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Effect of Pore Size and Particle Size Distribution on Granular Bed Filtration and Microfiltration

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Abstract: The paper reviews the effect of particle size distribution and pore size distribution on granular bed filter and crossflow microfiltration performance. The experimental results of the granular bed filter with pollen particles in suspension showed that the presence of large particles improved the filter efficiency of smaller particles in suspension. Microfiltration results with bi and tri-modal latex suspensions showed that the permeate flux and the quality were significantly affected by the particle size and its distribution, especially when the particle size was smaller than the pore size of the membrane. The mathematical model simulation results of granular bed filtration show that media pore size distribution is an important parameter of filtration for the particle removal and pressure drop across the filter.

Keywords: Microfiltration, bed filtration, pore size distribution, particle size distribution, modelling

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INTRODUCTION

Granular Bed Filtration

Granular bed filtration is used widely in municipal water treatment for the clarification of dilute suspensions of particles with a wide range of sizes. The particles have to be transported near the filter grains by different transport mechanisms and then adhered to the grain surfaces by various attachment mechanisms for their successful removal. Thus filtration is a complex process involving physico-chemical mechanisms and depends on various factors such as filtration rate, particle size, grain size, and influent concentration. Generally, filters have been designed based on past experiences, and no rational method has been followed due to the fact that most mathematical models available have been restricted to uniform sized media and particle size distribution. However, this is not the case in practice.

Crossflow Microfiltration

Crossflow microfiltration (CFMF) is a pressure-driven membrane process for separating particles, micro-organisms, large molecules and emulsion droplets and is increasingly important in water treatment and tertiary wastewater treatment. The filter medium is a micro-porous membrane with a separation limit in the range of 0.02 to 10 μm (which is between ultrafiltration and granular bed filtration). The performance of CFMF depends on the hydrodynamic conditions such as cross flow velocity and the applied pressure; and parameters such as particle size and its distribution, and membrane characteristics. Very little information is given in literature on the effect of particle size distribution on CFMF.

The paper reviews the experimental results on effect of particle size distribution on granular bed filter and CFMF performance. The effect of particle size distribution on deep bed filtration and CFMF performance is discussed based on the experimental results previously obtained by the authors with suspension containing different sizes of particles. The pore size distribution is another important parameter which is illustrated from a model simulation study (1).

EFFECT OF PARTICLE SIZE DISTRIBUTION IN GRANULAR BED FILTRATION

The results of experiments performed with sand as the filter medium (mean size = 0.11 μm; depth = 2 cm) and a suspension of pollen grains of different sizes in water are shown in Fig. 1. The size (D_{50}) of the pollen grains of Platane, Sorgho, Mais are 22 μm, 50 μm, and 85 μm respectively (2). This figure shows the results of bimodal (i.e. particles of two different

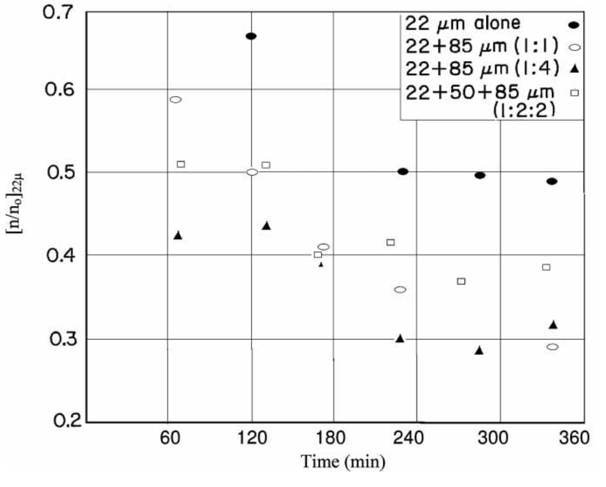

Figure 1. Effect of the combination of three particles (22, 50, and 85 μm) on the removal efficiency of 22 μm particles. ($d_c = 0.11$ cm, L = 2 cm, $C_o = 100$ mg/l and V = 5 m/h). Note $[n/n_o]_{22\ \mu m}$ is the effluent ratio of particles of 22 μm when particles of 22 μm are in combination with other particles.

sizes) and trimodal (particles of three different sizes) particles in suspension. Two and three different sizes of particles at a known concentration were mixed in the suspension, as shown in Fig. 1. The results obtained revealed that as the ratio of coarser particles to finer particles increases (in the case of bimodal particles), the removal efficiency of finer particles, which are in combination, also increases. It should be noted that the efficiency is expressed in terms of removal of fine particles (i.e. in this case, 22 μm particles). The results observed in the case of trimodal particles also confirm the above result. The results also show that the coarser the particles the better the removal efficiency of the finer particles. The improvement in the removal efficiency of particles of sizes 50 and 85 μm in the presence of 22 μm particles was not at all significant. The improvement was more when the ratio of coarse particles to fine particles increased.

EFFECT OF PARTICLE SIZE DISTRIBUTION IN CROSS FLOW MICROFILTRATION (CFMF)

Experiments were carried out with mono-dispersed latex suspensions (i.e. particles of one type) and with bimodal (i.e. particles of two different sizes) and tri-modal (particles of three different sizes) particles in suspension i.e. two and three different sizes of particles at a known concentration were mixed in the suspension as shown in Table 1 to study the effect of particle size and its distribution on CFMF performance. Details on the experimental

Table 1. Removal efficiency of combination of two particle sizes cross flow microfiltration (pressure = 70 kPa, cross flow velocity 1 m/s, Millipore membranes of 0.2 μm, latex particles of defined size and concentration in suspension)

Set no.	Run no.	Particle size (μm)	Concentration (mg/l)	Influent turbidity (NTU)	Permeate turbidity (NTU)	Removal efficiency (%)	Steady state flux ($L/m^2 \cdot h$)
1	1-1	0.6	20	124	0.09	99.9	450
	1-2	0.6 + 1.1	10 + 10	136	0.02	99.9	480
	1-3	0.6 + 3.0	10 + 10	108	0.03	99.9	525
2	2-1	0.1	20	40	5.4	86.5	220
	2-2	0.1 + 0.6	10 + 10	92	0.57	99.4	250
	2-3	0.1 + 1.1	10 + 10	57	2.6	95.4	350
	2-4	0.1 + 3.0	10 + 10	42	4.1	90.2	400

conditions are discussed elsewhere (3). The particle removal efficiency and steady state flux were found to increase with larger particle size in the range used in CFMF process (Table 1). Here steady state flux is the stable flux after initial flux decline and when the flux reaches a plateau.

Bimodal

Two different sets of experiments were conducted to study the effect of bimodal particles in suspension. The first set of experiments was carried out with two different size particles in suspension; both sizes are bigger than the pore size. The flux was found to be marginally higher with the increase in the ratio of particle size (Table 1). The permeate quality (turbidity) was also better when larger size of particles were used with 0.6 μm particles in suspension (i.e. Run No. 1-2 and 1-3, Table 2).

The second set of experiments was carried out with two different sizes of particles in the suspension. One particles size is bigger than the size of membrane pore and the other one smaller. Table 1 shows that the steady-state flux of the bimodal particles in suspension is lower than that of single-sized particle in suspension. The Table also shows that the flux decreased with the increase in the ratio of the particle sizes. The wider the ratio, the easier the passage of small particles through the packing formed by the large particles. Thus the fine particles (0.1 μm) could easily flow through the deposit layer on the membrane and finally cause the pore clogging. In the case of runs 2-3 and 2-4 (where the size ratios were 1:11 and 1:30), the particle removal efficiency in terms of turbidity was inferior compared to those of runs 1-2, 1-3, and 2-2 (where the size ratios were 1:2, 1:5, and 1:6). This could again be explained from the fact that in binary mixtures with a size ratio greater than 1:7, the small particles tend to pass through the packing formed by the large particles.

Trimodal

Another set of experiments was carried out with three different sizes of particles in suspension, where all the sizes of particles are bigger than the pore size. The other set of experiments was carried out with the suspension containing three sizes of particles where one size is smaller than the pore size. The results observed in the case of trimodal suspension also confirm the above finding (Table 2). The particle removal efficiency in terms of turbidity was much better in runs 3-1 to 3-3 (maximum size ratio 1:5) with respect to runs 4-3 and 4-4 where the maximum ratio is 1:30. The results obtained in these experiments also revealed the following: as the concentration ratio of coarser particles to finer particles decreases (Table 2) the steady state flux increases.

Table 2. Removal efficiency of combination of three particle sizes (pressure = 70 kPa, cross flow velocity 1 m/s, Millipore membranes of 0.2 μm, latex particles of defined size and concentration in suspension)

Set no.	Run no.	Particle size (μm)	Concentration (mg/l)	Influent turbidity (NTU)	Permeate turbidity (NTU)	Removal efficiency (%)	Steady state flux ($L/m^2 \cdot h$)
3	3-1	0.6 + 1.1 + 3.0	10 + 5 + 5	112	0.05	99.9	500
	3-2	0.6 + 1.1 + 3.0	10 + 10 + 5	156	0.05	99.9	450
	3-3	0.6 + 1.1 + 3.0	10 + 5 + 10	146	0.02	99.9	420
	3-4	0.6	20	124	0.03	99.9	550
4	4-1	0.1 + 1.1 + 3.0	10 + 5 + 5	50	5.4	89.8	470
	4-2	0.1 + 1.1 + 3.0	10 + 10 + 5	70	4.2	94	470
	4-3	0.1 + 1.1 + 3.0	10 + 5 + 10	62	5.5	91.9	200
	4-4	0.1	20	40	5.4	86.5	450

Detailed experiments conducted with particles of one, two, and three different sizes in suspension clearly showed that the particle size and size distribution has significant effect on permeate and effluent quality. The effect was more significant when the particles size (0.1 μm) is smaller than that of membrane pore (0.2 μm). The flux was found to increase with the increase in particle size for mono-dispersed suspensions. For poly-dispersed suspensions the flux decreased with larger particle sizes in the suspension. However, the filtrate quality improved due to the formation of deposit layer, consisting of particles of varying sizes. Therefore, by adding coarser particles in the suspension, (a size ratio less than 7:1) one could use membranes with a larger pore size to filter the fine particle.

FILTER MEDIA PORE SIZE DISTRIBUTION ON GRANULAR BED FILTRATION

Rapid sand filtration is affected by a large number of factors including

1. operating variables such as filtration rate and water temperature,
2. characteristics of the suspended particles such as geometry, size, size distribution, and concentration and
3. the characteristics of the filter media including pore shape, pore size, and pore size distribution (4). This means that it is unlikely that a relatively simple model incorporating the effect of all these variables can be established.

Granular filter beds have a variable pore structure. The manner with which the pore space is formed depends on, among many factors, the arrangement of the granular particles of the filter media. Pore sizes and their distribution in a filter bed are closely related with the size distribution of the granular particles of the filter media. The smaller the grains, the smaller are the pore sizes. Similarly, pore size distribution is also related to grain size distribution. A large distribution of grain sizes often results in a wide distribution of pore sizes, although through mixing could produce a relatively uniform pore size (5).

A granular bed may be considered to be composed of a series of unit bed elements of thickness (l), each of which, in turn, consists of a number of geometrically similar collectors (unit cells). For homogeneous beds, all the unit bed elements are identical. The geometry and size distribution of the unit cells depend on the porous media model used to characterize the bed, and there is a multitude of choice in this respect. Generally speaking, porous media models can be classified into two categories, namely, the external flow models and the internal flow models (6).

The external flow models are based on a geometric description of packing grains. Consideration of the pore space, therefore, can be made only in a secondary manner. The flow field resulting from such a formulation is of the external flow type. Among these models are the isolated sphere model, Brinkman's model, Happel's model, and the modified Happel's model, all of which have been used in the past to study the behavior of deep-bed filtration (1).

Internal flow models are formulated by focusing attention on the pores of the media. These pores, when connected, serve as flow channels, and the surface of the pore acts as the boundary across which transport processes take place. The advantage of the internal flow models is their capability for considering, in an explicit manner, the interaction between neighboring grains (1).

Another important parameter in the granular filter is the pore size distribution of the filter medium which is not often considered in the mathematical model. The basic premise in formulating such a model is to consider a filter bed as a network of randomly connected capillaries of various sizes. The flow behavior throughout the network can be estimated from both the capillary size and size distribution and from the mean flow behavior by using the effective medium approximation (EMA) theory (7).

The results on the flow through porous media obtained by Koplik (6) with EMA can be summarized as follows: A network representing a porous medium is composed of segments of capillaries of various sizes. The pressure drop across the capillaries of different sizes, which corresponds to a given superficial velocity, V, through the medium, constitutes a set of random parameters. For capillaries of a particular type (the jth type), the pressure drop, δP_j, is equal to the sum of a mean pressure drop, δP_m, and a fluctuating pressure drop, δP_{fi}, or

$$\delta P_j = \delta P_m + \delta P_{fj} \tag{1}$$

Where the fluctuating pressure drop, δP_{fj}, is given as

$$\delta P_{fj} = \frac{\delta P_m}{g_j + (Z/2 - 1)g_m} \tag{2}$$

Here, Z is the coordination number, g_j is the conductance of the capillary with radius r_j (the jth type), and g_m is the mean conductance of the network that satisfies the following condition:

$$\left[\frac{(g_m - g_j)}{g_j + (Z/2 - 1)g_m}\right]_{G(g_j)} = 0 \tag{3}$$

where, $G(g_j)$ is the conductance distribution function. Equations (2) and (3) are obtained from the analogy between electrical network and fluid flow, (6). The conductance, applied field (namely pressure drop), and volumetric flow rate are related by the expression

$$\text{Volumetric flow rate} = (\text{applied field})\,(\text{conductance}) \tag{4}$$

If the Poiseuille equation is assumed to apply for the flow through the cylindrical capillaries, then Equation (4) can be written as

$$Q_j = (\delta P_j \pi r_j^4)/8\mu l_j \tag{5}$$

where, μ is the fluid viscosity and l_j is the length of the capillary of type j. If the applied field is taken to be $(\delta P_j/\mu)$, then conductance, g_j is given as

$$g_j = \pi r_j^4/8\mu l_j \tag{6}$$

According to Equation (3), the mean conductance, g_m becomes

$$\sum_j \left[\frac{g_m - \pi r_j^4/8\mu l_j}{\pi r_j^4/8\mu l_j + \alpha' g_m}\right] f_j = 0 \tag{7}$$

where f_j is the number fraction of capillaries with radius r_j in the flow network and $\alpha' = (Z/2) - 1$.

The local flow through each type of capillary can be characterized by the average velocity through the capillary, u as

$$u_j = Q_j/\pi r_j^2 \tag{8}$$

Combining Equations (1), (5), and (8), the following equation for u_j is obtained:

$$u_j = \frac{(\delta P_m) r_j^2}{8\mu l_j}\left[1 + \frac{g_m - g_j}{g_j + \alpha' g_m}\right] = \frac{r_j^2 l_m}{8\mu l_j}\frac{(\delta P_m)}{l_m}\left[1 + \frac{g_m - g_j}{g_j + \alpha' g_m}\right] \tag{9}$$

The total number of open capillaries present in the volume element, N_t, can be calculated from the following:

$$N_t = (N_d \Delta L) = (\Delta L)\,\varepsilon\left[\sum f_j(\pi r_j^2) l_j\right]^{-1} \tag{10}$$

where N_d is the number of open capillaries per unit volume (i.e., capillary density) and ε is the effective porosity of the volume element.

The superficial velocity, V of suspension flow through the medium is then given as

$$\begin{aligned} V &= N_d \sum_j f_j u_j (\pi r_j^2) \\ &= N_d \sum_j f_j \frac{\delta P_m}{8\mu l_j} \pi r_j^4 \left[1 + \frac{g_m - g_j}{g_j + \alpha' g_m} \right] \\ &= N_d \sum_j f_j \frac{\delta P_m}{l_m} \frac{l_m}{8\mu l_j} \pi r_j^4 \left[1 + \frac{g_m - g_j}{g_j + \alpha' g_m} \right] \end{aligned} \tag{11}$$

Throughout a filtration cycle, V is kept constant (i.e., constant filtration rate condition). Furthermore, the mean pressure gradient across a mean capillary, ΔP_m, may be taken as the average pressure gradient over the volume element, ΔL, Thus, Equation (11) can be written as

$$V = N_d \sum_j f_j \frac{(\Delta P_m)}{\Delta L} \frac{l_m}{8\mu l_j} \pi r_j^4 \left[1 + \frac{g_m - g_j}{g_j + \alpha' g_m} \right] \tag{12}$$

The ratio of the pressure drop, ΔP, to its initial value, ΔP_o, at different times can be obtained by the following equation:

$$\frac{\Delta P}{\Delta P_o} = \frac{N_{do}}{N_d} \frac{\sum_j r_{jo}^4 f_{jo} (l_{mo}/l_{jo}) \left[1 + (g_{mo} - g_{jo})/(g_{jo} + \alpha' g_{mo}) \right]}{\sum_j r_j^4 f_j (l_m/l_j) \left[1 + (g_m - g_j)/(g_j + \alpha' g_m) \right]} \tag{13}$$

where the subscript o denotes the start of filter run. Accordingly, if the structure of the network is known (namely, capillary density [N_d], f_j, r_j, I_j, α'), then the flow distribution throughout the network can be determined from Equations (7) and (9). Furthermore, by knowing the change in the media structure, one can use Equation (13) to estimate the corresponding change in ΔP necessary to maintain a fixed flow rate.

Calculation of Pore Radius Reduction

The information necessary to define the structure of a flow network representing a filter medium are N_d and f_j. Assuming that the extent of restriction of capillaries of the same initial size remains the same, one may characterise the jth type capillary as that with an initial radius r_{jo}. According to this method of description the deposition alters the radii corresponding to various types of capillaries over time. The change in r_j can be estimated by the following expression:

$$-(2\pi r_j)l_j \frac{\partial r_j}{\partial t} = Q_j n_v \frac{\pi d_p^3}{6} \frac{\eta_{rcj}}{(1-\varepsilon_d)} \tag{14}$$

where η_{rcj} is the collection efficiency of capillaries of the jth type, ε_d is the porosity of the deposits formed by the collected particles, and n_v is the particle concentration of the suspension entering the capillary. Since the capillaries are situated over an axial distance between z and $z + \Delta L$, n_v is the arithmetic average of the concentration at depth z and $z + \Delta L$. Equation (14) is obtained on the assumption that the deposition on the cylindrical capillary is uniform along the surface of the capi1laries.

Estimation of the Collection Efficiency Including the Effect of Deposition

The rate at which the particles are removed by a single capillary equals the rate at which particles are removed on the wall of the capillary plus the rate at which particles are removed by the retained particles on the wall of the capillary minus the rate at which particles are detached from the capi1lary, (1),

$$\eta_{rcj} = \eta_j \alpha_j + N_j \alpha_p \left(\frac{d_p}{2r_j}\right)^2 - \beta_j \frac{J}{n} \sum_{i_1=1}^{i} \left(\eta_{rcj} n\right)_{i_1} - 1 \tag{15}$$

The change in the number of deposited particles per capillary of type j (N_j), with time, can be written as

$$\frac{\partial N_j}{\partial t} = \beta_j \eta_j \alpha_j n \cdot \pi r_j^2 \tag{16}$$

From the above two equations, one can write the filter efficiency as

$$\eta_{rcj}(i,k) = \eta_j \alpha_j \left[1 + \beta_j \alpha_{pj} \eta_p \sum_{i_1=1}^{i} (i_1 - 1, k) \cdot n(i_1 - 1, k) \cdot \frac{\pi d_p^2}{4} \Delta t\right] - \beta \frac{J(i-1,k)}{n(i,k-1)} \sum_{i_1=1}^{i} n(i_1 - 1, k) \cdot \eta_{rcj}(i_1 - 1, k) \tag{17}$$

Here, $\alpha j \eta_{rcj}$ is the single collector efficiency (through pores of type j) of clean bed filter. In this study an average value of $\alpha j \eta_{rcj}$ (i.e., $\alpha\eta$) is taken. The calculation procedure is explained elsewhere, (1), This average value can be calculated from equations describing clean bed filter efficiency or from experimentally observed removal efficiency of clean bed filter. In this study, typical $\alpha j \eta_{rcj}$ values obtained from experimental results were used in the simulation. ΔP appearing in hydraulic gradient term was calculated from Equation (13), while initial pressure drop ΔP_o, was calculated from Kozeny's equation.

Calculation of Local Concentration

Once $\eta_{rcj}(1, k)$ is calculated, one can estimate the concentration from the material balance equation

$$V[n(i, k) - n(i, k+1)] = \Delta L N_d \sum_j f_j \cdot \pi\left[r_j(i, k)\right]^2 \cdot u_j(i, k) \cdot n_v \eta_{rcj}(i, k) \quad (18)$$

Here, n is the average concentration values at k and k + 1.

The value of n(i, k + 1) can therefore be estimated from the following equation:

$$n(i, k + 1) = \frac{1 - (\pi \Delta L N_d)/(2V) \sum_j f_j\left[r_j(i, k)\right]^2 \cdot u_j(i, k) \cdot \eta_{rcj}(i, k)}{1 + (\pi \Delta L N_d)/(2V) \sum_j f_j\left[r_j(i,k)\right]^2 \cdot u_j(i, k) \cdot \eta_{rcj}(i, k)} \quad (19)$$

The η_{ij} was calculated from the following equation. (The values of α_η, β_2, and $\alpha\beta$ are known, which are taken from typical experimental results).

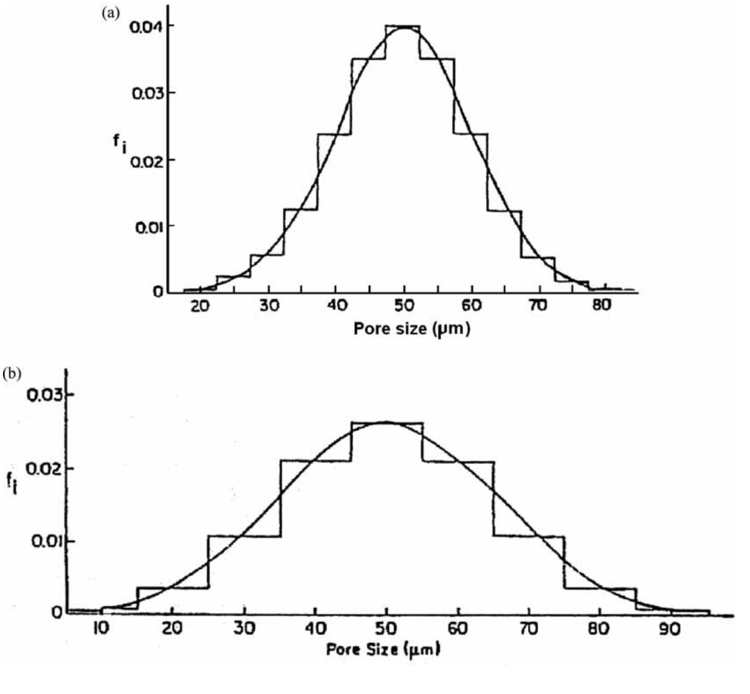

Figure 2. Pore size distribution used in the simulation study. (a) Normal distribution function (median pore size = 50 µm and step size = 10 µm), (b) Normal distribution function (median pore size = 50 µm and step size = 15 µm).

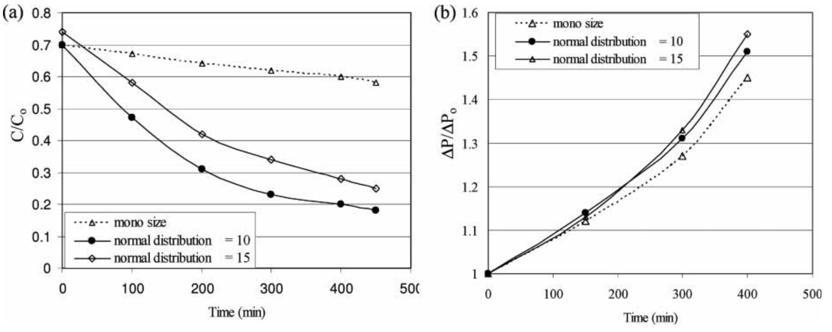

Figure 3. Effect of pore size distribution on filter performance. (a) variation of C/Co with time, (b) variation of $\Delta P/\Delta P_o$ with time $V = 0.15$ cm/s, $L = 2$ cm, $d_c = 0.05$ cm, $\eta\alpha = 0.0007$, $\alpha_p\beta' = 0.008$, $\beta' = 0.01$, $Z = 6$, $\varepsilon_d = 0.08$. The fractions are made 10 μm apart. C and Co are the effluent and influent concentrations.

The influent number concentration was calculated by taking the particle density and the size of 2,280 kg/m^3 and 1 μm, respectively. The influent concentration was taken as 100 mg/L in the simulation.

The pore size distribution used in the simulation is shown in Fig. 2. The results of simulation are shown in Fig. 3. C/Co refers to the concentration of the effluent relative to the concentration of the influent. As the sizes of cylindrical capillaries become distributed, (from monosize to normal distribution $\sigma = 10$) the effluent quality improved at the expense of a higher pressure ratio ($\Delta P/\Delta P_o$). This is attributed to smaller capillaries contributing to the removal of a larger portion of suspended particles. However, the effluent quality was reduced when the distribution of capillaries was broad (from normal distribution $\sigma = 10$ to 15). This implies that while a more distributed size of cylindrical capillaries improves the filter performance, there is appears to be a limit beyond which the performance begins to deteriorate.

CONCLUSION

Detailed granular bed filtration experiments conducted with particles of two and three different sizes in suspension clearly showed that there is an improvement in the fine particle removal efficiency when present with coarse particles. The improvement was more when the ratio of coarse particles to fine particles increased.

Detailed CFMF experimental results with particles of one, two, and three different sizes in suspension clearly showed that the particle size and size distribution has significant effect on permeate and effluent quality. The flux was found to increase with the increase in particle size for monodispersed suspensions. For polydispersed suspensions the flux decreased with the increase in particle sizes in the suspension. However, the filtrate quality improved due to the formation of deposit layer, consisting of particles of varying sizes. Therefore by adding coarser particles in the suspension, (a size ratio less than 7:1) one could go for membranes with larger pore size to filter the fine particle.

The model by Vigneswaran and Chang (1) which is an extension of O'Melia-Ali model (8) was modified to the internal flow model and was then incorporated with the model developed using effective medium approximation by Vigneswaran and Tien (9) to investigate the effect of pore size distribution in deep-bed filtration. Two pore size distributions were assumed and model simulations were performed. Significant variations of headloss as well as concentration were observed. This simulation indicated the clear effect of structure of porous media (i.e., pore size distribution) on the filter performance.

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