Separation of Fine Particle Dispersions Using Periodic Flows in a Spinning Coiled Tube

A centrifugal method for fractionating fine (<10 μm) particles by size and/or density is described. Periodic, bidirectional flow through a continuously spinning coiled tube splits fine particle suspensions into two fractions. Proof-of-principle experiments with zeolite powder have demonstrated successful concentration of aqueous dispersions.

J. S. Papanu, R. J. Adler, M. B. Gorensek, and M. M. Menon
Chemical Engineering Department
Case Institute of Technology
Case Western Reserve University
Cleveland, OH 44106

SCOPE

A new method for separating fine (<10 μm) particles in suspension is being developed. It can concentrate or fractionate according to size and/or density. Separation occurs in a spinning helical tube through which periodic, bidirectional fluid displacements are made. The mechanism involves repeated sedimentation and resuspension of particles, resulting in a net movement of all of the particles (concentration), or only a select portion (fractionation). Sedimentation results from the spinning motion, while resuspension is effected by secondary flow, which occurs only during axial flow and is due to the interaction of the helical geometry, steady rotation, and fluid motion.

This method is similar in some respects to parametric pumping (PP) (Wilhelm et al., 1966) and sedimentation field-flow fractionation (SFFF) (Giddings et al., 1974). It employs cyclic flow as does PP. However, PP separates molecules in solution, not particles in suspension, and uses a periodically varying intensive parameter such as temperature to effect separation instead of a steady centrifugal force field. SFFF also uses a steady centrifugal force field, but with a unidirectional rather than a periodic bidirectional flow. This gives a chromatographic-like separation, unlike the new method, which yields two streams of different composition.

Concentration experiments have been performed with dilute aqueous dispersions of ground (0.2-3.0 μm) molecular sieve powder. Although continuous operation is feasible, experiments were limited to batch operation. The major objective was to demonstrate that the method can concentrate suspensions. Some data on the effect of process parameters were also obtained.

CONCLUSIONS AND SIGNIFICANCE

Proof-of-principle experiments have demonstrated that this new separation method can concentrate suspensions of fine particles. Aqueous dispersions of molecular sieve powder were split into dilute and concentrated portions. Separation factors up to about 10 were obtained. Infinite separation factors are theoretically attainable in the absence of axial dispersion, but experiments were terminated short of steady state and the system was not optimized.

Fine particle separations are important in many large-scale industrial processes such as ore beneficiation (Somasundaran and Arbiter, 1979) and liquid synfuels production (Parekh and Goldberger, 1980). They are also crucial to the smaller scale but no less signifi-

The current address of J. S. Papanu is Chemical Engineering Department, University of California, Berkeley, CA 94720.
The current address of M. B. Gorensek is Halcon Research, One Phillips Parkway, Montvale, NJ 07645.

cant production of pigments, phosphors, metal oxides, and biochemicals. The new method may prove useful in these areas, especially where sharp separations are desired.

Introduction

Fine particles, defined here as having effective diameters less than 10 μm, must be separated from slurries or dispersions in many industries. One of the most important examples is minerals processing, in which finely ground ores are slurried to liberate the value from the gangue. Conventional solid-liquid separation techniques are often uneconomical when used to recover valuables from ore fines. This is especially true for slurries of particles smaller than 10 μm in size. In the phosphate industry, up to one-third of the phosphate is discarded as unrecovered slime, while an estimated one-fifth of the world's tin is lost in a similar manner (Fuersteneau et al., 1979). As reserves become depleted, lower quality ores will have to be utilized. A larger fraction of the valuables in these ores will be in the fine particle size range. Hence, processing with current technology may become uneconomical. Technological advances in this area clearly are needed.

A new way to separate fine particles that may overcome the limitations of present technology is being developed. This new method can separate or classify particles by size and/or density. It is inherently a multistage process, and therefore may be able to perform difficult and previously unattainable separations. The new method produces concentrated fractions, unlike elution methods, and promises to be effective in the micron to submicron range.

This paper describes the basic concepts of the new method and reports the results of proof-of-principle experiments.

Concepts and Principles of Operation

The distinguishing feature of the new method is a coiled tube spinning rapidly and continuously about its helical axis. Figure 1 depicts a batch version. The steadily rotating coil is connected at both ends to stationary ports by means of a spinning seal assembly. Each port leads to a reservoir, allowing flow to pass through the coil in either direction, from one reservoir to the other. At start-up, the system is filled with the fine particle suspension. Then, a periodic sequence of back-and-forth flows and rest periods is initiated. Particles in the spinning coil are periodically immobilized by sedimentation or resuspended by the flow. The result is a net movement of dilute liquid (or of one fraction) toward reservoir *I* and concentrated suspension (or the other fraction), toward reservoir *II*. After many cycles have transpired, reservoir *I* contains dilute, essentially particle-free fluid (or one particle fraction), while reservoir *II* holds the concentrated suspension (or other particle fraction). The number of cycles needed to perform a separation is determined by the cycle efficiency and by the volume of fluid pumped per cycle relative to the reservoir volume.

That particles are sedimented by centrifugal force is obvious. Particle resuspension by secondary flows, however, is more subtle. Flow through the coiled tube results in an imbalance between the centrifugal forces acting on fluid elements in different parts of the tube cross section. This provides the driving force for eddies or secondary flows, which scour particles from the tube wall. Figure 2 shows typical streamlines for these currents; the secondary flow phenomenon is discussed below.

Although this paper is primarily concerned with a batch system, both semicontinuous and continuous operation are possible. Figure 3 shows one way in which this can be achieved. Two coils with spinning seals are used, and the feed enters between them. An analogy to distillation can be drawn. The two coils are, effectively, two sections of a larger coil, much like the stripping and rectifying sections of a distillation column. The feed is superimposed on the back-and-forth pumping motion, which transports products and provides reflux, and product is drawn off continuously at both ends.

Whether batch, semicontinuous, or continuous, there are two basic modes of operation: concentration and fractionation. The fundamental principles involved in the sequence of flows needed to achieve separation in each mode are considered next.

Concentration

The concentration of suspensions is conceptually simpler than fractionation. Therefore, it is described first.

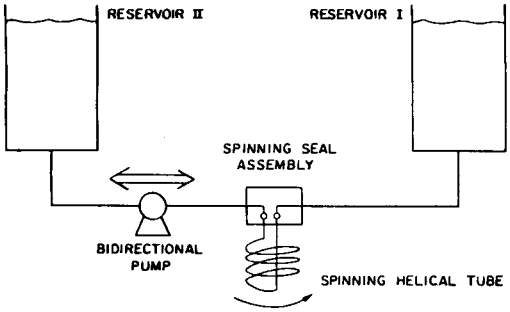

Figure 1. Batch system.

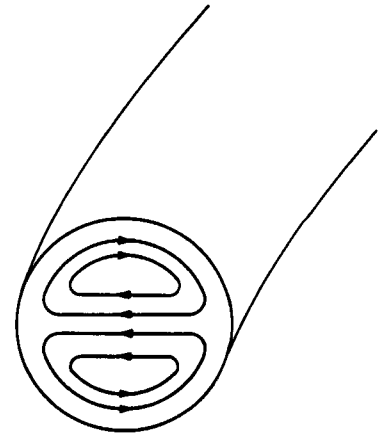

Figure 2. Secondary flows in a stationary helix.

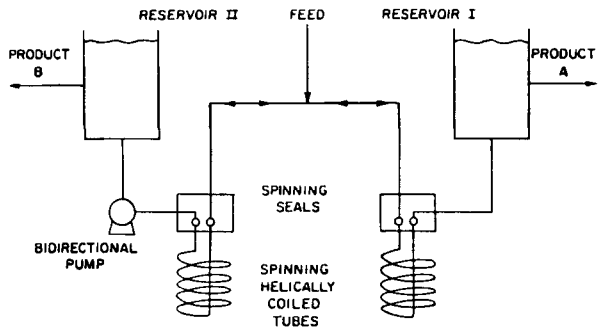

Figure 3. Configuration for continuous operation.

The steps comprising an ideal batch concentration are shown in Figure 4. Initially, the composition throughout the system is uniform. For simplicity, the coil volume is assumed equal to the volume of fluid pumped in either direction in a single pumping stroke, and the volume of the connecting lines is taken to be negligible. Thus, the entire coil contents are displaced during a pumping step, with the displaced liquid flowing directly into one reservoir, and the displacing liquid coming from the other.

Consider the fluid in the coil. The first step involves no flow. All of the particles move to the outer wall of the tube, provided the specific gravity of the solids is greater than that of the liquid. In the second step, fluid from reservoir *II* displaces the fluid in the coil, moving it into reservoir *I*. Since all of the particles have settled out of the fluid leaving the coil, reservoir *I* is diluted. The flow velocity is such that the induced secondary flows just begin to resuspend the sediment in the coil, mixing it with the fluid entering from reservoir *II*. Thus, at the end of the second step the suspension in the coil is more concentrated than that in reservoir *II*. In step three, the suspension is pumped back toward reservoir *II* (before any significant sedimentation occurs), increasing the concentration in that reservoir. At the conclusion of this step, the coil contains the same diluted suspension as reservoir *I*. The three-part sequence is then repeated. Overall, particles are transported from reservoir *I* to reservoir *II*, thereby diluting the former and concentrating the latter. Since equal volumes are pumped in each direction during a cycle, the net volume change in each reservoir is zero.

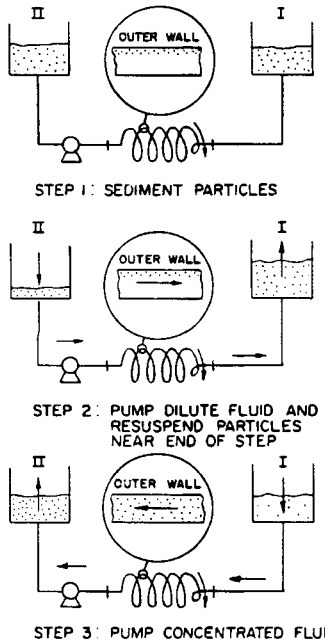

Figure 4. Concentration sequence.

The resuspension of settled particles may begin at any point during step two, from early on to not at all. The exact time depends on the strength of the secondary flows. Resuspension occurring too early would be undesirable, since particles then would be swept into reservoir *I*. The above sequence was termed "ideal" because of the assumption that complete resuspension occurs at the end of the second step. A slight modification of the sequence would be to use asymmetric pumping rates. The pumping rate in step two would be slow, insuring that few, if any, particles would be resuspended. Pumping in the reverse direction during the third step would be rapid, giving rise to strong secondary flows that would quickly resuspend the sediment. In terms of effect, there is no difference between complete resuspension occurring at the end of step two or at the beginning of step three.

Several assumptions other than those already mentioned have been implied. These include plug flow through the coil with negligible axial dispersion. Complete and instantaneous mixing between the fluid already in a reservoir and any fluid entering is also assumed. Finally, any settling that might occur in the coil during a flow step is taken to be negligible.

The simplifying assumptions made thus far are not essential for separation. In general, a separation may be slower or ultimately less sharp if any of the assumptions is relaxed. (One exception is partial displacement of fluid in the coil, which favors slower but sharper separations.)

Fractionation

The new method can also be used to fractionate a mixture of particles according to size and/or density. To obtain a separation, the particles must respond differently to the centrifugal field or to secondary flow, allowing them to be sedimented or resuspended to different degrees.

For most particle dispersions of interest, the Reynolds number for sedimentation is small enough so that the Stokes law,

$$v_s = \frac{2a^2(\rho_2 - \rho_1)G}{9\mu}, \tag{1}$$

applies. Thus the settling velocity, v_s, of an individual (spherical) particle is proportional to the square of the particle radius, a, as well as the density difference between the particle and the fluid, $\Delta\rho$. This can be exploited to separate particle mixtures according to size and density.

A five-step sequence based on this principle is illustrated in Figure 5. It presumes two particle species, A and B, with A having a larger value of $a^2\Delta\rho$ than B. Simply put, A sediments faster than B. In the first step, the fluid in the coil is allowed to sediment for a time long enough that A drops out of suspension completely, but short enough that a substantial portion of B remains suspended. Then, the coil contents are slowly displaced toward the right at a flow rate such that the sediment is undisturbed. The only particles moved are those of species B. This is followed by a complete sedimentation step in which all of the suspended particles drop out. The fourth step is a displacement to the left of a fraction of the volume of the coil at a flow rate slow enough

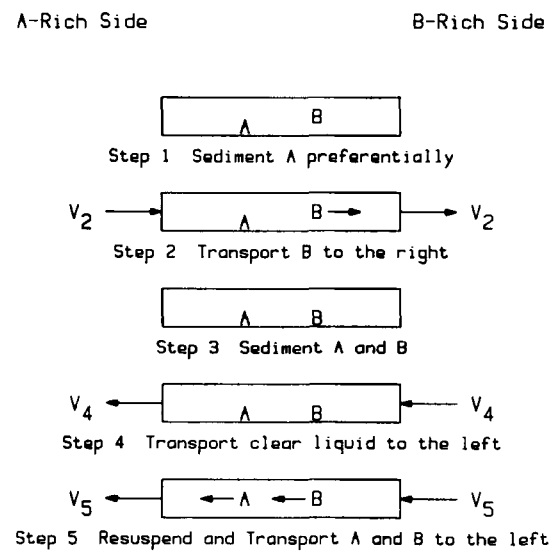

Figure 5. A fractionation sequence.
A sediments faster than *B*, $V_2 = V_4 + V_5$.

that the sediment stays undisturbed. No particles are moved. Finally, step five is rapid displacement to the left of the remainder of the coil volume at a flow rate large enough to insure complet resuspension of the sediment, restoring the reservoirs to their original levels. The net effect is to move *A* to the left (step 5) and *B* to the right (step 2 minus step 5), splitting the mixture into *A*-rich and *B*-rich portions.

Many other fractionation schemes are also possible. In addition to differences in sedimentation rates, steric effects can be used as a basis for separating particles during resuspension (Giddings and Myers, 1978).

Related separation methods

The new separation method has some features in common with two existing methods, parametric pumping and field-flow fractionation.

Parametric pumping (PP) was conceived by Wilhelm and coworkers (1966). It combines periodic, bidirectional fluid displacements in a column with coordinated periodic changes in an intensive physical parameter such as temperature. The PP principle has been applied commercially to gas separations in the pressure swing adsorption process. Periodic, alternating flow is the common feature of the new method and PP. One major difference is that PP is geared toward separating molecules in solution and not toward fractionating fine particle suspensions. Another difference is in mechanism. The oscillation of an intensive physical parameter in PP results in adsorption and desorption, which are the driving forces for the separation. The new method, however, employs sedimentation and resuspension, which are effected by a constant centrifugal field coupled with periodic secondary flows. Finally, since the mass transfer steps in PP are diffusion-limited, cycle times tend to be longer than in the new method (with the exception of pressure swing adsorption).

Field-flow fractionation (FFF), an elution technique for fractionating mixtures of fine particles or macromolecules, was conceived by Giddings (1966) at about the same time that Wilhelm developed PP. It employs an external field acting across a long, narrow channel through which the carrier fluid passes in laminar flow. A wide range of fields can be employed, leading to several subclasses of FFF (Giddings et al., 1981). The one most closely related to the new method is sedimentation field-flow fractionation (SFFF) (Giddings et al., 1974), which uses a centrifugal field. (A similar technique was independently developed by Berg and Purcell, 1967.) SFFF differs from the new method both in concept and application. It uses steady, unidirectional flow rather than periodic, alternating flow. Secondary flows are detrimental to SFFF and are suppressed by using channels of rectangular cross section with large width-to-height ratios. Finally, because it is a chromatographic-like technique which dilutes the separated species with carrier fluid, SFFF is more appropriate for analytical purposes, such as determining particle size distributions, than for large-scale processing of dispersions or slurries.

Secondary flow

Secondary flows are essential to the new separation method. Their existence has been recognized for over a century (Thompson, 1876), and a sizable literature has developed over the years.

Most investigators have considered secondary flows associated with steady flow through curved pipes or stationary coils (Eustice, 1910; Dean, 1928; White, 1929; Larrain and Bonilla, 1970; van Dyke, 1978; Manlapaz and Churchill, 1980); a comprehensive review of the subject has been made by Berger et al. (1983). These studies have revealed that secondary flows have a number of important features. Their strength can be appreciable, approaching that of the axial flow. They stabilize the laminar flow regime, with transition Reynolds numbers in coiled tubes being several times larger than in straight tubes (Taylor, 1929). Heat and mass transfer rates at the wall are augmented by secondary flows because they exchange fluid near the wall with fluid from the interior (Weissman and Mockros, 1968; Tarbell and Samuels, 1973). Also, axial dispersion is decreased in both the laminar and turbulent flow regimes due to the cross-sectional mixing (Koutsky and Adler, 1964).

Secondary flows in a coil oscillating about its helical axis with imposed axial flows have also been studied. Joseph and Adler (1975) obtained numerical solutions of the velocity field for a coiled tube of square cross section and found that the oscillatory motion induces strong secondary flows that have a periodic component. This work followed reports by Drinker et al. (1969) and Bartlett et al. (196), who developed an oscillating coil blood oxygenator. Using coiled tubes made from oxygen-permeable membranes, they obtained high oxygen transfer rates, confirming that the secondary flows enhance mass transfer at the wall.

No mention has been found in the literature for the case most relevant to this work: a steadily spinning coil with imposed axial flow. Some analytical work was done by Menon (1984). The Navier-Stokes equation for this case has been solved numerically by incorporating steady rotation into the approach used by van Dyke (1975, 1978). Secondary flow velocities were found to be up to an order of magnitude greater than the associated axial flow velocity. This is encouraging for the resuspension of fine particles.

A qualitative consideration of secondary flows in a spinning coil shows that unlike those in the stationary case, they depend on the direction of the axial flow relative to that of the coil rotation, as illustrated in Figure 6. They can also be much stronger.

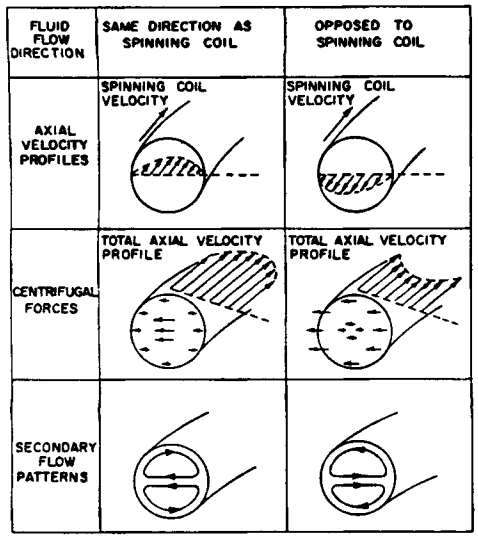

Figure 6. Secondary flows in a spinning helical tube.

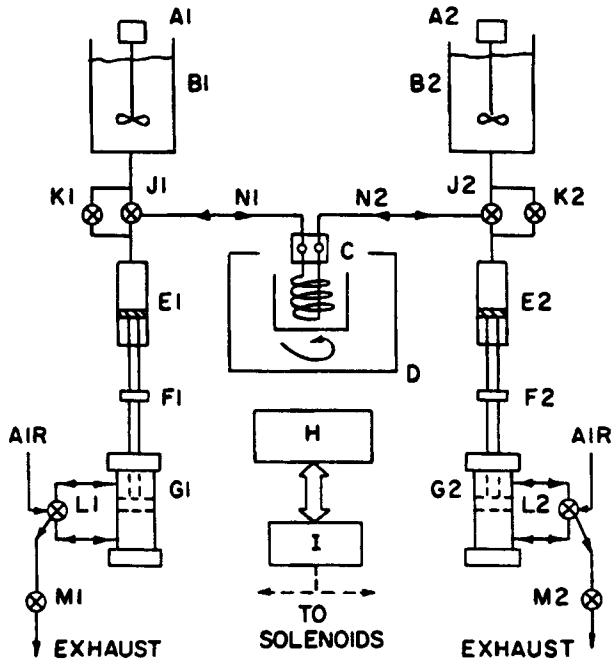

Figure 7. Experimental apparatus.

A1, A2 = Agitators
B1, B2 = Reservoirs
C = Spinning seal, coil, bowl
D = Centrifuge
E1, E2 = 20 mL syringes
F1, F2 = Couplings
G1, G2 = Compressed air actuators
H = Microprocessor
I = Interface/driver circuit
J1, J2 = 3-way solenoid valves
K1, K2 = Bypass solenoid valves
L1, L2 = Air line solenoid valves
M1, M2 = Manual metering valves
N1, N2 = Interconnecting lines, 6 mL each

If the axial flow and rotation are in the same direction, Figure 6, their speeds are additive. The resulting velocity profile is essentially parabolic, causing the greatest centrifugal forces to act on fluid elements near the tube center. This sets up a recirculating secondary flow similar to that which would occur in a stationary coil (see Figure 2), except that its magnitude is considerably greater. Let U be the tangential velocity of the tube wall, and let u be the maximum axial velocity of the fluid relative to the tube. The driving force for the recirculating flow is the difference between the centrifugal forces per unit mass acting on the fluid that has a total velocity $U + u$ (located somewhere in the interior) and on the fluid that has a total velocity U (located adjacent to the wall)

$$F \alpha \frac{(U+u)^2}{R} - \frac{U^2}{R} = \frac{2Uu + u^2}{R}.$$

This difference is proportional to $2Uu/R$, provided $U \gg u$. Thus, rapid rotation can cause strong secondary flows.

If axial flow opposes rotation, their speeds subtract. For the typical case of rotational velocity exceeding the maximum axial flow velocity, Figure 6, fluid elements near the wall have the highest net velocities and therefore experience the greatest centrifugal forces. They are forced outward, and the direction of circulation is reversed. Following similar reasoning, the driving force in this case is

$$F \alpha \frac{U^2}{R} - \frac{(U-u)^2}{R} = \frac{2Uu - u^2}{R},$$

which, as before, simplifies to $2Uu/R$, provided $U \gg u$. Again, rapid rotation causes strong secondary flows.

If the maximum flow velocity exceeds the spinning velocity, there is an annular region of fluid with zero angular speed. This fluid experiences no centrifugal force, and the resulting secondary flow is more complicated.

Experimental

Apparatus

A prototype of the new method was built for proof-of-principle experiments. The apparatus is illustrated in Figure 7. It features a centrifuge-driven rotating coil with a spinning seal assembly, syringe pumps, agitated reservoirs, and microprocessor-operated solenoid flow control valves.

The centrifuge (IEC model UV with a modified speed controller) can operate at speeds up to 4,800 ± 10 rpm. Several windings of Teflon semirigid tubing (4.4 mm ID, 6.6 mm OD) comprise the coil, which sits in a 20.4 cm ID stainless steel bowl mounted on the centrifuge rotor. Both ends of the plastic tube are connected to a spinning seal assembly, allowing flow connections to remain fixed as the coil rotates.

A pair of 20 mL disposable plastic syringes with compressed air-actuated plungers provides pumping. Air flow into each actuator is controlled by a three-way solenoid valve. Shaft collars limiting piston travel in the actuators are used to adjust the volume of fluid pumped per stroke, while travel rates, and hence pumping rates, are governed by air pressure and by throttling the exit gas through manual metering valves.

Two 500 mL burettes (4.5 cm ID) equipped with lab mixers to prevent settling are used as reservoirs. All interconnecting

lines are of polyethylene tubing (3.2 mm ID, 6.4 mm OD) with a total volume of 12 mL.

Liquid flow is directed with one two-way and two three-way solenoid valves. The three-way valves permit flow between either a reservoir and its associated syringe pump, or the coil and the syringe. Using these two valves alone, fluid cannot be pumped directly from one syringe to the opposite reservoir—it must be pumped into the opposite syringe first. The two-way solenoid provides a bypass for direct pumping into the opposite reservoir, which facilitates asymmetric pumping rates. A 6502-based microcomputer (Synertek SYM-1 with 4K of memory) is used to control all of the solenoids.

Methods and procedures

Concentration experiments were made with aqueous dispersions of Linde type 4A molecular sieve powder, a sodium aluminosilicate zeolite. This material has a dry density of 1.57 g/cm^3 (including pore volume), and a wet (water) density of 2.03 g/cm^3. The zeolite powder was ground with a mortar and pestle. About 50 g of this material was dispersed in 1 L of water, and the suspension was allowed to settle for 1 min to allow the largest particles to drop out. The supernatant was then decanted and found by gravimetric analysis to have a concentration of 0.052 g/mL. For experiments, aliquots of this stock suspension were diluted to about 4×10^{-4} g/mL. The approximate particle size range was determined with a Coulter model N4 laser light-scattering instrument. Those particles that remained in suspension after the dispersion was allowed to settle overnight had a mean size of 0.2 μm, with a standard deviation of 0.01 μm, while the settled particles had a mean size of 2.0 μ, with a standard deviation of 0.4 μm.

To prepare for an experiment, a fresh dispersion was poured into one reservoir and pumped toward the other, displacing any air in the lines. This was followed by rapid back-and-forth pumping for 50 cycles to assure homogeneity in case residue from previous experiments had remained in the lines or coil. In later runs, the first 80 mL pumped through the system during filling were discarded as an extra precaution. An initial sample was taken from each reservoir, and the contents of both were equalized before starting the centrifuge. During the separation, equal reservoir volumes were maintained by adjusting the shaft collars as needed.

The steps involved in each cycle of a typical concentration run with asymmetric pumping rates are shown in Table 1. This sequence is not optimal, but is intended as a working example.

Samples (4–5 mL) were taken intermittently to monitor concentration changes in each reservoir, and were not replaced. Although concentrations were not adjusted for volume reduction caused by sampling, this does not affect the results appreciably.

Analysis of samples was by measurement of optical transmission at 600 nm with a Cary model 219 spectrophotometer. To improve reproducibility, three trials with three readings per trial (at 0, 15, and 30 s after the instrument had stabilized) were performed for each sample, and an average percent transmission calculated. This was compared with a calibration curve, prepared for the ground powder to determine solids concentration.

Results and Discussion

Proof-of-principle experiments have demonstrated that the method is effective for concentrating suspensions. Figure 8 shows the result of a successful run. The initial concentration was 4.12×10^{-4} g/mL. A 62 s centrifugation time at 500 rpm was used. The final average concentration in the coil was midway between the final reservoir concentrations, and was 10% less than the initial concentration. Thus, particles did not accumulate in the coil. A final material balance including solids removed by sampling was able to account for 99.6% of the solids originally charged. The rate and degree of separation in this run, however, should not be interpreted as representing optimal operation.

A difficulty encountered in about one-third of the experiments was accumulation of particles in the coil. Particles were readily settled using adequate field strength or centrifugation time. However, secondary flows were not always strong enough

Table 1. Individual Operations, Run No. 22*

Step	Individual Operations, Fig. 7	Time, s
Step 1, Sediment	(a) No flow	58.0
	(b) Fill syringe E1 from reservoir B1	4.0
Step 2, Pump toward dilute reservoir	(c) Set valve J1 to connect E1 with coil; open valve K2 to allow direct flow into reservoir B2; set valve J2 to connect syringe E2 with the coil (E2 is empty and its plunger is pushed in)	0.1
	(d) Push fluid in E1 into coil, displacing fluid into B2*	30.0*
Step 3, Pump toward concentrated reservoir	(c) Close K2; set J2 to connect B2 with E2	0.1
	(f) Fill E2 with fluid from B2	4.0
	(g) Set J2 to connect E2 with coil; set J1 to connect E1 with the coil	0.1
	(h) Pump fluid in E2 into the coil, displacing fluid into E1*	1.0*
	(i) Set J1 to connect E1 with B1	0.1
	(j) Pump fluid in E1 into B1*†	0.1*†
	Total Sequence Time*	97.5*

*Times for operations (d), (h) and (j) and total sequence time vary from run to run as per Table 2.
†(j) requires the same time as (d); all but 0.1 s is in parallel with (a).

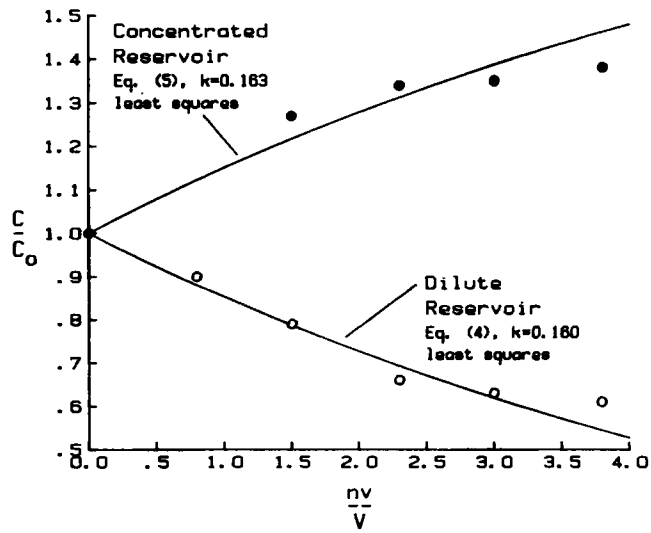

Figure 8. Proof-of-principle experiment, run 32.

to resuspend them. As a result, both reservoirs were sometimes diluted and particles accumulated in the coil.

In the sections that follow, the transient behavior during concentration trials, the effect of the major operating parameters, the importance of the particle size distribution, and colloidal effects are discused. The experimental conditions for all of the concentration trials cited are summarized in Table 2.

Transient behavior

The transient behavior of the reservoir concentration can be estimated. Simplifying assumptions include well-mixed reservoirs, negligible holdup in the lines connecting the reservoirs to the coil, negligible axial dispersion, and complete resuspension by the beginning of the second pumping stroke in each cycle.

Let $C_{n,d}$ be the concentration in the dilute reservoir at the end of the nth cycle, and let k be the fraction of particles in any coil segment that sediment during the first step in each cycle (assumed constant throughout the coil for the entire run). A material balance around the dilute reservoir yields

$$C_{n,d} = C_o\left(1 - \frac{kv}{V + v}\right)^n. \tag{2}$$

The behavior is exponential, and $C_{n,d} \rightarrow 0$ as $n \rightarrow \infty$ since $kv/(V + v) \ll 1$. If the amount of particles held within the coil is negligible compared to that contained in the reservoirs, the concentration in the concentrated reservoir after n cycles, $C_{n,c}$, must be

$$C_{n,c} = C_o\left[2 - \left(1 - \frac{kv}{V + v}\right)^n\right] \tag{3}$$

with $C_{n,c} \rightarrow 2C_o$ as $n \rightarrow \infty$.

A least-squares fit of the data in Figure 8 to Eqs. 2 and 3 yields $k = 0.160$ and 0.163 for the dilute and concentrated reservoirs, respectively. Given the spinning speed and centrifugation time used, these values are low. This is probably due to the nonideality of the system, especially with respect to axial dispersion and the volume of liquid in the interconnecting lines.

The term $kv/(V + v)$ is typically less than 0.1, with $v \ll V$. Thus, to a good approximation, Eqs. 2 and 3 may be rewritten

$$\frac{C_{n,d}}{C_o} = e^{-nkv/V} \tag{4}$$

$$\frac{C_{n,c}}{C_o} = 2 - e^{-nkv/V}. \tag{5}$$

For a given experiment, k has been assumed a constant. Therefore, the concentration change is a function of the number of reservoir volumes that have been displaced in each direction after n cycles, nv/V. This suggests that the dimensionless concentration C/Co be plotted against the dimensionless volume nv/V to provide a more meaningful basis for comparing data from different runs.

Process parameters

The main objective of the experimental work was to demonstrate that a suspension could be split into a dilute portion and a concentrated portion using the new method. Much of the work consisted of exploring combinations of operating parameters suggested by simple models. In general, a systematic study of the process parameters was not performed, and optimization was not attempted. Some comparative data, however, were obtained and are reported here.

The process parameters believed to be most important are centrifugation time, spinning velocity, rate and duration of pumping, direction of pumping relative to direction of spinning, and coil geometry. These are discussed below, although data showing their individual effects is limited in some cases. It should be emphasized that the effects of these variables are strongly interrelated, so that the separation depends on their net effect.

Centrifugation Time. According to the transient analysis presented above, the rate of separation depends on the fraction of

Table 2. Experimental Conditions

Run No.	Coil L/D[1]	Rotation Rate rpm	Field Strength g's	Init. Conc. g/mL × 10^4	Particle System[2]	Vol. Each Reservoir, mL	Pumping Rates, mL/s[3]		Duration of Operations, s[4]		Total Sequence Time, s[4]
							Step 2	Step 3	(d) and (j)	(h)	
22	355	2,000	442	4.02	Fresh dispersion	347	0.67	20	30.0	1.0	97.5
23	355	2,000	442	3.70	Fresh dispersion	347	0.62	40	32.3	0.5	99.3
25	355	2,000	442	3.93	Fresh dispersion	347	0.62	67	32.3	0.3	99.1
26	355	2,000	442	2.27	Run 25 reservoir contents	347	0.60	67	33.3	0.3	100.1
27	355	2,000	442	1.60	Run 26 reservoir contents	312	0.61	67	32.8	0.3	99.6
28	355	2,000	442	0.93	Run 27 reservoir contents	372	0.62	67	32.3	0.3	99.1
29	355	2,000	442	4.10	Fresh dispersion	312	0.62	50	32.3	0.4	99.2
32	355	500	27.6	4.12	Fresh dispersion	397	0.57	40	35.1	0.5	102.1
33	800	500	27.6	4.17	Fresh dispersion	387	0.57	40	35.1	0.5	102.1
34	800	500	27.6	4.07	Fresh dispersion	377	0.57	40	35.1	0.5	102.1

1 Coil is Teflon tube 4.4 mm ID, 6.6 mm OD. (23.6 mL internal volume for L/D = 355, 53.4 mL for L/D = 800). Helical radius is 9.87 cm (to center of Teflon tube).
2 See text.
3 During step 3 (resuspension) the directions of pumping and spinning coincide in all runs except run no. 34, where they are opposed. The volume pumped during both steps 2 and 3 is 20 mL.
4 Durations of operations (d), (j), and (h) and total sequence time. All other times are fixed as given in Table 1.

particles in any coil segment that sediment during each cycle. The degree of sedimentation is described by the dimensionless group

$$S = \frac{v_s t_s}{D} = \frac{\sigma G t_s}{D}, \tag{6}$$

which, in essence, is the number of tube diameters that particles with a settling velocity v_s or a sedimentation coefficient σ can settle through in a time t_s. Thus the primary effect of centrifugation time is to influence sedimentation in accordance with Eq. 6. This was verified in sedimentation tests, but no runs were made in which t_s was varied at constant centrifugal field strength G.

Polydisperse mixtures have a range of settling velocities and therefore are characterized by an S distribution. Mean values of S between 0.1 and 50 were used in the molecular sieve runs, with $S > 1$ typically chosen to insure adequate sedimentation. While S characterizes sedimentation, it does not fully characterize the overall separation because the field strength, G, also affects resuspension.

Spinning Velocity. Some runs were performed in which t_s was fixed while the spinning velocity was varied. Faster rotation tended to improve dilution and hinder concentration, as illustrated in Figure 9. For this experiment, the rotation rate was increased from 500 rpm (same data as Figure 8) to 2,000 rpm, keeping all else constant. Resuspension was less effective at 2,000 rpm despite stronger secondary flows, and solids accumulated in the coil.

As noted earlier, the driving force for secondary flow is directly proportional to the spinning velocity. However, the centrifugal field strength varies as the square of rotation rate. Apparently, the sixteenfold increase in the centrifugal field strength at 2,000 rpm overwhelmed the fourfold increase in secondary flow strength, holding the particles against the wall and inhibiting resuspension.

Pumping Rate. The pumping rate sets u and, in turn, the driving force for secondary flow. Asymmetric pumping rates were used to minimize resuspension during the first pumping step and to maximize it during the second. Sedimentation and resuspension tests showed that pumping rates below 2 mL/s were advantageous for the first pumping stroke. To obtain effective concentration, the system had to be operated at or near its maximum rate of 50 mL/s during the second stroke. These pumping rates correspond to Reynolds numbers in the coil of 600 and 14,500, respectively.

The inability to concentrate was the most common difficulty encountered in the experiments. Unfortunately, data on the effect on the separation of varying the rate of pumping in the second stroke is limited. A comparison of two runs in which the pumping rate for this step was changed is shown in Figure 10. Concentration was not obtained in either run. Nevertheless, the higher pumping rate definitely increased particle resuspension. The increased resuspension also reduced the dilution rate slightly. One possible explanation is that the increased resuspension caused a greater number of particles to be dispersed at the beginning of the sedimentation step. Thus, more particles could have remained in suspension at the end of sedimentation, causing them to be convected into the dilute reservoir by the subsequent fluid displacement.

Duration of Pumping. In the previous discussions the time dependence of particle resuspension was not mentioned, but its potential importance should not be overlooked. If a concentrated aqueous molecular sieve dispersion is allowed to settle for an hour, a layer of the heavier particles will form on the bottom of the flask. The settled particles can be resuspended by gentle swirling, which mimics the action of secondary flow in a coiled tube. At this low fluid velocity, the layer gradually erodes over a period of 10 to 15 s. However, erosion takes about 1 min for suspensions that have been quiescent for several days.

These observations suggest that effective particle resuspension depends on both the strength and duration of secondary flows. If only moderately strong flows can be obtained, then longer pumping strokes are required. The increased difficulty in resuspending particles that have settled for several days also

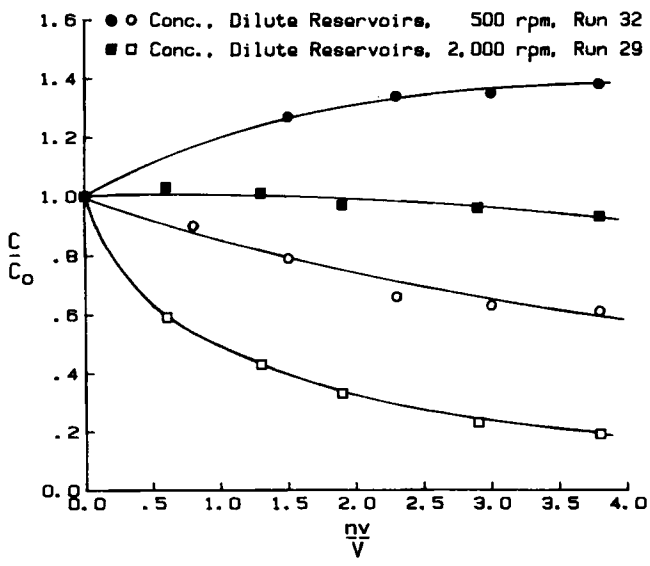

Figure 9. Effect of field strength.
Curves sketched only to aid visualization.

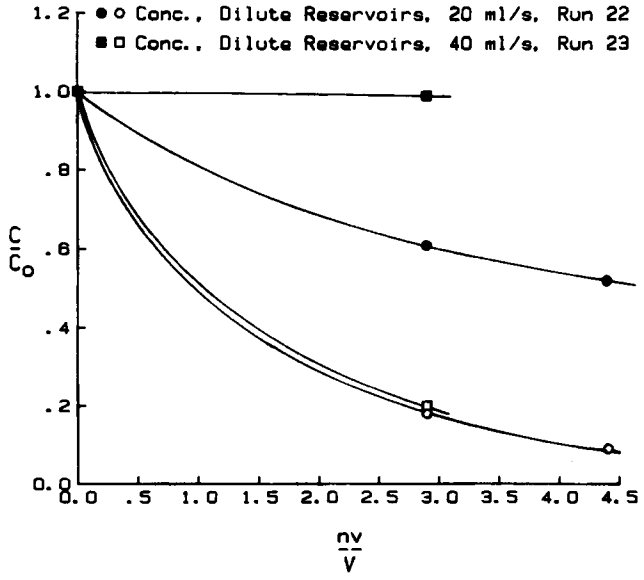

Figure 10. Effect of resuspension pumping rate.
Curves sketched only to aid visualization.

suggests that stronger or longer secondary flows may be needed for resuspension whenever t_s and/or G values are high.

Direction of Pumping. The orientation of axial flow relative to the direction of spin determines the direction in which secondary flows recirculate, as was noted previously. It is possible that one flow pattern is more effective than the other for resuspending particles.

Qualitative arguments can be made in favor of either flow pattern. When axial flow opposes spin, Figure 6, the secondary flow lifts particles from the outer wall directly into the tube center. However, when the spin and axial flow are in the same direction, particles must be moved along the tube circumference to the inner wall before being carried into the center of the tube. This requires more time and strong secondary flows. Furthermore, a stagnation point occurs at the centerline of the outer wall, where particles are most likely to collect as they sediment, so that velocity gradients near the settled particles may be quite small. Similarly, the flow pattern for axial flow opposing spin also has a dead zone along the centerline of the outer tube wall. Without a detailed understanding of the fluid mechanics involved, the more effective flow pattern, if indeed there is one, must be determined experimentally.

In all but two of the runs, the spin and axial flow during the resuspension step were in the same direction. The two exceptions, one made at 500 rpm and the other at 2,000 rpm, were duplicates of previous runs in which only the direction of pumping was changed. In both instances dilution was slightly improved and concentration was significantly reduced. Figure 11 illustrates the effect at 500 rpm.

Before jumping to conclusions, the difference in the strength of these flows should be considered. At 500 rpm the tube velocity is 532 cm/s. With a 0.44 cm ID Teflon coiled tube and a 40 mL/s pumping rate, the average bulk velocity is 263 cm/s. Based on the simple analysis presented above, the driving force for secondary flow when spin and axial flow are in the same direction is 3.0 times greater than for the opposite orientation.

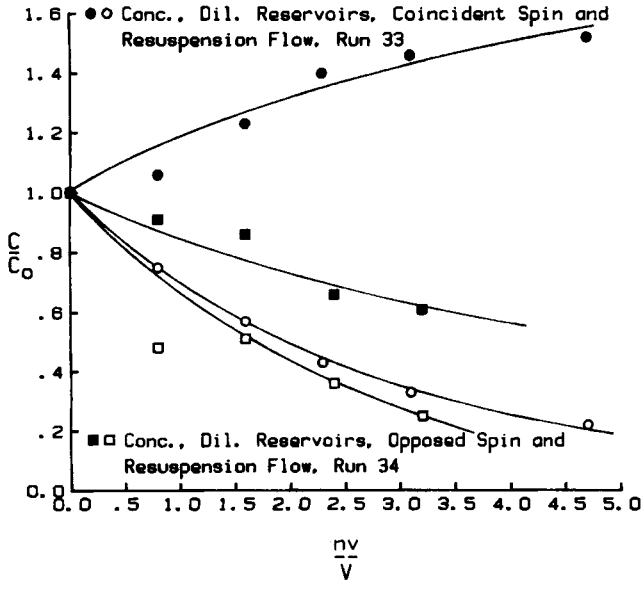

Figure 11. Effect of flow orientation during resuspension.
Curves sketched only to aid visualization.

At 2,000 rpm, the factor is 1.3. Since changing the direction of the axial flow apparently reduced the strength of the secondary flow, the data are not strictly conclusive. However, the run at 2000 rpm had a reduction factor of only 1.3. Thus, it appears that the secondary flows associated with spin and axial flow in the same direction may be more effective for resuspension.

Coil Geometry. Axial dispersion is strongly influenced by the ratio of length to tube diameter (L/D) of the coil. Increasing the number of turns in the coil for a given tube diameter diminishes axial dispersion and improves separation. This was verified experimentally by increasing L/D from 360 to 800. Both dilution and concentration were improved, as shown in Figure 12. Using a greater L/D value is expected to be beneficial up to a point, beyond which axial dispersion becomes negligible.

Particle size distribution

At the end of one run made with ground zeolite (run 33, Figures 11 and 12), the particle size distributions of the material in the coil and reservoirs were measured with a Coulter model N4 laser light-scattering instrument. Both dilution and concentration were obtained in this run, but some material also accumulated in the coil. The solids in the coil had a mean particle size of 1.3 μm, with a broad distribution. After several minutes of settling, the sample mean increased to 1.7 μm. (The light beam in the model N4 instrument passes through the sample holder near the bottom. Thus, for a sample with a broad size distribution that includes larger, faster settling particles, the observed mean particle size will increase and the standard deviation will decrease as the bigger particles settle.) The dilute reservoir had a mean particle size of 0.5 μm and a settled mean of 0.6 μm with a broad distribution. A mean particle size of 0.55 μm with a broad distribution was observed for the concentrated reservoir, changing to 2.0 μm with a standard deviation of 0.5 μm upon settling. For comparison, the feed had a particle size range of 0.2 to 3.0 μm.

The purpose of these measurements was to determine whether accumulation in the coil depended on particle size and to see if fractionation was occurring simultaneously with con-

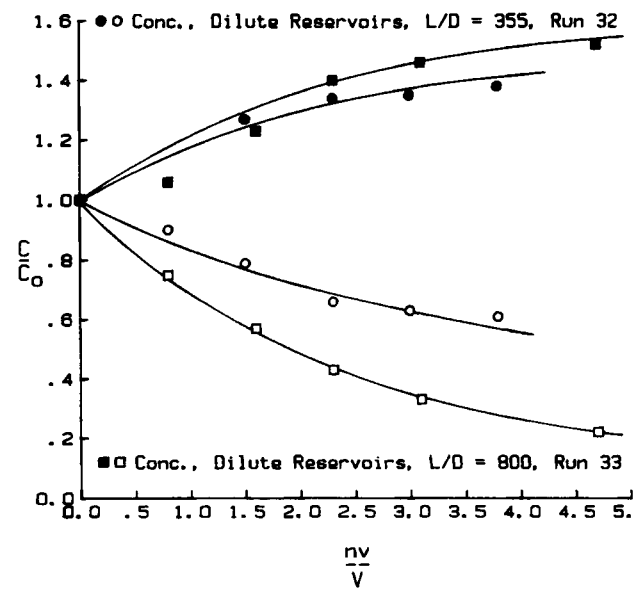

Figure 12. Effect of length-to-diameter ratio L/D.
Curves sketched only to aid visualization.

centration. The data indicate that larger particles accumulated in the coil. This suggests that they were more difficult to resuspend and/or that finer particles did not settle completely. The data also indicate that particles were simultaneously fractionated, but only to a modest extent. The large differences in mean particle size after settling do not necessarily imply a high degree of fractionation. As larger particles settle, they exclude smaller particles from the vicinity of the light path. This effect is more pronounced at higher concentrations.

The effect of differences in the particle size distribution was examined in a series of runs illustrated in Figure 13. The first experiment (run 25) was made with the original 0.2–3.0 μm ground zeolite suspension. The solids that accumulated in the coil were discarded, and the reservoir contents were combined to form the feed for the next run. This procedure was repeated three times in succession. Concentration was not obtained in run 25, but it was achieved in every subsequent run. Discarding the solids that accumulated in the coil lowered the average particle size and narrowed the distribution in every successive run.

Reducing the mean particle size at a fixed field strength had an effect similar to reducing the field strength for a fixed particle size distribution. Removing heavier particles improved concentration and resulted in a successful run. Comparing Figure 13 with Figure 9, in which runs at two different rotation rates are presented, illustrates this point. The data suggest that larger particles are more difficult to resuspend at higher field strengths, and that excessive centrifugal fields are undesirable when separating larger particles.

Colloidal phenomena

Colloidal effects must be considered when particles are in the submicron size range. Neither agglomeration nor adhesion to the coiled tube wall was encountered with the zeolite suspension. However, in a few early experiments with 0.65 μm polystyrene latex ($\sim 10^{-5}$ g/mL aqueous dispersions), particles that accumulated in the coil were found to adhere to the tubing. Zeolite particles that accumulated in the coil were easily removed by stopping the rotation or by flushing the tube with water. The latex particles, on the other hand, had to be scraped from the tube wall. Giddings (1974) added a surfactant to the polystyrene latex dispersion he used in his SFFF studies, probably to reduce particle-wall interactions. Although surfactants might have eliminated polystyrene particle adhesion to the tube wall in the coil, this approach was not pursued. A zeolite suspension, which exhibits minimal colloidal behavior, was used instead to focus on the process characteristics.

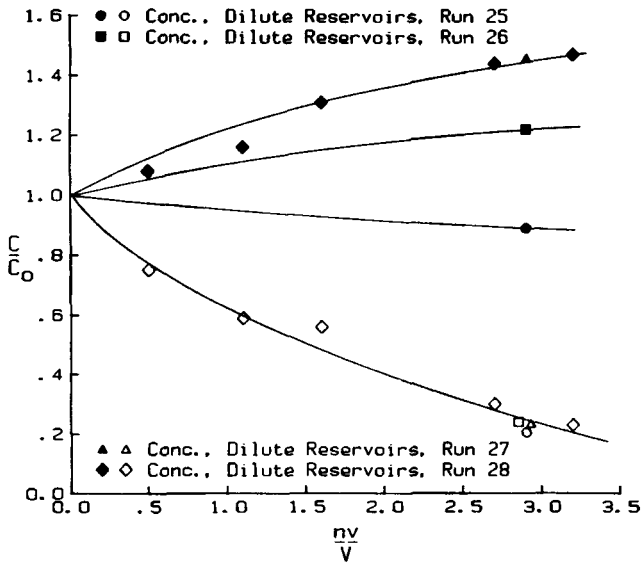

Figure 13. Effect of particle size distribution.
Particle size distribution narrows for each succeeding run. Curves sketched only to aid visualization.

Acknowledgment

Financial support for this work was received from the Minerals and Primary Materials Processing Program of the Chemical Process Engineering Division of the National Science Foundation, Grant No. CPE-8121096, and from Helipump Corporation, for which the authors are grateful. The authors also wish to acknowledge the assistance of Steve Bowen, Arthur Frankshteyn, and Jon Wineland.

Notation

a = particle radius
$C_{n,c}$ = concentration in concentrated reservoir at the end of the nth cycle
$C_{n,d}$ = concentration in dilute reservoir at the end of the nth cycle
C_o = initial concentration
D = inside diameter of tubing
F = driving force for secondary flow
G = field strength; 9.8 m/s^2 for gravity settling, $\omega^2 R$ for centrifugal sedimentation
k = fraction of particles in any coil segment that are sedimented during step 1
L = unwound length of coil
n = number of cycles
R = helical radius of coil
S = dimensionless sedimentation number, $v_s t_s/D$
t_s = centrifugation time
u = maximum axial fluid velocity
U = linear velocity of spinning tube
v = volume pumped in each direction per cycle
v_s = particle settling velocity
V = fluid volume in each reservoir

Greek letters

$\Delta\rho = \rho_2 - \rho_1$
μ = fluid viscosity (particle-free)
ρ_1 = liquid density
ρ_2 = particle density
σ = sedimentation coefficient
ω = rotational speed, radian/s

Literature Cited

Bartlett, R. H., et al. "Development of a Membrane Oxygenator: Overcoming Blood Diffusion Limitation," *J. Thoracic Cardiovasc. Surgery*, **58**, 795 (1969).

Berg, H. C., and E. M. Purcell, "A New Method for Separating According to Mass a Mixture of Macromolecules or Small Particles Suspended in a Field. III: Experiments in a Centrifugal Field," *Proc. Nat. Acad. Sci.*, **58**, 1,821 (1967).

Berger, S. A., L. Talbot, and L. S. Yao, "Flow in Curved Pipes," *Ann. Rev. Fluid Mech.*, **15**, 461 (1983).

Dean, W. R., "The Streamline Motion of Fluid in Curved Pipe," *Phil. Mag. S.7*, **5**, 673 (1928).

Drinker, P. A., et al., "Augmentation of Membrane Gas Transfer by Induced Secondary Flows," *Surgery*, **66**, 775 (1969).

Eustice, J., "Flow of Water in Curved Pipes," *Proc. Roy. Soc. A*, **84**, 107 (1910).

Fuerstenau, D. W., S. Chander, and A. M. Abouzeid, "The Recovery of Fine Particles by Physical Separation Methods," *Beneficiation of Mineral Fines—Problems and Research Needs*, P. Somasundaran and N. Arbiter, eds., NSF Workshop, Sterling Forest, NY (Aug., 1978); Final Report, Grant No. DAR-78-06999, 3 (1979).

Giddings, J. C., "A New Separation Concept Based on a Coupling of Concentration and Flow Nonuniformites," *Sep. Sci.*, **1**, 123 (1966).
Giddings, J. C., F. Yang, and M. N. Myers, "Sedimentation Field-Flow Fractionation," *Anal. Chem.*, **46**, 1,917 (1974).
Giddings, J. C., and M. N. Myers, "Steric Field-Flow Fractionation: A New Method for Separating 1 to 100 μm Particles," *Sep. Sci. Tech.*, **13**, 637, (1978).
Giddings, J. C., M. N. Myers, and K. D. Caldwell, "Field-Flow Fractionation: Methodological and Historical Perspectives," *Sep. Sci. Tech.*, **16**, 549 (1981).
Joseph, B., and R. J. Adler, "Numerical Treatment of Laminar Flow in Helically Coiled Tubes of Square Cross Secion. II: Oscillating Helically Coiled Tubes," *AIChE J.*, **21**, 974 (1975).
Koutsky, J. A., and R. J. Adler, "Minimization of Axial Dispersion by Use of Secondary Flow in Helical Tubes," *Can. J. Chem. Eng.*, **42**, 239 (1964).
Larrain, J., and C. F. Bonilla, "Theoretical Analysis of Pressure Drop in the Laminar Flow of a Fluid in a Coiled Pipe," *Trans. Soc. Rheol.*, **14**, 135 (1970).
Manlapaz, R. L., and S. W. Churchill, "Fully Developed Laminar Flow in a Helically Coiled Tube of Finite Pitch," *Chem. Eng. Comm.*, **7**, 57 (1980).
Menon, M. M., "Theoretical Analysis of Laminar Flow through a Coiled Tube Spinning about Its Helical Axis," M. S. Thesis, Case Western Reserve Univ. (1984).
Parekh, B. K., and W. M. Goldgerger, "Removal of Suspended Solids from Coal Liquefaction Oils," *Fine Particles Processing*, P. Somasundaran ed., Am. Inst. Mining, Metallurgical, and Petroleum Engineers, New York, **2**, 1,687 (1980).
Somasundaran, P., and N. Arbiter, eds., *Beneficiation of Mineral Fines—Problems and Research Needs*, NSF Workshop, Sterling Forest, NY (Aug., 1978); Final Report, Grant No. DAR-78-06999 (1979).
Tarbell, J. M., and M. R. Samuels, "Momentum and Heat Transfer in Helical Coils," *Chem. Eng. J.*, **5**, 117 (1973).
Taylor, G. I., "The Criterion for Turbulence in Curved Pipes," *Proc. Roy. Soc. A*, **124**, 243 (1929).
Thompson, J., "On the Origin of Windings of Rivers in Alluvial Plains, with Remarks on the Flow of Water Round Bends in Pipes," *Proc. Roy. Soc. A*, **28**, 5 (1876).
van Dyke, M., *Perturbation Methods in Fluid Mechanics*, Parabolic Pr., Stanford, CA (1975).
——— "Extended Stokes Series: Laminar Flow Through a Loosely Coiled Pipe," *J. Fluid Mech.*, **86**, 129 (1978).
Weisman, M. H., and L. F. Mockros, "Gas Transfer to Blood Flowing in Coiled Circular Tubes," *J. Eng. Mech. Div., Am. Soc. Civil Eng.*, **94**, 857 (1968).
White, C. M., "Stream-Line Flow through Curved Pipes," *Proc. Roy. Soc. A*, **123**, 645 (1929).
Wilhelm, R. H., A. W. Rice, and A. R. Bendelius, "Parametric Pumping: A Dynamic Principle for Separating Fluid Mixtures," *I&EC Fund.*, **5**, 141 (1966).

Manuscript received Jan. 17, 1985, and revision received Oct. 17, 1985.