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REMOVAL OF SUBMICRON SILICA PARTICLES FROM TERT-AMYL ALCOHOL BY DIELECTRIC/ELECTRIC PACKED BED FILTRATION

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ABSTRACT

A packed bed with an applied electric field is used to remove submicron and nanometer particles from a nonconducting or slightly conducting solution. Several studies have shown that the application of an electric field to a packed bed significantly increases the performance of the filtration. To enhance the electric-field filtration efficiency, it is desired that the packing materials have a

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higher dielectric constant than the solution so that the electric-field lines will be diverted into the packing materials.

In the present studies, a dc voltage of 0 to 8 kV/cm is applied to a packed bed (2.5-cm diameter and 3.0-cm length) filled with 1-mm-diameter glass beads. The filtration medium contains submicrometer or nanometer SiO_2 particles dispersed in *tert*-amyl alcohol. Two particle sizes are investigated: the average particle sizes are about 300 nm and 50 nm, respectively.

Visible light spectrophotometry is used to estimate the amount of SiO_2 particles in the effluent. The experimental results are presented as a series of breakthrough curves. The effect of the applied electric field on the breakthrough curve on two different particle sizes is presented. Depending on the applied electric field and the conductivity of the system, heating of the packed bed may occur. The operating current and temperature of the packed bed are also presented.

INTRODUCTION

Rapid removal techniques are required in the recovery and concentration of ultrafine precursor ceramic particles during the continuous production of dilute colloidal suspensions. Conventional methods (e.g., centrifugation and distillation) for the removal (or concentration) of ultrafine particles from (or in) liquids are expensive and are often large consumers of energy. Traditional filtration techniques are very inefficient or require large surface areas for the removal of ultrafine solids from liquids. Furthermore, as the size of the solid particles approaches the nanometer size regime, such processes are no longer viable unit operations for solid-liquid separations. Several studies have shown that application of a magnetic (1) or an electric field (2-5) to packed-bed filter media enhances the removal of aerosol particles from gas streams and removal of particles from nonconducting and conducting liquids. The criterion for efficient operation of dielectric/electric bed filtration (DEBF) is that the dielectric constant of the packing be higher than that of the continuous phase. Several forces act on colloids in packed-bed filter media. These include fluid drag force, charged particle electrical force-polarized filter media force by an external field, polarized particle-polarized filter media electrical force, electrostatic repulsive force between suspended and collected particles, and the London-van der Waals force. If the particles are above approximately 1 μm, gravitational forces will also be important.

An application of a rapid particle capture technique, such as dielectric/electric bed filtration, to the removal of particles from continuous liquid-phase precipitation processes is attractive because, in many cases, the beds are easily cleaned by turning off the electric field and allowing the particles and water to be washed from the bed. DEBF is also appealing because of low energy requirements.

The objectives of the present studies are to (a) determine if spherical glass beads can be used as the granular electrofiltration medium (ϵ = ~4-6) for the separation of silica particles from a low-molecular-weight alcohol and (b) provide a model colloidal dispersion for theoretical analysis of dielectric/electric bed filtration. The silica — *tert*-amyl alcohol system was chosen because of the low dielectric constant ($\epsilon = 5.85^{25}$) of the alcohol, the ability to easily prepare silica particles of various sizes (20 nm — 1000 nm) (6) and the excellent stability of the silica colloids in *tert*-amyl alcohol.

THEORY

It is assumed that the fluid phase containing the colloidal particles to be captured enters the bed and the particles are trapped on the packing material (collectors) between the electrodes, eventually breaking through the exit of the bed when the bed is filled. Typically, one observes an 'S'-shaped breakthrough curve for the materials. In modeling a bed, it is assumed that there is homogeneity in the bed, leading to concentration variations only in the axial direction (z) and with time, t. A mass balance over an element of the packed bed is performed on the colloids in the liquid phase and the corresponding deposited material leading to the following relationship (7):

$$\epsilon D_{zi}\frac{\partial^2 c_i}{\partial z^2}=\epsilon\frac{\partial c_i}{\partial t}+(1-\epsilon)\rho_b\frac{\partial \overline{q_i}}{\partial t}+\epsilon v\frac{\partial c_i}{\partial z} \qquad (1)$$

where

D_{zi} = dispersion coefficient, cm^2/s;
c_i = concentration of colloids in liquid, g/cm^3;
z = axial distance, cm;
t = time, s;
q = concentration of trapped particles, g particles/g packing material;
ϵ = void volume fraction in the bed;
ρ_b = density of packing material;
v = superficial velociby of liquid through the packed bed, cm s^{-1}.

Most models assume that axial dispersion is negligible; therefore, the left side of equation (1) is set equal to zero.

Another mass balance is done for the removal of colloidal particles from flowing suspensions toward a single stationary collector in an element. This process is governed by the convective diffusion equation (8):

$$\frac{\partial c}{\partial t}+\nabla\bullet J=Q \qquad (2)$$

where

J = particle flux vector, (see equation below), g $cm^{-2}\,s^{-1}$; and
Q = particle source term, g $cm^{-3}\,s^{-1}$.

The particle flux vector is computed by the equation

$$J = -D \bullet \nabla c + Uc + (1/kT) D \bullet Fc \tag{3}$$

Here **D** is the particle diffusion tensor (cm^2/s), **U** is the particle velocity vector induced by the flow of the suspending medium (cm/s), T is the absolute temperature (K), **F** is the external force vector and k is Boltzmann constant. The external force vector is the sum of the following forces:

$$F = F_E + F_{DD} + F_C + F_L \tag{4}$$

where

$\mathbf{F_E}$ = charged particle/polarized collector force,
$\mathbf{F_{DD}}$= polarized particle/polarized collector force,
$\mathbf{F_C}$ = charged particle/charged particle force,
$\mathbf{F_L}$ = London-van der Waals Force.

If the external forces and hydrodynamics are known, the rate of deposition of the particles can be computed. However, computing the behavior of these forces in a packed bed is a formidable task. The problem is simplified by doing the computation for the rate of deposition on a single spherical collector and assuming that all the particles in an element of the bed behave in a similar manner (5, 8-12).

Another problem is encountered when the collector and/or particles have free surface charges and/or a thin double layer. An external electric field can cause the migration of free surface charges or deformation of the diffuse lining of the double layer (13-20). This results in an "apparent" dielectric constant of the solids which exceeds their actual dielectric constants by several orders of magnitude (15).

Barker et al. (5) were the first to attempt to model granular electrofiltration and to compare the model with experimental results. Their model was in semiquantitative agreement with the experimental data. Inspection of the data suggests that the colloidal particles were highly agglomerated and irregular in shape. It is believed that model particles (spherical, monodisperse, and unagglomerated) are needed to further test and develop theoretical models for dielectric/electric bed filtration.

EXPERIMENTAL PROCEDURE AND APPARATUS

A flow diagram for DEBF unit is shown in Figure 1. The packed bed consists of 1-mm glass beads sandwiched between two electrodes. The electrodes are made from a stainless steel wire mesh to support the glass beads. In a normal operation, the suspension is passed upward through the bed. The bottom electrode is positive and the top one is negative if the surface of the particles in the suspension is negatively charged. Conversely, the electrodes are switched if the surface charge of the particles is positive. To reduce the void volume in the DEBF column, 3-mm glass beads were inserted below and above the electric bed filter region. The filter is also moved closer to the top of the column to minimize the volume between the top of the bed and the detector.

Colloidal silica particles were prepared by the ammonia-catalyzed hydrolysis of tetraethylorthosilicate (TEOS) in ethanol (6). Initial solutions containing 0.3 $\underline{M}$ TEOS, 6 $\underline{M}$ H_2O, and 0.7 $\underline{M}$ NH_3; and 0.15 $\underline{M}$ TEOS, 8.8 $\underline{M}$ H_2O, and 0.1 $\underline{M}$ NH_3 were prepared to produce particle sizes of approximately 50 and 300 nm, respectively. Transmission electron micrographs of the particles are shown in Figure 2. A 100- or 200-mL aliquot of the resulting colloidal suspension (Solutions A or B) was centrifuged at 4000 or 15,000 rpm for 30 min.

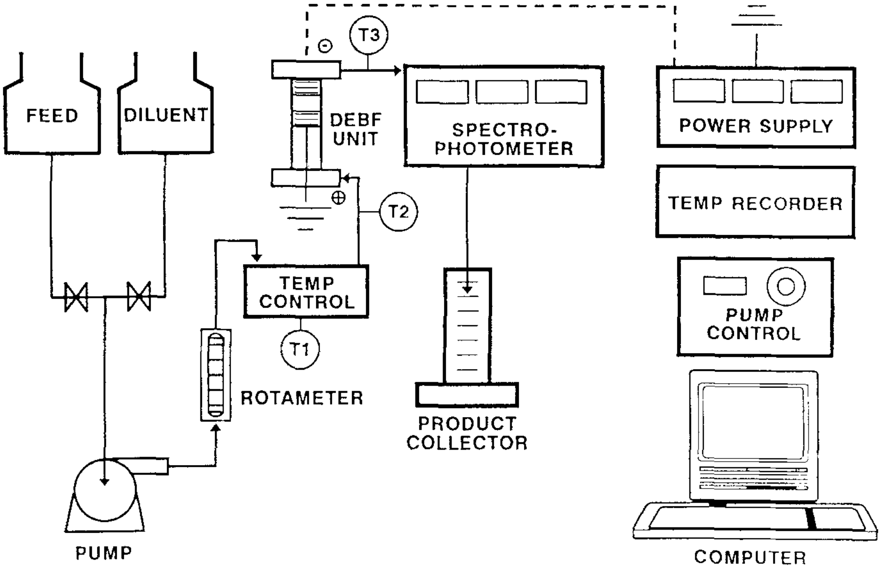

FIGURE 1. DEBF flow diagram.

After decanting the supernatant, the solids were resuspended in 1 L *tert*-amyl alcohol by ultrasonication. This suspension had a low conductivity and was stable for about a week. A high-conductivity suspension was prepared by directly diluting 100 mL of Solution A with 900 mL of *tert*-amyl alcohol. The SiO_2/*tert*-amyl alcohol suspensions contained 1.8 g SiO_2/L. These suspensions were diluted to prepare standards to calibrate the spectrophotometer (Perkin-Elmer Model 200). Measurements were taken at a wavelength of 420 nm.

Silica suspensions (1.8 g SiO_2/L) in *tert*-amyl alcohol were prepared and filtered in a packed bed (2.5-cm diameter x 3-cm length) of 1-mm-diameter glass beads. A voltage potential of 10 to 25 kV dc was applied across the bed (Glassman power supply model PS/ER30R10CDZ11, 0-30 kV and 0-10 mA). The influent and effluent temperature, electrical current, and applied voltage

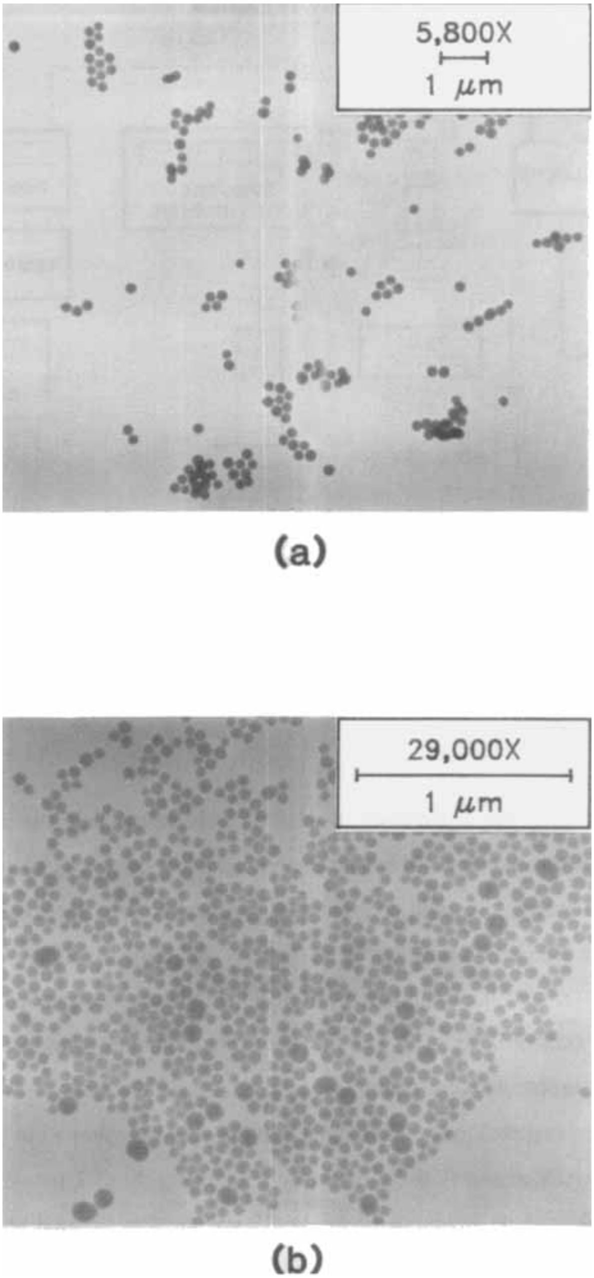

FIGURE 2. Silica particles produced by ammonia catalyzed hydrolysis of TEOS (a) 0.3M TEOS, 6M H_2O, 0.7M NH_3 (~300 nm); (b) 0.15M TEOS, 8.8M H_2O, 0.1M NH_3 (~50 nm).

across the bed and the percent transmittance of the effluent were monitored continuously by a computer data acquisition system.

RESULTS AND DISCUSSIONS

The purpose of the experiments was to demonstrate that electric-field packed-bed filtration can be used to remove submicrometer and nanometer particles from the nonconductive and slightly conductive liquids. Experimental results are presented as a series of breakthrough curves. Breakthrough curves for various conditions are shown in Figures 3, 4, and 5.

The effects of electric field on the breakthrough curves for 300 nm and 50 nm particles are shown in Figures 3 and 4, respectively. When no electric field is applied across the bed (i.e., 0 kV), the breakthrough time is about 8.6 min and 9.0 min for Figures 3 and 4, respectively.

As the electric field increases from 0- to 25-kV (Figures 3 and 4), the filtration efficiency increases, as indicated by the prolongation of particle breakthrough. Figure 3 shows an initial rapid increase and decrease of the particle concentration in the effluent stream for the 15-kV and 20-kV runs. After this period, the effluent concentration increases gradually and tends towards the influent concentration. In addition, several distinct slopes were observed in Figure 3 of the 20-kV run. These different zones of filtration efficiency might correspond to different filtration mechanisms. Future work will involve developing models to explore the mechanistic reasons for these zones.

Figures 6a & 6b show the electric current through the bed and the temperature of the effluent of runs in Figure 3, respectively. As the applied electric field increases, the current through the bed increases. Since the SiO_2 particles are negatively charged, a denser concentration of trapped particles are always observed near the positive electrode first as compared to the negatively charged electrode. As the particles start to build up on the bed, strings of particles appeared along the glass beads across the electrode, and these stringing

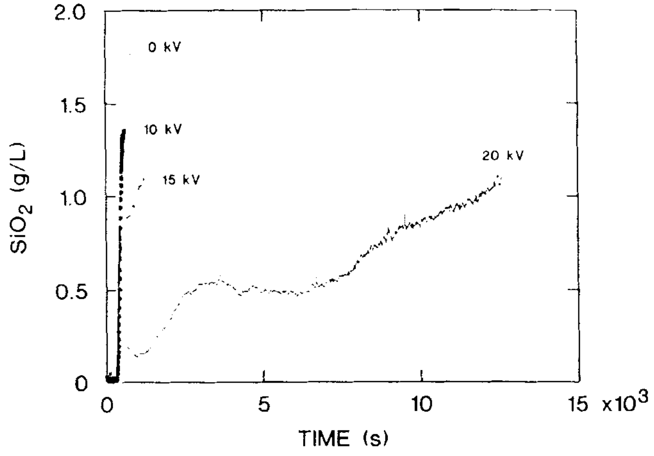

FIGURE 3. Breakthrough curves for 300 nm SiO_2 particles: Effect of applied voltage across filter bed (Q = 8.4 mL/min, c_o = 1.8 g/L SiO_2 in *tert*-amyl alcohol).

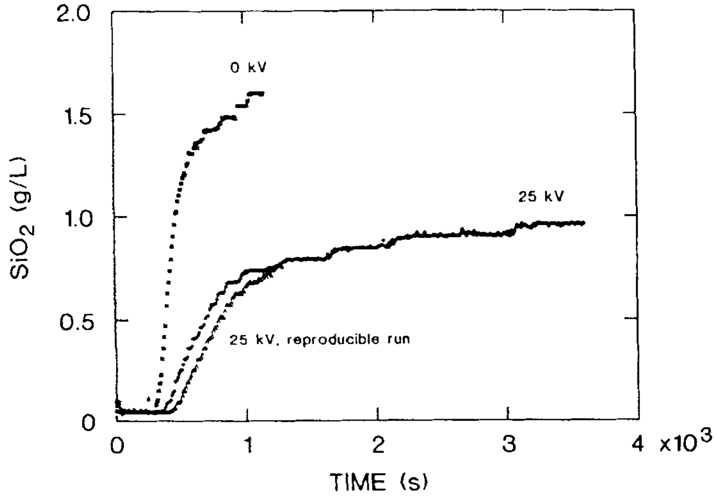

FIGURE 4. Breakthrough curves for 50 nm SiO_2 particles (Q = 6.4 mL/min, T = 25 °C, c_o = 1.6 g/L SiO_2 in *tert*-amyl alcohol).

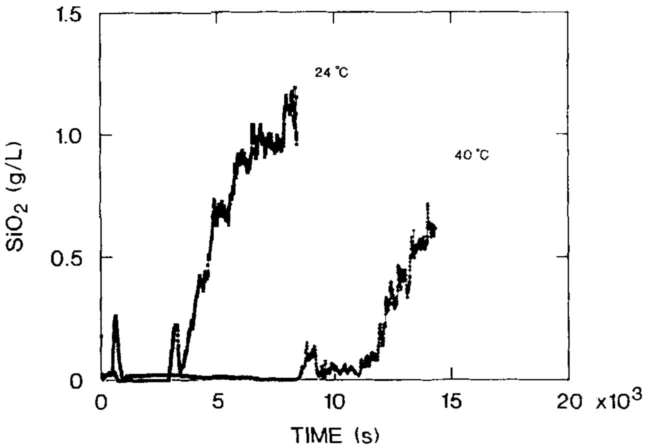

FIGURE 5. Breakthrough curves for 300 nm SiO_2 particles (Q = 6.4 mL/min, $V_{applied}$ = 25 kV, c_0 = 1.6 g/L SiO_2 in *tert*-amyl alcohol).

particles may increase the conductivity of the bed. As a result, the power dissipation to the filter increases, and the temperature of the bed increases, as indicated in Figure 6b.

The effect of the influent temperature on the breakthrough curve is presented in Figure 5. It was found that the filtration efficiency is strongly dependent on the operating temperature. The maximum loading in terms of volume percent of the filter bed void volume was found to be 7% and 22% for 24°C and 40°C, respectively. The increases in the filtration efficiency might be due to decreases of the dielectric constant of the liquid as the temperature increases. The relative dielectric constant of the glass bead and the suspension is very important to filtration efficiency (5). For the best filtration efficiency, it is desirable to have the dielectric constant of the packing materials higher than that of the suspension so that the electric force is diverted into the packing materials. As the dielectric constant of the liquid decreases and that of the solid remains constant, the electric capture force on the particulate increases; as a result, the filtration efficiency increases.

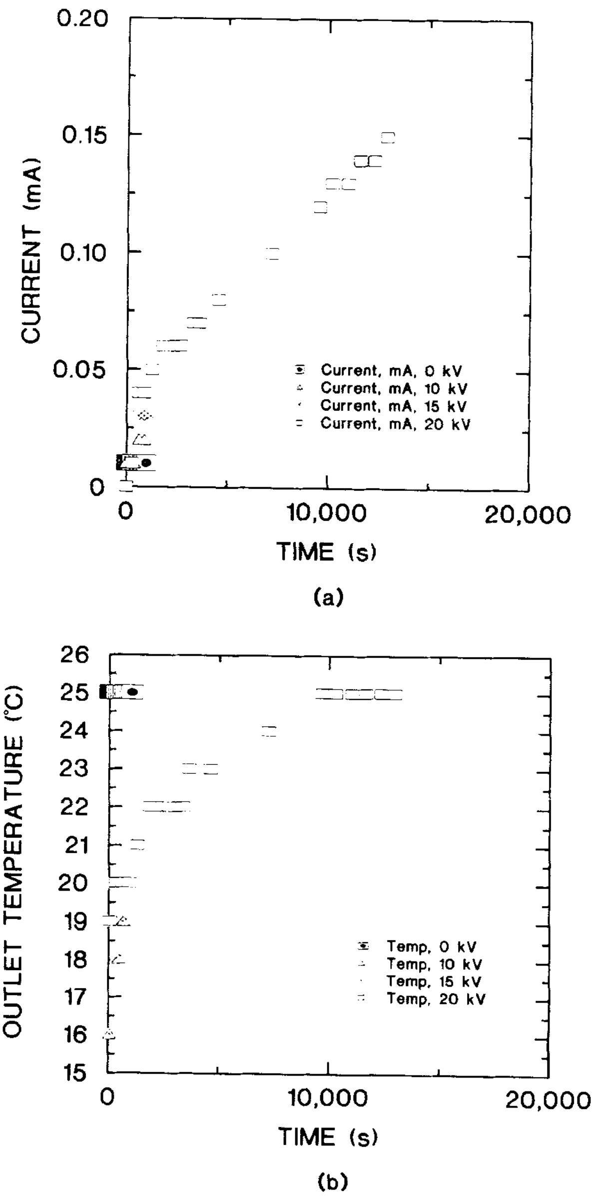

FIGURE 6. Electric current through filter bed and Temperature of effluent stream (Q = 8.4 mL/min, c_o = 1.8 g/L SiO_2 in *tert*-amyl alcohol;$V_{applied}$ = 0, 10, 15 and 20 kV): (a) electric current and (b) temperature.

The increase in filtration efficiency can also be due to the reduced drag that is caused by the decrease in viscosity with increasing temperature. Future experimental and theoretical investigations have been designed to evaluate the relative importance of the effects of polarization and viscous drag forces on the separation of silica particles from *tert*-amyl alcohol.

CONCLUSIONS

The performance of a packed-bed filter is greatly increased by applying an electric field across the bed. Experiments showed that this method is effective for removing submicrometer and nanometer particles from a suspension of nonconductive or slightly conductive solvent. Experimental results also showed that the filtration efficiency increased as the bed temperature increases. The increase in the filtration efficiency is due to the decrease of the dielectric constant and/or viscosity of the solvent as the temperature of the solvent increases through the filter bed.

Future studies will involve studying the effect of particle surface potential on the filtration efficiency. Experiments will also be carried out in a temperature-controlled environment. A theoretical model based on the convective diffusion equation will be developed to explain the experimental results.

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