THE EFFECTS OF ELECTROSTATIC AND INERTIAL FORCES ON THE DIFFUSIVE DEPOSITION OF SMALL PARTICLES ONTO LARGE DISKS: VISCOS AXISSYMMETRIC STAGNATION POINT FLOW APPROXIMATIONS

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Abstract—Fine particles can deposit on microelectronic parts during their manufacture and cause damage. Prediction of the rates of deposition is important in planning to reduce such losses. Analytic expressions were developed and Monte Carlo simulations (Brownian dynamics) were carried out to predict deposition rates of sub-micron particles in viscous axisymmetric stagnation point flow. Diffusive, inertial and electrostatic deposition mechanisms were incorporated in the analysis. The electrostatic forces considered included the Coulombic force on a charged particle in an electric field and the image force on a charged particle near a conducting plane. Analytical solutions to the convective-diffusion equation, in the absence of inertial effects, showed that for a Boltzmann charge equilibrium, the electrostatic image force did not appreciably enhance deposition for a free-stream velocity of 50 cm s$^{-1}$. However, the Coulomb electrostatic attraction of a charged particle towards a charged surface greatly enhanced deposition, assuming a field from the disk approximately equivalent to having a disk 20 cm in diameter (80 pF capacitance) at 2000 V. Some areas for future study are suggested by these results. Brownian dynamics simulations carried out for particles of the order of 1 $\mu$m in diameter at a much higher velocity (30 m s$^{-1}$) demonstrated an interaction between inertial and diffusive deposition that produced inertially enhanced deposition at Stokes values below the critical value, $St < 0.15$. Where inertia and electrostatic effects were negligible, the simulations matched the known solution for stagnation flow deposition due to diffusion. In the Appendix is presented a derivation of the Langevin equation governing single-particle motion in a fluid that is not uniform in temperature as well as in velocity.

NOMENCLATURE

These variables appear frequently in the text. Other variables are defined when they are first introduced:

a Potential flow constant in the velocity field for stagnation point flow
B Inverse mobility tensor
c Dimensionless concentration $c^*/c_0$
c_0 Reference concentration
D Einstein diffusion coefficient
F* External force
rpF*/kT Dimensionless external force
p* Local fluid velocity
Pe Peclet number
Rep Particle Reynolds number $(ar^2/v)$
rp Particle radius
Sc Schmidt number $(v/D)$
Sh Sherwood number $(dc/dz)|_{z=1+\delta}$
r* Radial coordinate
u* Fluid velocity field in the absence of particles
z* Axial coordinate
v Kinematic viscosity

1. INTRODUCTION: MICROCONTAMINATION

A major impact of contamination in the microelectronics industry is the failure of microelectronic parts due to the disruption of conductive paths by insulating particles, or the penetration of insulating regions by conductive particles, or the chemical contamination of small regions, due to unwanted material from particles. Contamination problems are not limited to the microelectronics field, but they are especially important to it. We seek a better understanding of particle deposition processes in order to minimize 'microcontamination.'

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Particles as small as one-tenth of the size of the features on a microelectronics chip can cause failures, and as these features become a micrometer and smaller in dimension, particles a tenth of a micrometer—and, eventually, even smaller particles—can be of concern. Such particles can be expected to deposit due to Brownian motion (diffusion) as well as to electrical and gravitational forces and the effects of interception and inertia. The analysis of such deposition requires consideration of these deposition factors in the context of a fluid velocity field appropriate to the physical conditions of interest. As particles approach within a few particle diameters of the surface on which they may deposit, the fluid resistance to their motion will increase, perhaps significantly. Furthermore, as dimensions in the problem become small compared to the mean free path of the gas molecules, alterations to the continuum approach may be needed.

Prediction of the rates of deposition of sub-micron particles under various conditions are useful in indicating the likely magnitude of particle contamination. The same methodology can be used to predict the outcomes of various approaches to lessening particle deposition. Before the predictive methods can be used this way, however, a set of conditions appropriate to the application need to be identified, and the predictive methods need to be tested, where possible, against data or verified theoretical results. This paper describes the use of both analytic theories and computer experiments (Brownian dynamics method) for predicting the deposition rate of Brownian particles onto planar surfaces under conditions of interest in contamination control in the microelectronics industry. The test conditions and outcomes, however, are presented in terms of dimensionless groups that have a far wider range of applicability than just the application that was our focus.

Our focus was the deposition of particles 0.01–1.0 μm dia on a disk 20 cm in diameter exposed to air at atmospheric temperature and pressure flowing towards the surface at 50 cm s⁻¹. This is one of the conditions that a silicon wafer with microelectronic chip structures on it would face in a manufacturing clean room. In studying the electrical interactions, we used the assumption of Boltzmann equilibrium charge distribution, and Table I gives the r.m.s. number of elementary charges expected on particles of the diameters we studied.

For simplicity in analyzing the relative effects of the numerous particle forces involved, we chose a simplified geometry of a thin circular disk lying flat on an infinite planar surface. The fluid velocity profile was taken to be that for axisymmetric viscous stagnation point flow (Schlichting, 1979), with the center of the disk at the stagnation point. It was further assumed that the disk-planar surface constituted an infinite conducting planar surface. Consequently, a charged particle near the conducting surface experiences a force due to any applied field (E₀) present and additionally a force due to the electric field induced by the particle charge (image force). Higher-order interactions also exist due to an induced dipole in the conducting particle and due to interactions of the induced dipole with gradients in the electric field (Hartmann et al., 1976; Kraemer and Johnstone, 1955). In the results presented here we have considered only the applied field force and the charged particle image force, and we have neglected the higher-order interactions.

<table>
<thead>
<tr>
<th>Diameter (μm)</th>
<th>Uncharged fraction</th>
<th>r.m.s. No. charges</th>
<th>Average absolute No. charges*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01</td>
<td>0.993</td>
<td>0.08</td>
<td>0.007</td>
</tr>
<tr>
<td>0.02</td>
<td>0.892</td>
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<td>0.05</td>
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<tr>
<td>0.10</td>
<td>0.423</td>
<td>0.94</td>
<td>0.67</td>
</tr>
<tr>
<td>0.20</td>
<td>0.299</td>
<td>1.34</td>
<td>1.00</td>
</tr>
<tr>
<td>0.50</td>
<td>0.189</td>
<td>2.11</td>
<td>1.64</td>
</tr>
<tr>
<td>1.00</td>
<td>0.134</td>
<td>2.99</td>
<td>2.34</td>
</tr>
</tbody>
</table>

* From Hinds (1982), for T = 293 K.
In the absence of particle inertial forces, the convective-diffusive deposition of particles onto surfaces can be studied through solutions to the Smoluchowski equation. Furthermore, because of the simplicity in the geometric arrangement assumed (axisymmetric stagnation point flow), various analytical solutions to the Smoluchowski equation exist, depending on the form of the particle-surface interaction force. In Section 2 we present these solutions in terms of the particle surface flux or deposition rate, including the particle-surface interaction forces of interest in contamination control.

With the additional factor of particle inertial forces, and for more complicated geometries, Brownian dynamics methods offer a convenient way of studying the particle deposition problem (Gupta and Peters, 1985, 1986). In Section 3 we present the results of these computations, including verification of the Brownian dynamics method by comparison with the solutions presented in Section 2.

2. ANALYTICAL THEORIES FOR PARTICLE DEPOSITION IN VISCOUS STAGNATION POINT FLOW

In the absence of inertial effects, the local concentration of particles near a collector or adsorbing surface is governed by the convective-diffusion equation, which can be written as

$$\frac{\partial c^*}{\partial t^*} + \mathbf{V}^* \cdot (B^{-1} \cdot \mathbf{p}^* \cdot c^*) = D \mathbf{V}^* \cdot (B^{-1} \cdot \mathbf{V}^* \cdot c^*) - \mathbf{V}^* \cdot (B^{-1} \cdot F^* D/kT \cdot c^*).$$

(1)

In the above equation, $D$ is the Einstein diffusion coefficient, $F^*$ is the external force acting on a particle, and $B^{-1}$ is the so-called mobility tensor. In the absence of hydrodynamic interactions between the particle and the surface we have that

$$B^{-1} = I$$

(2a)

and

$$\mathbf{p}^* = \mathbf{u}^*,$$

(2b)

where $I$ is the unit tensor and $\mathbf{u}^*$ is the fluid velocity vector in the absence of particles. The fluid velocity fields in the absence of particles for axisymmetric viscous stagnation point flow are given in Schlichting (1979) as

$$u^* = ar(d\phi/d\zeta)$$

(3a)

$$u^*_\zeta = -2av^{1/2}\phi,$$

(3b)

where $a$ is the constant appearing in the corresponding potential flow field solution and $\zeta = (a/v)^{1/2}z$. Near the collector surface the function $\phi$ may be approximated by

$$\phi \approx (1/2)\zeta^2.$$  

(4)

For axisymmetric and plane stagnation point flow, expressions for the mobility tensor and convective velocity of a particle, including hydrodynamic interactions between the particle and the planar surface, are summarized in Dabros and van de Ven (1983) and Chari and Rajagopalan (1985a). These hydrodynamic interactions extend over particle-surface separations of several particle diameters and have been shown to significantly affect colloidal particle deposition rates (Dabros and van de Ven, 1983; Chari and Rajagopalan, 1985a,b; and references cited therein). In the case of aerosols, however, the assumptions of a fluid continuum made in the evaluation of hydrodynamic interactions break down as the separation distance becomes comparable with the gas mean free path, hundredths of a micrometer at standard conditions. Further, the errors involved in applying the above-cited expressions for hydrodynamic interactions at small separation distances in a gas medium are amplified due to the exponential growth of these functions at close separation distances. For simplicity, we neglect hydrodynamic interactions in the analytical solutions to equation (1) presented below.

Introducing the dimensionless quantities $u = u^*/u_0$, $V = V^*/v_0$, neglecting the time derivative, and using equation (2), one can re-write equation (1) as

$$PeV \cdot (uc) = V^2c - V \cdot (Fc),$$

(5)
where \( Pe \) is the Peclet number \( (Pe = \ell_0 u_0/D) \) and \( F \) is the dimensionless external force \( (F = F_0 F^*/kT) \). An important simplification for mass transfer in stagnation point flow arises when the Schmidt number \( (Sc = \nu/D) \) is large, as is common in colloids and aerosols. Under such conditions the diffusion boundary layer thickness is uniform over the entire absorbing surface (Schlichting, 1979; Evans, 1968).

In reporting analytical solutions to the one-dimensional convective-diffusion equation for axisymmetric stagnation point flow \( (Sc \gg 1) \) it has been common to choose \( \ell_0 = r_p \), the particle radius, and \( u_0 = 2r_p^2a^{3/2}v^{-1/2} \), so that

\[
Pe = 2Re_p^{1/2}Sc,
\]

where \( Re_p \) is a particle Reynolds number \( (Re_p = ar_p^2/\nu) \). Of course, the choice \( \ell_0 = r_p \) is not an appropriate axial length scale and, for example, small values of \( Pe \) as defined by equation (6) do not necessarily imply negligible convective effects.

Using equation (3), and for \( Sc \gg 1 \), the one-dimensional problem in the infinite plane can be written from equation (5) as

\[
d^2c/dz^2 = a_1(z) dc/dz + b_1(z)c,
\]

where

\[
a_1(z) = -Pez^2/2 + F_z
\]

\[
b_1(z) = dF_z/dz.
\]

Solutions to equation (7) are subject to the boundary conditions

\[
c \rightarrow 1 \text{ as } z \rightarrow \infty, \text{ for all } r; \tag{10a}
\]

\[
c = 0 \text{ for } z = 1 + \delta, \text{ for all } r, \tag{10b}
\]

where \( \delta \) represents a small dimensionless distance (typically a few Angstroms, divided by the particle radius) at which Born repulsive forces prevent actual particle contact. (See the discussions in Dabros and van de Ven, 1983, and Chari and Rajagopalan, 1985a.)

Equation (7) can be solved numerically for various types of external forces (Dabros and van de Ven, 1983; Chari and Rajagopalan, 1985b). However, analytical solutions exist for the case of no external forces (Levich–Smoluchowski approximation; see the discussion in Dabros and Adamczyk, 1979), and for uniform, purely attractive forces (Chari and Rajagopalan, 1985a). Before passing to summarizing these solutions, we note that for short-range particle–surface interaction forces (i.e. a characteristic force length much less than the diffusion layer thickness) the so-called surface reaction model can be used for approximate analytical solutions (Prieve and Ruckenstein, 1976, and the references cited therein). The external field and image forces of interest here, however, extend throughout the diffusion layer, and the surface reaction model is not appropriate.

Solution for negligible interaction forces (Levich–Smoluchowski approximation)

It is of interest to compare the solutions to be given below, including various force combinations, to the zero-force interaction case. Under such conditions we set \( F_z = 0 \) in equations (8) and (9), and equation (7) admits a simple analytical solution. The concentration profile and flux for the zero-force interaction case can be readily shown to be (Levich, 1966):

\[
c = \int_{1+\delta}^{z} \exp \left( -Pez^3/6 \right) dz' \int_{1+\delta}^{z} \exp \left( -Pez^3/6 \right) dz'.
\]

\[
dc/dz = \exp \left( -Pez^3/6 \right) \int_{1+\delta}^{z} \exp \left( -Pez^3/6 \right) dz'.
\]

The denominator on the right-hand side of equations (11) and (12) can be integrated exactly if we assume \( \delta = 0 \) and neglect the so-called interception effect (tantamount to taking the lower integration limit as zero). With these simplifying assumptions, equations (11) and
Diffusive deposition of small particles onto large disks

(12) become
\[
c = \int_0^z \exp(-\alpha z^3/6) dz / 0.892993(6/\text{Pe})^{1/3}
\]
\[
dc/dz = \exp(-\alpha z^3/6) / 0.892993(6/\text{Pe})^{1/3}.
\]

Also, with the Sherwood number defined as
\[
Sh = (dc/dz)|_{z=0}
\]
we obtain from equation (14)
\[
Sh = 0.616\text{Pe}^{1/3}
\]
or, in terms of the particle Reynolds number and Schmidt number
\[
Sh = 0.776\text{Re}^{1/2}\text{Sc}^{1/3}.
\]

**Exact solution for a constant external force**

For a uniform external force, equation (7) can be solved exactly. The result in terms of the Sherwood number is (Dabros and van de Ven, 1983):
\[
Sh = \exp(-\alpha z^3/6 + zFz_0) / z = 1 + 6 \exp(-\alpha z^3/6 + z'Fz_0) dz',
\]
where \(Fz_0\) is a uniform (dimensionless) external force. An example of this type of force is a uniform applied electric field force acting on a particle of charge \(q\): \(F^* = qE\).

**Solution for attractive particle–surface interaction forces**

Chari and Rajagopalan (1985a) used the so-called JWKB technique to obtain approximate solutions to equation (7) for the particle concentration field and associated mass flux under attractive particle–surface interaction forces. Their analysis, as given, is restricted to attractive interaction forces which have the property \(F_z \to 0\) for \(z \to \infty\).

An example of this type of force is the charged particle image force discussed above. The final expression for the deposition rate, neglecting hydrodynamic corrections, can be written as
\[
Sh = 2E_1(-k(z))^{1/4} \exp(-\alpha z^3/12 - \Phi(z)/2)|_{z=1 + \delta},
\]
where
\[
Sh = dc/dz|_{z=1 + \delta},
\]
\[
F_z = -d\Phi/dz
\]
\[
k(z) = -\alpha z/2 - (1/2)dF_z/dz - (1/4)(-\alpha z^3/2 + F_z)^2
\]
\[
E_1 = (4)^{-1/3} \exp(-m(\infty) + W(z = 1 + \delta; \text{Pe}) - 1/3)\text{Pe}^{1/6}
\]

and, in equation (23),
\[
m(\infty) = \int_{1 + \delta}^{\infty} ((-k(z))^{1/2} - (-k'(z))^{1/2}) dz
\]
\[
k'(z) = (-\alpha z/2)(1 + \alpha z^3/8)
\]
\[
W(z; \text{Pe}) = (1/3)(z^{3/2}z(z) - \ln ((z(z) - v(z))/(\alpha(z) + v(z))))
\]

where
\[
\alpha(z) = (0.5\text{Pe} + 0.0625\text{Pe}^2z^3)^{1/2}
\]
\[
v(z) = 0.25\alpha(z)^{3/2}.
\]
Once the Peclet number and external force acting on a particle are specified, equations (19)-(28) can be used to calculate the Sherwood number. The solution is restricted to \( k(z) \) being a large, slowly varying function. Note that the Peclet number differs by a factor of 2 from that of Chari and Rajagopalan (1985a), who treated the problem of plane (not axisymmetric) stagnation flow.

**Specific results for a constant applied force and a particle electrostatic image force**

In order to examine the effects of particle electrostatic image and applied field forces on particle diffusive deposition rates, a test system was selected whose characteristic values are given in Table 2. Results of calculations of the Sherwood number for the constant applied field force and the particle image force are displayed in Figs 1–3. Note that in Figs 1–3 the particle diameter was varied from 0.05 \( \mu m \) at the smallest Peclet number shown to 1.00 \( \mu m \) at the largest Peclet number. The diffusion coefficient \( D \) was taken as \( D = kTC/6\pi\mu r_p \), where \( C \) is the Cunningham slip correction [see equation (31)].

Figure 1 shows the effect of a uniform applied electric field on the particle deposition rate. Here we have considered only the attraction of negatively charged particles with charge magnitude equal to one-half the average absolute charge from the Boltzmann distribution (Table 1). (One-half of the average absolute charge values was used because the force is proportional to the charge and is attractive only for one charge polarity.) The applied field

| Table 2. Values of the constants employed for the particle diffusive deposition calculations |
|---------------------------------|------|
| Constant | Value |
| \( a \) | 12.5 \( s^{-1} \) |
| \( T \) | 298 K |
| \( \varepsilon \) | 6.349 \( \times 10^{-8} \) cm |
| \( \mu \) | 1.8137 \( \times 10^{-4} \) g cm\(^{-1}\) s\(^{-1} \) |
| \( \mu' \) | 1.1843 \( \times 10^{-3} \) g cm\(^{-3} \) |
| \( \delta \) | 0.0 |

Fig. 1. Sherwood number (deposition group) vs Peclet number (convective-diffusion group) with and without a uniform electric field.
Diffusive deposition of small particles onto large disks

Fig. 2. Sherwood number vs Peclet number with electrostatic force group \( (K_e) \) as a parameter.

Fig. 3. Sherwood number vs Peclet number with and without electrostatic image force.

corresponds to a surface charge density of 30 elementary charges \( \mu m^{-2} \) or, roughly, a potential of 2000 V across a thin disk 20 cm in diameter and having a capacitance of 80 pF. As seen in Fig. 1 an applied field can have a dramatic effect on particle deposition rates; in this case, the field increased the Sherwood number (dimensionless deposition group) by two orders of magnitude for the negatively charged particles of the Boltzmann distribution.

In Fig. 2, we have given values of the deposition rate for a range of electric field forces expressed in terms of the dimensionless force \( F = K_e = qE_0r_p/kT \). Note that for values of
for the range of $Pe$ shown, the external force controls the deposition rate. In fact, equation (18) shows that for $|F| \gg Pe^{2/6}$, we have $Sh \approx |F|$.

The effects of the particle image force on the deposition rate are shown in Fig. 3. The image force is proportional to the square of the charge on the particle and inversely proportional to the square of twice the distance from the particle center to the surface. Since the particle image force is always attractive, we initially assumed that the particle charge is constant at the Boltzmann r.m.s. value (see Table 1) in analyzing the effects of this force. It was found, however, that for this level of particle charge the function $k(z)$ in equation (22) was not large throughout the region of interest. Therefore, twenty times the r.m.s. charge was used in generating the results shown in Fig. 3. As seen in Fig. 3, the particle image force, at the assumed level of charge, results in as much as an order of magnitude increase in the Sherwood number over the non-interacting (Levich–Smoluchowski approximation) for a given particle diameter.

In order to quantify further the effects of the particle image force on the Sherwood number, equation (7) was solved numerically. The differential equation in this case can be stiff, and thus a combined Gear's method and ‘shooting’ method was used (Chari and Rajagopalan, 1985b). A comparison of the numerical solution to the approximate analytical solution, at twenty times the Boltzmann r.m.s. charge on the particle, is shown in Fig. 3. As expected, the best agreement is obtained at larger Peclet numbers, where the function $k(z)$ defined by equation (22) is larger throughout the diffusion path. Numerical solutions obtained for the Boltzmann r.m.s. charge on the particle showed negligible differences with the zero-force case. This is also expected from the results shown in Fig. 3, since the image force is proportional to the square of the charge, and thus the force is two orders of magnitude smaller at the r.m.s. charge.

3. BROWNIAN DYNAMICS SIMULATIONS OF PARTICLE DEPOSITION ON DISKS

When particle sizes approach the micron and sub-micron range, the abrupt momentum changes that a particle experiences from collisions with surrounding fluid molecules, on time scales of the order of picoseconds or smaller, cannot be neglected. In the absence of particle–particle interactions and particle inertial forces, the macroscopic manifestation of this phenomenon is the convective-diffusion equation for the particles given by equation (1). A more general approach is to begin with an equation of motion for the particles which includes the ‘random’ force due to particle-fluid molecule collisions, the so-called Langevin equation. In general, the derivation of the Langevin equation is not a trivial matter. In general, the Langevin equation can be obtained from the Fokker–Planck equation and vice-versa (Chandrasekhar, 1945). The Fokker–Planck equation, in turn, is a contracted form of the Liouville equation, written for the particles and fluid molecules present in the system. Following this approach, we have given in the Appendix a derivation of the single-particle Langevin equation which includes fluid-molecular based expressions for the frictional and ‘random’ force terms, as well as the effects of non-uniformities in the host fluid due to temperature and velocity variations. This interpretation is of fundamental importance in aerosols, where gas rarefaction effects can lead to deviations from continuum fluid mechanical theories as discussed in Section 2.

Assuming a linear form for the frictional force term (valid at small particle Reynolds numbers), the single-particle Langevin equation can be written as (Appendix)

$$\frac{dv}{dt} + \beta(v - u_0) = \frac{F}{m_p} + A(t),$$

(29)

where $m_p$ is the mass of a single particle with velocity $v$; $\beta$ is the friction coefficient divided by the particle mass; $u_0$ is the undisturbed fluid velocity (i.e. in the absence of particles); $F$ is an external force acting on the particle; and $A(t)$ is the random or Brownian force per unit mass.

In the absence of hydrodynamic interactions between the particle and the adsorbing surface we have

$$\beta = \frac{6\pi \mu r_p}{C m_p},$$

(30)
where \( C \) is the Cunningham correction to Stokes law to account for the particle 'slip' through a molecular fluid, commonly expressed as

\[
C = 1 + (2\zeta/r_p)(2.514 + 0.800 \exp(-1.10r_p/\zeta)).
\]  

The integrated forms of the Langevin equation, equation (29), are given in the Appendix [also see Chandrasekhar (1945), Ermak and Buckholtz (1980), among others]. Application of these results to particle adsorption onto spherical surfaces has also been accomplished (Gupta and Peters, 1985, 1986). Here, we apply the integrated forms of the Langevin equation to the problem of particle adsorption onto a circular disk.

Figure 4 illustrates the physical features of the Brownian dynamics method as applied to the disk deposition problem. A disk of radius \( r_c \) is located on the surface of an infinite plane. The fluid flow is assumed to be viscous axisymmetric stagnation point flow. At a point \( z_0^* \) from the disk surface the particle concentration is \( C_0 \) and the fluid velocity in the \( z \)-direction is \( u_{z_0} \). A disk collection or target efficiency is defined as

\[
\eta = \frac{\langle r_{lim} \rangle^2}{r_c^2},
\]

where \( \langle r_{lim} \rangle \) is the average maximum \( r \)-distance, at the plane \( z^* = z_0^* \), which results in particle 'capture' by the disk. This value is determined by following the trajectories of Brownian particles, according to the integrated Langevin equation, beginning at the plane \( z^* = z_0 \) (for further details refer to the references cited above).

It is of interest to compare the Brownian dynamics results with the various solutions to the convective-diffusion equation for particle adsorption onto a large disk as given in the previous section. A relationship between the collection efficiency and surface concentration gradient, or Sherwood number, is therefore sought. Referring to Fig. 4, the total particle number flux through the circular surface with radius \( \langle r_{lim} \rangle \), located far from the surface where the diffusive flux of particles is negligible, can be written as

\[
N = \int_{\text{disk}} j \cdot dS = C_0 (u_{z_0} + F_{z_0} D/kT) \pi \langle r_{lim} \rangle^2,
\]

where we have included the possibility of a constant \( z \)-directional external force, \( F_{z_0} \). It is this total particle number flow which results in deposition onto the disk; hence,

\[
N = \int_{\text{disk}} j \cdot dS = D \int_0^{2\pi} \int_0^{r_c} (dc*/dz*)_{z^* = r_c (1 + \delta F^*)} d\theta.
\]

If the concentration gradient is independent of the radial coordinate, the above equation can be written as

\[
N = \left( Dc_0/r_p \right) (dc/dz)_{z = 1} + \delta (\pi r_c^2) = Sh(Dc_0/r_p)(\pi r_c^2).
\]  

\[\text{Fig. 4. Schematic of physical features of Brownian dynamics simulation.}\]
Combining equations (33) and (35), we obtain the desired relationship between the average disk target efficiency and the Sherwood number as

$$\eta = \frac{Sh(D/r_p)}{(u_\star^2 + F_\star D/kT)}.$$  \hspace{1cm} (36)

Thus, from values of $\langle r_{lim} \rangle$, obtained from Brownian dynamics, the Sherwood number can be obtained from equation (36).

In order to test the Brownian dynamics results against known analytical solutions, simulations were carried out in the non-inertial regime. The 'starting' plane, $z^* = z_0$, was selected to be one collector radius upstream from the disk surface. A disk diameter of 2 cm was selected for all computations given here, with a particle density chosen to be 3 gm cm$^{-3}$. Remaining constants were the same as those given in Table 2. With these parameters fixed, the particle radius was varied (and thus the particle Peclet number, $Pe$). The integration time step was $10^{-6}$ s.

Figure 5 shows the Brownian dynamics results obtained in the absence of external forces. The average values shown are based on ten repetitions of the 'experiment'. The error bars shown correspond to 90% confidence intervals for the means of the computer-generated data. The Peclet number variations in Fig. 5 correspond to particle diameter variations from 0.01 to 0.1 μm, well within the diffusion regime. As seen from Fig. 5, there is good agreement between the Brownian dynamics calculations and the Levich–Smoluchowski solution, equation (16).

One strength of the Brownian dynamics technique is that it allows combining diffusive deposition with inertial effects. Although inertial effects are not expected to be appreciable for particles having aerodynamic diameters a few microns or less in a flow of 50 cm s$^{-1}$ past objects a centimeter in dimension or larger, inertial effects might be appreciable in flows of a higher velocity, 30 m s$^{-1}$ for example, that could occur in impactors or in certain processing equipment.

Brownian dynamics simulations were run for particles about 3 μm in diameter and smaller for viscous axisymmetric stagnation point flow, with a flow velocity of $u_{\infty} = 30$ m s$^{-1}$, at a distance $z_0 = 1$ cm from the disk surface. The conditions are as in Table 2, except that $a = 1500$ s$^{-1}$ rather than 12.5 s$^{-1}$.
Figure 6 shows the Sherwood number vs Stokes number, \( St = \beta^{-1} u_{x_0}/2r_c \), with the inertialess results (Levich–Smoluchowski) given by dashes and the results with inertia included and calculated by Brownian dynamics (Monte Carlo simulation) given by solid dots. Effects due to inertia are evident even at \( St < 0.1 \).

Figure 7 shows \( Sh \) vs \( St \) in the vicinity of the critical value of \( St \) for inertial deposition. The solid dots are from the Brownian dynamics simulation. Dashed lines mark the diffusion
contribution without inertia. Solid lines mark the inertial contribution without diffusion, calculated for inviscid flow using equations from Ranz and Wong (1952), and calculated for viscous flow using our Brownian dynamics program without the diffusive contribution. The critical $St$ value for viscous flow we found to be $0.153 < St < 0.155$, higher than for inviscid flow. Note that the combination of diffusion and inertia produces much more deposition than would be expected from the sum of their individual contributions. The deposition is not additive, but synergistic.

The results shown in Figs 6 and 7 are in qualitative agreement with the inertia-diffusion studies of Fernandez de la Mora and Rosner (1981), which concerned deposition in plane stagnation point flow. The results of increased deposition at Stokes numbers below the critical $St$ over the zero-inertia values of deposition are qualitatively due to particle concentration increases ('inertial particle enrichment') outside the diffusion boundary layer. Figure 7 shows that dramatic increases in Sherwood number over the zero-inertia case are theoretically possible.

4. DISCUSSION

It should be mentioned that viscous stagnation point flow is often a good local approximation for a variety of flow situations. The two-dimensional version is applicable to the stagnation region caused by the impingement of a jet from a slit onto a flat plane and the stagnation region on the leading surfaces of long objects in a flow, where the object lies perpendicular to the flow. The three-dimensional version is applicable to the stagnation point region from the impingement of a jet from a round opening onto a flat plane and the stagnation point region at the leading surface of round objects such as spheres or disks. The flow strength, $a$, to be used is approximately the characteristic flow velocity divided by the characteristic flow dimension. For uniform flow of velocity $u^*$ toward a disk of radius $R^*$, the matching of the flow near the stagnation point with the viscous stagnation point flow model gives a value for the flow strength of (Cooper et al., 1989):

$$a = 2u^* / \pi R^*. \quad (37)$$

There are natural extensions to the work presented here: improvement to the stagnation point model, extension to other geometries and flow conditions, refinement of the particle–surface interactions. It would be valuable to extend the Brownian dynamics studies to the incorporation of electrostatics in the inertia-diffusion regime. It would be of practical interest to extend the perturbation solution of Fernandez de la Mora and Rosner in the inertia-diffusion regime to include electrostatic forces in viscous axisymmetric stagnation point flow. For practical applications, it would also be worthwhile to extend the calculations to the cases of finite disk perpendicular to the flow and finite disk parallel to the flow to compare with the analyses of Liu and Ahn (1987). Further characterization of the particle–surface interaction might include van der Waals attractive and Born repulsive forces, the longer-range forces of thermophoresis and perhaps diffusiophoresis, and more accurate determination of the resistance of the medium to the motion of the particle towards the surface at small distances from the surface.

5. CONCLUSIONS

In many cases of practical and theoretical interest, computer-intensive Brownian dynamics simulations need not be used to obtain solutions. Analytic solutions exist for many situations, and for others it is possible to obtain numerical solutions by numerical integration of differential equations. These non-simulation approaches allow solving for inertialless diffusive deposition in the presence of a uniform electric field, for example, for axisymmetric viscous stagnation point flow to the infinite plane as studied here.

The Brownian dynamics approach is an attractive method for the case where inertia is appreciable and so is diffusion. Our results show that inertia can enhance deposition with diffusion well below the critical Stokes parameter associated with inertial deposition without
diffusion. This would tend, for example, to make impactor collection efficiency vs Stokes inertial parameter less like a step function than would be predicted either ignoring diffusion or combining diffusion additively with impaction. Future work is planned to address the problem of having electrostatic forces and diffusion and inertia be significant simultaneously.

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APPENDIX. DERIVATION OF THE SINGLE-PARTICLE LANGEVIN EQUATION FOR A BROWNIAN PARTICLE IN A NON-UNIFORM HOST FLUID

The Langevin equation for a Brownian particle, or a system of interacting Brownian particles, can be obtained from the Fokker–Planck equation for the particles (Chandrasekhar, 1945; Ermak and McCammon, 1978; Ermak and Buckholtz, 1980). Furthermore, as shown, for example, by Murphy and Aguirre (1972) and Mazo (1969), the Fokker–Planck equation, in turn, can be obtained by contraction of the Liouville equation written for the system of fluid molecules and Brownian particles. A molecule-based Fokker–Planck equation has also been derived recently by Fernandez de la Mora and Mercer (1982), beginning with a two-component Boltzmann equation. The fundamental equivalence of this latter approach to the previously mentioned methods, based on direct contractions of the Liouville equation, is evident from derivations of the Boltzmann equation from the Liouville equation (see, for example, Andrews, 1962).

Following Fernandez de la Mora and Mercer (1982), we write the Fokker–Planck equation for a single, Brownian particle, as:

\[ \frac{\partial f^*}{\partial t^*} + v^* \cdot \nabla f^* = \frac{\beta}{\theta^*} (\nabla f^*) \cdot (v^* - D T \nabla \ln T^* f^* - u^*) f^* + (kT/m_p) \frac{\partial f^*}{\partial \theta^*} + m_p f^* \frac{\partial f^*}{\partial \theta^*} f^*, \]  

where \( f^* \) is the single-particle distribution function, \( v^* \) is the particle velocity at position \( x^* \), \( \beta \) is the Stokes friction coefficient divided by file particle mass, \( D \) is the diffusion coefficient, \( \theta^* \) is the thermal diffusion factor (see the above-cited reference for more details), \( u^* \) is the 'undisturbed' host fluid velocity field (i.e. in the absence of particles), and \( \theta^* \) is the potential of the external force acting on the particle.

The dimensionless variables are defined as: \( f = f^*/f_0, r = r^*/r_0, v = v^*/v_0, T = T^*/T_0, u = u^*/u_0, \) and \( \theta = \theta^*/\theta_0 \). Introducing these into equation (A1) gives:

\[ \frac{\partial f}{\partial t} + v \cdot \nabla f = \left( \frac{1}{S} \right) \frac{\partial f}{\partial \theta} \left( (v - N_T \nabla \ln T \nabla \theta - N_v u) f + N_K \frac{\partial f}{\partial \theta} \right) + N_p \left( \frac{\partial f}{\partial \theta} \right) \frac{\partial f}{\partial \theta}. \]  

(A2)

The dimensionless groups are: \( \beta' = \beta/kT_0, T = D_T/(\xi v_0), N_T = u_0/v_0, N_v = D_T/v_0, N_K = kT_0/m_p v_0^2, N_p = \phi_0/m_p v_0^2. \)

Note that the Fokker–Planck equation is valid for time intervals much greater than the average collision time between the host fluid molecules, \( r_0 \) (Chandrasekhar, 1945). For time intervals much larger than \( r_0 \), but smaller than the time over which there are appreciable variations in the host fluid macroscopic velocity, \( u^* \), temperature field, \( T^* \), and external force potential, \( \theta^* \), the Fokker–Planck equation can be integrated exactly. Following Chandrasekhar (1945) (see also Ermak and Buckholtz, 1980), the solution to (A2) that satisfies the initial conditions

\[ f = \delta(r - r_0) \delta(v - v_0) \]  

at \( t = 0, \)

(A3)

where \( r_0 \) and \( v_0 \) are the dimensionless initial position and velocity of the particle, can be shown to be

\[ f = (4\pi^2(F - H^2))^{-3/2} \exp \left( - (GI R^2 - 2H(R \cdot S) + F(S^2) - 2(F G - H^2)) \right), \]  

(A4)
where

\[ R = r - r_0 - Stv_0(1 - \exp(-t/St)) - (N_r \partial \ln T/\partial \tau + N_r u - StN_r \partial \Phi/\partial \tau) (t - St(1 - \exp(-t/St))) \]  
(A5)

\[ S = v - v_0 \exp(-t/St) - (N_r \partial \ln T/\partial \tau + N_r u - StN_r \partial \Phi/\partial \tau) (1 - \exp(-t/St)) \]  
(A6)

\[ G = 3N_K (1 - \exp(-2t/St)) \]  
(A7)

\[ H = StN_K (1 - \exp(-t/St))^2 \]  
(A8)

\[ F = (St)^2 N_K ((2t/St) - 3 + 4 \exp(-t/St) - \exp(-2t/St)) \]  
(A9)

Since \( f \) is a bivariate Gaussian distribution, the particle velocity and displacement change over the time interval, \( t \), can be calculated from

\[ v = v_0 \exp(-t/St) + (N_r \partial \ln T/\partial \tau + N_r u - StN_r \partial \Phi/\partial \tau) (1 - \exp(-t/St)) + B_1 \]  
(A10)

\[ r = r_0 + Stv_0(1 - \exp(-t/St)) + (N_r \partial \ln T/\partial \tau + N_r u - StN_r \partial \Phi/\partial \tau) (t - St(1 - \exp(-t/St))) + B_2 \]  
(A11)

in which \( B_1 \) and \( B_2 \) are random vector functions that follow a bivariate Gaussian distribution with the following mean and variance properties:

\[ \langle B_1 \rangle = 0, \quad \langle B_1^2 \rangle = 3G \]

\[ \langle B_2 \rangle = 0, \quad \langle B_2^2 \rangle = 3F \]

\[ \langle B_1 \cdot B_2 \rangle = 3H \]  
(A12)

For a system with a uniform temperature, the dimensional form of the time-differential equation of (A10) is the equation of motion for the particle, equation (29).