Effect of Particle Stratification on the Performance of Fluidized Adsorption Beds

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INTRODUCTION

The application of fluidized granular activated carbon columns for treating liquid waste has become a common engineering practice during the last two decades. The dynamics of the process is complicated by the growth of bacterial films on the surfaces of carbon particles. The films have certain beneficial effects, including direct uptake of biodegradable substrates from the liquid phase and bioregeneration of the carbon. However, film growth also has certain harmful effects. It creates extra mass transfer resistance to adsorption and the inevitable bed height expansion as the apparent size and density of carbon particles change.

The presence of bacterial films outside carbon particles also complicates the fluid-mechanical behavior of the fluidized columns. The apparent size and density of carbon particles change as the film thickness increases. Since the film coverage of carbon particles is inevitably nonuniform, a certain degree of carbon particles stratification according to the extent of bacterial growth can be expected. It is conceivable that this stratification may affect the performance of the adsorption column.

Andrews and Tien (1982) proposed a model for analyzing the dynamics of fluidized adsorption columns taking into account some of the complications which arise from the presence of bacterial growth. As a simplification of their analysis, the distribution of carbon particles was assumed to be either one of two limiting situations: complete stratification, such that the placement of carbon particles in a column is according to the values of their settling velocities; or complete mixing, such that the same size distribution of carbon particles is observed throughout the bed.

Patwardhan and Tien (1984) have recently established a procedure to estimate the distribution of particles of different types (different either in density or particle size, or both) in liquid fluidized beds. Thus, by incorporating that method into the earlier model of Andrews and Tien (1982) one can perform a more exact analysis of fluidized adsorption columns.

RELEVANT EQUATIONS

When a mixture of N types of particles is fluidized by liquid, the axial distribution of particles is described by the following set of equations (Patwardhan and Tien, 1984):

$$-D_1 \frac{dC_i}{dz} + (u/\varepsilon + v_i)\, C_i = 0 \qquad i = 1, \ldots N \tag{1}$$

subject to the condition

$$V_i = \int_o^H C_i \, dz \tag{2}$$

where C_i is the volume concentration on the ith type particles and D_i and v_i are, respectively, the diffusivity and relative velocity (with respect to liquid) of the ith type particles. D_i has been reported experimentally (Al-Dubouni and Garside, 1979), and v_i can be estimated from the following equations.

$$\frac{v_i}{u_{t,i}} = \varepsilon^{n_i - 2}(\rho_i - \rho_m)/(\rho_i - \rho_f) \tag{3}$$

$$\frac{5.1 - n_i}{n_i - 2.7} = 0.1\, [N_{Re,t}]_i^{0.9} \tag{4}$$

where $u_{t,i}$ is the terminal velocity of a single particle of the ith type. The Reynolds number is based on $u_{t,i}$ and d_i.

The performance of an adsorption column is given by its effluent concentration history, which can be obtained from the solution of the conservation equation of substrate in the continuous phase.

$$\frac{d}{dz}\left[\varepsilon D_d \frac{dS}{dz}\right] - \left(u - \frac{d}{dt}\int_o^z \varepsilon \, dz\right)\frac{dS}{dz} - \overline{R}_j = \varepsilon \frac{dS}{dt} \tag{5}$$

If the bacterial growth rate is low, both the terms $(d/dt) \int_o^z \varepsilon \, dz$ and $\varepsilon\, (dS/dt)$ can be ignored. In other words, the pseudo-steady-state solution at any given time is assumed to provide a reasonable description of the dynamics of the process at that time. Thus, Eq. 1 becomes

$$\varepsilon D_d \frac{d^2S}{dz^2} - \left[u - \frac{d}{dz}(\varepsilon D_d)\right]\frac{dS}{dz} = \overline{R}_j \tag{6}$$

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The boundary conditions are

$$\frac{dS}{dz} = 0 \qquad \text{at } a = 0 \tag{7}$$

$$S_{\text{in}} - S + D_d \frac{dS}{dz} = 0 \qquad \text{at } z = H \tag{8}$$

Note that the z coordinate is taken downward from the top of the column. The liquid stream enters the bed at $z = H$ and the fluid velocity is negative.

The effluent concentration is given by the value of S at $z = 0$. The time-dependent behavior of the effluent history arises from the fact that the extent of film coverage, x, and therefore the particle distribution, the bed porosity and, more important, $\overline{R}_f$, all vary with time. Thus, the value of S at $z = 0$ varies.

The quantity $\overline{R}_f$ represents the substrate uptake rate by bacterial films per unit bed volume. Since film coverage of particles at any point within the bed is not uniform, this quantity has to be obtained by including the substrate uptake rates from carbon particles with all kinds of film coverage.

In an earlier work, Andrews and Tien (1981) postulated that for an adsorbent particle with a film coverage x (defined as the ratio of the film volume to the clean particle volume) exposed to a solution of substrate concentration S, the total substrate uptake rate, N_T, and the substrate uptake rate by adsorbent, N_A, (on the basis of unit volume of clean particles) are:

$$N_T = \frac{k_v}{k_1}\frac{S\cosh k_1 x - c_s}{\sinh k_1 x} \tag{9}$$

$$N_A = \frac{k_v}{k_1}\frac{S - c_s\cosh k_1 x}{\sinh k_1 x} = k_s(q_s^* - q) = k_s\left(\frac{a c_s}{b + c_s} - q\right) \tag{10}$$

with the meanings of the symbols given in the Notation.

The above two expressions relate the rates of substrate uptake to the conditions of the adsorbent particle (i.e., film coverage, x, and the degree of saturation, q) and the substrate concentration of the solution to which the film is exposed.

Before deriving the expression of $\overline{R}_f$, it is necessary to decide upon a method to classify adsorbent particles in a fluidized bed. The most direct way would be to group the particles according to their values of x. To accomplish this, it is necessary to identify each and every particle with its initial film coverage, x_o, and the history of its movement throughout the bed. This approach is impractical because of the very large number of particles involved, and impossible because exact knowledge of particle mixing in liquid fluidized beds is lacking.

To circumvent this difficulty, the following assumption is used. In view of the low rate of bacterial growth, over a short time interval, Δt, the conditions within a fluidized bed may be assumed to be constant. On the other hand, Δt is sufficiently large to allow a given particle to move around throughout the bed. For two identical particles (same x and q), the time each spends in a given part of the bed during Δt is the same. This assumption is a generalization of that used by Andrews and Tien (1982) in the case of a completely mixed (solid) fluidized bed. In that instance, particles were assumed to spend equal amounts of time in every part of the bed.

By using this assumption, one can see that for the case of monosized adsorbent particles with different initial film coverages, particles which have the same initial film coverage, x_o, will always have the same x and q at any time, even though both x and q change with time. Thus, one may classify particles according to x_o. The ith type of particles, for example, are particles with their initial film coverage equal to $x_{i,o}$.

When particles are so classified, the quantity V_i of Eq. 2 at any given time (or any given film coverage, x) is

$$V_i = V_{i,o}(1 + x_i)$$

where $V_{i,o}$ is the volume of the ith type particle initially. The density and size of the ith type particles and the density of the liquid-solid mixture are

$$d_i = d_o(1 + x_i)^{1/3} \tag{11}$$

$$\rho_i = (\rho_o + x_i\rho_b)/(1 + x_i) \tag{12}$$

$$\rho_m = \rho_f(1 - \varepsilon) + \sum_i \rho_i C_i \tag{13}$$

The total bed height, H, is given as (Andrews and Tien, 1979; Wang and Tien, 1983):

$$H = \frac{1 - \varepsilon_c}{1 - \varepsilon_{\text{av}}}(1 + x_{\text{av}})H_o \tag{14}$$

$$x_{\text{av}} = \frac{\sum_i V_{i,o}x_i}{\sum_i V_{i,o}} \tag{15}$$

$$\varepsilon_{\text{av}} = \frac{1}{H}\int_o^H \varepsilon\, dz \tag{16}$$

For a given type of particle, the film coverage increase over Δt in terms of the organic carbon content of the bacterial film is $\rho\Delta(V_{i,o}x_i)$.

The extent of the bacterial growth can be related to the quantity $N_T - N_A$, namely, the difference between the total substrate uptake and the uptake by adsorption. Although the particles move throughout the bed, it is reasonable to assume that they spend more time in parts of the bed where their presence is dominant. Thus, if the bed is divided into N compartments, one may assume that the time spent by a particle of the ith type in a given compartment is

$$\frac{\Delta t}{N}\frac{C_i}{\frac{1}{H}\int_o^H C_i\, dz}$$

The amount of substrate consumed by a given type of particle as a result of the bacterial growth is, therefore,

$$\begin{aligned}\sum_j (V_{i,o})[N_T - N_A]_{\substack{x=x_i\\ z=j\Delta z}}\frac{\Delta t}{N}\frac{C_i H}{\int_o^H C_i\, dz} &= \Delta t\frac{V_{i,o}}{\int_o^H C_i\, dz}\sum(N_T - N_A)_{\substack{x=x_i\\ z=j\Delta z}} C_i(\Delta z) \qquad (17)\\ &= \frac{\Delta t\, V_{i,o}}{\int_o^H C_i\, dz}\int_o^H (N_T - N_A)\, C_i\, dz\end{aligned}$$

A balance of the substrate in the growth process yields the following expression

$$\frac{d(V_{i,o}x_i)}{dt} = \frac{Y}{\rho}\frac{1}{1 + x_i}\int_o^H (N_T - N_A)\, C_i\, dz - (k_b + k_w)V_{i,o}\, x_i \tag{18}$$

The last term accounts for film wash-off and decay. Similarly, the rate of the saturation of adsorbent particles is

$$\frac{d(V_{i,o}q_i)}{dt} = \frac{1}{1 + x_i}\int_o^H C_i\, N_A\, dz \tag{19}$$

The substrate uptake term, $\overline{R}_J$, of Eq. 6, on the other hand, is the average uptake rate for all types of adsorbent particles. For a unit bed volume, the volume occupied by the ith type particle in its film-free state is $C_i/(1 + x_i)$. Thus, the substrate uptake rate by the biofilm outside the ith type particle per unit bed value is $[C_i/(1 + x_i)]\ (N_T)_{x=x_i}$. The total substrate uptake rate, including all types of adsorbent particles, $\overline{R}_J$, is

$$\overline{R}_J = \sum_i \frac{C_i}{1 + x_i} (N_T)_{x=x_i}$$

$$= \sum_i \frac{C_i}{1 + x_i} \frac{k_v}{k_1} \frac{S \cosh k_i x_i - c_{s,i}}{\sinh k_i x_i} \tag{20}$$

Substituting Eq. 20 into Eq. 6, one has

$$\varepsilon D_d \frac{d^2S}{dz^2} - \left[u - \frac{d}{dz}(\varepsilon D_d)\right] \frac{dS}{dz}$$

$$= \sum_i \frac{C_i}{1 + x_i} \frac{k_v}{k_1} \frac{S \cosh k_i x_i - c_{s,i}}{\sinh k_i x_i} \tag{21}$$

In summary, the relevant equations describing the various aspects of fluidized adsorption column problems are:

Substrate Concentration Profile in Liquid Phase

$$\varepsilon D_d \frac{d^2S}{dz^2} - \left[u - \frac{d}{dz}(\varepsilon D_d)\right] \frac{dS}{dz}$$

$$= \sum_i \frac{C_i}{1 + x_i} \frac{k_v}{k_1} \frac{S \cosh k_1 x_i - c_{s,i}}{\sinh k_1 x_i} \tag{21}$$

with boundary conditions

$$\frac{ds}{dz} = 0, \text{ at } z = 0 \text{ (Eq. 7) and } S_{in} - S + D_d \frac{dS}{dz}$$

$$= 0 \text{ at } z = H \text{ (Eq. 8).}$$

Particle Concentration Profile

$$-D_i \frac{\partial C_i}{\partial z} + (u/\varepsilon + v_i)\, C_i = 0 \text{ for } i = 1, 2, \ldots n \tag{1}$$

$$V_i = V_{i,o}(1 + x_i) = \int_o^H C_i\, dz \tag{2}$$

with v_i given by Eqs. 3 and 4, and densities from Eqs. 12 and 13. The bed height, H, can be found from Eq. 14.

Film Coverage and Saturation

$$\frac{d}{dt}(v_{i,o} x_i) = \frac{Y}{\rho} \frac{1}{1+x_i} \int_o^H (N_T - N_A) C_i\, dz - (k_b + k_w) V_{i,o} x_i \tag{18}$$

$$\frac{d}{dt}(V_{i,o} q_i) = \frac{1}{1+x_i} \int_o^H C_i N_A\, dz \tag{19}$$

where

$$x_i = x_{i,o} \text{ at } t = 0 \text{ (Eq. 22) and } q_i = 0 \text{ at } t = 0 \tag{23}$$

Relationship between x_i, S, $c_{s,i}$, and q_i

$$\frac{k_v}{k_1} \frac{S - c_{s,i} \cosh k_1 x_i}{\sinh k_1 x_i} = k_s \left(\frac{a\, c_{s,i}}{b + c_{s,i}} - q_i\right) \tag{10}$$

SOLUTIONS AND RESULTS

The equations presented above provide a complete description of the dynamics of fluidized adsorption columns. A numerical method for their solution was developed as follows. Assuming that x_i and q_i for $i = 1, 2, \ldots N$ are known at a given time, t, then the total bed height, H can be calculated from Eq. 14. The particle concentration profile, $C_i(z)$, can be found by solving the system of Eqs. 1 and 2, and the profile of the substrate concentration in the liquid phase from Eqs. 21, 7, and 8. The former solution was obtained by using an interative procedure developed by Patwardhan and Tien (1984), while the substrate concentration profile was obtained by first discretizing Eq. 21 and then solving the resulting nonlinear algebraic equations by the Newton-Raphson method. The axial dispersion coefficient, D_d, was estimated from the correlation of Chung and Wen (1968). To solve Eq. 21, the substrate concentration at the film-adsorbent interface was required for each type of particles. This information was obtained through Eq. 10. Once the substrate and particle concentration profiles were known, the rate of the film coverage growth and that of adsorbent saturation were estimated from Eqs. 18 and 19, and the value of x_i and q_i at time $t + \Delta t$ were readily calculated (e.g., by the fourth-order Runge-Kutta method). The effluent concentration history (i.e., S at $z = H$ and various times) provided the description of the column performances.

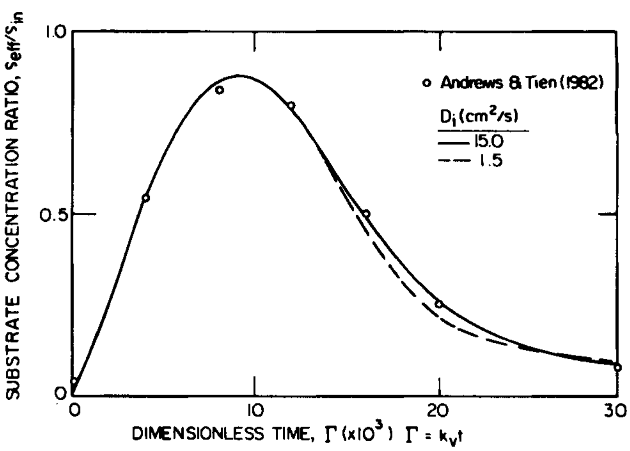

Figure 1. Prediction of effluent concentration.

A number of computations were made and a typical set of results is shown here. The calculations were made using parameter values and x_o identical to those listed in Table 1 of Andrews and Tien (1981) and two values of D_i to account for the different degree of solid mixing. The results are shown in Figures 1 through 4. Also included in these figures are the results of Andrews and Tien based on complete solid mixing.

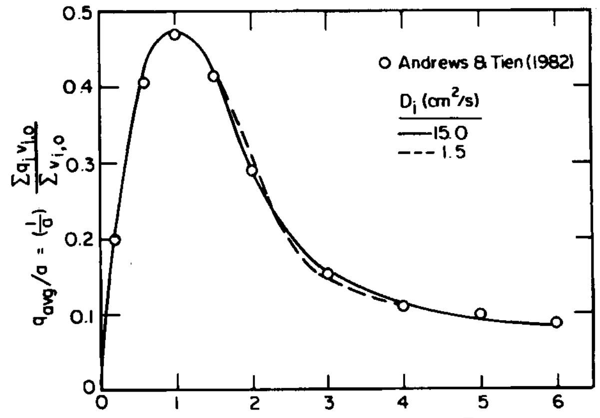

Figure 2. Variation of average adsorbent saturation with time.

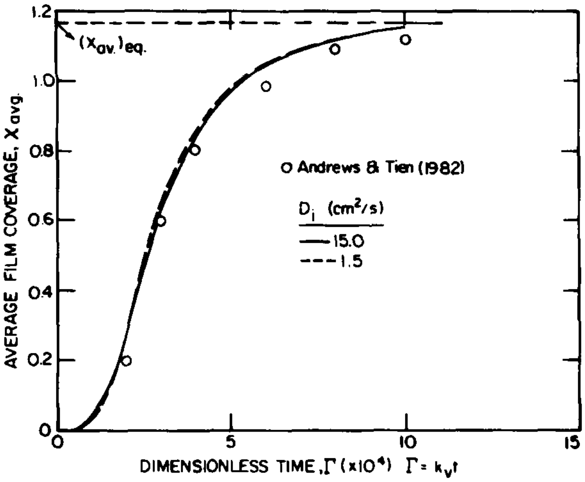

Figure 3. Variation of average film coverage with time.

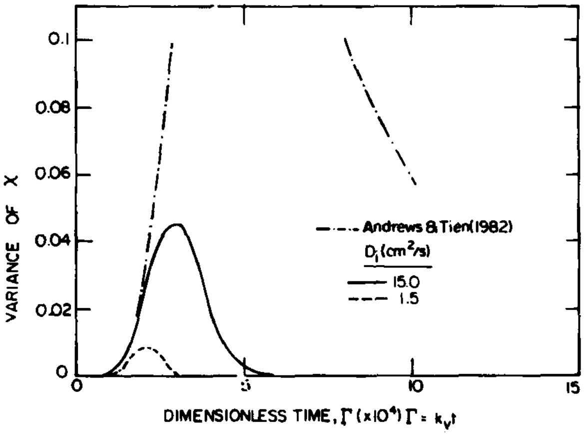

Figure 4. Variation of the variance of film coverage with time.

It is obvious that with the exception of the variance of x, the effect of particle distribution is insignificant. Thus, in predicting the performance of fluidized adsorption columns with nearly monosized adsorbent particles, the use of the complete mixing assumption is warranted.

The results of the calculation do not necessarily indicate that particle distribution has no effect on adsorption column performance. The calculations merely show that for the case of monosized particles, the difference in film coverage does not provide sufficient effect. On the other hand, if the adsorbent particles cover a wide size range, the effect of particle segregation is expected to be important. The degree of particle segregation, however, will not change substantially with time. The particle distribution corresponding to the clean particle state obtained from the solution of Eq. 1 would remain essentially correct throughout the duration of the operation.

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NOTATION

a,b = Langmuir isotherm expression constant
C_i = particle concentration (vol/vol) for the ith type particle
c_s = substrate concentration at the film-adsorbent interface
D_d = axial dispersion coefficient in the liquid phase
D_i = diffusivity of the ith type particles
d_i = diameter of the ith type particles
d_o = diameter of film-free particles
H = bed height
H_o = initial bed height
k_1 = defined as $\alpha\sqrt{k_v/D}$ where D is the substrate diffusivity in film
k_b = film decay rate constant due to basal metabolism
k_v = substrate uptake rate constant in the film
k_w = Film wash-off rate constant
N_A = rate of substrate uptake due to adsorption per unit clean particle volume
$N_{Re,t}$ = Reynolds number of particle based on particle terminal velocity u_t
N_T = total substrate uptake rate per unit clean particle volume
n = exponent
q = average adsorbed substrate concentration
q_s = adsorbed substrate concentration at the film-adsorbent interface
$\overline{R}_J$ = total substrate uptake rate given by Eq. 20
S = substrate concentration in the liquid
S_{in} = inlet value of S
t = time
u = axial superficial velocity of liquid
u_{ti} = terminal velocity of particles of the ith type
V_i = volume of the ith type of particle
$V_{i,o}$ = volume of the ith type particle in the film-free state
v_i = relative velocity (to liquid) of the ith type particles
x_{av} = average value of x_i
x_i = film coverage of the ith type particles
$x_{i,o}$ = initial film coverage of the ith type particles
Y = yield coefficient
z = axial distance

Greek Letters

α = volume/surface ratio for clean particles
ε = porosity
ε_{iv} = average porosity
ε_c = clear bed porosity
ρ = organic carbon density of bacterial film
ρ_f = fluid density
ρ_i = density of particle of the ith type
ρ_m = density of fluid-particle mixture

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