Advances in Deep Bed Filtration

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The main problems which are relevant to a fundamental understanding of deep bed filtration are the nature of and the conditions leading to the retention of particles throughout a filter bed, the change of the filter media structure due to deposition, and its effect on filter performance. The purpose of this review is to discuss in a systematic manner the more recent advances in the investigation of all these problems. A reasonably complete understanding of the pertinent phenomena is essential for the establishment of a comprehensive deep bed filtration theory which can be used as a basis of rational design.

SCOPE

Deep bed filtration is an engineering practice of long standing and is usually employed to remove fine and colloidal particulates from dilute liquid suspensions. This process is intrinsically transient, as deposited material changes both the geometry of the interstitial space of the filter and the nature of the collector (filter grain) surfaces. These changes are reflected in the variations of the filtration efficiency and of the pressure drop. Typically, an initial increase of the efficiency is observed followed by a monotonic decrease. The major phenomena that need to be understood are the mechanisms through which particles are deposited on the collector surfaces, the deposit morphology and its evolution, the conditions under which deposited particles may become reentrained, the effects of electrolytes and polyelectrolytes on particle capture and reentrainment, and ultimately the changes of the filtration efficiency and pressure drop across the filter bed due to particle deposition and reentrainment. Accordingly, deep bed filtration is often treated empirically, without benefit of a thorough understanding of the phenomena involved. A review of recent advances in the study and modeling of depth filtration is presented here.

CONCLUSIONS AND SIGNIFICANCE

Recent advances in the study of deep bed filtration have shown that in spite of the inherent complexities of the process, the problem can be studied fruitfully based on fundamental consideration. Resurgent research efforts of the past two decades in this relatively old engineering practice have already yielded important results of practical significance. In general, the study of deep bed filtration has been made using two different yet often complementary approaches: phenomenological and theoretical. The phenomenological approach describes the dynamic behavior of deep bed filters with the use of a set of partial differential equations and characterizes the filtration mechanism by means of several model parameters the values of which, for a given application, can be obtained from appropriate bench scale experiments. The solution of these equations provides the basis of design, scale-up and optimization. Although this approach affords only limited insight about the physical process of filtration and is therefore not entirely satisfactory from scientific viewpoints, it represents a practical and rational methodology to the design of industrial scale depth filters, provided the bench scale experiments from which the relevant parameters are obtained can be conducted with sufficient accuracy.

The ultimate objective of the theoretical approach is to derive mathematical formulations which quantitatively describe the complete dynamics of deep bed filtration, taking into account the complexed interaction between the filter grains and liquid solution particles to be removed as well as the effects of operating and system variables including the addition and presence of polyelectrolytes and electrolytes. At this stage of its development, it has been possible to predict with reasonable accuracy the efficiency of clean filters. Furthermore, limited success in predicting the transient behavior of filtration has also been achieved. The role of polyelectrolytes is understood from a qualitative point of view. Much work remains to be done before the results of these theoretical analyses can be translated directly to the practical design calculation.

This review examines both the phenomenological and theoretical studies conducted during recent years. It reveals that deep bed filtration is a challenging and fascinating subject of study and warrants further attention and effort from scientists and engineers.

Deep bed filtration may be defined as a process in which a fluid suspension is passed through a filter composed of granular or fibrous materials for the purpose of removing the fine and/or colloidal particles present in the fluid. The fluid stream may be either liquid or gas. Because of space limitations, the attention of this review will be restricted to liquid systems.

The flow of a suspension through a packed bed of grains (sand, garnet, anthracite, active carbon, etc.) or fibers results in the deposition of the suspended particles on the grain or fiber surfaces and ultimately the separation of these particles from the fluid. The particles penetrate into the porous medium and deposit at various depths. This is the reason for which this type of filtration is called deep bed (or depth) filtration and should not be confused with the more familiar cake filtration (at least to chemical engineers). In contrast to cake filtration which is commonly encountered in chemical process industries and used to separate particles from relatively dense suspensions with solid volume fractions exceeding say, 2 000 ppm, deep bed filtration is the most effective and economical in treating large quantities of liquids containing relatively low solid volume fractions (below 500 ppm) of particles with fine or colloidal size (less than 30 μm). A most notable application of depth filtration is the use of sand filters for the treatment of water and wastewater. In addition, several important chemical processes rely on this technique of solid-liquid separation for their smooth and trouble free operation. For instance, deep bed filters are used to remove solids from feedstocks to catalytic reactors in order to prevent an increase of pressure drop and operating temperature and the need for frequent backwashing.

The rational design of a depth filtration process must be based on reliable predictions of the histories of the effluent concentration and of the pressure drop required to maintain a given throughput. Furthermore, since filter cleaning is required once a filter becomes sufficiently clogged, understanding of the mechanisms of filter clogging and development of effective technology for filter cleaning are equally important to a proper process design.

Depth filtration theories are therefore mainly concerned with the elucidation and understanding of these phenomena and ultimately their quantitative prediction. Broadly speaking, these theories can be grouped into two categories: macroscopic and microscopic. The macroscopic studies are aimed at the phenomenological description of the filtration process, the prediction of its dynamic behavior, and the development of methodology and techniques for design, calculation, and optimization. Microscopic theories in deep bed filtration are intended to provide information and insight about the mechanisms of particle deposition, the conditions under which deposition may be facilitated, and the effect of deposition on the structure of filter media and characteristics of filter grains. Separate reviews on these topics will be presented in the present paper.

PHENOMENOLOGICAL THEORIES

Macroscopic Description of Deep Bed Filters

The fundamental equations describing the particle retention of a filter bed are the macroscopic conservation equation and the rate equation. The conservation equation is similar to that used in any fixed bed process involving the transport of matters from the mobile to the stationary phase. For an axial flow filter with a constant cross-sectional area, this is written as

$$u\left(\frac{\partial c}{\partial z}\right)_t + \left[\frac{\partial(\sigma + \epsilon c)}{\partial t}\right]_z = 0 \qquad (1)$$

The independent variables are the axial distance z and the time t, which begins with the flow of the suspension into the bed. The other assumptions involved are one-dimensional plug flow and negligible axial dispersion of particulate matters. The porosity (or void fraction) of the bed ϵ changes with time as particle accumulation within the bed increases. If the deposited matters form relatively smooth coatings outside the filter grains, ϵ and σ can be related by the simple expression

$$\epsilon = \epsilon_0 - \frac{\sigma}{1 - \epsilon_d} \qquad (2)$$

On the other hand, if the deposit morphology is largely of the blocking type (as discussed in later sections), a different expression relating ϵ, σ, and the interstitial velocity results.

Similar to all fixed bed processes, it is more meaningful to employ a corrected time variable θ defined as

$$\theta = t - \int_0^z \frac{\epsilon}{u}\, dz \qquad (3)$$

In terms of the variables (z, θ), Equation (1) becomes

$$u\left(\frac{\partial c}{\partial z}\right)_\theta + \left(\frac{\partial \sigma}{\partial \theta}\right)_z = 0 \qquad (4)$$

The difference between t and θ is usually small. However, it may become important in the interpretation of data from small experimental filters.

To complete the description, the rate expression of filtration needs to be specified. In general, this can be expressed as

$$\left(\frac{\partial \sigma}{\partial \theta}\right)_z = -u\left(\frac{\partial c}{\partial z}\right)_\theta = G[\Upsilon, c, \sigma] \qquad (5)$$

Based on slow sand filter data, Iwasaki (1937) proposed the expression

$$-\left(\frac{\partial c}{\partial z}\right)_t = \lambda c \qquad (6a)$$

namely, the profile of the particulate concentration in the liquid phase throughout a filter bed is logarithmic. If one ignores the difference between t and θ, combining Equations (5) and (6a) yields

$$\left(\frac{\partial \sigma}{\partial \theta}\right)_z = u\lambda c \qquad (6b)$$

In other words, use of Iwasaki's assumption is tantamount to the use of a first-order expression for the rate of filtration. Later, Ives (1960) applied Equation (6a) to rapid sand filters successfully. The validity of this expression was also shown experimentally by Ison and Ives (1969). λ is commonly referred to as the filter coefficient and has dimensions of reciprocal length. Typical values of λ for clean filters from a number of systems are listed in Table 1.

From physical considerations one would expect λ to be a function of the state of the media. This is confirmed by experimental observations. For example, if one plots ln c vs. z, a linear relationship is obtained initially. As time increases, a graduate departure from linearity occurs. To account for the variation of λ due to particle accumulation in the bed, one may write

$$\lambda = \lambda_0(\mathbf{x}) f_\lambda(\alpha, \sigma) \qquad (7)$$

with

$$f_\lambda(\alpha, 0) = 1$$

where λ_0 is the value of λ when the bed is clean. $\mathbf{x}$ is a parameter vector (including quantities such as interstitial

Table 1. Typical Values of Clean Filter Coefficients

Filter medium	Grain size, mm	Liquid	Suspended matter	Particle size, μm	u, cm/min	λ_o, cm^{-1}	Investigator
Calcium carbonate	Not uniform	Water	Ferric floc	10	8.2	0.1	Mackrle (1960) as determined by Ives (1963)
Calcium carbonate	Not uniform	Water	Ferric floc	10	16.7	0.044	Mackrle (1960) as determined by Ives (1963)
Anthracite	0.77	Water	Quartz powder	2-22	8.2	0.064	Robinson (1961) as determined by Ives (1963)
Sand	0.54	Water	Chlorella	5	8.2	0.34	Ives (1961)
Sand	0.647	Water	Fuller's earth	6	7.87	0.363	Deb (1969)
Granular carbon	0.594	Water	Clay (EPK)	4-40	8.47	0.102	Mehter (1970)

velocity, grain size, density difference, etc.) on which λ_0 depends. The change in the value of λ is governed by the extent of particle retention as well as another parameter vector $\boldsymbol{\alpha}$ which determines the mode of deposit morphology.

The empirical determination of the functional form of $f_\lambda(\boldsymbol{\alpha}, \sigma)$ from experimental data has been attempted by a number of investigators. Extensive reviews on this can be found in Agrawal (1966), Ives (1969, 1971), and Herzig et al. (1970). Table 2 lists some of the expressions proposed. Generally speaking, most of the proposed expressions can be derived from a general equation proposed by Ives (1967, 1969) and Mohanka (1969)

$$f_\lambda = \left(1 + \frac{\sigma}{1-\epsilon_0}\right)^{\alpha_1} \left(1 - \frac{\sigma}{\epsilon_0}\right)^{\alpha_2} \left(1 - \frac{\sigma}{\sigma_{\max}}\right)^{\alpha_3} \tag{8}$$

in which α_1, α_2, and α_3 are empirical constants. $\sigma_{\max}$ is the maximum value of σ to be achieved in filtration. It is implicitly assumed that filter media will ultimately become nonretentive owing to the change of surface characteristics, increased reentrainment, etc. Experimental data indicate that $\sigma_{\max}$ varies from system to system but is usually in the range of 0.2 ϵ_0 to 0.4 ϵ_0. Equation (8) makes it possible to represent experimental values of λ which either decrease monotonically with σ or show an initial increase followed by a decrease as shown in Figure 1.

A different approach was adopted by Mints (1951) and later by Litwiniszyn (1967). The rate expression is assumed to be

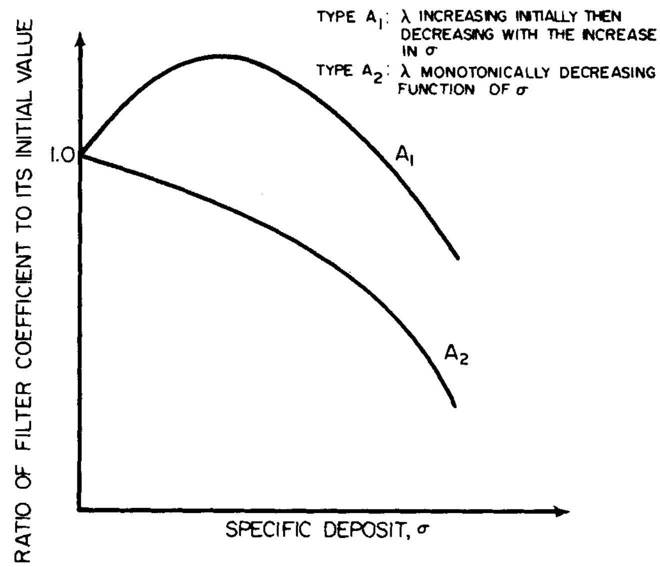

Fig. 1. Possible dependence of λ on σ.

$$-\left(\frac{\partial c}{\partial z}\right)_t = \lambda_0 c - \frac{a\sigma}{u} \tag{9}$$

The first term of the equation represents particle deposition as before, whereas the second term is introduced to account for the scouring of deposits, evidence of which is given by the observation that in a shallow filter, the effluent was found to contain particle aggregates signifi-

Table 2. Some Analytical Solutions of Filtrate Equations

Investigator	Rate expression	Solution of filtrate quality
Iwasaki (1937)	$\frac{\partial c}{\partial z} = -\lambda_0 c$	$\frac{c}{c_0} = e^{-\lambda_0 z}$
Mints (1951)	$\frac{\partial c}{\partial z} = \lambda c - \frac{\alpha}{u}\sigma$	$\frac{c}{c_0} = \sum_{n=1}^{\infty} e^{-\lambda z}\frac{(\lambda z)^{n-1}}{(n-1)!}T_n e^{-\alpha\theta}$; $T_n = T_{n-1} - \frac{(\alpha\theta)^{n-2}}{(n-2)!}$; $T_1 = e^{\alpha\theta}$
Shekhtman (1961)	$\frac{\partial c}{\partial z} = -\lambda_0(1 - j\sigma)c$	$\frac{c}{c_0} = \frac{e^{\lambda_0 j u c_0 \theta}}{e^{\lambda_0 z} + e^{\lambda_0 j u c_0 \theta} - 1}$
Ives (1963)	$\frac{\partial c}{\partial z} = -(a - b\sigma^2)c$	$\frac{c}{c_0} = \frac{Be^{-az}}{(B^2 - 1 + e^{-2az})^{1/2}}$; $B = \frac{e^{2uc_0\theta(ab)^{1/2}} + 1}{e^{2uc_0\theta(ab)^{1/2}} - 1}$

cantly greater than those present in the influent stream (Mints et al., 1967). Mints also argued that improved filtration efficiency, due to the application of polyelectrolytes, can be attributed in part to the fact that polyelectrolytes increase the bonding strength among deposits, therefore reducing the extent of scouring.

The shortcoming of these expressions is that all the model parameters can be determined only empirically. Some attempts have been made to develop generalized correlations for λ_0. For example, Ives (1971) suggested the following form of correlation:

$$\lambda d_g = P_1\left(\frac{d_p}{d_g}\right)^{e_1}\left(\frac{kT}{6\pi\mu a_p u d_g}\right)^{e_2}\left(\frac{2|\rho_p-\rho|a_p^2 g}{9\mu u}\right)^{e_3}\left(\frac{\rho u d_g}{\mu}\right)^{e_4} \quad (10)$$

However, there do not exist sufficient data or, more importantly, accurate data to establish such a correlation. Rajagopalan and Tien (1977*a*) also pointed out the lack of theoretical validity for an expression such as that of Equation (10). Accordingly, the prediction of filter coefficients, as well as of the relevant functional expressions, must largely rely upon theoretical means.

For a clean filter bed, the pressure drop vs. flow rate relationship is governed by the Carman-Kozeny equation; that is

$$-\left(\frac{\partial p}{\partial z}\right)_o = \frac{150}{d_g^2}\mu u\frac{(1-\epsilon_o)^2}{\epsilon_o^2} \quad (11)$$

To account for the change in pressure gradient due to filter clogging, either one of the following expressions may be used:

$$\frac{(\partial p/\partial z)}{(\partial p/\partial z)_o} = F_1(\boldsymbol{\beta},\sigma) \quad (12a)$$

or

$$\frac{(\partial p/\partial z)}{(\partial p/\partial z)_o} = 1 + F_2(\boldsymbol{\beta},\sigma) \quad (12b)$$

A number of specific functional forms have been proposed for F_1 (or F_2) (Ives, 1969; Herzig et al., 1970; Ives and Pienvichitr, 1965). Ives and Pienvichitr (1965) pointed out that most of these expressions can be derived from a general form:

$$F_1(\boldsymbol{\beta},\sigma) = \frac{1}{(1-\beta_2\sigma)^{\beta_1}} = 1+\beta_1\beta_2\sigma + \frac{\beta_1+(\beta_1+1)}{2}\beta_2^2\sigma^2+\ldots \quad (13)$$

Although theoretical expressions similar to that of Equation (13) have been obtained, these expressions are based on specific deposit morphology which may or may not be valid for the problem at hand. In practice, the coefficients (β_1 and β_2, for example) have to be determined from experimental data.

If the specific functional form of F_1 is known, and furthermore if $\sigma(z,\theta)$ is known from the solution of the conservation equation [that is, Equation (4)] and the rate expression [Equation (5)], the pressure distribution can be obtained from a simple integration as follows:

$$p - p_o = \left(\frac{dp}{dz}\right)_o \int_0^z F_1[\boldsymbol{\beta},\sigma(z',\theta)]\,dz' \quad (14)$$

Method of Solution

The equations which give a macroscopic description of the dynamic behavior of deep bed filtration, as discussed above, are

$$u\left(\frac{\partial c}{\partial z}\right) + \frac{\partial\sigma}{\partial\theta} = 0 \quad (4)$$

$$\frac{\partial\sigma}{\partial\theta} = G[\boldsymbol{\Upsilon}, c, \sigma] \quad (5)$$

$$p - p_o = \left(\frac{dp}{dz}\right)_o \int_0^z F_1[\boldsymbol{\beta},\sigma(z',\theta)]\,dz' \quad (14)$$

with the following initial and boundary conditions:

$$c = c_o(\theta), \quad z = 0 \quad (15)$$

$$\sigma = \sigma_o(z), \quad c = c_i(z) \qquad z>0, \quad \theta<0 \quad (16)$$

In most cases of practical interest, $c_o(\theta) = c_o =$ constant, and $\sigma_o = c_i = 0$; that is, the bed initially is free of particulate matters.

The solution of these equations yields values of c, σ, and p as functions of time and position. Specifically, one can obtain histories of filtrate concentration and pressure drop across the bed, which, in turn, can be used as a basis of design and optimization.

The simultaneous solution of the conservation equation and the rate equation perhaps deserves some comment. These equations are of the same type as those found in a variety of engineering applications such as fixed bed adsorption, cross flow heat exchanger, heat regenerators, etc. If the rate expression is of a certain kind, analytical solutions may be possible. A summary of available analytical solutions is presented in Table 2.

For the more general case, integration of Equations (4) and (5) can be carried numerically using the method of characteristics (Lapidus, 1962; Lister, 1962). A more accurate algorithm based on a higher-order Taylor series expansion developed by Vanier (1970) and applied to fixed-bed adsorption calculation (Hsieh, Turian, and Tien, 1976) has also been found useful in the integration of these equations (Payatakes et al., 1970, 1975).

Herzig, Leclerc, and LeGoff (1970) have demonstrated that if the rate expression is first order in c

$$\left(\frac{\partial\sigma}{\partial\theta}\right) = -u\left(\frac{\partial c}{\partial z}\right) = u\,\lambda_0\, f_\lambda(\boldsymbol{\alpha},\sigma)c \quad (17)$$

then the conservation equation [that is, Equation (4)] and the rate equation [that is, Equation (17)] are equivalent to a pair of ordinary differential equations:

$$\frac{d\sigma_i}{d\theta} = c_o u \lambda_0 f_\lambda(\boldsymbol{\alpha},\sigma_i) \quad (18a)$$

with

$$\sigma_i = 0, \quad \text{at} \quad \theta = 0 \quad (18b)$$

and

$$\frac{\partial\sigma}{\partial z} = -\lambda_0 f_\lambda(\boldsymbol{\alpha},\sigma)\sigma \quad (19a)$$

with

$$\sigma = \sigma_i \quad \text{at} \quad z = 0 \quad (19b)$$

Also

$$\frac{\sigma}{\sigma_i} = \frac{c}{c_o} \quad (20)$$

where σ_i denotes the value of σ at $z = 0$.

Thus, by two integration steps, the effluent quality and the specific deposit distribution throughout the bed can be estimated as functions of time. This approach gives a significant simplification of deep bed filtration computations.

An important implication of the development of Herzig et al. (1970) is the existence of what the authors termed as a "constant clogging front." Equation (19a) suggests that for a fixed value of σ, the slope of σ vs. z curve (that is, specific deposit profile) remains constant. In other words, the specific deposit profiles corresponding to different times θ are interchangeable by translation, and the profiles retain the same shape. The front velocity can be shown to be

$$V_F = \left(\frac{\partial z}{\partial \theta}\right)_\sigma = u\frac{c}{\sigma} \tag{21}$$

This conclusion, however, is not substantiated by experimental results (Letterman, 1977). A more detailed study into this question is desirable.

Parameter Estimation

The phenomenological description of deep bed filtration presented above requires the knowledge of functional forms of f_λ and F_1 and parameter values which have to be determined from experimental data. For the determination of these parameters and functions, most of the earlier investigators have employed what may be classified as a differential method which applies Equation (5) to experimental measurements. The main advantages of this method are its relative simplicity and the fact that it does not require a priori knowledge of the proper functional form. The main drawback is the lack of accuracy resulting from the required numerical (or graphical) differentiations.

In contrast to the differential method, a more rigorous technique, termed integral method, was developed by Payatakes et al. (1970, 1975). The term integral is derived from the fact that the integrated form of the phenomenological equation was used. For particular functional forms [that is, $G(\boldsymbol{\alpha}, c, \sigma)$ or $f_\lambda(\boldsymbol{\alpha}, \sigma)$, $F_1(\boldsymbol{\beta}, \sigma)$], optimal parameter values can be estimated such that the predicted behavior of the filter bed provides the best fit of experimental observations. Standard optimization techniques can be applied to search for these parameter values. To initiate the search, an initial guess of the parameter values has to be made. This represents its major but by no means decisive disadvantage.

Multimedia Filters and Polydispersity of Particles

The discussion given above has been limited to the consideration of filter media composed of uniform filter grains for the removal of monodispersed particles. In single-media filters, the retention of particulate matters is often confined to a relatively small section near the inlet. This means that a substantial part of the filter remains underutilized or not utilized at all when the filter requires backwashing because of excessive clogging at the top. This situation can be improved substantially by the use of multimedia filters, namely, filters composed of several distinctive layers. The entrance layer must have the lowest collection efficiency, and the subsequent layers must have progressively increasing efficiencies. This can be achieved by using coarse grains to form the entrance layer and increasingly finer ones for subsequent layers. Furthermore, the densities of these filter grains should be such that the desired stratification can be maintained after backwashing.

The phenomenological equations describing the behavior of any given layer of a multilayer filter are exactly the same as those given before. However the system parameters [ϵ, u, $(\partial p/\partial z)_o$] and the model parameters (λ_0, β_1, β_2, etc.) are different from layer to layer. Integration of these equations can be carried out sequentially noting that the effluent from the i^{th} layer is the influent to the $(i+1)^{\text{th}}$ layer.

To account for the polydispersity of the particulate matters, the relevant equations should be modified as follows:

$$u\frac{\partial c_j}{\partial z} + \frac{\partial \sigma_j}{\partial \theta} = 0 \tag{22}$$

$$\frac{\partial c_j}{\partial z} = G_j\left[\boldsymbol{\alpha}, c_j, \sum_{j=1}^{N} \sigma_j\right] \tag{23}$$

$$p - p_o = \left(\frac{\partial p}{\partial z}\right)_o \int_0^z F_1\left[\boldsymbol{\beta}, \sum_{j=1}^{N} \sigma_i(z', \theta)\right] dz'$$
$$j = 1, 2, \ldots, N \tag{24}$$

Namely, for each size fraction, a set of equations [Equations (23) and (24)] is required for its description. The total deposit $\sum_{i=1}^{N} \sigma_i$ is required to account for the effect of deposition on the rate of filtration as well as the pressure drop across the filter bed. A detailed discussion on this subject has been given by Payatakes (1977a).

PARTICLE DEPOSITION IN FILTER MEDIA

Model Representation of Filter Bed

The phenomenological equations discussed above describe the dynamic behavior of deep bed filtration in terms of model parameters and functions. The importance of estimating these quantities with sufficient accuracy is rather obvious. Since no generalized correlations for the estimation of these quantities have been established, one has to resort to theoretical calculation. Furthermore, theoretical analysis has the added advantage of providing a more fundamental understanding of the deposition phenomenon.

The principle involved in the theoretical calculation is as follows. A filter bed can be viewed to be an assembly of particle collectors. Using the terminology of Payatakes, Tien, and Turian (1973a), a filter bed can be considered to be a series of unit bed elements (UBE), each of which, in turn, is composed of a number of collectors. Since the thickness of a unit bed element l is always small, Equation (6a) can be applied to a UBE with the assumption that the filter coefficient remains constant throughout the element. Considering the i^{th} UBE, let c_{i-1} and c_i be the concentration of the influent and effluent streams, respectively. The following result is obtained:

$$\ln\frac{c_{i-1}}{c_i} = \lambda l \tag{25}$$

Similarly, the capacity of a UBE to remove particulate matters from the fluid stream flowing through it can be characterized by its collection efficiency η, defined as

$$\frac{c_{i-1} - c_i}{c_{i-1}} = \eta \tag{26}$$

Comparing Equations (25) and (26), we get

$$\lambda = \frac{1}{l}\ln\frac{1}{1-\eta} \tag{27}$$

Note that Equation (27) applies to any unit bed element regardless of its degree of particle deposition.

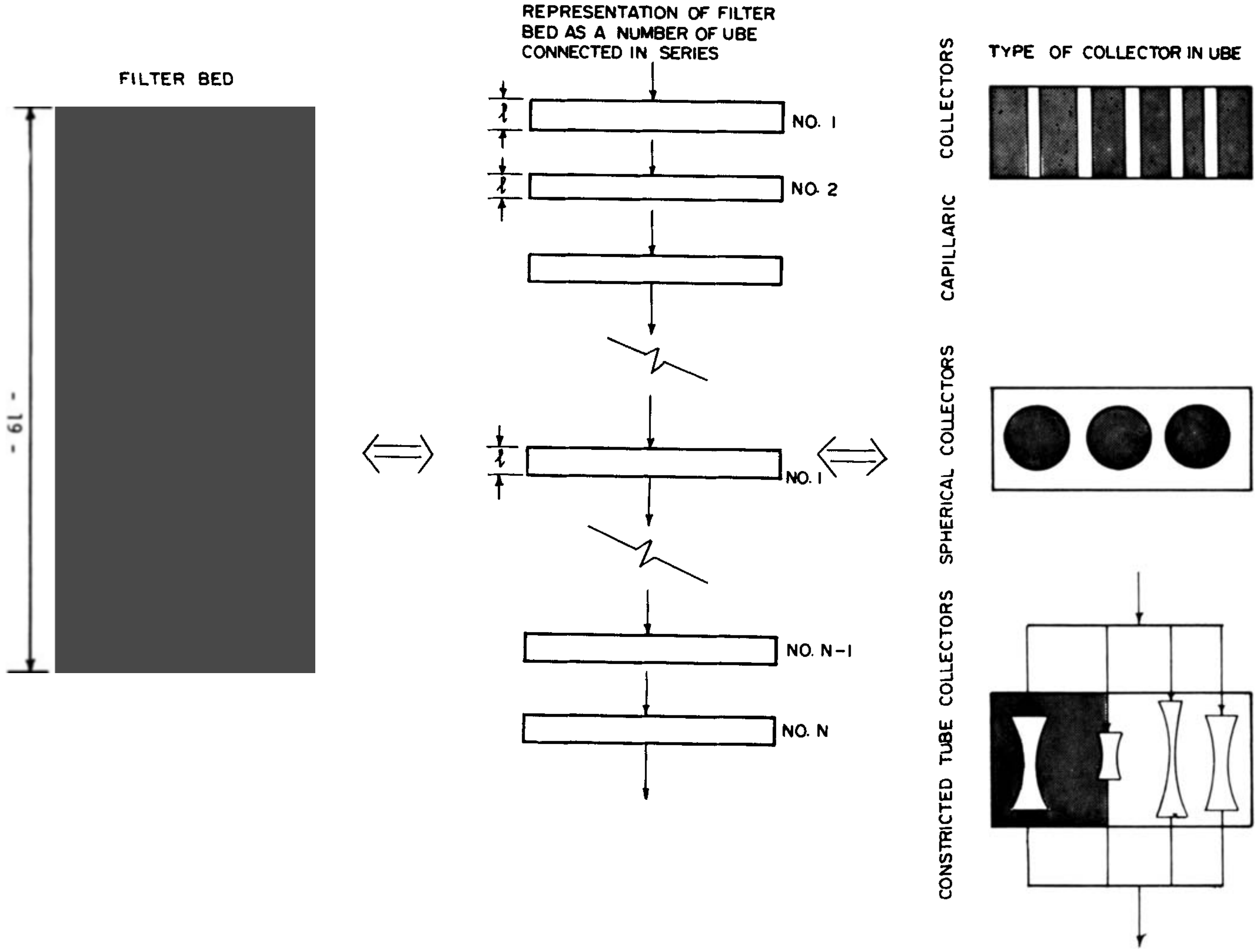

Fig. 2. Representation of filter bed as an assembly of collectors.

The general idea of depicting a filter bed as a series of UBE is illustrated in Figure 2. Specification of geometrical configurations for the collectors present in a UBE is admittedly somewhat arbitrary. In principle, any model which adequately describes the flow field in the porous media can be used. This possibility of multiple selection, however, is limited by practical considerations that only relatively simple geometrical entities should be used. The models employed by various investigators can be grouped into three categories: capillaric collector (Payatakes, Rajagopalan, and Tien, 1974; Hung and Tien, 1976), spherical collector (Yao, Habibian, and O'Melia, 1971; Spielman and Fitzpatrick, 1973; Payatakes, Rajagopalan, and Tien, 1974; Rajagopalan and Tien, 1976, 1977*a*), and the constricted tube collector (Payatakes, Tien, and Turian, 1974*a*, *b*).

Using the collector concept, the retention of particulate matters within a filter medium can be considered in terms of the deposition of particles from a suspension flowing past collectors of specified geometry. The mechanisms of deposition are known to be inertial impaction, sedimentation, interception, Brownian diffusion, and straining. For liquid systems, the effect of inertial impaction is negligible, and the relative importance of other mechanisms depends upon a number of variables, the most significant of which is perhaps particle size. Although explicit rate expressions of deposition for some of the individual mechanisms are available (see, for example, Davies, 1973), in most practical situations deposition takes place according to more than one mechanism.

Trajectory Calculation

The most versatile method for estimating the rate of particle deposition is the so-called trajectory calculation. As particles move toward a collector, their trajectories deviate from streamlines, and some of them may intersect with the collector. If one assumes that deposition occurs once a particle makes contact with the collector,* the particle deposition flux can be estimated if particle trajectories are known. These trajectories can be determined in turn from the appropriate equations of particle motion, with the knowledge of the forces acting on the particle.

The idea of determining the rate of particle deposition from particle trajectories was first advanced more than 40 yr ago in connection with air filtration (Sell, 1931; Albrecht, 1931). However, the possibility of extending this concept to liquid filtration was recognized only rather recently (O'Melia and Stumm, 1967). The use of trajectory calculation for determining particle deposition is illustrated in Figure 3 in which unit cells of the constricted tube type are considered as collectors. The amount of particle deposition can be determined from the location of the limiting trajectory which is defined as the trajectory that makes contact with the unit cell at its exit.

* This is tantamount to the assumption that particles do not bounce off the collector, which is true in liquid filtration.

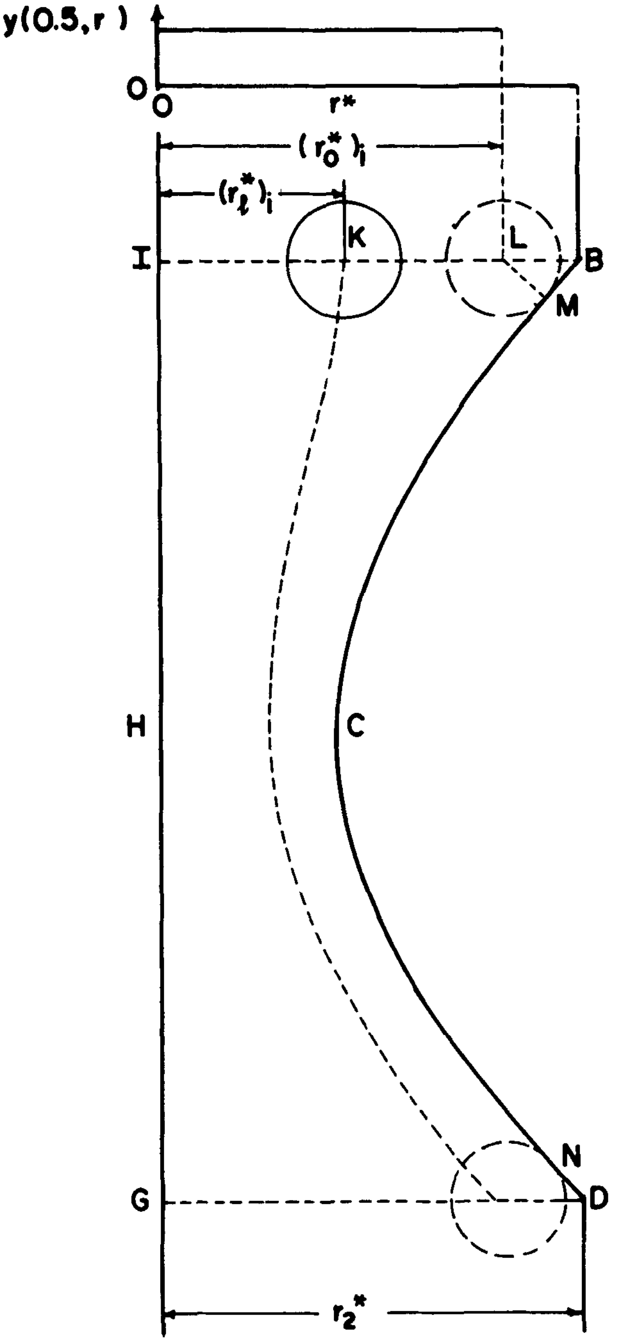

Fig. 3. Schematic representation of limiting trajectory and of particle position closest to the wall at the entrance of the unit cell.

In Figure 3, this is represented by particle K with an inlet radial position $(r_l^*)_i$. Any particle entering the unit cell at a position with radial coordinate between $(r_l^*)_i$ and $(r_o^*)_i$ will become deposited, while those with initial radial positions less than $(r_l^*)_i$ will flow out of the cell. Also, there can be no particles with initial radial positions greater than $(r_o^*)_i$. The collection efficiency of the constricted tube collector η_{oi}, by definition, is given by

$$\eta_{oi} = \frac{\psi^*\left[\frac{1}{2}, (r_l^*)_i\right] - \psi^*\left[\frac{1}{2}, (r_o^*)_i\right]}{\psi^*\left[\frac{1}{2}, 0\right] - \psi^*\left[\frac{1}{2}, (r_o^*)_i\right]} \qquad (28)$$

where ψ^* is the dimensionless stream function for the flow inside the constricted tube (Payatakes, Tien, and Turian, 1973*b*). Since a unit bed element contains a number of constricted tubes of different sizes (see Figure 2), the collection efficiency of the entire element (or unit collector) is expressed as

$$\eta_o = \frac{1}{<d_c^3>} \sum_{j=1}^{I_o} \eta_j\, d_j^3\, \eta_{oj} \qquad (29)$$

Trajectory Calculation Results

A detailed discourse on the determination of particle trajectories (or of limiting trajectories in particular) is beyond the scope of the present paper, and only a brief outline will be presented. To initiate trajectory calculation, the following procedures have to be followed:

1. Specification of collector geometry, collector size, and size distribution.
2. Specification of flow field around or within the specified collector.
3. Specification of forces acting on particles under consideration.

Thus, a variety of combinations of conditions can be used for the calculation of collection efficiency, a fact which accounts for the differences among the various studies on this subject carried out during the past decade. A summary of these studies is given in Table 3. Most of these studies were based on four dissertations completed in recent years at North Carolina (Yao, 1968), Harvard (FitzPatrick, 1972), and Syracuse (Payatakes, 1973; Rajagopalan, 1974). A brief account of these studies is presented below.

1. The particle trajectories are obtained from the integration of the appropriate equations of particle motion, which are obtained from the balance of forces and torques acting on the particle. The exact form of these equations varies with the geometry of the collector, but they are first-order differential equations because of the omission of the inertial force. Furthermore, the Brownian diffusion force cannot be included owing to its stochastic nature. The forces which have been considered are gravity, hydrodynamic drag (with or without wall correction), and different kinds of surface forces. The effect of interception is included as a boundary condition to determine particle deposition.

2. The pertinent equations of particle motion, irrespective of their specific forms, contain a large number of dimensionless parameters, a listing of which is given in Table 4. In general, these equations do not permit analytical solutions but can be solved effectively with a number of algorithms such as the fourth-order Runge-Kutta method or the Adams-Moulton method. However, for two cases, isolated sphere model (Rajagopalan and Tien, 1977*a*) and the sphere-in-cell model (Rajagopalan and Tien, 1976), collection efficiency values obtained from numerical calculation have been correlated empirically with relevant dimensionless parameters, thus overcoming the major objection about the use of trajectory calculation for the estimation of the filter coefficient, namely, the method being tedious and time consuming. These expressions are:

Sphere-in-cell model (Rajagopalan and Tien, 1976)

$$\eta_o = 0.72\, A_s N_{Lo}^{1/8} N_R^{15/8} + 2.4 \times 10^{-3} A_s N_G^{1.2} N_R^{-0.4}$$

$$\text{for} \quad N_R < 0.09 \qquad (30a)$$

where

$$A_s = 2(1 - P^5)/w \qquad (31)$$

$$P = (1 - \epsilon_0)^{1/3} = a_c/b \qquad (32)$$

$$w = 2 - 3P + 3P^5 - 2P^6 \qquad (33)$$

Isolated sphere model (Rajagopalan and Tien, 1977*a*)

TABLE 3. SUMMARY OF RESULTS ON TRAJECTORY CALCULATIONS FOR DEEP BED FILTRATION

Model	Investigator	Remarks
Single-sphere model	Yao, Habibian, and O'Melia (1971)	Surface interaction not included; drag correction neglected.
Single-sphere model	Rajagopalan and Tien (1977*a*)	Surface interaction with retardation effect for London force. Drag correction considered.
Capillaric model	Payatakes, Rajagopalan, and Tien (1974*b*)	Surface interaction with retardation effect for London force. Drag correction considered.
Capillaric model	Hung and Tien (1976)	Surface interaction and drag correction included; nonvanishing fluid velocity across collecting surface.
Sphere-in-cell model (Happel's model)	Spielman and FitzPatrick (1973)	Approximate fluid velocity expression valid for small particles. No retardation effect.
Sphere-in-cell model (Brinkman model)	Payatakes, Rajagopalan, and Tien (1974*b*)	Surface interaction and drag correction included.
Sphere-in-cell model (Happel's model)	Rajagopalan and Tien (1976)	Surface interaction with retardation effect for London force and drag correction included.
Constricted tube model	Payatakes, Tien, and Turian (1974)	Surface interaction with retardation effect for London force and drag correction included. Constricted nature of flow channels considered.

TABLE 4. DIMENSIONLESS GROUPS APPEARING IN TRAJECTORY ANALYSIS

Name	Symbol	Definition
Double layer group	N_{DL}	κa_p
Electrokinetic group No. 1	N_{E1}	$\vec{\epsilon}\kappa[\psi^2_{01} + \psi^2_{02}]/12\pi\,\mu\,V_\infty$
Electrokinetic group No. 2	N_{E2}	$2\,\psi_{01}\,\psi_{02}/[\psi^2_{01} + \psi^2_{02}]$
Gravitational group	N_G	$2\,a_p^2(\rho_p - \rho)\,g/9\,\mu\,V_\infty$
London group	N_{Lo}	$H/9\,\pi\,\mu\,a_p^2\,V_\infty$
Relative size group	N_R	a_p/a_c
Retardation group	N_{Rtd}	$2\,\pi\,a_p/\lambda_e$
Brownian diffusion group (Peclet number)	N_{Pe}	$V_\infty(2a_c)/D_{BM}$

$$\eta_o = N_G + 1.5\,N_R^2 \tag{34}$$

The definitions of the dimensionless groups are given in Table 4. These expressions are based on results obtained under the assumption that the surface interactions (vander Waal's force and double layer force combined) are favorable.

3. The calculated filter coefficient values are found to vary with the collector model. However, with the exception of the capillaric model which gives significantly lower values of λ_0 (Payatakes, Rajagopalan, and Tien, 1974), calculated values of λ_0, according to different collector geometries, usually were found to be comparable in magnitude. Furthermore, the calculated values are of the same order of magnitude as corresponding experimental values. A comparison between the experimental values of λ_0 obtained by Ison (1967) and Ison and Ives (1969) and the corresponding theoretical values, ac-

TABLE 5. COMPARISON OF EXPERIMENTAL DATA (ISON, 1967; ISON AND IVES, 1969) AND CALCULATED λ_0 BASED ON HAPPELS' AND CONSTRICTED TUBE MODELS (RAJAGOPALAN AND TIEN, 1976)

$\lambda \times 10^2$, cm^{-1}

	$d_p = 2.75\ \mu$m			$d_p = 4.5\ \mu$m			$d_p = 9.0\ \mu$m		
Run No.	Expt'l data	Payatakes (1973)	Rajagopalan (1974)*	Expt'l data	Payatakes (1973)	Rajagopalan (1974)*	Expt'l data	Payatakes (1973)	Rajagopalan (1974)*
I	6.0	2.4	2.4	7.6	3.2	3.4	8.8	7.1	9.6
II	8.1	3.1	2.6	11.0	4.6	3.9	14	10.5	10.4
III	11.0	4.1	2.7	15.0	6.2	4.1	16.5	14.2	10.4
IV	3.1	1.8	2.2	4.6	2.5	3.7	6.4	5.0	9.2
V	3.1	1.2	1.8	4.4	1.7	3.3	5.6	3.7	9.1
VI	4.5	2.1	1.5	5.8	3.3	2.2	6.3	7.7	6.0
VII	3.9	1.9	1.1	4.4	3.1	1.5	4.6	8.2	3.5
VIII	2.7	1.7	0.7	3.9	3.1	1	5.3	8.2	2.3

* The corresponding values based on Spielman and Fitzpatrick (1973) are given in Payatakes et al. (1974*b*) and are markedly higher, especially for large d_p (for example, for $d_p = 9\ \mu$m, they are about four to ten times higher than those based on the rigorous application of the sphere-in-cell model). See also Rajagopalan and Tien (1976).

cording to Happel's and constricted tube models, is given in Table 5.

4. The dependence of η_o on the various dimensionless groups listed in Table 4 varies, of course, with the models used. Qualitatively, they exhibit essentially the same behavior to a remarkable degree. Based on the constricted tube model, Payatakes, Tien, and Turian (1974*b*) undertook a case study by varying the values of all pertinent dimensionless groups one at a time for a particular set of values of these groups. Their conclusions were:

a. η_o is a very strong increasing function of N_G, especially at values of N_G around zero.

b. When the London-van-der Waals force dominates the double layer interaction force at all separations, the effect of groups N_{E1} and N_{E2} is negligible.

c. There are critical values at which small changes in N_{E1}, N_{DL}, N_{Lo}, and N_{Ret} produce a dramatic decrease in η_o, in step function manner. A more detailed discussion on the effect of surface interaction on particle deposition will be given in later sections.

d. The η_o vs. N_{RS} curve ($N_{RS} = 1/2N_R$) has a very pronounced minimum. This is also observed by Rajagopalan and Tien (1976) and has been suggested by them as a possible explanation for the often observed initial increase of λ.

5. The statements made above should not be construed that there is no difference among the various models used for the characterization of deep bed filters. There exist subtle and significant differences. For example, according to the single-sphere model (Rajagopalan and Tien, 1977*a*), η decreases monotonically with the increase of N_R, whereas the results based on the Happel's model indicate that for $N_G > 10^{-3}$, the η vs. N_R plot exhibits a minimum. It should also be mentioned that even though the work by Spielman and FitzPatrick (1973) was presented to be a study of deposition based on the Happel's model, the various approximation introduced by these investigators caused the results to be significantly different from those obtained by a correct and consistent use of the Happel's model (see Payatakes, Rajagopalan, and Tien, 1974; Rajagopalan and Tien, 1976). Furthermore, it should be emphasized that based on geometrical grounds, the Happel's model cannot be applied to particles with diameters greater than $[1 - (1 - \epsilon_o)^{1/3}]a_c/(1 - \epsilon_o)^{1/3}$, that is, about 0.09 d_g. In reality, for the trajectory calculation to be valid, particle diameters must be much less than that value.

6. Among all the models employed in the study of particle deposition, only the constricted tube model provides an adequate framework to consider the effect of deposition on filter performance. The isolated sphere model completely ignores the effect due to the presence of neighboring grains. Happel's model considers this effect to a certain extent, but incompletely. A rigorous study of the important phenomena associated with straining due to lodgement of large particles or particle aggregates at pore constrictions is possible only with the use of the constricted tube model.

7. The collection efficiency obtained from trajectory calculation is often referred to as the initial collection efficiency or clean collector efficiency. Significant particle deposition may change the collector configuration, alter its surface characteristics, and cause the flow field around the collector to be different from that around a clean collector to such an extent as to render the collection efficiency calculated with the omission of these factors to be erroneous. The principle of trajectory calculation, however, remains valid. The problem is that of taking into account all these changes in the formulation of the trajectory equation.

Deposition by Brownian Diffusion

As pointed out earlier, in the formulation of the particle trajectory equations, the Brownian movement of the particle is omitted. Consideration of the random motion caused by Brownian bombardment of the particles by liquid molecules would make the resulting equation stochastic in nature. Although technically a solution can still be found and be known as the trajectory of the particle, it would not be deterministic but would be characterized by probability distributions. The appropriate setting for the analysis of stochastic trajectories is the theory of diffusion processes. Such an approach was applied to deep bed filtration by Rajagopalan (1974). One of the main results of his analysis is that the contribution of Brownian diffusion to deposition η_{BM_o} can be simply added to that of all other mechanisms combined (η_o) to give the total fraction collected:

$$\eta_{T_o} = \eta_o + \eta_{BM_o} \tag{35}$$

This method was used by Yao (1968), FitzPatrick (1972), and Payatakes et al. (1974*a*, *b*) without proof. That such an analysis is adequate enough for the consideration of diffusion in a filter bed was also shown by Prieve and Ruckenstein (1974).

Cookson (1970) calculated particle transfer and deposition rates in packed beds based on the results of Pfeffer and Happel (1964). Assuming that particle concentration vanishes on the grain surfaces (instantaneous irreversible adsorption), Cookson obtained

$$\eta_{BM_o} = 4A_s^{1/3} N_{Pe}^{-2/3} \tag{36}$$

where A_s is defined by Equation (31). The definition of the Peclet number N_{Pe} is given in Table 4. Equations (35), (30), and (36) together give the Rajagopalan-Tien correlation

$$\eta_{T_o} = 0.72\, A_s N_{Lo}^{1/3} N_R^{15/8} + 2.4 \times 10^{-3} A_s N_G^{1.2} N_R^{-0.4} + 4A_s^{1/3} N_{Pe}^{-2/3} \quad \text{for} \quad N_R < 0.09 \tag{30b}$$

which includes all mechanisms accounted for by Equation (30*a*) as well as Brownian diffusion.

Capture by Straining

Filtration systems in which straining is a dominant mechanism from the outset are not operating under optimal conditions, since they are characterized by very shallow penetration into the filter and rapid clogging, requiring frequent backwashing. In such a case, the filter should be replaced with one composed of larger grains, so that the degree of straining may be reduced and better utilization of the filter achieved. In view of this, it may appear that the study of filtration systems with predominant straining is without practical importance. This is not so. Its study is useful both from a theoretical and an operational point of view. It provides a more complete model of filtration, applicable to particles of all sizes. More importantly, even in the absence of particles large enough to be captured by straining, reentrained particle agglomerates may very well be removed by straining, a phenomenon which is believed to be of major importance in determining the transient behavior of depth filtration systems.

Based on experimental evidence, Craft (1969) claimed that straining is important for particles with diameter

$$d_p > 0.2 \langle d_g \rangle \tag{37}$$

This result is in essential agreement with a similar criterion proposed by Maroudas and Eisenklam (1965). It is worth noting that 0.2 $\langle d_g \rangle$ is approximately equal to

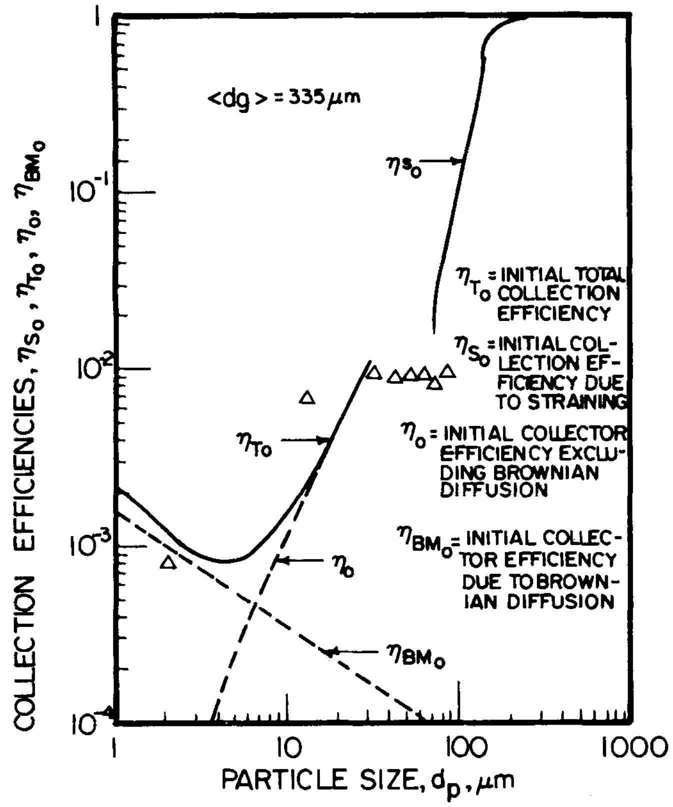

Fig. 4(a). Comparison between data by craft and the corresponding calculated values (Payatakes and Tien, 1974).

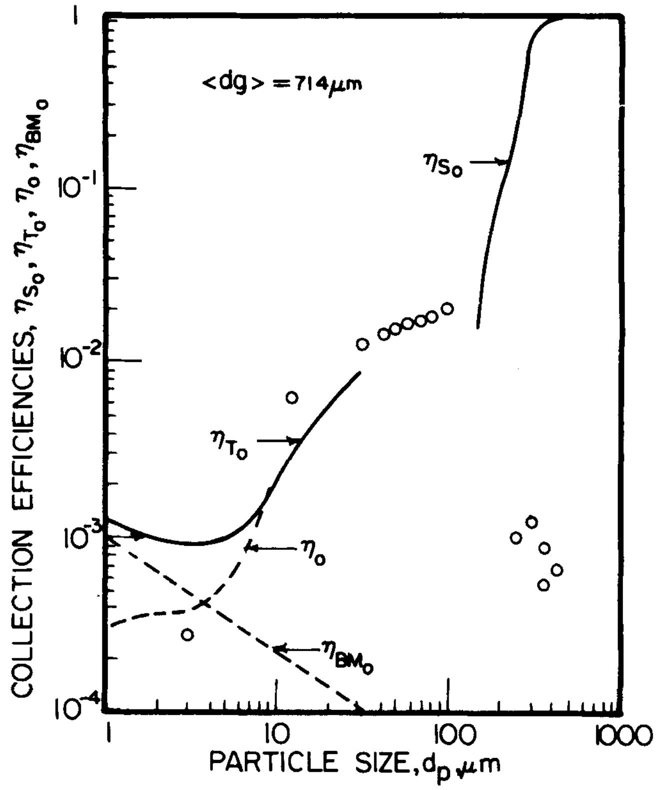

Fig. 4(b). Comparison between data by craft and the corresponding calculated values (Payatakes and Tien, 1974).

the diameter of the smallest constriction of the unit collector corresponding to a packed bed (see Table 8.4, Payatakes, 1973). Thus, the experimental criterion expressed by Equation (37) is in agreement with the constricted tube model.

Payatakes and Tien (1974) developed a simple model for filtration due to straining. Consider a monosized particle suspension with d_p larger than the minimum constriction diameter of the bed. It is assumed that particles which enter a unit bed element composed of constricted tube collectors at the same time and which escape capture will exit the unit collector at the same time. Based on a geometrical consideration and the assumption that the probability of a particle entering a particular constricted tube is proportional to the bulk flow rates, the collection efficiency is found to be

$$\eta_s = \frac{1}{<d_c^3>} \sum_{i=1}^{I_s} n_i d_i^3 \tag{38}$$

where d_1, d_2 ... d_{Ic} denote the constriction diameters of the constricted tubes present on the unit bed element in ascending order. n_i denotes the number fraction of the tubes with constriction diameter d_i. $<d_c^3>$ is the average value of d_i^3, and I_s is the index value defined by $d_{I_s} \leq d_p < d_{I_s+1}$.

A comparison of Equation (38) with experimental data reported by Craft (1969) is shown in Figures 4*a* and *b*. The pertinent constriction size distribution data of the filter media are those given by Payatakes (1973) (see Table 8.4 of the references). Also included in the figures are the collection efficiencies obtained from trajectory calculation. A remarkable degree of consistency among these results is observed. It is interesting to note that the same approach was used by Donaldson et al. (1977) in their study of the transport of particles in sandstone with the

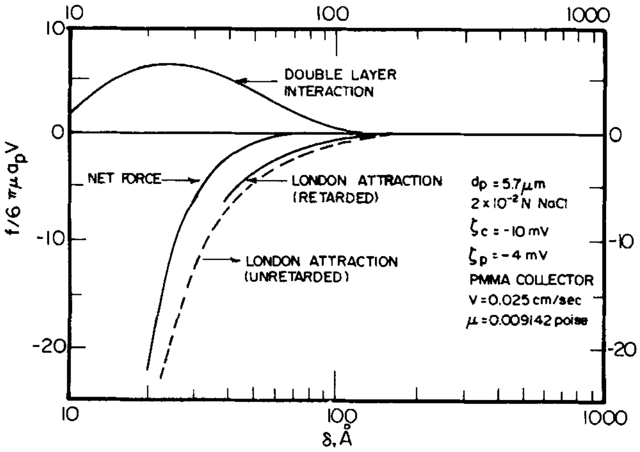

Fig. 5(a). Surface interaction force (for the sphere-plane system).

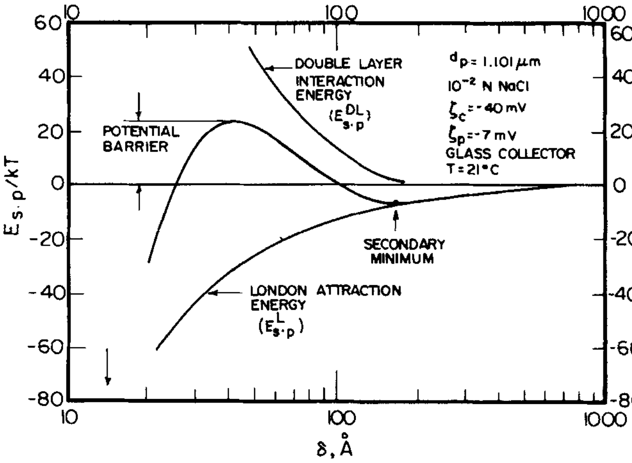

Fig. 5(b). Surface interaction energy (for the sphere-plane system) corresponding to an 'unfavorable' condition.

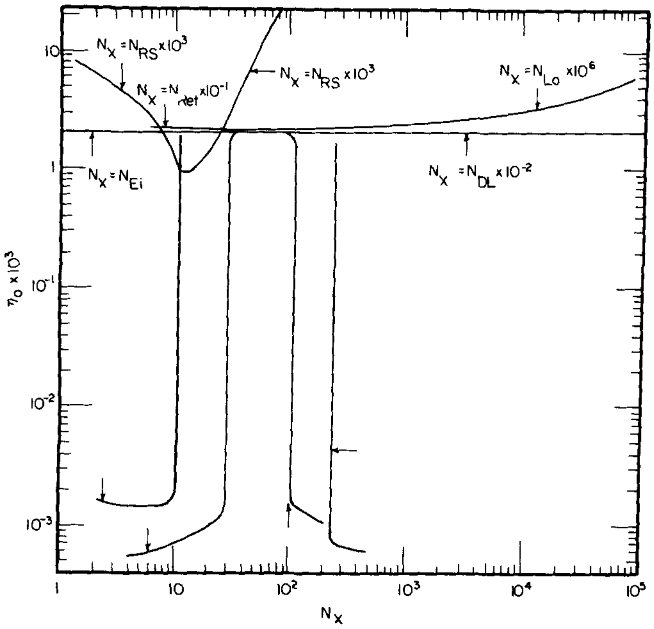

Fig. 6. Effect of surface interaction on η_0 according to trajectory (Payatakes, Tien and Turian, 1974b).

difference that capillaries of various sizes instead of constricted tubes were used in the analysis.

EFFECT OF SURFACE INTERACTION ON DEPOSITION

Under conditions commonly encountered in liquid deep bed filtration, when particles move sufficiently close to filter grain surfaces, a significant manifestion of surface forces is expected. The surface forces involved are principally of two kinds, the molecular dispersion force (commonly known as London van-der Waal's force) and the double ionic layer interactive force. The former is given as

$$F_L = -\frac{2Ha_p^3}{3\delta^2(2a_p+\delta)^2}\,\alpha_{sp}(\delta, a_p, \lambda_e) \tag{39}$$

Exact but cumbersome expressions for α_{sp} and simple expressions for $a_p > 1$ μm have been obtained by Payatakes (1973).

The double ionic layer force between two approaching bodies under conditions of constant surface potentials, according to Hogg et al. (1966), can be expressed as

$$F_E = \frac{\bar{\epsilon}\, a_p\kappa(\psi_{01}^2+\psi_{02}^2)}{2}\left[\frac{2\psi_{01}\,\psi_{02}}{(\psi_{01}^2+\psi_{02}^2)} - e^{-\kappa\delta}\right]\frac{e^{-\kappa\delta}}{1-e^{-2\kappa\delta}} \tag{40}$$

The double layer reciprocal thickness κ is defined as

$$\kappa = \sqrt{\frac{4\pi e^2}{\bar{\epsilon}\, kT}\sum_j m_j z_j^2} \tag{41}$$

Equations (40) and (41), upon comparison with the numerical solutions of Derjaguin (1954) and Devereux and Bruyn (1963), are found to yield accurate results of ψ_{01} and ψ_{02} up to 60 mv and κa_p larger than 10.

Both of the force expressions [that is, Equations (39) and (40)] are obtained with the assumption that the particle-grain system can be approximated by a sphere-plate system, namely, that the size of the collector is much greater than that of the particle. Both forces act along the direction normal to the collector surface.

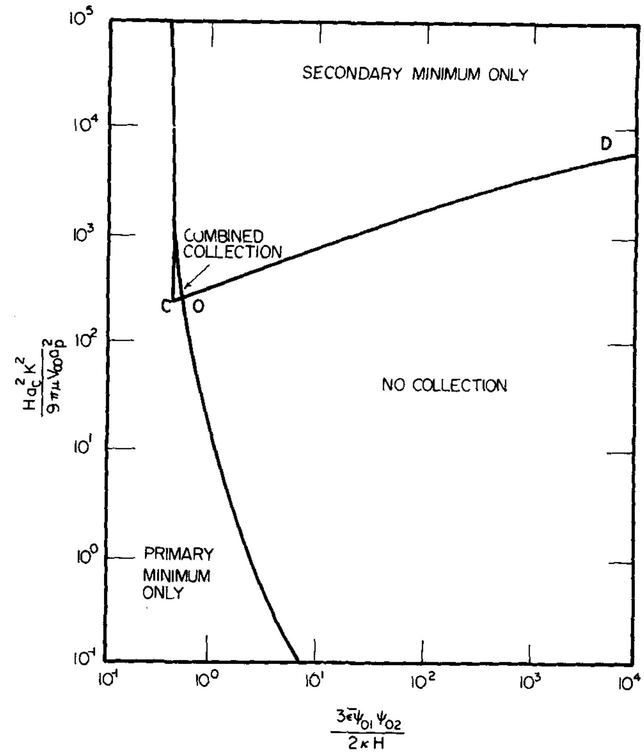

Fig. 7. Delineation of collection regimes according to Spielman and Cukor (1973).

The net surface interaction is the algebraic sum of these two forces and can be either favorable (that is, attractive for all separations) or unfavorable (that is, repulsive at certain separations) as shown in Figures 5*a* and *b*. The London force is invariably attractive. On the other hand, as seen in Equation (40), the sign of the double-layer force depends on whether the signs of the two surface potentials are the same or not. The magnitude of the force, on the other hand, is influenced by the ionic concentration of the liquid. Thus, there exists the possibility of altering both the magnitude and sign of the double-layer force between particles and collector surface through the use of appropriate surface coatings and by changing the *p*H values of the liquid medium. The expectation that a more attractive force between particle and collector would lead to enhanced deposition has provided incentive for the study of the effect of surface interaction.

Theoretical Studies of non-Brownian Particles

Theoretical trajectory calculations including both the double-layer force and the London force (with retardation effect) have shown that the presence of a repulsive force barrier near the collector surface prevents particles from being collected, and this is true irrespective of the model used for filter representation (Payatakes, Tien, and Turian, 1974*b*; Rajagopalan and Tien, 1976, 1977*a*). This conclusion is shown in Figure 6 in which the results of trajectory calculations based on the constricted tube model are presented in the form of plots of η_o vs. the various dimensionless parameters characterizing surface forces. Figure 6 indicates that the value of η_o undergoes a step function change to virtually zero at certain critical values of the relevant parameter corresponding to the presence of a repulsive force barrier near the collector surface. The presence of a barrier force of sufficient magnitude makes deposition impossible. In reality one may expect the transition to be more gradual owing to a number of factors including nonuniformity of surface po-

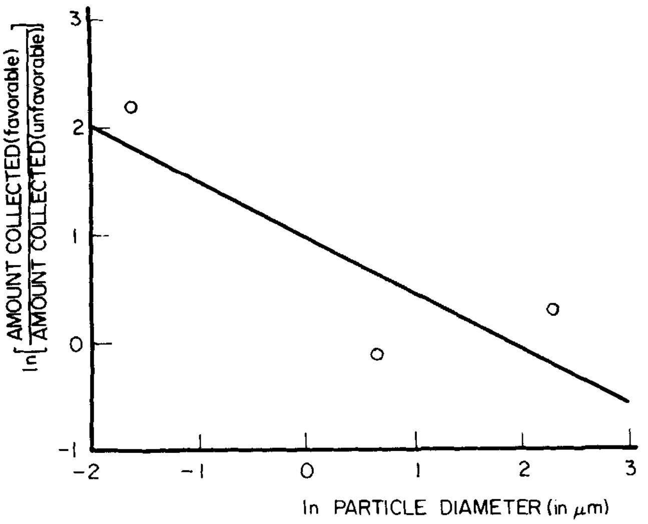

Fig. 8. Onorato's results.

tentials of particles and filter grains, the presence of Brownian motion, etc. Furthermore, in the theoretical calculation, both the collector and the particles are assumed to be perfectly smooth, while in reality their surface roughness is likely to be of the same order of magnitude as the range of surface interactions. It would be improper to interpret the trajectory calculation results without taking these factors into account.

A different approach to consider the effect of surface interactions on particle deposition was advanced by Spielman and Cukor (1973). It was argued that deposition does not require physical contact between particle and collector. Particles become collected if, through a balance of forces, they are held stationary in close proximity of a collector. Based on this argument, and a close examination of the various forces acting on a particle, a delineation of the nature and mechanism of deposition was established. The results are shown in Figure 7.

Brownian Diffusion of Submicron Particles

The studies cited above apply to particles of relatively large size (that is, diameter greater than 1 μm). For submicron particles, the Brownian force becomes important, as particle deposition can be considered to be equivalent to that of convective diffusion. The effect of surface interactions on Brownian diffusion has been studied by a number of investigators (Clint et al., 1973; Ruckenstein and Prieve, 1973; Spielman and Friedlander, 1974; Wnek, 1973; Wnek et al., 1977) for both spherical collectors and rotating disk systems. The presence of a repulsive force barrier near the collector surface can be shown to be equivalent to the occurrence of a first-order chemical reaction at the collector surface. The equivalent rate constant is expressed in terms of the potential of the surface interaction forces. The rate of deposition is given as

$$J = \frac{J^0}{1 + \left(\frac{J^0}{K C_0}\right)} \tag{42}$$

According to Spielman and Friedlander (1974), K is given as

$$K^{-1} = \frac{1}{D} \int_0^\infty (e^{\phi/kT} - 1)\, dx \tag{43}$$

Single Collector Experiment

Several investigators (Marshall and Kitchener, 1966; Hull and Kitchener, 1969; Clint et al., 1973) have mea-

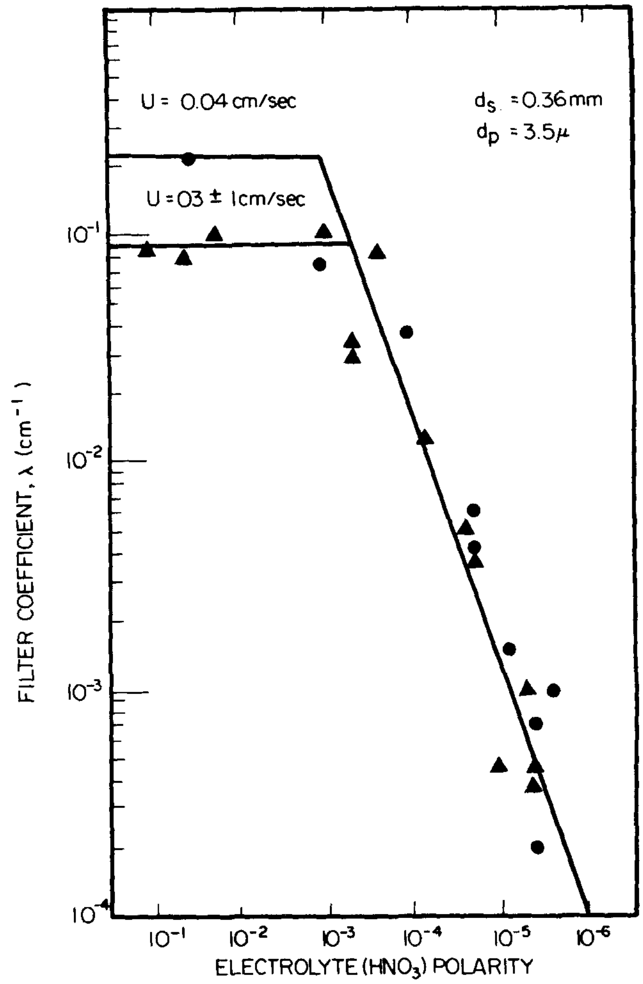

Fig. 9. Effect of surface interaction due to pH change in filter coefficient (Fitzpatrick, 1972).

sured the rate of deposition from colloidal suspensions to a rotating disk surface. These investigations were carried out as extensions of studies of colloidal stability, since they provided a more flexible system in which both the nature of the interacting bodies and the sign and magnitude of their charges can be varied. The particle sizes in these measurements were in the submicron range so that Brownian diffusion was the dominant mechanism of deposition. The results indicate that the rate of deposition is a strong function of surface conditions, but no agreement was obtained with theoretical predictions when a repulsive

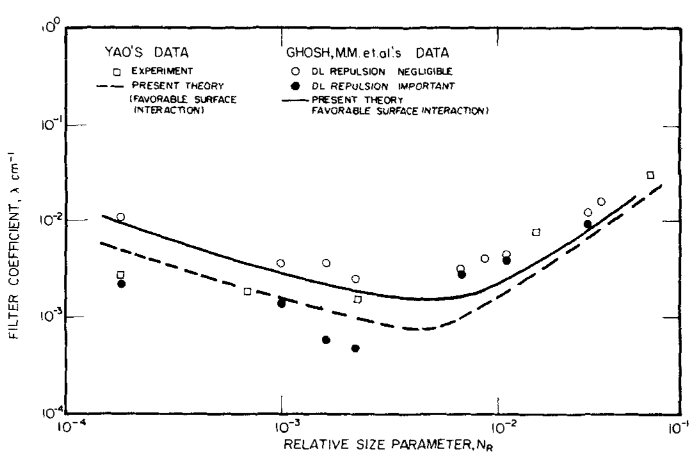

Fig. 10. Comparison of theoretical results with experimental data of Ghosh et. al. (1975) and Yao (1968).

double-layer force was present. There also appears to be some uncertainty with respect to the data. For example, the rate of deposition was found to decrease after an initial period (Hull and Kitchener, 1969), but no explanation was offered for this time dependent behavior.

Measurements of the deposition of particles on collectors with geometries similar to those found in deep bed filters have been made by Chang (1973), Rajagopalan (1974), Rajagopalan and Tien (1977*b*), and Onorato (1978), Chang's work was concerned with single fibers. The amount of deposition, determined from direct counting of deposited particles under a microscope, was found to vary with the surface treatment afforded to the fiber. Rajagopalan's work was carried out using spherical collectors. The surface potentials of the collector and particles were varied by coating them with surfactants. Thus, it was possible to obtain both favorable interaction (net attractive force) between collector and particles as well as unfavorable interactions (presence of repulsive force). With favorable interaction, his experimental data agree well with trajectory calculations. With the presence of repulsive barriers, the amount of deposition was reduced but not to the extent predicted by trajectory calculation. The reduction was more pronounced for smaller particles. Similar but more extensive measurements have recently been conducted by Onorato (1978). His results indicate that the presence of repulsive force barrier decreases the rate of depostion but not to the extent predicted by theory. Furthermore, this reduction diminishes with increasing particle size. The results are shown in Figure 8.

Measurement with Filter Bed

Hunter and Alexander (1963) and more recently FitzPatrick (1972) and Ghosh et al. (1975) conducted measurements with granular filters to study the effect of surface interactions. In FitzPatrick's work, surface potentials of filter grains were varied through the addition of electrolytes. In one series of measurements, it was shown that the filter coefficient suffered a catastrophic decline as the surface potential of the filter grain exceeded a certain value which corresponds to the presence of a repulsive barrier between the particles and the filter grains (see Figure 9). A semiempirical relationship was proposed, the validity of which remains to be tested since it was based on a rather small amount of data. The data of Ghosh et al. (1975) and Yao (1968) (see Figure 10) indicate a decrease in λ with the establishment of a double-layer repulsive force. The magnitude of the decrease was rather small and well within experimental error. Hunter and Alexander's experiment was conducted using kaolinite suspensions in beds of silica particles. The amount of deposition was found to vary with the magnitude of the surface interaction, and this was attributed to the effect of surface potentials on the yielding strength of the deposited kaolinite sol and the hydrodynamic drag force exerted by the flowing suspension on the deposits formed within the bed.

A number of studies on improved filtration performance with use of precoated filter aids have been reported in literature (Oulman et al., 1964; Burns et al., 1970 Bauman and Oulman, 1970; Martinola, 1973). Filter aid materials (diatomite, for example) coated with desired polyelectrolytes were added to the suspension before the commencement of the filtration process. A prima facie case on the importance of surface interaction on particle deposition was made from the improved filtration results. It is, however, difficult to obtain quantitative relationships between the increase in deposition rate and the state of surface interactions from these data because of the inherent complexities associated with the growth of filter cakes.

EFFECT OF PARTICLE DEPOSITION ON FILTER PERFORMANCE

The retention of particulate and colloidal matters within a filter bed causes changes in the microstructure of the filter medium. As a result, the behavior of the flow of fluid through the medium and the deposition of particles from suspensions to filter grains can be significantly affected.

Qualitatively speaking, the major changes resulting from particle deposition involves the effective size and geometry of filter grains, the surface characteristics of the grains, the porosity, and the effective porosity of the bed. The last quantity is defined as the fraction of the connected void space which is open to suspension flow. This is an important variable in the understanding of the effect of deposition on filter performance, since particle deposition may result in the blocking of certain flow passages of the bed. A complete filtration theory should provide information on these changes as functions of operating and system variables. Once these changes are known, it would be possible, at least in principle, to account for these changes in predicting filtration rates and pressure drop requirements.

The development of modern filtration theories, of course, has not reached such a stage as to permit a complete and quantitative description of the effect of particle deposition. Sufficient progress, however, has been made to allow a semiquantitative treatment. The pertinent works in this regard will be reviewed below.

Deposit Morphology

The importance of the knowledge of deposit morphology in understanding the effect of particle deposition is rather obvious. For example, the amount of particle deposits required to completely fill a given pore space in a filter bed and that required to block the constriction of that pore space are quite different, although their effects on fluid flow through the medium are identical. Failure to recognize this fact is the main reason for the lack of success of earlier investigations in developing generalized expressions relating the effect of deposition to the amount of deposition [such as Equation (7) for filter coefficients and (12*a*, *b*) for pressure drop].

Stein (1940), in a study which employed the use of an experimental filter composed of cylindrical rods, observed the formation of relatively smooth coatings over the collecting bodies during filtration of ferric floc. A pattern of smooth but nonuniform coating extending only to the front portion of the filter grains was found in the case of calcium carbonate suspensions by Cleasby and Baumann (1962) and for kaolinite particles by Ison and Ives (1969). Maroudas and Eisenklam (1965) performed a very ingenious series of experiments using a specially designed two-dimensional filter consisting of a number of capillary segments (alternating in vertical and horizontal positions) connected in series. These investigators observed that even with the same filter and the same kind of particles, differences in flow rate and particle size may lead to three different results: gradual constriction of all junctions leading to nonretention, blocking of some junctions leading to nonretention, and blocking of some junctions leading to a complete blocking of the bed. A washing out technique was used by Hsiung (1974) to determine the porosity of deposits. Knowledge of the deposit porosity alone is not sufficient to define the effect of particle deposition. Furthermore, the possibility that deposit characteristics may be altered by the washout technique casts some uncertainty on the validity of his experimental findings.

Payatakes et al. (1977) calculated the increase in pressure drop of clogged filters as a function of the specific deposit and the morphology of deposits. Their calculations suggest that constriction clogging is dominant in the filtration of clay suspensions. On the other hand, in the filtration of ferric and alum flocs, deposits are likely to be in the form of smooth coatings. In a more recent study, Pendse et al. (1978) developed a diagnostic technique for the determination of deposit morphology based on dispersion measurements in clogged filter beds. The method is inferential, and its principle can be summarized as follows. Based on pressure drop measurements of a clogged filter bed and assumed deposit morphology, a quantitative description of the structural change of the filter bed due to particle deposition can be obtained. Based on this information, the response of the bed to an injection of a tracer pulse at its inlet, namely, the traveling of the tracer pulse throughout the bed, can be predicted. The prediction can then be compared with the appropriate tracer dispersion measurement, and an agreement between prediction and measurement can be construed as a validation of the assumed morphology. Similarly, discrepancy between theory and measurement would suggest that the assumed morphology is incorrect. The use of dispersion measurements in a clogged filter bed for the purpose of studying the effect of particle deposition on the structural charge of clogged filter media has been attempted earlier by Mints and Meltser (1970). What distinguishes the work of Pendse et al. from that of Mints and Meltser is the recognition that the effect of particle deposition on filter media cannot be deduced from dispersion measurements alone, and the dispersion characteristics of a clogged filter differ from those of a clean filter. Mints and Meltser's interpretation of dispersion data tacitly assumed that particle deposits form a relatively smooth coating outside filter grains which certainly is not always true as shown by the dispersion measurements of Pendse et al. (1978).

Quantitative Relationships

A number of investigators (Camp, 1964; Mohanka, 1969; Deb, 1969; Ives, 1969; Sakthivadivel et al., 1972) have presented relationships between the local pressure gradient of a clogged filter bed and the extent of particle deposition. The starting point was usually the Carman-Kozeny equation. The effect of deposition was assumed to increase the grain size and to decrease the local porosity on the basis that deposition over filter grains is uniform, resulting in the formation of a smooth deposit layer outside the grains. In general, their results can be expressed as

$$\frac{\partial p/\partial z}{(\partial p/\partial z)_o} = \prod_{i=1}^{M} (1 - p_i\sigma)^{a_i} \tag{44}$$

The number of terms involved M, the coefficients p_i's, and the exponents a_i's depend upon the particular model used to characterize the filter bed.

Pendse et al. (1978), based on assumed deposit morphology, obtained relationships between the extent of deposition and the structural variables characteristic of filter media. Two types of deposit morphology corresponding to two limiting cases were considered. In the first case, particle deposition is considered to be in the form of uniform smooth coating outside filter grains (smooth coating mode). In the second case, the deposition of particles means the lodgement of particles within pore constrictions of the bed such that the pore constrictions become clogged (blocking mode). The following expressions were obtained.

Smooth Coating Mode. The result of deposition is to form a layer of smooth coatings outside filter grains. Consequently, filter grain size increases, and the porosity decreases. No flow passage is completely blocked unless the pores are completely filled:

Grain size:

$$a_c = a_{c_o}\left[1 + \frac{\sigma}{(1-\epsilon_o)(1-\epsilon_d)}\right]^{1/3} \tag{45}$$

Porosity:

$$\epsilon = \epsilon_o - \frac{\sigma}{1-\epsilon_d} \tag{46}$$

Interstitial velocity:

$$V_\infty = V_{\infty o}\left\{\frac{1}{1 - \dfrac{\sigma}{1-\epsilon_d}}\right\} \tag{47}$$

To be consistent with the assumptions that deposits form smooth coatings, it is essential that the deposited particles should be contiguous to each other.

Blocking Mode. The result of deposition is the lodgement of particles within pore constrictions. Certain pore constrictions, therefore, become clogged and not open to flow. It is obvious that an overall property such as porosity ϵ cannot be used to describe the extent of bed clogging if blocking mode is the dominant mechanism. The use of an effective porosity to distinguish the part of void space available for suspension flow from the total void space is necessary. More conveniently, the problem can be considered in terms of the unit bed element concept with the constricted tube model. Let N_o be the number of unit cells (that is, constricted tube) per unit cross sectional area of a clean bed and N be the number of unit cells remaining open when the bed has become partially clogged. The extent of bed clogging can be characterized by the value of N, and a quantitative relationship between N, N_o, and σ can be derived. If all the unit cells are of the same size, one has

$$N = N_o - \frac{l\,\sigma}{m} \tag{48}$$

If the particle aggregates lodged in pore constrictions can be approximated by a spherical geometry, the minimum amount of deposit to block off one unit cell is

$$m_{\min} = \frac{4\pi}{3} r_c^3 (1-\epsilon_d) \tag{49}$$

To allow for the fact that particle aggregates lodging in the constriction may be more than the minimum amount, an adjustable parameter β can be introduced, or

$$m = \frac{4}{3}\pi r_c^3 (1-\epsilon_d)\beta \tag{50}$$

According to the constricted tube model of Payatakes et al. (1973*a*), l is found to be

$$l = 2a_c\left[\frac{\pi}{6(1-\epsilon_o)}\right]^{1/3} \tag{51}$$

Combining Equations (48), (49), and (51), we get

$$\frac{N}{N_o} = 1 - \frac{3a_c\sigma}{2\beta(1-\epsilon_d)r_c^3[6(1-\epsilon_o)]^{1/3} N_o \pi^{2/3}} \tag{52}$$

The relationships governing other pertinent filter media variables are

$$a_c = a_{c_o} \tag{53}$$

$$\frac{V_\infty}{V_{\infty o}} = \frac{N_o}{N} \tag{54}$$

Equations (45) to (46) and (51) to (54) can be used to obtain expressions relating the amount of deposition σ and the changes of pressure gradient and the filter coefficient. Based on the Carman-Kozeny equation, the following results are obtained.

For smooth coating mode:

$$\frac{(\partial p/\partial z)}{(\partial p/\partial z)_o} = \left[\frac{1}{1 - \dfrac{\sigma}{\epsilon_o(1-\epsilon_d)}} \right]^3 \left[1 + \frac{\sigma}{(1-\epsilon_d)(1-\epsilon_o)} \right]^{4/3} \tag{55}$$

For blocking mode:

$$\frac{(\partial p/\partial z)}{(\partial p/\partial z)_o} = \frac{V_\infty}{V_{\infty o}} = \left[1 - \frac{3a_c\sigma}{2\beta(1-\epsilon_d)r_c^3[(1-\epsilon_o)]^{1/3} N_o \pi^{2/3}} \right]^{-1} \tag{56}$$

The same relationship [that is, Equations (45) to (46), (53) to (55)] can also be used to assess the effect of particle deposition on the rate of filtration. Note that under either hypothesis, particle deposition has resulted in the change of the interstitial velocity, the dimension of the collector, and the overall porosity but has no effect on the geometrical configuration of the collector. The results of trajectory calculation therefore can be applied to a clogged filter provided these changes are properly taken into account.* Based on this argument and the results of Rajagopalan and Tien (1976), the following expressions relating σ, the specific deposit, and λ, the filter coefficient, are obtained (Tien et al., 1979).

For smooth coating mode:

$$\frac{\lambda}{\lambda_o} = B_1\left(\frac{A_s}{A_{s_o}}\right)\left[1 + \frac{\sigma}{(1-\epsilon_o)(1-\epsilon_d)}\right]^{17/24} + B_2\left(\frac{A_s}{A_{s_o}}\right)\left[1 + \frac{\sigma}{(1-\epsilon_o)(1-\epsilon_d)}\right]^{4.4/3} + B_3\left(\frac{A_s}{A_{s_o}}\right)^{1/3}\left[1 + \frac{\sigma}{(1-\epsilon_o)(1-\epsilon_d)}\right]^{4/9} \tag{57}$$

$$B_1 = \frac{(1-\epsilon_o)^{2/3}}{\eta_o} A_{s_o} N_{Lo_o}{}^{1/8} N_{R_o}{}^{15/8} \tag{58}$$

$$B_2 = \frac{3.376 \times 10^{-3}}{\eta} (1-\epsilon_o)^{2/3} A_{s_o} N_{G_o}{}^{1.2} N_{R_o}{}^{-0.4} \tag{59}$$

$$B_3 = \frac{4A_{s_o}{}^{1/3} N_{Pe_o}{}^{-2/3}}{\eta_o} \tag{60}$$

$$\left(\frac{A_s}{A_{s_o}}\right) = \left[\frac{1-(1-\epsilon)^{5/3}}{1-(1-\epsilon_o)^{5/3}}\right] \left[\frac{2-3(1-\epsilon_o)^{1/3}+3(1-\epsilon_o)^{5/3}-2(1-\epsilon_o)^2}{2-3(1-\epsilon)^{1/3}+3(1-\epsilon)^{5/3}-2(1-\epsilon)^2}\right] \tag{61}$$

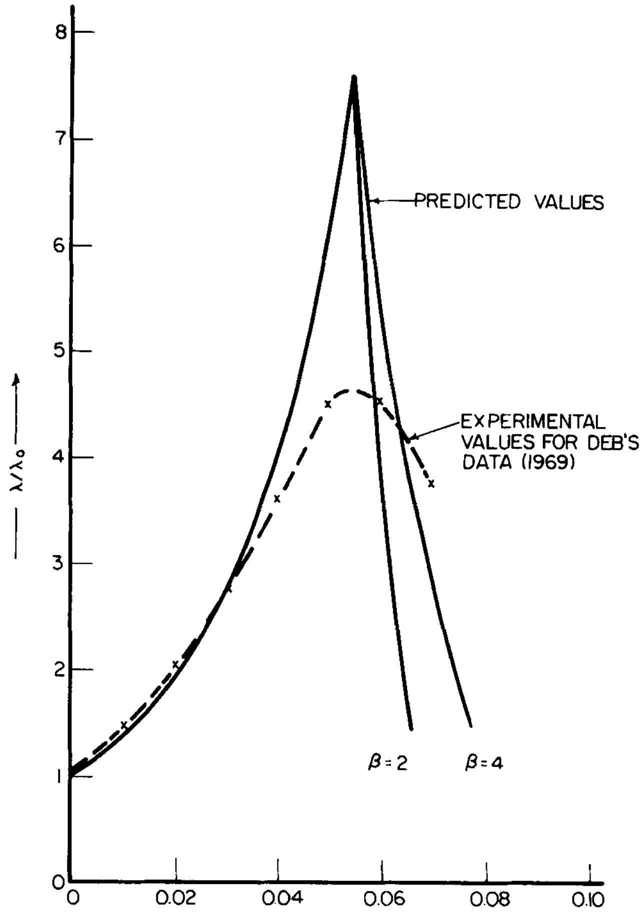

Fig. 11. Variation of filter coefficient with specific deposit (Tien et. al., 1979).

For blocking mode:

$$\frac{\lambda}{\lambda_o} = B_1\left[1 - \frac{\sigma}{2\beta\pi^{2/3}r_c^3[6(1-\epsilon_o)]^{1/3}(1-\epsilon_d)N_o}\right]^{1/8} + B_2\left[1 - \frac{\sigma}{2\beta\pi^{2/3}r_c^3[6(1-\epsilon_o)]^{1/3}(1-\epsilon_d)N_o}\right]^{1/2} + B_3\left[1 - \frac{\sigma}{2\beta\pi^{2/3}r_c^3[6(1-\epsilon_o)]^{1/3}(1-\epsilon_d)N_o}\right]^{2/3} \tag{62}$$

It was found (Tien et al., 1978) that for conditions encountered in deep bed filtration, Equation (57) predicts that λ increases with increasing σ. The opposite results are obtained from Equation (62). A typical example is shown in Figure 11. It is interesting to note that it has been observed experimentally (Ives, 1961; Camp, 1964; Deb, 1969) that λ first increases with σ and then decreases, suggesting the possibility that the deposit morphology may undergo a change from one type to another during a filtration cycle depending upon the extent of particle deposition. This point will be discussed in some detail in later sections.

Dynamics of Particle Deposition

The results of Equations (55), (56), (57), and (62), which relate the extent of deposition with filter performance, were obtained on the basis of assumed morphology. The selection and application of these expressions for predicting filter performance, therefore, cannot be made without additional evidence or assumption on deposit morphology. A complete solution to the problem requires

* It may be argued that the presence of a deposit layer outside the collector surface may change the collector-particle surface interaction. However, in view of the inconclusive results we have on the effect of surface interaction on deposition, this possible change cannot be considered at the present time.

a theory which provides detailed information about the deposition process, including the formation and growth of particle deposits as a function of time. Such a theory is not yet available, although some progress has been made along this direction. Payatakes and Tien (1976) and Payatakes (1977*b*) have presented mathematical models for aerosol particle deposition with dendritelike pattern and related collection efficiency and pressure drop as functions of the degree of deposition. It is likely that the same approach can be applied to hydrosol systems. Tien, Wang, and Barot (1977) and Wang et al. (1977) argued that deposition phenomena, in general, can be studied in terms of the two properties which are characteristic of deposited particles and approaching particles, respectively. The shadow effect refers to the fact that once a particle is captured by a collector, certain parts of the collector surface near the deposited particle would no longer be accessible to approaching particles (and these areas are called shadow areas). The degree of uniformity of deposited matters (or the lack of it) is a direct consequence of the magnitude of these shadow areas. The morphology of particle deposits can be determined by the fact that the spatial distribution of particles in a dilute suspension follows Poisson's law, and the presence of deposited particles allows approaching particles to be collected by the collector as well as by the already deposited particles. This principle was adopted by Beizaie (1977) in his study of deposition on single isolated spherical collectors. His results indicate that the deposition process, in general, consists of three steps. The first, or the initial stage, may be called the clean collector stage during which the presence of deposited particles have negligible effect. The second stage, or the dendrite growth stage, is characterized by the formation and growth of particle dendrites. During this stage, an increasing amount of particle deposition is effected by deposition on deposited particles. The third stage of the process is termed the open structured solid growth stage. Particle collection is entirely due to deposition on deposited particles, and individual particle dendrites become less distinguishable as they become merged and interconnected. A direct application of Beizaie's results to filter clogging, of course, would be inappropriate, since the use of the isolated sphere model precludes consideration of the effect due to the presence of neighboring grains. However, with the use of a more realistic porous media model (such as the constricted tube model), Beizaie's method on deposition dynamics can be applied advantageously to the study of filter clogging.

Another aspect of the problem which warrants consideration is the reentrainment of deposited particles due to hydrodynamic drag force. Both Shekhtman (1961) and Mackrle et al. (1965) included reentrainment in the phenomenological rate expression of deep bed filtration and assumed the degree of reentrainment to be proportional to the extent of deposition. Payatakes et al. (1977) advanced the hypothesis that filter clogging can be described by the reentrainment of particle deposits and subsequent redeposition of the particle aggregates in pore constrictions, resulting in the closure of certain flow elements. Leclerc and Vu (1972) studied the retentivity of filter media but provided no mechanistic description of reentrainment.

To initiate a rigorous study of the reentrainment phenomenon, methods which can be used to estimate the hydrodynamic drag force experienced by particle aggregates of various configurations in shear flow field have to be developed. Studies of this kind have not been reported in literature. It also requires knowledge of the adhesive and cohesive forces between collector and particle deposits and within particle aggregates, which, for most systems, are not available.

DESIGN AND OPTIMIZATION

The practical concern in the study of deep bed filtration is the development of design methods and optimization techniques. Design calculations, in essence, require prediction of filter performance from a given set of operating and system variables. Such a prediction can be obtained from the solution of the phenomenological equations using appropriate rate and pressure gradient expressions. This is, in fact, the approach adopted by Ives (1960) in his pioneering work on the rational design of deep bed filtration. Subsequent studies (Mohanka, 1969; Deb, 1970; Ives, 1962, 1969) have refined and extended Ives' earlier work by the use of different rate expressions and the inclusion of other factors such as the inhomogeneity of multilayer filter media. The major limitation of this approach is that the rate and pressure gradient expressions can be obtained only if relevant experimental data are available. Even for the same system, extrapolation of results obtained from a small experimental filter to larger units is often questionable.

Two recent studies which aim at the development of a truly predictive method of deep bed filtration were made by Wnek, Gidaspow, and Wasan (1975) and Tien, Pendse, and Turian (1979). In the work of Wnek, Gidaspow, and Wasan, the rate of deposition was based on the results of Yao et al. (1971) and corrected for the presence of surface forces by Equation (42). A charge balance was made as part of the phenomenological description, which accounts for the variation of particle-grain interaction due to particle deposition. The pressure gradient expression was formulated on the basis that deposits are in the form of relatively smooth coatings. The work of Tien et al. (1979) considered the effect of deposit morphology and its evolution during the course of filtration. Their formulation is based on the observations of Maroudas and Eisenklam (1965) and their own experiments (Pendse et al., 1978). The main features of their work are:

1. The rate of deposition is assumed to be given by the expression developed by Rajagopalan and Tien (1976) from trajectory calculation results. The pertinent variables, such as grain diameter and interstitial velocity, are considered to vary with the amount of deposition and with the resulting deposit morphology.

2. Both the constricted tube model and also Happel's model are used to represent the filter bed. Happel's model is used during the initial period of deposition when the deposit morphology is of the smooth coating mode and the constricted tube model for the later stages of deposition when transition to the blocking mode is more likely.

3. The transition from one type of morphology (smooth coating mode) to another (blocking mode) is taken to occur when the specific deposit reaches the transition value σ_{tran}. This transition in morphology is consistent with observed behavior, namely, that the filter coefficient initially increases with the specific deposit σ and subsequently decreases. The value of σ_{tran} thus represents the value of σ which gives a maximum for λ. It is estimated that σ_{tran} is within the range of 0.03 to 0.05.

4. The amount of deposit to block a unit cell is expressed in terms of the equivalent volume of multiples of spheres having diameters equal to that of the constriction of the unit cell. From physical arguments, this value is estimated to be within the range of 2 to 4.

5. Deposition ceases if the interstitial velocity exceeds a critical value v_{cr} in accordance with the findings of Maroudas and Eisenklam (1965).

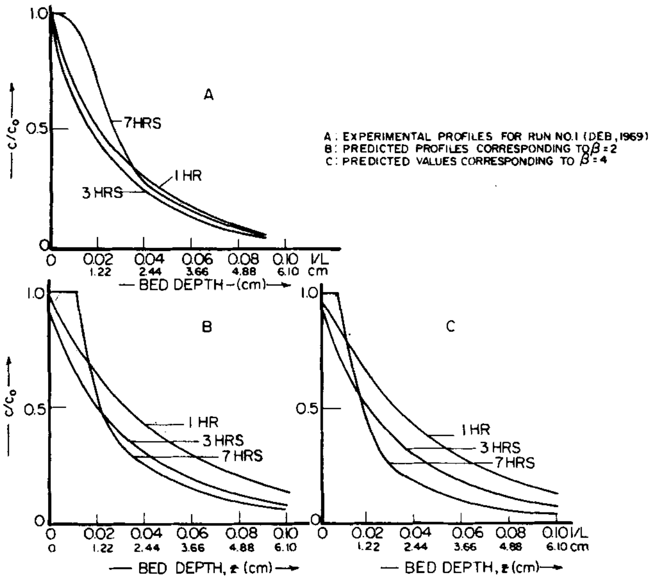

Fig. 12. Comparison of experimental and predicted concentration profiles (Tien et. al., 1979).

6. The Carman-Kozeny equation, together with the assumed morphology, can be used to estimate the pressure drop increase.

7. The algorithms developed by Herzig et al. (1970) are used to integrate the pertinent filter equations to obtain histories of filtrate quality and pressure drop.

Comparisons of simulated results with experimental data from five earlier studies were made (Deb, 1969; Ives, 1961; Camp, 1964; Mehter, 1970; Rimer, 1968). These results should be assessed in the context that the accuracy of these data is, on the whole, not high and that furthermore, some of the measurements were taken under rather poorly controlled conditions. The comparisons with Deb's work are given in Figures 12 and 13. It should be noted from Figure 12, for example, that the reversal in filtrate quality observed experimentally was confirmed partially by prediction. For the pressure drop increase, both theory and experiment show a break in the Δp vs. time curve at 4 hr even though the respective values differ by a factor of 2. The basic framework, therefore, appears to be valid, although further refinements of this method undoubtedly will be made when more accurate experimental data become available.

In contrast to the rational approach is the use of statistical models for filter design calculations. Hudson (1963) developed a functional design method which assumes that the performance of a filter can be characterized by the quality of filtrates and the length of filter run. It is assumed that empirical expressions relating these two quantities with system and operating variables (flow rate, porosity, grain size, floc strength, etc.) can be established from plant or pilot plant data and are therefore available. Hsiung (1967) and Hsiung and Cleasby (1968), based on data obtained from filtration of ferric floc in a shallow filter bed, proposed a statistical model for filter performance, and their results have been successfully applied to design calculations (Hwang and Bauman, 1971; Conley and Hsiung, 1969). Again, the limitations are the requirement of extensive experimental data and the difficulty of generalizing the model beyond a particular system.

In design calculations, optimum conditions often imply minimum cost. The many unresolved problems in the study of deep bed filtration make such an undertaking impractical at present time. Mints (1968) argued that

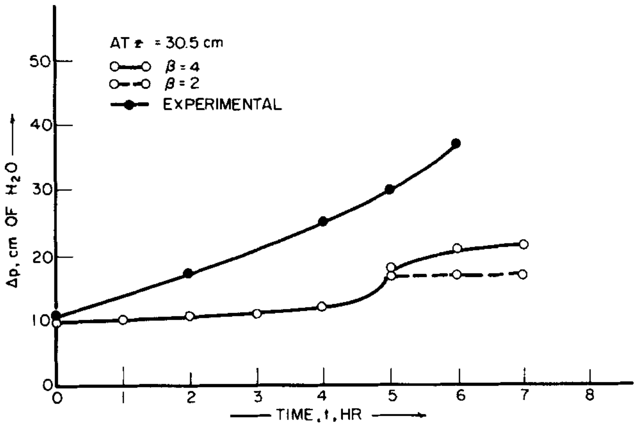

Fig. 13. Comparison of experimental and predicted values for pressure drop data (Deb, 1969) (Tien et. al., 1979).

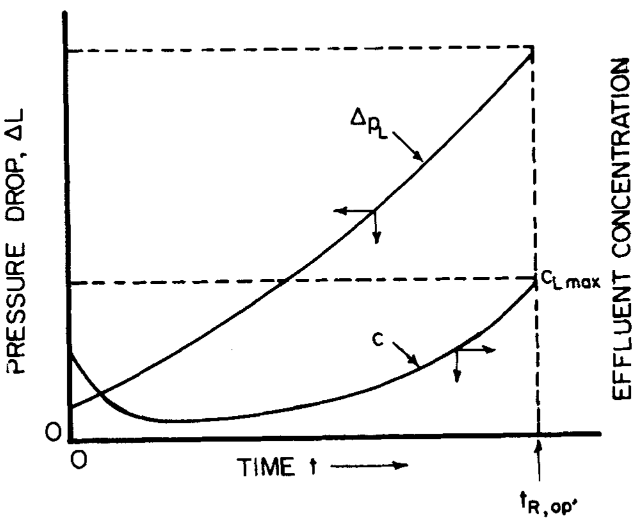

Fig. 14. 'Optimized' filter according to Mints (1968). Head loss and residual volume fraction reach their limiting values simultaneously.

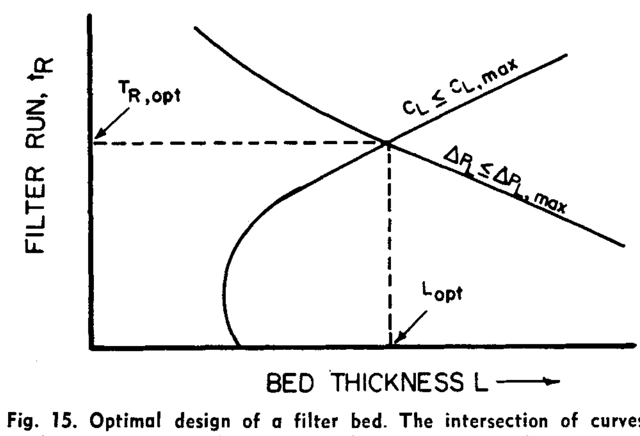

Fig. 15. Optimal design of a filter bed. The intersection of curves for limiting residual volume fraction ($c_L \leq_{L,max}$) and limiting head loss ($\Delta P_L \leq \Delta P_{L,max}$) gives the optimal filter depth and optimal duration of the filter run.

an operating optimum is obtained if both the filtrate quality and pressure drop reach their allowable upper limit at the same time (see Figure 14). This optimum can be determined by the solution of the phenomenological equations to obtain the values of filter bed height L and filtrate run θ under the constraints that the filtration quality and overall pressure drop should not exceed preassigned limits and with specific grain size and filtrate rate. Two such curves can be obtained as shown in Figure 15, and their intersecting point represents the opti-

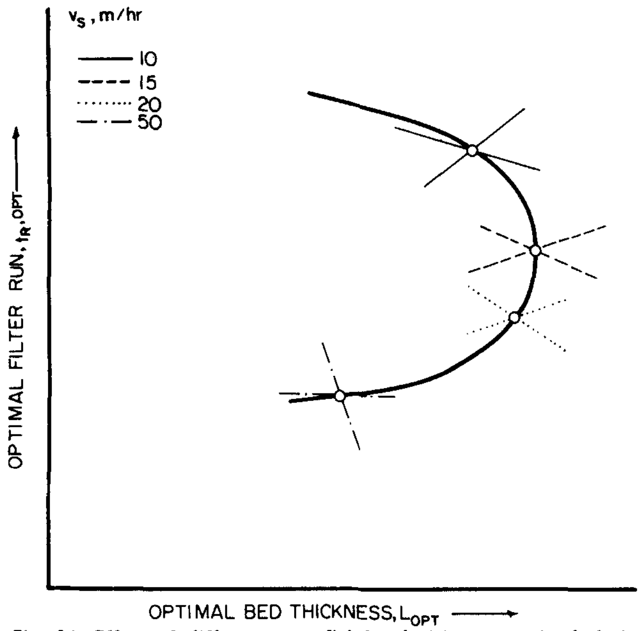

Fig. 16. Effect of different superficial velocities on optimal design ($L_{opt}, t_{r,opt}$).

mum defined by Mints. Variations of the filtrate rate would produce different but similar intersecting points, the loci of which would yield a curve as shown in Figure 16. Families of such curves can be obtained if the grain size is considered to be a variable. This method has been discussed in detail by Ives (1969) and has been applied by Kreisol et al. (1968). Letterman (1976) pointed out that the optimum method based on Mint's hypothesis has certain limitations, particularly when it is possible to achieve optimum conditions through pretreatment. The concept of deposit distribution was introduced as a basis of achieving operational optima. Experimental data of filtration of clay suspensions with different pretreatment were presented to substantiate his proposition.

PRETREATMENT

Addition of coagulants into dilute solid-liquid suspensions prior to depth filtration can lead to increased capture efficiency and, with proper filter design, even increased capacity. Early reviews of pretreatment methods were given by Mints (1969) and O'Melia and Stumm (1967).

There are basically two types of coagulants used: alums [such as $M_2{}^{I}SO_4M_2{}^{III}\,(SO_4)_3\,(H_2O)_{24}$ or $M^{I}M^{III}\,(SO_4)_2\,(H_2O)_{12}$ where M^{I} denotes a monovalent metal such as $N_a{}^{+}$, K^{+} and M^{III}, a trivalent metal such as Al^{+3} and

TABLE 6. EXAMPLES OF CATIONIC, NONIONIC, AND ANIONIC POLYELECTROLYTES
(SOURCE: GRUTSCH AND MALLATT, 1977)

Polyelectrolyte description	Structural type	Functional group	Example	
Cationic	Amines	$—\overset{\oplus}{N}(H)(R)—R$	$\sim(—CH_2—CH_2—\overset{\oplus}{N}H_2—)_x\sim$ $Cl^{\ominus}$	Polyethylenimine hydrochloride
	Quaternary	$—\overset{\oplus}{N}(R)(R)—R$	$\sim(—CH_2—CH—)_x\sim$, CH bearing a 4-pyridinium ring with $\overset{\oplus}{N}—CH_3$ $Cl^{\ominus}$	Poly(N-methyl-4-vinyl) pyridium chloride
Nonionic	Polyamide	$—C(=O)—NH_2$	$\sim(—CH_2—CH(C{=}O\,NH_2)—)_x\sim$	Polyacrylamide
	Polyalcohol	—OH	$\sim(—CH_2—CH(OH)—)_x\sim$	Polyvinylalcohol
Anionic	Carboxylic	$—C(=O)—O^{\ominus}$	$\sim(—CH_2—C(R)(C{=}O\,O^{\ominus})—)_x\sim$	Poly(meth)acrylic acid
	Sulfonic	$—S(\rightarrow O)(\rightarrow O)—O^{\ominus}$	$\sim(—CH_2—CH(O{\leftarrow}S{\rightarrow}O\,O^{\ominus})—)_x\sim$	Polyvinylsulfonate

Fe^{+3}] and polyelectrolytes (cationic, nonionic or anionic). Polyelectrolytes are macromolecules with large numbers of functional groups which ionize in aqueous solution. Typical polyelectrolytes are listed in Table 6, taken from Grutsch and Mallatt (1977).

Role of Flocculants in Depth Filtration

The effects of flocculants on deep bed filtration are manifold, and not all of them are completely understood. They change the feed characteristics by destabilizing the colloidal particles and inducing flocculation. Furthermore, they facilitate the capture of suspended flocs by the grains of the filter bed. In the case of polyelectrolytes, the deposits formed are very strong and can withstand high shear stresses without reentrainment. The same is not true in the case of pretreatment by alum; the flocs formed then are readily torn by small shear stresses.

Stumm and Morgan (1962) and O'Melia and Stumm (1967) showed that the action of alums is primarily due to their hydrolysates, namely, the various metalhydroxo complexes formed in aqueous solution. The relative concentrations of the various hydrolysates are strong functions of *p*H. Flocculation with the aid of these materials takes place in two steps: neutralization of the surface charges of particles by chemisorption or simple adsorption of metal-hydroxo complexes of opposite charge sign and formation of flocs composed of particles encapsulated by adsorbed metal-hydroxo complexes and interconnected with bridges of metal-hydroxo polymers. Such flocs are weak and contain substantial amounts of water trapped in interstitial spaces.

The action of polyelectrolytes is explained as follows (LaMer and Healy, 1963). To be effective in destabilization, a polymer molecule must contain chemical groups which can interact with sites on the surface of the colloidal particle. When a polymer molecule comes into contact with a colloidal particle, some of its active groups get attached to the particle, while the rest of the molecule extends away from the particle into the liquid. If a second particle or a collector with some vacant adsorption sites comes into contact with this extended segment, the two bodies form a particle-polymer-particle (or grain) complex, with the polymer acting as a bridge. If a long time passes without the first particle coming into contact with another body, the groups of the extended part of the adsorbed polymer may also adsorp on sites of the same particle, so that the polymer is no longer available to serve as a bridge.

Optimal Dosage

One of the most important steps in applying a flocculant is determination of the optimal dosage. Very small dosages may not be sufficient, while too large a dosage usually leads to inversion of the particle charge, restabilization of the colloid, and consequently poor filtration.

This is shown dramatically in Figure 17, where the effect of polyelectrolyte dosage on the ζ potential of particles suspended in API separator effluent is shown, and in Figure 18, where the analogous study is made for coke fines in hydraulic decoking waters (Grutsch and Mallatt, 1977). Such curves are extremely useful. They can be applied to determine the optimal dosage for each electrolyte, since this is the amount of polyelectrolyte per unit volume at which the ζ potential goes through zero. They can also be used in a cost effectiveness comparison of various electrolytes. Finally, they indicate the region of dosages which corresponds to good destabilization conditions (-5 mV $< \zeta <$ 5 mV). A similar approach can be used for alums. Alternatively, results obtained from jar tests may provide an approximate indication about the optimum dosage in the absence of other data.

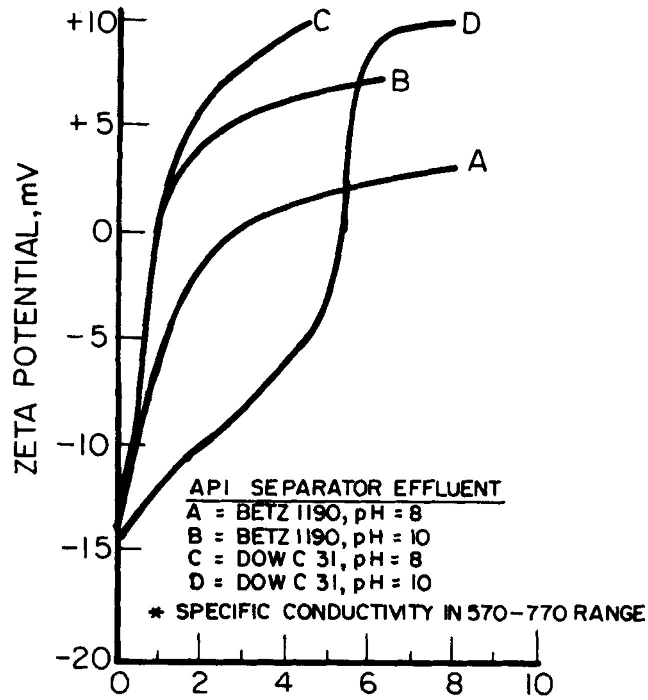

Fig. 17. Zeta potential-cationic polyelectrolyte titration curves of API separator effluent (Grutsch and Mallatt, 1977).

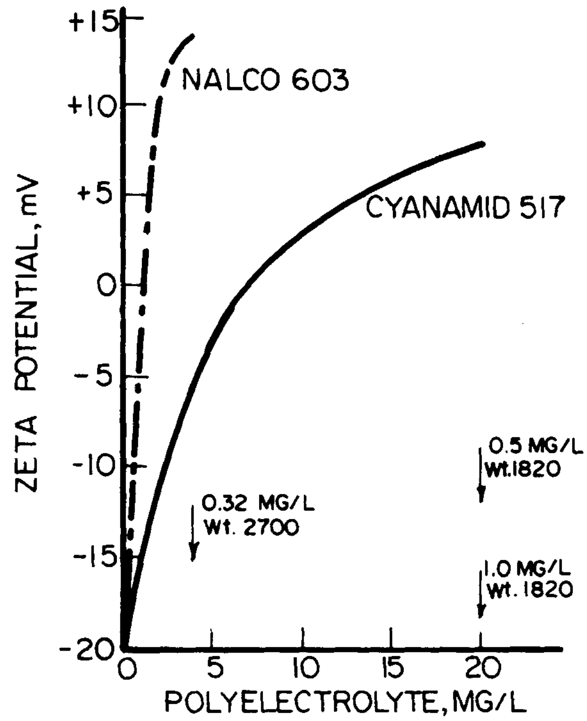

Fig. 18. Zeta potential-cationic polyelectrolyte titration curves of coke fines in hydraulic decoking waters (Grutsch and Mallatt, 1977).

Overdosing should be avoided meticulously not only because of the danger of restabilization and breakthrough, but also because of the concomitant increase in operational costs. Not only higher coagulant costs are incurred (especially in the case of polyelectrolytes), but larger amounts of sludge (from backwashing) must be treated and disposed of, especially when alum is used.

Polyelectrolytes vs. Alum

A comprehensive study of the relative merits of using alum or some polyelectrolyte is still unavailable.

An interesting study of this problem was made by Adin and Rebhun (1974) using two different sand beds (coarse: $\langle d_g \rangle = 1.21$ mm; medium: $\langle d_g \rangle = 0.62$ mm) and suspensions of kaolinite in softened Haifa tap water. Two flocculants were compared: the cationic polyelectrolyte polydiallyldimethyl ammonium halide and conventional alum. Their findings are summarized below, since they are thought to be of somewhat general validity.

1. There is a clear overdose effect at more than the optimal dosage of 0.5 mg/l of polyelectrolyte. The optimal dosage for deep bed filtration is the same with that determined from jar tests. However, the polymer dose range giving good removal in deep bed filtration is substantially wider than the corresponding range in volume flocculation.

2. The residual turbidity C_L/C_o vs. time curve shows an initial decrease (initial increase of the filtration coefficient λ) when either alum or polyelectrolyte is used. The stage of initial increase of the filter coefficient with alum is shorter and sharper than with polymers. However, use of polymers provided better efficiency during the advanced stages. The breakthrough at the last stages after treatment with polymer is milder than with alum. Breakthrough with alum occurs earlier than with polymers.

3. Plots of the total pressure drop ΔP_L vs. accumulated filtrate volume per unit filter area indicated that in the case of medium sand ($\langle d_g \rangle = 0.62$ mm) and treatment with alum, the increase in head loss is nearly linear, becoming slightly curved only at the end. It is also substantially lower than the nearly exponential increase in head loss when polymers are used in pretreatment. For coarse grain beds ($\langle d_g \rangle = 1.21$ mm), the run with alum was interrupted early owing to concentration breakthrough. The run with polymer pretreatment did not show a breakthrough; it was stopped only when the maximum pressure drop was reached.*

4. The total filtrate output for alum treatment was about 30% less in the case of the coarse grain filter than that obtained with the medium grain filter. On the contrary, the total filtrate output for polyelectrolyte treatment using the coarse grain filter was at least 500% higher than that achieved using the medium grain filter. This increase in capacity is due to the fact that a greater part of the filter length participated in the filtration process.

It follows that deep bed filters of coarse sand with polyelectrolyte pretreatment can give efficient filtration combined with high capacity.

5. The velocity of the clogging front is substantially higher when alum is used. This, again, is due to the ease of reentrainment of alum flocs.

6. Changing the superficial velocity u_s from 5 to 10 m/hr decreased the cycle output by about 33% with alum and only by about 8% with polymers.

The change of velocity did not affect penetration when polyelectrolytes were used, but with alum the effect was to change the criterion for terminating the cycle from reaching the limiting pressure drop to having concentration breakthrough.

Increasing the superficial velocity from 10 to 20 m/hr led to immediate breakthrough, showing alum to be inadequate as a filtration aid under these conditions. The same velocity change, when using polyelectrolytes, increased the cycle output by 25% and decreased the need for backwash by about 20%.

7. The optimal depth for polymers is much lower than for alum. The difference increases with increasing superficial velocity.

8. Under conventional conditions (medium or fine grains, say $d_g < 0.6$ mm, and relatively small superficial velocities, say $u_s < 10$ m/h) alum is advantageous. However, for coarse grain filter beds and high velocities, polyelectrolytes are clearly superior. An advantage of the latter design is increased cycle output (increased capacity); this results in less frequent backwashing and more concentrated sludge.

Grutsch and Mallatt (1977) favor the use of polyelectrolytes over that of alum for the depth filtration of intake water, primary effluent, and activated sludge plant effluent in oil refineries. The main reason cited is that the optimal polyelectrolyte dosage is directly proportional to the amount of colloidal material and that the sludge produced from backwashing is of minimal amount and easy to handle. On the contrary, massive amounts of alum are necessary for the same applications, the amount and nature of the colloidal material being almost irrelevant. This type of destabilization (sweep floc) results in generating large amounts of wet alum (or iron) sludges, which are difficult and costly to dewater. These authors conclude that alum is to be avoided in granular media filtration (even though it is widely used in water clarification) because of the sludge problem and the fact the use of polyelectrolytes results in significantly lower operating and capital costs.

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NOTATION

a = coefficient of filtration rate expression, Equation (9)
a_p = particle radius
a_c = collector radius
a_i = exponents of pressure gradient expression, Equation (44)
A_s = quantity defined by Equation (31)
b = radius of the outer shell of the sphere-in-cell model
B_1, B_2, B_3 = quantities defined by Equations (58), (59), and (60)
c = volume concentration of particulate matters in liquid
c_j = volume concentration of particles of size j in liquid
c_o = volume particle concentration of inlet stream
c_i = volume particle concentration on liquid initially present in bed
c_L = volume concentration of particles in exit stream with bed height L
d_g = grain diameter
d_i = constriction diameter of size i
d_j = constriction size
d_p = particle diameter
$\langle d_{3_i}{}^3 \rangle$ = average value of the cubic power of constriction diameter
D = particle diffusion coefficient in liquid
e = electronic charge
e_1, e_2, e_3, e_4 = exponents of Equation (10)
$f_\lambda(\alpha, \sigma)$ = function describing the effect of particle depo-

* The behavior of the head loss curves for the coarse filter indicates that constriction clogging by particle cluster straining was not a dominant factor until near the end of the run, even when polyelectrolytes were used. This is due, apparently, to the large size of the constrictions in such a bed.

sition on filter coefficient
$F_1(\boldsymbol{\beta}, \sigma)$, $F_2(\boldsymbol{\beta}, \sigma)$ = functions describing the effect of particle deposition on pressure gradient
F_E = double layer force
F_L = London force
$G(\boldsymbol{\gamma}, c, \sigma)$ = function expression for filtration rate
H = Hamaker's constant
I_c = number of different sizes of constricted tube in a unit bed element
J = particle flux
J^0 = particle flux for case of no surface force
$J^0{}_s$ = particle flux for case of surface force controlling
k = Boltzmann's constant
K = equivalent rate constant defined by equation (43)
l = physical dimension of unit bed element
m = amount of deposit required to block one constriction
$m_{\min}$ = quantity defined by Equation (49)
m_j = ionic concentration of the j^{th} ionic species
M = number of terms of the pressure gradient expression, Equation (44)
n_i = number fraction of constricted tube of size i in a unit bed element
n_j = number fraction of unit cells with constriction size j
N_o = number of constricted tubes per unit cross-section area of clean filter bed
N = number of constricted tubes open to flow per unit cross-section area of filter bed
N_{R_s} = dimensionless group defined as $\frac{1}{2}N_R$
N_L, N_{E1}, N_{E2}, N_G, N_{Lo}, N_{Pe}, N_R, N_{Ret} = dimensionless groups defined in Table 4
p = pressure
p_j = coefficients of Equation (44)
P_1 = coefficient of Equation (10)
P = quantity defined by Equation (32)
r_c = radius of constriction
t = time
T = temperature
u = superficial velocity of filter bed
$V_{\infty o}$ = interstitial velocity of clean filter bed
V_∞ = interstitial velocity of filter bed
V_F = clogging fluid velocity given by Equation (19)
z = axial distance
z_j = valency of j^{th} species

Greek Letters

$\boldsymbol{\alpha}$ = parameter vector of function $f_\lambda(\boldsymbol{\alpha}, \sigma)$, Equation (7)
α_{sp} = retardation factor for London force
α_1, α_2, α_3 = exponents of Equation (8)
$\boldsymbol{\beta}$ = parameter vector of functions $F_1(\boldsymbol{\beta}, \sigma)$ and $F_2(\boldsymbol{\beta}, \sigma)$
β_1, β_2 = components of $\boldsymbol{\beta}$
β = parameter defined by Equation (50)
$\boldsymbol{\gamma}$ = parameter vector of function $G(\boldsymbol{\gamma}, c, \sigma)$
δ = separation distance
ϵ = porosity of filter bed
ϵ_d = porosity of deposits
ϵ_o = porosity of clean filter bed
$\bar{\epsilon}$ = dielectric constant of liquid
ψ_{01}, ψ_{02} = surface potentials of particle and grain
ψ^* = dimensionless stream function for flow within a constricted tube
η = collection efficiency of collector
η_o = collection efficiency of clean collector
η_{BM} = collection efficiency due to Brownian motion
η_s = collection efficiency due to straining
η_{oi} = collection efficiency of constricted tube of type i
θ = corrected time defined by Equation (3)
ϕ = potential of surface interaction forces
κ = double-layer reciprocal thickness
λ_e = wavelength of electron oscillation
λ = filter coefficient
λ_o = clean filter coefficient
μ = viscosity
ρ = density of liquid
ρ_p = density of particles
σ = specific deposit (volume of deposited matter per unit volume of bed)
$\sigma_{\max}$ = ultimate specific deposit

Subscripts

o = clean filter

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Estimation of Phase Diagrams and Solubilities for Aqueous Multi-ion Systems

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Calculation of the phase diagram for saturated aqueous multi-ion solutions of strong electrolytes, based upon use of the thermodynamic solubility products, is illustrated for the four-ion system: Na^+, K^+, Mg^{++}, $SO_4^=$. Solid phases are identified along with the ion concentrations of the associated saturated solutions.

SCOPE

Solubility information for strong electrolytes in aqueous solutions containing several ions is often desired, such as when producing sodium hydroxide by reacting a sodium carbonate solution with lime, evaporating a natural brine containing various alkali and alkaline earth chlorides and sulfates to precipitate individual salts, calculating the vapor pressure of dissolved hydrochloric acid over a