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Microbial Film Model for the Interaction between Adsorption and Bacterial Activity in Fixed Bed Processes

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ABSTRACT

A chemical reaction - diffusion film model is presented for the description of the growth and presence of microbial film outside adsorbents in fixed bed processes. The model was tested against experiments in which an aqueous solution of valeric acid was passed through a carbon bed and the removal of valeric acid from the solution was effected through carbon adsorption as well as aerobic and anaerobic bacteria present in the bed.

INTRODUCTION

The use of solid adsorbents for the removal of certain dissolved substances present in liquid streams is an engineering practice of long standing. Adsorption processes are cyclic in nature, beginning with fresh adsorbents and terminating when the adsorbents become nearly saturated. For fixed bed operations, if the saturation phase is prolonged (i.e. the duration of contact between adsorbents and liquid is length) and furthermore, if some of the species present in the liquid are biodegradable in addition to being adsorbable, the presence of bacteria near adsorbent surfaces becomes likely, leading to the formation of microbial film outside adsorbents. Such a situation occurs in the application of

granular activated carbon for water and waste water treatment.

The interaction between the bacterial activity as exemplified by the microbial film outside adsorbents and the adsorption processes is myriad and indeed complex. A qualitative description of the interaction can be stated as follows:

(1) Microbial film offers additional resistance to mass transfer. Species to be removed by adsorption must first diffuse across the film before adsorption can take place.

(2) Microbial film reduces the void space of adsorption beds, thus causing bed clogging and increasing the pressure drop necessary to maintain a given throughput.

(3) Wash-off microbial film may occur under certain conditions (e.g. when film thickness exceeds certain values). Consequently the quality of effluents deteriorates.

(4) Bacterial activity of microbial film contributes to the overall performance of adsorption columns. The species can be removed by either adsorption or biological degradation.

(5) The presence of microbial films outside adsorbent surfaces causes the bioregeneration phenomenon. Experimental data indicate that with bacterial activity, saturated adsorbents can be regenerated (1). Similarly, in fixed bed experiments, with microbial film forming outside adsorbents, a complete saturation of adsorbents cannot be achieved in certain cases.

Certain aspects of the interaction are clearly advantageous to the intended separation processes, while others are definitely deleterious. An optimum operation of adsorption processes with bacterial activity therefore, requires the maximum exploitation of the potential advantages as well as the suppression or reduction of the harmful factors. To accomplish the optimization, a theoretical framework which analyzes the interaction phenomenon and describes it quantitatively is obviously required. The present work provides a brief summary of such an effort.

DESCRIPTION OF MODEL

A number of investigators (2, 3, 4, 5) have considered the model description of microbial film growth outside adsorbent surfaces primarily in connection with carbon treatment of water and waste water. The conceptual model used here was first formulated by Andrews and Tien (5, 6, 7) and is shown in Figure 1. The presence of bacterial activity leads to the presence of microbial film outside adsorbents in the form of uniform coating whose thickness increases with time. The time dependence behavior can be directly related with the extent of bacterial activity.

The removal of the species from the liquid stream is effected through the species uptake by the film, N. On the other hand, only part of this flux leads to adsorption. The adsorption flux can be calculated by the concentration profile of the species across the film at the film base.

Figure 1.b shows a form of bioregeneration built into the model. As the film grows beyond a certain thickness, an inversion of the species concentration profile in contrast to the monotonic type as shown in Figure 1.a may occur. If the adsorption process is reversible, this means desorption of species previously adsorbed. Adsorbents therefore become less saturated.

Since the growth of the film is slow in general, the use of the pseudo steady state assumption is justified. The concentrations of species i, and S_i are described by the following set of conditions:

$$D \frac{d^2 S_i}{dx^2} + R_i = 0 \qquad \text{for } 0 < x < \ell \tag{1}$$

$$(N)_i = - D_i \left(\frac{dS_i}{dx}\right)_{\ell} \tag{2}$$

$$(N_A)_i = - D_i \left(\frac{dS_i}{dx}\right)_{o} \tag{3}$$

$$\ell = f[(N)_i - (N_A)_i] \tag{4}$$

$$\ell = o, \qquad t = o \tag{5}$$

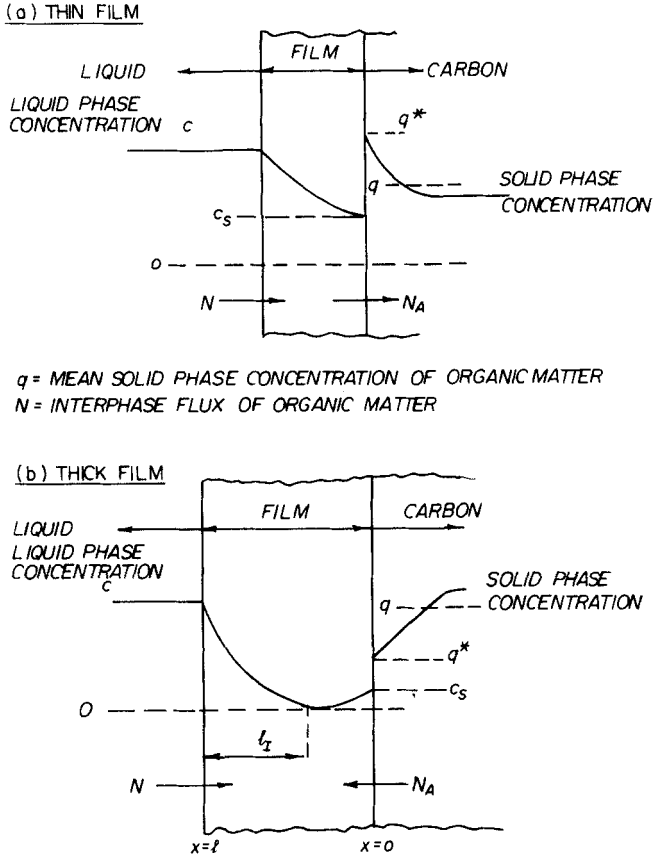

FIG.1 GROWTH OF A BACTERIAL FILM ON AN ADSORBENT SURFACE

where S_i denotes the concentration of the i-th species and R_i is the rate of consumption of the i-th species due to bacterial activity. The concentration profile of the i-th species can be obtained from the solution of Equation (1) with appropriate boundary conditions.

Once the concentration profile is known, the species uptake rate, N and the rate of adsorption, N_A can be found readily from Equations (2) and (3). The growth rate of the film can be found from Equation (4).

ADOPTION OF THE MODEL TO FIXED BED PROCESSES

The conceptual model described above has been applied to a number of situations (7, 8) under the assumption that the nature of the bacterial activity is independent of the film thickness. For example, liquid wastes which require treatment with activated carbon inevitably contain a certain amount of dissolved oxygen (DO). Accordingly, for thin microbial film, the biological activity is likely to be aerobic respiration. As the film thickness increases, the depletion of DO in the outer portion of the film renders the bacterial activity in the inner part of the film anaerobic. The inclusion of this bi-layer behavior of microbial film is considered in the present work.

The process considered here can be described as follows: A liquid solution containing an adsorbable and biodegradable species as well as other necessary inorganic salts is passed through a column packed with granular adsorbents. The assumptions used are:

(1) The column has a uniform cross section and the adsorbents are spherical and uniform in size.

(2) The dispersion effect is negligible.

(3) The rate of the bacterial activity is first order with respect to the concentration of the adsorbable species.

(4) The bacterial activity of the microbial film is aerobic as long as there is dissolved oxygen present. Wherever the dissolved oxygen is completely depleted, the bacterial activity becomes anaerobic leading to the removal of nitrate salt from the solution.

(5) In both aerobic and anaerobic growth, the biological pathways are invariant. The consumption of substrates (adsorbable species) follows a fixed stoichiometric relationship.

(6) The effect of outward diffusion of biological end products across the film is negligible.

(7) There is little wash-off and basal metabolism from the film.

In addition, the assumptions commonly used in fixed bed processes such as isothermal, steady-state one dimensional plug flow, uniform packing, etc. are involved.

The macroscopic conservation equation for the adsorbable species, the dissolved oxygen and nitrate in the liquid phase can be written as

$$u \frac{\partial [c]}{\partial z} + (1 - \varepsilon) \frac{3}{a_p} N_c = 0 \tag{6.a}$$

$$u \frac{\partial [O_2]}{\partial z} + (1 - \varepsilon) \frac{3}{a_p} N_{O2} = 0 \tag{6.b}$$

$$u \frac{\partial [NO_3]}{\partial z} + (1 - \varepsilon) \frac{3}{a_p} N_{NO_3} = 0 \tag{6.c}$$

where $[c]$, $[O_2]$ and $[NO_3]$ are the concentrations of the adsorbable species, the dissolved oxygen and nitrate salt respectively. The independent variable z denotes the axial distance measured from the inlet. u is the superficial velocity. ε is the void fraction of the carbon bed. a_p is the radius of each carbon granule.

To estimate substrate uptake fluxes of the biofilm, N_c, N_{02}, and N_{NO_3}, Equation (1) can be applied. The relevant equations are

In the region $\ell_I < x < \ell$

$$D_c \frac{d^2 S_c}{dx^2} = k_v S_c \tag{7.a}$$

$$D_o \frac{d^2 S_{O_2}}{dx^2} = \beta k_v S_c \tag{7.b}$$

$$D_N \frac{d^2 S_{NO_3}}{dx^2} = 0 \tag{7.c}$$

In the region $0 < x < \ell_I$

$$D_c \frac{d^2 S_c}{dx^2} = \alpha k_v S_c \tag{8.a}$$

$$S_{O_2} = 0 \tag{8.b}$$

$$D_N \frac{d^2 S_{NO_3}}{dx^2} = \gamma k_v S_c \tag{8.c}$$

where ℓ_I is the thickness of the anaerobic film. α, β and γ are the reaction rate ratios of nitrate respiration, oxygen reduction and denitrification to the aerobic respiration respectively.

The boundary conditions of Equations (7) and (8) are as follows

At $x = \ell$ $$S_c = [C] \tag{9}$$

$$S_{O_2} = [O_2] \tag{10}$$

$$S_{NO_3} = [NO_3] \tag{11}$$

At $x = 0$ $$D_c \frac{dS_c}{dx} = \frac{a_p}{3} \frac{dq}{d\theta} = k_p (q_i - q) \tag{12}$$

$$\frac{dS_{NO_3}}{dx} = 0 \tag{13}$$

At $x = \ell_2$ $$\frac{dS_{O_2}}{dx} = 0 \tag{14}$$

when $\ell_I > 0$, in addition to the boundary conditions, the interface conditions are

$$S_c \Big|_{\ell_I^+} = S_c \Big|_{\ell_I^-} \tag{15}$$

$$S_{O_2} \Big|_{x < \ell_I} = 0 \tag{16}$$

$$S_{NO_3} \Big|_{\ell_I^+} = S_{NO_3} \Big|_{\ell_I^-} \tag{17}$$

$$\frac{dS_c}{dx} \Big|_{\ell_I^+} = \frac{dS_c}{dx} \Big|_{\ell_I^-} \tag{18}$$

$$\frac{dS_{NO_3}}{dx} \Big|_{\ell_I^+} = \frac{dS_{NO_3}}{dx} \Big|_{\ell_I^-} \tag{19}$$

$$q_i = f(S_c \Big|_{x=0}) \tag{20}$$

where f(S) is the adsorption isotherm. In other words, equilibrium condition is assumed at the film base. The film growth rate can be related with the bacterial activity throughout the film as

$$(4\pi a_p^2)\rho \frac{d(\ell-\ell_I)}{d\theta} = \frac{4}{3}\pi a_p^3 \rho_p \frac{d\sigma_b^{(1)}}{\theta} = Y_1 4\pi a_p^2 \int_{\ell_I}^{\ell} k_v S_c dx \qquad (21.a$$

$$(4\pi a_p^2)\rho \frac{d\ell_I}{d\theta} = \frac{4}{3}\pi a_p^3 \rho_p \frac{d\sigma_b^{(2)}}{d\theta} = Y_2 4\pi a_p^2 \int_{0}^{\ell_I} \alpha k_v S_c dx \qquad (21.b$$

and

$$\sigma_b = \sigma_b^{(1)} + \sigma_b^{(2)} \qquad (22)$$

where σ_b is the biofilm organic carbon per unit weight of carbon granule and ρ is the organic carbon density of the film and ρ_p is the carbon granule density. Superscripts (1) and (2) denote the regions of $x > \ell_I$ (aerobic) and $x < \ell_I$ (anaerobic), where Y_1 and Y_2 are the corresponding yield coefficients. $\theta(=t - z\varepsilon/u)$ is the corrected time.

Equations (16) - (22) constitute a complete description of bacterial growth, adsorption and substrate reduction in the GAC column. The algorithms developed by Vanier (9) can be used for the numerical solution.

PARAMETER ESTIMATION

The model discussed above is formulated with the use of ten biological parameters (Y_1, Y_2, k_v, α, β, γ, ρ, D_c, D_o and D_N) as well as the usual adsorption parameters (isotherm parameters, k_p). The adsorption parameters can be obtained from the appropriate adsorption measurements. For the determination of the biological parameters, Andrews (6) proposed a procedure involving the use of a fluidized bed reactor and non-adsorbing particles (coal). The fluidized bed apparatus functions as a completely stirred tank reactor (CSTR) from which concentration histories of dissolved organic carbon (DOC), dissolved oxygen (DO), nitrate, and total organic carbon (TOC) as well as the history of bed height expansion can be measured. The apparatus can be operated under either aerobic or anaerobic conditions.

Under the condition of anaerobic growth, the conservation equations become

$$\tau \frac{d[C]}{dt} = [C]_{in} - [C] - \frac{W}{FY_2} \frac{d\sigma^{(2)}}{dt} \tag{23}$$

$$\tau \frac{d[NO_3]}{dt} = [NO_3]_{in} - [NO_3] - \frac{W}{FY_2} \frac{\alpha}{\alpha} \frac{d\sigma_b^{(2)}}{dt} \tag{24}$$

$$\tau \frac{d[TC]}{dt} = \frac{W}{F} \frac{d\sigma_b^{(2)}}{dt} - \Delta(TC) \tag{25}$$

$$\tau \frac{d\sigma_b^{(2)}}{dt} = \frac{3Y_2}{a_p \rho_p} \sqrt{\alpha k_v D_c}\, [C] \tanh \sqrt{\frac{\alpha k_v}{D_c}} \frac{1}{\rho} \frac{a_p \rho_p}{3} \sigma_b^{(2)} \tag{26}$$

In the fluidized reactor, the biofilm volume can be related to the bed height expansion as (10)

$$\sigma_b^{(2)} = \frac{\rho}{\rho_p} \frac{\frac{H}{H_c} - 1}{1 - E} \tag{27}$$

where H and H_c are bed height and clean bed height respectively and E is the expansion factor.

The solution of the above equations, together with the data of the concentration histories and bed height histories enable the determination of Y_2, $\frac{Y}{\alpha}$, ρ, αk_v, and D_c. Also, if the inlet solution is not deoxygenated, the data obtained during the initial period of measurement will provide the values of Y, β, k_v and D_{o_2}. The basis used to evaluate the parameters is the application of the biofilm equations [i.e. Equations (7) - (21)] to a completely stirred tank reactor system (CSTR) which can be used to approximate the fluidized bed reactor with large recycle ratio. Since the measurements are conducted with non-adsorbing particles, the right hand side of Equation (12) equals zero.

EXPERIMENTAL WORK AND COMPARISON WITH MODEL

Some preliminary experiments were performed and compared with model predictions. The experimental work consisted of passing an aqueous solution of valeric acid-nitrate salt through a granular carbon column (see Fig. 2 for experimental apparatus). The efflu-

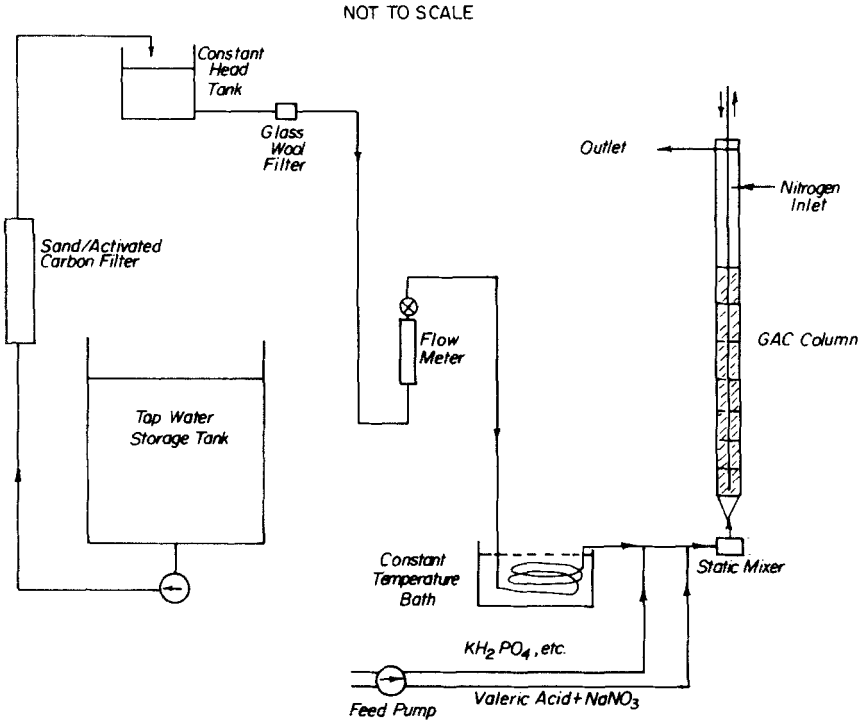

Fig. 2. Fixed Bed Apparatus

ent concentrations and bed height expansion were monitored. The initiation of bacterial activity was made by introducing a fixed quantity of culture at the beginning of each run. The experimental conditions are summarized in Table 1.

As an indication of the validity of the bi-layer model, Fig. 3 shows the comparison between the experimentally determined effluent concentration histories (dissolved oxygen, nitrate and dissolved organic carbon) and the prediction from the solutions of Equations (6) - (22). Both the nitrate concentration history and the dissolved organic carbon concentration history give reasonably good agreement. For the dissolved oxygen concentration, the comparison was marginal. However, it should be noted that none of the concentrations can be determined with sufficient accuracy. More importantly, in making

TABLE 1
Experimental Conditions

Carbon Particle Size	0.3 mm
Void Fraction of Bed	app. 0.42
Volumetric Flow Rate	27 cm^3/sec.
Bed Height	30 cm

Composition of Influent (Basis 1 ℓ tap water)

KH_2PO_4	17 mg	Na_2SO_4	106.5 mg for aerobic growth
K_2HPO_4	43.5 mg	$Co(NO_3)_2$-$6H_2O$	0.23 mg
Na_2HPO_4-$7H_2O$	66.8 mg	Valeric Acid	30 - 40 mg DOC
NH_4Cl	70 mg	$NaNO_3$/Valeric Acid	6.5 g/g
Na_2SO_3	94.5 mg for anaerobic growth	Dissolved Oxygen-Up to 7 mg	

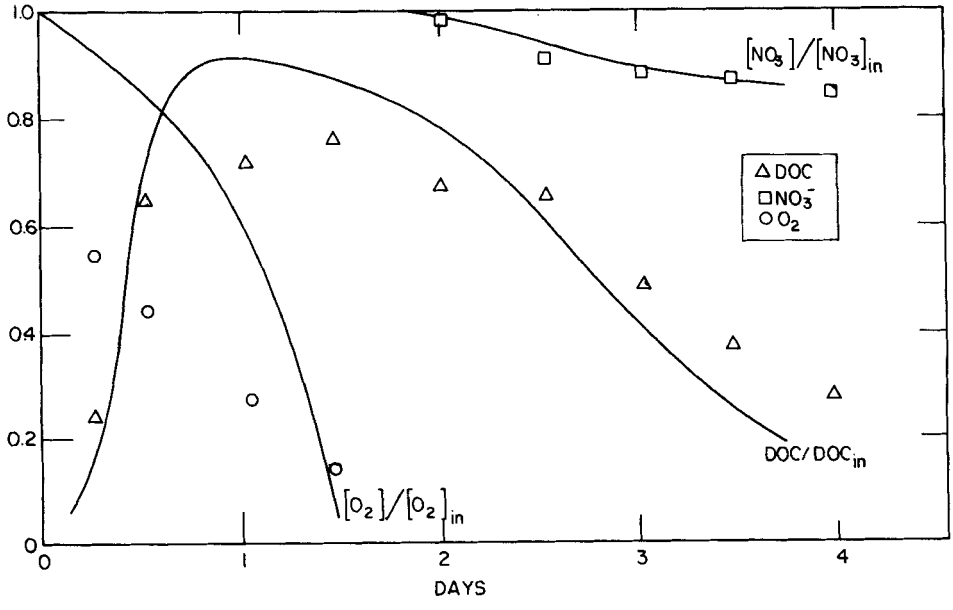

Fig. 3. Theoretical and Experimental Substrate Concentrations - Run 1

the prediction, all the model parameters were determined or estimated according to a well-prescribed procedure and there was no attempt to improve the fit by use of adjustable parameters. Under the circumstances the degree of agreement observed may provide optimism as to the validity of the basic model. A more definitive conclusion cannot be made until more extensive data becomes available.

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NOMENCLATURE

a_p	particle radius
D	diffusivity of substrate in the microbial film
F	volumetric flow rate through reactor
k_v	biological reaction rate constant
k_p	particle phase mass transfer coefficient
ℓ	film thickness
ℓ_1	thickness of the outer portions of the film, where aerobic respiration dominates
N	substrate uptake rate of microbial film from liquid
N_A	rate of adsorption
q_i	adsorbed phase concentration at adsorbed surface
q	average concentration of adsorbent
R	rate of biological degradation of substrate
S	substrate concentration in film
u	superficial velocity
x	distance measured from adsorbent surfaces

Y_1, Y_2	yield ratio of anaerobic and aerobic respirations respectively
z	axial distance
W	weight of coal particles in fluidized reactor

Greek Letters

α, β, γ	stoichiometric ratios of bacterial degradation reactions
σ_b	bacterial organic carbon per unit of adsorbent
ρ	organic carbon density
θ	time
τ	reactor residence time

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