

A Rotating Annular Chromatograph for Continuous Separations

Preparative, multicomponent liquid chromatographic separations have been achieved by using a slowly rotating annular sorbent bed with fixed multiple feed points and fixed product withdrawal locations. The cation exchange separation of copper, nickel, and two cobalt complexes has been extensively used to study the effects of rotation rate, eluent rate, bed loading, and column size on separation performance. Experimental results have been compared with three theoretical models: plate theory, an analytical solution which incorporates multicomponent Langmuir isotherms, and an extension of the analytical solution to include dispersion effects.

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SCOPE

With its versatility, high resolution capabilities, and near-universal potential applicability, chromatography lacks only throughput capacity to make it an ultimate separation technique. Typical chromatographic separations are made in fixed columns and, as such, are inherently batch in nature. Various attempts have been made to increase the capacity of chromatographic devices either through cyclic operation of large-diameter columns or through continuous feeding of a mixture and continuous removal of the components in moving-bed systems. Several reviews of methods which attempt to achieve continuous chromatographic separations have appeared in the literature, e.g., Rendell (1975), Sussman and Rathore (1975), and Sussman (1976). These include counterflow, moving-bed systems,

crossflow moving-bed systems, and simulated moving-bed systems.

The objective of this work was to describe the performance results obtained by using a rotating annular sorbent bed, which has been successfully used to achieve continuous separations. The effects of feed rate, eluent velocity, bed loading, and rotation rate on the separation performance of such a device are discussed. Also discussed is the application of plate theory and an extension of the analytical solution describing a rotating annular sorbent bed. With such information, engineers should be able to make large-scale continuous chromatographic separations possible.

CONCLUSIONS AND SIGNIFICANCE

This study was made to obtain experimental data showing the effects of feed rate, feed concentration, rotation rate, and eluent velocity on the separation performance of a rotating annular sorbent bed. This bed receives a constant flow of a feed mixture and continuously separates the mixture into its respective solutes.

Both plate theory and an extension to the analytical solution of Rhee et al. (1970) to include dispersion effects were applied to the rotating bed. While plate theory is simpler to apply, the extended model is particularly useful when a nonlinear adsorption isotherm must be considered. Agreement between the extended model and experimental concentration profiles was excellent. When a conventional column was used to obtain the necessary adsorption and dispersion parameters of a desired

separation, both models adequately predicted the experimental effects of varying the rotation rate and eluent velocity. The extended model, however, with its use of the nonlinear, multicomponent, competitive Langmuir adsorption isotherm, was also capable of accurately predicting the effects of varying the feed rate and concentration.

With these models in hand, a design engineer should be able to plan confidently new and larger rotating annular chromatographs for performing continuous separations on a preparative scale. With its continuous feed and products withdrawal, its adaptability to large-scale operations, and its capability to separate many solutes by using one or a number of eluents, the rotating annular chromatograph should make chromatography a more competitive process in the industrial sector.

INTRODUCTION

A number of investigators have studied the development of a continuous chromatographic system for use in preparative separations. Martin (1949) was the first to propose the concept of an annular chromatograph rotating with respect to a continuous feed stream and product collection points, as shown in Figure 1. The sorption bed results from packing the annular space formed between the two concentric cylinders. This entire bed is slowly rotated while the feed material to be separated is continuously introduced at a stationary point at the top of the bed with eluent fed everywhere else around the annulus. The resulting elution chromatog-

raphy, coupled with rotation of the sorbent bed, causes the separated components to appear as helical bands, each of which has a characteristic stationary exit point.

Figure 1 depicts the separation of two components, with the slope of each being dependent on three factors: (1) the distribution coefficient of the species under the given feed, eluent, and sorbent bed conditions; (2) the eluent velocity; and (3) the rotation rate of the annulus. As long as these factors remain constant, the slope of each constituent (and hence its exit point) will also remain constant. The part of the bed that does not receive feed at a given time is undergoing either elution or regeneration. Thus feed can be continuously pumped into the device and stationary collectors can continuously withdraw separated components for any desired period, thereby making the rotating annular chromatograph a truly continuous process.

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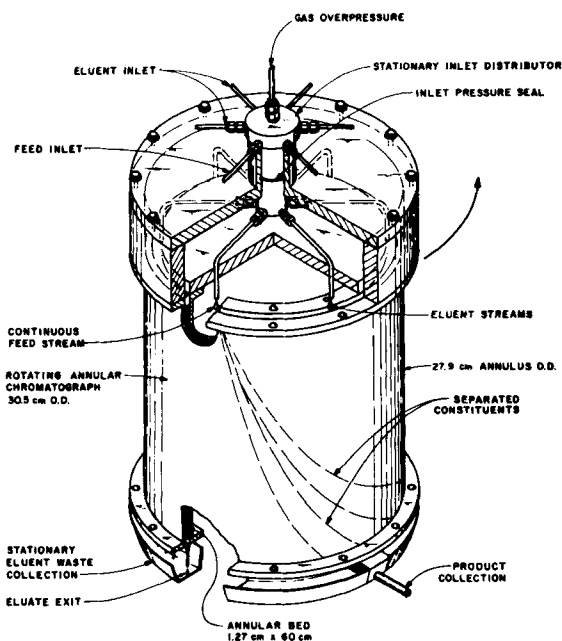


Figure 1. Schematic of the 279-mm-diam continuous annular chromatograph (CAC-II).

Work has been in progress at ORNL since 1974 to demonstrate the feasibility of this continuous chromatography concept for making preparative separations. Scott and coworkers (1976) were the first to assemble a continuous annular chromatograph (CAC) capable of pressurized operation. Canon and Sisson (1978) described some equipment modifications which improved the consistency of the performance data and, more importantly, allowed a gradient elution mode of operation. Canon et al. (1980) presented experimental results on several cation exchange separations, including: (1) the separation of iron and aluminum in ammonium sulfate-sulfuric acid solutions, (2) the separation of hafnium from zirconium in sulfuric acid solutions, and (3) the separation of copper, nickel, and cobalt in ammonium carbonate solutions. Investigation of the difficult separation of zirconium and hafnium, which is important to the nuclear industry, was continued by Begovich and Sisson (1983), who studied the effects of feed flow rate and metal feed concentration on the separation of the two metals. They concluded that a 279-mm-diam. device, such as that shown in Figure 1, could produce 150 kg of nuclear-grade (<100 ppm hafnium) zirconium per year. Begovich and Sisson (1982) also presented experimental results detailing the effects of rotation rate, feed bed loading, and column size on CAC performance. A more complete description of the background of annular chromatography has been provided by Begovich (1982).

THEORY

Modeling of a CAC has been done by using a variety of methods. Plate theory, as modified by Scott et al. (1976) to apply to a CAC, will be discussed first. This discussion will be followed by the analytical solution of Rhee et al. (1970) to the idealized partial differential equations. Finally, an extension to this solution to include dispersion effects will be presented.

Stage Model

Scott et al. (1976) mathematically described a CAC by a theoretical-plate approach similar to that developed for conventional chromatographic columns. After the dimensional translation from time to angular position has been made, as follows:

$$\theta = \omega t, \quad (1)$$

the solute exit position can be related to the operating parameters and the distribution coefficient by the expression,

$$\bar{\theta} = \frac{\omega L}{V} [\epsilon + (1 - \epsilon)K]. \quad (2)$$

By recognizing the contribution of the initial feed bandwidth, the final exit bandwidth could be expressed as:

$$W = \sqrt{W_0^2 + 16\bar{\theta}^2/N}. \quad (3)$$

The initial feed bandwidth can be obtained by visual observation or can be estimated from the following relationship:

$$W_0 = 360Q_f/(Q_f + Q_E). \quad (4)$$

The use of this equation assumes that as the feed leaves the feed nozzle, its velocity is changed so that it matches the eluent velocity. Thus, the feed bandwidth would occupy that part of the total annulus as indicated by the ratio of its flow rate to the total flow rate. This also assumes that the feed is distributed uniformly over the entire calculated bandwidth. The use of Eq. 4 has been found to correlate very well with the initial bandwidths observed visually.

The resolution between two chromatographic peaks is defined as the ratio of the distance separating two solute peaks to their average bandwidth. Thus,

$$R = \frac{\bar{\theta}_2 - \bar{\theta}_1}{\frac{1}{2}(W_1 + W_2)} = \frac{2(\bar{\theta}_2 - \bar{\theta}_1)}{W_1 + W_2}. \quad (5)$$

For Gaussian peaks, a resolution value of unity indicates no dead volume between two species but virtually no overlapping either. If the distribution coefficients and plate heights are known for a pair of solutes, the resolution between them can be predicted through use of Eqs. 2 through 5.

Differential Model

Wankat (1977) has described the relationship between conventional chromatographic columns and a rotating CAC. He noted that conventional chromatography is an unsteady-state one-dimensional process which separates a feed mixture on a time basis, whereas chromatography in a rotating annular chromatograph is a steady-state two-dimensional process which separates the feed mixture in a spatial direction. When radial gradients in velocity and concentration can be ignored, the mass balance on a differential element in a conventional packed bed with constant eluent velocity results in the following:

$$\frac{\partial c}{\partial t} + \left(\frac{1 - \epsilon}{\epsilon} \right) \frac{\partial n}{\partial t} = D_z \frac{\partial^2 c}{\partial z^2} - \frac{V}{\epsilon} \frac{\partial c}{\partial z}, \quad (6)$$

where the initial condition is:

(1) for $t = 0$, all z : $c = n = 0$,

and the boundary conditions are:

(2) for $z = 0$, $0 < t \leq t_f$: $c = c_f$
 $t_f < t$: $c = 0$,

(3) $\lim_{z \rightarrow \infty} c = 0$.

The steady-state mass balance in a rotating CAC, again ignoring any radial gradients, yields the following:

$$\omega \frac{\partial c}{\partial \theta} + \omega \left(\frac{1 - \epsilon}{\epsilon} \right) \frac{\partial n}{\partial \theta} - D_\theta \frac{\partial^2 c}{\partial \theta^2} = D_z \frac{\partial^2 c}{\partial z^2} - \frac{V}{\epsilon} \frac{\partial c}{\partial z}, \quad (7)$$

where the boundary conditions are:

(1) for $\theta = 0$, all z : $c = n = 0$

(2) for $z = 0$, $0 < \theta \leq \theta_f$: $c = c_f$
 $\theta_f < \theta \leq 360 \text{ deg}$: $c = 0$

(3) at $z = -\infty$, $0 < \theta \leq \theta_f$: $c = c_f$
 $\theta_f < \theta \leq 360 \text{ deg}$: $c = 0$

(4) at $z = \infty$, all θ : $c = 0$.

A comparison between Eqs. 6 and 7 shows a term-by-term correspondence with the exception of the angular dispersion term in Eq. 7. If the angular dispersion is considered negligible and the transformation of Eq. 1 is made, then Eqs. 6 and 7 are identical. Thus, under conditions of negligible angular dispersion, the unsteady-state one-dimensional process is analogous to the steady-state two-dimensional rotating process. As confirmed by a number of investigators (Begovich, 1982), angular dispersion in a rotating CAC has indeed been found to be negligible.

If axial dispersion can be neglected, Eq. 6 can be simplified to the following:

$$\frac{\partial c}{\partial t} + \left(\frac{1-\epsilon}{\epsilon} \right) \frac{\partial n}{\partial t} + \frac{V}{\epsilon} \frac{\partial c}{\partial z} = 0. \quad (8)$$

Rhee et al. (1970) obtained an analytical solution to this equation with the assumption that local chemical equilibrium exists throughout the sorption bed. They used the nonlinear Langmuir competitive solute-resin binding isotherms common to many separations to relate concentrations of the liquid- and solid-phase solutes as follows:

$$n_i = \frac{N_c K_{Li} c_i}{1 + \sum_{j=1}^m K_{Lj} c_j} \quad (9)$$

Note that Eq. 9 adequately describes the saturability of a sorbent as the concentration of solute in the liquid phase is increased. The basic concept assumes that the sorbent particles have only a finite number of binding sites and the tendency of any particular solute to bind decreases as sites become filled. In multicomponent chromatography, this means that the presence of a solute with a higher affinity for the sorbent (and thus a larger K_L value) will effectively displace solutes with lower sorbent affinities. Linear isotherm models cannot include this effect. At low liquid solute concentrations, the Langmuir isotherm parameter and distribution coefficient are related by the following approximation:

$$K \approx N_c K_L \quad (10)$$

The use of the Langmuir competitive isotherm in Eq. 8 means that m partial differential equations must be solved simultaneously. Rhee et al. (1970) analytically solved these equations by setting them up as a Riemann's problem and determining the m generalized Riemann invariants. Bratzler and Begovich (1980) described a computer program that applied the solution of Rhee et al. to a rotating CAC using the transformation of Eq. 1. This solution, applied to an experimental run in the 279-mm-diam. CAC-II, is shown in Figure 2. The experiment involved the cation exchange separation of copper (Cu), nickel (Ni), and two cobalt species (represented as Co-1 and Co-2). The top half of the figure shows the predicted band profiles from the initial bandwidth at the top of the column to their exit at the bottom of the CAC. The bottom half of the figure shows the exit concentration profile of each solute. The solid curves represent the experimental data obtained from the chromatogram, while the dotted lines represent the predicted profiles from the solution of Rhee et al. The predicted baseline bandwidth of each component can be seen to match the widths of the band profiles at the bottom of the column shown in the upper half of the figure. These predicted bandwidths, however, are much narrower than those experimentally observed. This leads to an overprediction of the peak height of each solute as compared with the experimental peak height. Both the overpredicted peak heights and the underpredicted bandwidths are the result of the assumption of negligible axial dispersion made in Eq. 8.

Extension to the Idealized Analytical Model

Chromatographic peaks have often been observed to be Gaussian shaped. As such, the mean of the curve and its variance completely describe the normal Gaussian curve. Levenspiel (1972) related the Gaussian curve variance to a vessel dispersion number (VDN) through the following equation:

$$\sigma^2 = 2\epsilon D_z / VL = 2 \cdot \text{VDN} \quad (11)$$

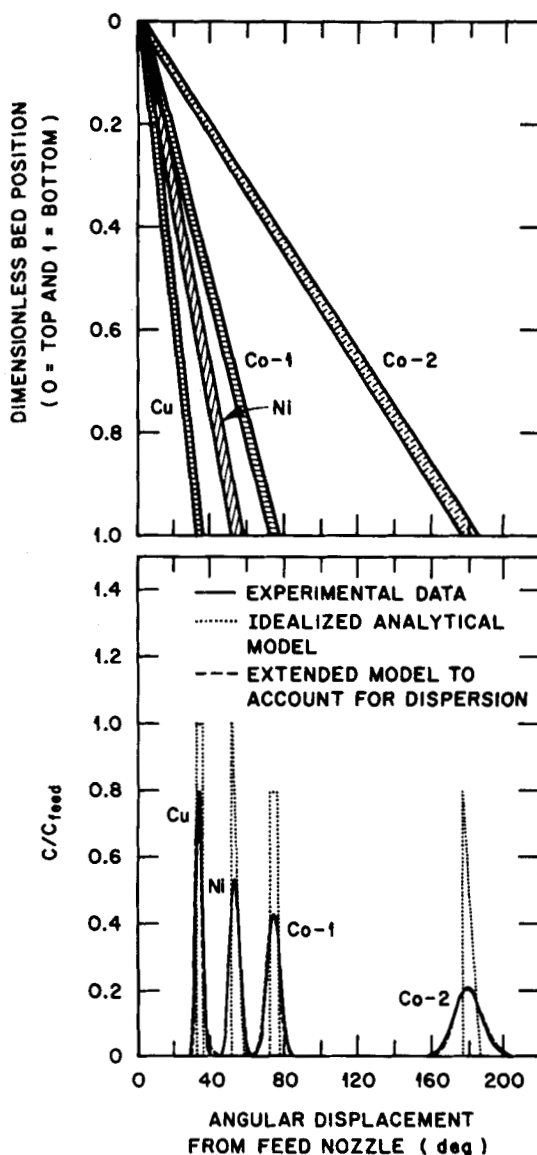


Figure 2. Comparison of experimental concentration profiles with model predictions.

Using this information, it is now possible to extend the solution of Rhee et al. (1970) to include estimates of the effects of dispersion. Essentially, this was accomplished by superimposing a variance (i.e., dispersion) onto concentration profiles predicted by the model of Rhee et al. In practice, the predicted concentration profile for each solute was divided into 0.25-deg increments. Next, the area of the first increment was calculated. This increment was then spread out into a Gaussian-shaped curve using the known mean and area of the increment, along with a given variance. These two procedures were subsequently repeated for all of the increments for a particular solute. The final dispersed curve for the solute was determined by summing all of the incremental Gaussian curves. By comparing the experimental concentration profiles with the dispersed predicted profiles, it was possible to determine variance values for each solute that gave a best fit. It should be noted that variance values obtained by this procedure are really lumped parameters, representing the combined effects of dispersion, mass transfer, and end effects.

The dashed lines shown in the lower half of Figure 2 represent the extended model, using the dotted lines of the Rhee et al. solution as an initial starting point. Agreement between the extended model and the experimental data is excellent. The computer program by Bratzler and Begovich (1980) has been modified to include this extended model. The modified program, CACMCS (Continuous

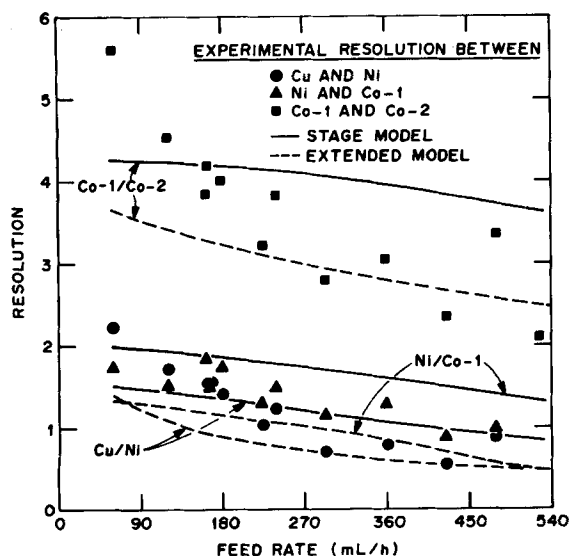


Figure 3. Effect of feed rate on resolution.

Annular Chromatographic Multicomponent Separations), has been described by Begovich (1982). The same procedure was applied to more than 140 runs made in CAC units whose diameters ranged from 89 to 445 mm. In general, the agreement between the extended model and the experimental concentration profiles was similar to that shown in Figure 2, i.e., excellent.

The extended model was then compared with each of 13 conventional column experimental runs. The variance value for each solute was allowed to vary to obtain a best fit between the experimental and predicted profiles. These runs were performed using the same cation exchange separation system of copper, nickel, and cobalt mentioned previously, in columns of varying bed length (0.61 to 1.22 m) and with varying eluent superficial velocity (0.40 to 2.44 mm/s). Although the solute variance values tended to increase as the solute retention time increased, the VDN values for each solute were very similar. When the solute VDN values were averaged and regressed against the eluent Reynolds number, the following correlation was obtained:

$$\overline{VDN}_x = 1.06 \times 10^{-2} \cdot Re^{0.77} \quad (12)$$

This correlation was found to adequately represent the average solute VDN values obtained when the extended model was applied to each of the experimental runs made in the CAC units.

Thus, in the case of a new separation system for which a CAC design is desired, the logical starting place is a conventional column. The distribution coefficients for the system, as well as the dispersion values, can be determined in the conventional column. The dispersion values can be average solute VDN values, similar to those correlated in Eq. 12, or they can be direct variance or dispersion coefficients, depending on the conventional column results for the particular separation system. These values can then be input to the program CACMCS, and the performance of a CAC can be predicted.

COMPARISON OF MODELS WITH EXPERIMENTAL RESULTS

The stage model, as represented by plate theory, and the extended model of Rhee et al. to include dispersion effects can now be compared with CAC experimental results. In particular, the effects of feed rate, feed concentration, rotation rate, and eluent velocity on the resolution between pairs of solutes will be described. All other parameters were held constant during each set of experiments. Constant K and K_L values for each solute were used in the two models, while an average N value was used in the stage model and Eq. 12 was used in the extended model.

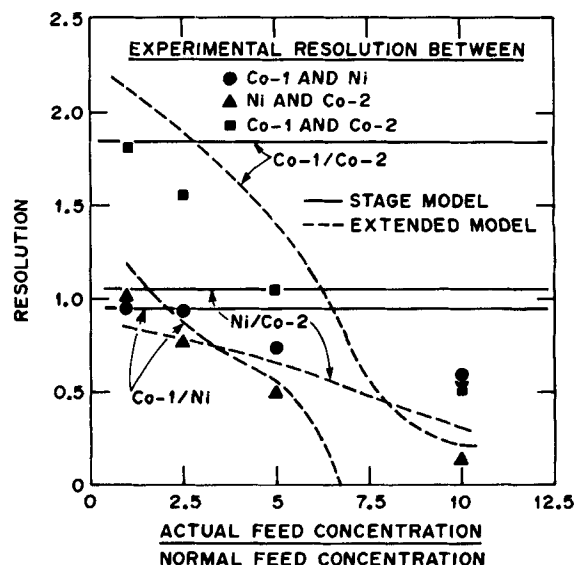


Figure 4. Effect of feed concentration on resolution.

Effect of Feed Rate

Figure 3 shows that the resolution between adjacent solute pairs decreases as the feed rate is increased, with all other parameters held constant (i.e., eluent velocity and rotation rate). This effect was expected since the bandwidths increase as the bed loading is increased, making overlapping of the bands more likely. The stage model, represented by the solids curves, tends to fit the experimental data best at low feed rates, while the extended model fits the data better at high feed rates. Plate theory is limited at high feed rates or concentrations because of its use of a linear adsorption isotherm. Of the two models, the extended model tends either to predict the resolution well or to underpredict it, leading to conservative design values.

Effect of Feed Concentration

The effect of feed concentration on resolution is shown in Figure 4. In this set of experiments, performed on the 279-diam ORNL CAC by Torres et al. (1981), a normal feed concentration was defined as 4 g Ni/L and 1 g/L each of Co-1 and Co-2. Thus, an abscissa value of 10 means that the actual feed mixture used for their experiment contained 40 g Ni/L and 10 g/L each of Co-1 and Co-2.

As the feed concentration was increased, the bed loading was increased and in turn the resolution values decreased. Plate theory, with its assumption of a linear isotherm, predicted constant resolution as the feed concentration was varied. Such predictions are obviously in error at the higher concentrations used in this set of experiments. On the other hand, the nonlinear multicomponent competitive Langmuir isotherm used in the extended model correctly predicted the decrease in resolution as the concentration was increased.

Effect of Rotation Rate

When the feed and eluent rates were held constant and the rotation rate was varied from 15 to 90 deg/h, the resolution increased with increasing rotation rate, as shown in Figure 5. The increase in resolution can be explained by considering the decrease in bed loading as the rotation rate was increased while the feed rate was held constant. Plate theory predicts, in agreement with the experimental data, that the resolution values will initially increase but will level off with increasing rotation rate. However, it also appears to predict that this leveling of the resolution will occur at lower rotation rates than those actually observed. The extended

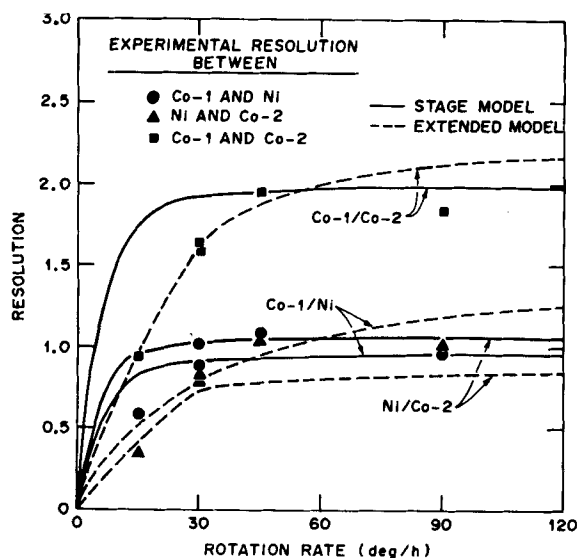


Figure 5. Effect of rotation rate on resolution using a constant feed rate.

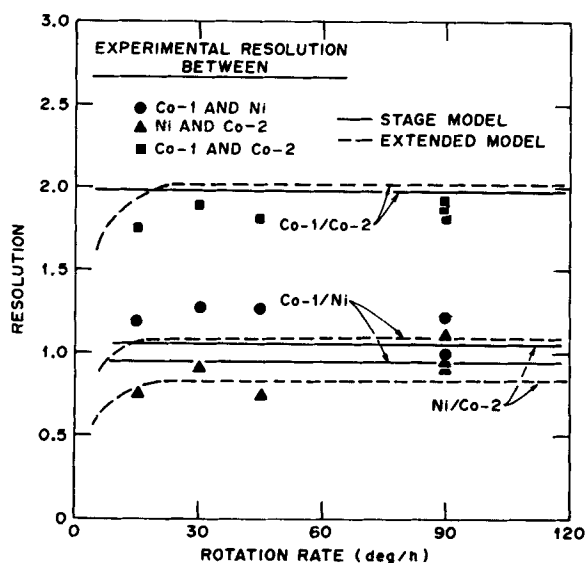


Figure 6. Effect of rotation rate on resolution using a constant feed rate to rotation rate ratio.

model, as would be expected in the higher bed loading region, appears to fit the data at low rotation rates quite well.

When the bed loading was held constant by keeping the ratio of feed rate to rotation rate constant, the experimental resolution values were unaffected by changes in the rotation rate, as shown in Figure 6. Both plate theory and the extended model predict this behavior correctly, although the extended model indicates that the resolution values would start to decrease if rotation rates of less than 15 deg/h were used.

Effect of Eluent Velocity

Figure 7 shows the effect on resolution of increasing the eluent velocity while keeping the rotation rate and ratio of feed to eluent rates constant. Plate theory accounts for changes in the eluent rate by varying the plate height of a theoretical plate, while the extended model sees such changes as varying the vessel dispersion number in Eq. 12. Both models predict the separation between Ni and Co-1 quite satisfactorily, while the extended model more accurately represents the Cu/Ni and Co-1/Co-2 separations.

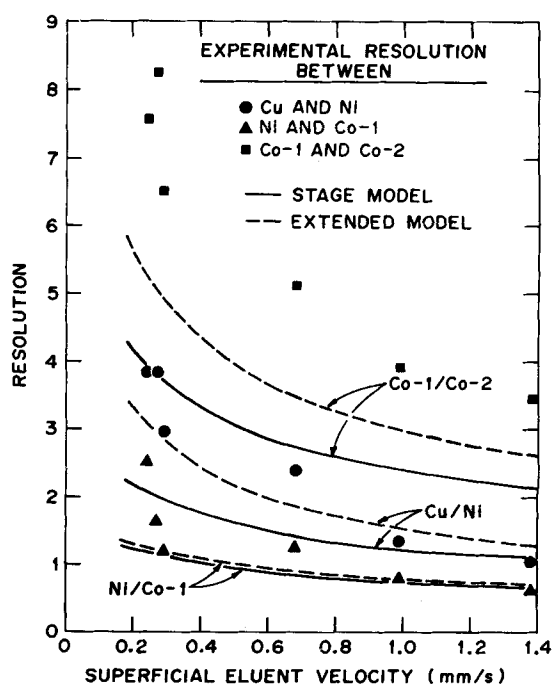


Figure 7. Effect of eluent velocity on resolution.

DISCUSSION

Within the limitations imposed by its assumption of a linear isotherm, plate theory can be applied to predict the performance of a CAC unit with fair confidence. As noted by Begovich and Sisson (1982b), plate theory was quite useful in scaling up a CAC to both larger sizes and higher operating pressures.

When a nonlinear adsorption isotherm must be considered, the multicomponent Langmuir isotherm used by Rhee et al. (1970) should clearly give superior results. However, the lack of a dispersion term in their solution causes the solute concentration profiles to be poorly predicted. By extending the solution of Rhee et al. to include dispersion effects, excellent agreement between predicted and experimental concentration profiles could be obtained. In addition, the effects of feed rate, feed concentration, rotation rate, bed loading, and eluent velocity could be accurately predicted.

Both models require knowledge of the solute distribution coefficients and a dispersion term. Plate theory incorporates the dispersion term in its plate height, while the extended model considers dispersion to be related to a band's variance in a correlation of the form of Eq. 12. For both models, these parameters are most easily obtained by performing the desired separation in a conventional column. With such information and the two models in hand, a design engineer should be able to plan new and larger CAC units for performing continuous separations on a preparative scale.

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NOTATION

c	= solute concentration in the liquid phase, mol/m ³ solution
c_f	= liquid-phase feed concentration, mol/m ³ solution
d	= sorbent particle diameter, m
D_z	= axial dispersion coefficient, m ² /s
D_θ	= circumferential dispersion coefficient, deg ² /s
K	= distribution coefficient, m ³ solution/m ³ sorbent

K_L = Langmuir isotherm parameter, m^3 solution/mol
 L = bed length, m
 m = number of components
 n = solute concentration in the solid phase, mol/ m^3 sorbent
 N = number of theoretical plates
 N_c = sorbent capacity, mol/ m^3 sorbent
 Q_f = feed rate, m^3/s
 Q_E = eluent rate, m^3/s
 R = resolution
 Re = $\rho dV/\epsilon\mu$ = eluent Reynolds number
 t = time, s
 t_f = time feed is applied to column, s
 V = superficial eluent velocity, m/s
 VDN = $\epsilon D_z/VL$ = vessel dispersion number
 VDN_x = $D_z \theta/\omega L^2$ = solute vessel dispersion number
 W = solute baseline bandwidth, deg
 W_o = initial feed bandwidth, deg
 z = axial coordinate, m

Greek Letters

ϵ = bed void fraction, m^3 voids/ m^3 bed
 θ = angular coordinate, deg
 $\bar{\theta}$ = solute peak position, deg
 θ_f = angle feed is applied to column, deg
 μ = eluent viscosity, kg/m-s
 ρ = eluent density, kg/ m^3
 σ^2 = variance
 ω = rotation rate, deg/s

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Influence of Hydrodynamics on Crystal Growth and Dissolution in a Fluidized Bed

Mass transfer coefficients for the growth and the dissolution of potassium alum and sodium thiosulfate in a laboratory-scale fluidized-bed apparatus were determined and correlated in a form of the Frössling equation. The surface reaction rate for potassium alum was found to be dependent on the crystal size.

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SCOPE

The mass transfer in liquid-solid systems has been studied extensively for the dissolution process. Only a few reports, however, exist concerning the mass transfer from the liquid phase into the solid phase. This process is only rarely linearly proportional to the driving force and thus cannot be regarded as a simple reciprocity of the dissolution. The method introduced previously (Karpinski, 1980), based on the two-step growth concept, allows us to determine: (1) a mass transfer coefficient

for the crystal growth, and (2) a surface reaction rate constant. The method has not yet been fully verified by joint dissolution-crystallization experiments.

The purpose of this work is to compare the mass transfer coefficients for both dissolution and diffusional steps of crystal growth and their dependence on the hydrodynamics, as well as to verify a possible dependence of the surface reaction rate on the crystal size.

A fluidized bed has been selected as the experimental system since it seemed to ensure well-defined hydrodynamic conditions, particularly when monodimensional crystals were used.

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