Introduction

In recent years, microreaction systems made using combined microfabrication and chemistry technologies have found wider use in practice because of their advantages over conventional chemical reaction systems. The main advantages are that high rates of heat and mass transfer are possible in micro (or nano)-scale devices, leading to higher yields of products, and further provides new reaction pathways that are not possible in larger scale reactions [1].

One example of a microreaction system is the micro-packed bed reactors that are used for gas phase or gas-liquid phase reactions [2-8]. The packed bed reactors are designed to provide efficient contact between the gas and the catalysts (packings) or between the gas and the liquid. An important problem in this process is to determine the pressure drop across the bed.

Fluid flow through a packed bed of particles has been examined extensively in the literature, both theoretically and experimentally. Probably the first systematic approach was reported by Carman [9] who modeled the void space in the packed bed by using straight capillaries whose diameter was taken to be a function of both the volume fraction and the size of the particles. He proposed a correlation by adjusting the capillary model to fit empirical data. An additional empirical extension of Carman’s correlation for a wider range of Reynolds numbers is the so-called Ergun equation [10], which is widely used in practice for predicting the pressure drop across the bed. Later analytical efforts were directed at computing the pressure drop-velocity relationship for a well-defined geometry of packed beds of equal-sized spheres arranged in a periodic array [11-13]. The case of random arrays was considered by Ladd [14] and Mo and Sangani [15].

Our present study was concerned with determining the pressure drop across the micro-packed bed of spheres in random arrays by fluid flow. Fluid flow through the micro-packed bed satisfies the well-known Stokes equations of motion because the Reynolds number based on the diameter of the spheres and the average fluid velocity is small in practice. The Stokes equations of motion are solved for fluid flow past spheres in a random array using a multipole expansion method. The numerical simulation results are compared with the experimental results obtained by Ajimera and coworkers [2,4,5] and the estimations by simple correlations.

Formulation of the Problem and Method

We consider an incompressible fluid flow through a
micro-packed bed of spheres. The fluid velocity satisfies the well-known Stokes equations of motion and the continuity equation given by

\[ -\nabla p + \mu \nabla^2 \mathbf{u} = 0 \]  
\[ \nabla \cdot \mathbf{u} = 0 \]  

where \( p \) is the pressure in the fluid, \( \mathbf{u} \) the fluid velocity, and \( \mu \) is the viscosity of the fluid. To model an infinitely extended fixed bed of spheres, we followed the standard practice and assumed the bed to consist of a periodic array with each unit cell of the array containing \( N \) spheres whose positions were generated using a specified spatial distribution law. The above boundary conditions must be satisfied on the surface of each sphere. In addition, the fluid velocity \( \mathbf{u} \) must be spatially periodic. An additional constraint to be satisfied is

\[ \frac{-1}{\tau} \int \mathbf{u} \, dV = \mathbf{U} \]  

where \( \mathbf{U} \) is the superficial fluid velocity through the bed, \( \tau \) is the volume of the unit cell, and \( V_f \) is the volume occupied by the fluid within the fixed unit cell.

We used the method outlined in Mo and Sangani [15] for determining \( \mathbf{u} \). Briefly, the method consists of writing a formal solution of Stokes equations of motion in terms of derivatives of a periodic fundamental singular solution of Stokes equations. This formal solution containing a number of undetermined coefficients satisfies the periodicity and the governing Stokes equations of motion. The coefficients are subsequently determined by expanding the formal solution around the surface of each sphere and satisfying the boundary conditions on the surfaces of spheres.

The expansion near a representative sphere \( a \) is expressed in terms of spherical harmonics according to the well-known Lamb's solution [16]:

\[ \mathbf{u}(x) = \sum_{n=-\infty}^{\infty} \left[ (c_n \nabla^2 \mathbf{p}_n + b_n \mathbf{r} \mathbf{p}_n^9) + \nabla \times (\mathbf{r} x_n) + \nabla \Phi_n^9 \right] \]  

with \( \mathbf{r} = x - x_0 \) and

\[ c_n = \frac{n+3}{2(n+1)(2n+3)}, \quad b_n = \frac{-n}{(n+1)(2n+3)} \]  

Here, \( p_n, a_n, \) and \( x_n \) in (4) are the spherical harmonics of order \( n \). The harmonics of negative order are singular at \( r = 0 \), and we express them as

\[ p_n^{a-1} = \sum_{m=0}^{n} (P_{n}^{a,m} Y_{m} + P_{n}^{a,0} Y_{m}^0) r^{n-1} \quad (n \geq 0) \]  

In (6), \( P_{n}^{a,m} \) and \( P_{n}^{a,0} \) are the coefficients of the singular harmonics and

\[ Y_{m} = P_{n}^{a,m} (\cos \theta) \cos m\phi, \quad Y_{m}^0 = P_{n}^{a,0} (\cos \theta) \sin m\phi \]  

are the surface harmonics with \( P_{n}^{a,m} \) being the associated Legendre polynomial and \( \theta \) and \( \phi \) the polar and azimuthal angles defined by \( x_1 - x_1^0 = r \cos \theta, \quad x_2 - x_2^0 = r \cos \phi, \) and \( x_3 - x_3^0 = r \sin \phi \). The singular harmonics \( x_{-n-1} \) and \( e_{-n-1} \) are likewise expressed in terms of coefficients \( T_{n}^{a,-n}, T_{n}^{a,0}, \Phi_{n}^{a,-n}, \) and \( \Phi_{n}^{a,0} \).

The harmonics with non-negative \( n \) are expressed as

\[ p_n^a = \sum_{m=-\infty}^{\infty} (P_{n}^{a,m} Y_{m} + P_{n}^{a,0} Y_{m}^0) r^m \quad (n \geq 0) \]  

with similar expressions for \( x_n^a \) and \( e_n^a \).

To satisfy the boundary conditions for the velocity at \( r = a \), it is convenient to use

\[ \mathbf{u}_r = \sum_{n=-\infty}^{\infty} \left[ (n c_n + b_n) p_n + (n/r) \Phi_n \right] \]  
\[ \nabla_s \cdot \mathbf{u}_s = - \sum_{n=-\infty}^{\infty} n(n+1) [c_n \mathbf{p}_n + \Phi_n/r] \]  
\[ \mathbf{e}_r \cdot (\nabla \times \mathbf{u}_s) = \sum_{n=-\infty}^{\infty} n(n+1)x_n/r \]  

where \( \mathbf{u}_r \) is the radial component of the velocity, \( \mathbf{u}_s = u_\theta \mathbf{e}_\theta + u_\phi \mathbf{e}_\phi \) is the tangential velocity at the surface of the sphere, and

\[ \nabla = \mathbf{e}_r \frac{\partial}{\partial r} + \frac{1}{r} \nabla_s. \]  

Using the orthogonality of surface harmonics and the expressions (9) ~ 11), the boundary conditions at \( r = a \) yield

\[ [(n+1)c_{-n-1} + b_{-n-1}] P_{n}^{a,n+1} - n c_{-n} P_{n}^{a,n-1} - \sum_{m=-n}^{n} \Phi_{n}^{a,m} \Phi_{n+1}^{a,m+1} = P_{n+1}^{a,1} \]  
\[ -n(n+1) [ P_{n}^{a,n+1} + P_{n}^{a,n-1} - \Phi_{n}^{a,n+1} \Phi_{n}^{a,n-1} ] = 0 \]  
\[ -n(n+1) \left[ T_{n}^{a,n+1} + T_{n}^{a,n-1} \right] = T_{n+1}^{a,1} \]  

plus similar equations involving the coefficients of \( Y_{n}^{0} \).

The singular coefficients in the equations above represent the effect of other spheres and the imposed
flow. The terms on the right-hand side are given by the imposed flow. As mentioned earlier, the velocity can be expressed in terms of fundamental periodic singular solution of Stokes equations $V$ as

$$u(x) = U + \sum_{\alpha=1}^{N} G^{\alpha} \cdot v(x - x^\alpha),$$

where $G^{\alpha}$ is a differential operator defined in terms of singular coefficients, such as $P_{\gamma}^{\alpha}$ and $\Phi_{\gamma}^{\alpha}$, in such a way that $G^{\alpha} \cdot v(x - x^\alpha)$ corresponds exactly to the singular terms in (4) as $x \to x^\alpha$ [15]. The coefficients of the regular terms in Lamb’s solution, e.g., $P_{\gamma}^{\alpha}$ and $T_{\gamma}^{\alpha}$, are related to various derivatives of the regular part of $u$ at $x = x^\alpha$.

Equations (13) – 5) were rearranged and truncated to solve for these coefficients (e.g., $P_{\gamma}^{\alpha}$). The force on sphere $a$ in the $x_1$ direction is related to $P_{\gamma}^{\alpha}$ by

$$F^a = -4\pi \mu P_{\gamma}^{\alpha}$$

The results for the average force on a sphere are expressed in terms of coefficients $K$

$$F = 6\pi \mu a L K$$

where $K$ represents the non-dimensional drag on the sphere in a packed bed as a function of $\phi$, the volume fraction occupied by the spheres. The pressure gradient in the gas is related to the force by

$$-\frac{d\rho}{dx_1} = nF = \frac{3\phi}{4\pi a^2}$$

where $n$ is the number of spheres per unit volume of the bed.

### Results and Discussions

The results for the pressure drop, or equivalently $K$, for random as well as periodic arrays are given in Table 1. The results for random arrays were obtained by averaging over 20 configurations generated using a hard-sphere molecular dynamics code that employed 16 particles per unit cell. These results were in excellent agreement with the results obtained previously [14,15].

The drag coefficients $K$ obtained by numerical calculations, and some correlations given by Carman [9] and Ergun [10], are plotted in Figure 1 as a function of $\phi$. Carman’s correlation gives the following simple expression for $K$:

$$K = \frac{10\phi}{(1-\phi)^{3/2}}$$

More widely used in practical process design is the Ergun equation, which is an empirical extension of Carman’s correlation for a wider range of Reynolds numbers. Thus, the Ergun equation is reduced to Carman’s correlation when the Reynolds number is small, but the coefficient in the Ergun equation has been re-corrected from Carman’s original work to fit empirical data. For low-Reynolds-number flow, the Ergun equation can be written as

$$K = \frac{25\phi}{3(1-\phi)^{3/2}}$$

which is also called the Blake-Kozeny equation. We see that there is an excellent agreement between Carman’s correlation and the exact calculations at $\phi > 0.4$, whereas the Ergun equation slightly underpredicts.

We compared the rigorous calculations and estimations...
above with the experimental results for pressure drop across a micropacked bed by a gas (nitrogen) flow. The experiment was performed by Ajimera and coworkers [2,4,5] while working on designing a variety of microreaction systems. An example is their experiment for phosgene synthesis using a micro-packed bed reactor loaded with active carbon particles (60 µm in diameter) [2]. The dimensions of the packed bed are a width of 625 µm, a depth of 300 µm, and a length of 20 mm. Nitrogen gas was used for measuring the pressure drop across the bed. The nitrogen gas flow rates for the experiment ranged from 0 to 20 cm$^3$/min. The actual flow rates for the mixture of carbon monoxide and chlorine for phosgene synthesis were 4.5 and 8.0 cm$^3$/min. At these flow rates, the Reynolds numbers based on the particle radius were ca. 0.8 and 1.4, respectively. According to their paper [2], Ajimera and coworkers mentioned that they could obtain a good agreement between the experimental results and the predictions when using the Ergun equation for the pressure drop across the bed when the volume fraction of particles was set at ca. 0.6. However, specific experimental data was not provided in that paper.

Later, Ajimera and coworkers [4,5] presented experimental data for the gas pressure drop across a micropacked bed reactor packed with 60-microns glass beads, as shown in Figure 2. The maximum gas flow rate for the experiment was 100 cm$^3$/min and the Reynolds number for the gas flow was less than 1. The dimension of the packed bed were a length of 400 µm, a depth of 500 µm, and a width of 25.55 mm. Nitrogen gas was injected into the packed bed through 64 bifurcated inlet microchannels. We obtained the experimental data from 'Figure 6' in Ref. [4]. These data are re-plotted in Figure 3 for comparison with those obtained by numerical simulation and by Carman's correlation. The pressure drop across the bed is plotted as a function of the gas velocity.

For comparison with the simulations and estimations, the volume fraction of spheres in the packed bed should be known. Because the volume fraction of spheres in a random array depends on the configurations of the spheres, it is reasonable to take a range of volume fractions. Chong and coworkers [17] experimentally obtained a value of $\phi$ of 0.6 for closely packed spherical glass beads. Batchelor and O'Brien [18] gave 0.62 and Ma and Almahdi [19] took 0.64356 as the maximum volume fraction of equal-size spheres in a random array. Thus, in our present study we used the range from $\phi=0.6$ to 0.65 for calculation of the pressure drop through Carman's correlation and through numerical simulation. The value of $K$ obtained using the numerical simulation at $\phi=0.65$ corresponds to the extrapolated value. Figure 3 shows that the numerical simulation results are within the range of the experimental data for the pressure drop. Although the numerical simulation results are acceptable in practice, on average they are slightly smaller than the experimental results. This feature may be due to the assumption of equal-sized spheres, which is difficult to obtain in reality. Digression from this assumption can increase the actual volume fraction of the spheres, which results in enhancing the drag acting on the spheres. Overall, the numerical simulations were in reasonably good agreement with the actual experimental results for the pressure drop across the micropacked bed by single-phase flow. Therefore, we see that the simple Carman correlation is practically useful for predicting the pressure drop by single phase flow through micropacked beds filled with spherical particles.

**Summary**

A single-phase Stokes flow through a packed bed of
spheres was solved numerically to determine the drag acting on spheres and, hence, the pressure drop across the micropacked bed. The numerical simulation was performed from dilute to highly dense concentrations of spheres in random arrays. Some correlations were employed for comparison with the numerical simulation results. The estimations by Carman’s correlation were in excellent agreement with the numerical simulation results for $\phi > 0.4$. Finally, these numerical calculations were compared with the experimental results for a micropacked bed [2,4,5]. A good agreement exists between the two, which ultimately means that the pressure drop across the micropacked bed can be predicted with reasonable accuracy by using Carman’s simple correlation.

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Nomenclatures

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>$a$</td>
<td>radius of particle</td>
</tr>
<tr>
<td>$F$</td>
<td>drag force acting on spheres</td>
</tr>
<tr>
<td>$G$</td>
<td>differential operator</td>
</tr>
<tr>
<td>$K$</td>
<td>non-dimensional drag</td>
</tr>
<tr>
<td>$n$</td>
<td>number density of particles</td>
</tr>
<tr>
<td>$p$</td>
<td>pressure in the fluid</td>
</tr>
<tr>
<td>$p_n$</td>
<td>spherical harmonic function of order $n$</td>
</tr>
<tr>
<td>$P_{nm}$</td>
<td>coefficient of the harmonic function $p_n$ (superscript $s$: singular part; superscript $r$: regular part)</td>
</tr>
<tr>
<td>$P_n^m$</td>
<td>associated Legendre polynomial</td>
</tr>
<tr>
<td>$r$</td>
<td>radial distance from the center of the particle at origin</td>
</tr>
<tr>
<td>$u$</td>
<td>velocity of the fluid</td>
</tr>
<tr>
<td>$U$</td>
<td>superficial velocity of the fluid through the bed</td>
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<tr>
<td>$v$</td>
<td>fundamental periodic singular solution of Stokes equation</td>
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<tr>
<td>$x$</td>
<td>position vector</td>
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Greek Letters

<table>
<thead>
<tr>
<th>Symbol</th>
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<tbody>
<tr>
<td>$x_n$</td>
<td>spherical harmonic function of order $n$</td>
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<td>$\phi$</td>
<td>volume fraction of the particles</td>
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<tr>
<td>$\phi_n$</td>
<td>spherical harmonic function of order $n$</td>
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<td>$\mu$</td>
<td>viscosity of fluid</td>
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<td>$\tau$</td>
<td>unit cell volume</td>
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References