A Filtration Model of Microporous Membrane Filters in Liquids

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A theoretical study on particulate retention by microporous membrane filters during liquid filtration has been investigated. A model to predict particle retention using a multi-layer concept for filter retention by sieving and using flow resistance for cake filtration was developed to predict particle retention as a function of particle diameter and particle loading. Filter efficiency for the 0.45 μ m rated membranes predicted 99.95% for the particle diameter of 0.453 μ m. Filter efficiency by sieving model decreased with increased particle loading on the filter. After substantial particle loading, filter efficiency was found to increase with increased particle loading due to the combined effect of sieving and cake filtration. Theoretical particle retention modeling showed good agreement with experimental results.

Key Words: Liquid Filtration, Sieving Model, Cake Filtration Model, Membrane Filters, Pore Size, Monodisperse and Polydisperse Spheres

Nomenclature		n	: Constant
		$n(t,D_f)$: Pore number in pore size from D_f to D_f	
Α	: Filter face area		$+ dD_f$
a, b, c	: Constants	Q	: Flow rate through a capillary
C_{eff}	: Effluent particle concentration	Q_p	: Flow rate through pores of size larger
C_{in}	: Influent particle concentration		than particle size
C_i	: Particle concentration of the i th filter	Q_t	: Total flow rate through a filter layer
	layer	P	: Overall penetration
CMD	: Count median diameter	P_i	: Particle penetration of the i th filter
D_f	: Pore size		layer
D_P	: Particle size in diameter	R	: Filter medium resistance
E_c	: Filter efficiency by cake filtration	R_c	: Cake resistance
$f(D_f)$: Lognormal frequency function	Т	: Thickness of a single filter layer
K, k	: Constants	t	: Time
L	: Filter thickness	V_f	: Void volume of the filter
LRV	: Log reduction value	ω	: Mass of cake deposited per unit area
N	: Total number of filter layers	ε	: Media's porosity
N_{ci}	: Number of pores clogged at time t in	ρ_p	: Particle density
	the i th filter layer	μ	: Fluid viscosity
N_o	: Initial total pore number of the filter	0g	: Geometric standard deviation
		$\varDelta P$: Pressure drop across the filter
* Department of Mechanical and Precision Engi-		Λt	: Time interval

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1. Introduction

Filtration, particularly membrane filtration,

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plays an important role for removal of any unwanted particulate matter suspended in gases and liquids. Liquid filtration can be described as a particle/fluid separation process in which the liquid to be filtered is passed through the porous media that retains the particles as they come in contact with the structural elements of the media. Liquid filtration is less efficient than gas filtration because particles of a given size diffuses more slowly in a liquid and generally does not undergo capture by inertial impaction. The only effective mechanism that can be relied upon to remove suspended particles in a liquid is sieving, although the actual efficiency of filtration will be modified by charge effects due to electrical double-layer, adhesion, adsorption, and hydrophobic or hydrophilic properties of the filter. Membrane filters are widely used to remove particles from liquids and gases and have been shown to be highly effective in reducing particle levels(Brock, 1983; Rubow and Liu, 1986).

The purpose of this study is to investigate theoretically the particle removal efficiency of the microporous membrane filters using monodisperse and polydisperse spheres. A predictive model is developed using a multi-layer concept for filter retention by sieving and using flow resistance for cake filtration during monodisperse and polydisperse particle challenge. Also the theoretical results will be compared with the experimental results.

2. Previous Works

Many investigators have developed several models to predict filter efficiency for removing liquid-borne particles. Grant and Liu(1991) modified the model developed by Soo and Radke(1986) to predict the retention of small monodisperse latex particles by 0.45 μ m rated membrane filters as a function of particle loading. They showed that as particle loading increased, the retention decreased. The rate of decrease was shown to depend on a number of factors including particle diameter, filter thickness, and particle loading.

Soo and Radke(1986) have developed a sim-

plified filtration model describing the flow of stable, dilute emulsions in porous media. They described that the initial reduced dimensionless filter coefficient and the flow redistribution parameter were functions of the incoming particle size, the filter porosity, and pore size distribution.

Rajagopalan and Tien(1976) developed a model for the initial particle deposition in a deep bed filter with a sphere-in-cell porous media model. They found that the collection rate depended on essentially four process parameters which characterized the ratio of the particle size to the grain size, the magnitude of the gravitational collection, the extent of the attractive surface force, and the magnitude of Brownian diffusion. Payatakes et al.(1973) developed a model for granular porous media. The media was modeled as consisting of a series of statistically identical unit elements each of which in turn consists of a number of unit cells connected in parallel.

3. Particle Capture Mechanisms in Liquids

Particle capture in liquid filtration is usually described as sieving, depth, and cake filtration. In sieving, if the size of the particles present is larger than the pore opening in the filter, the particles will be captured and retained. Although sieving is commonly pictured as occurring on the surface of a membrane, sieving can also occur in the interior of the membrane when small interior pores are present, as depicted in Fig. 1(a). The mechanism is also known as mechanical sieving or straining. Retention by sieving will theoretically be unaffected by the test or process conditions. The particle removal efficiency is expected to increase with increasing membrane thickness and to be independent of face velocity(Lee, 1992; Grant and Liu, 1991). Sieving is considered to be the most important particle capture mechanism because it can be relied upon to positively remove suspended particles from a liquid. Sieving, therefore, can be considered as absolute. On the other hand, mechanisms that rely on electrostatic attraction and charge effects are influenced by the charge characteristics of the membrane surface, as well as



Fig. 1 Particle capture mechanisms in liquid filtration

the fluid, and particle properties. Since the latter properties may vary from one application to another, these mechanisms cannot always be relied upon to remove suspended particles. Their effectiveness is, therefore, not absolute or guaranteed.

Because the absolute particle retention characteristics of filtration by sieving, sterilizing grade membrane filters have been developed. For example, the 0.22 μ m rating of sterilizing grade membrane filters is based upon retention(>99. 9999999%) of the bacteria Pseudomonas diminuta, an organism which is thought to be 0. 22 μ m in diameter(Leahy and Sullivan, 1978). Removal of particles >0.22 μ m is considered to be absolute by such filters.

In contrast to sieving, particle capture in depth filtration is by particle/membrane contact throughout the depth of the filter, as depicted in Fig. 1(b). Particles smaller than the pores are retained as long as they are brought to physical contact with the membrane surface by such processes as diffusion, interception, impaction, and gravitational settling, and are held onto the membrane surface by the van der Waals and electrostatic attractive forces. Depth filtration by diffusion, interception, and impaction is very important in gas filtration, because of the effectiveness of the van der Waals attractive forces between particles and the filter surface. However, depth filtration is less important in liquid filtration because particle adhesion to the membrane surface are influenced strongly by electrostatic repulsion and hydrodynamic interactions between the particles and the membrane surface. Depth filtration can become important in liquid filtration if the particles can be effectively retained by the membrane through hydrophobic and electrostatic adsorption, but the effectiveness of the mechanism is likely to be affected by the electrical properties of the liquid as well as those of the particulate contaminants in the liquid.

In cake filtration, as depicted in Fig. 1(c), the accumulation of a thick layer of collected particulate material on the upstream face of the filter provides a porous medium through which the liquid must pass. This porous medium can cause additional particle capture by the conventional mechanisms of sieving and depth filtration. If particles are larger than the filter pores, the cake can form readily on the filter surface. When the particles are substantially smaller than the pores, cake formation must be preceded by a bridging process. Cake filtration will continue until the pressure drop across the cake exceeds the maximum permitted by the specific system. A very important factor in cake filtration, therefore, is the permeability or the resistance of the cake. which is determined by the properties of the filtered particles and the structure of the cake formed.

4. Theoretical Modeling

The most important mechanisms of particle capture in filtration of liquids through microporous membrane filters are sieving, electrostatic adsorption, and cake filtration. Double layer repulsion can prevent particle and filter surface contact, so that the only operating mechanism of liquid filtration is sieving. This mode of operation significantly reduces the filter collection efficiency. Hence, capture by sieving under the condition of double layer repulsion represents "worst case" filter performance in liquids. The proposed model is based on the particle capture by sieving only, followed by the cake filtration.

4.1 Sieving Model

A predictive sieving model has been developed using a multi-layer concept for filter retention as a function of particle diameter and filter pore size. Fig. 2 shows the schematic representation of the proposed multilayer model to predict the filter efficiency. A homogeneous, randomly packed medium can be considered as a number of unit filter layers connected in series. Each filter layer of thickness, T, contains an assembly of capillaric collectors or pores of specified geometry and size distribution. Thickness of the unit filter layer depends on the porosity value and the pore count median diameter (CMD). It can be described as T $=a \ CMD/(1-\varepsilon)^b$, where a and b are constants, ε is the media's porosity. The efficiency of the filter layer is determined by the efficiencies of each collector and depends on the flow field through the collectors and the mechanism of the transport of particles from the suspension to the collector surface.

The proposed sieving model is based on the following assumptions :

1. The initial pore size distribution of the filter media is lognormally distributed and the pores have circular openings.

2. Each filter layer has the same pore size

distribution and filter properties.

3. The thickness of a filter layer is twice the CMD.

4. The flow rate through each capillary can be estimated using the well established Poiseuille equation for laminar flow, $Q = K D_f^4$ where K is a constant and D_f is the pore size.

5. There is no flow through pores clogged by particles.

 Porous media and particles are rigid(no shape change is allowed).

7. Particle suspensions are dilute and stable.

8. There is no particle entrainment.

9. Particle clogging pores do not interfere with adjacent pores.

The overall penetration, P, describing the macroscopic description of filtration is defined as

$$P = \frac{C_{eff}}{C_{in}} \tag{1}$$

where C_{in} and C_{eff} denote the influent and effluent particle concentrations, respectively. The penetration of the *i* th filter layer, P_i , is C_i/C_{i-1} , where C_i and C_{i-1} are the particle concentration of the suspension exiting the *i* th and i-1 th filter layer. Therefore, overall penetration can easily be expressed in terms of the unit filter layer penetration as follows;

$$P = (P_1) \cdot (P_2) \cdot (P_3) \cdots (P_i) \cdots (P_N) \quad (2)$$

where N is the total number of filter layers and



Fig. 2 Schematic representation of a microporous membrane for proposed multilayer model

should be taken as the integer closest to the value of L/T, where L is the filter thickness.

In sieving, particles larger than the pore size are retained in the media. Particle penetration depends on the flow through the open pores as no flow is assumed through the pores clogged by particles. The penetration of the i th single filter layer, P_i , due to sieving in a filter layer may also be expressed as

$$P_i = \frac{Q_P}{Q_t} \tag{3}$$

where Q_p is the flow through pores of size larger than the challenge particle size and Q_t is the total flow through a filter layer. Q_p strongly depends on the pore size distribution and challenge particle size. The liquid flow rate through a filter layer, assumed as consisting of a lognormal pore size distribution, can be obtained with the assumption of the established Poiseuille equation regarding flow rate through a capillary tube. The flow rate through a pore varies as the fourth power of its radius. Liquid flow through larger pores is much greater than through pores of a smaller diameter. Therefore, initial penetration of the *i* th single filter layer, P_i , can be expressed as

$$P_i = \frac{\int_{D_f}^{\infty} D_f^4 f(D_f) dD_f}{\int_0^{\infty} D_f^4 f(D_f) dD_f}$$
(4)

where D_p and D_f are the monodisperse challenge particle diameter and the pore diameter, respectively, and $f(D_f)$ is the lognomal frequency function as described in Eq. (9). With the above expression, the overall initial particle penetration of the filter can be obtained by the Eq. (2).

The particle penetration of the i th filter layer after filter loading at time t, can be calculated using the pore size distribution taking into account the pores clogged by particles. As particles larger than the pore size are retained in the media in sieving, the pores smaller than test particle size are clogged by particles. The number of open pores within each filter layer changes as a function of time. The details of pore clogging will be described later in the section of pore size determination. The particle penetration of the i th filter layer after filter loading at time t, described in Eq. (4), can be calculated from the determination of pore size distribution at time t.

This sieving model is also applicable to the filtration of polydisperse spheres. The penetration through each filter layer takes into account the polydisperse particle sizes. The amount of flow through pores of size larger than particle size described in Eq. (3) is the summation of overall particle sizes.

4.2 Cake Filtration

A predictive model for cake filtration is developed using flow resistance as a function of particle loading. In cake filtration, particles are deposited in the form of a cake on the upstream filter surface, and filtration results from the bed of deposited particles. A cake starts to build on the surface of the filter so that, gradually, a greater proportion of the available pressure drop is due to the cake. This results in an effective increase in the resistance leading to a gradual drop in permeability. The filtrate flow rate, Q, and pressure drop across the filter, P, are related as :

$$Q = \frac{A \Delta P}{\mu (R + R_c)} \tag{5}$$

where A, μ , R, and R_c are the face area, fluid viscosity, medium resistance, and cake resistance, respectively. Eq. (5) is called the modified Darcy' s equation describing the relationship of the pressure drop and flow rate(Brock, 1983). The medium resistance should be constant in a given filter and depends on the membrane thickness and various morphological features such as the tortuosity, porosity, and pore size distribution. The cake resistance increases with time and can be assumed to be dependent on the amount of cake deposited and the properties of the cake. It can be described as follows :

$$R_c = k(\Delta P)^n \omega \tag{6}$$

where k is the empirical constant, n is dependent upon cake properties, and ω is the mass of cake deposited per unit area. From Eqs. (5) and (6), the mass of cake deposited per unit area can be obtained as follows:

$$\omega = \frac{1}{k(\Delta P)^n} (\frac{\mu Q}{A\Delta P} - R) \tag{7}$$

From the above equation, k and n were deter-

mined from the measurement of the pressure drop across the filter as 9.4 and 14.1, respectively. The filter efficiency by cake filtration, E_c , can be obtained from the mass of cake deposited per unit area, ω , as follows:

$$E_c = \left(\frac{\omega A}{\rho_p D_p^3/6}\right) \frac{1}{C_{in}} \tag{8}$$

where ρ_p is the particle density. Therefore, filter efficiency by cake filtration can be obtained by Eqs. (7) and (8) through the measurement of pressure drop across the filter(Lee, 1992).

4.3 Pore Size Determination

The distribution of pore size in a filter provides information on a filter's ability to remove particles of known sizes from liquid streams. There is no reliable way of directly measuring the pore sizes of membrane filters because pore size distribution in a filter medium is composed of a polydisperse and randomly connected array of pores. Several test methods have been developed for measuring pore size distributions, for example, direct microscopic examination, bubble point test, mercury intrusion method, and particle challenge test. The pore size ratings of most commercially available liquid filters are determined by bubble-point extrapolation techniques and reflect the membrane's retention efficiency. Bubble point integrity testing is not only a method to determine filter integrity, but also serves as a nominal quantitative measure of pore size. However, bubble point values do not directly yield pore sizes. Mercury intrusion method is a more accurate method for characterizing filters. However, some deformation of the membrane filter matrix may occur, causing changes in pore size or structure at the high pressures that must be used(Brock, 1983).

In the proposed sieving model, the pore size distribution is required to predict the filter efficiency. This model was developed using a multilayer concept of filter retention as described above. A lognormal pore size distribution is used to describe a model for presenting a reasonable physical description of pore sizes. The lognormal frequency function(Hinds, 1982), $f(D_f)$, is

$$f(D_f) = \frac{1}{\sqrt{2\pi} D_f l n \sigma_g} \exp(-\frac{(l n D_f - l n C M D)^2}{2(l n \sigma_g)^2})$$
(9)

where σ_g is the geometric standard deviation. The two parameters describing a lognormal pore size distribution are *CMD* and σ_g . These can be determined through the best fit of experimental and calculated initial penetration results as follows. Penetration based on particle removal via sieving in a filter layer can be described by Eq. (4). This equation has three unknown parameters; initial penetration(P_i), *CMD* and σ_g . Through the measurements of initial filter penetration for monodisperse particles ranging from 0.368 to 0. 741 μ m in diameter (Lee, 1992), *CMD* and σ_g of 0.45 μ m rated filter can be calculated by the numerical method of best fit.

The initial total pore number, N_o , in a filter layer can be obtained using lognormal frequency function and filter porosity(75%), ε , known as the ratio of void volume divided by the volumes of the filter. The void volume, V_f , can be described as

$$V_f = \varepsilon A L = N_o L \int_0^\infty \frac{\pi}{4} D_f^2 f(D_f) dD_f \quad (10)$$

Therefore, the initial total pore number from the above equation can be described as

$$N_o = \frac{\varepsilon A}{\int_0^\infty \frac{\pi}{4} D_f^2 f(D_f) dD_f}$$
(11)

Since ε , A, and $f(D_f)$ are known, N_o can be obtained by the numerical method. The predicted total pore numbers was 1.86×10^9 for the 0.45 μ m rated clean PVDF filter.

The initial pore number, $dn(0, D_f)$, at the range of pore size from D_f to $D_f + dD_f$ should be first calculated using the following equation :

$$dn(0, D_f) = N_o f(D_f) dD_f$$
(12)

Since initial total pore number, N_o , and lognomal frequency function, $f(D_f)$, are known, the number of pores as a function of pore size can be obtained by Eq. (12).

The number of open pores in each filter layer changes with increased particle loading. The pores smaller than test particle size are clogged by particles. The number of pores clogged at time t, $N_{ci}(t)$, in the *i* th filter layer can be described as where P_i is the particle penetration of the *i* th filter layer, C_{i-1} is the particle concentration upstream of the *i* th filter layer and Δt is the time interval. The pore number for pore size larger than test particle size does not change, but changes for pore size smaller than test particle size. Therefore, the pore number in pore size ranging from D_f to $D_f + dD_f$ after particle loading at time *t* and in the *i* th filter layer, $dn(t, D_f)$, can be obtained as

$$dn(t, D_f) = dn(t-1, D_f) - N_{ci}(t) \frac{n(t-1, D_f)D_f^4 dD_f}{\int_0^{D_f} D_f^4 n(t-1, D_f) dD_f}$$
(14)

Therefore, the particle penetration of the filter after particle loading at time t may be obtained using Eq. (4) and the pore size distribution taking into account the pores clogged by particles.

5. Results and Discussion

5.1 Predicted Filtration Efficiency

The filter efficiency using the log reduction value(LRV) can be described as a function of pore volumes of particles removed which is equivalent to the fraction of the volume of the particles retained in the filter pore volume. The LRV is defined as the logarithm to the base 10 of the number of particles in the challenge to the number of particles in the filtrate : $LRV = \log_{10}(C_{in}/C_{eff})$, where C_{in} and C_{eff} are the particle concentration at the filter inlet and outlet, respectively. Retention efficiencies of 90, 99, 99.9 and 99.99% are equivalent to LRV of 1, 2, 3, and 4, respectively.

Figure 3 shows the predicted filter performance based on the combined effect of sieving and cake filtration using the multi-layer concept of the proposed model. Filter efficiency based on the sieving model decreases exponentially with increased particle loading while filter efficiency due to cake filtration increases. Therefore, the filtration sequence is a sieving dominant stage in the membrane, followed by transition to a cake filtration dominant stage.

Predicted filter efficiency by sieving alone for the 0.45 μ m rated filter with the monodisperse spheres is shown in Fig. 4. Values of parameters used in this study are the porosity of 75%, the thickness of 110 μ m, the filter layers of 122, and the face area of 13.38 cm². The model fit yielded a pore size distribution with a CMD of 0.45 μ m and σ_g of 2.2 based on the proposed multi-layer sieving model described before. Filter efficiency was strongly dependent upon test particle size and the LRV increased with increasing particle size. The predicted initial LRVs for 0.368, 0.453, 0. 741, and 0.913 μ m particle diameter were 1.80, 3. 29, 10.36, and 15.53, respectively. The LRVcurves for all particle sizes decrease exponentially with increased particle loading. Similar results were obtained for the 0.22 μ m rated filters(Lee, 1992).

Figure 5 shows the predicted filter efficiency of the 0.45 μ m rated filter by sieving and cake filtra-



Fig. 3 Predicted filter performance by sieving and cake filtration

Fig. 4 Predicted filter efficiency for 0.45 μ m rated filters by sieving alone

tion as a function of particle sizes. Filter efficiency initially decreases exponentially with in-

Fig. 5 Predicted filter efficiency for 0.45 μm rated filters by sieving and cake filtration

Fig. 6 Predicted filter efficiency during polydisperse sphere challenge by sieving alone

Fig. 7 Predicted filter efficiency during polydisperse sphere challenge by sieving and cake filtration

creased particle loading, then gradually decrease, and the LRV increases with increasing particle size due to cake filtration.

Figure 6 shows the predicted filtration efficiency of sieving alone with the polydisperse sphere suspensions for discrete particle size intervals. The trend of filter efficiency was similar to monodisperse sphere suspensions as shown in Fig. 4. Retention of polydisperse particles was higher than that of monodisperse. For example, the LRV for particle size of 0.913 μ m in diameter at the pore volumes of particles removed of 0.01 was 2.5, while for $0.6 \sim 0.8 \,\mu m$ particle size channel of the polydisperse suspension the LRV was 4. Therefore, the breakthrough was found to occur more slowly when the challenge suspension was polydisperse. The predicted initial LRVs for $0.3 - 0.4, 0.4 - 0.5, 0.5 - 0.6, and 0.6 - 0.8 \ \mu m$ particle size channel were 1.69, 3.45, 5.76, and 10.1, respectively. Figure 7 shows the predicted filter efficiency during polydisperse sphere challenge by sieving and cake filtration. The trend of filter efficiency was similar to monodisperse sphere suspensions as shown in Fig. 5.

5.2 Comparison of the Theoretical and Experimental Results

Figure 8 presents the comparison of the predicted and measured initial filter efficiency as a function of time with the 0.45 μ m rated filter. The initial predicted *LRVs* were based on sieving model only, and the measured initial *LRVs* were obtained from results described by Lee (1992). An

Fig. 8 Comparison of predicted and measured initial filter efficiency with the 0.45 μ m rated filter

experimental study of particulate matter retention by 0.45 µm rated membranes during liquid filtration was made with polystyrene latex spheres using an automated filter test system and a laser particle counter. The system fluid was deionized water containing 0.1% non-ionic surfactant(Triton X-100). Experimental results showed good agreement with theory, but discrepancy between theory and experiment increased as test particle size decreased. For the particle size below 0.2 μ m in diameter, additional particle capture by diffusion might be expected in the experimental results. Particle capture by diffusion under the unfavorable deposition condition was dominant for the particle size below 0.2 μ m in diameter. For the particle size above 0.7 μ m in diameter, an experimental difficulty might be expected due to

Fig. 9 Comparison of predicted and measured filter performance during monodisperse sphere challenge

Fig. 10 Comparison of predicted and measured filter performance during polydisperse sphere challenge

high filter efficiency. Typical particle concentration upstream of the filter was 1.4E7 #/cc(Lee, 1992). A few particles downstream of the filter were measured by the optical particle counter because the filter efficiency was >99.9999%.

Figure 9 shows the comparison of predicted and measured filter performance during the monodisperse particle challenge for 0.45 μ m rated filter as a function of particle loading. Experiment results for particle sizes similar to the rated pore size of the filter were in good agreement with theory, but the discrepancy between theory and experiment increased as the test particle size increased. The model fit yielded a pore size distribution with a *CMD* of 0.45 μ m and σ_{g} of 2.2 based on the proposed multi-layer sieving model.

Figure 10 shows the comparison of predicted and measured performance of 0.45 μ m rated filter during the polydisperse particle challenge. Polydisperse suspension was a mixture of 0.741, 0. 576, 0.453, and 0.305 μ m sphere particles(Lee, 1992). The model for predicting the filter performance was less accurate than for the case of monodisperse particles. Also, the model predicted good agreement with experiment for smaller particles and a less rapid breakthrough of larger particles than was experimentally observed. Similar results were obtained for the 0.22 μ m rating filter. The accuracy of the model prediction was strongly a function of the particle size distribution. For particle sizes similar to the given pore size rating, the model accurately predicted filter performance for both monodisperse and polydisperse particles.

6. Summary and Conclusions

Particle removal by microporous membrane filters has been investigated theoretically. These studies are based on particle capture by sieving alone, followed by cake filtration on the upstream side of the filter.

A model to predict particle retention using a multi-layer concept for filter retention by sieving and using flow resistance for cake filtration was developed to predict particle retention as a function of particle diameter and loading. Filter efficiency based on the sieving model decreased exponentially with increased particle loading while filter efficiency due to cake filtration increased. Therefore, the filtration sequence predicted a sieving dominant stage in the membrane, followed by transition to a cake filtration dominant stage. Retention of polydisperse particles was higher than that of monodisperse.

Theoretical modeling results were in good agreement with experiment, but the discrepancy between theory and experiment increased as the test particle size increased. For particle sizes similar to the given pore size rating, the model accurately predicted filter performance for both monodisperse and polydisperse particles.

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