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DISPERSION/REACTION MODEL OF AEROSOL FILTRATION BY POROUS FILTERS

MICHAEL SHAPIRO and HOWARD BRENNER*

Faculty of Mechanical Engineering, Technion–Israel Institute of Technology, Haifa 32000, Israel
*Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, U.S.A.

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Abstract—Filter efficiency is rigorously calculated without *ad hoc* assumptions pertaining to aerosol distribution within the filter bed and even without the very concept of single-element efficiency. In particular, aerosol filtration processes are treated by formulating the particle transport problem at a pointwise (interstitial) level throughout the whole filter bed. These microscale processes ultimately govern aerosol transport and collection at the coarser Darcy scale. At the latter level of description the filter bed is viewed as a continuum in which aerosol propagation and deposition processes are characterized by three position-independent 'global' phenomenological coefficients: the mean aerosol velocity vector, dispersivity dyadic and mean volumetric aerosol deposition rate coefficient. Calculation of these three global aerosol coefficients is effected via a rigorous application of Taylor–Aris convective dispersion theory to a lattice model of a porous filter bed. The filter efficiency is easily and explicitly expressed in terms of these three transport coefficients, thereby completely eliminating evaluation of the single-element efficiency as an intermediate step in the calculations. Circumstances are outlined in which the coarse-scale aerosol diffusivity may be neglected and the concomitant aerosol collection rate uniquely characterized by Leer's filtration length parameter [Leers, R. (1957) *Staub* 17, 402.], relating filter bed thickness to total filtration efficiency. The scheme developed herein is illustrated by numerically computing the three Darcy-scale aerosol transport coefficients (via a finite element technique) for a fibrous filter subjected to typical filtration operating conditions. These coefficients are subsequently used to calculate the characteristic filtration length, whose values are then compared with existing theoretical results and available experimental data. The practical relevance of our theoretical development is outlined with respect to: (i) establishing the limitations of current filtration models; (ii) interpreting experimental data and (iii) optimal filter design.

NOMENCLATURE

a	characteristic collector size
C	Cunningham's correction factor
d_p	aerosol particle diameter
$\mathbf{D}, D$	Brownian diffusivity of aerosol particle
$\bar{\mathbf{D}}^*$	Darcy-scale dispersivity dyadic
$\bar{D}_f$	effective axial aerosol dispersivity
$\bar{\mathbf{D}}_{xx}, \bar{\mathbf{D}}_{qq}$	tensor components of the dispersivity $\bar{\mathbf{D}}^*$
E	dimensionless potential energy function
f	initial aerosol spatial concentration distribution
$\mathbf{F}$	external force
$\mathbf{i}_x$	unit vector in x direction
$\mathbf{J}$	aerosol flux vector operator
k	Boltzmann's constant or microscale reaction constant
$\bar{K}^*$	Darcy- or macroscale aerosol deposition rate coefficient
$\bar{K}_f$	axial aerosol deposition rate coefficient
l	characteristic microscale length
l_q	characteristic cross-sectional length
l_f	characteristic filtration length, $\bar{U}_f/\bar{K}_f$
$\boldsymbol{l}_i, (\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3,)$	unit basic lattice vectors
L	filter thickness
$\bar{L}$	dimensionless filter thickness, $L\,\bar{U}_f/\bar{D}_f$
m	integer number
$\mathbf{M}, M$	aerosol particle mobility
M_0	zero-order moment of aerosol concentration distribution
M_{x0}	zero-order axial moment of coarse-scale concentration
$\mathbf{n}=\{n_1, n_2, n_3\}, n_k$	positive or negative integer numbers, including zero
P	aerosol number density microscale concentration field
$\bar{P}$	aerosol number density macroscale concentration field
$\bar{P}_f$	cross-sectionally averaged macroscale aerosol concentration

$P_{\text{in}}, P_{\text{out}}$	aerosol concentrations at the filter inlet and outlet
Pe	microscale Peclet number, $2aU_o/D$
$\mathbf{q}_o$	filter cross-sectional domain
$\mathbf{q} \equiv (q_1, q_2)$	curvilinear coarse-scale coordinates within the filter cross-section
$\partial\mathbf{q}_o$	filter cross-sectional periphery
r_{p}	aerosol particle radius
$\mathbf{r}$	intracellular (local) position vector
$\mathbf{r}_{\text{f}}$	unit cell interstitial fluid domain
$\partial\mathbf{r}_k$	k^{th} unit cell face
$\mathbf{R}$	microscale position vector
$\mathbf{R}_{\mathbf{n}}$	global (discrete) position vector specifying the location of the **n**th unit-cell
$\bar{\mathbf{R}}$	macroscale position vector; mean displacement vector
$\overline{(\Delta\mathbf{R})^2}$	mean-square displacement dyadic
S	filter cross-sectional area
$S_{\text{c}}, S'_{\text{c}}$	actual and virtual bed particle (collector) surface
t	time
T	absolute temperature
$\mathbf{U}$	aerosol particle and fluid velocity vector
U_o	superficial filter velocity
$\bar{\mathbf{U}}^*$	mean Darcy- or macroscale aerosol velocity vector
$\bar{U}_{\text{f}}$	mean axial aerosol velocity
V	external force potential
$\mathscr{V}_\infty$	infinite (continuous space) domain
x	coarse-scale axial coordinate
$\bar{x}$	mean axial displacement
$\bar{x}_{\text{f}}$	dimensionless axial coordinate
α	constant, equation (1.3)
ε_{f}	dimensionless deposition rate parameter, $\bar{K}_{\text{f}}\bar{D}_{\text{f}}/\bar{U}_{\text{f}}^2$
η	filtration efficiency
η_{c}	unit bed (or single-element) efficiency
λ	Iwasaki's filter coefficient
λ_1, λ_2	constants, equations (6.5a, b)
μ	air viscosity

1. INTRODUCTION

This study concerns the transport and deposition of submicrometer aerosol particles in porous granular and fibrous filters. Collection of such particles is usually governed by diffusional, interceptional and gravitational deposition mechanisms. Existing filtration theory determines aerosol collection rates by solving the particle transport and deposition problem posed within a representative region of a porous bed, thereby evaluating the so-called unit-bed or single-element efficiency, η_{c}. The latter calculation of η_{c}, as well as its use in determination of the total filter efficiency, hinges upon several *ad hoc* assumptions pertaining to the aerosol distribution within the filter bed.

Aerosol particles are generally regarded as posing significant health hazards (Friedlander, 1977). A major source of air pollution arises via the discharge of various types of particulates into the atmosphere from industrial plants. Removal of dust particles from air streams prior to discharge constitutes an important engineering problem, one whose resolution lies at the focus of major scientific and engineering efforts (Davies, 1973; Dorman, 1973; Strauss, 1975).

Various techniques have been proposed for aerosol elimination. Electrostatic precipitators are among the most widely used air pollution control devices. However, they are very expensive, in addition to failing in the filtration of submicrometer and high-resistivity dusts (Strauss, 1975; Dietz, 1977). Space-charge precipitators and self-agglomerators are ineffective unless the pollutant to be removed is of extremely high density. Cyclones operate effectively for coarse dusts (≥ 10 μm), but prove ineffective in the removal of submicrometer and micrometer particles. Scrubbers, while effective in cleaning air, create auxiliary problems of water pollution (Hetsroni, 1982).

Submicrometer aerosol particles are ineffectively collected by conventional air pollution control devices. Such particles constitute primary health hazards when inhaled into upper and lower human respiratory tracts (Mercer, 1973; Melandri *et al.*, 1975). Filtration of submicrometer particles thus poses an important engineering challenge.

Both micrometer and submicrometer aerosols can be effectively collected by porous granular (Tardos *et al.*, 1978; Gutfinger and Tardos, 1979; Tien, 1989) and fibrous (Fuchs, 1964; Kirsch and Stechkina, 1978) filters. Such filters display high filtration efficiencies over a broad range of particle sizes, in addition to being characterized by low capital costs and operating expenses. These facts motivated the present study, which is devoted to developing the basic physical principles prerequisite to the rational design of porous filters capable of removing submicrometer particles from flowing gas streams.

A porous filter is depicted schematically in Fig. 1. Consider a dusty gas flowing within a cylindrical channel of arbitrary, but uniform cross-sectional shape. The filter is created by filling with a porous substance (sand, glass beads, cotton, wool, etc.) a region of length L lying between any two channel cross-sections. Dusty gas flowing within the filter bed interstices deposits aerosol particles onto the filter's collecting elements (collectors).

Filtration efficiency, η, is defined as

$$\eta = \frac{\text{mass of aerosol captured by the filter}}{\text{mass of aerosol entering the filter}}. \tag{1}$$

Alternatively, in terms of the aerosol concentrations P_{in} and P_{out} prevailing, respectively, at the filter inlet and outlet,

$$\eta = 1 - \frac{P_{\text{out}}}{P_{\text{in}}}. \tag{2}$$

This filtration efficiency depends upon a variety of operating parameters, the most important being the aerosol particle radius, r_p, characteristic collector size, a, superficial stream velocity, U_0 and filter thickness, L. The particle diameter $2r_p$ constitutes the primary factor governing the dominant mechanism of aerosol collection. Fine particles, those possessing diameters below 0.2 μm, are affected by Brownian motion. Accordingly, their diffusion to the collector's surface represents the dominant deposition mechanism. However, for larger particles, namely those possessing diameters between 0.2 and 2 μm, this diffusional mechanism is less effective in the removal process. Such particles are nevertheless insufficiently coarse to be significantly affected by inertial forces; (these forces provide an efficient deposition mechanism only for particle sizes above 2 μm). Consequently, porous filters—especially granular ones—are usually less effective in the collection of aerosols lying within the intermediate size range ($0.2\ \mu\text{m} \le 2r_p \le 2\ \mu\text{m}$). Calculation of the collection rate of such aerosols is usually effected by supplementing the diffusional particle deposition with

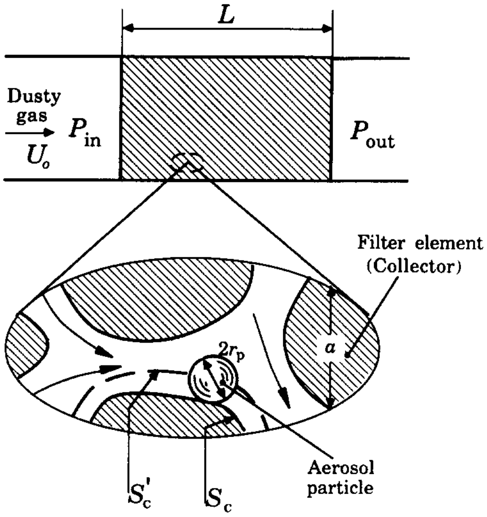

Fig. 1. Porous filter.

other, secondary collection mechanisms (Gutfinger and Tardos, 1979). Included among the latter is interception, namely the 'geometric' removal of particles moving along those streamlines whose minimum approach distance to the collector surface is less than the particle's radius. Other secondary deposition mechanisms include gravitational (Tardos *et al.*, 1979) and electrostatic (Pich, 1966; Davies, 1973; Ariman and Tang, 1976; Melcher *et al.*, 1977; Fan and Gentry, 1979; Tardos and Pfeffer, 1979; Tardos *et al.*, 1979; Henry and Ariman, 1981a, b; Shapiro, 1984; Shapiro and Laufer, 1984a, b; Shapiro *et al.*, 1983, 1986a–c, 1988a) forces.

Calculations of the filtration efficiencies of porous filters usually invoke several simplifying assumptions common to classical filtration theory (Pich, 1966, 1987; Löffler, 1971; Dorman, 1973). Therein, each individual filter element is assumed to manifest identical filtration behavior irrespective of bed position, allowing the cumulative aerosol collection rate for the filter bed as a whole to be obtained by merely summing the respective collection rates for each of the individual collectors composing the bed.

Classical filtration theory begins with a proper description of aerosol transport and deposition at the microscale, i.e. interstitial level, although only for a so-called 'single' representative unit-bed element (UBE) (Shapiro *et al.*, 1988a; Tien, 1989). In order to calculate the total aerosol collection rate for the entire set of UBEs, a set of simplifying assumptions is introduced pertaining to: (i) the microstructure of the individual collectors and bed, as a whole; (ii) the flow field and external force field prevailing at the microscale (i.e. within the UBE); (iii) the formulation of the aerosol microtransport problem within a single UBE; and (iv) the calculation of the total collection rate and filtration efficiency by transition from the single UBE results to that for the filter as a whole, i.e. the bed composed of a sequence of UBEs. It is, in essence, only in the last two—but nonetheless critical—steps that our approach departs from that of classical theory.

The essence and significance of these assumptions, as well as the scheme for calculating filtration efficiency according to existing theory, is schematically depicted in Fig. 2. Filter microstructure is characterized by specifying a UBE within which the detailed velocity and external force fields are calculated. The scheme ultimately calculates the aerosol collection rate achieved by such a single UBE. Towards this goal the aerosol microtransport problem is formulated within one UBE, while imposing an *ad hoc* boundary condition specifying a uniform inlet aerosol concentration at its upstream boundary. As a result, a so-called unit-bed efficiency,* η_c, (Tien, 1989) is calculated.

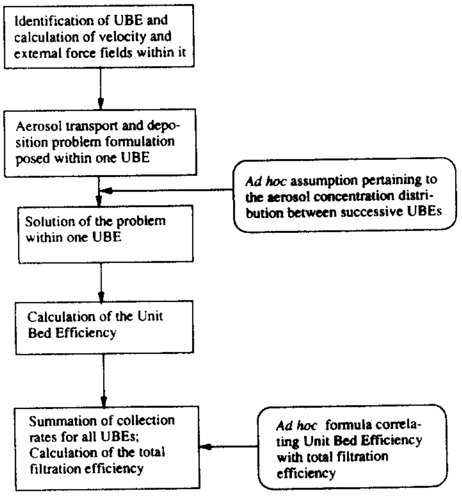

Fig. 2. *Ad hoc* scheme for computing the total filtration efficiency employed in existing filtration theory.

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* In cases where the UBE is composed of a single collector element, the unit-bed efficiency is termed the single-element efficiency (Davies, 1973; Tardos, 1977).

In order to cumulatively sum the aerosol collection rates over all collector elements composing the bed, and thus calculate the total filter efficiency, other *ad hoc* assumptions have been invoked (Tardos *et al.*, 1978; Shapiro *et al.*, 1986b) pertaining to the filter bed structure and spatial aerosol concentration distribution existing within the filter bed interstices. As a result, the following formula was proposed, correlating the total and single-element efficiencies:

$$\eta = 1 - \exp(-\alpha\eta_c L/a). \tag{3}$$

The exact nature of the non-dimensional porosity-dependent coefficient, α, is controversial (Dietz, 1977; Tardos, 1977). In many cases the choice of α was simply phenomenologically dictated so as to effect a better correlation of the theory with experimental data obtained for selected aerosols (Zahedi and Melcher, 1976, 1977; Zieve *et al.*, 1978). Other formulas correlating η with η_c have also been proposed in the literature (see e.g. Tien, 1989), each being based upon several *ad hoc* assumptions pertaining to the aerosol distribution existing within the filter bed.

In order to secure agreement between theoretical filtration efficiencies calculated from (3) and experimental data obtained for aerosols (covering a wide range of particle diameters), various geometric models of unit-bed elements have been proposed. Among these are capillaries (Jackson and Calvert, 1966), constricted tubes (Payatakes and Neira, 1977) and sphere- (or circular cylinder-) in-cell (Happel, 1958; Kuwabara, 1959; Neale and Nader, 1974) models, each characterized by its own distinctive local velocity field and external force field prevailing within the UBE.

More realistic models of porous filter beds consist of periodic arrays of spheres and cylinders, within which microvelocity fields have been calculated for several spatially periodic geometries (Snyder and Stewart, 1966; Sørensen and Stewart, 1974; Sangani and Acrivos, 1982a, b; Zick and Homsy, 1982). For such periodic models of porous filters the UBE is identified with the elementary unit-cell of the spatially periodic array. Aerosol collection in spatially periodic models of porous filters has been considered by Gal *et al.* (1985) for granular and by Kao *et al.* (1986) for fibrous filters. In spite of the apparently realistic representative velocity field calculated for such models, subsequent calculations of filtration efficiency performed during each of these two studies are nevertheless still described by the *ad hoc* scheme depicted in Fig. 2. Indeed, the aerosol collection problem was formulated within the single UBE, ultimately resulting in calculations of the unit-bed efficiency, which—upon using equation (3)—was correlated with the total filtration efficiency.

Existing *ad hoc* filtration models, described by the scheme shown in Fig. 2, suffer from their inability to theoretically substantiate the fundamental assumptions underlying calculations of unit-bed (and, subsequently, total) filtration efficiences. In fact, the efforts of Gal *et al.* (1985) and Kao *et al.* (1986) to employ more realistic spatially periodic velocity field and external force field models in calculating the unit-bed efficiency are meaningless and inconsistent with the very concept of the latter quantity, as in all cases their aerosol collection problem within the UBE was formulated on an *ad hoc* basis.

A precise definition of unit-bed efficiency may be unambiguously assigned only to a truly isolated collector, i.e. one embedded in an infinitely-extended, otherwise undistributed flow field (and/or external force field) possessing a spatially uniform aerosol concentration at the far upstream ('inlet') location. In our opinion this concept is inapplicable to dense porous filters, being physically inconsistent. Rather, it is only the total filter efficiency that possesses physical reality, as it is only this global quantity that can be experimentally measured.

The present contribution describes a novel approach to the problem of diffusion-controlled aerosol filtration processes in porous filters. While utilizing the conventional microscale description of aerosol transport and deposition processes accepted in classical filtration theory, it explicitly avoids *ad hoc* summation of the respective collection rates characterizing each individual UBE; it does not even utilize the concept of unit-bed efficiency. Rather than formulate the microscale aerosol filtration problem within the one representative UBE, the present theory begins with a comparable description of the aerosol transport and collection processes within the whole porous filter bed. Accordingly, instead of the artificial (indeed, physically incorrect) boundary conditions imposed upon the aerosol concentration field between neighboring UBEs (as currently employed in the existing theory), our more physically realistic model imposes conditions of continuity of aerosol concentration and flux across adjacent faces separating contiguous UBEs—these being the unit-cells of the spatially periodic filter bed model.

The proposed scheme ultimately describes the filter bed at a Darcy, rather than interstitial scale; as such, the bed is viewed at the former level of description as being a homogeneous convective-diffusive-'reactive' continuum, possessing uniform (i.e. position-independent) phenomenological transport coefficients. [The first-order, irreversible, chemical 'reaction'-like behavior at the macroscale arises from the permanent removal of the aerosol particles from the interstitial (microscale) fluid by deposition onto the filter bed collector elements.] These macrotransport coefficients consist, respectively, of the Darcy-scale mean velocity vector, $\bar{\mathbf{U}}^*$, dispersion dyadic, $\bar{\mathbf{D}}^*$, and scalar aerosol volumetric deposition rate coefficient, $\bar{K}^*$. They are demonstrated to be independent of the filter macroscale geometry and can be rigorously calculated by employing generalized Taylor dispersion phenomena theory (Brenner, 1980a; 1982a). Recently, techniques for analysing the flow, dispersion and 'disappearance' of inert (Brenner, 1980b) and chemically reactive (Shapiro and Brenner, 1988) solute species in spatially periodic models of porous media have been proposed. In the present study we exploit these models to determine aerosol collection rates governed by combined diffusion, interception and external-force (gravitational, electrostatic, magnetic, etc.) filtration mechanisms prevailing within porous filters.

The sequential process of calculating the total filtration efficiency via the proposed model is described schematically in Fig. 3. After a precise physico-mathematical formulation of the aerosol microtransport and deposition processes in the entire spatially periodic (model of the) filter bed, the three Darcy-scale transport coefficients are explicitly calculated (while introducing no *ad hoc* assumptions, such as is done in the classical theory). These Darcy-scale coefficients are subsequently used to describe the gross, coarse-scale filtration process at a continuum level. Solution of the coarse-scale transport and reaction problem thereby posed, immediately yields the total filtration efficiency [without utilizing *ad hoc* formulas of the type represented by equation (3)].

The requisite macro- or Darcy-scale transport and 'reaction' coefficients required above are ultimately calculated from a rigorous formulation and analysis of the physico-mathematical steady-state problem of aerosol transport and deposition within a single unit-cell of the bed. However, in contrast with the *ad hoc* formulation posed by the comparable aerosol

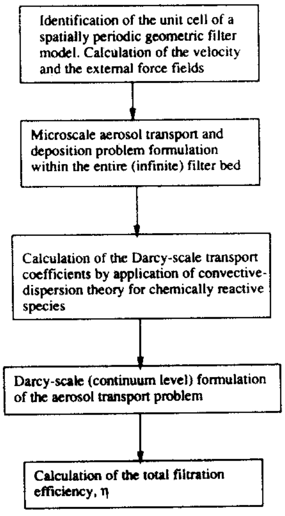

Fig. 3. Calculation of filtration efficiency by application of the proposed dispersion/reaction model.

transport problem within the single UBE (employed in the classical theory), the present unit-cell formulation derives rigorously from the general aerosol transport problem posed over the whole filter bed. Methodologically, our unit-cell analyses parallel existing computational schemes for determining the unit-bed efficiency, η_c, within the framework of classical filtration theory. In fact, our unit-cell analyses preserve the same aerosol collection mechanisms underlying the latter well-studied problems. However, in contrast with such previous studies, the present more fundamental approach for determining the filtration efficiency, η, is free of the artificial decomposition of the problem into sequentially: (i) establishing the unit-bed efficiency; and (ii) subsequently summing the individual collection rates. In our scheme, these two artificially distinct steps appear more naturally in combination as the single inseparable physical process that they indeed are in nature.

Practical applications of the proposed model abound. Its use exposes the conceptual limitations implicit in current models of diffusion-controlled aerosol filtration processes in granular bed and fibrous filters. Specifically, effects arising from collector packing arrangement, bed porosity and other filtration parameters may be systematically investigated. Moreover, the validity of fundamental assumptions underlying classical filtration theory [such as 'sticky' collector surfaces and the concomitant absence of particle re-entrainment (Tardos, 1977)] may also be scrutinized by comparison of the predictions of such theories with experimental data.

2. MACROSCALE DESCRIPTION OF THE FILTRATION PROCESS

This section addresses the microscale transport and deposition of aerosol particles within the filter bed, viewed at the interstitial continuum length scale, comparable to (or less than) the characteristic linear dimension 'a' of a collector element. This geometrical/kinematical description will be effected in its most general form, without explicitly assigning any *a priori* lattice/particle configuration to the filter bed. Explicitly, in this section we re-examine the equations governing the aerosol particles' motion in the filter interstices, as well as their collection on bed elements, in accordance with classical filtration theory. The main postulates and assumptions of that theory at this microscale level of description are also adopted in the dispersion model proposed in the present study.

Consider identical, non-interacting, spherical aerosol particles of radii r_p ($r_p \ll a$) moving within the interstices of a porous filter (Fig. 1). Here, we consider inertialess Brownian aerosol particles whose stochastic motion is usually described by a Eulerian equation governing the microscale aerosol concentration P (Levich, 1962; Fuchs, 1964). Motion of larger (normally micrometer sized) particles is affected by inertial forces, neglected here. If deemed relevant, this force may also be incorporated into the model as an external force exerted upon the suspended particles (Fernandez de la Mora and Rosner, 1982).

It will henceforth be supposed that all modes of interaction between neighboring aerosol particles (hydrodynamic, electric, magnetic, etc.) are negligible, so that each is effectively transported through the filter bed interstices as an isolated (Brownian) particle, independently of the presence of the others. Moreover, any cumulative effects of the aerosol cloud upon either the flow field or external force field existing within the filter are neglected. These assumptions (usually accepted in the filtration theory) are justified when the aerosol concentration P is everywhere small.

With these hypotheses the microscale conservation equation governing the temporal evolution of the aerosol particles' number density, $P(\mathbf{R}, t)$, within the interstitial fluid region may be written as

$$\frac{\partial P}{\partial t} + \nabla \cdot (\mathbf{J}P) = 0. \tag{4}$$

Here, with $\mathbf{J}$ an operator, $\mathbf{J}P$ is the single-particle aerosol flux density vector, given generally for the dilute systems to which our analysis is valid by the constitutive relation (Bird *et al.*, 1960)

$$\mathbf{J}P = (\mathbf{U} + \mathbf{M}\cdot\mathbf{F} - \mathbf{D}\cdot\nabla)P. \tag{5a}$$

In general, due to wall effects arising from the aerosol particle's proximity to the collector surface, the (force-free) particle velocity vector $\mathbf{U}(\mathbf{R})$ at point $\mathbf{R}$ will differ from the undisturbed fluid velocity vector that would prevail in the absence of the particle. For this same reason, the respective spherical particle mobility, $\mathbf{M}$, and molecular diffusivity, $\mathbf{D}$, [which are related through the Stokes–Einstein equation, cf. (6)] will generally each be functions of $\mathbf{R}$. Moreover, again by virtue of wall effects, they will generally be dyadics (Brenner and Gaydos, 1977) rather than scalars, reflecting the generally anisotropic nature of these quantities, even for spherical aerosol particles (Hirschfeld *et al.*, 1984; Falade and Brenner, 1985, 1988). However, the assumption that $r_p/a \ll 1$ permits us to neglect such wall proximity effects provided that, simultaneously, the aerosol particle is not too close to the wall compared with its radius r_p (Falade and Brenner, 1985, 1988). (Though the latter may appear to preclude the actual physical deposition of a particle onto the collector surface—which is, in fact, the primary subject of interest here—the deposition process will eventually be simulated [cf. equations (9) and (16)] by invoking the irreversible adsorption, $P = 0$, boundary condition on the collector surface.) Accordingly, at the level of description of the transport process at which this (mesoscale) boundary condition applies, the constitutive equation (5a) may be replaced by

$$\mathbf{J}P = \mathbf{U}P + M\mathbf{F}P - D\nabla P, \tag{5b}$$

in which $\mathbf{U}(\mathbf{R})$ is now identical to the fluid velocity, and the scalars M and D are $\mathbf{R}$-independent constants, interrelated through the Stokes–Einstein equation

$$D = kTM, \tag{6}$$

in which kT is Boltzmann's factor. The aerosol particle mobility is given by the expression

$$M = C/6\pi\mu r_p, \tag{7}$$

where μ is the viscosity of the suspending gas and C is Cunningham's slip-correction factor (Davies, 1973). For simplicity the system is regarded as isothermal, so that aerosol transport by thermophoresis (Hidy and Brock, 1970; Batchelor and Shen, 1985) is assumed absent.

In cases where the external force, $\mathbf{F}$, appearing in equation (5b) is derivable from a potential energy function $V(\mathbf{R})$ (i.e. $\mathbf{F} = -\nabla V$), equation (5b) may be rewritten as

$$\mathbf{J}P = \mathbf{U}P - De^{-E(\mathbf{R})}\nabla(e^{E(\mathbf{R})}P), \tag{8}$$

with $E(\mathbf{R}) \equiv V(\mathbf{R})/kT$, a non-dimensional potential.

At the same microscale level of description that allows equation (5b) to appear in place of (5a), so too does the external force field $\mathbf{F} \equiv \mathbf{F}(\mathbf{R})$ appearing in (5b) differ from that in (5a). In particular, (5b) incorporates only those 'long-range' forces (gravitational, electrostatic, magnetic, etc.) exerted upon the aerosol particle when the distance of its center from the collecter surface is 'large' compared with its radius r_p. Present in (5a), but excluded from (5b) [and consequently from (8)], are the 'short-range', so-called dispersion forces (Mahanty and Ninham, 1976) exerted on the particle by the collector when the separation distance between them is comparable to, or less than the aerosol particle size r_p. It is these forces that are important in causing the particle's ultimate attachment to the collector surface (Fichman and Pnueli, 1985), but they are regarded as non-existent in describing the aerosol transport* at the coarser level of description of equation (5b) (Shapiro *et al.*, 1990a).

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* These forces may, however, be important in the collection of fine particles from liquids (Prieve and Ruckenstein, 1974), corresponding to hydrosols rather than aerosols.

Though excluded from consideration in the latter, these forces are, nevertheless, implicitly included in the boundary condition imposed upon P at the collector surfaces S_c. Formulation of this condition requires several further hypotheses, usually accepted in classical filtration theory. Explicitly, it is assumed (Löffler, 1971; Tardos, 1977) that when the center of an aerosol particle approaches the collector's surface to within a distance comparable to its radius r_p, it collides with (and permanently adheres to) the collector—never to be re-entrained. This permanent attachment arises (Friedlander, 1977) from the action of several dispersion forces, primarily of the van der Waals type. In the case of electrically charged aerosols, this attachment is facilitated by the electrostatic image force (Shapiro *et al.*, 1983). Such situations are modelled by the assumption of zero aerosol concentration on the collector surface (Levich, 1962; Gupalo *et al.*, 1985):

$$P = 0 \quad \text{on } S_c. \tag{9a}$$

For the submicrometer aerosol particles here considered, this boundary condition has been rigorously substantiated by Shapiro *et al.* (1989) by adapting the matched asymptotic expansion scheme (Van Dyke, 1975) of Brenner and Leal (1982) to such interfacial problems. This condition is sometimes reformulated (Ruckenstein and Prieve, 1973; Tardos, 1977) as applying instead on the hypothetical or virtual surface S'_c, situated at a distance r_p beyond S_c:

$$P = 0 \quad \text{on } S'_c. \tag{9b}$$

The latter boundary condition presumably takes account of the effect of interception upon the diffusionally-controlled aerosol deposition process, a phenomenon whose quantification will be separately addressed in a later publication.*

The preceding assumption of a 'sticky' collector surface, from which surface the possibility of re-entrainment is excluded and from which assumption derives boundary condition (9a) or (9b), is, in fact, strictly valid only for a 'clean' filter where the collector shapes and sizes have not been significantly altered by the cumulative aerosol deposition process (Payatakes and Gradon, 1980; Tien, 1989). For the dilute aerosols here assumed, such contamination of the collector surfaces occurs relatively slowly over time, thus justifying continued use of the 'bare' surface boundary condition (9a) or (9b).

3. DARCY-SCALE DESCRIPTION OF THE FILTRATION PROCESS

3.1. *Darcy-scale aerosol transport coefficients*

In principle, when the microscale aerosol concentration P is known everywhere within the filter's interstitial region, the filtration efficiency may be determined by integrating the aerosol collection rate (i.e. the flux $\mathbf{n}\cdot\mathbf{J}P$, where $\mathbf{n}$ is the unit normal vector on S_c or S'_c) over all of the collecting surfaces contained within the filter. However, the prior computation of P at each point $\mathbf{R}$ within the filter bed constitutes a formidable task, even if the geometric structure and packing arrangement of the porous filter are completely known, which in practice they generally are not. Even if such pointwise aerosol concentration distribution data, P, were indeed available, such exhaustively detailed information is generally unnecessary to determine gross parameters such as filtration efficiency. A more fruitful approach, pursued here, seeks to effect only a coarse-scale description of the aerosol convection, diffusion and deposition processes within the filter bed. Nevertheless, as will be shown, this

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* An alternative and more general formulation of the boundary condition on the collector surface arises upon viewing the aerosol particles as a species undergoing a first-order irreversible surface reaction with reactivity k (Shapiro and Brenner, 1988; Dungan *et al.*, 1990); explicitly, $\mathbf{n}\cdot\mathbf{J}P = kP$ on S_c (or S'_c), with $\mathbf{n}$ the unit outer normal vector on S_c (or S'_c). Our analysis can easily be extended to encompass this more general situation. However, in the interest of maintaining simplicity in this introductory paper, we prefer using boundary conditions of the type (9a) [or (9b)].

less detailed scheme suffices to establish the filtration efficiency. Implementation of this scheme is based upon the following model problem.

Suppose that clean (particle-free) air flows through the filter. At time $t = 0$ let a cloud of aerosol particles be instantaneously introduced into some interstitial region of the filter bed. Each particle will be transported through the interstices by diffusion and convection, with some being deposited on the collector surfaces. We will not focus on the pointwise aerosol concentration, P, *per se*, but rather will attempt to characterize the aggregate transport of the aerosol cloud as a whole by considering the temporal evolution of the following three moments of this distribution:

$$M_0 = \int_{\mathscr{V}_\infty} P(\mathbf{R}, t) \mathrm{d}^3\mathbf{R}, \tag{10a}$$

$$\bar{\mathbf{R}} = \frac{1}{M_0} \int_{\mathscr{V}_\infty} \mathbf{R} P(\mathbf{R}, t) \mathrm{d}^3\mathbf{R}, \tag{10b}$$

$$\overline{(\Delta \mathbf{R})^2} = \frac{1}{M_0} \int_{\mathscr{V}_\infty} (\mathbf{R} - \bar{\mathbf{R}})^2 P(\mathbf{R}, t) \mathrm{d}^3\mathbf{R}, \tag{10c}$$

with $\mathrm{d}^3\mathbf{R}$ a microscale volume element. Each of the above integrations is to be performed over the effectively infinite interstitial region $\mathscr{V}_\infty$ of the entire bed.

$M_0(t)$ represents the 0th moment of the concentration distribution, namely the total number of aerosol particles remaining within the interstitial filter bed domain at time t.† $\bar{\mathbf{R}}$ is the displacement at time t of the aerosol cloud's center from its original position at $t = 0$, whereas the dyadic $\overline{(\Delta \mathbf{R})^2}$ represents the mean square displacement of the aerosol cloud about this moving center. Because, on average, P diminishes with time owing to the removal of some aerosol particles from the gas stream by the collectors (i.e. M_0 is not conserved), $\bar{\mathbf{R}}$ and $\overline{(\Delta \mathbf{R})^2}$ are defined above, not in an absolute sense (which would only be appropriate in the absence of deposition), but rather in a normalized sense with respect to the total number M_0 of residual aerosol particles remaining within the filter at time t. This normalization is effected by the appearance of M_0 in the respective denominators of equations (10b, c).

The three global quantities represented by the trio of equations (10a–c) depend upon the time, t, as well as in general upon the initial microscale spatial distribution of aerosol particles (the 'cloud') within the interstitial region. We seek to investigate those circumstances for which the corresponding trio of phenomenological coefficients defined as (Brenner and Gaydos, 1977; Brenner, 1980b; Shapiro and Brenner, 1988):

$$\bar{K}^* = -\lim_{t \to \infty} \frac{\mathrm{d}}{\mathrm{d}t} \ln M_0, \tag{11a}$$

$$\bar{\mathbf{U}}^* = \lim_{t \to \infty} \frac{\mathrm{d}\bar{\mathbf{R}}}{\mathrm{d}t}, \tag{11b}$$

$$\bar{\mathbf{D}}^* = \frac{1}{2} \lim_{t \to \infty} \frac{\mathrm{d}}{\mathrm{d}t} \overline{(\Delta \mathbf{R})^2}, \tag{11c}$$

not only exist, but are also independent of the initial aerosol distribution. When such circumstances obtain, these coefficients constitute macroscale phenomenological parameters, characterizing the mean transport of the aerosol cloud as a whole.

Explicitly, $\bar{K}^*$ asymptotically describes the long-time rate of diminution of the total number of aerosol particles present in the cloud; $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$, respectively, represent the velocity vector of the aerosol cloud's number center and its dispersivity dyadic. When $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ prove to be independent of the initial spatial distribution of aerosol particles

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† Particles deposited on the collector surfaces are, of course, regarded as having been permanently removed from the interstitial region $\mathscr{V}_\infty$, so that M_0 necessarily diminishes monotonically with time.

within the filter—in particular, of the initial number of such particles introduced—these coefficients then constitute fundamental phenomenological properties of the aerosol transport and deposition processes described at the macro- or Darcy-scale. At this scale of description the filter may be regarded as a continuum within which the aerosol is transported at a mean velocity, $\bar{\mathbf{U}}^*$, dispersed about this mean velocity at a rate governed by the aerosol dispersivity, $\bar{\mathbf{D}}^*$, and permanently 'removed' from this effective continuum at a rate governed by the (specific) 'reactivity' $\bar{K}^*$ of the volumetrically homogeneously distributed aerosol 'sinks'.

3.2. *Aerosol filtration process viewed at the coarse scale*

As a consequence of the above facts, the 'Lagrangian' constants $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ may be interpreted (Shapiro and Brenner, 1988) as phenomenological transport coefficients appearing in the following Eulerian convection-diffusion-reaction equation governing the transport and temporal evolution of an averaged or mean Darcy-scale, aerosol number density concentration, $\bar{P}$, (yet to be precisely defined):

$$\frac{\partial \bar{P}}{\partial t} + \bar{\mathbf{U}}^* \cdot \bar{\nabla} \bar{P} - \bar{\mathbf{D}}^* : \bar{\nabla}\bar{\nabla}\bar{P} + \bar{K}^* \bar{P} = 0. \tag{12}$$

Herein, $\bar{\nabla} \equiv \partial/\partial\bar{\mathbf{R}}$ represents the gradient with respect to the coarse-scale (i.e. continuum-scale) position vector, $\bar{\mathbf{R}}$, for which the magnitude of the 'differential' displacement vector $\Delta\bar{\mathbf{R}}$ satisfies the inequality $|\Delta\bar{\mathbf{R}}| \geq a$; $\bar{P}$ is correlated with the microscale aerosol number density concentration, P, by averaging of the latter over a representative volume within the filter bed, the linear dimension of which volume exceeds the mean collector size a. The precise definitions of both $\bar{P}$ and $\bar{\mathbf{R}}$, as well as the precise physico-mathematical meaning of the phrase 'asymptotically long-time' introduced in (11), remain uncertain in the absence of any quantification of the filter's geometrical structure. These issues will be resolved in the next section, at least for a spatially periodic geometric model of a filter bed.

It is noteworthy that, whereas at a microscale level of description the aerosol may be viewed as a species undergoing surface reaction at an effectively infinite rate upon the individual collector surfaces, at the Darcy-scale level the aerosol appears rather as a species undergoing a volumetric reaction at a finite rate (characterized by the bulk reactivity coefficient $\bar{K}^*$).

Once the three phenomenological coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ are known, calculation of the filtration efficiency may be effected by solving a steady-state version of equation (12) (cf. Section 6). Calculation of this filtration efficiency thus effectively reduces to determining these three coefficients for given aerosol physical properties and a prescribed microscale filter bed configuration. The problem thereby posed is solved in the next section for a spatially periodic model of the filter bed.

4. THEORETICAL CALCULATION OF THE DARCY-SCALE AEROSOL DEPOSITION AND TRANSPORT COEFFICIENTS $\bar{K}^*$, $\bar{\mathbf{U}}^*$ AND $\bar{\mathbf{D}}^*$ FOR A LATTICE MODEL OF A POROUS FILTER

The fundamental phenomenological coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ represent the asymptotic Darcy-scale manifestation of the microscale aerosol transport and deposition processes occurring in the filter bed interstices. Accordingly, their calculation devolves upon knowledge of both the filter's microscale geometry and the aerosol's microtransport phenomenological coefficients appearing in the interstitial microtransport equation. In addition to the aerosol's molecular diffusion coefficient, D, and mobility, M, (which in most cases of interest may be regarded as known constants), knowledge of the fluid velocity field and external force field exerted on the aerosol particles within the filters is also required. These depend *inter alia* upon the collector configuration and packing arrangement. Substantial conceptual difficulties arise when attempting to determine $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ without any *a priori* assumptions (Crapiste *et al.*, 1986) regarding the geometric structure of the bed.

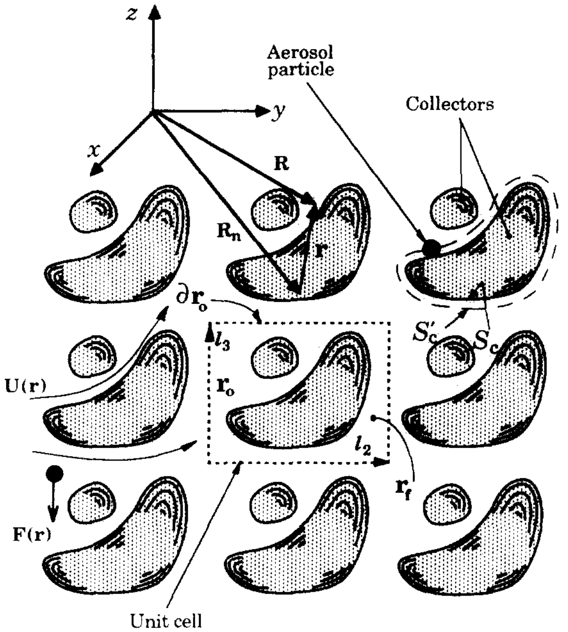

Fig. 4. Spatially periodic porous filter. Microscale view.

4.1. *Spatially periodic filter model*

Without assuming any specific collector shape(s) or sizes, it will nevertheless be supposed that the filter bed possesses a spatially periodic or lattice structure[*] (Brillouin, 1953; Brenner, 1980b). Equivalently, the filter may be represented by an arbitrarily chosen unit-cell (Fig. 4), indefinitely repeated along the directions of the three basic lattice vectors $\mathbf{l}_i\,(i = 1, 2, 3)$. Within each unit-cell the number of collector elements, as well as their shapes and sizes, may be selected to most closely approximate the disordered microstructure of the real filter bed of interest.

Due to the property of spatial periodicity, a general position vector $\mathbf{R}$ may be decomposed into a respective sum of local, bounded ($\mathbf{r}$) and global, unbounded ($\mathbf{R}_n$) position vectors, in accordance with the relation

$$\mathbf{R} = \mathbf{R}_n + \mathbf{r}. \tag{13}$$

Here, the discontinuous global position vector, $\mathbf{R}_n$, locating the $\mathbf{n}$th unit-cell in the lattice is related to the trio of basic lattice vectors by the expression

$$\mathbf{R}_n = n_1 l_1 + n_2 l_2 + n_3 l_3, \quad (n_k = 0, \pm 1, \pm 2, \pm 3, \ldots; k = 1, 2, 3). \tag{14}$$

Owing to the assumed spatially periodic representation of the filter bed geometry, the microscale external force field, $\mathbf{F}(\mathbf{R})$, may also be assumed to be a spatially periodic field, and hence to be a function, $\mathbf{F}(\mathbf{r})$, only of the local position vector, $\mathbf{r}$, independent of $\mathbf{n}$. Moreover, in circumstances where the interstitial air flow is effectively incompressible and the unidirectional macroscale fluid velocity vector homogeneous (i.e. constant $\mathbf{R} \equiv \mathbf{R}_n$), we may

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[*] Although aerosol filtration in porous filters possessing spatially periodic structures has been considered in several prior studies (Havlicek, 1961; Davies, 1973; Gal *et al.*, 1985; Kao *et al.*, 1986), granular and fibrous filters are not spatially periodic. This fact does not, however, exclude use of the proposed model for analysing practically important (i.e. disordered) systems. Spatially periodic models of porous beds constitute a more faithful physical representation of real filter structures than do commonly used cellular (spherical or cylindrical) models (Davies, 1973; Tardos *et al.*, 1978; Shapiro *et al.*, 1986b, 1988a). The proposed filter configuration is understood to be a simple model of the actual geometric mode of arrangement of the collectors comprising the filter bed; this bed may, in fact, be disordered.

also suppose $\mathbf{U}(\mathbf{R})$ to be spatially periodic, and hence of the functional form $\mathbf{U}(\mathbf{r})$. On the other hand, the aerosol concentration, P, is not a spatially periodic function and hence is viewed as being of the functional form $P = P(\mathbf{R}_{\mathbf{n}}, \mathbf{r}, t)$.

The interstitial velocity, $\mathbf{U}(\mathbf{r})$, may, in principle, be calculated (albeit numerically, rather than analytically) for a specified collector geometry, filter packing arrangement and prescribed mean Darcy-scale flow rate (filter-face velocity) or mean Darcy-scale pressure gradient (pressure drop across the filter). Indeed, results for macroscopically uniform flows through simple arrays of spheres (Sørensen and Stewart, 1974; Sangani and Acrivos, 1982a; Zick and Homsy, 1982) and cylinders (Sangani and Acrivos, 1982b; Larson and Higdon, 1986; Edwards *et al.*, 1988) are already available. Moreover, when the external force experienced by the particle is caused by an externally applied electrostatic field, its potential E may be obtained by solving Laplace's equation for the spatially periodic particle array (Jackson, 1971).

With the microscale fields, $\mathbf{U}(\mathbf{r})$, $\mathbf{F}(\mathbf{r})$, explicitly available and aerosol phenomenological coefficients, D and M, respectively, given by equations (6) and (7), the propagation and deposition of an aerosol cloud through the filter bed of a specified spatially periodic geometry is governed by equation (4), which, in the present circumstances, adopts the form

$$\frac{\partial P}{\partial t} + \nabla \cdot (\mathbf{J}P) = 0 \qquad (\mathbf{r} \in \mathbf{r}_f\{\mathbf{n}\}, t \geq 0), \tag{15}$$

where the aerosol flux density, $\mathbf{J}P$, is given by equation (5b); here, $\nabla \equiv \partial/\partial\mathbf{r}$ denotes the intracell gradient operator.

Boundary condition (9a) may be rewritten in the intracellular form

$$P = 0 \qquad \text{for } \mathbf{r} \in S_c\{\mathbf{n}\}. \tag{16}$$

Here, $\mathbf{r}_f\{\mathbf{n}\}$ and $S_c\{\mathbf{n}\}$, respectively, represent the interstitial (fluid) space and the collector surface contained within the $\mathbf{n}$th unit-cell.

Continuity conditions (Brenner, 1980b)

$$P(\mathbf{R}_{\mathbf{n}} - \boldsymbol{l}_k, \mathbf{r} + \boldsymbol{l}_k, t) = P(\mathbf{R}_{\mathbf{n}}, \mathbf{r}, t) \qquad (\mathbf{r} \in \partial\mathbf{r}_k), \tag{17a}$$

$$\nabla P(\mathbf{R}_{\mathbf{n}} - \boldsymbol{l}_k, \mathbf{r} + \boldsymbol{l}_k, t) = \nabla P(\mathbf{R}_{\mathbf{n}}, \mathbf{r}, t) \qquad (\mathbf{r} \in \partial\mathbf{r}_k), \tag{17b}$$

are imposed across the superficial (generally curvilinear) unit-cell surfaces $\partial\mathbf{r}_k$ ($k = 1, 2, 3$).

Boundary conditions at infinity,

$$|\mathbf{R}_{\mathbf{n}}|^m (P, \mathbf{J}P) \to (0, \mathbf{0}) \quad \text{as} \quad |\mathbf{R}_{\mathbf{n}}| \to \infty \qquad (m = 0, 1, 2, \ldots), \tag{18}$$

assure sufficiently rapid decay of the aerosol density at distant points.

Formulation of the microscale problem defining P is completed by imposing the initial condition

$$P(\mathbf{R}_{\mathbf{n}}, \mathbf{r}, 0) = f(\mathbf{R}_{\mathbf{n}}, \mathbf{r}) \tag{19}$$

at time $t = 0$, with $f(\mathbf{R}_{\mathbf{n}}, \mathbf{r})$ the initial aerosol distribution within the filter bed.

In contrast with the existing approach, where aerosol transport is considered only within the single UBE, equations (15)–(19) describe exactly the real physical process of propagation and deposition of the aerosol particles throughout the whole (spatially periodic) filter bed. As a well-posed mathematical problem, equations (15)–(19) may (at least in principle) be explicitly solved. This solution may be directly and rigorously used [without the necessity of utilizing the *ad hoc* formula (3)] to calculate the filter efficiency. However, as previously discussed, for the latter goal such an exhaustively detailed microscale solution P is unnecessary. Accordingly, we seek instead to obtain only the coarse-grained macroscale aerosol concentration field $\bar{P}(\bar{\mathbf{R}}, t)$, governed by the transport and reaction/deposition equation (12). Towards this goal we will employ the method of moments (Aris, 1956; Brenner, 1980b) to effect the calculation of the three phenomenological coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ required in this macrotransport equation, other than via their fundamental definitions (10)–(11), which require *a priori* knowledge of the detailed distribution, P. These calculations are performed in Appendix A. The computational effort required necessitates calculation of only the leading eigensolutions (Φ_0, Ψ_0) and λ_0 of the pair of adjoint eigenvalue problems (A11)–(A12), together with a determination of the time-independent vector **B**-field, governed by equations (A17a–d)—all within but a single unit-cell of the infinitely extended porous filter bed.

4.2. *Darcy-scale aerosol concentration distribution*

The average or Darcy-scale aerosol concentration, $\bar{P}$, is exactly defined for our spatially periodic model via integration of the microscale concentration field, P, over the interstitial unit-cell volume:

$$\bar{P}(\mathbf{R}_n, t) \overset{\text{def.}}{=} \frac{1}{\tau_0} \int_{\tau_f} P(\mathbf{R}_n, \mathbf{r}, t) \mathrm{d}^3\mathbf{r}, \tag{20}$$

where τ_0 is the unit-cell superficial volume. It may be shown (Shapiro and Brenner, 1988) that for asymptotically long times ($t \gg l^2/D$) $\bar{P}$ is governed by the Darcy-scale equation (12) possessing phenomenological transport coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ determined in Appendix A, provided that the coarse-grained position vector $\bar{\mathbf{R}}$ appearing in (12) is identified with the discrete global position vector, $\mathbf{R}_n$, (Brenner, 1980b).

4.3. *Validity of the Darcy-scale description*

As discussed above [see also equation (A7)], the coarse-scale transport coefficients exist for times greatly exceeding the characteristic intracell transport time, i.e.

$$t \gg l^2/D, \tag{21}$$

where l is the characteristic unit-cell dimension. Physically, this represents the time interval during which the microscale aerosol density, P, itself attains a kind of self-similar, asymptotic, intracell distribution. Consequently, the latter inequality delineates the range of validity of the Darcy-scale equation (12). Observe, however, that this characteristic microscale time is much less than the characteristic time for Darcy- or macroscale transport, i.e. the characteristic time required for translational motion within the filter bed of the aerosol cloud as a whole. The latter time scale is given by L/U_o, with U_o the superficial air velocity and L the filter thickness. Thus, the condition of applicability of the proposed coarse-scale description of aerosol transport and deposition in filters of finite thickness may be formulated as

$$U_o l/D \ll L/l. \tag{22}$$

The left- and right-hand sides of (22), respectively, represent the characteristic microscale Peclet number and number of collector layers contained within the filter. Because l is generally small ($l \approx a$), the inequality (22) is normally satisfied by both fibrous (Davies, 1973) and granular (Tardos, 1977) filters when collecting submicrometer aerosols.

5. ONE-DIMENSIONAL FILTER MODEL

5.1 *Mean cross-sectional aerosol concentration distribution*

Knowledge of the phenomenological macrotransport coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ appearing in the macrotransport equation (12) governing $\bar{P}$, permits their eventual use in calculating the filtration efficiency, η. It is demonstrated (see Appendix A) that these three coefficients are independent of the initial aerosol spatial concentration distribution. As such, they may be regarded as material properties of the macrocontinuum, characterizing the Darcy-scale deposition and transport processes for a given aerosol and filter bed, irrespective of the filter's (finite) macroscopic cross-sectional shape, $\mathbf{q}_o$, as well as of the inlet aerosol concentration distribution. In principle, equation (12) may be solved pointwise to obtain the macroscale aerosol distribution, $\bar{P}$, at each point, $\bar{\mathbf{R}}$, within the filter bed. In turn, this

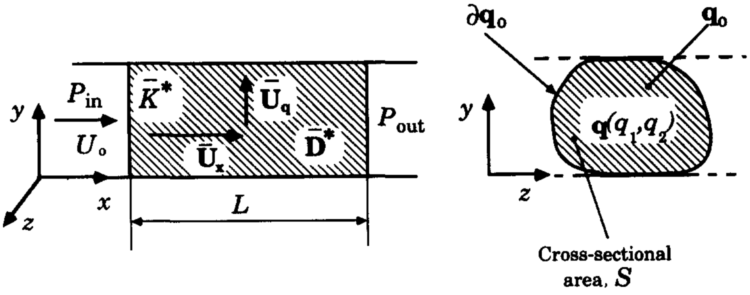

Fig. 5. Porous filter. Macroscale view.

detailed information may be used to calculate the filtration efficiency η. In practice, however, this pointwise aerosol distribution (even at this coarse level of description) is still unnecessarily detailed if our sole purpose is to calculate η. This goal may be achieved by considering the mean cross-sectional aerosol concentration

$$\bar{P}_f(x,t) \overset{\text{def.}}{=} \frac{1}{S}\int_{\mathbf{q}_o} \bar{P}(x,\mathbf{q},t)\,\mathrm{d}\mathbf{q}, \tag{23}$$

where $\mathbf{q}_o$ is the two-dimensional domain characterizing the filter's areal cross-section, S, and x is the axial cartesian coordinate measured along the air flow direction (see Fig. 5).

5.2. *One-dimensional axial aerosol transport equation*

The coarse-scale aerosol concentration within $\mathbf{q}_o$ may be described by the vector $\mathbf{q} = \mathbf{i}_1 q_1 + \mathbf{i}_2 q_2$, characterized as in Fig. 5 by the two (generally curvilinear) coordinates (q_1, q_2) (Happel and Brenner, 1973). Consequently, the generic macroscale position vector $\bar{\mathbf{R}}$ may be decomposed into the orthogonal sum

$$\bar{\mathbf{R}} = \mathbf{i}_x x + \mathbf{q}. \tag{24}$$

The objective of this section is to eliminate details of the $\mathbf{q}$-dependence of the aerosol concentration $\bar{P}$, thereby passing to the cross-sectionally averaged description $\bar{P}_f$ of the aerosol concentration field, defined by equation (23). Analogous to our earlier remarks concerning $\bar{P}$, it is not $\bar{P}_f$ itself that constitutes the main focus of interest, but rather the effective phenomenological coefficients $\bar{K}_f$, $\bar{U}_f$ and $\bar{D}_f$ governing the aerosol's axial depositional diminution and transport rates. These scalar coefficients, respectively, represent the mean aerosol axial reactivity, velocity and dispersivity. All three appear in the one-dimensional axial transport equation

$$\frac{\partial \bar{P}_f}{\partial t} + \bar{U}_f \frac{\partial \bar{P}_f}{\partial x} - \bar{D}_f \frac{\partial^2 \bar{P}_f}{\partial x^2} + \bar{K}_f \bar{P}_f = 0 \tag{25}$$

governing the axial aerosol distribution $\bar{P}_f(x,t)$, subject to the appropriate initial condition $\bar{P}_f(x,0)$ [obtained by subsequent integrations of $f(\mathbf{R}_n, \mathbf{r})$ appearing in (19) over the interstitial unit-cell volume, via equation (20), and over the filter cross-sectional area, via equation (23)]. In this section we will determine these three phenomenological coefficients in terms of the comparable Darcy-scale phenomenological coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$.

5.3. *Mean axial aerosol transport coefficients*

Consider (as in Section 3) the model problem of the propagation, diffusion and deposition of an aerosol cloud within the filter. Now, however, we will describe this process at the macroscale using equation (12) with coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$, here regarded as known constants.

The nature of the aerosol transport process within the filter cross-section is materially different from that along the filter-bed axis. Aerosol transport within the filter cross-section, $\mathbf{q}_o$, is hindered by the presence of the filter walls $\partial \mathbf{q}_o$. Therefore, for sufficiently long times, the cross-sectional aerosol distribution effectively attains an asymptotic terminal state. However, aerosol transport in the axial direction is undisturbed, provided that sufficient filter thickness obtains.

Thus, for times greatly exceeding the characteristic time for cross-sectional transport, the aerosol cloud as an integral entity is transported only along the x axis. In such circumstances the axial aerosol transport process is grossly quantified by the following global quantities (Brenner, 1980a; Shapiro and Brenner, 1986, 1987):

$$M_{x0} = \int_{\mathbf{q}_o} \mathrm{d}\mathbf{q} \int_{-\infty}^{\infty} \mathrm{d}x \, \bar{P}(x, \mathbf{q}, t), \tag{26a}$$

$$\bar{x} = \frac{1}{M_{x0}} \int_{\mathbf{q}_o} \mathrm{d}\mathbf{q} \int_{-\infty}^{\infty} \mathrm{d}x \, x \, \bar{P}(x, \mathbf{q}, t), \tag{26b}$$

$$\overline{(x - \bar{x})^2} = \frac{1}{M_{x0}} \int_{\mathbf{q}_o} \mathrm{d}\mathbf{q} \int_{-\infty}^{\infty} \mathrm{d}x \, (x - \bar{x})^2 \, P(x, \mathbf{q}, t), \tag{26c}$$

respectively, expressing the total aerosol 'mass', the axial position of the center of mass of the aerosol cloud and the aerosol's axial dispersion with respect to this moving mass center, all at time t. Further remarks will be offered below as to how large the filter bed thickness must be to justify the infinite-domain definitions (26).

The scalar global phenomenological coefficients $\bar{K}_f, \bar{U}_f$ and $\bar{D}_f$ governing the cross-sectionally averaged aerosol deposition and transport rates are expressed in terms of the three global parameters defined in equations (26a–c) via the following relationships (Brenner, 1980a; Shapiro and Brenner, 1986, 1987):

$$\bar{K}_f = -\lim_{t \to \infty} \frac{\mathrm{d} \ln M_{x0}}{\mathrm{d}t}, \tag{27a}$$

$$\bar{U}_f = \lim_{t \to \infty} \frac{\mathrm{d}\bar{x}}{\mathrm{d}t}, \tag{27b}$$

$$\bar{D}_f = \frac{1}{2} \lim_{t \to \infty} \frac{\mathrm{d}}{\mathrm{d}t} \overline{(x - \bar{x})^2}. \tag{27c}$$

As before, the coefficients $\bar{K}_f$, $\bar{U}_f$ and $\bar{D}_f$ are expected to prove independent of the mode of initial introduction of the aerosol cloud into the filter, as well as of the total initial aerosol content.

In Appendix B the mean axial transport coefficients $\bar{K}_f$, $\bar{U}_f$ and $\bar{D}_f$ are explicitly expressed in terms of the Darcy-scale coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ [see equations (B13), (B14)]. In particular, for the cross-sectionally isotropic filter bed, $\bar{U}_f$ and $\bar{D}_f$ are given by the respective axial components $\bar{U}_x$ and $\bar{D}_{xx}$ of the Darcy-scale velocity $\bar{\mathbf{U}}^*$ and dispersivity $\bar{\mathbf{D}}^*$ [see equations (B16a, b) together with decomposition (B1a, b).

5.4. *Applicability of equation* (25) *to filter beds of finite thickness*

Several remarks are in order concerning the application to a bed of finite thickness, L, of the one-dimensional equation (25), whose phenomenological coefficients $\bar{K}_f$, $\bar{U}_f$ and $\bar{D}_f$ were originally derived for an infinitely-extended filter bed in the axial direction. Generally speaking, equation (25)—with phenomenological coefficients derived as indicated above—is valid for times, t, exceeding the characteristic time $l_q^2/||\bar{\mathbf{D}}_{qq}||$ for cross-sectional aerosol transport, with l_q a characteristic linear cross-sectional dimension and $||\bar{\mathbf{D}}_{qq}||$ an appropriate norm of the cross-sectional dyadic $\bar{\mathbf{D}}_{qq}$ [see decomposition (B1b)]. On the other hand, this characteristic time must be much less than that required for axial aerosol transport through the entire bed (of length L), namely $L/\bar{U}_f$; otherwise, the aerosol particle may have exited from the bed before having had the opportunity to 'equilibrate transversely' by attaining its Boltzmann distribution [cf. equation (B10)] in $\mathbf{q}_o$. Thus, a necessary condition governing the applicability of (25) is obviously

$$\frac{l_q \bar{U}_f}{\|\bar{\mathbf{D}}_{qq}\|} \ll \frac{L}{l_q}. \tag{28}$$

Usually, as a result of convective dispersion (Taylor, 1953, 1954a, b), the dispersivity norm $\|\bar{\mathbf{D}}_{qq}\|$ appearing in (28) greatly exceeds the aerosol's molecular diffusivity, D. Consequently, the limitation (28) imposed upon the filter's linear dimensions, L and l_q, as well as upon its operating conditions, is not as severe as might superficially appear.

In circumstances where the axial and transverse (cross-sectional) aerosol transport processes are not coupled [cf. equations (B2a, b) and (B15)], the aerosol redistribution occurring within each filter cross-section $x = \text{const.}$ towards the Boltzmann distribution does not preclude the use of (25) for times preceding the cessation of this redistribution process. In this case the effective axial equation (25) is valid almost immediately following introduction of the aerosol cloud into the filter bed. (In this case the equilibrium time is of the order of the characteristic intracell aerosol transport time.) Therefore, when (B15) obtains, the restriction (28) becomes inoperative and hence need not be imposed; instead, only the milder condition (22), established earlier, limits the domain of applicability of (25).

6. FILTRATION EFFICIENCY

6.1. *Steady-state aerosol filtration problem*

Equation (25) governs the asymptotically long-time, transversely-averaged concentration distribution of an aerosol cloud propagating and depositing within a porous filter that extends indefinitely in the axial direction. As described above, this process is inherently time dependent. On the other hand, real porous filters possess finite thicknesses and their performance is characterized by an almost steady-state deposition process. While the latter problem differs formally from the model problem of propagation and deposition of an aerosol cloud in a porous medium, we observe that the steady-state aerosol filtration process may be viewed as a continuous source problem (Gill and Sankarasubramanian, 1972) in which each subsequent portion of aerosol particles entering the filter propagates and deposits within the filter medium independently of any particles earlier introduced. Superposition of all contributions issuing from such successive portions is described by the steady-state version of equation (25), namely

$$\bar{D}_f \frac{d^2 \bar{P}_f}{dx^2} - \bar{U}_f \frac{d\bar{P}_f}{dx} - \bar{K}_f \bar{P}_f = 0. \tag{29}$$

The applicability of equation (29) was formally substantiated and discussed by Shapiro *et al.* (1988b).

Continuity of the aerosol flux across the filter inlet requires imposing the boundary condition

$$U_o P_{in} = \bar{U}_f \bar{P}_f - \bar{D}_f \frac{d\bar{P}_f}{dx} \quad \text{at } x = 0. \tag{30a}$$

Analogously, at the filter outlet,

$$U_o P_{out} = \bar{U}_f \bar{P}_f - \bar{D}_f \frac{d\bar{P}_f}{dx} \quad \text{at } x = L. \tag{30b}$$

Thus, the ratio P_{out}/P_{in} will necessarily appear as an independent parameter governing the axial aerosol concentration distribution $\bar{P}_f$.

In contrast, classical filtration theory fails to predict any dependence of $\bar{P}_f$ upon the ratio P_{out}/P_{in}. This disparity is here resolved by noting that the classical model supposes the aerosol to be 'diffusion' (dispersion)-free at the macroscale, despite the fact that it undergoes diffusion at the microscale (see e.g. Tardos, 1977; Tien, 1989). The present model rectifies this omission by incorporating macroscale dispersion into the description. Inclusion of this additional transport mechanism necessitates the use of boundary conditions at both $x = 0$ and $x = L$ when formulating the filtration problem in order that the mathematical boundary-value problem governing the second-order equation (29) be uniquely posed.*

The engineering significance of the functional dependence of $\bar{P}_f$ upon the ratio P_{out}/P_{in} derives from the dependence of this concentration distribution upon the detailed manner in which the penetrant aerosol is removed at the filter outlet. The latter effect may be important for very short filters, but is generally insignificant in situations of practical importance, where the filter is sufficiently long. For such cases the boundary condition at the filter outlet is specified here in the form (Danckwerts, 1953; Wehner and Wilhelm, 1956):

$$\frac{d\bar{P}_f}{dx} = 0 \qquad \text{at } x = L. \tag{30c}$$

This boundary condition is further discussed and justified in the theory of chemical reactors by Pearson (1959), Bischoff (1961), Fan and Ahn (1962) and Bass *et al.* (1988). Use of this condition instead of (30b) eliminates the dependence of $\bar{P}_f$ upon the ratio P_{out}/P_{in}.

6.2. *Solution of the aerosol filtration equations*

Equations (21), (22a) and (22c) possess the solution

$$\frac{\bar{P}_f(\bar{x}_f)}{P_{in}} = \frac{U_o}{\bar{U}_f}\,\frac{\lambda_1 \exp[\lambda_2(\bar{x}_f - \bar{L})] - \lambda_2 \exp[\lambda_1(\bar{x}_f - \bar{L})]}{\lambda_1^2 \exp(-\lambda_2 \bar{L}) + \lambda_2^2 \exp(-\lambda_1 \bar{L})}, \tag{31}$$

wherein

$$\bar{x}_f = x\bar{U}_f/\bar{D}_f, \qquad \bar{L} = L\bar{U}_f/\bar{D}_f, \tag{32a, b}$$

$$\lambda_1 = \frac{1}{2}[1 + (1 + 4\varepsilon_f)^{1/2}], \qquad \lambda_2 = \frac{1}{2}[1 - (1 + 4\varepsilon_f)^{1/2}], \tag{33a, b}$$

with

$$\varepsilon_f = \bar{K}_f\bar{D}_f/\bar{U}_f^2 \tag{34}$$

which are dimensionless quantities. These results may be used to obtain the filtration efficiency, defined by (1) or (2), in the form

$$\eta = 1 - \frac{\lambda_1 - \lambda_2}{\lambda_1^2 \exp(-\lambda_2 \bar{L}) + \lambda_2^2 \exp(-\lambda_1 \bar{L})}. \tag{35}$$

6.3. *Limiting cases of aerosol filtration*

Two limiting cases are of special interest.

Case (i): $\varepsilon_f >> 1$. Here, (35) reduces to

$$\eta = 1 - 2\varepsilon_f^{-1/2} \exp\left[-\left(\frac{\bar{K}_f}{\bar{D}_f}\right)^{1/2} L\right]. \tag{36}$$

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* In fact, as indicated by (2), the ratio P_{out}/P_{in} completely determines the filter efficiency (and the converse). This observation, as well as formula (2) itself, implicitly assumes that the aerosol transport process preceding the filter inlet and after the filter outlet is dispersion free. In more general situations, where the latter assumption does not necessarily obtain, the filtration efficiency must be calculated by solving the aerosol transport problem within the porous filter bed simultaneously with that in the two effectively semi-infinite regions, respectively, preceding the filter inlet and following the filter outlet. In this case P_{in} and P_{out}, appearing in equation (2) defining the filtration efficiency, should be understood, respectively, as aerosol concentrations existing at locations far upstream from the filter inlet and far downstream from the filter exit (Bass *et al.*, 1988).

The inequality (i) and concomitant efficiency formula (36) are obviously valid when the filter face velocity, U_o, is sufficiently small, whereupon axial aerosol transport occurs primarily by diffusion. [Strictly, for this case, one has to substantiate the applicability of the Danckwerts boundary condition (22c), which is normally regarded as valid in circumstances wherein aerosol convective transport dominates (or is at least comparable to) diffusive transport. However, when the dimensionless parameter $(\bar{K}_f/\bar{D}_f)^{1/2} L$ appearing in (36) is much larger than unity, corresponding to a sufficiently long filter, the boundary condition (22c) continues to seem appropriate.] Further investigations are obviously required to delineate the extent to which case (i) describes any practically important aerosol filtration regimes.

Case (ii): $\varepsilon_f << 1$. Here, equation (35) adopts the elementary form

$$\eta = 1 - \exp\left(-\frac{\bar{K}_f L}{\bar{U}_f}\right), \tag{37}$$

in which circumstance the filtration efficiency is independent of the macroscale axial dispersivity, $\bar{D}_f$. It would appear that many practically important filtration operating conditions fall within the purview of case (ii). Indeed, calculations of the mean axial transport coefficients $\bar{K}_f$, $\bar{U}_f$ and $\bar{D}_f$ reported in the next section, and subsequent evaluation of ε_f, show that the inequality (ii) governing this case is achieved at sufficiently high values of the characteristic microscale Peclet number.

6.4. *Characteristic filtration length, l_f*

Equation (37) may be rewritten alternatively as

$$\eta = 1 - \exp(-L/l_f), \tag{38}$$

with

$$l_f = \bar{U}_f/\bar{K}_f \tag{39}$$

the characteristic filtration length (Leers, 1957). For the case $\varepsilon_f << 1$ considered here, l_f appears as the single intrinsic filtration parameter characterizing the efficiency of the collection process for a given aerosol and for specified operating conditions, irrespective of the filter's macroscale geometry (i.e. the linear size and cross-sectional shape of the filter bed as a whole).

Equation (38) may be also compared with an analogous exponential form derived by integration of the filtration-rate expression

$$\frac{d\bar{P}_f}{dx} = -\lambda \bar{P}_f \tag{40}$$

first suggested by Iwasaki (1937) [see also Tien (1989)] after analysing experimental data obtained for sand filters. The parameter λ appearing in (40) (which possesses units of reciprocal length) is termed the filter coefficient. Comparison of the solution of (40) with equation (38) yields $l_f = \lambda^{-1}$. In Iwasaki's (1937) approach [as developed in Tien's (1989) monograph] the filter coefficient, λ, is regarded as a fundamental experimental parameter, which is related to the unit-bed efficiency. In contrast with the above cited works of Leers and Iwasaki, in our contribution the existence of l_f (or λ) is rigorously demonstrated from the fundamental aerosol transport and deposition equations; moreover, its numerical value may be rationally derived via equation (39) from the fundamental axial aerosol transport/reaction coefficients $\bar{U}_f$ and $\bar{K}_f$, rather than having to be obtained experimentally.

It is noteworthy that a functional similarity exists between the respective efficiency formulas (3) and (38), each predicting an exponential dependence of filtration efficiency upon filter-bed thickness, L. However, whereas (3) represents an *ad hoc*, virtually semi-empirical expression, its rational alternative (38) was derived by rigorous physico-mathematical arguments applied under well-defined limiting asymptotic conditions to a physically realistic (albeit idealized) geometrical model of the filter bed. Although only the two phenomenological coefficients $\bar{K}_f$ and $\bar{U}_f$ seem to be required in the filtration efficiency calculation (38), explicit knowledge of $\bar{D}_f$ is nevertheless also required to assure fulfillment of the inequality $\varepsilon_f << 1$ underlying the validity of (38).

7. EXAMPLE. FILTRATION OF DIFFUSIONAL AEROSOLS BY FIBROUS FILTERS

The theory developed here was implemented by calculation of the filtration efficiency of a fibrous filter collecting fine aerosol particles possessing diameters $d_p \leq 0.1$ μm. Specifically, we studied the situation where Brownian diffusion dominates over all other deposition mechanisms, namely, over interception, inertia and external forces, which were neglected. For simplicity, the geometric microstructure of the filter bed was modelled by a square array of circular cylinders with their axes perpendicular to the mean air flow direction (see Fig. 6). As a first step, calculations of the spatially periodic Navier–Stokes velocity field in such an array were performed for various bed void fractions. These calculations, as well as the subsequent numerical integration of the pertinent unit-cell boundary-value problems [see equations (A11a)–(A12d) and (A17a–d)] were performed by finite element methods. Details of the computational scheme employed, together with the results obtained for the flow field, are presented by Edwards *et al.* (1989).

Results. The coarse-scale aerosol transport coefficients were calculated as functions of the characteristic microscale Peclet number $\mathrm{Pe} = 2aU_0/D$. For simplicity, the characteristic microscale Reynolds number was chosen to be zero. A comprehensive parametric investigation of the effects of Reynolds number, as well as of other characteristic filtration parameters, will be reported in a forthcoming contribution (Shapiro *et al.*, 1990b).

For the preceding filter-bed geometry the Darcy-scale aerosol velocity vector, $\bar{\mathbf{U}}^*$, was determined to lie parallel to the mean air flow direction. The Darcy-scale aerosol dispersivity dyadic was characterized by its longitudinal ($\bar{D}_{xx}$) and lateral ($\bar{D}_{yy}$) components, both of which were computed. As the geometric configuration of the filter bed considered is characterized by the absence of coupling between aerosol transport in the longitudinal and lateral directions, the mean axial aerosol velocity, $\bar{U}_f$, and dispersivity, $\bar{D}_f$, are, respectively, given by their Darcy-scale counterparts, $\bar{U}^*_x$ and $\bar{D}_{xx}$ [see equations (B16a, b)]. Moreover, the calculated data showed for $\mathrm{Pe} \gg 1$ that the macroscale aerosol transport coefficients obey the inequality $\varepsilon_f \ll 1$ [see equation (34) and case (ii) of section 6]. Consequently, the high Peclet number aerosol collection process may be uniquely characterized solely by the characteristic filtration length, l_f.

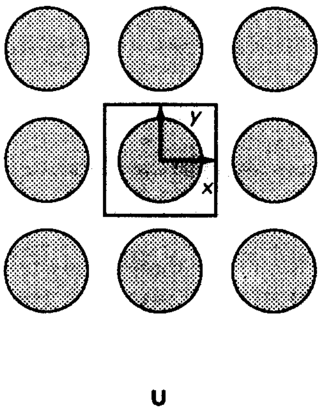

Fig. 6. Spatially periodic model of a fibrous filter used in computations.

Table 1. Comparison of characteristic filtration lengths, l_f, with available data

Aerosol particle diameter d_p (μm)	Filter-face velocity U_0, (cm s^{-1})	Characteristic filtration length, l_f (cm): Present theory	Stechkina and Fuchs (1966): Formula (43)	Stechkina and Fuchs (1966): Formula (44)	Experimental, Lee (1977)
0.035	10	0.105	0.048	0.040	0.128
0.035	30	0.315	0.099	0.084	0.203
0.05	3	0.081	0.039	0.033	0.124
0.05	10	0.267	0.088	0.075	0.191
0.07	3	0.116	0.050	0.043	0.168
0.07	10	0.386	0.113	0.096	0.298
0.1	1	0.098	0.045	0.038	0.279
0.1	3	0.289	0.093	0.079	0.384
0.1	10	0.856	0.210	0.178	0.800

7.1. *Comparison with previous results*

Calculated values of l_f were compared with theoretical results of Stechkina and Fuchs (1966) and with the experimental data tabulated by Lee (1977). Computations of the characteristic filtration length were specifically performed for the filtration parameters (reported in the latter study), corresponding to the collection of diffusional aerosol particles by Dacron filters of thickness $L = 0.354$ cm composed of 11 μm diameter fibers. Results for a filter characterized by a void fraction of $\varepsilon = 0.849$ which was collecting 0.035–0.1 μm diameter dioctyl phthalate aerosol particles are summarized in Table 1. Experimental values of l_f were calculated from Lee's (1977) filtration efficiency data by the formula [cf. (38)]:

$$l_f = -\frac{L}{\ln(1-\eta)}. \tag{41}$$

The theoretical data of Stechkina and Fuchs (1966), who calculated the single-fiber efficiency for diffusional aerosol collection (from the Kuwabara flow field) in the form

$$\eta_c = 2.9\,\mathrm{Ku}^{-1/3}\mathrm{Pe}^{-2/3} + 0.624\,\mathrm{Pe}^{-1}, \tag{42}$$

with $\mathrm{Ku} \equiv -1/2\ln(1-\varepsilon) - \varepsilon/2 - \varepsilon^2/4$ the Kuwabara number is also included in Table 1. As already mentioned in the Introduction, the correlation (3) between the single fiber efficiency, η_c, and the total filter efficiency, η, depends upon the exact form of the coefficient α appearing therein. Davies (1973) used $\alpha = 2(1-\varepsilon)/\pi$, which in combination with (3) and (38) yields the following expression for l_f:

$$\frac{l_f}{2a} = \frac{1}{1-\varepsilon}\frac{\pi}{4\eta_c}. \tag{43}$$

On the other hand, Lee (1977) and Lee and Liu (1982a, b) suggested another formula for α, which differs from that of Davies (1973) by the factor ε. With Lee's expression for α, l_f becomes

$$\frac{l_f}{2a} = \frac{\varepsilon}{1-\varepsilon}\frac{\pi}{4\eta_c}. \tag{44}$$

The values of l_f computed from both (43) and (44), with η_c evaluated from (42), are listed in Table 1. As can be seen, the values of the characteristic filtration length calculated using the theory of Stechkina and Fuchs (1966) [and either equation (43) or (44)] are significantly lower than the experimentally measured l_f. As such, the existing 'single fiber efficiency' theory of Stechkina and Fuchs' systematically overestimates the total efficiency of the filter. On the other hand, the values of l_f calculated using the proposed dispersion/reaction theory, reasonably fit the experimental data of Lee (1977)—even for the oversimplified, parallel fiber, filter-bed microstructure chosen for our computations.

Of course, the limited computational results presented in this section are insufficient to fully assess the merits of the filtration model proposed in this study. A much more comprehensive investigation of aerosol filtration based upon the present model will be presented in a subsequent paper (Shapiro *et al.*, 1990b).

8. DISCUSSION AND SUMMARY

The proposed dispersion/reaction model of aerosol transport and deposition phenomena in porous media constitutes a novel fundamental contribution to the classical theory of aerosol filtration by porous filter beds. This contribution frees the classical theory of many implicit and explicit *ad hoc* assumptions. Central points underlying the proposed scheme consist of: (i) representing a filter bed as a spatially periodic porous medium; (ii) viewing the aerosol filtration process as consisting of combined interstitial convective and dispersion phenomena, accompanied by irreversible deposition on the filter bed collector particles; and (iii) subsequently passing from the microscale or interstitial view to a coarser macro- or Darcy-scale description of the phenomena.

Instead of the misleading concept of a unit-bed efficiency, η_c, we naturally introduce and rigorously calculate three Darcy-scale aerosol macrotransport coefficients. These constitute physically objective, experimentally accessible quantities, in terms of which the filtration efficiency, η, is easily determined. These Darcy-scale phenomenological coefficients (each of which is, in principle, separately measurable rather than being lumped together) provide a new conceptual framework for understanding aerosol transport and filtration processes within porous filters; the underlying analysis derives, in part, from our earlier theory (Shapiro and Brenner, 1988; Dungan *et al.*, 1990) of heterogeneous chemical reactions in porous media—in particular involving irreversible, infinitely rapid, surface reaction phenomena on the bed particles. The present approach can be used to delineate the range (if indeed any exists) for which the classical concept of unit-bed efficiency results in a viable, albeit somewhat empirical, method for designing porous aerosol filters.

Existing filtration theory separates, on an *ad hoc* basis, the influence of local bed microstructure from that of global or macroscale filter thickness, L. In contrast, our proposed model fuses these distinct geometric influences via a rigorous physico-mathematical scheme that clearly describes the circumstances under which the aerosol filtration process can be described by Leers' (1957) characteristic filtration length, l_f, as an intrinsic filtration parameter.

Despite its apparent complexity compared with standard unit-bed efficiency methods, the proposed dispersion/reaction macrotransport model is based upon a well-posed aerosol microtransport problem, which can accordingly be treated by standard analytical and/or numerical methods. Computations, performed by finite element methods, of the coarse-scale transport coefficients characterizing the diffusional aerosol particles in spatially periodic fibrous media, yielded values of the characteristic filtration length in accordance with the available experimental data of Lee (1977).

While the proposed dispersion/reaction model of aerosol filtration bodes useful in examining several fundamental assumptions underlying the transport and collection of micrometer and submicrometer aerosols, its real advantage lies more, perhaps, in its ability to systematically rationalize the quantitative effects of aerosol properties, bed structure and filtration operating parameters upon aerosol filtration efficiency. These questions will be addressed in a future publication (Shapiro *et al.*, 1990b).

Moreover, the proposed model is able to account for the following parameters and processes (which cannot strictly be addressed by current single-element efficiency theories): (i) polydisperse structure of the filter bed; (ii) irregularity of collector shapes; (iii) non-spherical nature of the aerosol particles; and (iv) kinetics of chemical conversion of aerosols upon collector surfaces (in circumstances where actual, rather than fictitious chemical reactions occur). These research agenda items represent longer-term areas of investigation of potential value arising from the proposed filtration model.

By placing the transport and deposition of fine particles within a porous media upon a firmer physico-mathematical basis than has hitherto existed, our theory potentially impacts adjacent engineering areas concerned with the behavior of fluid-particle systems; included are deep-bed filtration (Payatakes *et al.*, 1973; Rajagopalan and Tien, 1979) and particle separation processes (Watson, 1973; Lin and Benguigui, 1983). Related research areas are addressed elsewhere (Shapiro and Brenner, 1988).

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APPENDIX A. METHOD OF MOMENTS

Define intracellular local and total polyadic moments of the aerosol number density, P, by the respective expressions

$$\mathbf{P}_m(\mathbf{r},t)=\sum_{\mathbf{n}}\mathbf{R}_{\mathbf{n}}^m P(\mathbf{R}_{\mathbf{n}},\mathbf{r},t), \tag{A1a}$$

$$\mathbf{M}_m(t)=\int_{\mathbf{r}_f}\mathbf{P}_m(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}, \tag{A1b}$$

for $m = 0, 1, 2, \ldots$. Here

$$\sum_{\mathbf{n}} \equiv \sum_{n_1=-\infty}^{\infty}\ \sum_{n_2=-\infty}^{\infty}\ \sum_{n_3=-\infty}^{\infty}.$$

The zero-order total moment, M_0, defined by (A1) with $m=0$, represents a version of the equivalent formula (10a) for the spatially periodic porous filter model. Similarly, the displacement, $\bar{\mathbf{R}}$, of the aerosol cloud's center of mass as well as its dispersion, $\overline{(\Delta\mathbf{R})^2}$, at time t, respectively, defined by (10b, c), may be expressed in terms of the first three total moments $M_0, \mathbf{M}_1, \mathbf{M}_2$ as (Brenner, 1980b; Shapiro and Brenner, 1988)

$$\bar{\mathbf{R}} \equiv \frac{\mathbf{M}_1}{M_0}, \tag{A2a}$$

$$\overline{(\Delta\mathbf{R})^2} \equiv \frac{\mathbf{M}_2}{M_0} - \frac{\mathbf{M}_1\mathbf{M}_1}{M_0^2}. \tag{A2b}$$

Because the phenomenological coefficients $\bar{K}^*, \bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ required in the theory are to be calculated via (11) from knowledge of the long-time behavior of M_0, $\bar{\mathbf{R}}$ and $\overline{(\Delta\mathbf{R})^2}$, their calculation can obviously be effected by establishing the comparable asymptotic properties of M_0, $\mathbf{M}_1$ and $\mathbf{M}_2$.

Towards this end, we consider first the asymptotic behavior of P_0, $\mathbf{P}_1$ and $\mathbf{P}_2$. Multiply equations (15) and (16) by $\mathbf{R}_\mathbf{n}^m$, sum over all of the infinitely many unit-cells, and use boundary conditions (18) to obtain (Brenner, 1980b)

$$\frac{\partial\mathbf{P}_m}{\partial t} + \nabla\cdot(\mathbf{J}\mathbf{P}_m) = \mathbf{0}, \tag{A3a}$$

$$\mathbf{P}_m = \mathbf{0} \text{ on } S_c. \tag{A3b}$$

$\mathbf{P}_m$ must also obey certain so-called 'jump' conditions imposed across the unit-cell faces $\partial\mathbf{r}_k$. These are obtained by employing the continuity conditions (17) to obtain (Brenner, 1980b)

$$\llbracket P_0 \rrbracket = 0, \quad \llbracket \nabla P_0 \rrbracket = \mathbf{0}, \tag{A4a, b}$$

$$\llbracket \mathbf{P}_1 \rrbracket = -\llbracket \mathbf{r}P_0 \rrbracket, \quad \llbracket \nabla\mathbf{P}_1 \rrbracket = -\llbracket \nabla(\mathbf{r}P_0) \rrbracket, \tag{A4c, d}$$

$$\llbracket \mathbf{P}_2 \rrbracket = \llbracket \mathbf{P}_1\mathbf{P}_1/P_0 \rrbracket, \quad \llbracket \nabla\mathbf{P}_2 \rrbracket = \llbracket \nabla(\mathbf{P}_1\mathbf{P}_1/P_0) \rrbracket. \tag{A4e, f}$$

Generically, $\llbracket \mathbf{F} \rrbracket$ denotes the change in the value of the tensor-valued function, $\mathbf{F}$, across equivalent points lying on opposite cell faces, defined explicitly by the expression

$$\llbracket \mathbf{F} \rrbracket_k = \mathbf{F}(\mathbf{r} + \boldsymbol{l}_k) - \mathbf{F}(\mathbf{r}). \tag{A5}$$

To complete the problem statement defining the local moments, $\mathbf{P}_m$, the initial condition (19) is employed to obtain

$$\mathbf{P}_m(\mathbf{r}, 0) = \sum_\mathbf{n} \mathbf{R}_\mathbf{n}^m f(\mathbf{R}_\mathbf{n}, \mathbf{r}). \tag{A6}$$

Temporal evolution and intracellular transport of the local moments, $\mathbf{P}_m$, within a unit-cell is governed by the system of equations (A3)–(A6). Although none of the local moments exhibits a steady-state solution, all achieve asymptotic, long-time, limiting forms for times, t, satisfying the inequality

$$t \gg l^2/D, \tag{A7}$$

where $l = \max\{|\boldsymbol{l}_1|, |\boldsymbol{l}_2|, |\boldsymbol{l}_3|\}$.

A.1. *Zero-order moments*

Consider the zero-order moment problem for P_0 posed by equations (A3a, b), (A6) (with $m=0$) and (A4a, b). Shapiro and Brenner (1988) showed that the asymptotic long-time solution of this problem may be expressed as

$$P_0(\mathbf{r}, t) \simeq A_0 h(\mathbf{r})\Phi_0(\mathbf{r})\exp(\lambda_0 t)(1 + \exp), \tag{A8}$$

where 'exp', appearing without an argument, denotes a function decaying exponentially rapidly with time; additionally,

$$h(\mathbf{r}) = \exp[-E(\mathbf{r})], \tag{A9}$$

$$A_0 = \int_{\mathbf{r}_f} f_0(\mathbf{r})\Psi_0(\mathbf{r})\mathrm{d}^3\mathbf{r}. \tag{A10}$$

In (A8) and (A10) the quantities λ_0, $\Phi_0(\mathbf{r})$ and $\Psi_0(\mathbf{r})$, respectively, represent the common leading eigenvalue and corresponding eigenfunctions (Courant and Hilbert, 1966) of the following pair of adjoint eigenvalue problems (EVPs):

(i) Characteristic EVP:

$$\nabla\cdot(h\mathbf{J}\Phi) + \lambda h\Phi = 0 \quad \text{in } \mathbf{r}_f, \tag{A11a}$$

$$\Phi = 0 \quad \text{on } S_c, \tag{A11b}$$

$$\llbracket \Phi \rrbracket = 0, \quad \llbracket \nabla\Phi \rrbracket = \mathbf{0}; \tag{A11c, d}$$

(ii) Adjoint EVP:

$$h\mathbf{U}\cdot\nabla\Psi + \nabla\cdot(hD\nabla\Psi) = \lambda h\Psi \quad \text{in } \mathbf{r}_f, \tag{A12a}$$

$$\Psi = 0 \quad \text{on } S_c, \tag{A12b}$$

$$\llbracket \Psi \rrbracket = 0, \quad \llbracket \nabla\Psi \rrbracket = \mathbf{0}. \tag{A12c, d}$$

The leading eigenfunctions Φ_0 and Ψ_0, together with their common eigenvalue λ_0, may be explicitly calculated for a prescribed unit-cell geometry and given microscale phenomenological functions $\mathbf{U}(\mathbf{r})$, $E(\mathbf{r})$, D. Note that Φ_0 and Ψ_0 are each non-negative within $\mathbf{r}_f$ (Shapiro and Brenner, 1988) thus assuring the same property for P_0 (at least for asymptotically long times). These eigenfunctions (which are generally defined only to within arbitrary constant multipliers) are here chosen so as to satisfy the following integral normalization condition:

$$\int_{\mathbf{r}_f} h(\mathbf{r})\Psi_0(\mathbf{r})\Phi_0(\mathbf{r})\mathrm{d}^3\mathbf{r} = 1. \tag{A13}$$

The asymptotically long-time [cf. (A7)] expression for the zero-order total moment, M_0, obtained by integration of (A8) over the unit-cell interstitial domain $\mathbf{r}_f$, is

$$M_0(t) = \bar{A}_0 \exp(\lambda_0 t)(1 + \exp), \tag{A14}$$

where

$$\bar{A}_0 = A_0 \int_{\mathbf{r}_f} h(\mathbf{r})\Phi_0(\mathbf{r})\mathrm{d}^3\mathbf{r}. \tag{A15}$$

A.2. *First-order moments*

Subject to *a posteriori* verification, assume the following asymptotic functional form for $\mathbf{P}_1$:

$$\mathbf{P}_1(\mathbf{r},t) \simeq A_0 h(\mathbf{r})\Phi_0(\mathbf{r}) \exp(\lambda_0 t)[\bar{\mathbf{U}}t + \mathbf{B}(\mathbf{r}) + \exp], \tag{A16}$$

with $\bar{\mathbf{U}}$ a constant vector and $\mathbf{B}(\mathbf{r})$ an intracell vector field, each to be determined. Introduce (A16) into (A3) (with $m = 1$) and (A4c, d), and use (A8) to derive the following system of equations governing the **B** field:

$$\nabla \cdot [h\mathbf{J}(\Phi_0\mathbf{B})] + \lambda_0 h\Phi_0\mathbf{B} = -h\Phi_0\bar{\mathbf{U}} \quad \text{in } \mathbf{r}_f, \tag{A17a}$$

$$\mathbf{B} = \text{finite on } S_c, \tag{A17b}$$

$$\llbracket \mathbf{B} \rrbracket = -\llbracket \mathbf{r} \rrbracket, \quad \llbracket \nabla\mathbf{B} \rrbracket = \mathbf{0}. \tag{A17c, d}$$

To obtain the requisite value of $\bar{\mathbf{U}}$, multiply both sides of (A17a) by $\Psi_0(\mathbf{r})\mathrm{d}^3\mathbf{r}$, integrate over $\mathbf{r}_f$, and use equations (A11a)–(A12d) together with the divergence theorem and jump conditions (A17c, d). This eventually yields

$$\bar{\mathbf{U}} = \int_{\mathbf{r}_f} h[\mathbf{U}\Phi_0\Psi_0 + D(\Phi_0\nabla\Psi_0 - \Psi_0\nabla\Phi_0)]\mathrm{d}^3\mathbf{r}. \tag{A18}$$

Integration of (A16) over the interstitial unit-cell domain furnishes the first-order total moment

$$\mathbf{M}_1(t) \simeq \bar{A}_0 \exp(\lambda_0 t)(\bar{\mathbf{U}}t + \bar{\mathbf{B}} + \exp), \tag{A19}$$

with $\bar{\mathbf{B}}$ given by

$$\bar{\mathbf{B}} = A_0 \bar{A}_0^{-1} \int_{\mathbf{r}_f} h(\mathbf{r})\Phi_0(\mathbf{r})\mathbf{B}(\mathbf{r})\mathrm{d}^3\mathbf{r}. \tag{A20}$$

It may be demonstrated (Shapiro and Brenner, 1988) that the **B**-field problem posed by (A17a–d) possesses a solution which is uniquely determined only to within an arbitrary additive constant vector. This ambiguity will be shown to be without effect upon the mathematical expressions for the final physical results, explicitly upon the values of the phenomenological coefficients $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$.

A.3. *Second-order moments*

Subject to *a posteriori* verification, assume a long-time trial solution for $\mathbf{P}_2$ of the form

$$\mathbf{P}_2(\mathbf{r},t) \simeq A_0 h(\mathbf{r})\Phi_0(\mathbf{r}) \exp(\lambda_0 t)[\bar{\mathbf{U}}\bar{\mathbf{U}}t^2 + 2\mathscr{I}t + 2\,\mathrm{sym}(\bar{\mathbf{U}}\mathbf{B})t + \mathbf{H}(\mathbf{r}) + \exp], \tag{A21}$$

with $\mathscr{I}$ a constant dyadic, $\mathbf{H}(\mathbf{r})$ an intracellular dyadic field to be determined and in which the operator 'sym' denotes the symmetric portion of the dyadic argument. To determine $\mathbf{H}(\mathbf{r})$, introduce (A21) into the problem posed by equations (A3a, b) (with $m = 2$) and (A4e, f) to obtain the following set of equations governing the **H**-field:

$$\nabla \cdot [h\mathbf{J}(\Phi_0\mathbf{H})] - \lambda_0 h\Phi_0\mathbf{H} = 2[\mathscr{I} + \mathrm{sym}(\bar{\mathbf{U}}\mathbf{B})]h\Phi_0, \tag{A22a}$$

$$\mathbf{H} = \text{finite on } S_c, \tag{A22b}$$

$$\llbracket \mathbf{H} \rrbracket = \llbracket \mathbf{BB} \rrbracket, \quad \llbracket \nabla\mathbf{H} \rrbracket = \llbracket \nabla(\mathbf{BB}) \rrbracket. \tag{A22c, d}$$

To obtain the required value of $\mathscr{I}$, multiply both sides (A22a) by $\Psi_0\mathrm{d}^3\mathbf{r}$, integrate over $\mathbf{r}_f$, and use equations (A11a–A12d) and (A17a–d) together with the divergence theorem and jump conditions (A22c, d) to obtain

$$\mathscr{I} = -\frac{1}{2}\int_{\partial\mathbf{r}_o} \mathrm{d}\mathbf{s} \cdot \{\mathbf{U}h\Phi_0\Psi_0\mathbf{BB} + Dh[(\nabla\Psi_0)\Phi_0\mathbf{BB} - \Psi_0\nabla(\Phi_0\mathbf{BB})\} - \mathrm{sym}(\bar{\mathbf{U}} \int_{\mathbf{r}_f} h\Phi_0\Psi_0\mathbf{B}\,\mathrm{d}^3\mathbf{r}). \tag{A23}$$

The **H**-field, defined by equations (A22a–d) with $\mathscr{I}$ given by (A23), may be shown to possess a solution which is determined uniquely only to within an arbitrary additive constant dyadic. However, explicit knowledge of the **H**-field will prove unnecessary in deriving the values of the three Darcy-scale phenomenological coefficients. Accordingly, this lack of uniqueness will prove physically irrelevant.

Integration of (A21) over the interstitial unit-cell domain furnishes the second-order total moment, $\mathbf{M}_2$, as $t \to \infty$ in the form

$$\mathbf{M}_2(t) \simeq \bar{A}_0 \exp(\lambda_0 t)[\bar{\mathbf{U}}\bar{\mathbf{U}}t^2 + 2\mathscr{I}t + 2\,\mathrm{sym}(\bar{\mathbf{U}}\bar{\mathbf{B}})t + \text{const.} + \exp]. \tag{A24}$$

A.4. *Darcy-scale phenomenological coefficients*

Having established the asymptotic behavior of the total moments M_0, $\mathbf{M}_1$, $\mathbf{M}_2$ in equations (A14), (A19) and (A24), respectively, the definitions (11a–c) in conjunction with equations (A2a, b) now permit determination of $\bar{K}^*$, $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$ as

$$\bar{K}^* = -\lambda_0, \quad \bar{\mathbf{U}}^* = \bar{\mathbf{U}}, \quad \bar{\mathbf{D}}^* = \mathscr{I}, \tag{A25a, b, c}$$

with $\bar{\mathbf{U}}$ given by (A18) and $\mathscr{I}$ by (A23). Thus, all three Darcy-scale aerosol deposition and transport coefficients are each now expressed in terms of solutions of the unit-cell problems posed for $\Phi_0(\mathbf{r})$, $\Psi_0(\mathbf{r})$ and $\mathbf{B}(\mathbf{r})$ [as well as the prescribed microscale phenomenological data, namely $\mathbf{U}(\mathbf{r})$, $E(\mathbf{r})$ and D].

The dispersivity, $\bar{\mathbf{D}}^*$, may be computed from its alternative representation (Shapiro and Brenner, 1988)

$$\bar{\mathbf{D}}^* = D\int_{\tau_f} h\Phi_0\Psi_0(\nabla\mathbf{B})^\dagger : (\nabla\mathbf{B})\mathrm{d}^3\mathbf{r}, \tag{A26}$$

which requires knowledge only of $\nabla\mathbf{B}$ rather than of $\mathbf{B}$ itself; here $(\nabla\mathbf{B})^\dagger$ is the transpose of the dyadic $\nabla\mathbf{B}$. Since the $\mathbf{B}$-field is uniquely determined only to within an arbitrary additive constant vector [in consequence of being a solution of the problem posed by equations (A17a–d)], it is evident that the alternative form (A26) of $\mathbf{D}^*$ [cf. (A25c) jointly with (A23)] is unaffected by this arbitrariness and is hence uniquely determined. Equation (A26) shows $\bar{\mathbf{D}}^*$ to be both symmetric and positive definite.

APPENDIX B. CALCULATION OF THE CROSS-SECTIONAL AVERAGED AXIAL AEROSOL TRANSPORT COEFFICIENTS

To establish the long-time behavior of the global quantities (27a–c), consider the aerosol cloud propagation through the filter continuum, as described by the Darcy-scale equation (12). In the most general situation the velocity vector, $\bar{\mathbf{U}}^*$, may possess both x- and $\mathbf{q}$-components, leading to the decomposition

$$\bar{\mathbf{U}}^* = \bar{\mathbf{U}}_x + \bar{\mathbf{U}}_q \tag{B1a}$$

into respective axial and transverse components. Similarly, the dispersion dyadic, $\bar{\mathbf{D}}^*$, may include all nine (non-zero) scalar components (of which only six are independent), corresponding to its decomposition into the partitioned matrix

$$\bar{\mathbf{D}}^* = \left[\begin{array}{c|c} \bar{\mathbf{D}}_{xx} & \bar{\mathbf{D}}_{xq} \\ \hline \bar{\mathbf{D}}_{qx} & \bar{\mathbf{D}}_{qq} \end{array}\right]. \tag{B1b}$$

Here, the direct or 'diagonal' dyadics $\bar{\mathbf{D}}_{xx}(\equiv \mathbf{i}_x\mathbf{i}_x\bar{D}_{xx})$ and $\bar{\mathbf{D}}_{qq}$ represent dispersivities parameterizing the respective axial and cross-sectional Fickian aerosol fluxes, whereas the off-diagonal dyadics, $\bar{\mathbf{D}}_{xq}$ and $\bar{\mathbf{D}}_{qx} \equiv \bar{\mathbf{D}}_{xq}^\dagger$, constitute comparable coupling dispersivities. Constitutive equations for the cross-sectional ($\mathbf{J}_q\bar{P}$) and axial ($\mathbf{J}_x\bar{P}$) aerosol flux components may be written in the respective forms

$$\mathbf{J}_x\bar{P} = \bar{\mathbf{U}}_x\bar{P} - \bar{\mathbf{D}}_{xx}\cdot\nabla_x\bar{P} - \bar{\mathbf{D}}_{xq}\cdot\nabla_q\bar{P}, \tag{B2a}$$

$$\mathbf{J}_q\bar{P} = \bar{\mathbf{U}}_q\bar{P} - \bar{\mathbf{D}}_{qx}\cdot\nabla_x\bar{P} - \bar{\mathbf{D}}_{qq}\cdot\nabla_q\bar{P}, \tag{B2b}$$

where $\nabla_x \equiv \mathbf{i}_x\partial/\partial x$ and $\nabla_q \equiv \partial/\partial\mathbf{q}$ denote respective gradient operators in the x- and $\mathbf{q}$-directions.

Introduce into (12) the aerosol concentration, $\bar{P}'$, defined via the transformation (Brenner, 1980a; Shapiro and Brenner, 1987)

$$\bar{P}'(x,\mathbf{q},t) = \bar{P}(x,\mathbf{q},t)\exp(\bar{K}^*t), \tag{B3}$$

thereby obtaining

$$\frac{\partial\bar{P}'}{\partial t} + \nabla_q\cdot(\mathbf{J}_q\bar{P}') + \nabla_x\cdot(\mathbf{J}_x\bar{P}') = 0. \tag{B4}$$

This equation must be solved subject to the boundary condition

$$\mathbf{n}\cdot\mathbf{J}_q\bar{P}' = 0 \quad \text{on } \partial\mathbf{q}_o \tag{B5}$$

on the filter walls, as these walls are impermeable to penetration by the aerosol. Comparable axial boundary conditions are

$$|x|^m(\bar{P}', \mathbf{J}_x\bar{P}') \to (0, \mathbf{0}) \quad \text{as } |x| \to \infty \tag{B6}$$

for $m = 0, 1, 2, \ldots$. Specification of the initial condition,

$$\bar{P}'(x,\mathbf{q},0) = \bar{f}(x,\mathbf{q}), \tag{B7}$$

with $\bar{f}(x,\mathbf{q}) \equiv \bar{f}(\mathbf{R})$ [cf. (24)] a coarse-scale analog of the pointwise initial distribution (19), thereby completes the formulation of the problem posed for the modified aerosol concentration $\bar{P}'$.

It is easily established by integration of (B4) over the filter volume $\{\mathbf{q}_0\} \cup \{-\infty < x < \infty\}$ using boundary conditions (B5) and (B6) that

$$\int_{-\infty}^{\infty}\mathrm{d}x\int_{\mathbf{q}_0}\bar{P}'(x,\mathbf{q},t)\mathrm{d}\mathbf{q} = \text{const.} \quad (t > 0). \tag{B8}$$

Use of (B8) together with (B3), (26a) and (27a) demonstrates that

$$\bar{K}_f = \bar{K}^*. \tag{B9}$$

The problem posed for $\bar{P}'$ by equations (B4)–(B7) does not explicitly entail any information pertaining to the aerosol diminution within the filter (other than that implicitly embodied in the macroscale transport coefficients $\bar{\mathbf{U}}^*$ and $\bar{\mathbf{D}}^*$); as such, the problem is identical to one already treated by Brenner (1982b) using generalized Taylor dispersion phenomena theory formalism. Although $\bar{U}_f$ and $\bar{D}_f$ may be explicitly calculated using the latter formalism, we will derive these coefficients by invoking the following simpler, albeit less rigorous, physical arguments. It may be shown (Brenner, 1982b) that for sufficiently long times (explicitly specified below) the spatial aerosol distribution within each filter cross-section $x=\text{const.}$ reaches the asymptotic state characterized by the condition

$$\mathbf{J}_q\bar{P}'=\mathbf{0}. \tag{B10}$$

The latter no-flux condition satisfies the requirement (B5) of impermeability of the filter walls to aerosol particles. It may be viewed as a generalization of the equilibrium Boltzmann distribution to the more general case where coupling exists between the cross-sectional and axial aerosol transport processes. Introduce into the latter equation the constitutive expression (B2b) [rewritten for $\mathbf{J}_q\bar{P}'$ with the use of (B3)] and pre-multiply both sides by $\bar{\mathbf{D}}_{qq}^{-1}$ to obtain

$$\nabla_q\bar{P}'=-\bar{\mathbf{D}}_{qq}^{-1}\cdot\bar{\mathbf{U}}_q\bar{P}'+\bar{\mathbf{D}}_{qq}^{-1}\cdot\bar{\mathbf{D}}_{qx}\cdot\nabla_x\bar{P}'. \tag{B11a}$$

Substitution of this expression for $\nabla_q\bar{P}'$ into equation (B2a) [rewritten for $\mathbf{J}_x\bar{P}'$] thereby yields

$$\mathbf{J}_x\bar{P}'=(\bar{\mathbf{U}}_x-\bar{\mathbf{D}}_{xq}\cdot\bar{\mathbf{D}}_{qq}^{-1}\cdot\bar{\mathbf{U}}_q)\bar{P}'-(\bar{\mathbf{D}}_x-\bar{\mathbf{D}}_{xq}\cdot\bar{\mathbf{D}}_{qq}^{-1}\cdot\bar{\mathbf{D}}_{qx})\cdot\nabla_x\bar{P}'. \tag{B11b}$$

Upon comparison of the latter equation with the effective convective-dispersive constitutive equation†

$$\mathbf{J}_x\bar{P}'=\mathbf{i}_x\bar{U}_f\bar{P}'-\bar{D}_f\nabla_x\bar{P}' \tag{B12}$$

for the axial flux, one obtains the following expressions for the axial transport coefficients $\bar{U}_f$, $\bar{D}_f$:

$$\bar{U}_f=\mathbf{i}_x\cdot(\bar{\mathbf{U}}_x-\bar{\mathbf{D}}_{xq}\cdot\bar{\mathbf{D}}_{qq}^{-1}\cdot\bar{\mathbf{U}}_q) \tag{B13}$$

and

$$\bar{D}_f=\frac{\det\bar{\mathbf{D}}^*}{\det\bar{\mathbf{D}}_{qq}}. \tag{B14}$$

Moreover, for commonly encountered isotropic filter beds,

$$\bar{\mathbf{D}}_{qx}=\bar{\mathbf{D}}_{xq}=\mathbf{0}. \tag{B15}$$

This causes (B13) and (B14) to adopt the elementary forms

$$\bar{U}_f\equiv\mathbf{i}_x\cdot\bar{\mathbf{U}}^*\equiv\bar{U}_x^*,\quad\bar{D}_f\equiv\bar{D}_{xx}. \tag{B16a, b}$$

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† In fact, the axial flux equation (B12) describes the constitutive relationship existing for the asymptotically long-time, coarse-scale, 'pointwise' aerosol concentration $\bar{P}'$. This contrasts with the comparable axial flux constitutive equation implicitly embodied in equation (25) pertaining only to the cross-sectionally averaged concentration $\bar{P}_f$.