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Electrically Stabilized Expanded Beds for Sorption Separations

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ABSTRACT

Recent studies have shown that when high electric fields are imposed on expanded fluidized beds, motion of the particles can cease. This provides the potential for fixed bed operations with the void fraction sufficiently large that only moderate pressure drops are required, even with small particles. This paper explores the dielectric forces that inhibit particle motion in alternating-current electric fields and the potential and limitations of the concept for adsorption operations. The dielectric constants of the particles and the fluid, the size of the particles, the density and viscosity of the fluid, and the imposed electric field gradient are the variables of most importance to bed stabilization. In adsorption tests, the expanded-stabilized beds perform essentially as well as densely packed fixed beds but with only a fraction of the pressure drop.

INTRODUCTION

Adsorption processes that employ solid sorbent to separate solutes from fluids are often hindered by the need to carry out high throughput operations without excessive losses of energy from the

fluid pressure drop that is required to force the fluid through the sorbent particles. For effective separations, it is usually necessary to prevent axial motion of the solid sorbent and to minimize axial mixing (dispersion) of the fluid. This allows sharp concentration profiles in both the solid and fluid phases and is usually achieved by using packed beds of sorbents where the solid particles are restrained by fixed screens, usually the bottom bed support screens when the fluid flows downward.

When adsorption rates are controlled by diffusion resistance within the solid and, to a lesser extent, when rates are controlled by diffusion through fluid "film" resistance outside the particles, it is desirable to minimize the diffusion path in the solid and to maximize the surface area of the sorbent. This usually involves decreasing the diameters of nominally spherical sorbent particles, but somewhat similar results can be achieved by fabricating selected sorbent particle shapes that are thin and thus have high surface areas and short diffusion paths. The use of ever-decreasing particle sizes results in rapidly increasing pressure drops through the sorbent bed, and thus a practical limit is eventually encountered. In cases where the sorbent bed cannot be fabricated into special shapes because of either the costs or the difficulties in fabrication of strong structures, other means of decreasing pressure drop such as increasing bed porosity are needed. One potential means for doing this involves the use of sorbents with ferro- or paramagnetic properties. Rosensweig (1) and Siegel (2) have shown that such particles can be restrained within a magnetic field without resorting to the use of densely packed beds. Rosensweig (3) presented a mathematical description of the forces involved in magnetic stabilization of fluidized beds. Katz and Sears (4); Byers, Watson, and Sisson (5); and Johnson and Melcher (6) also reported that similar restraints can be placed on particles in direct-current electric fields, where the need for using special sorbents with suitable magnetic properties is thus avoided. Another difference between the two approaches is the energy consumption in creating the field. While a current is needed to create a magnetic field, essentially no current is required in the maintenance of a high-voltage electric field. However, the electric fields required can be relatively high, and this technique is limited to use in nonconducting fluids such as gases and many organic liquids. The limitation on solids in the electrically stabilized bed case is that they be nonconducting.

This paper extends the work of Byers, Watson, and Sisson (5) by examining the use of alternating-current fields and by providing further investigation into the nature and magnitude of the forces restraining the particles. The use of an alternating-current field has two potential advantages. First, it eliminates electrostatic effects, which can either be detrimental to the bed performance or at best mask the dielectric forces that are desired to restrain particle motion. Second, alternating-current fields permit the convenient use of alternating currents to measure the degree of electrical polarization. This is potentially important and useful to understanding the forces restraining the particles.

DESCRIPTION OF DIELECTRIC RESTRAINING FORCES

Quantitative description of how electric fields restrain the motion of sorbent particles in expanded beds is complicated by the random arrangement of the particles, but considerable insight can be gained by examining the forces involved. Rosensweig (3) gave a description of the dynamics of the restraining forces involved in magnetically stabilized beds. His analysis is of most interest in the high gas throughput region where the bed is fluidized but the formation of bubbles is reduced or eliminated. On the other hand, this study is concerned with lower flow rates, where the motion of the particles is essentially eliminated; thus a simpler description of particle restraint is presented.

An electric field induces charge separation in dielectric particles, and the net force on the particle can be nonzero if the electric field is nonuniform. The net force can be expressed as

$$\overline{F} = \frac{\alpha V}{2} \nabla |\overline{E}|^2,$$

where

α = polarizability of particles,

V = volume of a particle, and

\overline{E} = electric field strength.

This equation has a form similar to that for the force on paramagnetic particles in magnetic fields. Although the electric field applied to a bed of particles may be uniform, the presence of the particles will introduce local distortions in the electric field. The separated charges within the particles introduce additive fields that are inhomogeneous. These are likely to result in some preferred orientation of the particles into "string" or "net" type arrangements because oppositely charged regions of particles are mutually attracted.

An expression for the forces restraining randomly spaced particles in an electric field would require a number of simplifications before it could be handled mathematically, but some insight can be gained by an examination of the equation. Note that the forces on particles in identical geometric arrangements are proportional to the square of the applied field and directly proportional to the dielectric constant of the particles. The forces are also proportional to the volume of the particles, but note that larger forces are required to restrain larger particles. If the spatial relationship of particles remains the same as the bed is expanded, the separation between particles increases with bed expansion. However, increased bed expansion need not necessarily result in simple geometric separation of particles; there could be an increase in the alignment of particles and a decrease in "cross-linking" interactions between lines of particles. Random

inhomogeneities caused by small slugs of continuous medium could be "frozen" in place. Thus it is difficult to predict how the restraining forces will vary with bed expansion.

Restraint of bed motion should be viewed in two contexts: (1) in terms of the general motion of all particles, and (2) in terms of uniformity (i.e., are all particles restrained equally?). Studies of magnetic stabilization of expanded beds have explored the flow regions just above the minimum fluidization velocity where general bed motion stops, and a second region at still higher fluid velocities where general particle motion resumes. In this higher velocity region, fluidization resumes, but the presence of "bubbles" in the bed is greatly reduced. This study of dielectric stabilization is focused on the lower velocity region where the particles are nominally stationary. However, with randomness in particle arrangement in the bed, the dielectric forces may not always be equal among all particles. Some particles may even be mobile. No mobility has been observed in the gas and liquid studies in electric fields, but the field of view is limited to the outer layers of the bed. With fluid flowing down toward the porous restraining plate, loosely restrained particles can be swept down to the restraining plate, forming a thin packed bed. Mathews and Fan (7) recently showed that if even a small portion of the top (outlet end) of a fluidized bed is forced into a packed bed configuration, the overall performance of the bed in adsorption operations can approximate that of a packed bed in the initial phases of operation. Thus, although a thin layer of particles at the bottom of an electrically stabilized bed in downflow may increase the pressure drop slightly, there may be some compensating gains in mass transfer performance because the layer of particles acts like a packed bed.

EXPERIMENTAL

The experimental apparatus used in this study is shown in Figs. 1 and 2. It consisted of a 4.6-cm ID glass column 30 cm long filled with Davidson 4A molecular sieve particles 500 to 600 μm in diameter, a photograph of which is Fig. 1. The particles were approximately spherical (selected from a batch of nominally spherical particles by a shape classifier) (5). The electric fields were axial in direction and were applied by electrodes located just below and just above the bed. The lower electrode was fixed in place just below the glass frit that supported the bed of particles. The upper electrode could be moved in the axial direction and was positioned to correspond to the top of the bed of particles after expansion. Both electrodes were constructed from stainless steel sieve plates with 0.1-cm-diam openings. The plates offered negligible resistance to gas flow; indeed, the openings were sufficiently large to minimize any restraint of the bed.

The gas used throughout the study was nitrogen, and small concentrations of CO_2 (1000 ppm) were added for mass transfer measurements. The gas rate was measured with a calibrated rotameter, and

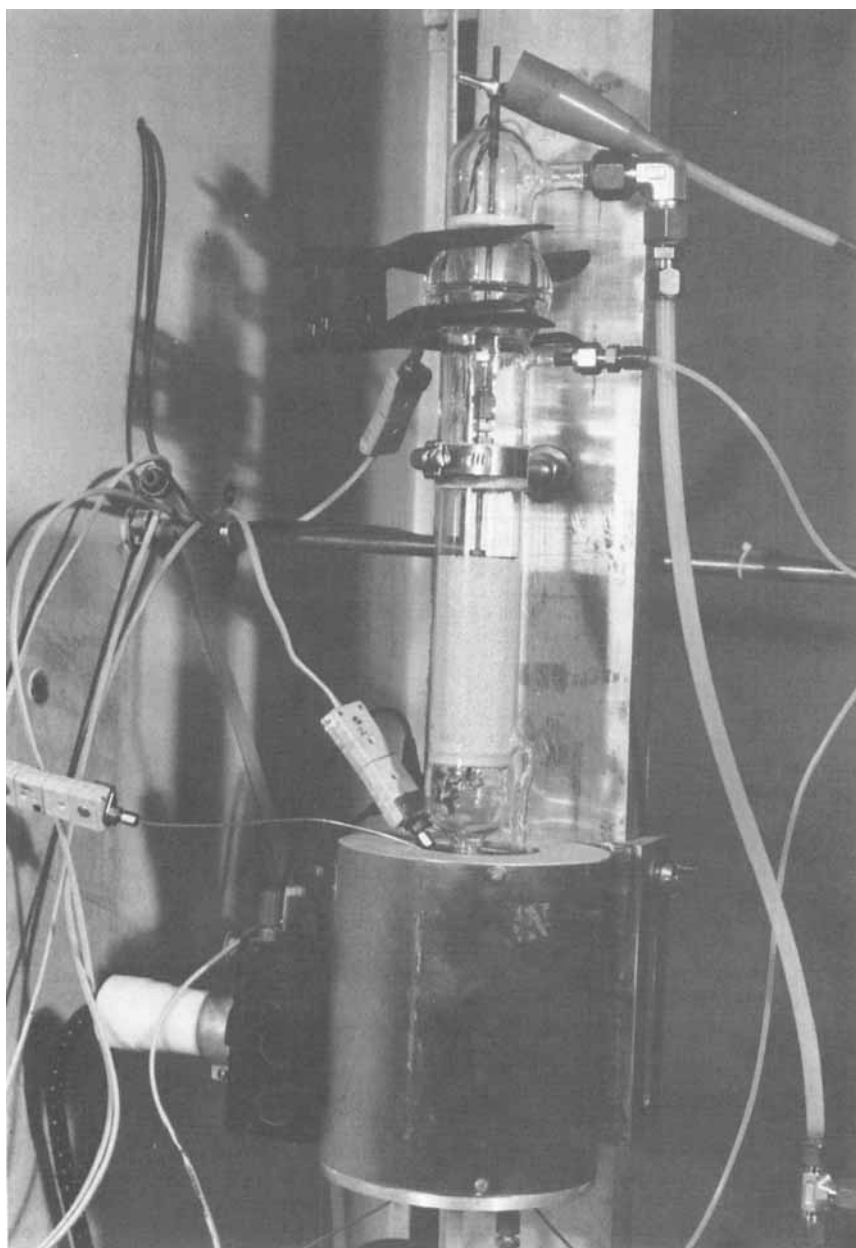


Fig. 1. Photograph of the experimental bed.

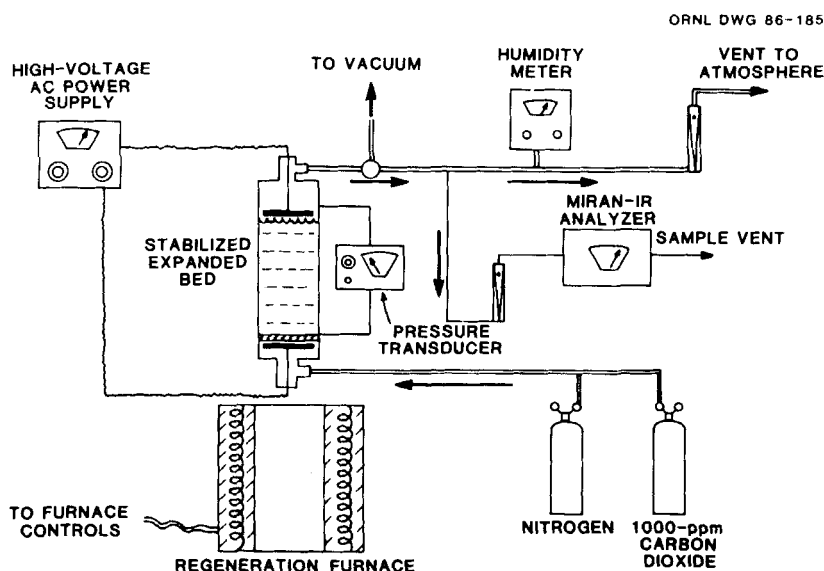


Fig. 2. AC-field stabilized bed experimental system.

the concentration of CO_2 in the gas was monitored with a Miran 101 infrared analyzer. The system was valved for either upflow or downflow through the column. The beds were regenerated by heating to 400°C for several hours, usually overnight, under vacuum.

The alternating-current voltage applied between the electrodes was obtained with a custom-designed "step-up" transformer which operated at 60 Hz. The maximum voltage used for stabilizing the bed was 20 kV. The transformer operated at higher voltages, but the current drawn by the transformer became excessive in the particular configuration and the power supply used in this study. The output voltage was stable over the several hours required for the longest runs.

RESULTS AND DISCUSSION

The ability of the alternating-current field to restrain particle motion is illustrated in Fig. 3. Pressure drop characteristics provide an excellent indication of bed fluidization and of changes in void fraction. In the latter case, pressure drop is a very sensitive indication of minor changes in void fraction. Pressure drop data are reported for a 4.2-cm-diam (ID) bed containing 56.84 g of molecular sieve; this corresponds to a settled bed depth of approximately 5.5

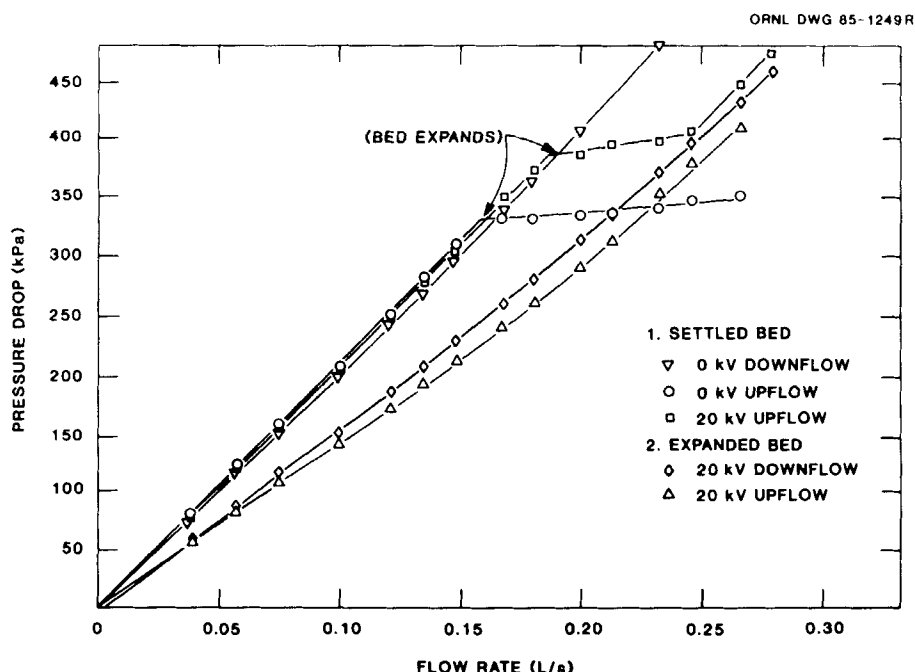


Fig. 3. Pressure drop at different bed conditions.

cm. Two curves in Fig. 3 provide a baseline with no voltage applied to the bed. One curve corresponds to downflow of gas through the bed while the second is the corresponding upflow case. These represent the pressure resistance of a packed bed of the particles. At low flow rates, the upflow and downflow curves are similar, as expected. At approximately 0.15-L/s or 165-KPa (24-psi) pressure drop, the bed fluidizes and the upflow curve levels at a value where the pressure drop vs the bed cross section equals the weight of the particles.

The remaining three curves in Fig. 3 present data with a 20-kV applied alternating-current voltage to upflow and downflow experiments. For all of these curves, the electrodes were located 6.1 cm apart. When the 20 kV was applied to the settled bed, the upflow pressure drop continued following that of the packed bed (downflow with no applied voltage) well beyond the point where fluidization would normally occur. For expanded bed studies, the bed was first expanded to fill the void between the electrodes prior to application of the voltage. Once the alternating-current field was applied, motion of the particles appeared to stop completely, similar to

what had been observed with direct-current fields (5). The slopes of the two 20-kV curves are not exactly identical, but the slopes are much less than for the packed-bed cases (no applied voltage). On a constant weight basis, a 25 to 30% decrease in pressure drop can be expected in the stabilized-bed case as compared to the equivalent-flow fixed-bed case. Byers, Watson, and Sisson (5) have shown that this effect is significantly greater in the case of smaller particles and obviously where greater bed expansion can be achieved. Expansions in bed volume by as much as 50% have been reported by the Exxon group in magnetically stabilized beds (1, 2, 3). We have some preliminary indications that control of beds with similar expansion can be achieved with electric stabilized beds. However, the sorption characteristics of these beds require further investigation. The fact that pressure drop is a very sensitive function of the void fraction of the bed is particularly significant in this technology, because even small bed expansions (changes in the void fraction) can alter the energy loss significantly.

The linear dependence of the pressure drop with flow rate extends beyond the normal fluidization point. This is to be expected because the particles are "fixed" by the electric field and thus should behave more like a normal packed bed where the particles are restrained by being closely packed with neighboring particles.

The small difference between the upflow and downflow curves for expanded beds probably reflects a characteristic of electrostabilized beds; the dielectric forces that restrain particle motion are finite. Restraining forces are believed to result from interactions of dielectric charges induced in the particles. The strength of these forces depends on the relative locations of particles with respect to surrounding particles. Because the particles are initially in an approximately random arrangement before the electric field is applied, all particles are not expected to be subjected to the same restraining force.

One explanation for the difference between the upflow and downflow curves in Fig. 3 is that some of the less restrained particles are removed from the bed by the flowing gas. When the gas is in downflow, these particles can work their way down toward the bottom of the column to the glass frit that supports the bed. There they can form a thin fixed bed. This can then give a higher pressure drop. With the gas flowing upward, any particles moved by the flowing gas are likely to go upward. Because there is no frit to restrain them, they tend to remain as individual particles near the top of the bed, and thus to have pressure drop characteristics of a fluidized bed. Although we did not observe any particle motion, nor segregation of granules, the interior of the bed was not visible.

Comparisons of the expanded-pressure bed drop measurements with two popular correlations for packed beds are given in Fig. 4. Here both the upflow and downflow pressure drop data are plotted, along with predictions from correlations proposed by Leva (8) and by Ergun

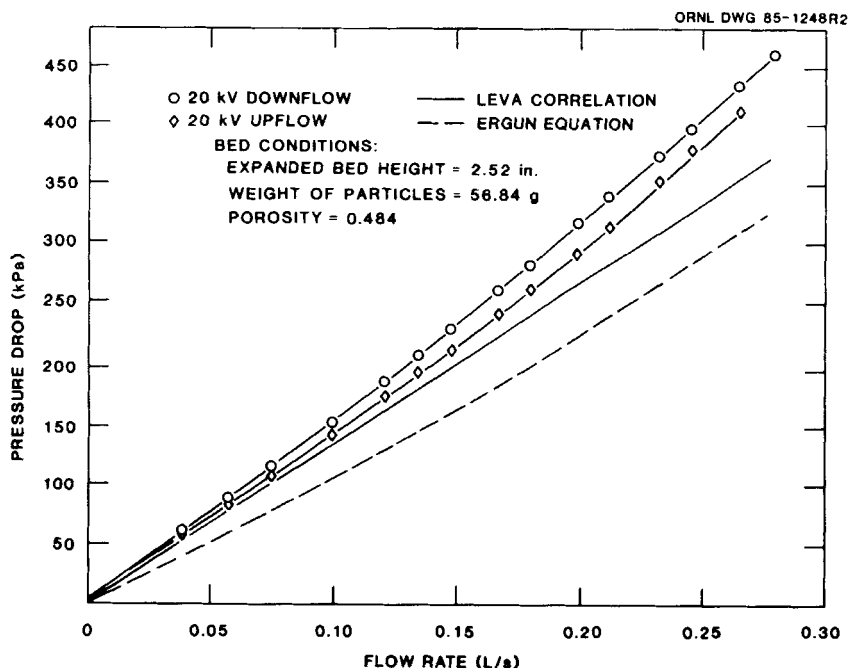


Fig. 4. Comparisons of calculated and experimental values for the expanded bed.

(9). The predicted curves are estimated based on the estimated void fraction of the expanded bed. The prediction from the Leva correlation is only approximately 8% lower than the upflow and 15% lower than the downflow data. The agreement between the data and the correlations is within the range expected. Differences of this magnitude may result because the particles were neither perfectly uniform in size nor perfect spheres. Since the shapes of all curves are quite similar, it would appear that our estimate of one or more of the three parameters (average diameter, sphericity, or void fraction) might correlate differently from the original equation, or be estimated inaccurately.

Because the electric fields are expected to result in some rearrangement of the particles from the relatively random arrangements expected for packed beds on which the correlations were based, exact agreement with the correlations should not be expected. In fact, the agreement is essentially the same as that observed between the correlations and packed beds with no electric fields. This similarity must not be viewed as an indication that the electrically stabilized beds are the same as expanded packed beds; it only means that in this one way they are similar.

Results of the CO_2 adsorption experiments are shown in Figs. 5 and 6. These figures present breakthrough curves produced with 56.84 g of molecular sieve in the bed and with flow rates of 0.122 L/s (Fig. 5) and 0.181 L/s (Fig. 6). The lower flow rate (0.122 L/s) is below the fluidization point, and the higher rate (0.181 L/s) is above the fluidization flow rate. All of the curves in Fig. 5 represent conditions which should produce the same breakthrough curves, if the electric field itself has no effect upon sorption. The one curve which is slightly poorer than the others is the upflow with no field applied. This is the least restrained case, and even that breakthrough is only marginally sooner than the others and could result from small differences in experimental conditions, particularly differences in the inlet gas composition. Since we have significantly lower pressure drops in the expanded bed cases, the overall performances of the expanded beds are substantially improved.

Fluidization is reflected clearly in the upflow curve shown in Fig. 6 with no applied voltage. In this case, the particles are largely mixed while the gas phase may be in plug flow, with some mixing. The immediate breakthrough and long tail are totally unacceptable for most sorption applications. Again, the downflow curve represents the performance of a packed bed with no motion of the particles. The shape of the breakthrough curve results from equilibria relations, mass transfer resistances, and possibly some dispersion in the gas phase. It appears from the curves in Fig. 5 that the behavior is almost totally controlled by the solid phase in nonfluidized situations.

Figure 6 illustrates some differences among the various stabilized modes of operation. The downflow case with the establishment of the field after the bed was fully expanded is almost indistinguishable from the ideal (0 kV downflow). From an overall point of view, the performance of the stabilized beds are an enormous improvement over the fluidized bed from a sorption viewpoint. In the best of cases, one can approach fixed bed sorption performance with substantial pressure drop improvement. The differences in upflow cases may result from several factors. These include:

1. small motion (mixing) of the solid particles,
2. increased dispersion (perhaps even channeling) of the gas flow, or
3. less favorable mass transfer behavior due to different arrangements of the particles.

With the data shown, there is no significant difference between mass transfer rates in the downflow (fixed-bed) cases and those in the electrically stabilized expanded cases. The slight differences in the locations of the curves on the graphs result principally from small differences in the CO_2 concentration and the nonreproducible flow rates used in each run.

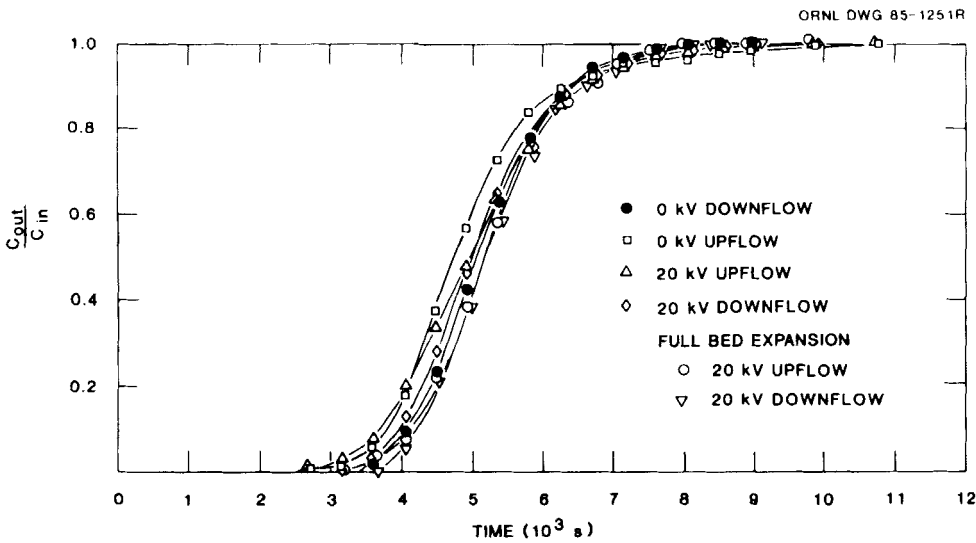


Fig. 5. Mass transfer with 0.122 L/s gas rate.

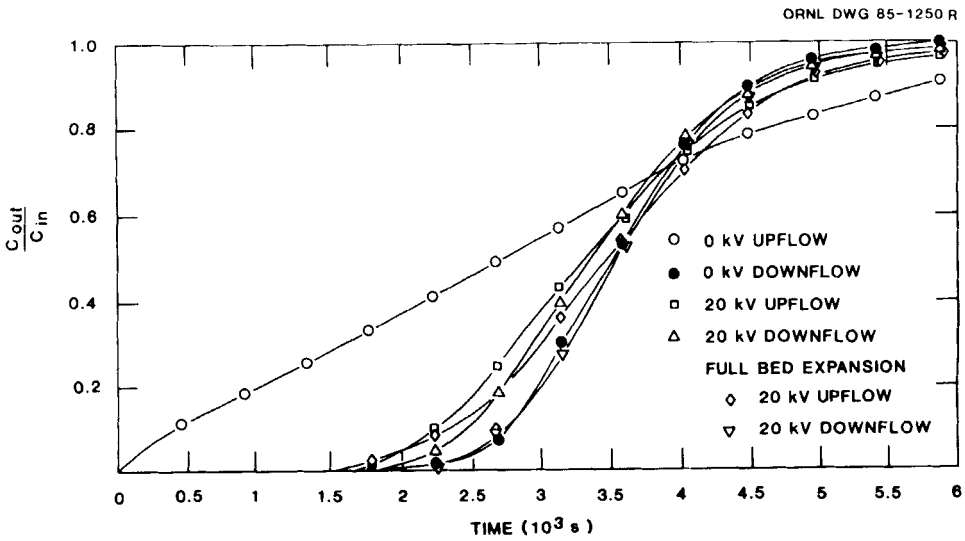


Fig. 6. Mass transfer with 0.181 L/s gas rate.

Differences between the mass transfer characteristics were estimated by calculating the number of transfer units in the column in the expanded and "fixed" states. The mass transfer resistance was approximated by a linear form. Any differences between the number of transfer units calculated for the fixed bed (downflow) and the expanded bed (upflow or downflow) were insignificant and well within the uncertainty of the results when the gas flow rate was below the fluidization rate (Fig. 5). When the gas rate was above the fluidization rate, there was again no difference between the fixed bed and the expanded bed with downflow; however, the upflow curve did indicate a detectably lower number of transfer units. These results show that dielectric stabilization of expanded beds can give mass transfer performance which is equivalent to that of fixed beds, but with significantly lower pressure losses.

CONCLUSIONS

High voltage alternating-current electric fields can be effective in restraining the motion of dielectric sorbent particles in an expanded bed. The degree of particle restraint is not qualitatively different from that observed previously with direct-current fields, and the elimination of electrostatic forces (from charged particles) appears to reduce undesirable attraction of the particles to one of the electrodes. Particle motion and fluid (axial) dispersion are reduced sufficiently that beds expanded by as much as 10% perform essentially as well as packed beds, but with reduced pressure losses.

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