

- Chen, H. T., and F. B. Hill, "Characteristics of Batch, Semi-continuous, and Continuous Equilibrium Parametric Pumps," *Separ. Sci.*, **6**, 411 (1971).
- Grevillot, Georges, and Daniel Tondeur, "Equilibrium Staged Parametric Pumping: I—Single Transfer Step per Half-cycle and Total Reflux; the Analogy with Distillation," *AIChE J.*, **22**, 1055 (1976).
- Sweed, N. H., and R. A. Gregory, "Parametric Pumping: Modeling Direct Thermal Separations of Sodium Chloride—Water in Open and Closed Systems," *ibid.*, **17**, 171 (1971).
- Sweed, N. H., and R. H. Wilhelm, "Parametric Pumping: Separations via Direct Thermal Mode," *Ind. Eng. Chem. Fundamentals*, **8**, 221 (1969).
- Thompson, D. W., and B. D. Bowen, "Equilibrium Theory of the Parametric Pump. Effect of Boundary Conditions," *ibid.*, **11**, 415 (1972).
- Wakao, N., H. Matsumoto, K. Suzuki, and A. Kawahara, "Adsorption Separation of Liquid by Means of Parametric Pumping," *Kagaku Kogaku*, **32**, 169 (1968).
- Wankat, P. C., "Liquid-Liquid Extraction Parametric Pumping," *Ind. Eng. Chem. Fundamentals*, **12**, 372 (1973).
- Wilhelm, R. H., D. W. Rolke, and N. H. Sweed, "Parametric Pumping: A dynamic Principle for Separating Fluid Mixtures," *ibid.*, **7**, 337 (1968).

## APPENDIX

The limit regimes of Figures 2 to 9 and the separation factors of Figures 10, 11, and 12 have been calculated cycle by cycle with a computer using extended precision. The nonlinear isotherms are described by Equations (3a) and (3b) and the linear isotherms by Equation (4a) and (4b). The parameter values are given below.

Figures	Isotherms		$\alpha$	$\alpha'$	$x_0$	$y_0$	$\rho$
2	Nonlinear		3.5	1.5	0.2	0.467	1
3, 4, 5	Nonlinear		3.5	1.5	0.4	0.7	1
6, 7, 8	Linear		3.5	1.5	0.01	0.035	1
9, 10	Nonlinear		3.5	1.5	0.4	0.7	1
11	Nonlinear		3.5	1.5	0.4	0.7	0.1
12	Nonlinear		3.5	1.5	0.4	0.7	Varies*

\*  $\rho = P_s \cdot N = N$  for staged-reservoir pumps,  $\rho = P_m \cdot N/n = N/n$  for mixed-reservoir pumps.

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# A Solid / Liquid Separation Process Based on Cross Flow and Electrofiltration

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The cross flow/electrofiltration process combines migration of particles in the presence of electrical and shear fields to increase filtration rates. A mathematical model and the experimental data are presented for both a kaolin clay suspension and an oil in water chemically stabilized emulsion. When the filter is operated in a regime above the critical voltage, filtration rate-electric field strength dependence becomes linear, and both electroosmosis in the filtration medium and electrophoresis in the liquid film are the controlling mechanisms of transport. Both experimental data and the mathematical model indicate that the fluid circulation rate tangential to the filtration media does not necessarily increase filtration rate depending on the regime of operation. The model is used in conjunction with the experimental data to separate the liquid film resistance adjacent to the filter cake which is determined by fluid shear and electric field from the cake and filter medium resistances which are influenced by electroosmosis.

## SCOPE

The cross flow/electrofilter employs two particle transport mechanisms to minimize accumulation of particles at the filter medium. The process combines particle migration away from the filter due to fluid shear (cross-flow filtration) and electrophoretic migration (electrofiltration). In addition to these two mechanisms, electroosmosis in the filter medium, filter cake, or both can influence the filtration rate. The process offers the potential of both improved filtration rates and steady state operation for filtration of suspensions of micron or submicron sized particles. Most particles in aqueous media are negatively charged. The process should find application in filtration

of suspensions where there is relatively low conductivity in the continuous phase. This is true for low ionic strength, aqueous media, and nonaqueous suspending media. The combination of the electrical field with a shear field should be particularly attractive for suspensions of charged, shear sensitive particles. This paper deals entirely with the filtration rate improvement aspects of the cross flow/electrofilter. In principle, this process can be used for particle-particle fractionation of multicomponent particulate suspensions.

The principal objective of this investigation was to experimentally test a mathematical model which permits separation of the effects of the particle concentration polarization in the liquid film on the feed side of the filtration medium, the filter medium resistance including electroosmotic effects, and the filter cake resistance which

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can also be influenced by electroosmosis. The experimental data were obtained from a cross flow/electrofilter which permitted variation of the electric field strength from 0 v/cm to 50 v/cm, circulation rate with a Reynolds number from 1 200 to 3 600, and pressure driving force from 0.7 to  $2.8 \times 10^4$  N/M<sup>2</sup>. Performance data were obtained both for a particulate suspension (Kaolin clay/water) and a chemically stabilized oil/water emulsion.

Bier (1959) was the first to develop a filtration technique using an electrical field to dewater colloidal suspensions. Subsequently, Cooper et al. (1965), Moulik et al., (1967), and Bier (1971) modified the batch filtration equations to include the effect of particle mobility in the slurry adjacent to the filtration medium and included a correction for electroosmosis both in the filter cake and in the filter medium. They did not consider the influence of fluid shear tangential to the filter medium and did

not incorporate concentration polarization concepts in their mathematical model or in the interpretation of their data. Giddings (1966) and Grushka et al. (1973) have extensively investigated a process called electrical field flow fractionation to fractionate either mixed particle suspensions or solutions of macromolecules. Field flow fractionation involves the idea of electrical field normal to an axial flow between two flat plates. It does not simultaneously involve a filtration flux in the direction normal to the flow. This is the distinguishing feature between electrical field flow fractionation and cross flow/electrofiltration. Further, Giddings' principal objective was to fractionate suspended or dissolved solutes as opposed to the primary emphasis being placed on filtration rate improvement. Reis et al. (1976) have investigated a similar process which they call electropolarization chromatography.

## CONCLUSIONS AND SIGNIFICANCE

Owing to the fact that both the Kaolin particles and the oil droplets are negatively charged in aqueous suspensions, a direct electric field will always give higher filtration rates than cross-flow filtration alone. The level of improvement depends on the intensity of fluid shear and the electric field strength.

A mathematical model for the cross flow/electrofiltration process was developed which includes the influence of the electrophoretic mobility of either the particles or suspension drops on the film resistance, medium resistance, and cake resistance. In the case of the latter two resistances, electroosmotic effects were included. It is possible to separate these three resistances by using both the experimental data and the mathematical model. For the slurries investigated, the performance of the cross flow/electrofilter was dominated by both electroosmosis and electrophoresis at high voltages. Both the electroosmotic coefficient of the filter medium and the electrophoretic mobility of the charged particle can be used with the mathematical model to calculate the slope of the filtration

flux-voltage dependence at high voltage. This calculated value of the slope at high voltages agrees well with the value that was obtained from the experimental data. Experimental data also confirmed that at voltages below critical, the experimental filtration rate increased with the Reynolds number or the tangential velocity past the filtration medium. At the critical voltage, there was no dependence of the filtration rate on the circulation velocity. Finally, at voltages above critical, increases in the tangential circulation velocity reduced the filtration rate.

This process combines three distinct transport mechanisms to improve filtration rate: electrophoretic mobility in the liquid adjacent to the filter particle, transport induced by the high circulation velocity, and electroosmosis. The mathematical model permits separation of these effects so that performance data can be interpreted. The model, however, does not predict a priori the film resistance which is dominated by the concentration polarization phenomena. This aspect of the cross flow/electrofiltration process is presently being investigated and will be the subject of a future paper.

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## CROSS FLOW/ELECTROFILTRATION CONCEPT

The cross flow/electrofilter employs two particle transport mechanisms to minimize the accumulation of particles at the filter medium. The process combines particle migration away from the filter due to fluid shear (cross-flow filtration) and electrophoretic migration (electrofiltration). The process concept is illustrated in Figure 1.

There are three factors (bulk flow, fluid shear, and electrophoretic migration) which can cause particle transport normal to the filter medium. The movement of the liquid phase towards and through the filter medium can cause a particle transport in that direction due to bulk flow. In fact, in a conventional filtration process, this leads to an accumulation of filter cake next to the filter medium and at constant driving force produces the filtration rate-time decay. Depending on the regime of operation, fluid shear can cause particle transport either away from or toward the filter medium (see later discussion of the regimes of operation). Most particles in aqueous suspension have a negative charge. Consequently, an electrical field with

appropriate polarity will cause the particles to migrate away from the filtration medium. In contrast to conventional dead-end filtration, the cross flow/electrofiltration process can be operated at steady state, that is, with no filtration rate-time decay. At steady state there is a balance between the rates of migration of particles towards the filter medium due to bulk flow and the rate of migration of particles away from the filter medium due to both shear and electrophoretic migration effects.

Depending on the regime of operation, there is a possibility that electrodeposition of particulate material can occur at the electrode which is away from the filtration medium; that is, the particles which are transferred away from the filtration medium by both shear and electrical forces may accumulate at the other electrode. There are several approaches to avoiding or minimizing electrodeposition. These will be discussed in a later section.

The combination of particle transport away from the filter medium both by shear and electrical effects offers the following potential process advantages:

1. Increased filtration rate over that which can be obtained by either conventional pressure filtration or cross flow or electrofiltration alone.

2. Steady state operation.

3. Filtration rates are high enough with microporous filtration media so that micron sized gelatinous particles can be dewatered without a filter aid. This is an important consideration when the product is the particles; that is, it is possible to avoid contamination of the product with filter aid.

4. The electrical field increases the filtration rate over that which can be obtained with cross-flow filtration alone. This is of particular interest when there is a desire to minimize the shear degradation of the particles which are being processed.

## PREVIOUS WORK

The cross-flow filtration process has been investigated extensively. Henry (1972) has discussed the status of cross-flow filtration module development, reviewed many applications of the cross-flow filter, and discussed various mathematical models which have been used to interpret cross-flow filtration performance data. Virtually all mathematical models for the cross-flow filtration process are based on a representation of the particle concentration polarization phenomenon that occurs in the liquid upstream to the filter medium. Dahlheimer et al. (1970) have used cross-flow filtration to dewater suspended solids such as clay slurries. Henry and Allred (1972) used the cross-flow filtration process to concentrate bacterial cells, and Porter (1972) has presented performance data for a large number of suspensions.

Manegold (1937) was the first to appreciate the potential of combining the processes of conventional pressure filtration and electrophoresis. Beechold (1926) utilized a combination of electroosmosis and electrophoresis to purify colloids in an apparatus he called an electro ultrafilter. Much later Bier (1959) developed a membrane technique using an electrical field to dewater colloidal suspensions. Cooper et al. (1965), Moulik et al., (1967), and Bier (1971) have modified the batch filtration equations to include the effect of the particle mobility in the slurry adjacent to the filter medium and included a correction for electroosmosis both in the filter cake and filter medium. They did not consider the influence of fluid shear tangential to the filter medium and did not incorporate concentration polarization concepts in their mathematical models.

Giddings (1966) has extensively investigated a process called field flow fractionation which is in some ways similar to the cross flow/electrofiltration process. Grushka et al. (1973) investigated the case where an electrical field was applied. If we consider the electrical field case as an example, the field flow fractionation concept involves introducing a laminar flow between parallel plates and imposing an electric field in a direction normal to the flow. The electrical field causes dissolved solutes and/or particles which have charge to concentrate in a region near one of the walls. The lower velocity of the laminar flow field in the region next to the wall means that materials that are concentrated in that region will be retarded, while materials that are near the center line will move down the tube to the higher velocity. This process is very much like conventional chromatographic separations in that solute or particle bands can be developed and eluted from the flow channel.

The major differences between field flow fractionation and the cross flow/electrofiltration process include the fact that the cross flow/electrofiltration process not only uses an electrical field, but in addition there is a bulk flow of

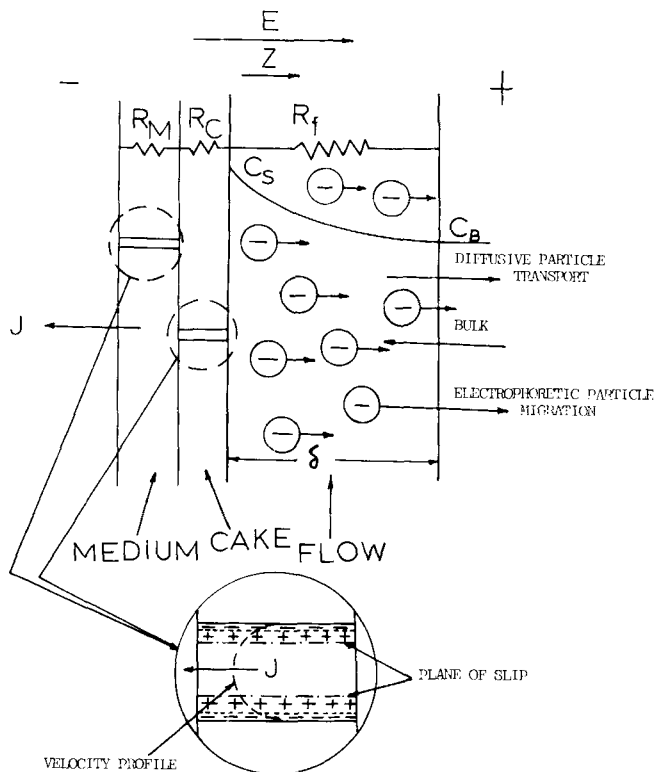


Fig. 1. Cross flow/electrofiltration concept.

fluid toward one of the walls which is the filtration medium. In addition, when the cross flow/electrofilter is operating in a mode for concentration of a suspension, there is no requirement for periodic elution. Giddings principal objective in electrical field flow is to fractionate either dissolved solutes or suspended particles. The principal objective of the research described in this paper by comparison is to concentrate an entire suspension and take advantage of the improved filtration rate that is produced in the cross flow/electrofiltration process.

Giddings et al. (1976) have investigated another variation of their process called flow field-flow fractionation which utilizes a filtration barrier as one of the walls of the flow channel so that the bulk flow of liquid toward that wall acts to concentrate particles in the low axial velocity region near to the filtration medium. Again, this process has some of the elements of the cross flow/electrofiltration process but does not include the simultaneous application of the electric field in a direction normal to the axial flow.

Reis et al. (1976) have recently described a process which they call electropolarization chromatography. They have shown that a hollow fiber geometry is particularly effective for fractionating dissolved proteins. The fiber is immersed in a batch of circulating buffer. The electric field is applied in the traverse direction across the fiber. Electropolarization chromatography is essentially the same as the electric field flow fractionation process of Giddings.

## THEORY

In general, the filtration flux  $J$  can be expressed by the following resistance equation:

$$J = \frac{\Delta P}{R_T} \quad (1)$$

The total resistance is expressed as a function of the film, medium, and cake resistances:

$$R_T = R_f \Delta P + R_M + R_C \quad (2)$$

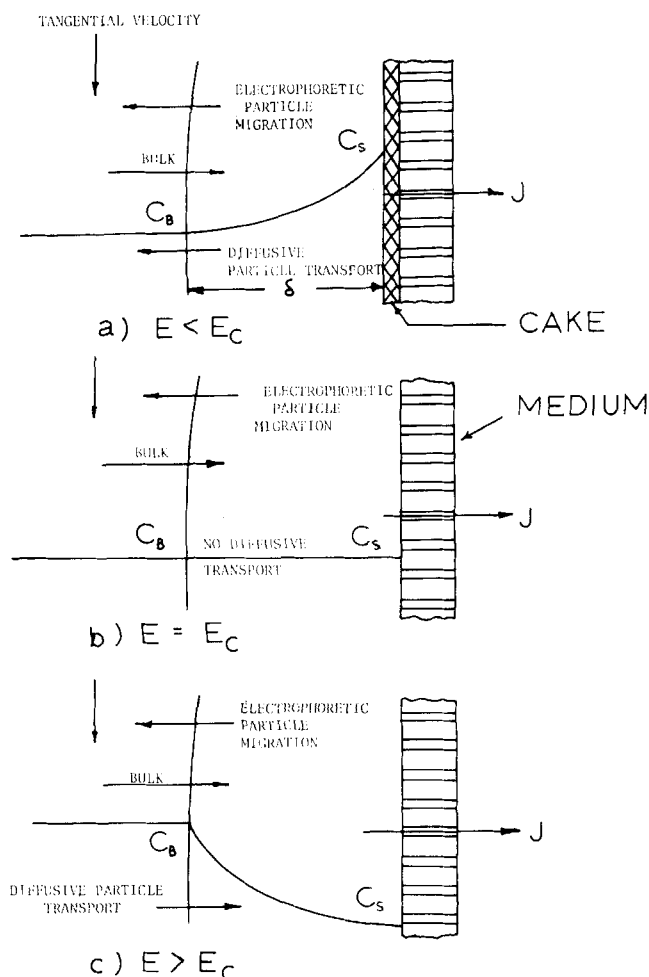


Fig. 2. The three regimes of operation of a cross flow/electrofilter.

The film resistance is multiplied by the pressure driving force because in the case of film or concentration polarization control, the flux must be independent of this driving force. This is discussed in detail below. The definitions of medium or cake resistances include the corresponding medium or cake thickness. Since steady state experimental data will be used, the thickness will be constant. There is no advantage to defining the resistance separate from the thickness.

The principal objective of the model is to separate the effects of the film resistance, medium resistance, and cake resistance, and, in particular, to distinguish between the influence of the electrical and shear fields on the transport mechanisms.

#### Film Resistance

Particle transport in the film on the feed side of the filter medium includes bulk flow of particles due to the movement of filtrate toward the filter, diffusion of particles due to the concentration gradient and fluid shear effects, and migration of particles due to electrophoretic mobility in the presence of direct electrical field.

There are three distinct regimes of operation of a cross flow/electrofilter. These are illustrated in Figure 2. Before they are discussed, it is necessary to define the concept of the critical voltage  $E_c$ . The critical voltage is defined as the voltage at which the net particle migration velocity toward the filtration medium is zero, that is, at the critical voltage, there is a balance between the electrical migration and velocity away from the filtration medium in the velocity at which the particles are swept toward the filter by bulk flow.

Figure 2a illustrates the transport in the liquid film for the case where the voltage is less than critical. In this case, there is a net migration of particles toward the filtration medium, and a cake is formed. In addition, the concentration of the particles is higher next to this filter cake than in the bulk stream. The tangential velocity past the filtration media (cross-flow filtration) leads to an increase in filtration rate in this case because particles diffuse at a higher rate down their concentration gradient away from the filtration medium.

The case where the voltage is equal to the critical voltage is illustrated in Figure 2b. In this case, there is a balance between the particle migration velocity due to the bulk flow and the reverse velocity due to the electrical field. Note that there is no tendency for particles to concentrate at any point in the liquid film. Consequently, there is no concentration gradient. The fluid shear tangential to the filter media in the case of critical voltage has no influence on the filtration rate. The fluid shear can only improve the transport of particles down a concentration gradient, and, in this case, there is no concentration gradient.

Finally, the case of voltage greater than critical is illustrated in Figure 2c. In this case, the electrophoretic migration velocity is greater than the velocity caused by the bulk flow. Particles concentrate away from the filtration medium; that is, the particle concentration is lowest next to the filtration medium. In the case of the applied voltage greater than critical, the influence of fluid shear still improves the transfer of particles down the concentration gradient, but in this case, it is toward the filtration medium. Consequently, increases in the velocity tangential to the membrane will reduce rather than increase the filtration rate. Bier (1971) has recognized the concept of critical voltage and three regimes of operations described above. He did not include in the cases for the voltage either above or below critical the influence of fluid shear, that is, the cross-flow filtration effect.

Mathematically, modeling of the concentration polarization phenomenon in the liquid film is exceedingly complex if it is attempted in any generality. In addition to the effects of the concentration polarization phenomenon and the electrophoretic migration of particles in the liquid film, the effect of radial migration of particles away from the filter medium is conceivably important if the particle sizes are large. Since the primary objective of this study was to separate the filter medium, cake, and liquid film resistances, radial migration effects are not included in our analysis. A more detailed investigation of the liquid film resistance is underway in our laboratory. Henry (1972) has discussed the details for the development of this model for the case of cross-flow filtration, that is, where there is no electrical field present. The following equation is obtained if the electrophoretic particle migration is included:

$$J = k \ln \left( \frac{C_s}{C_b} \right) + \mu_E E \quad (3)$$

Here, the electrophoretic mobility  $\mu_E$  ( $E$  is not a constant) is assumed to be constant. Equation (3) would represent the filtration rate for a cross flow/electrofilter only if the film resistance were controlling. Lawler (1976) has presented the details of the derivation of Equation (3). Equations (1), (2), and (3) can be combined for the case of film control to obtain the following expression for the film resistance:

$$R_f = \frac{1}{k \ln \left( \frac{C_s}{C_b} \right) + \mu_E E} \quad (4)$$

### Filter Medium Resistance

The membrane resistance includes both the effects of filter medium permeability and electroosmosis. Electroosmosis occurs if the pores of the filtration medium are charged. The contributions to the filtration flux due to permeability and electroosmotic effects are additive. This is illustrated by Equation (5):

$$J_M = J_{0M} + K_M E \quad (5)$$

Two resistances associated with the filtration medium can now be defined:  $R_M$  which is the resistance in the presence of both liquid permeability and electroosmotic effects and  $R_{0M}$  which is the resistance in the absence of the electrical field. Equations (6) and (7) define these two resistances:

$$J_M = \frac{\Delta P}{R_M} \quad (6)$$

$$J_{0M} = \frac{\Delta P}{R_{0M}} \quad (7)$$

Equations (5), (6), and (7) can be combined to obtain the following expression for the medium resistance:

$$R_M = \frac{R_{0M}}{1 + \frac{K_M E R_{0M}}{\Delta P}} \quad (8)$$

Both the resistance  $R_{0M}$  and the electroosmotic coefficient  $K_M$  are assumed constant.

### Cake Resistance

There is a possibility of forming a filter cake when the electric field strength is less than critical. In this case, there is a net migration of particles towards the filtration media. Often during a start-up of a cross flow/electrofilter, there is some accumulation of particles in a very thin layer next to the filtration media. The cake resistance at zero field strength  $R_{0C}$  and the electroosmotic coefficient  $K_C$  will be assumed constant. The assumption of constant cake resistance at zero voltage assumes that any filter cake that forms is incompressible. In most of the experiments where the pressure is held constant, it is not necessary to consider cake compressibility effects to separate the effects of electroosmosis in the filter cake, filter medium, and the influence of the electrical field on the film resistance. In the event that it would be necessary to consider the influence of the pressure driving force on the cake resistance, there are relatively straightforward procedures for representing the cake resistance dependence on the pressure driving force (see Tiller, 1962, for details).

In most experiments, the filtration rate is either film or medium resistance controlled. In these situations, the influence of cake resistance and the subtlety of including cake compressibility effects would not improve the mathematical model. In fact, it is doubtful that a true cake exists in the sense that particles are in intimate contact in the presence of the shear and electrical induced migration of particles.

An expression for the cake resistance in the presence of an electrical field can be obtained in a manner analogous to the expression developed above for the medium resistance. Equation (9) illustrates the effect of the electric field strength on the cake resistance:

$$R_C = \frac{R_{0C}}{1 + \frac{K_C E R_{0C}}{\Delta P}} \quad (9)$$

### Combination of the Filtration Resistances

The resistances discussed above for the liquid film, filter medium, and filter cake can be combined using Equations

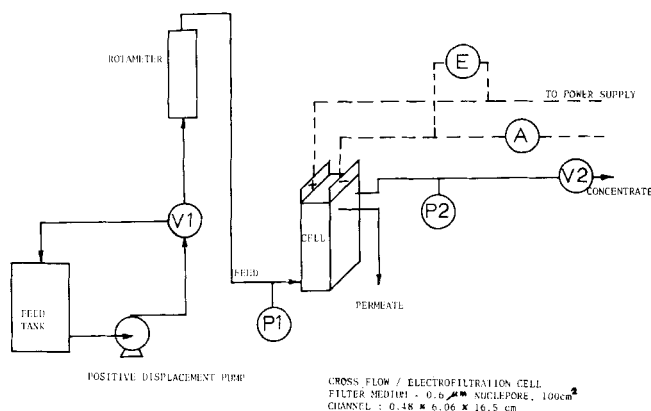


Fig. 3. Experimental cross flow/electrofiltration system.

(1), (2), (4), (8) and (9) to obtain the following expression for the filtration flux for the cross flow/electrofiltration process:

$$J = \Delta P \left/ \left[ \frac{\Delta P}{k \ln \left( \frac{C_S}{C_B} \right) + \mu_E E} + \frac{R_{0M}}{1 + \frac{K_M E R_{0M}}{\Delta P}} + \frac{R_{0C}}{1 + \frac{K_C E R_{0C}}{\Delta P}} \right] \right. \quad (10)$$

It should be noted that  $R_{0C}$  is not a constant. Based on the defining equation for  $R_{0C}$ , it is a function of the cake thickness which can be influenced by the electric field strength opposed across the cake. Sufficiently high field strengths may cause expansion of the cake and thus decrease the resistance. At an electric field strength greater than the critical voltage, the cake resistance can totally be eliminated; that is, Equation (10) will become

$$J = \frac{\Delta P}{\frac{\Delta P}{k \ln \left( \frac{C_S}{C_B} \right) + \mu_E E} + \frac{R_{0M}}{1 + \frac{R_{0M} K_M E}{\Delta P}}} \quad \text{for } E > E_C \quad (11)$$

and if both  $\mu_E E \gg k \ln(C_S/C_B)$  and  $K_M E \gg \Delta P/R_{0M}$ , Equation (11) can be rearranged and simplified in a form

$$J = \left[ \frac{\mu_E K_M}{\mu_E + K_M} \right] E + \frac{K_M k \ln \left( \frac{C_S}{C_B} \right) + \mu_E \frac{\Delta P}{R_{0M}}}{\mu_E + K_M} \quad (12)$$

or

$$J = M E + M_0$$

where

$$M = \left[ \frac{\mu_E K_M}{\mu_E + K_M} \right]; \quad M_0 = \frac{K_M k \ln \left( \frac{C_S}{C_B} \right) + \mu_E \frac{\Delta P}{R_{0M}}}{\mu_E + K_M} \quad (13)$$

At an electric field strength greater than the critical voltage, the filtration rate-electric field strength dependence is a linear function of the electric field strength  $E$ , with a slope which is determined by  $\mu_E$  and  $K_M$ .

TABLE 1. FEED SYSTEMS

	Liquid liquid	Solid liquid
Dispersed phase	Paraffin oil	Kaolin
Continuous phase	Distilled water	Distilled water
Surfactant	Glycerol monooleate	None
Mobility, cm/s/ V/cm	$1.6\text{--}5.5 \times 10^{-4}$	$3.9\text{--}6.6 \times 10^{-4}$
Particle size, $\mu\text{m}$	0.5-2.0	2.5 avg
Concentration, p.p.m., wt.	300-400	500

## DESCRIPTION OF EXPERIMENTS

### Apparatus

The cross flow/electrofiltration system is illustrated in Figure 3. The system is designed so that the following variables can be independently varied: electrical field strength, circulation rate or Reynolds number of the slurry, applied pressure drop, and feed concentration. The cross flow/electrofiltration cell is a modified electrofiltration cell manufactured by Canaco, Inc. This electrofiltration cell is based on the original designs of Bier. The cell utilizes a variety of polymer spaces and platinum mesh electrodes. Three 1/16 in. spacers were used to form the flow channel on the feed side of the membrane. The flow channel depth, width, and length were 0.48, 6.06, and 16.5 cm, respectively. The filter medium area was approximately 100 cm<sup>2</sup>. The large channel depth was chosen to assure that the Reynolds number or circulation rate could be varied without changing the pressure driving force; that is, the flow channel depth was selected to give negligible frictional pressure drop. The filter medium in all experiments was a Nuclepore polycarbonate filter having a pore size of 0.6 cm.

### Feed Systems

Two distinctly different feed systems were investigated. These included a chemically stabilized oil and water emulsion and an aqueous suspension of Kaolin clay. The properties of both these systems are summarized in Table 1. The conductivity of the suspensions ranged from 10 to 20  $\mu\text{mhos/cm}$  for various feed batches.

### Procedures

All operating procedures were designed so that variable changes could be made in such a manner that hysteresis could be avoided. The principal cause of any hysteresis effects is the formation of very small thickness filter cakes or oil layers next to the filtration medium. Consider the case when the voltage is less than critical. In this situation, the particles have a net migration velocity toward the filter medium. If the parameter under study were the Reynolds number, it is important during start-up to be sure that the Reynolds number is held constant or is higher than the steady state value to eliminate hysteresis effects, that is, insure that the cake resistance is never higher during start-up than at steady state. Since the primary objective of these experiments was to conduct parameter studies to reveal the different regimes of operation and mechanisms of transport, a new Nuclepore filter and fresh feed were used in each experiment.

The electrophoretic mobility measurements were made on all feed samples with a Zeta meter. The dispersed oil concentration was measured using light transmittance (Baush and Lomb Spectronic 20 to 520 nm). The Kaolin clay concentrations were determined by filtration on a 0.45  $\mu\text{m}$  millipore filtration and weighing.

Detailed descriptions of the experimental apparatus, procedures, and analytical methods have been summarized by Lawler (1976).

## RESULTS

Filtration rate data were obtained to determine the influences of electric field strength and the Reynolds number. These data are used in conjunction with the mathematical model to illustrate the effects of electroosmosis on the filter resistance and electrophoresis on the film resistance.

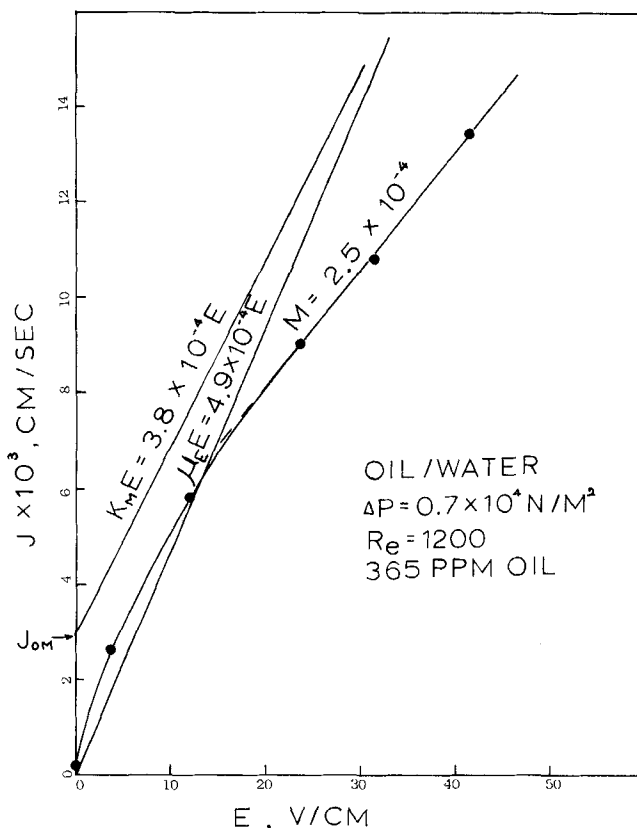


Fig. 4. The filtration-electric field strength dependence for the oil/water system.

### Filtration Rate—Electric Field Strength Dependence

The filtration rate increases monotonically with the field strength for both the oil/water and the clay/water systems. Data for the oil/water system are presented in Figure 4. The curve with the data points represents the filtration rate data. Note that the curve becomes essentially linear when

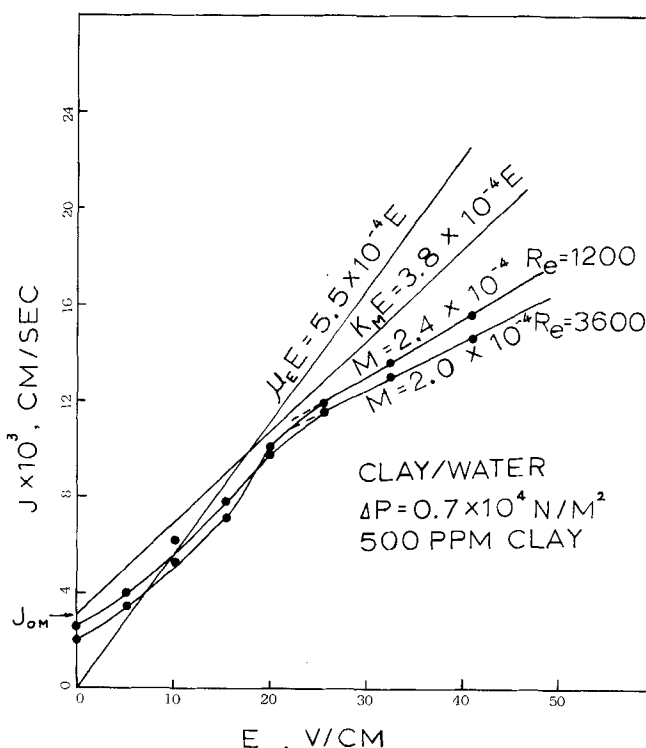


Fig. 5. The filtration-electric field strength dependence for the clay/water system.

TABLE 2. COMPARISON OF EXPERIMENTAL AND CALCULATED SLOPE OF THE FILTRATION RATE—FIELD STRENGTH DEPENDENCE AT HIGH VOLTAGE

System	Reynolds number	Slope, $M = \left( \frac{\mu_E K_M}{\mu_E + K_M} \right)$		
		Expt.	Calc'd.	% diff.
Oil/water	1 200	$2.5 \times 10^{-4}$	$2.2 \times 10^{-4}$	-12%
Clay/water	1 200	$2.4 \times 10^{-4}$	$2.3 \times 10^{-4}$	-4%
Clay/water	3 600	$2.0 \times 10^{-4}$	$2.3 \times 10^{-4}$	+15%

the voltage is above 20 V/cm. This implies that at a voltage above 20 V/cm, there is no oil layer existing next to the filtration medium that is, the system is operated in a regime above the critical voltage. Both electroosmosis in the filtration medium and electrophoresis in the liquid film are the major mechanisms of transport in this region. The slope of the line above critical voltage can be independently calculated from the known electrophoretic mobility  $\mu_E$  of the charged oil droplet and electroosmotic coefficient  $K_M$  of the filter medium from Equation (14).

Filtration rate-electric field strength data for the clay/water system are presented in Figure 5 for two Reynolds numbers. Both these curves become linear at a voltage approximately above 30 V/cm. Again, the linear slopes for two Reynolds numbers can be calculated by the known electrophoretic mobility  $\mu_E$  of the charged clay particle and electroosmotic coefficient  $K_M$  of the filter medium from Equation (14). These calculated and experimental slopes for both the oil/water and the clay/water systems are shown in Table 2. The results indicate that slopes calculated from Equation (14) agree reasonably with the experimental data for both systems.

The slight deviations of slopes at the two different Reynolds numbers for the clay/water system are believed to be due to the effect of fluid shear in the film resistance. In the case where the applied voltage is greater than the critical voltage, increasing fluid shear decreases the filtration rate.

Below the critical voltage, the shape of the filtration rate-field strength curves for the clay/water system are complicated. This is likely due to the occurrence of cake electroosmosis at the low field strengths. The curvature for clay/water systems (see Figure 5) is likely caused by an expansion in the cake with increasing field strengths. Note that the particles are migrating away from the filtration media that is, as the field strength is increased, the cake becomes less consolidated and finally reaches a point where a discrete cake does not exist.

#### Filtration Rate—Reynolds Number Dependence

The experimental filtration rate-Reynolds number dependence was obtained by holding other parameters such as field strength and pressure driving force constant. Data are presented in Figure 6 for the oil/water systems. In order to avoid the possibility of breakthrough of oil droplets below critical voltage, all of the data were obtained for this system above the critical voltage. Note that in all cases, the filtration rate decreases with increasing Reynolds number. This is consistent with the discussion of the three regimes of operation of a cross flow/electrofilter presented earlier. Above the critical voltage, the concentration of the oil droplets decreases as the filtration medium is approached. Consequently, any improvement in convection will lead to the transport of more oil droplets toward the filter medium.

Filtration rate-Reynolds number data are presented for the clay/water system in Figure 7. In the upper curve, the

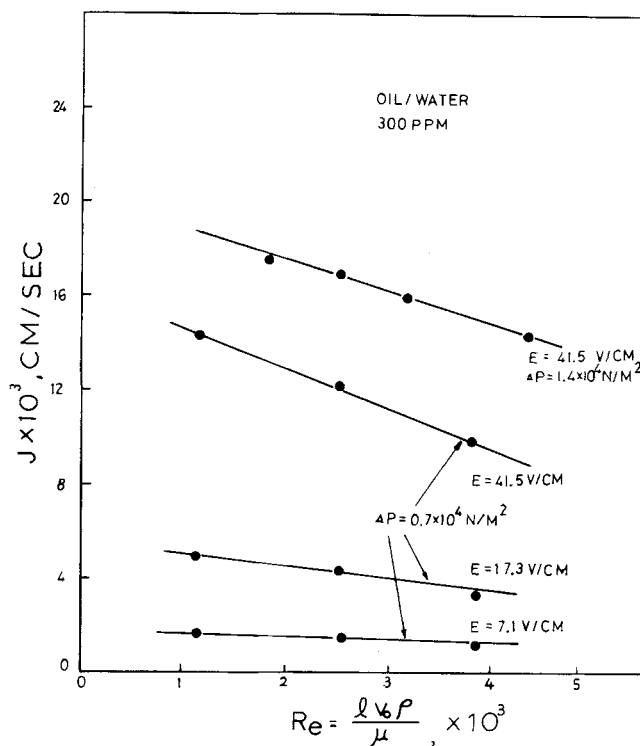


Fig. 6. The filtration rate-reynolds number dependence for the oil/water system.

voltage is above critical, and again the filtration rate decreases with increasing Reynolds numbers as predicted by the model. The voltage was below critical in the lower curve. The data points represent a sequence of experiments starting at the higher Reynolds number. Note that when the voltage was below critical value, the filtration increases with increasing Reynolds number. This again is due to improved convection in the liquid film and results because

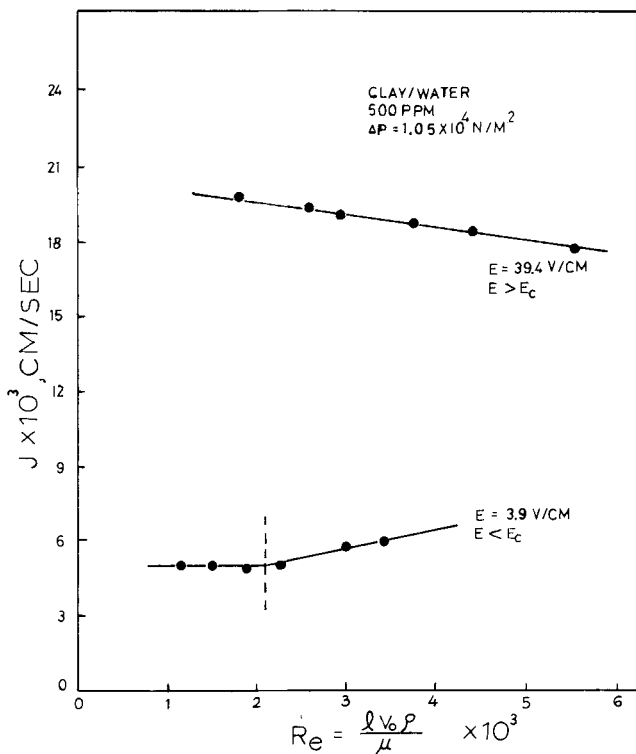


Fig. 7. The filtration rate-reynolds number dependence for the clay/water system.

the concentration of particles is highest next to the filter. Consequently, improved convection increases the rate of transport of particles away from the filter. The voltage is constant for the lower curve, but as the filtration rate decreases, a point is reached where the critical voltage is attained. After this, the filtration rate becomes independent of Reynolds number. This is consistent with the idea that at critical voltage there is no concentration profile in the liquid film, and consequently the film resistance is zero.

## DISCUSSION AND SUMMARY

The mathematical model was used to interpret cross flow/electrofiltration performance data to separate the effects of the electric field strength on the film and medium resistances. This is an essential starting point prior to comparing filter performance data with more detailed models for the transport characteristics in the liquid film, for example, convective mass transfer and radial migration effects.

The data obtained with both the oil/water system and the clay/water system indicate that both electroosmosis and electrophoresis control the performance of the filter above the critical field strength. However, this observation cannot be generalized to all operating conditions. The relative importance of contributions of the film resistance and medium electroosmosis above the critical voltage will depend not only on the conditions of operation such as Reynolds number, but also the feed concentration and the type of feed suspensions. The electroosmosis coefficient is a function of both ionic strength or specific conductivity and pH. Film resistance, as defined in Equation (4), may be controlled by the field strength, particle electrophoretic mobility, particle concentration polarization, and radial migration effects. As the particle size is increased, the radial migration of particles in the liquid film may become the major mechanism which enhances the mass transfer of particles away from the filter surface, and the particle diffusion may become less significant. In such a case, an increase in Reynolds number will always increase the filtration rate for all three regimes of operation.

The particle size in both the oil/water system and the clay/water system are so small that the radial migration of particles is negligible (see Table 1) compared with the particle concentration polarization. In general, radial migration velocity depends on the fourth power of the particle diameter/cell width ratio. The experimental data confirm that the influence of Reynolds number or fluid shear can either increase or decrease the filtration rate, depending on the regime of operation. This would be the case if radial migration were significant.

The filtration rate feed concentration dependence that is implied by the concentration polarization phenomenon in the liquid film suggests that multiple filtration units could be staged similarly to the methods that are used for either cross flow or ultrafiltration cells. Note, however, that the regime of operation must be considered; for example, at the critical voltage there would be no incentive to stage sequential units because in this regime there is no filtration rate-concentration dependence. The cross flow/electrofilter should find applications for suspensions which have a low background ionic strength or specific conductivity and/or systems which have shear sensitive particles.

The fact that electrodeposition can occur at the electrode away from the filtration medium was mentioned only in passing. In the case when the voltage is high enough so that there is a migration velocity of particles towards the electrode, a number of approaches such as reverses in polarity, protection of the electrode with a porous membrane or filter medium, and/or utilization of high shear can minimize electrodeposition.

Electrical heating effects have not been mentioned because they are negligible at the Reynolds numbers (axial velocities) employed in the experiments.

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## NOTATION

$C_B$	= concentration of oil or particles in the bulk fluid
$C_S$	= concentration of oil or particles at the surface of filter medium or filter cake if one exists
$E$	= electric field strength, V/cm
$E_C$	= critical electric field strength, V/cm
$J$	= filtration flux, cm/s ( $\text{cm}^3/\text{cm}^2/\text{s}$ )
$J_M$	= filtration flux defined in Equation (6), cm/s
$J_{OM}$	= filtration flux defined in Equation (7), cm/s
$k$	= mass transfer coefficient defined in Equation (3), cm/s
$K_C$	= electroosmotic coefficient in the filter cake, cm/s/V/cm
$K_M$	= electroosmotic coefficient in the filter medium, cm/s/V/cm
$l$	= length of the filter, cm
$M$	= parameter defined in Equation (13), cm/s/V/cm
$M_o$	= parameter defined in Equation (13), cm/s
$\Delta P$	= pressure driving force, N/M <sup>2</sup>
$R_C$	= filter cake resistance, N/M <sup>2</sup> /cm/s
$R_{OC}$	= filter cake resistance at zero electric field strength, N/M <sup>2</sup> /cm/s
$R_e$	= inlet Reynolds number, dimensionless
$R_f$	= liquid film resistance ( $\text{cm/s}$ ) <sup>-1</sup>
$R_M$	= filter medium resistance, N/M <sup>2</sup> /cm/s
$R_{OM}$	= filter medium resistance at zero electric field strength, N/M <sup>2</sup> /cm/s
$R_T$	= total resistance to filtration, N/M <sup>2</sup> /cm/s
$v_o$	= inlet velocity, cm/s
$\delta$	= boundary layer thickness, cm
$\mu$	= liquid viscosity, g/cm-s
$\mu_E$	= electrophoretic mobility, cm/s/V/cm
$\rho$	= liquid density, g/cm <sup>3</sup>

## LITERATURE CITED

- Beechold, H., "Ultrafiltration and Electro-Ultrafiltration," in *Colloid Chemistry*, Vol. I, J. Alexander, ed., The Chemical Catalog Company (1926).
- Bier, M., ed., *Electrophoresis*, Vol. I, p. 263, Academic Press, New York (1959).
- , "Electrokinetic Membrane Processes," in *Membrane Processes in Industry and Biomedicine*, Plenum Press, New York (1971).
- Cooper, F. C., Q. M. Mees, and M. Bier, "Water Purification by Forced Flow Electrophoresis," *J. San. Eng. Div., A.S.C.E.*, **91**, 13 (1965).
- Dahlheimer, J. A., D. G. Thomas, and K. A. Kraus, "Application of Woven Fiber Hoses to Hyperfiltration of Salts and Cross Flow Filtration of Suspended Solids," *Ind. Eng. Chem. Process Design Develop.*, **9**, No. 4, 566 (1970).
- Giddings, C. J., "A New Separation Concept Based on a Coupling of Concentration and Flow Nonuniformities," *Separation Science*, **1**, 123-125 (1966).
- , F. J. Yang, and M. N. Myers, "Theoretical and Experimental Characterization of Flow Field Flow Fractionation," *Analytical Chemistry*, **48**, No. 8, 1126-1132 (1976).
- Grushka, E., K. D. Caldwell, M. N. Myers, and J. C. Giddings, "Field Flow Fractionation," *Separation and Purification Methods*, **2**, No. 1, 127-151 (1973).
- Henry, J. D., "Cross Flow Filtration," in *Recent Developments in Separation Science*, Vol. II, pp. 205-225, N. N. Li, ed.,



the Chemical Rubber Company, Ohio (1972).  
 —, and R. C. Allred, "Concentration of Bacterial Cells by Cross Flow Filtration," *Dev. Ind. Microbiol.*, 13 (1972).  
 Lawler, L. F., M.S. Thesis, in progress, W. Va. Univ., Morgantown (1976).  
 Manegold, E., "The Effectiveness of Filtration, Dialysis, Electrolysis, and their Intercombinations as Purification Processes," *Trans. Faraday Soc.*, 33, (1937).  
 Moulik, S. P., F. C. Cooper, and M. Bier, "Forced Flow Electrophoretic Filtration of Clay Suspensions," *J. Col. Int. Sci.*, 24, 427 (1967).

Porter, M. C., "Ultrafiltration of Colloidal Suspensions," *AIChE Symposium Ser.*, 68, 120 (1972).  
 Reis, J. F. G., and E. N. Lightfoot, "Electropolarization Chromatography," *AIChE J.*, 22, No. 4, 770-785 (1976).  
 Tiller, F. M., and H. Cooper, "The Role of Porosity in Filtration, Part V: Porosity Variation in Filter Cakes," *ibid.*, 8, 445-449 (1962).

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# The Relationship Between One-Dimensional and Two-Dimensional Separation Processes

One-dimensional, time dependent separations are analogous to two-dimensional, steady state separations, and the equations for the former can be transformed to the latter by the transform  $t \rightarrow \theta/w$ . This analogy is used to develop and provide the solutions for the two-dimensional analogues of several variations of chromatography, parametric pumping, and pressure swing adsorption.

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## SCOPE

Many different separation techniques have been developed for the separation of complex mixtures using adsorbents and various chromatographic packings. For preparative separations, a variety of operating techniques including straightforward scale-up of elution development, cyclic operation, and two-dimensional operation (reviewed by Conder, 1973; Wankat, 1974; Sussman, 1976, respectively) have been developed. These methods are all attempts to increase throughput and/or continuously separate multicomponent mixtures. The prototype of the two-dimensional chromatograph is a packed annulus which is rotated past a fixed feed point while solvent or carrier gas is continuously fed to the entire annulus. The schematic of this rotating cylinder system is shown in Figure 1. The solutes move up the annulus in helical flow paths depending upon the rotational velocity, the carrier velocity, and the amount they are adsorbed. Since the steady state separation occurs in the  $z$  and  $\theta$  directions, the process is commonly called a two-dimensional separation. This two-dimensional flow path contrasts with usual chromatographic and adsorption systems, where separation occurs in only the  $z$  direction, but is also time dependent.

The purpose of this study was to formalize the relationship between time dependent, one dimensional separa-

tion processes such as elution development in chromatography or cyclic operation of adsorption columns and the two-dimensional separation techniques. Then this formalized analogy will be used to compare existing processes, and, finally, the analogy is used in a systematic study to develop new two-dimensional separation techniques. This work thus serves as a bridge to connect three intensively studied separation techniques and helps to explain the somewhat unfamiliar two-dimensional techniques. In addition, a powerful tool for looking for new separation techniques is developed.

The relationship between time dependent, one-dimensional and steady state, two-dimensional processes has been discussed briefly in the literature. Martin (1949) used the analogy between these processes to suggest the first two-dimensional chromatograph. This relationship between the rotating annulus device and usual elution development has been occasionally mentioned in the literature since then. Wankat (1972) extended this analogy to staged systems when he noted the relationship between countercurrent distribution and two-dimensional stage systems. This analogy was further extended to cyclic systems when Wankat et al. (1976) noted the analogy between cycling zone adsorption and continuously regenerated two-dimensional systems.

## CONCLUSIONS AND SIGNIFICANCE

Comparison of the material and energy balances, equilibrium relation, and mass transfer expression for the time dependent, one-dimensional and the steady state, two-dimensional separations in the geometry of Figure 1 shows that the equations and appropriate boundary conditions for the one-dimensional system can be transformed into the equations and boundary conditions for the two-dimensional system if the transform  $t \rightarrow \theta/w$  is

made where  $t$  is time,  $\theta$  is the angular coordinate,  $\theta/w$  is the angular velocity, and diffusion and dispersion can be neglected. Thus, under condition of negligible diffusion and dispersion, the two techniques are mathematically similar. The separations obtained will thus be similar. The analogy is applied to the limiting periodic state of cyclic operations and the steady state of the two-dimensional apparatus. This analogy thus makes it possible to apply