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A Comparison Between Theory and Experiment

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Particle Filtration: A Comparison Between Theory and Experiment

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Abstract

The process of filtration of non-charged, submicron particles represents an example of transport in homogeneous and heterogeneous porous media that can be analyzed using the method of volume averaging. In this article we develop the local volume averaged particle transport equation for a homogeneous filter and compare the results with experimental data. The particle continuity equation is represented in terms of the *first correction* to the Smoluchowski equation that takes into account particle inertia effects for small Stokes numbers. This leads to a cellular efficiency that contains a minimum in the efficiency as a function of the particle size, and this allows us to identify the *most penetrating particle size*. Comparison of the theory with experimental results indicates that the first correction to the Smoluchowski equation gives reasonable results for the most penetrating particle size and for smaller particles; however, results for larger particles clearly indicate the need to extend the Smoluchowski equation to include higher order corrections. The influence of local heterogeneities on the measured filter efficiency may account for some of the observed differences between theory and experiment.

1. Introduction

The process of filtration takes place in an hierarchical porous media (Cushman, 1990) and we have illustrated this in Figure 1. In order to design a filter, one needs a particle transport equation in which the porosity heterogeneities have been *spatially smoothed*. This suggests the use of the first averaging volume shown in Figure 1 along with the method of large-scale averaging (Quintard and Whitaker, 1987, 1988, 1990; Plumb and Whitaker, 1988, 1990). Large-scale averaging requires the use of local volume averaged equations that are associated with the second averaging volume shown in Figure 1. These equations are sometimes referred to as the *Darcy-scale* transport equations and they represent the point in the hierarchical process at which the governing differential

Figure 1
Hierarchical View of the Filtration Process

γ-phase

σ-phase

γ-phase

κ-phase

β-phase

BOUNDARY CONDITION
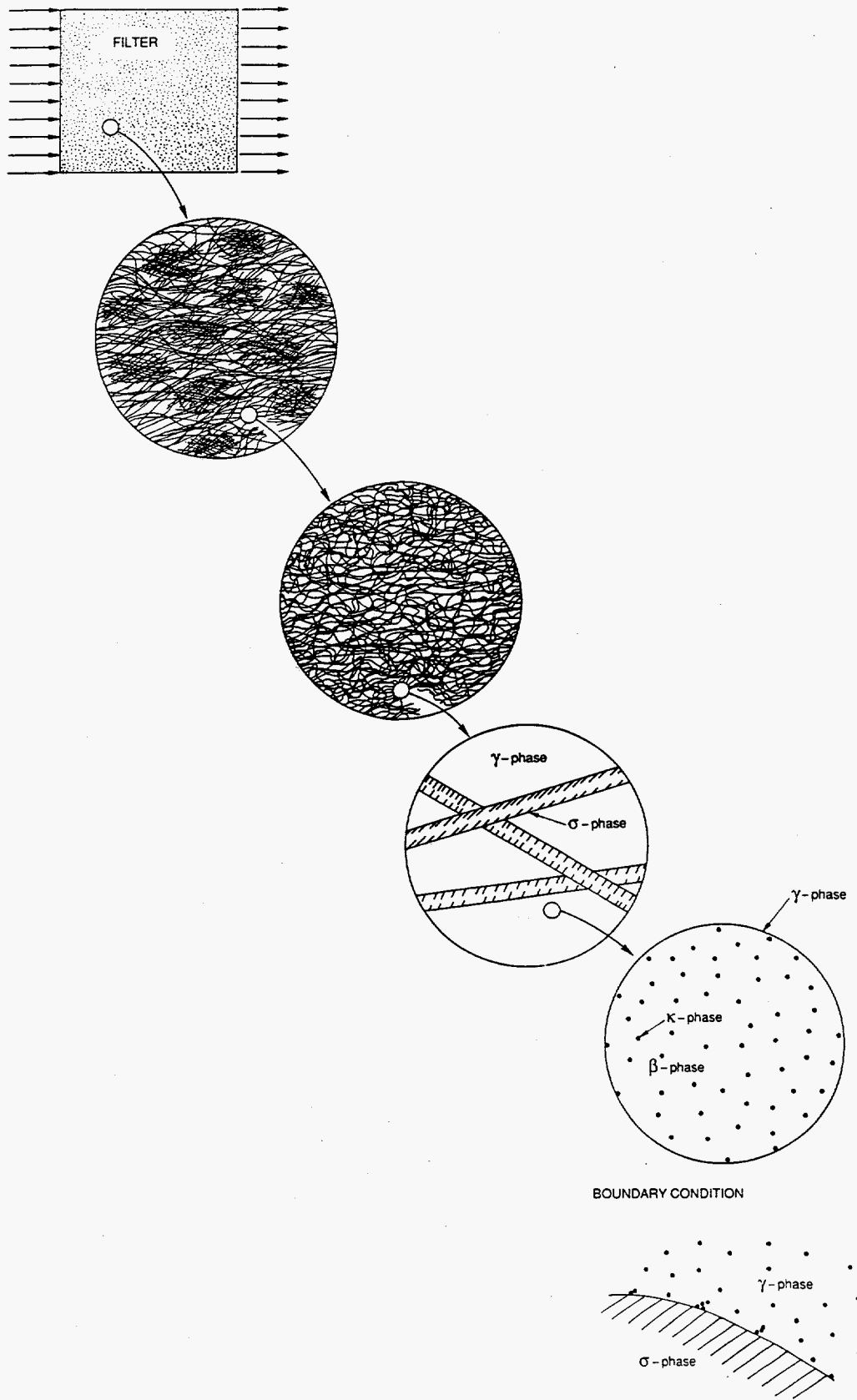
γ-phase

σ-phase

equations and boundary conditions are *joined*. These boundary conditions are imposed at the γ-σ interface illustrated in the third volume contained in Figure 1 where we have identified the fibers as the σ-phase and the fluid as the γ-phase. The governing equation for the fluid velocity in the γ-phase will be taken to be Stokes' equations, while the governing equation for the particle concentration is represented by a Fokker-Planck equation for the probability density function. This idea is suggested in the last volume illustrated in Figure 1 where we have identified the particles as the κ-phase and the pure fluid as the β-phase. The fact that we are going to use Stokes' equations to described the velocity of the γ-phase indicates that the volume fraction of the particles is much, much less than one (Russell, 1981).

Macroscopic transport equations for filtration are often introduced heuristically in the form of a convective-dispersion equation with a source term accounting for the particle deposition. This source term requires knowledge of the *filter collection efficiency* that can be determined by experiments. The starting point for a theoretical derivation of the filter collection efficiency is a pore-scale description of the particle transport which must be subjected to both local volume averaging and large-scale averaging in order to obtain a filter transport equation. The particle transport equation must account for the various mechanisms that affect the particle deposition process such as Brownian diffusion, inertial deposition, electrostatic effects, etc. Results published in the literature [see Ramarao and Tien (1991) for an extensive review] can be classified according to the various assumptions made in describing the particle transport, as well as the methodology used in proceeding from the pore-scale equations to the macroscopic description.

If one assumes pure Brownian diffusion, particle transport can be viewed as equivalent to the process of convective-diffusion in a porous medium with heterogeneous chemical reaction (Friedlander, 1977; Shapiro and Brenner, 1990). However, it has been recognized for some time that the existence of a *most penetrating particle size* is the result of a complex interaction between Brownian diffusion and inertial effects. Because of this, it is important to make use of a description of the particle transport process that accounts for particle inertia effects.

Several approximate methods have been proposed to determine the particle velocity field. Deterministic particle trajectory calculations can be used in the convective-diffusion equation (Fuchs, 1964; Yuu and Jotaki, 1978; de la Mora and Rosner, 1981). More precise solutions of this stochastic process can be obtained by a continuous approach (de la Mora and Rosner, 1982; Yeh and Liu 1974; Banks and Kurowski 1983) which will be described later in this paper. More recently, direct solutions of the Langevin equations (the so-called Brownian dynamics calculations) have been used to simulate particle motions around single fibers (Kanaoka et al., 1983; Gupta and Peters. 1985, 1986; Ramarao, Tien, and Mohan 1994).

Brownian dynamics represents the most reliable method of analyzing the particle transport process since the physics can be incorporated directly into the calculations for a

single fiber (or multi-fiber) efficiency. However, the design of a filter requires an analysis that faithfully transmits the physics from the particle scale illustrated in Figure 1 to the filter scale, and this is difficult to accomplish via Brownian dynamics. This suggests the development of continuum equations that can accurately simulate the phenomena described by Brownian dynamics. Our approach will be to derive both the macroscopic equation and the macroscopic coefficients from the pore-scale description. This approach has been used successfully in dealing with several problems of transport phenomena in porous media. In particular, valuable results have been obtained for the process of passive dispersion (Lee, 1979; Brenner. 1980; Carbonell and Whitaker, 1983; Eidsath et al., 1983; Rubinstein and Mauri 1986), active dispersion (Zanotti and Carbonell, 1994; Quintard and Whitaker. 1994a), and dispersion with chemical reaction (Mauri, 1989; Edwards et al., 1993). In this paper, the macroscopic forms of the pore-scale equations are obtained by the method of volume averaging and effective transport properties are determined by two independent closure problems that are solved for periodic arrays of cylinders.

PARTICLE MOTION

Our description of the motion of the particles begins with the single particle Langevin equation

$$m_p \frac{d\mathbf{v}_p}{dt} = -\frac{3\pi\mu d_p}{c_s}(\mathbf{v}_p - \mathbf{v}) + \mathbf{F}_r(t) \qquad (1.1)$$

in which m_p is the mass of the particle and $\mathbf{v}_p$ is the velocity of the particle. The first term on the right hand side of Eq. 1.1 describes the Stokes' drag on a single isolated particle with c_s representing the Cunningham correction factor (Tien, 1989). The use of this form for the force indicates that we are ignoring particle-particle interactions and the fluid mechanical complications that arise when a particle approaches a solid surface (Peters and Ying, 1991). These effects should certainly be taken into account; however, in this initial study it is our attention to keep the analysis as simple as possible.

It is convenient to express Eq. 1.1 as

$$\frac{d\mathbf{v}_p}{dt} = -\gamma(\mathbf{v}_p - \mathbf{v}) + \Gamma(t) \qquad (1.2)$$

in which γ is inversely proportional to the Stokes' number to be defined later.

$$\gamma = \frac{3\pi\mu d_p}{m_p c_s} \sim St^{-1} \qquad (1.3)$$

There are many particle transport processes for which the Stokes' number is small compared to one and this so-called *high friction limit* represents an important case for

filtration. The Fokker-Planck equation associated with the stochastic process described by Eq. 1.2 takes the form (Risken, Sec. 10.1, 1989)

$$\frac{\partial W_p}{\partial t} + \frac{\partial}{\partial \mathbf{r}}\left(\mathbf{v}_p W_p\right) - \frac{\partial}{\partial \mathbf{v}_p}\left[\gamma(\mathbf{v}_p - \mathbf{v})\right] = \left(\frac{\gamma k T}{m_p}\right)\frac{\partial}{\partial \mathbf{v}_p}\cdot\frac{\partial}{\partial \mathbf{v}_p}\left(W_p\right) \qquad (1.4)$$

and this result is often known as Kramer's (1940) equation. The dependent variable represents a probability density function

$$W_p = W_p(t, \mathbf{r}, \mathbf{v}_p) \qquad (1.5)$$

and the particle number density is given by

$$n_p = \int\limits_{-\infty}^{+\infty} W_p(t, \mathbf{r}, \mathbf{v}_p)\, d\mathbf{v}_p \qquad (1.6)$$

The solution of Eq. 1.4 for the high friction limit is based on matrix continued fraction methods (Risken, Sec 10.4, 1989) which yield

$$\frac{\partial n_p}{\partial t} = \nabla\cdot\left\{\left[-\mathbf{v} + D_p\nabla\right]n_p\right\} + \gamma^{-1}\nabla\cdot\left\{\left[\mathbf{v}\cdot\nabla\mathbf{v} - (\nabla\cdot\mathbf{v})D_p\nabla\right]n_p\right\} + \qquad (1.7)$$

$$+ \gamma^{-3}\nabla\cdot\left\{[\dots\dots]n_p\right\} + \mathbf{O}(\gamma^{-5}) + \dots\dots$$

in which D_p is the Brownian diffusivity. It is important to keep in mind that this result *does not take into account* the complex fluid mechanics that occur when a particle approaches a solid surface. The first two terms on the right hand side of Eq. 1.7 were obtained by Titulaer (1978) and by Skinner and Wolynes (1979) using different methods. If only the first term on the right hand side of Eq. 1.7 is retained we have the Smoluchowski equation (Gardiner, Sec. 6.4, 1990) and under these circumstances the mean motion of the particles follows the fluid streamlines. As we shall see in Sec. 3, this leads to a situation that can not predict a crucial characteristic of many filtration processes, thus we will retain the second term on the right hand side of Eq. 1.7 so that our particle transport equation takes the form

$$\frac{\partial n_p}{\partial t} + \nabla\cdot\left\{\left[\mathbf{v} - \gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}\right]n_p\right\} = \nabla\cdot(D_p\nabla n_p), \quad \textit{in the } \gamma\textit{-phase} \qquad (1.8)$$

This correction to the Smoluchowski equation has been used by Yeh and Liu (1974), Banks and Kurowski (1983), and others for the study of particle transport in the filtration process, and derivations are available from de la Mora and Rosner (1981, 1982) and from

Peters and Ying (1991) in addition to the references cited above. Equation 1.8 represents the first smoothing process in the hierarchy of averaging processes illustrated in Figure 1. The next step in this process requires that we form the local volume average of Eq. 1.8.

2. Volume Averaging

At this point we are ready to express the complete transport problem under consideration, and we list the governing differential equations and boundary conditions as

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \left\{ \left[\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right] n_p \right\} = \nabla \cdot (D_p \nabla n_p) \tag{2.1}$$

B.C. 1 $\qquad n_p = 0, \quad$ *at the γ-σ interface* $\tag{2.2}$

$$0 = -\nabla p + \rho \mathbf{g} + \mu \nabla^2 \mathbf{v} \tag{2.3}$$

B.C. 2 $\qquad \mathbf{v} = 0, \quad$ *at the γ-σ interface* $\tag{2.4}$

$$\nabla \cdot \mathbf{v} = 0 \tag{2.5}$$

It should be clear that we have already used Eq. 2.5 with Eq. 1.7 in order to simplify that result to Eq. 1.8, and we will need to use Eq. 2.5 again in our analysis of the convective transport term in Eq. 2.1. The boundary condition represented by Eq. 2.2 must be thought of as a limiting case which will create an upperbound for the filtration efficiency. This boundary condition has been used by Ruckenstein and Prieve (1973), Shapiro and Brenner (1990), and others.

The method of volume averaging (Anderson and Jackson, 1967; Marle, 1967, Slattery, 1967; Whitaker, 1967) begins by associating with every point in space (in both the γ-phase and the σ-phase) an averaging volume that we denote by $\mathcal{V}$. Such a volume is illustrated in Figure 2 where we have located the centroid of the averaging volume by the position vector $\mathbf{x}$, the radius of the averaging volume by r_o, and the characteristic length of the γ-phase by ℓ_γ. We will make use of two averages in our analysis of Eq. 2.1 and the first of these is the *superficial* volume average which can be expressed as

$$\langle \psi_\gamma \rangle = \frac{1}{\mathcal{V}} \int_{V_\gamma} \psi_\gamma dV \tag{2.6}$$

Here ψ_γ is any function associated with the γ-phase and V_γ is the volume of the γ-phase contained within the averaging volume, $\mathcal{V}$. In addition to the superficial average, we will also make use of the *intrinsic* volume average that is defined by

Figure 2
Positions Vectors and Length Scales Associated with the Averaging Volume
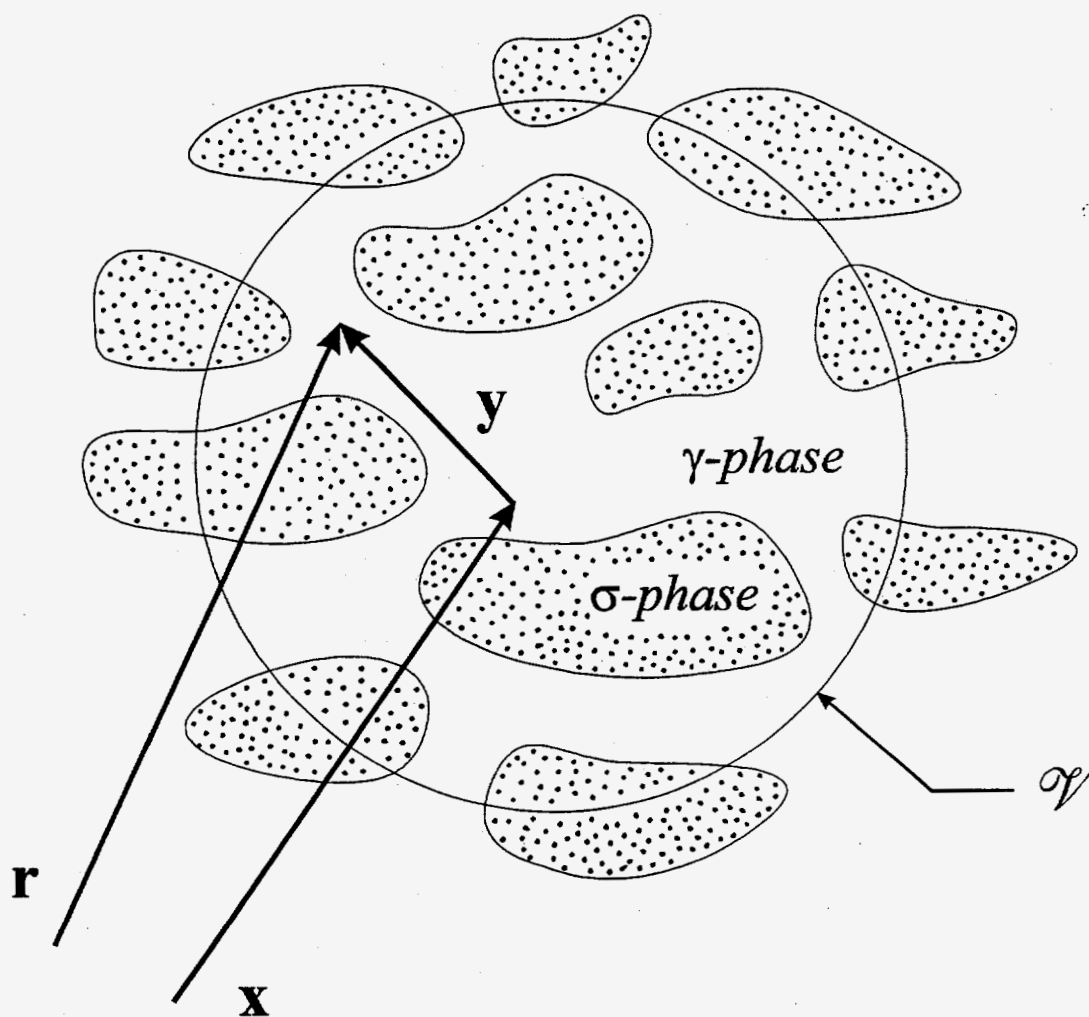
$$\langle \psi_\gamma \rangle^\gamma = \frac{1}{V_\gamma} \int_{V_\gamma} \psi_\gamma dV \tag{2.7}$$

These two averages are related by

$$\langle \psi_\gamma \rangle = \varepsilon_\gamma \langle \psi_\gamma \rangle^\gamma \tag{2.8}$$

The average velocity is often represented in terms of the *superficial* velocity, while the average particle concentration is typically represented in terms of an *intrinsic* average. To avoid confusion between these two averages we will always make use of the nomenclature indicated in Eqs. 2.6 through 2.8.

When we form the volume average of Eq. 2.1 we will encounter averages of gradients and we will need to convert these to gradients of averages by means of the spatial averaging theorem (Howes and Whitaker, 1985) which we represent as

$$\langle \nabla \psi_\gamma \rangle = \nabla \langle \psi_\gamma \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \psi_\gamma dA \tag{2.9}$$

We begin the analysis of the particle transport process by expressing the superficial average of Eq. 2.1 as

$$\left\langle \frac{\partial n_p}{\partial t} \right\rangle + \left\langle \nabla \cdot \left\{ [\mathbf{v} - \gamma^{-1}\mathbf{v} \cdot \nabla \mathbf{v}] n_p \right\} \right\rangle = \left\langle \nabla \cdot D_p \nabla n_p \right\rangle \tag{2.10}$$

and note that the first term can be written as

$$\left\langle \frac{\partial n_p}{\partial t} \right\rangle = \frac{\partial \langle n_p \rangle}{\partial t} \tag{2.11}$$

since V_γ is independent of time. The convective transport term in Eq. 2.10 requires the use of the averaging theorem which leads to

$$\left\langle \nabla \cdot (\mathbf{v} - \gamma^{-1}\mathbf{v} \cdot \nabla \mathbf{v}) n_p \right\rangle = \nabla \cdot \langle (\mathbf{v} - \gamma^{-1}\mathbf{v} \cdot \nabla \mathbf{v}) n_p \rangle \tag{2.12}$$

on the basis of the no-slip condition given by Eq. 2.4. The diffusive term on the right hand side of Eq. 2.10 provides us with

$$\langle \nabla \cdot D_p \nabla n_p \rangle = \nabla \cdot \langle D_p \nabla n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA \qquad (2.13)$$

Use of Eqs. 2.11 through 2.13 along with Eq. 2.8 in the form

$$\langle n_p \rangle = \varepsilon_\gamma \langle n_p \rangle^\gamma \qquad (2.14)$$

allows us to express the superficial average particle transport equation as

$$\underbrace{\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t}}_{accumulation} + \underbrace{\nabla \cdot \langle \mathbf{v} n_p \rangle}_{convection} - \underbrace{\gamma^{-1} \nabla \cdot \langle (\mathbf{v} \cdot \nabla \mathbf{v}) n_p \rangle}_{\substack{particle \\ inertia\ convection}}$$

$$\underbrace{\nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} n_p dA \right] \right\}}_{diffusion} + \underbrace{\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA}_{particle\ capture} \qquad (2.15)$$

Here we have identified the correction to the Smoluchowski equation as the *particle inertia convection*, and it is this term that plays a key role in the determination of the "most penetrating particle size" in the filtration process.

In order to eliminate the point value of the particle concentration in the diffusion term, we use the following decomposition (Gray, 1975) for the particle density

$$n_p = \langle n_p \rangle^\gamma + \tilde{n}_p \qquad (2.16)$$

and later we will use the velocity decomposition given by

$$\mathbf{v} = \langle \mathbf{v} \rangle^\gamma + \tilde{\mathbf{v}} \qquad (2.17)$$

One can use Eq. 2.16 in the diffusion term and follow the type of analysis given by Whitaker (Sec. 2, 1986a) or Quintard and Whitaker (Sec. II, 1993) to obtain

$$\nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} n_p dA \right] \right\} = \nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] \right\} \qquad (2.18)$$

This development makes use of the lemma extracted from Eq. 2.9

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$$\nabla \varepsilon_\gamma = -\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} dA \qquad (2.19)$$

along with the two length-scale constraints given by

$$\ell_\gamma \ll r_o \qquad (2.20)$$

$$\frac{r_o^2}{L^2} \ll 1 \qquad (2.21)$$

Here one should think of L as the smallest characteristic length associated with a macroscopic quantity such as ε_γ, $\langle n_p \rangle^\gamma$, $\nabla\langle n_p \rangle^\gamma$, etc. These length-scale constraints were originally developed by Carbonell and Whitaker (Sec. 2, 1984) and a more thorough discussion is available in the more recent work of Quintard and Whitaker (1994b). One can also use the decomposition given by Eq. 2.16 in order to express the particle capture term as

$$\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA = -(\nabla\varepsilon_\gamma) \cdot D_p \nabla\langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \qquad (2.22)$$

Use of Eqs. 2.17 and 2.21 in Eq. 2.15 leads to

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla\cdot\langle \mathbf{v}n_p \rangle - \gamma^{-1}\nabla\cdot\langle (\mathbf{v}\cdot\nabla\mathbf{v})n_p \rangle = \qquad (2.23)$$

$$\nabla\cdot\left\{ \varepsilon_\gamma D_p \left[\nabla\langle n_p \rangle^\gamma + \frac{1}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma}\tilde{n}_p dA \right] \right\} - (\nabla\varepsilon_\gamma)\cdot D_p\nabla\langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\tilde{n}_p dA$$

and in order to complete the averaging procedure we would like to express the convective transport in terms of $\langle n_p \rangle^\gamma$ and $\tilde{n}_p$. One can follow Carbonell and Whitaker (1983, 1984) in order to represent the traditional convective transport as

$$\langle \mathbf{v}n_p \rangle = \underbrace{\varepsilon_\gamma \langle \mathbf{v} \rangle^\gamma \langle n_p \rangle^\gamma}_{\substack{traditional \\ convective\ transport}} + \underbrace{\langle \tilde{\mathbf{v}}\tilde{n}_p \rangle}_{\substack{traditional \\ dispersive\ transport}} \qquad (2.24)$$

The so-called inertial contribution to the convective transport is algebraically more complex, and it is convenient to use only the decomposition given by Eq. 2.16 to obtain

$$\langle(\mathbf{v}\cdot\nabla\mathbf{v})n_p\rangle = \underbrace{\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle\langle n_p\rangle^\gamma}_{\substack{particle\\inertia\ convection}} + \underbrace{\langle(\mathbf{v}\cdot\nabla\mathbf{v})\tilde{n}_p\rangle}_{\substack{particle\\inertia\ dispersion}} \qquad (2.25)$$

On the basis of Eqs. 2.24 and 2.25 we can express the volume averaged particle transport equation as

$$\underbrace{\varepsilon_\gamma\frac{\partial\langle n_p\rangle^\gamma}{\partial t}}_{accumulation} + \underbrace{\nabla\cdot\left(\varepsilon_\gamma\langle\mathbf{v}\rangle^\gamma\langle n_p\rangle^\gamma - \gamma^{-1}\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle\langle n_p\rangle^\gamma\right)}_{convection} + \underbrace{\nabla\cdot\left(\langle\tilde{\mathbf{v}}\tilde{n}_p\rangle - \gamma^{-1}\langle(\mathbf{v}\cdot\nabla\mathbf{v})\tilde{n}_p\rangle\right)}_{dispersion}$$

$$= \nabla\cdot\underbrace{\left\{\varepsilon_\gamma D_p\left[\nabla\langle n_p\rangle^\gamma + \frac{1}{V_\gamma}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\tilde{n}_p dA\right]\right\}}_{diffusion} + \underbrace{\frac{1}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\tilde{n}_p dA}_{particle\ capture} \qquad (2.26)$$

Here we have imposed the simplification

$$(\nabla\varepsilon_\gamma)\cdot D_p\nabla\langle n_p\rangle^\gamma \ll \frac{1}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\tilde{n}_p dA \qquad (2.27)$$

and the length-scale constraint associated with this restriction is given by

$$\frac{\ell_\gamma^3\ell_{bl}}{\ell_\sigma^2 L_\varepsilon L_n} \ll 1 \qquad (2.28)$$

The derivation of this result requires an estimate of $\tilde{n}_p$ and definitions of the various length scales, and the analysis is given by Quintard and Whitaker (1995).

Before moving on to the closure problem we should list the volume average forms of the Stokes' equation given by Eq. 2.3 and the continuity equation that was presented earlier as Eq. 2.5. The volume averaged forms of these two equations have been developed in detail elsewhere (Whitaker, 1986b; Quintard and Whitaker, 1994b) and we simple list the volume averaged form of Eq. 2.3 as

$$\langle \mathbf{v} \rangle = -\frac{\mathbf{K}}{\mu} \cdot \left(\nabla \langle p \rangle^\gamma - \rho \mathbf{g} \right), \quad Darcy's \ law \tag{2.29}$$

in which $\langle \mathbf{v} \rangle$ represents the *superficial* volume averaged velocity and $\langle p \rangle^\gamma$ represents the *intrinsic* volume averaged pressure. The volume averaged continuity equation can be expressed either in terms of the superficial average velocity

$$\nabla \cdot \langle \mathbf{v} \rangle = 0 \tag{2.30}$$

or in terms of the intrinsic average velocity

$$\nabla \cdot (\varepsilon_\gamma \langle \mathbf{v} \rangle^\gamma) = 0 \tag{2.31}$$

Here we have made use of the nomenclature given by Eqs. 2.6 and 2.7 and the relation between the two averages indicated by Eq. 2.8.

In order to obtain a closed form of Eq. 2.26, we need to develop the boundary value problem for $\tilde{n}_p$ and we need to show how $\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle$ can be determined by Darcy's law. In the *general* analysis of the filtration process we definitely need to take porosity variations into account via the method of large-scale averaging (Quintard and Whitaker, 1987); however, in the development of the closure problem it is permissible to consider a local homogeneous region in which variations of ε_γ can be ignored. Under these circumstances we can divide Eq. 2.26 by ε_γ to obtain

$$\frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v} \rangle^\gamma \langle n_p \rangle^\gamma - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \langle n_p \rangle^\gamma \right) + \nabla \cdot \left(\langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^\gamma - \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^\gamma \right)$$

$$= \nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle^\gamma + \frac{1}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] \right\} + \frac{\varepsilon_\gamma^{-1}}{\mathcal{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \tag{2.32}$$

With this intrinsic form of the particle transport equation we are ready to begin the derivation of the closure problem.

3. Closure Problem

In order to develop the governing differential equation for $\tilde{n}_p$ we recall Eq. 1.8

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \left\{ \left[\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right] n_p \right\} = \nabla \cdot (D_p \nabla n_p) \tag{3.1}$$

and remember Eq. 2.16 so that Eq. 2.32 can be subtracted from Eq. 3.1 to obtain

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left\{ \left[\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right] n_p - \left[\langle \mathbf{v} \rangle^\gamma - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right] \langle n_p \rangle^\gamma \right\}$$

$$-\nabla \cdot \langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^\gamma + \gamma^{-1} \nabla \cdot \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^\gamma = \nabla \cdot (D_p \nabla \tilde{n}_p) -$$

$$-\nabla \cdot \left[\frac{D_p}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] - \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \qquad (3.2)$$

The second and third terms in this result can be arranged as

$$\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) n_p - \left(\langle \mathbf{v} \rangle^\gamma - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \langle n_p \rangle^\gamma =$$

$$\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \tilde{n}_p - \left[\left(\langle \mathbf{v} \rangle^\gamma - \mathbf{v} \right) - \gamma^{-1} \left(\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma - \mathbf{v} \cdot \nabla \mathbf{v} \right) \right] \langle n_p \rangle^\gamma \qquad (3.3)$$

If we neglect variations of the porosity in the closure problem, we can use the various forms of the continuity equation to obtain

$$\nabla \cdot \left(\langle \mathbf{v} \rangle^\gamma - \mathbf{v} \right) = 0 \qquad (3.4)$$

and this allows us to substitute Eq. 3.3 into Eq. 3.2 and obtain the following transport equation for $\tilde{n}_p$

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \tilde{n}_p \right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \cdot \nabla \langle n_p \rangle^\gamma}_{source} - \underbrace{\left\{ \nabla \cdot \left[\gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \right\} \langle n_p \rangle^\gamma}_{source}$$

$$- \underbrace{\nabla \cdot \langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^\gamma + \gamma^{-1} \nabla \cdot \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^\gamma}_{non\text{-}local\ convective\ transport} = \nabla \cdot \left(D_p \nabla \tilde{n}_p \right) - \underbrace{\nabla \cdot \left[\frac{D_p}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right]}_{\substack{non\text{-}local\ diffusive \\ transport}}$$

$$- \underbrace{\frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA}_{particle\ capture} \qquad\qquad (3.5)$$

Here we see terms representing

1. The classic effects of accumulation, local convection, and local diffusion.
2. Non-local convection and non-local diffusion.
3. Sources proportional to $\nabla \langle n_p \rangle^\gamma$ and $\langle n_p \rangle^\gamma$.
4. Particle capture.

We use the word *non-local* to describe those terms which involve integrals of $\tilde{n}_p$, and one can draw upon previous studies (Carbonell and Whitaker, 1984; Whitaker, 1986a; Quintard and Whitaker, 1993; Quintard and Whitaker, 1994a) to argue that these terms are negligible when length-scale constraints such as those indicated by Eqs. 2.20 and 2.21 are valid. The analysis consists of comparing the non-local terms with the associated local terms and demonstrating that the former are smaller than the latter by a factor of ℓ / L. This occurs because the non-local terms involve the derivatives of *average quantities* while the local terms always contain the derivatives of *point quantities*. Because ℓ / L is always small compared to one, the non-local terms can be neglected and Eq. 3.5 simplifies to

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left[(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1} (\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle) \right] \cdot \nabla \langle n_p \rangle^\gamma}_{source} - \underbrace{\left\{ \nabla \cdot \left[\gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \right\} \langle n_p \rangle^\gamma}_{source}$$

$$= \nabla \cdot \left(D_p \nabla \tilde{n}_p \right) - \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \qquad\qquad (3.6)$$

The last term in this result is also a non-local term; however, it is not negligible since it represents the rate at which particles are captured per unit volume.

Use of the boundary condition given by Eq. 2.2 and the decomposition represented by Eq. 2.16 leads to the following boundary condition

$$\text{B.C. 1} \qquad\qquad \tilde{n}_p = - \underbrace{\langle n_p \rangle^\gamma}_{source} , \qquad at\ the\ \gamma\text{-}\sigma\ interface \qquad\qquad (3.7)$$

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and in order to determine the $\tilde{n}_p$-field in some local, representative region, we are forced to accept the spatially periodic model of a porous medium and impose the following periodicity condition.

Periodicity:
$$\tilde{n}_p(\mathbf{r} + \ell_i) \;=\; \tilde{n}_p(\mathbf{r}), \qquad i \;=\; 1,2,3 \tag{3.8}$$

In addition, when the length-scale constraints indicated by Eqs. 2.20 and 2.21 are valid, Carbonell and Whitaker (1984) have shown that the average of the spatial deviation can be set equal to zero and we express this idea as

Average:
$$\langle \tilde{n}_p \rangle^\gamma \;=\; 0 \tag{3.9}$$

Strictly speaking, we need an initial condition for $\tilde{n}_p$ to complete our problem statement; however, both the volume averaged equation given by Eq. 2.26 and the closure equation represented by Eq. 3.6 can be treated as quasi-steady, thus the initial condition for both $\langle n_p \rangle^\gamma$ and $\tilde{n}_p$ can be ignored. This means that the closure problem takes the quasi-steady form given by

QUASI-STEADY CLOSURE PROBLEM

$$\nabla \cdot \left[(\mathbf{v} - \gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v})\tilde{n}_p \right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1}(\mathbf{v}\cdot\nabla\mathbf{v} - \langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle) \right]\cdot\nabla\langle n_p\rangle^\gamma}_{source} - \underbrace{\left\{ \nabla\cdot\left[\gamma^{-1}\left(\mathbf{v}\cdot\nabla\mathbf{v} - \langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle^\gamma\right)\right]\right\}\langle n_p\rangle^\gamma}_{source}$$

$$= \;\; \nabla\cdot\left(D_p \nabla\tilde{n}_p \right) \;-\; \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma}\cdot D_p \nabla\tilde{n}_p \, dA \tag{3.10a}$$

B.C. 1
$$\tilde{n}_p \;=\; \underbrace{-\langle n_p\rangle^\gamma}_{source}, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{3.10b}$$

Periodicity:
$$\tilde{n}_p(\mathbf{r} + \ell_i) \;=\; \tilde{n}_p(\mathbf{r}), \qquad i \;=\; 1,2,3 \tag{3.10c}$$

Average:
$$\langle \tilde{n}_p \rangle^\gamma \;=\; 0 \tag{3.10d}$$

The form of this boundary value problem suggests a representation for $\tilde{n}_p$ given by

$$\tilde{n}_p \;=\; \mathbf{b}\cdot\nabla\langle n_p\rangle^\gamma - s\langle n_p\rangle^\gamma \tag{3.11}$$

in which **b** and *s* are referred to as the *closure variables* or the *mapping variables* since they map the sources onto the spatial deviation concentration. One can draw upon a series of studies associated with the closure problem (Eidsath et al., 1983; Nozad et al., 1985; Crapiste et al., 1986; Ochoa et al, 1986; Quintard and Whitaker, 1993 and 1994a) to conclude that the vector **b** and the scalar *s* are determined by two boundary value problems that are analogous to the problem given by Eqs. 3.10. The first of these problems determines the vector, **b**, and this problem is given by

PROBLEM I

$$\nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \mathbf{b} \right] + \left[\tilde{\mathbf{v}} - \gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^{\gamma} \right) \right] \quad =$$

$$\nabla \cdot \left(D_p \nabla \mathbf{b} \right) + \frac{\varepsilon_{\gamma}^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \mathbf{b} \, dA \tag{3.12a}$$

B.C. 1 $\qquad\qquad \mathbf{b} = 0 , \quad$ *at the γ - σ interface* $\tag{3.12b}$

Periodicity: $\qquad \mathbf{b}(\mathbf{r} + \ell_i) = \mathbf{b}(\mathbf{r}), \quad i = 1,2,3 \tag{3.12c}$

Average: $\qquad\qquad \langle \mathbf{b} \rangle^{\gamma} = 0 \tag{3.12d}$

For the process of filtration, the second problem is much more important than the first since it is used to determine the capture coefficient, k_{eff}. This problem is given by

PROBLEM II

$$\nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) s \right] + \nabla \cdot \left[\gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^{\gamma} \right) \right] \quad =$$

$$= \nabla \cdot \left(D_p \nabla s \right) - \frac{\varepsilon_{\gamma}^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla s \, dA \tag{3.13a}$$

B.C. 1 $\qquad\qquad s = 1 , \quad$ *at the γ - σ interface* $\tag{3.13b}$

Periodicity: $\qquad s(\mathbf{r} + \ell_i) = s(\mathbf{r}), \quad i = 1,2,3 \tag{3.13c}$

Average: $\qquad\qquad \langle s \rangle^{\gamma} = 0 \tag{3.13d}$

These two closure problems can be solved using the numerical methods described by Quintard and Whitaker (1993), and their solution allows us to determine the mapping variables for the *spatial deviation* particle concentration represented by Eq. 3.11.

Substitution of Eq. 3.11 into the volume averaged transport equation given by Eq. 2.26 leads to the closed form of that equation which contains effective transport and capture coefficients which are determined by Problems I and II.

CLOSED FORM

The closed form of Eq. 2.26 can be expressed as

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left[\left(\langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle \right) \langle n_p \rangle^\gamma \right] - (\mathbf{d} + \mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma =$$

$$\nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{eff} \langle n_p \rangle^\gamma \qquad (3.14)$$

and it is important to recognize that this is a *superficial average* transport equation. This means that each term represents a certain quantity *per unit volume of the porous medium* and not per unit volume of the fluid phase. This is obvious for the accumulation term; however, it can be confusing for the particle capture term. The various coefficients that appear in Eq. 3.14 are defined as

$$\mathbf{u} = \frac{1}{\mathcal{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \mathbf{b} \, dA \qquad (3.15)$$

$$\mathbf{d} = -D_p \left[\frac{1}{\mathcal{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} s \, dA \right] + \left[\langle \tilde{\mathbf{v}} s \rangle - \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) s \rangle \right] \qquad (3.16)$$

$$\mathbf{D}^* = \varepsilon_\gamma D_p \left(\mathbf{I} + \frac{1}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \mathbf{b} \, dA \right) - \langle \tilde{\mathbf{v}} \mathbf{b} \rangle - \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \mathbf{b} \rangle \qquad (3.17)$$

$$k_{eff} = \frac{1}{\mathcal{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla s \, dA \qquad (3.18)$$

The last of these coefficients represents the principle objective of this study, thus it is Problem II that will provide results that can be compared with Brownian dynamics and laboratory experiments. In order to determine k_{eff} it is convenient to transform Eqs. 3.13 by means of the following representation

$$s = 1 + k_{eff}S \tag{3.19}$$

Here the quantity S has units of time and the boundary problem for this new variable is given by

PROBLEM II

$$\nabla \cdot (vS) - \nabla \cdot \left[\left(\gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) S \right] = \nabla \cdot \left(D_p \nabla S \right) - \varepsilon_\gamma^{-1} \tag{3.20a}$$

B.C. 1 $S = 0$,   *at the γ-σ interface* $\tag{3.20b}$

Periodicity: $S(\mathbf{r} + \ell_i) = S(\mathbf{r})$, $i = 1,2,3$ $\tag{3.20c}$

Average: $k_{eff} = - \left(\langle S \rangle^\gamma \right)^{-1}$ $\tag{3.20d}$

In deriving this result from Eqs. 3.13 we have used the continuity equation given by Eq. 2.5, and we have also made use of the simplification indicated by

$$\nabla \cdot \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma = 0 \tag{3.21}$$

This is consistent with the simplification used in the closure problem that variations in the porosity can be neglected.

In order to complete the closure of Eq. 3.14 we must represent the term $\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle$ in a form that can be determined by the use of Darcy's law as given by Eq. 2.29. From the closure problem for Darcy's law (Whitaker, Sec. 4, 1986b) we know that the point velocity is given in terms of the intrinsic average velocity by (Barrère et al., 1992)

$$\mathbf{v} = -\left(\mathbf{D} \cdot \mathbf{K}^{-1} \right) \cdot \langle \mathbf{v} \rangle \tag{3.22}$$

Here $\mathbf{D}$ is a second order mapping tensor that is determined by a modified closure problem that has been solved by a variety of authors (Sangani and Acrivos, 1982; Snyder and Stewart, 1966; Zick and Homsy, 1982), and $\mathbf{K}$ is the Darcy's law permeability tensor that appears in Eq. 2.29. In order to use this result with the convective transport term in Eq. 3.14, we make use of the continuity equation, the averaging theorem, and the no-slip condition to obtain

$$\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle = \langle \nabla \cdot (\mathbf{v}\mathbf{v}) \rangle = \nabla \cdot \langle \mathbf{v}\mathbf{v} \rangle \tag{3.23}$$

Use of Eq. 3.22 in Eq. 3.23 leads to

$$\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle = \nabla \cdot \left[\langle \mathbf{DD} \rangle : \left(\mathbf{K}^{-1} \mathbf{K}^{-1} \right) : \langle \mathbf{v} \rangle \langle \mathbf{v} \rangle \right] \tag{3.24}$$

in which the proper dyadic multiplication can be inferred from Eq. 3.22. When this result is used in Eq. 3.14 we obtain the *completely closed form* of the volume averaged particle transport equation given by

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left\{ \left(\langle \mathbf{v} \rangle - \gamma^{-1} \nabla \cdot \left[\langle \mathbf{DD} \rangle : \left(\mathbf{K}^{-1} \mathbf{K}^{-1} \right) : \langle \mathbf{v} \rangle \langle \mathbf{v} \rangle \right] \right) \langle n_p \rangle^\gamma \right\} - (\mathbf{d} + \mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma =$$

$$\nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{eff} \langle n_p \rangle^\gamma \tag{3.25}$$

For simplicity we define a volume averaged particle velocity according to

$$\langle \mathbf{v}_p \rangle = \langle \mathbf{v} \rangle - \gamma^{-1} \nabla \cdot \left(\langle \mathbf{DD} \rangle : \left(\mathbf{K}^{-1} \mathbf{K}^{-1} \right) : \langle \mathbf{v} \rangle \langle \mathbf{v} \rangle \right) \tag{3.26}$$

so that Eq. 3.25 takes the form

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v}_p \rangle \langle n_p \rangle^\gamma \right) - (\mathbf{d} + \mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma = \nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{eff} \langle n_p \rangle^\gamma \tag{3.27}$$

In order to understand the filtration process, we need to examine the effective coefficients in Eq. 3.27; however, it is the capture coefficient, k_{eff}, that dominates the filtration process, and the details concerning $\mathbf{d}$, $\mathbf{u}$, and $\mathbf{D}^*$ can be found in Appendix B of Quintard and Whitaker (1995). Solution of the closure problem to determine k_{eff} is described in the next section.

## 4. Determination of the Capture Coefficient

The simplest model of a fibrous porous medium is a regular array of cylinders such as that shown in Figure 3. For most practical cases, the Reynolds number for flow in fibrous filters is less than one, thus we can use Stokes' equations to determine the fluid velocity field. For the case of a *macroscopically uniform flow* one can justify the use of spatially periodic boundary conditions (Sanchez-Palencia, 1980), and the fluid mechanical problem to be solved is given by (Barrère et al., 1992)

$$\nabla \cdot \mathbf{v} = 0 \tag{4.1}$$

$$0 = -\nabla p + \rho \mathbf{g} + \mu \nabla^2 \mathbf{v} \tag{4.2}$$

B.C. 1 $\mathbf{v} = 0$,   *at the $\gamma - \sigma$ interface* \hfill (4.3)

Figure 3
Spatially Periodic Array of Cylinders
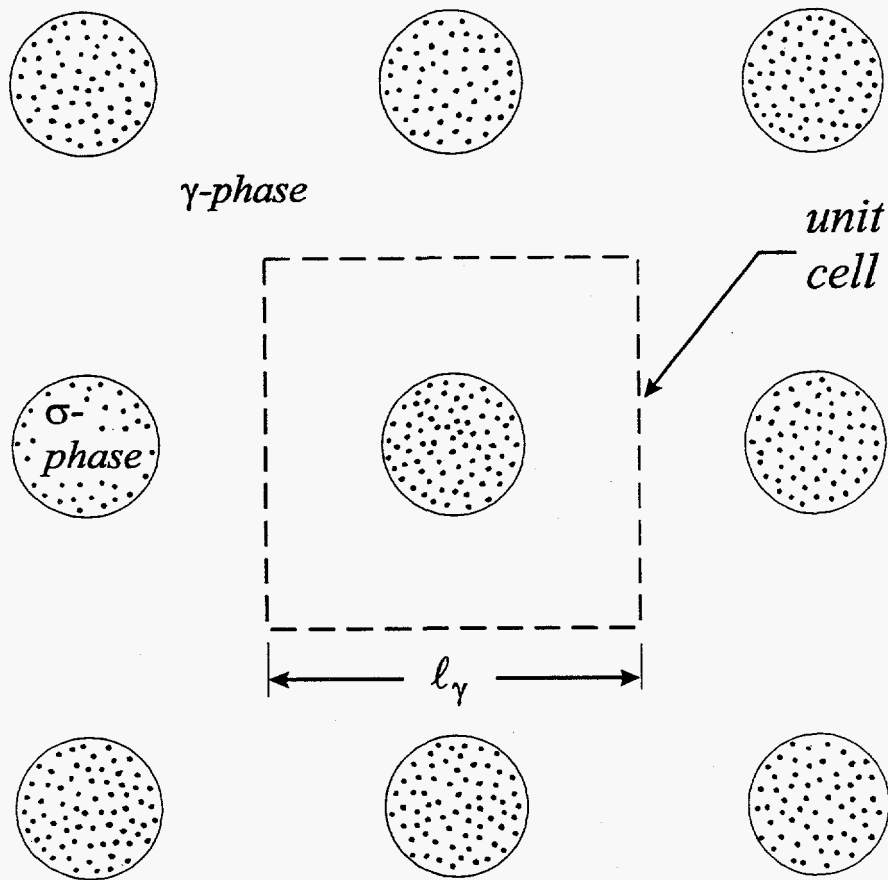
Periodicity: $\mathbf{v}(\mathbf{r}+\ell_i)=\mathbf{v}(\mathbf{r})$, $i=1, 2,$ and 3   (4.4)

Here we have imposed the no-slip condition with the thought that slip will be unimportant when the fiber diameter is larger than several micrometers. It is well known that the entrance length for the Stokes' flow problem is on the order of the characteristic length, ℓ_γ, thus the velocity field determined by Eqs. 4.1 through 4.4 is representative of the conditions essentially everywhere in the uniform array shown in Figure 3. However, the *particle velocity* field need not have the same entrance region as the *fluid velocity* field, and it is of some interest to know how many unit cells are required in order to develop a spatially periodic particle velocity since this is required in order that the periodicity conditions represented by Eqs. 3.10c, 3.12c, and 3.13c be valid. The entrance region associated with the particle concentration field has been explored by Quintard and Whitaker (1995) and their results indicated that the entrance length for the particle velocity field is on the order of ten unit cells. This represents a small portion of any fibrous filter, thus the entrance region for the particle velocity field can be neglected and we can make use of spatially periodic conditions (Shapiro and Brenner, 1990) in order to solve the two closure problems given by Eqs 3.12 and 3.20. This will provide us with theoretical values of the effective coefficients in the volume averaged particle transport equation which we list here as

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v}_p \rangle \langle n_p \rangle^\gamma \right) - (\mathbf{d}+\mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma \;\; = \nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{\text{eff}} \langle n_p \rangle^\gamma \qquad (4.6)$$

One must remember that the velocity, $\langle \mathbf{v}_p \rangle$, *does not* represent the volume average of the particle velocity that appears in the Langevin equation, but instead it represents the inertia corrected average velocity defined by Eq. 3.21 which can also be expressed as

$$\langle \mathbf{v}_p \rangle \;\; = \;\; \langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle \qquad (4.7)$$

For the special case of a *macroscopically uniform flow*, Eq. (3.24) requires that;

$$\langle \mathbf{v}_p \rangle = \langle \mathbf{v} \rangle \qquad (4.8)$$

however, this need not be the case for heterogeneous filters and in future studies we will examine this matter more carefully. The non-traditional contributions to the convective transport represented by the "velocities", $\mathbf{d}$ and $\mathbf{u}$, are the result of the particle capture process, and this effect has been thoroughly documented by Paine et al. (1983) for the case of adsorption and chemical reaction in capillary tubes. The values of $\mathbf{d}$ and $\mathbf{u}$ are determined by Eqs. 3.15 and 3.16 and it is important to know the magnitude of these two terms relative to $\langle \mathbf{v}_p \rangle$. Calculated values of the x-components of $\mathbf{d}$ and $\mathbf{u}$ are given by Quintard and Whitaker (1995) and they show that these terms may contribute as much as

10% to the convective transport in Eq. 4.6. While this is not negligible, it is not a key issue in terms of the comparison of the theory with laboratory experiments. Thus we will discard these terms, along with the dispersive transport so that our superficial volume averaged transport equation takes the form

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v} \rangle \langle n_p \rangle^\gamma \right) = - k_{eff} \langle n_p \rangle^\gamma \tag{4.9}$$

This result will be quasi-steady when the following constraint is satisfied

$$k_{eff} t \gg 1 \tag{4.10}$$

and for incompressible flows one can use the continuity equation given by Eq. 2.30 in order the express Eq. 4.9 as

$$\langle \mathbf{v} \rangle \cdot \nabla \cdot \langle n_p \rangle^\gamma = - k_{eff} \langle n_p \rangle^\gamma \tag{4.11}$$

One should think of this result as being a reasonable approximation for homogeneous filters; however, real filters are heterogeneous and the terms that have been discarded in going from Eq. 4.6 to Eq. 4.11 will be retained in future studies of heterogeneous porous media.

CELLULAR EFFICIENCY

In order to present our results for k_{eff} in a traditional form, we will write Eq. 4.11 as

$$\langle v_x \rangle \frac{d \langle n_p \rangle^\gamma}{dx} = - k_{eff} \langle n_p \rangle^\gamma \tag{4.12}$$

and note that for our unit cell calculations, or any homogeneous porous filter, we have

$$\mathbf{i} \cdot \langle \mathbf{v}_p \rangle = \mathbf{i} \cdot \langle \mathbf{v} \rangle = \langle v_x \rangle \tag{4.13}$$

We can solve Eq. 4.12 in order to represent the change in particle concentration that takes place across a unit cell as

$$\frac{\langle n_p \rangle^\gamma \big|_{x=0} - \langle n_p \rangle^\gamma \big|_{x=\ell_\gamma}}{\langle n_p \rangle^\gamma \big|_{x=0}} = 1 - e^{-\frac{k_{eff} \ell_\gamma}{\langle v_x \rangle}} \tag{4.14}$$

It is convenient to define the left hand side of this result as the *cellular efficiency*, η_c , in order to distinguish it from the *single fiber efficiency*, and this leads to

$$\eta_c = 1 - e^{-\frac{k_{eff}\ell_\gamma}{\langle v_x \rangle}}, \quad \textit{cellular efficiency} \quad (4.15)$$

The closure problem given by Eqs. 3.20 was solved using numerical methods similar to those described by Quintard and Whitaker (1993, 1994b). To illustrate the general nature of the solutions for the cellular efficiency, η_c, calculations were carried out for the parameters listed in Table 1.

Table 1 Physical Properties

particle diameter	d_p = 0.5 and 0.1 μm
fiber diameter	$2a$ = 0.5 μm
particle density	ρ_p = 4.0 g/cm^3
temperature	283.0 K
viscosity	1.8×10^{-5} Pa s
porosity	ε_γ = 0.95

The illustrative values for η_c are shown in Figure 4 as a function of the particle diameter. The Stokes is defined by

$$St = \frac{\langle v_x \rangle^\gamma \rho_p d_p^2 c_s}{18\mu\ell_\gamma}$$

and the curve for $St = 0$ represents the purely diffusive case and does not exhibit a minimum. This means that the Smoluchowski equation for the particle concentration cannot be used to determine the most penetrating particle size. The curve for finite Stokes numbers, which range from 10^{-4} to almost 10^{-1}, indicates a minimum cellular efficiency for particle diameters on the order of 1 to 2 micrometers. In thinking about the results shown in Figure 4, one must be careful to remember that the additional convective transport represented by the coefficients **u** and **d** has been neglected along with the dispersive transport. Inclusion of these effects would change the values presented in Figure 4 but not the conclusion that the corrected Smoluchowski equation exhibits a minimum in the cellular efficiency as a function of particle diameter.

If a filter can be thought of as a series of unit cells, the cellular efficiency determined on a theoretical basis can be determined with the filter efficiency determined on an experimental basis. This approach neglects the heterogeneities illustrated in Figure 1 and the inclusion of the effects of those heterogeneities will be the subject of future studies. In Figure 5 we have shown a comparison with the work of Lee and Liu (1982)

Figure 4
Cellular Efficiency as a Function of Particle Diameter

$(2a = 0.5 \ \mu m, \ \rho_p = 4000 \ kg/m^3$
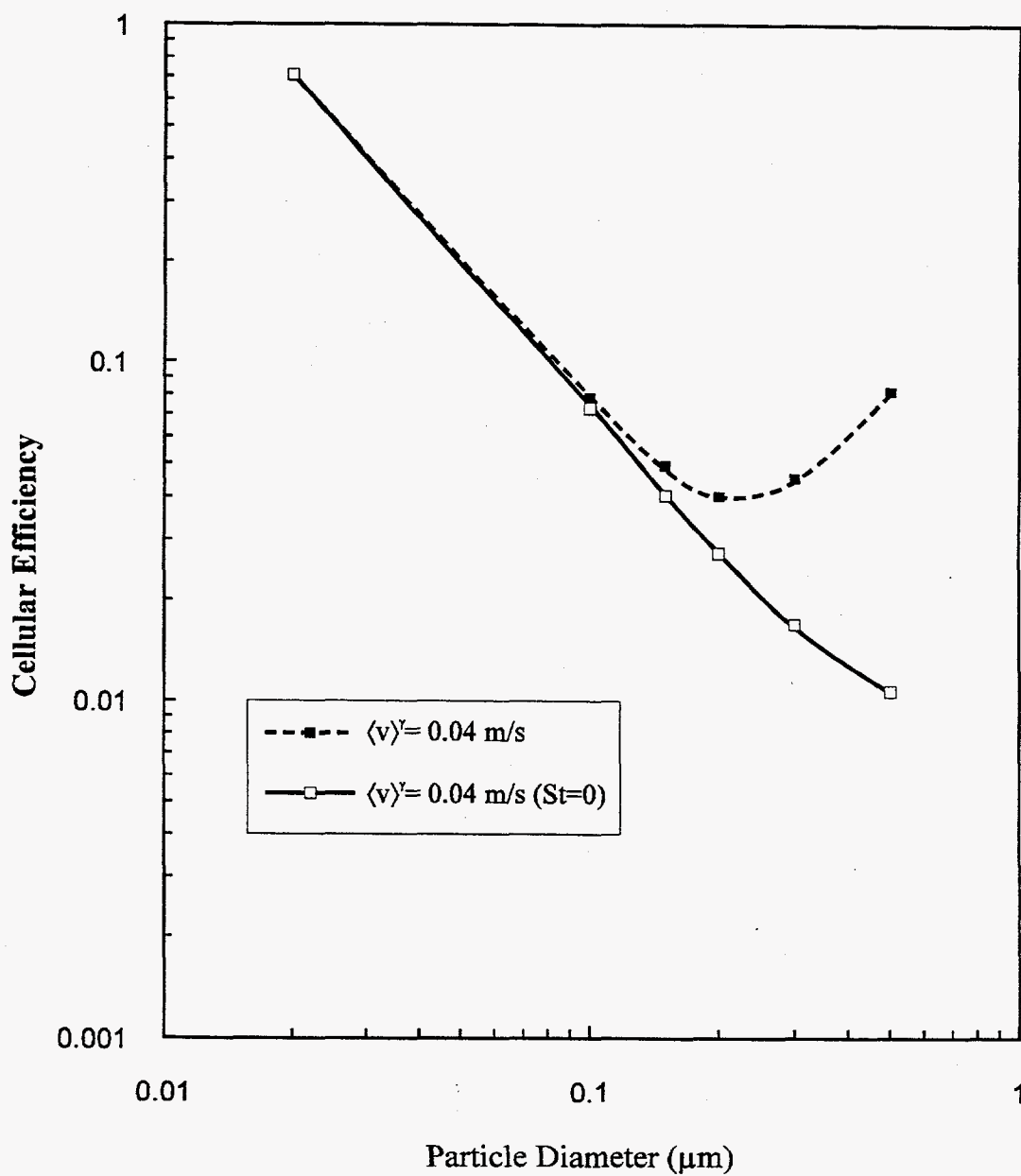$T=283 \ K, \ \mu=1.8 \ 10^{-5} \ Pa \ s, \ \varepsilon_\gamma=0.95)$

Figure 5
Comparison with Laboratory Experiments

$(2a = 11 \ \mu m, \ \rho_p = 1000 \ kg/m^3$
$T=278 \ K, \ \mu=1.83 \ 10^{-5} \ Pa \ s, \ \varepsilon_\gamma=0.849)$
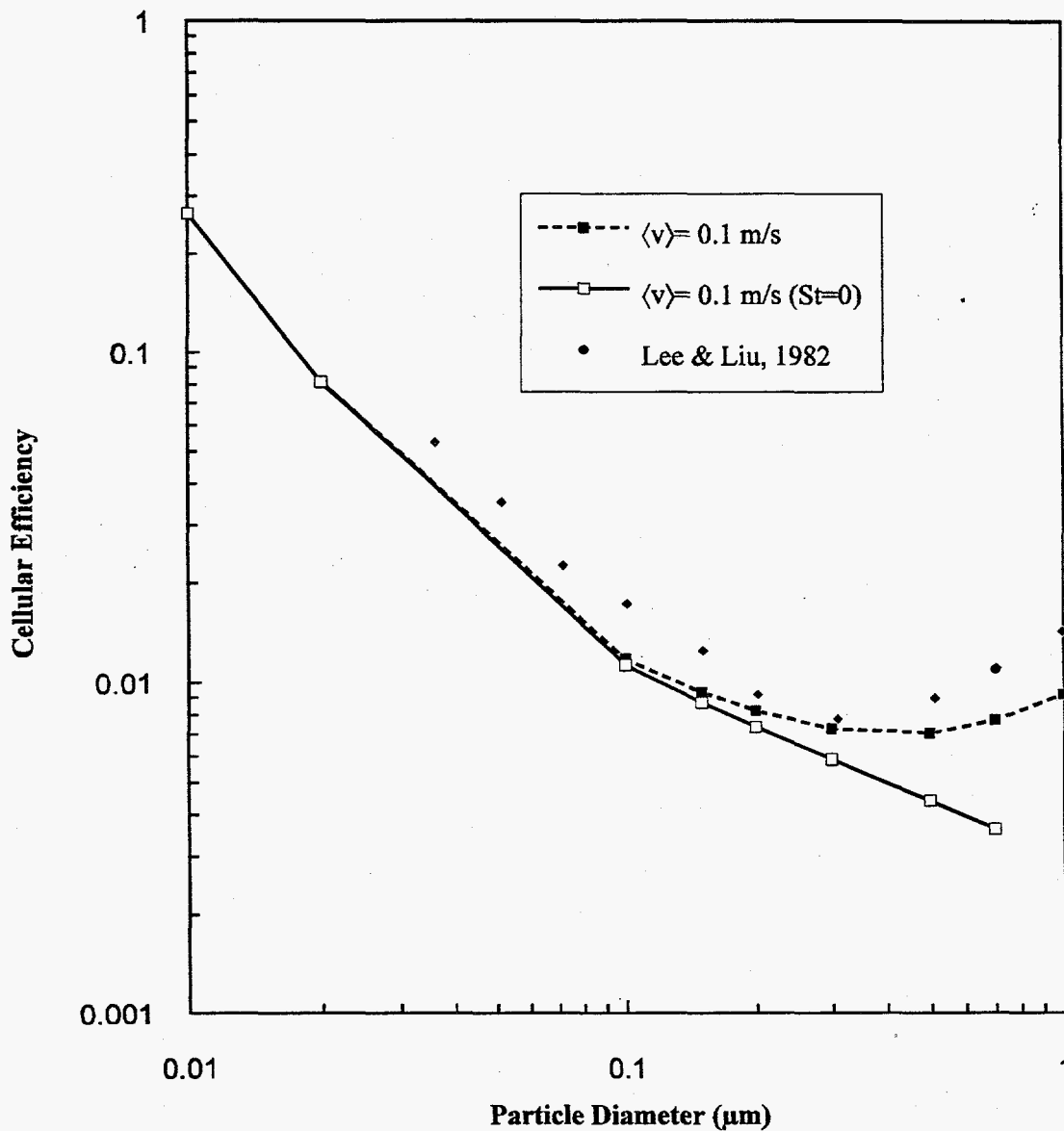
Figure 6
Comparison with Laboratory Experiments

$(2a = 11 \ \mu m, \ \rho_p = 1000 \ kg/m^3$
$T=278 \ K, \ \mu=1.83 \ 10^{-5} \ Pa \ s, \ \varepsilon_\gamma=0.849)$
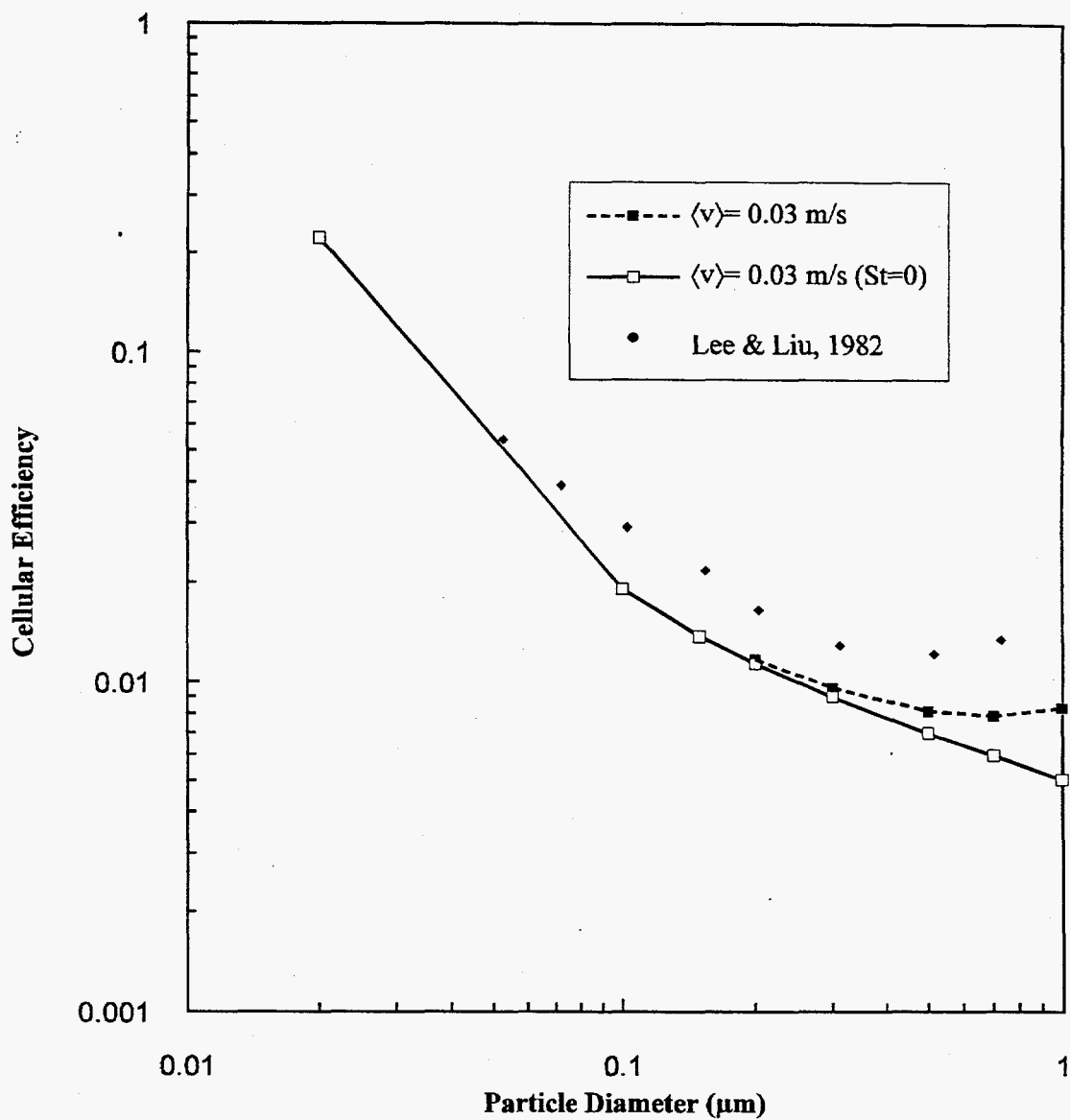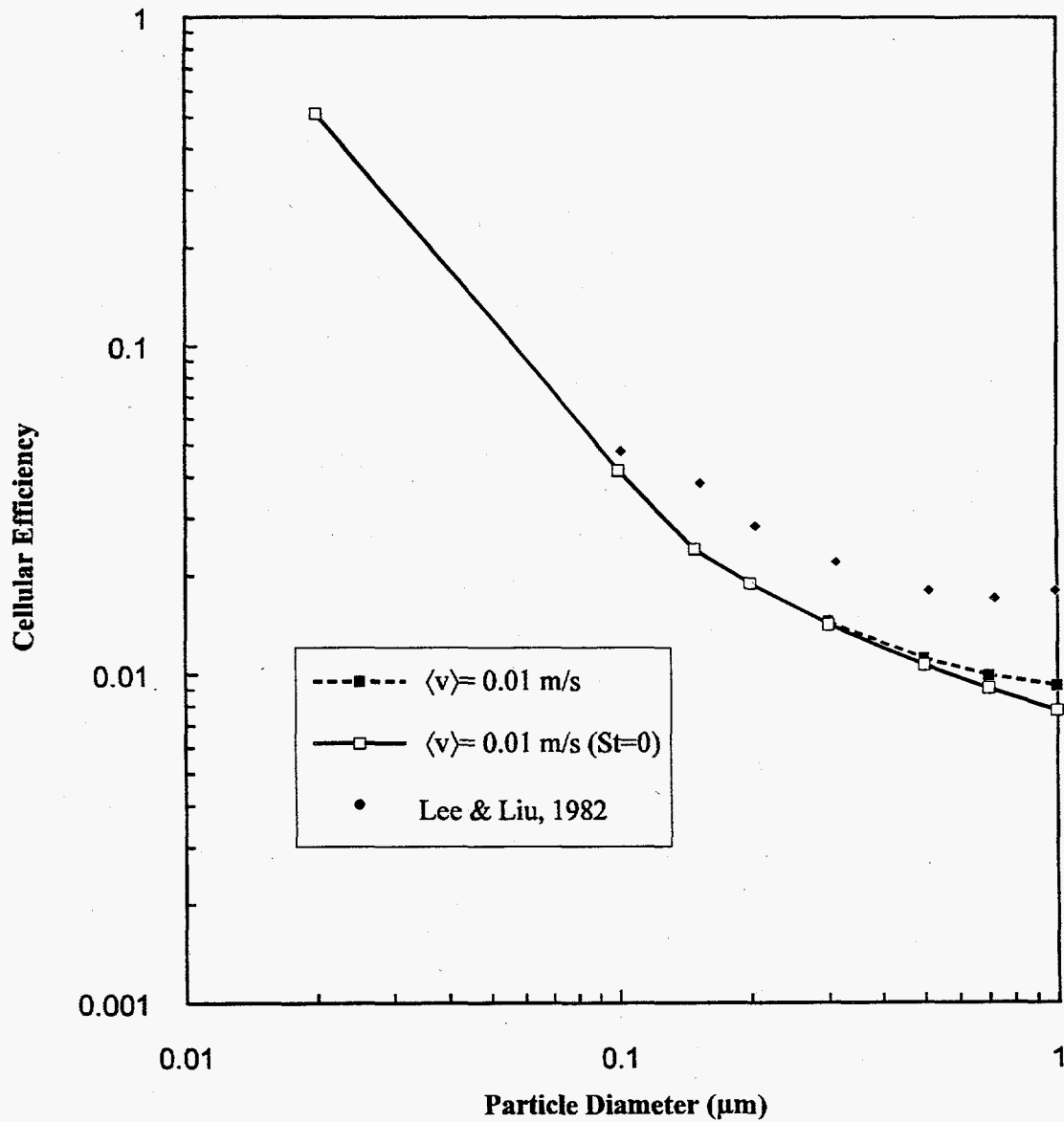
Figure 7
Comparison with Laboratory Experiments

$(2a = 11 \ \mu m, \ \rho_p = 1000 \ kg/m^3$
$T=278 \ K, \ \mu=1.83 \ 10^{-5} \ Pa \ s, \ \varepsilon_\gamma=0.849)$

for an average velocity given by $\langle v \rangle = 0.1 m/s$, and there one can see attractive agreement between theory and experiment for particle diameters that are equal to or smaller than the diameter of the most penetrating particle. For larger particles, the agreement diminishes and this would appear to confirm our suspicions concerning the importance of the higher order terms in Eq. 1.7. In Figure 6 the comparison between theory and experiment in shown for $\langle v \rangle = 0.03 m/s$ and we again see reasonable agreement for the smaller particles. The comparison for an even smaller velocity given by $\langle v \rangle = 0.01 m/s$ is shown in Figure 7 and here it becomes apparent that the theory is less reliable at lower velocities. We have no explanation for this observation; however, one must remember that the model illustrated in Figure 3 cannot possibly capture all the characteristics of a real filter and further studies using more complex unit cells are certainly in order. In addition, the influence of local heterogeneities must be determined and that is the objective of a subsequent study.

5. Conclusions

In this work we have used the first correction to the Smoluchowski equation to describe the effects of particle inertia, and the resulting particle transport equation has been used to develop a *local volume average transport equation* that includes the effects of non-traditional convective transport, dispersion, and particle capture. A spatially periodic model of a fibrous filter has been used, along with two closure problems, to calculated the effective coefficients that appear in the volume averaged transport equation. This leads to a direct calculation of the cellular efficiency and results were determined for a unit cell containing a single fiber that is orthogonal to the mean flow field. The results are in reasonably good agreement with experiments for the most penetrating particle size; however, higher order corrections need to be included in the Smoluchowski equation in order to predict the behavior of large particles.

6 Acknowledgment

7 Nomenclature

Roman Letters

a fiber radius, m.

$A_{\gamma\sigma}$ area of the γ-σ interface contained within the averaging volume, m^2.

$\mathbf{b}$ a closure variable that maps $\nabla\langle n_p \rangle^\gamma$ onto $\tilde{n}_p$, m.

d_p particle diameter, m.

$\mathbf{d}$ a velocity-like coefficient, m/s

D_p Brownian diffusivity, m²/s.

$\mathbf{D}^*$ dispersion tensor, m²/s

$\mathbf{F}_r(t)$ Brownian or random force, N.

$\mathbf{g}$ gravity vector, m/s²

k_{eff} effective rate coefficient for particle capture, s^{-1}

ℓ_γ characteristic length for a unit cell, m.

ℓ_σ characteristic length for the σ-phase ($= 2a$), m.

ℓ_i $i = 1, 2,$ and 3, lattice vectors, m.

L_ε characteristic length for the porosity, m.

L_n characteristic length for $\langle n_p \rangle^\gamma$, m.

L generic characteristic length for volume averaged quantities, m.

m_p mass of a particle, kg.

n_p particle density, number/m³.

$\tilde{n}_p$ spatial deviation of the particle of the particle, number/m³.

$\langle n_p \rangle^\gamma$ intrinsic average particle concentration, number/m³.

$\mathbf{n}_{\gamma\sigma}$ unit normal vector pointing from the γ-phase toward the σ-phase.

p fluid pressure, N/m².

$\langle p \rangle^\gamma$ intrinsic average pressure in the γ-phase, m³.

$\mathbf{r}$ position vector, m.

s a closure variable that maps $\langle n_p \rangle^\gamma$ onto $\tilde{n}_p$.

St $\langle v_x \rangle^\gamma \rho_p d_p^2 c_s / 18\mu\ell_\gamma$, the Stokes number.

t time, s.

$\mathbf{u}$ a velocity-like coefficient, m/s.

$\mathscr{V}$ averaging volume, m³.

V_γ volume of the γ-phase contained in the averaging volume, m³.

$\mathbf{v}$ fluid velocity vector, m/s.

$\langle \mathbf{v} \rangle^\gamma$ intrinsic average fluid velocity, m/s.

$\langle \mathbf{v} \rangle$ $\varepsilon_\gamma \langle \mathbf{v} \rangle^\gamma$, superficial average fluid velocity, m/s.

$\tilde{\mathbf{v}}$ $\mathbf{v} - \langle \mathbf{v} \rangle^\gamma$, spatial deviation fluid velocity, m/s.

$\mathbf{v}_p$ particle velocity, m/s.

$\bar{\mathbf{v}}_p$ deterministic particle velocity, m/s.

$\langle \mathbf{v}_p \rangle$ $\langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla\mathbf{v} \rangle$, inertia corrected volume average velocity for the particles, m/s.

$\mathbf{y}$ position vector relative to the centroid of the averaging volume, m.

$\mathbf{x}$ position vector locating the centroid of the averaging volume, m.

Greek Letters

μ fluid viscosity, Ns/m².

π 3.1416.......

ε_γ porosity.

γ $3\pi\mu d_p / m_p$, s^{-1}

ρ fluid density, kg/m^3.

ρ_p particle density, kg/m^3.

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