Electrokinetic Phenomena in the Filtration of Colloidal Particles Suspended in Nonaqueous Media

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Filtration of carbon black particles from tetralin through a bed of sand with simultaneous measurement of bed streaming potential shows that the charge on the filter and its neutralization by depositing colloidal particles plays an important role in filter performance. Filter capacities up to the point of charge neutralization, calculated from knowledge of the colloid and filter zeta potentials using a semitheoretical model for variation of filter surface charge characteristics, are found to be in good agreement with experimental values.

SCOPE

There has been a recent surge of interest in coal liquefaction processes for converting high sulfur coals to clean liquid fuels. Separation of the residual mineral matter and coal particles from the final product is a major problem in these processes. Conventional separation processes have not yielded the solution due to difficulties caused by the high viscosity of the liquid product and the fineness of the solids, a large fraction of which are submicron in size. Recently, suspended particles in coal-derived liquids have been shown to carry a charge (Henry and Jacques, 1977; Rodgers, 1978; Lee et al., 1979). Asphaltenes and preasphaltenes in coal liquids have been found to affect the suspended particle size (Briggs et al., 1980), and Rodgers (1980) has related this phenomenon to the changes in particle charge caused by asphaltenes depositing on the particle surface. Lee et al. (1979) enhanced sedimentation rates by applying an electric field and also presented (Lee et al., 1980) a design of a cross-flow electrofilter which utilized the electrophoretic motion of the particle to prevent filter clogging. Thus, the charge residing on the residual mineral matter and the coal particles can be used to advantage in a number of separation processes.

Deep bed, or granular media, filtration is a widely used separation process for economical and efficient removal of low concentrations of fine solids from water. The need for improved filtration techniques for treating the products of coal liquefaction warrants a study of deep bed filtration with special emphasis on nonaqueous systems.

Removal of colloidal particles in deep bed filtration is achieved by deposition onto the filter grains constituting the bed. The particles are transported to the filter grain (collector) surface by the mechanism of interception, sedimentation and Brownian diffusion. Whether or not the particle finally attaches itself to the collector surface depends on the net resultant of the short-range forces which are the attraction due to London-Van der Waal forces, and the force due to electrical double layer interactions. Repulsive double layer force between similarly charged particle and filter medium can be strong enough to prevent deposition. The extent to which the surface charges affect deposition rates is also determined by the distance to which their influence extends. By virtue of their low dielectric constant and low electrical conductivity, organic liquids have very thick interfacial double layers as compared to aqueous solutions. Therefore, the effects of surface charges are expected to manifest themselves more strongly in the filtration of organic liquids than aqueous solutions.

Recently, Wnek et al. (1975) proposed a model which predicts filter performance based on surface charge characteristics of the filter and its variation due to particle deposition. In this paper, the changes in the double layer around the filter grains due to particle deposition have been monitored by means of the streaming potential, and the effect of these changes on the performance of the filter has been studied. The system chosen for the study was a suspension of carbon in tetralin, which is a coal-derived product.

CONCLUSIONS AND SIGNIFICANCE

1. Simultaneous monitoring of filtrate turbidity and streaming potential during the filtration of carbon black from tetralin through a bed of sand shows that there is a marked decline in filter efficiency at about the time the streaming potential drops to zero. This close correlation between the filter breakthrough curves and the streaming potential measurements suggests the importance of charge neutralization phenomena in the prediction of filter performance.

2. Specific deposits up to the point of charge neutralization, calculated using a filter performance model that included variations in surface charge characteristics of the filter due to particle deposition, were found to be in good agreement with experiment. Thus, for filtration systems in which surface phenomena have a major influence on deposition rates, filter capacities up to the point of charge neutralization can be estimated from knowledge of the zeta potentials of the suspension and filter particles.

3. Filtration breakthrough and streaming potential curves have been calculated based on the concept of charge neutralization and found to be in reasonable agreement with experimental data.

The above results are of particular significance in the design of filters for situations where pressure drop or other considerations dictate the need for shallow but efficient filters. In such situations, the proper choice of a surfactant and a filter medium to obtain desirable surface charge characteristics may be the key to optimization.

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BACKGROUND

Existence of charge at solid-liquid interfaces, the concept of the electrical double layer, and the resulting electrokinetic phenomena in aqueous media have been extensively studied. Fundamental knowledge about charged interfaces has found a major application in the theory of colloid stability (Kruyt, 1952). The charge residing on the surface of colloidal particles in a liquid results in a repulsive energy barrier to collision between particles in the suspension. Therefore, the magnitude of this barrier, and hence of the charge, plays an important role in determining the stability of the colloid.

It is only in recent years that the concepts and theory of colloid stability have been applied to a limited extent in the study of deep bed filtration. The importance of surface forces in the filtration of water through granular media has been pointed out by a number of investigators. Polyelectrolytes have been used to change the filter and colloid surface characteristics and aid in particle attachment to the filter surface (Burns et al., 1969; Yao et al., 1971). The forces due to electrical double layer interactions have been incorporated into the theoretical expressions for the local deposition rate (Tien and Payatakes, 1979). Experimental tests of such expressions in aqueous media, though yielding experimental rates much higher than theoretically predicted for repulsive double layer interactions, are in good qualitative agreement with the theory (Rajagopalan and Tien, 1977; Bowen and Epstein, 1979). Quantitative disagreement is attributed mainly to the difficulty of achieving in practice the ideal conditions of particle shape, surface smoothness, and uniformity of surface potential assumed in the theory.

The major problem in the modeling of deep bed filtration has been in accounting for the changes in various characteristics of the filter that occur due to particle deposition. Though most attempts at modeling deep bed filter performance have concentrated on the changes in deposit morphology (Tien and Payatakes, 1979), some investigators (Heertjes and Lerk, 1967; Wnek et al., 1975) highlighted the effect of changes in filter surface characteristics that occur due to deposition of particles of colloidal size.

Wnek et al. (1975) incorporated the surface charges of particles and filter media into their model by making a charge balance on the filter. Their theoretical treatment assumes that suspension particles carrying a charge opposite to that of the filter grains deposit on the filter medium until the charges are neutralized and the filter eventually acquires the same charge as the suspension particles. Thereafter, repulsive surface forces come into play, hindering further deposition, so that the filter medium ceases to be effective at the time when the charge on the filter medium has been neutralized by the depositing colloid. In a recent study of deposition rates of negatively charged colloidal particles from an aqueous suspension in laminar flow through a smooth parallel plate channel with positively charged walls, Bowen and Epstein (1979) found the rate of deposition to decrease with time and attributed the decrease in part to changes in the double layer at the wall caused by particle deposition. In addition, they found that the effect of double-layer interactions on the deposition rate was greater at larger double-layer thicknesses. This last result is particularly significant in view of the fact that double-layer thicknesses encountered in nonaqueous systems are much larger than those encountered in aqueous systems.

The major differences between aqueous and nonaqueous solutions are in the values of dielectric constant and electrical conductivity. Organic liquids have a very low dielectric constant (~ 2 for hydrocarbons) as compared to water (~ 80) and also have specific conductivities several orders of magnitude lower than those encountered in aqueous solutions. The existence of ions and charged interfaces in hydrocarbons was a topic of controversy until a few decades ago when Minnie and Hermanie (1952) measured the electrophoretic velocity of carbon black in benzene under conditions that excluded the possibility of motion due to other phenomena unrelated to surface charge.

Besides electrophoresis, the phenomena of streaming potential (Kitahara, 1971; Lorenz, 1953; Briggs et al., 1978) and streaming current (Rutgers et al., 1959) have also been used for measurement of zeta-potential in nonaqueous media. Electrophoresis has been the most common method, however, due to its suitability for measurements on the small particles encountered in work on colloids. Colloid stability has been the focus of much interest in nonaqueous systems too, and a number of investigators (Koelmans and Overbeek, 1954; Lewis and Parfitt, 1966; McGown et al., 1965) have found predictions by the DLVO theory (Verwey and Overbeek, 1948) for colloid stability to be in good qualitative agreement with experiments. Trace quantities of water in hydrocarbon media have been found to have a marked effect on colloid stability owing to the tendency of water to adsorb at the interface, thus altering and sometimes even reversing the charge. (Kitahara et al., 1967; Romo, 1966) Parfitt and Peacock (1978) and Kitahara (1973) have reviewed colloid stability in nonaqueous media and have tabulated the sign of the particle charge for a number of systems.

No work is available on the application of these concepts to the study of particle deposition in granular media filters for nonaqueous liquids. The large distances to which the effects of surface charge extend in nonaqueous systems should cause the double layer interactions to have a marked effect on deposition rates. The streaming potential provides a convenient method for monitoring the changes in the average surface potential of a filter bed (Wnek et al., 1975a). The use of this technique in the present work has made it possible to verify the applicability of the concept of filter charge neutralization and its effect on deposition rates, as put forth by Wnek et al. (1975), to the removal of carbon black from tetralin by deposition onto a sand bed.

EXPERIMENTAL

Apparatus

Figure 1 shows a schematic of the apparatus used for filtration and simultaneous measurement of streaming potential. The principal component of the apparatus is the teflon cell assembly in which a porous plug of filter grains can be formed between two perforated platinum disc electrodes. The plug is about 1 cm in diameter and can be up to 4 cm deep. Either a reservoir of particle free liquid, *A*, or a reservoir of particle suspension, *B*, can be connected to the liquid flow system by means of the switching valve *C*. Liquid flow is maintained by pressurizing the reservoir with nitrogen, and the flow rate is measured by a rotameter, *G*. The gas space in reservoir *A* is also connected to a metering bleed valve, *H*, which can be used to obtain continuously falling pressures, as in the measurement of bed ζ-potential, which is described in the section on procedure. The pressure drop across the bed is measured by a differential pressure transducer (Validyne DP7, range ± 5 psid) and the streaming potential by a high impedance electrometer (Keithley 610 C). Filtrate samples, withdrawn from the sampling valve *F*, were analyzed for particle concentration using a turbidimeter.

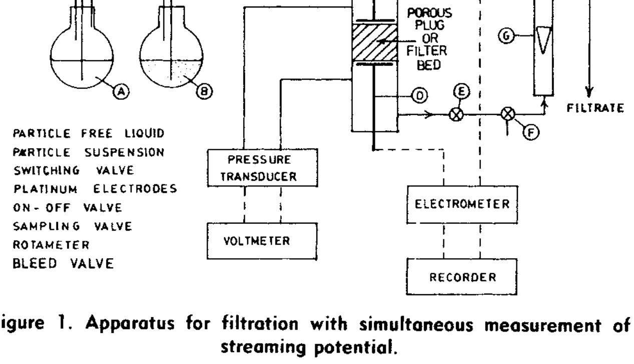

Figure 1. Apparatus for filtration with simultaneous measurement of streaming potential.

TABLE 1. SYSTEM PROPERTIES

Mean diameter of carbon black particles	$2a_p = 0.3\ \mu m$
Mean diameter of filter grains	$2a_F = 81\ \mu m$
Zeta potential of carbon black	$\zeta_p = -57.3$ mV
Density of carbon black	$\rho_p = 2.55 \times 10^3 kg/m^3$
Density of sand	$\rho_F = 1.65 \times 10^3 kg/m^3$
Initial void fraction in the bed	$\phi = 0.35$
Density of tetralin	$\rho = 0.98 \times 10^3 kg/m^3$
Viscosity of tetralin	$\mu = 2.0 \times 10^{-3} kg/(m)(s)$
Dielectric constant of tetralin	$\epsilon = 2.7$
Bulk conductivity of 10mM/L Aerosol *OT* in tetralin	$\sigma_b = 1.0\text{-}1.7 \times 10^{-8}\ ohm^{-1}m^{-1}$

A "Zeta-Meter," a commercial instrument manufactured by Zeta Meter Co., was used for measuring the ζ-potential of the colloidal particles in the feed to the filter. The instrument is provided with a voltage booster to develop the high electric fields necessary to obtain measurable electrophoretic velocities in nonaqueous systems but its application is limited to colorless liquids since visual measurement of the velocity is required. The instrument is designed to give a uniform field in a cylindrical cell. The particle velocity reversed on reversal of the polarity of the electrodes, and was found to be equal in both directions, indicating the absence of significant dielectrophoretic effects in the electrophoresis cell. Lee et al. (1979) used the same equipment for measurements of the electrophoretic mobility of alumina particles in xylene. The strong variation of measured electrophoretic mobility with surfactant concentration observed by them also demonstrates the predominance of electrophoretic effects over dielectrophoretic effects. Zeta potentials are calculated from the electrophoretic mobility by the Huckel formula, which is generally used in nonaqueous systems where the double layer thickness is large compared to the colloidal particle radius. In SI units the formula is:

$$\zeta = \frac{3}{2} EM \cdot \frac{\mu}{\epsilon\epsilon_0} \tag{1}$$

The dielectric constant, ϵ, of the liquid was measured using a chemical oscillometer and a Canon Fenske viscometer was used for determining the bulk viscosity, μ.

Materials

Silica sand, obtained from Wedron Silica, was sieved, washed and dried for use as the filter medium. Carbon-black, obtained from the Sargent Welch Scientific Co., was suspended in tetralin containing 10 m mol/L Aerosol OT for use as feed to the filter. Aerosol OT, a widely used surfactant for both aqueous and nonaqueous media, serves as a dispersant while at the same time increasing the conductivity of tetralin to a value such that measured streaming potentials are stable. Solutions of Aerosol OT in tetralin are colorless, making the system suitable for determination of the particle electrophoretic mobility on the Zeta Meter and the particle concentration using a turbidimeter. Feed slurries of various solid concentrations were prepared by diluting a concentrated stock suspension containing carbon particles that remained in suspension after larger particles had been removed by sedimentation for about ten hours. The mean diameter of the colloidal particles in the feed, obtained from electron micrographs was about 0.3 micron. Some relevant properties of the system under study are listed in Table 1.

Procedure

The sand filter bed was prepared by sedimentation in the teflon cell filled with particle free liquid. Liquid was then streamed through the bed at a constant flow rate until the measured streaming potential reached a steady value. Before starting the filtration run, data required for evaluation of the initial zeta potential of the sand bed was collected as follows. After pressurizing the reservoir A, the system was isolated from the nitrogen supply and a continuously falling liquid flow rate was obtained by bleeding the gas through the metering valve *H*. Plots of streaming potential versus pressure drop across the bed were automatically obtained by connecting the outputs from the pressure transducer and the electrometer to the two axes of an X-Y recorder, a procedure adopted from Parreira (1965). The specific conductivity of the liquid in the filter bed was determined by measuring the electrical resistance between the electrodes, the cell constant having been previously determined using an identical bed with 0.1 N KCl of known

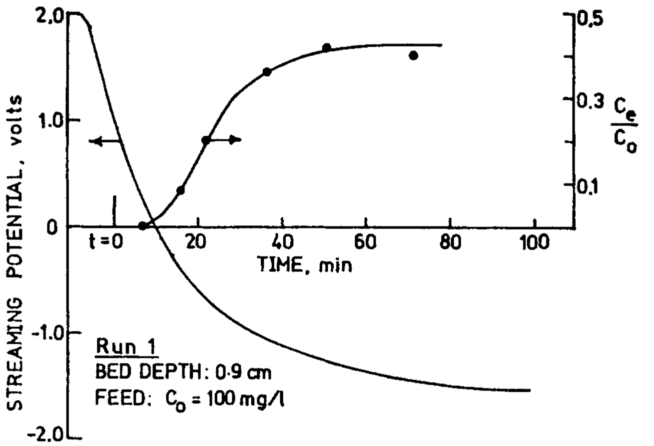

Figure 2. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol OT through a bed of sand.

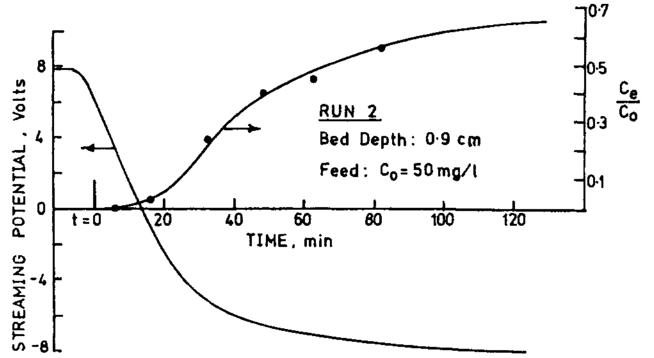

Figure 3. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol OT through a bed of sand.

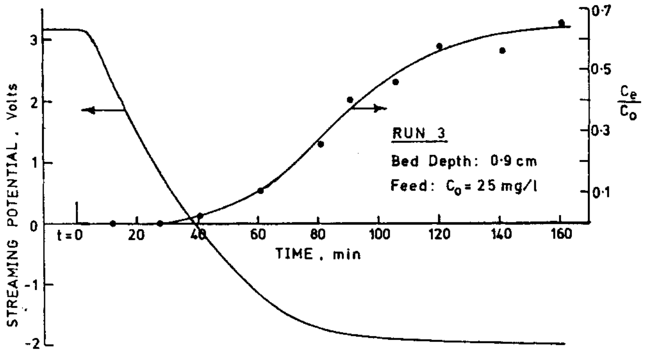

Figure 4. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol *OT* through a bed of sand.

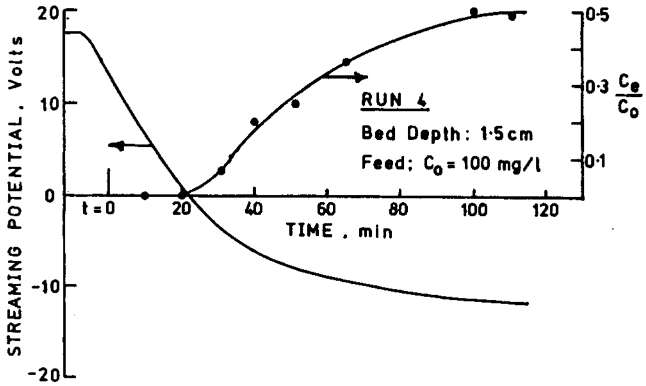

Figure 5. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol *OT* through a bed of sand.

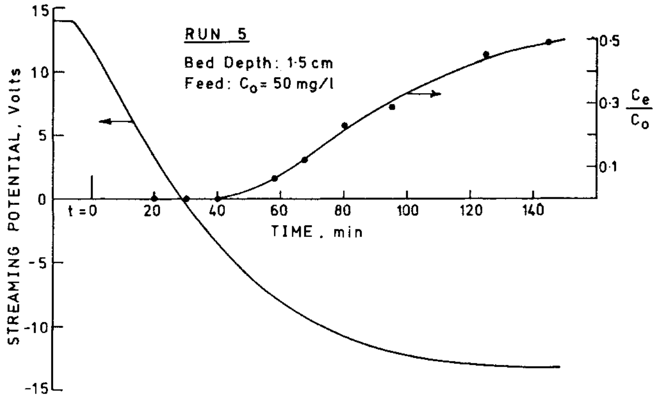

Figure 6. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol *OT* through a bed of sand.

TABLE 2. NEUTRALIZATION OF FILTER CHARGE BY DEPOSITING COLLOID

Run No.	Bed Depth (cm)	Carbon Black Concentration in Feed (kg/m³) ×10³	Initial Zeta Potential of Filter ζ_{Fo}(mV)	Specific Deposit for Neutralization of Filter Charge, $\bar{S}^o$ (kg/m³ filter)		$\frac{\zeta_{Fo}}{\bar{S}^o_{\text{expt}}}$
				Experimental	Theoretical	
1	0.9	100	+21.5	0.73	0.30	29.4
2	0.9	50	+16.9	0.49	0.23	34.5
3	0.9	25	+23.0	0.71	0.32	32.4
4	1.5	100	+26.3	0.91	0.37	28.9
5	1.5	50	+18.8	0.61	0.26	30.8
6	1.5	25	+23.8	0.75	0.33	31.7

specific conductivity. The initial zeta potential of the filter medium is calculated from the slope of the streaming potential versus pressure drop plot using the Smoluchowski equation which reads, in SI units:

$$\zeta = \frac{\mu\sigma}{\epsilon\epsilon_o}\frac{\Delta E}{\Delta P} \tag{2}$$

ϵ_o is the permittivity of vacuum, and the specific conductivity, σ, is that measured in the bed, in order to account for surface conductance effects.

The filtration run was begun by switching the valve *C* to feed the slurry to the filter. Thereafter the streaming potential was recorded as a function of time, and filtrate samples were analyzed for solid concentration in a turbidimeter which had been previously calibrated using suspensions of known solid content. The filtrate flow rate was monitored and kept constant by increasing the nitrogen pressure in the reservoir.

RESULTS AND DISCUSSION

Filtration runs were carried out with bed depths of 0.9 and 1.5 cm, feed concentrations of 25, 50 and 100 mg/L carbon black, and a liquid superficial velocity of 0.01 cm/s. In all cases the plots of streaming potential of the bed and effluent solids concentration as functions of time were similar in shape. Typical results are shown in Figures 2-7. Effluent solid concentrations have been plotted as a fraction of the feed concentration. Measurement of the variation of streaming potential with time, as shown in Figures 2-7, was started at the time the switching valve *C* was switched to allow the suspension to flow into the filter. The origin of the time axis (denoted by "t = 0" in the figures) has, however, been chosen as the time at which the suspension would reach the top of the filter bed if it were in ideal plug flow.

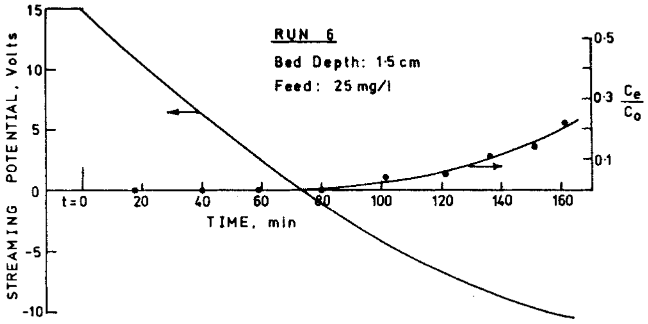

Figure 7. Filtrate turbidity and streaming potential curves for the filtration of carbon black from tetralin + 10 millimoles per liter Aerosol *OT* through a bed of sand.

Concept of Charge Neutralization

The plots show clearly that the filter initially removes carbon black particles efficiently enough to reduce the effluent solids concentration to a negligibly low value. After some time, however, the effluent solid concentration rises to a high value, indicating a decline in the efficiency of particle removal by the filter. Most models of deep bed filtration have explained fall in filter efficiency in terms of changes in the deposit morphology, or mode of deposition. Based on experimental evidence in aqueous media filtration Tien et al. (1979) assume that deposition changes from a "smooth coating mode" to a "blocking mode" at a specific deposit of 0.05 m³ deposit/m³ filter, resulting in a fall in filter efficiency. In our filtration runs the maximum value of specific deposit attained before the filter became inefficient was about 4 × 10^{-4} m³ deposit/m³ filter. This value is too low for changes in deposit morphology to be significant and, in fact, lies in the range where the theory predicts an increase in filter efficiency during the smooth coating mode of deposition. The observed phenomenon of filter "clogging" at low specific deposits cannot therefore be explained in terms of changes in deposit morphology above and the consequent changes in the flow field within the bed.

A plausible explanation for the phenomenon is provided by the plot of streaming potential measured simultaneously during the filtration run. The streaming potential versus time plot in Figures 2-7 shows that during the process of filtration the average charge on the filter drops from an initial positive value to zero and eventually attains a constant negative value. The most significant observation from the experimental results shown in Figure 2 and the subsequent figures is the fact that for all the runs the effluent solid concentration rises sharply at about the time the streaming potential reaches zero. This is in agreement with the mechanism for filtration proposed by Wnek et al. (1975). Deposition of negatively charged carbon black on the initially positively charged sand neutralizes the charge on the sand, causing it to eventually attain a constant negative value. This change in the colloid-collector double layer interactions from attractive to repulsive in nature causes a sharp decline in the deposition rate. As mentioned earlier, similar variations in deposition rate were observed in flow of an aqueous suspension through a parallel plate channel by Bowen and Epstein (1979).

Since unfavorable electrokinetic interactions cause the filter to become inefficient after its streaming potential reverses sign, the amount of colloids removed up to the time when the average filter charge reduces to zero is approximately the capacity of the filter. If the neutralization of filter charge is occurring due to particle deposition, the initial surface condition, or charge, of the filter should determine its capacity. That this is indeed the case is shown in Table 2, where a striking correlation is evident between the initial zeta potential ζ_{Fo}, of the filter for each run and the experimental specific deposit, $\bar{S}^o_{\text{expt}}$, up to the point of exact neutralization of filter medium charge. The variation in the initial zeta potential for the various runs was probably due to trace amounts of water in the system. The values of $\bar{S}^o_{\text{expt}}$ shown in the table are a measure of the filter capacity since the filter is inefficient after charge reversed. The strong dependence of the filter capacity on the initial filter charge is further emphasized by the fact that the ratio $\zeta_{Fo} \div \bar{S}^o_{\text{expt}}$ is nearly constant for the various runs. It should, therefore, be possible to estimate the capacity of the filter from knowledge of its initial zeta potential, and that of the depositing colloid.

The results of a theoretical prediction of filter capacity by a very simplistic charge neutralization approach are shown in the table. The values were calculated assuming that each depositing particle neutralizes a quantity of charge equal to its own charge. Then, the specific deposit for charge neutralization is given by

$$\bar{S}^o = \begin{pmatrix}\text{No. of filter grains}\\ \text{per unit vol. of filter}\end{pmatrix} \times \begin{pmatrix}\text{Mass deposited}\\ \text{per filter grain}\end{pmatrix}$$

$$= \frac{3(1-\phi)}{4\pi a_F^3} \times \frac{4}{3}\pi a_p^3 \rho_p N^o$$

$$= \frac{a_p^3}{a_F}\rho_p(1-\phi)N^o \tag{3}$$

N^o, the number of particles deposited on each filter grain, is given by

$$N^o = \frac{q_{Fo}}{q_p} \tag{4}$$

where the initial charge q_{Fo} on a filter particle is calculated from the condenser formula valid for large particle diameters:

$$q_{Fo} = 4\pi a_F^2\, \epsilon\epsilon_o \kappa \zeta_{Fo} \tag{5}$$

The charge on a carbon black particle was calculated using the equation

$$q_p = 4\pi a_p \epsilon\epsilon_o \zeta_p (1 + \kappa a_p) \tag{6}$$

Equations 5 and 6 are both written in SI units. Equation 4, though strictly valid only for $\zeta_p < 25$ mV, does not introduce much error for larger values of ζ_p when the double-layer thickness is also large. The reciprocal double-layer thickness κ was calculated using the expression

$$\kappa = \left(\frac{2e^2 n}{\epsilon\epsilon_o kT}\right)^{1/2} \tag{7}$$

The ionic concentration, n, is obtained as

$$n = \frac{\sigma F}{e\Lambda_o} \tag{8}$$

and the equivalent conductance for solutions of Aerosol OT in tetralin calculated using Walden's Law (Kitahara et al., 1967).

$$\Lambda_{o(nonaq)}\mu_{(nonaq)} = \Lambda_{o(aq)}\mu_{(aq)} \tag{9}$$

For all the experiments conducted the value of double-layer thickness $1/\kappa$ calculated as above was found to be approximately 1.2 microns. Comparison of the experimental and theoretical quantities of carbon black required for filter charge neutralization shows agreement within a factor of about 2. Considering the extremely simplistic picture adopted for the neutralization phenomenon, even this close an agreement is encouraging and suggests that at least order of magnitude estimates of filter capacity can be made from knowledge of zeta potentials of the filter and suspension particles.

It is interesting to note here that the theoretical value of the filter capacity $\bar{S}^o$ is consistently lower than the experimental value. A number of factors contribute to this deviation. The neutralizing effect of a depositing colloid may be less than the total charge carried by it. Also, filtration by mechanisms like straining, which do not contribute to the neutralization of filter charge, would tend to give a larger experimental capacity compared to the theoretical, calculated based on a charge balance. These points are discussed in detail in the following sections.

Model

A model for the prediction of variation in filter performance due to changes in surface charge was presented by Wnek and coworkers (1975). The model uses a charge balance on the filter to account for the neutralization of filter charge by the depositing colloid. For a system like the one under study where charge effects are significant, a charge balance on a filter grain yields (Wnek et al., 1975)

$$\frac{\partial q_F}{\partial t} = \hat{q}_p J \tag{10}$$

where J is the deposition rate of colloidal particles on a filter grain. J varies with time and along the bed depth due to variations in the particle concentration in the liquid and the surface characteristics of the filter. $\hat{q}_p$ is the "effective charge" of a colloidal particle.

A mass balance on the suspension yields (Wnek et al., 1975)

$$\frac{\partial C}{\partial z} = -\frac{3(1-\phi)}{4\pi a_F^3 V} J \tag{11}$$

The deposition rate, J, was calculated as the sum of theoretically predicted individual deposition rates based on the mechanisms of Brownian Diffusion (Levich, 1962) and Interception and Sedimentation (Spielman and Fitzpatrick, 1973). Rajagopalan and Tien (1979) have listed various other models available for calculation of the initial rate, and their relative merits. The effect of surface forces was incorporated into these expressions by treating the surface of the filter as possessing first order intermediate reaction kinetics for which the rate constant was a function of the stability ratio of colloid chemistry. An expression of the form

$$J = \frac{J^o}{1 + \dfrac{J^o}{J_S^o}} \tag{12}$$

was derived, which relates the deposition rate J to the values under conditions of favorable surface interaction, J^o, and repulsive surface interactions controlling, J_S^o, for both of which theoretical expressions are available.

Effective Charge on a Colloidal Particle

In order to obtain a model free of any empirical factors Wnek et al. (1975) assumed the effective charge of a colloidal particle in Eq. 10 to be equal to its actual surface charge q_p. This is not a justifiable assumption since the change in surface characteristics of the filter grain is not in reality a simple neutralization of electrostatic charges on the grain surface by depositing charges. On the other hand, the phenomenon is not amenable to an exact theoretical analysis at this stage owing to the complex nature of the charge distribution on the solid surfaces, coupled with the fact that we are not dealing with point charges. It becomes necessary, therefore, to use the charge balance Eq. 10 with the introduction of a degree of empiricism into the model in the form of an "effectiveness factor", f, to take into account the complex nature of interactions involved in the charge neutralization process. Thus,

$$\frac{\partial q_F}{\partial t} = f q_p J \tag{13}$$

The factor f is a measure of how effective a colloidal particle is in neutralizing the filter charge. If, on deposition, it neutralizes a charge equal to its own charge, then $f = 1$. A physical significance may be given to the factor f by realizing that fq_p represents the total charge on the area of the filter effectively excluded from further deposition by a single deposited particle. Generally, in systems where changes in surface characteristics of the filter are important it is reasonable to expect a value of $f < 1$ since the deposited particle physically covers only $4\,a_p^2$ of the filter area.

Estimation of Specific Deposit

In order to compare the theoretical predictions of the above model with experiment, Eqs. 11 and 13 were solved, with J given by Eq. 12, and the boundary conditions

$$t = 0,\ z \geq 0,\ q_F = q_{F0} \tag{14}$$

$$t = 0,\ z \geq 0,\ C = 0 \tag{15}$$

$$t \geq 0,\ z = 0,\ C = C_0 \tag{16}$$

The pair of hyperbolic partial differential Eqs. 11 and 13 has characteristics along the axes. The equations can therefore be reduced to the ordinary differential equations

$$\frac{dC}{dz} = -\frac{3(1-\phi)}{4\pi a_F^3 V} J \tag{17}$$

along t = constant, and

$$\frac{dq_F}{dt} = f q_p J \tag{18}$$

along z = constant

Equations 17 and 18 were solved simultaneously using second order Runge Kutta algorithms to obtain the variation of filter charge and suspension concentration with time and along the bed. The factor f used in the computations had to be determined from the experimental results. Since the streaming potential of the filter bed is a direct consequence of the electrical double layer, it is convenient to use its variation with time in order to determine the effective charge of depositing particles. The value of the "effectiveness factor" f was determined from one of the experimental runs (Run No. 5) by matching the theoretical and experimental times for the streaming potential to reduce to zero. The theoretical value of streaming potential at any time was calculated based on an average charge in the filter which is given by

$$\bar{q}_F = \frac{1}{L}\int_0^L q_F dz \tag{19}$$

The streaming potential was assumed proportional to the average charge, so that the theoretical streaming potential at any time could be calculated from

$$\Delta E = \left(\frac{\bar{q}_F}{q_{F0}}\right)\Delta E_0 \tag{20}$$

where the initial filter charge is obtained from Eq. 5. Equation 20 can be derived by integrating the Smoluchowski equation over the length of the bed at any time t, under the assumption of a constant pressure drop.

The factor f is expected to be a function of the system properties including shape and size of the filter grains and colloidal particles. The use of the experimental results of Run No. 5 to evaluate the value of f for the system under study yielded a value of $f = 0.394$, which was used thereafter in the prediction of filter capacities for the other runs. The filter grains and colloidal particles were assumed spherical and uniform in size, with diameters equal to the mean values listed in Table 1. Due to the small values of specific deposit involved, the void fraction ϕ was assumed to be constant and equal to its initial value. A value of 10^{-19} Joules was used for the Hamaker constant.

Theoretical filter capacities based on the above model were calculated as follows. The number of particles deposited at any time, t, on a filter grain at a depth z in the filter is given by

$$N = \int_0^t J dt \tag{21}$$

The average specific deposit in the filter at any time can then be calculated as

$$\bar{S} = \left(\frac{a_p}{a_F}\right)^3 \rho_p \frac{1-\phi}{L}\int_0^L N dz \tag{22}$$

The filter capacity was taken to be the average specific deposit at the time when the theoretical streaming potential of the filter dropped to zero. Theoretical filter capacities computed as above are compared with the experimental value in Table 3.

TABLE 3. COMPARISON OF THE MODIFIED MODEL OF WNEK ET AL. WITH EXPERIMENT (f = 0.394 FROM RUN 5)

	Average Specific Deposit at Zero Streaming Potential $\bar{S}^0$ (kg/m³ filter)			Time at Zero Streaming Potential $t_{\Delta E=0}$ (mins)		
Run No.	Experimental	Theoretical	% Deviation	Experimental	Theoretical	% Deviation
1	0.73	0.692	−5.2%	10	10.2	+2.0%
2	0.49	0.469	−4.3%	14	13.7	−2.1%
3	0.71	0.793	+11.7%	39	46.4	+19.0%
4	0.91	0.833	−8.5%	22	21.5	−2.3%
5	0.61	0.570	−6.6%			
6	0.75	0.753	+0.4%	73	77.2	+5.8%
	Average Deviation −2.1%			Average Deviation +4.5%		

The agreement between the theoretical and experimental values for the various runs is very good, indicating the suitability of the above model for prediction of filter capacities in the system under study. Table 3 also shows a comparison of the experimental times for the streaming potential to drop to zero with the theoretically calculated values. These values too are in good agreement. It is apparent from the results that the assumption of a constant "effectiveness factor" f is valid.

Filter Breakthrough Curves

Filter breakthrough and streaming potential profiles calculated by the above theory were plotted for the various runs. The results for run Nos. 3 and 5 are shown in Figures 8 and 9 respectively. The plots for the other runs are similar. It is obvious that, in addition to its ability to predict reasonably well the specific deposit up to the neutralization of filter charge, the

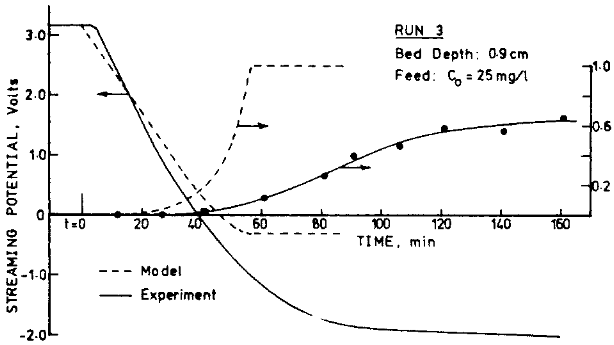

Figure 8. A comparison of experimental filter breakthrough and streaming potential curves with theoretical curves calculated using the modified model of Wnek et al.

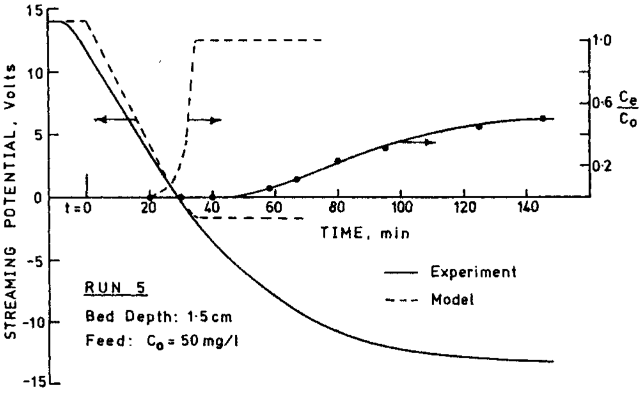

Figure 9. A comparison of experimental filter breakthrough and streaming potential curves with theoretical curves calculated using the modified model of Wnek et al.

theory is in agreement with experiment in its prediction of a reduction of filter efficiency at about the time the streaming potential drops to zero. However, the effluent solids concentration as a function of time shows only qualitative agreement with the experiment. The experimental values of effluent concentration never reach a value equal to the feed concentration, contrary to the theoretical prediction. Actually, the concentration seems to reach a plateau value and the filter continues to remove particles even after charge reversal, though at a much slower rate.

This is not surprising when one considers that quantitative agreement between theory and experiment in phenomena which are strongly influenced by surface interactions is rare at this stage, even in situations far simpler than deep bed filtration. In a situation as complicated as a packed bed, deposition occurs by a host of mechanisms, including straining, dendrite formation (Tien et al., 1977) etc., a number of which may not be affected by changes in the electrical double layer. Besides, one has also to bear in mind the factors of surface roughness and possible nonuniformity of the surface potential distribution within the bed. It is reasonable to assume, in systems like the present one where charge effects are important, that changes in filter performance occur due to variation of filter charge as well as deposit morphology; the first effect being felt over a much smaller time scale than the latter. Thus, if our filtration runs had been continued long enough, changes in deposit morphology could have been expected to cause another fall in filter efficiency and the effluent concentration would eventually have reached the feed value.

Thus, an improvement in the prediction of effluent solids concentration profiles by the model of Wnek and co-workers can be achieved by modifying it to account for deposition by "charge independent" mechanisms and by adopting a more realistic view of the manner in which charge neutralization affects deposition rates. At the present state of the art in deep bed filtration, such modifications entail an increase in the degree of empiricism of the model. Empirical improvements of a model are not totally useless in that they help in opening new areas for future theoretical development. Some modifications of the model in order for it to more closely represent a real filtration system are discussed in the following sections.

The basis of the new model is still the concept of charge neutralization, since the results based on the modified model of Wnek et al. show the importance of charge neutralization effects in nonaqueous systems. Geometrical effects have been suspected to overshadow charge neutralization effects in some studies of deposition in packed beds and rotating discs (Rajagopalan, 1980; Rajagopalan and Chu, 1980). These results on *aqueous media* are characterized by theoretical capacities based on a charge balance (with $f = 1$) which are higher than experimental capacities. In other words, the experimentally determined value of f is greater than one for these systems. A simple charge balance on the data of Bowen and Epstein (1979) also yield a value of f greater than one. Such a large effectiveness of a depositing colloid in preventing further deposition is indicative of the presence of significant geometric effects in addition to the charge neutralization effects. However, in the present study on *nonaqueous media*, values of f less than one have been found, suggesting the adequacy of the charge neutralization concept in explaining changes in deposition rate for nonaqueous systems. Owing to the large double layer thickness in nonaqueous media, the charge neutralization effect of a deposited particle extends to a far greater distance around it than in aqueous media, resulting in a decreased significance of the role played by geometrical effects in reducing the deposition rate.

Deposition rates

Based on the discussion of the experimental results, the total deposition rate J on a filter grain is assumed to be the sum of two independent deposition rates:

(a) A rate J_d which depends on the nature of the double layer around the filter grain, and also contributes to changes in the double layer. This rate eventually drops to zero.

(b) A rate J_c which is independent of the surface characteristics of the colloid and filter grains and also does not in any way contribute to changes in the electrical double layer. Thus, J_c accounts for particle deposition by straining and by dendrite formation on initially deposited particles (Tien et al., 1977). This rate may be taken as a constant during the filter operating times under consideration since, as discussed before, the time scale for variations in J_c is much larger than that for J_d. For the purpose of the model it is necessary to estimate the deposition rates J_d and J_c.

The rate J_d varies with the amount of deposition. Its initial value on the clean filter, J_{do}, was estimated considering deposition by Brownian diffusion, interception and sedimentation as in (Wnek et al., 1975). A value of $A = 10^{-19}$ Joules was used for the Hamaker constant. To account for the change in J_d due to change in surface characteristics Wnek et al. (1975) used the theory of colloid stability for the colloid particle-filter grain interactions, with the variation of filter grain charge given by Eq. 10. However, Eq. 12 does not realistically account for the reduction in rate on a "loaded" filter grain.

A more realistic approach is to assume that each particle depositing on the filter surface affects the electrical double layer in its neighborhood in such a manner that it excludes an area A_e of the filter from further deposition. A similar approach was suggested by Bowen and Epstein (1979). Assuming the deposition rate to be proportional to the area unoccupied, one may write

$$J_d = J_{do}\left(1 - \frac{N_d A_e}{A_F}\right) \tag{23}$$

where N_d is the total number of particles deposited by the processes contributing to charge neutralization, and A_F is the total area of a filter grain.

Now, if N_{df} is the number of particles required to completely exclude a filter grain from further charge dependent deposition,

$$A_F = N_{df} A_e \tag{24}$$

Equation 23 may then be written as

$$J_d = J_{do}\left(1 - \frac{N_d}{N_{df}}\right) \tag{25}$$

The charge on each filter grain falls from an initial value q_{Fo}, reverses sign and eventually reaches a value q_{Ff}, whereafter no further deposition occurs. Total monolayer coverage of the filter surface is not necessary before repulsive double layer forces exclude enough filter area from further deposition by the processes accounted for by J_{do}. The experiments of Bowen and Epstein (1979) on deposition rates in parallel plate channels showed less than 10% area coverage before saturation. Since the change in the filter grain charge is assumed proportional to the number of particles deposited, one may write

$$\frac{N_d}{N_{df}} = \frac{q_F - q_{Fo}}{q_{Ff} - q_{Fo}} \tag{26}$$

where q_F is the charge at a time when N particles have deposited. Using Eq. 26, Eq. 25 reduces to

$$J_d = J_{do}\frac{q_F - q_{Ff}}{q_{Fo} - q_{Ff}} \tag{27}$$

Filter Performance

The equations representing the filter bed can be written as

$$\frac{\partial q_F}{\partial t} = f q_p J_d \tag{28}$$

$$\frac{\partial C}{\partial z} = -(\lambda_c + \lambda_d)C \tag{29}$$

Table 4. Estimates of Parameters for the Proposed Model

Run No.	Bed Depth (cm)	Feed Conc. (kg/m^3) $\times 10^3$	Final Filter Coefficient λ_c(m^{-1})	Final Zeta Potential of Filter ζ_{Ff}(mV)	Particle Charge "Effectiveness Factor" f
1	0.9	100	95.1	−16.7	0.531
2	0.9	50	47.9	−17.3	0.429
3	0.9	25	49.6	−14.6	0.544
4	1.5	100	46.2	−18.0	0.476
5	1.5	50	46.2	−17.8	0.479
6	1.5	25		−16.7	0.517

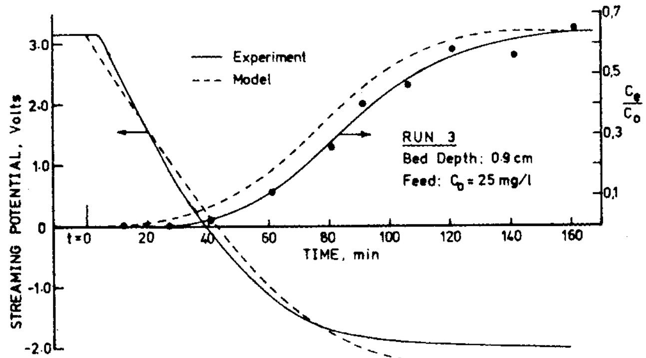

Figure 10. A comparison of experimental filter breakthrough and streaming potential curves with theoretical curves calculated using the proposed model.

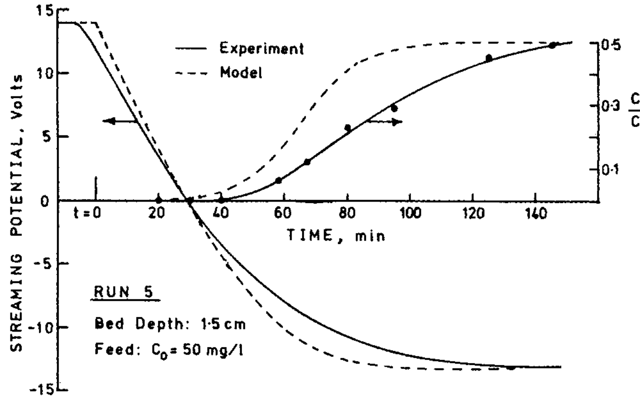

Figure 11. A comparison of experimental filter breakthrough and streaming potential curves with theoretical curves calculated using the proposed model.

Table 5. Comparison of the Proposed Model with Experiment (Model Parameters from Run 5 (Table 4)

Run No.	Average Specific Deposit at Zero Streaming Potential $\bar{S}^o$ (kg/m^3 filter) Experimental	Theoretical	% Deviation	Time at Zero Streaming Potential $t_{\Delta E=0}$ (mins) Experimental	Theoretical	% Deviation
1	0.73	0.671	−8.1%	10	9.5	−5.0%
2	0.49	0.451	−8.0%	14	12.6	−10.0%
3	0.71	0.768	+8.2%	39	43.2	+10.8%
4	0.91	0.872	−4.2%	22	22.0	0
5	0.61	0.577	−5.4%			
6	0.75	0.782	+4.3%	73	78.5	+7.5%
	Average Deviation −2.2%			Average Deviation +0.7%		

where Eq. 29 is the same as Eq. 11 but written in terms of filter coefficients which are related to the corresponding local deposition rates by the expression (Wnek et al., 1975)

$$\lambda = \frac{3(1-\phi)}{4\pi a_F^3 V C} J \qquad (30)$$

Equation 28 indicates that the filter charge will no longer change once J_d is zero, though particle retention will continue to increase due to a finite J_c. This is because particles deposited by the mechanisms like straining and dendrite formation (accounted for by J_c) are unlikely to have an effect on the nature of the electrical double layer near the surface of the filter grain available for deposition by the processes accounted for by J_{do}. Since λ_c in Eq. 29 remains constant even after λ_d drops to zero, Eq. 29 accounts for the fact that the effluent concentration never attains a value equal to the feed value, as observed in Figures 2-7. Equations 28 and 29 were solved with the boundary and initial conditions (Eqs. 14-16) by the procedure described earlier to obtain streaming potential and concentration profiles. The values of λ_c, q_{Ff} and f are needed to carry out the computations and have to be determined from the experimental results. f was determined by the procedure described earlier. The final filter grain charge q_{Ff} can be estimated from the final streaming potential of the filter bed, using Eq. 20. The filter coefficient, λ_c, is estimated as follows. At sufficiently large values of time when λ_d has reduced to zero, the filter performance is governed solely by the "charge independent" filter coefficient, λ_c, which has been assumed constant. Thus, the effluent solids concentration at large times is given by

$$C_f = C_o \exp(-\lambda_c L) \qquad (31)$$

Since C_f can be estimated from the experimental results, λ_c can be obtained from the above equation.

It is reasonable to assume that the values of f, λ_c and q_{Ff} should not vary with feed concentration and bed depth for a particular system under study. Table 4 shows this to be reasonably true for the data available. The value of λ_c for run 6 was not calculated since the effluent concentration had not reached a steady value when the filtration was stopped. The exceptionally large value of λ_c for run 1 is not an effect of the feed concentration, as is obvious from the value of λ_c for run 4 which has the same feed concentration. The reason for the large deviation in run 1 is not known, but is probably due to some error in turbidimeter calibration.

Filter breakthrough and streaming potential curves were obtained for the various runs using the parameter values estimated from run 5 and were found to be in reasonably good agreement with the experiments. Figures 10 and 11 show the results for runs 3 and 5 respectively. A similar agreement between theory and experiment was found for the other runs. Table 5 compares theoretical values of the filter capacity up to zero streaming potential and the time for the streaming potential to fall to zero, with the experimental values. The agreement is found to be good, indicating the constancy of the factor f for the various runs.

The main factor affecting the shape of the theoretical effluent concentration curves is the mode of variation of the deposition rate J_d (Eq. 23). Though the model is not totally free of empirically determined factors, the reasonably good agreement between the model and experiment lends support to the arguments leading to the equations used in the model.

ACKNOWLEDGMENT

This study was supported by the National Science Foundation under grant number CPE-79-19189 and ENG-76-06495.

NOTATION

a_F = radius of a filter grain, m
a_p = radius of a colloidal particle, m
A_c = area of filter surface excluded from further deposi-

tion by the deposition of a colloidal particle, m^2

A_F = total surface area of a filter grain, m^2
C = solid concentration in suspension, particles/m^3
C_e = solid concentration in filtrate, particles/m^3
C_f = final value of effluent solid concentration, particle/m^3
C_o = solid concentration in feed, particles/m^3
e = electronic charge = 1.602×10^{-19} coulombs
EM = electrophoretic mobility, (m/s)/(V/m)
ΔE = streaming potential, V
ΔE_o = initial streaming potential, V
f = effectiveness factor for charge neutralization by a depositing particle, dimensionless
F = Faraday constant = 96,500 coul/(gm-equiv.)
J = total deposition rate on a filter grain, particles/s
J_c = charge independent deposition rate on a filter grain, particles/s
J_d = charge dependent deposition rate on a filter grain, particles/s
J_{do} = charge dependent deposition rate on a clean filter grain, particles/s
J^o = deposition rate in the absence of electrical double layer interaction, particles/s
J_s^o = deposition rate for surface forces controlling, particles/s
k = Boltzman constant = 1.3805×10^{-23} J/°K
L = depth of the filter bed, m
n = ionic concentration, ions/m^3
N = number of particles deposited on a filter grain
N_d = number of particles deposited on a filter grain by charge dependent mechanisms
N_{df} = maximum number of particles that can be deposited on a filter grain by charge dependent mechanisms
N^o = number of colloidal particles required to neutralize the charge on a filter grain
ΔP = pressure drop, N/m^2
q_F = surface charge on a filter grain, coul
$\bar{q}_F$ = charge per filter grain, averaged over the filter bed, coul
q_{Ff} = final charge per filter grain, coul
q_{Fo} = initial charge on a filter grain, coul
$\hat{q}_{Fo}$ = initial charge density on a filter grain, coul/m^2
q_p = charge on a suspension particle, coul
$\bar{S}$ = average specific deposit in the filter bed, kg/m^3 filter
$\bar{S}^o$ = average specific deposit at charge neutralization, kg/m^3 filter
t = time, s
T = temperature, °K
V = liquid superficial velocity, m/s
z = coordinate along filter depth, m

Greek Letters

ϵ = dielectric constant, dimensionless
ϵ_o = permittivity of vacuum = 8.805×10^{-12} $coul^2\ N^{-1}\ m^{-2}$
ζ = zeta potential, V
ζ_{Ff} = final zeta potential of the filter, V
ζ_{Fo} = initial zeta potential of the filter, V
ζ_p = zeta potential of a colloidal particle, V
κ = reciprocal double-layer thickness, m^{-1}
λ = filter coefficient corresponding to total deposition rate = $\lambda_c + \lambda_d$, m^{-1}
λ_c = filter coefficient corresponding to charge independent deposition rate, m^{-1}
λ_d = filter coefficient corresponding to charge dependent deposition rate, m^{-1}
Λ_o = equivalent conductance at infinite dilution, $ohm^{-1}\ m^2\ (gm\text{-}equiv.)^{-1}$
μ = liquid bulk viscosity, kg/(m) (s)
ρ_p = density of colloidal particles, kg/m^3
σ = specific conductivity of the liquid in the bed, ohm^{-1}, m^{-1}
ϕ = void fraction in the filter bed

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Manuscript received April 28, 1980; revision received November 24, and accepted December 10, 1980.

Analysis of Particulate Removal in Venturi Scrubbers—Effect of Operating Variables on Performance

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A theoretical model of venturi scrubber performance consisting of the governing differential equations for momentum, material, and energy exchange is solved using a Milne fourth-order predictor—Hamming corrector numerical integration procedure. The model provides for specification of the operating variables, including scrubber geometry, throat gas velocity, liquid to gas loading ratio, and collector droplet and particle-size distributions. Liquid loading ratio and gas velocity are shown to be the two most important operating variables, while the dispersity of the droplet size distribution only slightly affects collection efficiency over the operating range normally encountered. Location of the liquid injection site and length of the throat are also important design considerations. While particle collection in venturi scrubbers has typically been assumed to occur in the potential flow regime, the present results show that collection can also occur under conditions corresponding to viscous flow about the collecting droplets. The transition Reynolds number used to change from the potential to viscous flow equations affects the predicted performance.

SCOPE

High energy scrubbers have long been useful for particulate removal and control, especially in the metallurgical and chemical process industries. Despite this generally successful use of the venturi, reliable design criteria are generally not available; there is considerable reliance on prior experience and pilot scale tests. The primary objective of the present study was to develop a realistic computer model that predicts particulate collection efficiency in a venturi scrubber.

The model requires specification of liquid flow rate, gas flow rate, venturi geometry, entering conditions of the liquid and gas, and particle size distribution. The dust particles are assumed to move along the gas streamlines and are removed by inertial impaction, interception, and other secondary mechanisms. The equations of motion of the collector droplets and the mass balances for the droplet and dust phases are numerically solved. The flux of the dust at the exit of the venturi is integrated over the cross-sectional area and compared to the dust input to determine the collection efficiency. The model is evaluated in computer simulations by varying the important parameters and comparing the results obtained with some of the limited data available in the literature.

CONCLUSIONS AND SIGNIFICANCE

In this research, a simulation model was developed to more realistically model venturi scrubber performance. In particular, allowance was made for a complete description of all operating variables, scrubber geometry, and parameters relating to properties of the droplet and dust size distributions. Following are specific conclusions pertaining to the results of this investigation.

1. The data with which to validate models describing venturi scrubber performance are extremely limited. The present model was compared to the data presented by Ekman and Johnstone (1951) and Brink and Contant (1958). In most cases, experimentally determined collection efficiencies were 10-35% of theoretical. It has been pointed out that the data of Ekman and Johnstone may not be typical of industrial scale venturi scrubbers due to the small size of their laboratory scale venturi (cf., Boll, 1973). Also, the data of Brink and Contant may have been taken under conditions where poor spray coverage in the throat section existed. In any event, the predic-

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0001-1541-81-4913-0984-$2.00.