lower pressure, heating, or adding more purge columns. If heterogeneous adsorption makes a contribution to tailing, close attention will have to be paid to adsorbent selection to minimize or eliminate this source.

A more limiting problem is the desorption of the less strongly adsorbed component (*A*) from the column behind the feed column. If all *A* is not sent to the next column, it will come out with the *B* product stream in the next cycle. This problem has been addressed for the continuous countercurrent separator (Fish et al., 1987), where it has been shown that when adsorption follows a Langmuir isotherm, there is an upper limit on the feed rate, above which the separator floods. To avoid contamination of product *A* with *B*, the solids velocity must be increased, but then product *B* becomes contaminated with *A*. (Similar behavior would be observed for any nonlinear, favorable isotherm.) A similar situation must occur for the SCMCS. The switching rate (a pseudo-solids velocity) can always be adjusted to stay ahead of the *B* front breakthrough time in the feed column; however, under flooding conditions some component *A* will be found in the *B* product stream, because it is carried more slowly than the switching rate. It is

necessary to ensure nonflooded operation, and in the experiments here care was taken to avoid flooding by maintaining low feed rates, which also access the approximately linear region of the isotherm for adherence to the model assumption.

The effect of the convex isotherm is to shorten elution times at the higher concentrations desirable for practical operations, and the data suggest that with the consequent short advancement times satisfactory separations may not be possible. It then becomes necessary to either decrease the carrier flow rate or design longer columns. It is also possible that elevated temperature operation might be beneficial, since isotherm nonlinearity usually becomes less pronounced with increasing temperature. Although elution times would be fast and have to be compensated by fast switching, tailing would be expected to be less severe and satisfactory separations might be obtained.

This investigation shows that the optimum number of columns in a SCMCR is four when the ratio of breakthrough times is two. This may also seem to be the optimum for more difficult separations, since, in principle, a stream of less strongly adsorbed component can be removed from the feed column as long as there is a difference in breakthrough times. The issue of the number of columns required to ensure good purity of the more strongly adsorbed component when the adsorptivities are not too dissimilar needs to be addressed before this conclusion can be affirmed.

Acknowledgment

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Literature Cited

- Barker, P. E., and R. E. Deeble, "Production Scale Organic Mixture Separation Using a New Sequential Chromatographic Machine," *Anal. Chem.*, **45**, 1121 (1973).
- Barker, P. E., and R. E. Deeble, "Sequential Chromatographic Equipment for the Separation of a Wide Range of Organic Mixtures," **8**, 67 (1975).
- Barker, P. E., and G. Ganetsos, "Production of High Purity Fructose from Barley Syrups Using Semi-continuous Chromatography," *J. Chem. Tech. Biotechnol.*, **35B**, 217 (1985).
- Barker, P. E., and E. K. E. Abusabah, "The Separation of Synthetic Mixtures of Glucose and Fructose and also Inverted Sucrose Feedstocks Using Countercurrent Chromatographic Techniques," *Chromatographia*, **20**, 9 (1985).
- Barker, P. E., and S. Thawait, "Separation of Fructose from Carbohydrate Mixtures by Batch and Semi-continuous Chromatographic Operation," *Chem. Eng. Res. Des.*, **64**, 302 (1986).
- Broughton, D. B., "Production-Scale Adsorptive Separations of Liquid Mixtures by Simulated Moving-Bed Technology," *Sep. Sci. Technol.*, **19**, 723 (1984-85).
- Ching, C. B., and D. M. Ruthven, "An Experimental Study of a Simulated Counter-Current Adsorption System: I. Isothermal Steady-State Operation," *Chem. Eng. Sci.*, **40**, 877 (1985a).
- Ching, C. B., and D. M. Ruthven, "An Experimental Study of a Simulated Counter-Current Adsorption System: II. Transient Response," *Chem. Eng. Sci.*, **40**, 887 (1985b).
- Ching, C. B., D. M. Ruthven, and K. Hidajat, "An Experimental Study of a Simulated Counter-Current Adsorption System: III. Sorbex Operation," *Chem. Eng. Sci.*, **40**, 1411 (1985c).
- Ching, C. B., and D. M. Ruthven, "An Experimental Study of a Simulated Counter-Current Adsorption System: IV. Non-Isothermal Operation," *Chem. Eng. Sci.*, **41**, 3063 (1986a).
- Ching, C. B., and D. M. Ruthven, "An Improved Adsorption Process for the Production of High-Fructose Syrup," *AIChE J.*, **32**, 1876 (1986b).

- Ching, C. B., and D. M. Ruthven, "Experimental Study of a Simulated Counter-Current Adsorption System: V. Comparison of Resin and Zeolite Adsorbents for Fructose-Glucose Separation at High Concentration," *Chem. Eng. Sci.*, **42**, 2547 (1987).
- Ching, C. B., and D. M. Ruthven, "Experimental Study of a Simulated Counter-Current Adsorption System: VI. Non-linear Systems," *Chem. Eng. Sci.*, **43**, 703 (1988).
- Fish, B. B., R. W. Carr, and R. Aris, "The Continuous Countercurrent Moving-bed Separator," *AIChE J.*, **35**, 737 (1987).
- Fish, B. B., R. W. Carr, and R. Aris, "Computer-aided Experimentation in Countercurrent Reaction Chromatography and Simulated Countercurrent Chromatography," *Chem. Eng. Sci.*, **43**, 1867 (1988).
- Hashimoto, K., S. Adachi, H. Noujima, and H. Maruyama, "Models for the Separation of Glucose/Fructose Mixtures Using a Simulated Moving Bed Adsorber," *J. Chem. Eng. Japan*, **16**, 400 (1983a).
- Hashimoto, K., S. Adachi, H. Noujima, and Y. Ueda, "A New Process Combining Adsorption and Enzyme Reaction for Producing Higher Fructose Syrup," *Biotech. and Bioeng.*, **XXV**, 2371 (1983b).
- Maki, H., H. Fukuda, and H. Morikawa, "The Separation of Glutathione and Glutamic Acid Using a Simulated Moving-Bed Adsorber System," *J. Fermentation Technol.*, **65**, 61 (1987).
- Paryjczak, T., *Gas Chromatography in Adsorption and Catalysis*, Chap. 4, Wiley, New York (1986).
- Ruthven, D. M., "Adsorption Separation Processes: II. Continuous Countercurrent Systems," *Principles of Adsorption and Adsorption Processes*, Chap. 12, Wiley, New York (1984).
- Szepesy, L., Z. Sebestyen, I. Feher, and Z. Nagy, "Continuous Liquid Chromatography," *J. Chrom.*, **108**, 285 (1975).
- Wankat, P., "Improved Efficiency in Preparative Chromatographic Columns Using a Moving Feed," *I & EC Fund.*, **16**, 468 (1977).

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