MATHEMATICAL MODELING OF NATURAL IN SITU BIOREMEDIATION TO ESTIMATE INITIAL CONTAMINANT CONCENTRATION EFFECT

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Abstract   A mathematical model has been simulated to describe natural *in situ* bioremediation of contaminated soil. The model equations consist of a system of three non-linear partial differential equations. Sensitivity analysis conducted by numerically solving them, has demonstrated the effect of initial contaminant concentration on the time and mechanism of remediation. The result of simulation indicates that microbiological degradation of contaminant within the soil aggregate may play a significant role in remediation so that most of contaminants, which are initially present within the aggregates, are consumed before they reach the outer surface.

Key Words   Soil, Bioremediation, Mathematical Model

چکیده   یک مدل ریاضی به منظور توصیف فرآیند درمان بیولوژیکی طبیعی خاکهای آلوده در محل، مورد مطالعه و شبیه سازی قرار گرفته است. معادلات مدل شامل سه معادله دیفرانسیل جزیی غیرخطی می‌باشد. بررسی حساسیت مدل به دنبال حل عددی آن، تاثیر غلظت اولیه آلاینده را بر زمان و مکانیزم پاکسازی (درمان) نشان داده است. نتایج شبیه سازی نشان می‌دهد که تجزیه بیولوژیکی آلاینده‌ها توسط میکروارگانیزمهای طبیعی در داخل خاک دانه‌ها، نقش اساسی و مهمی در پاکسازی داشته و کسر قابل توجهی از آلاینده‌ها قبل از رسیدن به سطح مورد مصرف میکروارگانیزمها قرار می‌گیرد.

1. INTRODUCTION

Intrinsic bioremediation, also called natural attenuation or passive bioremediation, arises from the observation that in nature, microorganisms are ubiquitous and they will degrade organic materials at some rate, even if it is not very rapid [1,2]. Since the rate of movement of many organic materials in subsurface environments is not very rapid, the rate of degradation at some sites can be demonstrated to be at least as rapid as the movement of the organic materials in the subsurface. Hence, as long as there are no pathways of exposure (means of contact) to the contaminant plume, an argument can be made to leave everything alone and the problem will eventually go away by itself [3].

In certain cases, natural (or intrinsic) in situ bioremediation may provide a low-cost alternative to other methods of site restoration.

However, the effectiveness of intrinsic bioremediation depends on the relationship between the contaminant biodecay rate and the contaminant movement rate.

If natural biodecay is insufficient, then an engineered system such as biostimulation or bioaugmentation may be designed to supply nutrients, oxygen or introduced microorganism for enhanced in-situ bioremediation. If the enhanced process is inadequate, then more extreme measures such as hydraulic or physical containment may be needed. Due to variability in biological processes, evidence suggests that bioremediation potential must be evaluated on a model basis, and mathematical transport models can assist in such evaluations [4].

Bioremediation of contaminated soil has been modeled by a number of researchers [4-10].

Although numerous models are available in the literature, a little attempt to describe the ability of naturally occurring microorganisms to

attenuate the contaminants in the soil aggregates, are made.

2. MODEL DESCRIPTION

Let us, consider the soil as a collection of spherical aggregates with a single size. The microorganisms are present as suspended forms in the pore liquid and are also attached as microcolonies to the solid surface. They utilize organic contaminants, which uniformly deposited in soil aggregates, as carbon and energy source, in this way; they grow by consuming organic contaminants and oxygen. Because of the presence of immobile phase in the microvoids of aggregates, the transport is directed by diffusion only.

In the present model the primary emphasis is placed on simulating the biodegradation of hydrocarbons and other organics which are degradable only in the presence of oxygen using as few model parameters as feasible.

Compound which require oxygen for degradation is common in nature and include many petroleum derivatives.

Assumptions The following major assumptions are made in deriving the model equations:
(a) The temperature is constant in the bed, and the aggregates are, homogenous and isotropic; (b) water in pores of the soil aggregate constitutes the liquid phase and the remaining part of aggregate is considered as the solid phase. No gas phase exists because the aggregate is saturated with water; (c) only three components, substrate, oxygen and biomass, are involved in biodegradation; (d) the flow rate of oxygen rich water having a zero concentration of substrate and a low value of biomass is sufficiently high in the macrovoids of the bed; This maintains fixed concentrations of substrate, oxygen and biomass at exterior of the aggregate at r = R; (e) the dissolved concentration, X, and the adsorbed concentration, $M,$ are related through the local adsorption - desorption equilibrium in the aggregate : $M = K_d X$ (Freundlich isotherm, exponent = 1); and (f) the transport resistances of substrate and oxygen to and through the microcolonies attached to the surface of soil particles are negligible.

3. MATHEMATICAL MODELLING

The mass balance of component n in a spherical aggregate gives rise to Equation 1,

$$\varepsilon_a \frac{\partial X_n}{\partial t} + \rho \frac{\partial M_n}{\partial t} = \frac{D_n \varepsilon_a}{\tau r^2} \frac{\partial}{\partial r}(r^2 \frac{\partial X_n}{\partial r}) + \quad (1)$$
$$\frac{D_{n,so}}{r^2} \rho \frac{\partial}{\partial r}(r^2 \frac{\partial M_n}{\partial r}) + \varepsilon_a r_{n,l} + \rho r_{n,s}$$

where subscript n stands for substrate, oxygen and biomass. According to described assumption about the relation between, dissolved concentration and adsorbed concentration, the general equation for component n in the aggregate can be gained from Equation 1.

$$(\varepsilon_a + \rho K_{dn})\frac{\partial X_n}{\partial t} =$$
$$\frac{D_n \varepsilon_a / \tau + D_{n,so} \rho K_{dn}}{r^2} * \frac{\partial}{\partial r}(r^2 \frac{\partial X_n}{\partial r}) + \varepsilon_a r_{n,l} + \rho r_{n,s}$$
$$(2)$$

The aerobic microbial metabolism can be limited by a lack of substrate (carbon and energy source), oxygen (electron acceptor) or both simultaneously. This fit nicely with the modified Monod relationship for microbial growth kinetics given by Bailey and Ollis [11] therefore the rate of substrate consumption by the microorganisms in the pore liquid, $-r_{s,l}$ is expressed as

$$-r_{s,l} = \frac{\mu_m}{Y_s} X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_0 + X_0}) \quad (3)$$

where subscripts s, o and b refer to the substrate, oxygen, and biomass, respectively. The rate of substrate consumption by the microcolonies attached to the surface of soil particles, $-r_{s,s}$, is

$$-r_{s,s} = \frac{\mu_m}{Y_s} M_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o}) \quad (4)$$

The rate of oxygen consumption in the pore liquid

space, $-r_{o,l}$ and that on the surface of soil particles, $-r_{o,s}$ are expressed, respectively, as

$$-r_{o,l} = \frac{\mu_m}{Y_o} X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o}) \qquad (5)$$

$$-r_{o,s} = \frac{\mu_m}{Y_o} M_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o}) \qquad (6)$$

The rate of biomass growth in the form of suspended microorganisms in the pore liquid, $r_{b,l}$ and that at the interface, $r_{b,s}$ are expressed, respectively, as

$$r_{b,l} = \mu_m X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_0}) - k_d X_b \quad (7)$$

$$r_{b,s} = \mu_m M_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o}) - k_d M_b \quad (8)$$

Substituting the above kinetic expressions for substrate consumption, oxygen consumption, and biomass growth into Equation 2 gives rise to

$$(\varepsilon_a + \rho K_{ds})\frac{\partial X_s}{\partial t} = \frac{D_s \varepsilon_a / \tau + D_{s,so}\rho K_{ds}}{r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial X_s}{\partial r})$$

$$-(\varepsilon_a + \rho K_{db})\frac{\mu_m}{Y_s} X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o})$$

$$(9)$$

The adsorption of oxygen in the solid phase is neglected; thus,

$$\varepsilon_a \frac{\partial X_o}{\partial t} = \frac{D_o \varepsilon_a}{\tau r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial X_o}{\partial r})$$

$$-(\varepsilon_a + \rho K_{db})\frac{\mu_m}{Y_o} X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o}) \qquad (10)$$

The diffusion of biomass on the solid phase is negligible; hence, $D_{b,s} = 0$

$$(\varepsilon_a + \rho K_{db})\frac{\partial X_b}{\partial t} = \frac{D_b \varepsilon_a}{\tau r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial X_b}{\partial r})$$

$$+ (\varepsilon_a + \rho K_{db})\mu_m X_b (\frac{X_s}{K_s + X_s})(\frac{X_o}{K_o + X_o})$$

$$- (\varepsilon_a + \rho K_{db})k_d X_b$$

$$(11)$$

Equations 9 through 11 are governing equations for this model.

To better understand the effect of the model parameters on the solution, it is desirable to rewrite equations in dimensionless forms [10].

Thus, the following dimensionless variables and parameters are defined.

Dimensionless concentrations:

$$X_b^* = \frac{X_b R_b}{X_s^\circ R_s Y_s}, X_o^* = \frac{X_o}{X_o^f}, X_s^* = \frac{X_s}{X_s^\circ}$$

Dimensionless diffusion coefficients:

$$D_s^* = \frac{D_s/\tau + D_{s,so}(R_s - 1)}{D_s / \tau}, D_o^* = \frac{D_o}{D_s}$$

$$D_b^* = \frac{D_b}{D_s}$$

Thiele moduli:

$$\phi_g = R / 3\sqrt{\frac{\mu_m \tau X_s^\circ X_o^f}{K_s K_o D_s}}, \phi_d = R / 3\sqrt{\frac{k_d \tau}{D_s}}$$

Saturation parameters:

$$\beta_o = \frac{X_o^f}{K_o}, \beta_s = \frac{X_s^\circ}{K_s}$$

Retardation factors:

$$R_s = 1 + \frac{\rho K_{ds}}{\varepsilon_a}, R_b = 1 + \frac{\rho K_{db}}{\varepsilon_a}$$

Oxygen supply factor:

$$W = \frac{X_s^\circ R_s Y_s}{X_o^f Y_o}$$

Dimensionless time:

$$T = \frac{tD_s}{R^2 \tau}$$

Dimensionless radius:

$$r^* = \frac{r}{R}$$

In these dimensionless expressions, X_s° is the initial concentration of substrate in the bed, and X_o^{f} is the concentration of oxygen at exterior of the aggregate. By transforming Equations 9 through 11, three equations as dimensionless forms are obtained.

$$\frac{\partial X^*{}_s}{\partial T} = \frac{D_s^*}{R_s}\left(\frac{2}{r^*}\frac{\partial X_s^*}{\partial r^*} + \frac{\partial^2 X^*{}_s}{\partial r^{*2}}\right) \qquad (12)$$
$$- 9\phi_g{}^2 X_b^*\left(\frac{X_s^*}{1 + \beta_s X_s^*}\right)\left(\frac{X_o^*}{1 + \beta_o X_o^*}\right)$$

$$\frac{\partial X_o^*}{\partial T} = D_o^*\left(\frac{2}{r^*}\frac{\partial X_o^*}{\partial r^*} + \frac{\partial^2 X_o^*}{\partial r^{*2}}\right) \qquad (13)$$
$$- 9\phi_g{}^2 W X^*{}_b\left(\frac{X_s^*}{1 + \beta_s X_s^*}\right)\left(\frac{X_o^*}{1 + \beta_o X_o^*}\right)$$

$$\frac{\partial X_b^*}{\partial T} = \frac{D_b^*}{R_b}\left(\frac{2}{r^*}\frac{\partial X_b^*}{\partial r^*} + \frac{\partial^2 X_b^*}{\partial r^{*2}}\right)$$
$$+ 9\phi_g{}^2 X_b^*\left(\frac{X_s^*}{1 + \beta_s X_s^*}\right)\left(\frac{X_o^*}{1 + \beta_o X_o^*}\right) - 9\phi_d{}^2 X_b^*$$
$$(14)$$

Initial and Boundary Conditions The appropriate initial and boundary conditions for this mathematical model are as follows:

At   T = 0 $X_o^* = 0.05$ $X_s^* = 1.00$

$X_b^* = X_b^{*\circ}$ $For \ 0 \leq r^* \leq 1$

At $r^* = 0$ $\frac{\partial X_n^*}{\partial r^*} = 0$ $n = o, s, b$

$For \ T \geq o$

At $r^* = 1$ $X_o^* = 1.00$ $X_s^* = 0$

$X_b^* = X_b^{*\circ}$ $For \ T > 0$

4. NUMERICAL SIMULATION

The model equations investigated in the preceding section consist of three Partial Differential Equations (PDEs), which are coupled through the nonlinear reaction terms.

The system of non-linear partial differential equations has been solved using implicit finite difference method. The method employs finite difference to solve initial and boundary value problems to replace each of the equations by central, forward, and backward difference quotient, approximation formula. Gauss - Seidel method has been used for the solution of the resulting tridiagonal system of non - linear algebraic equations.

Since the PDEs are solved using implicit method, regardless of the values, mesh size and time step the solution method is converging the values mesh size and time step, which affect the accuracy of the solution.

In order to capture dynamic behavior of the process completely and accurately, simulation have been conducted with different values of mesh size and time step to check the accuracy of simulations.

5. RESULTS AND DISCUSSION

The values of the parameters for simulation are summarized in Table 1.

To render the numerical values of the parameters as realistic as possible, literature values were extensively reviewed. While a multitude of sources are available for several of the parameters, only one of the sources is listed for each of these parameters on the basis that values

TABLE 1. Parameter Values For Numerical Simulation.

Parameter	Value	Reference
R	$1.0 cm$	[10]
K_{ds}	$10.0 cm^3/g$	[10]
ε_a	0.35	[10]
X°_b	$1.0 \times 10^{-7}\ g/cm^3$	[12]
X^f_o	$8.0 \times 10^{-6}\ g/cm^3$	[11]
D_b	$1.0 \times 10^{-6}\ cm^2/s$	[15]
D_o	$2.0 \times 10^{-5}\ cm^2/s$	[11]
D_s	$7.8 \times 10^{-6}\ cm^2/s$	[13]
$D_{s.so}$	0	[18]
k_d	$5.9 \times 10^{-7}\ /s$	[11]
K_o	$1.0 \times 10^{-8}\ g/cm^3$	[16]
K_s	$1.0 \times 10^{-6}\ g/cm^3$	[11]
K_{db}	$30.0 cm^3/g$	[14]
Y_o	$1.0\ g/g$	[11]
Y_s	$0.4\ g/g$	[11]
μ_m	$5.9 \times 10^{-5}\ /s$	[11]
ρ	$1.6 g/cm^3$	[17]
τ	1.25	[17]

obtained are realistic within the range of experimental conditions.

The initial contaminant concentration is one of the most important parameter that affects significantly the time of remediation in the aggregates.

To estimate the effect of this parameter, 5 cases, with different initial contaminant concentrations, were investigated and are listed in Table 2.

We varied only three numbers (W, ϕ_g, β_s) by changing the initial contaminant concentration and kept the other dimensionless numbers constant.

Tables 3 and 4 show the variable and constant dimensionless numbers for these 5 cases. During each run, the simulation was terminated as soon as the contaminant concentration through out the aggregate reached 1 mg/kg (ppm) or below, and t_b was determined. The results of simulation are presented in Figures 1 through 6, for three cases 1, 3 and 5 with low, middle and high initial

TABLE 2. Effect of Initial Contaminant Concentration on the Time For Remediation.

Case No.	Initial Contaminant Concentration $M_s^\circ, mg/kg$	Bioremediation Time $t_b, Days$	Diffusion Time $t_d, Days$	The Ratio of t_d/t_b
1	12.5	1.32	72.1	54.6
2	125	7	93.8	13.4
3	500	13.3	107.4	8.1
4	1250	18.3	115.13	6.3
5	12500	39.7	137.2	3.45

contaminant concentration, respectively. The bioremediation time for the concentration of the adsorbed substrate to drop below 1ppm at the center of the aggregate for the 5 cases investigates are listed in Table 2. The time

TABLE 3. Variable Dimensionless Numbers.

Case No.	W	ϕ_g	β_s
1	3.18	32	1.25
2	31.8	102.5	12.5
3	127.5	205	50
4	318	324	125
5	3180	1025	1250

TABLE 4. Constant Dimensionless Numbers.

$R_s = 51$	$\beta_\circ = 800$	$D_b^* = 0.13$	$\phi_d = 205$
$R_b = 151$	$D_\circ^* = 2.6$	$D_s^* = 1$	

taken by the contaminants to diffuse out of aggregated t_d, are also given in this table. The values of t_d have been determined by solving Equation 12 for the limiting cases without bioremediation.

As the Table 2 shows, the bioremediation time is less than the diffusion time for all cases.

When the initial contaminant concentration is small (Case 1), the diffusion time is much larger than bioremediation time (t_d/t_b is large).

A small initial contaminant concentration requires a small oxygen supply factor (3.18 for Case 1), thus rendering the overall process to be reaction limited (Figures 1 and 2). The oxygen requirement being low, the oxygen is made available rapidly in the aggregate, and the reaction in the aggregate proceeds quite uniformly. In the case of middle initial contaminant concentration (Case 3), the oxygen requirement being higher, in this way the overall process not to be only, reaction limited (Figures 3 and 4). A large initial contaminant concentration requires more oxygen. The Case 5 demonstrates that for a large initial contaminant concentration, the rate of contaminant diffusion out of the aggregate is significant and microbial growth occurs in the region where both oxygen and substrate are present (Figures 5 and 6). As the contaminant concentration decreases, the zone of active growth moves further towards the center of the aggregate (Figure 6). Because of endogenous metabolism,
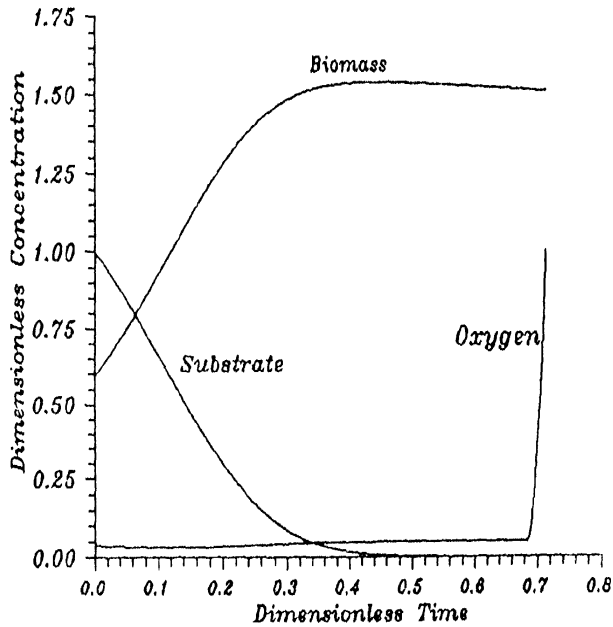
Figure 1. Concentration in the aggregate vs. time for Case 1 at $r^* = 0$, $W = 3.18$, $\varphi_g = 32$ and $\beta_s = 1.25$.
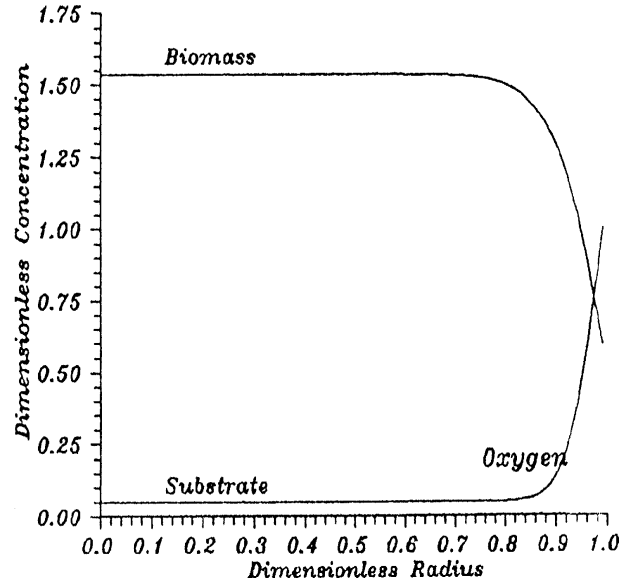

Figure 2. Concentration profiles in the aggregate for Case 1 at $T = 0.5$, $W = 3.18$, $\phi_g = 32$ and $\beta_s = 1.25$.

the dimensionless concentration of biomass is relatively small in Figures 5 and 6. Biomass accumulation does not appear to be sufficient to affect the diffusivity of oxygen. For a large initial substrate concentration, the saturation parameter, β_s, is also large (Table 3), thus indicating that the reaction is of zero order with respect to substrate, and is controlled, by the rate of oxygen diffusion in to the aggregate.

The build up of biomass is low when the initial concentration of contaminant is low. This reduces the biodegradation rate, thereby prolonging the time of remediation.

6. CONCLUSIONS

The natural in situ bioremediation of soil aggregate has been simulated to determine the initial contaminant concentration effect. The results of simulation indicate that microbiological degradation of contaminants within the aggregate may play a significant role in remediation; most of the
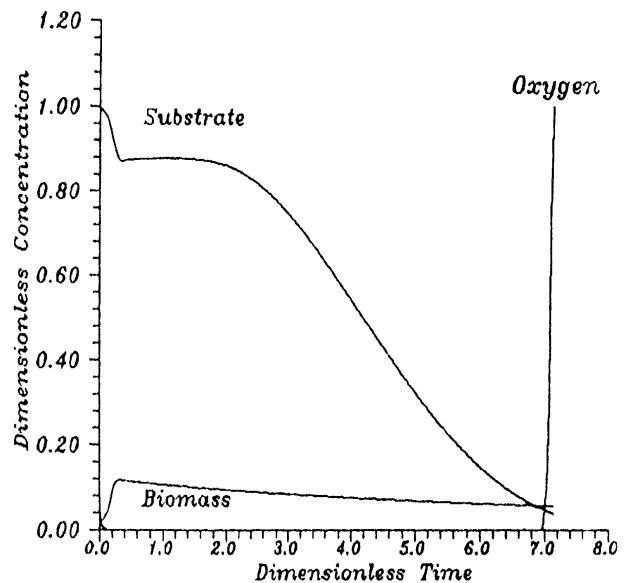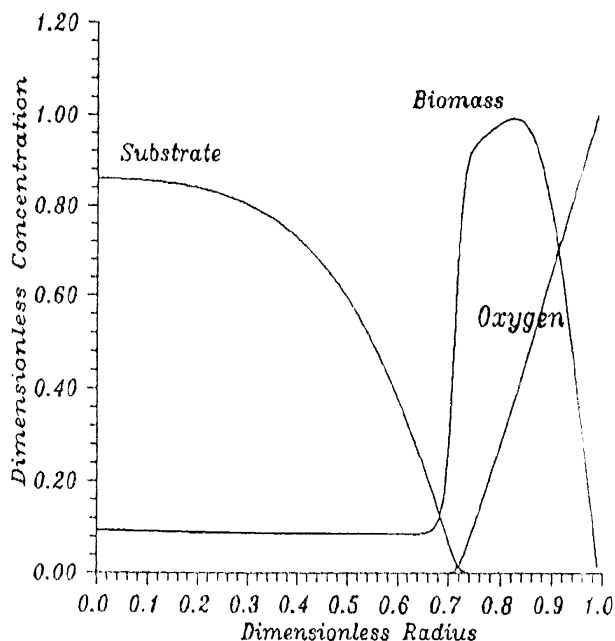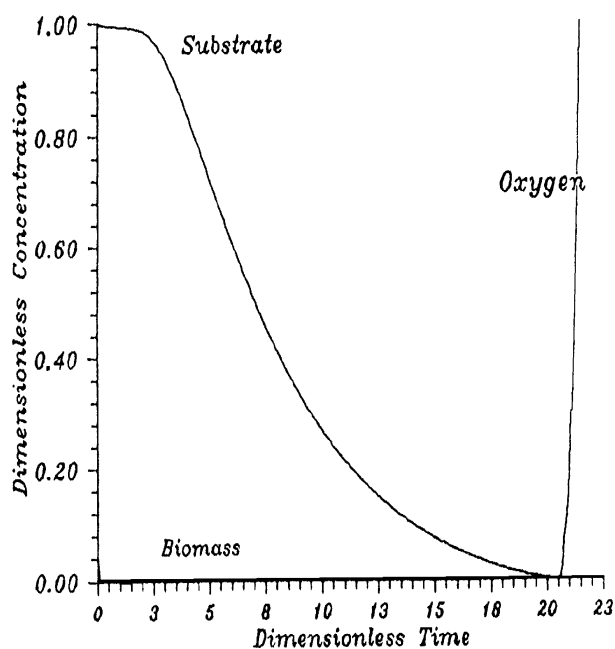

Figure 3. Concentration in the aggregate vs. time for Case 3 at $r^* = 0$, $W = 127.5$, $\phi_g = 205$ and $\beta_s = 50$.

contaminants, which are present initially within the aggregates, are consumed before they reach the outer surface.

Figure 4. Concentration profiles in the aggregate for Case 3 at $T = 0.2$, $W = 127.5$, $\phi_g = 205$ and $\beta_s = 50$.

Figure 5. Concentration in the aggregate vs. time for Case 5 at $r^* = 0$, $W = 3180$, $\phi_g = 1025$ and $\beta_s = 1250$.

On the other hand the results of simulation indicate that treatment methods, which transport oxygen and nutrients to the surface of aggregate, are superior to pump and treat methods, which depend upon the surface treatment of effluent pumped from the contaminated region. Thus, stimulating bioremediation within aggregate has the potential to reduce the treatment time compared to diffusion of the contaminant out of aggregate.

Finally it is pointed out through the simulation approach to be interesting from the engineering point of view and include some findings of the effect of diffusion rates of substrate and oxygen and the biodegradation rate on attenuation of chemicals. This model can be simulated to estimate the remediation time if reasonable values of the diffusivity and the kinetic parameter for biodegradation are available.

7. NOMENCLATURE

$X_b^{*\circ}$	dimensionless concentration of biomass originally present in the pore liquid
X_n	concentration of component n in the pore liquid, M/L^3
X_n^{*}	dimensionless concentration of component n in the pore liquid
X_o^{f}	concentration of oxygen at the exterior of the aggregate, M/L^3
D_n^{*}	dimensionless diffusion coefficient of component n
D_n	diffusion coefficient of component n in the pore liquid, L^2/T
$D_{n,so}$	surface diffusion coefficient of component n in the solid phase, L^2/T
k_d	reaction rate constant for the decay of biomass, T^{-1}
K_o	saturation constant of oxygen, M/L^3
K_s	saturation constant of substrate, M/L^3
K_{dn}	linear partition coefficient of component n, L^3/M
M_n	concentration of component n, in the solid phase, M/M
r	radial position in the aggregate, L
R	radius of the aggregate, L
R_n	retardation factor for component n
$r_{n,l}$	reaction rate in liquid phase, $M/L^3/T$

$r_{n,s}$	reaction rate at the interface, M/M/T
t	time, T
t_d	time taken to diffuse the substrate out of the aggregate, T
t_b	time of bioremediation of the aggregate, T
W	oxygen supply factor
Y_o	yield factor of oxygen, M/M
Y_s	yield factor of substrate, M/M

Greek Letters

β_n	saturation parameter of component n
ε_a	volumetric fraction of liquid in the aggregate: (porosity)
μ_m	maximum specific growth rate of biomass, T^{-1}
ρ	bulk density of the aggregate particle, M/L^3
T	dimensionless time
ϕ_g	Thiele modulus for the growth of biomass
ϕ_d	Thiele modulus for the decay of biomass
τ	tortuosity of pores in the aggregate

Subscripts

n	b,o,s for biomass, oxygen, and substrate , respectively
l	pore liquid
s	solid interface
o	solid phase

Figure 6. Concentration profiles in the aggregate for Case 5 at $T = 1$, $W = 3180$, $\phi_g = 1025$ and $\beta_s = 1250$.

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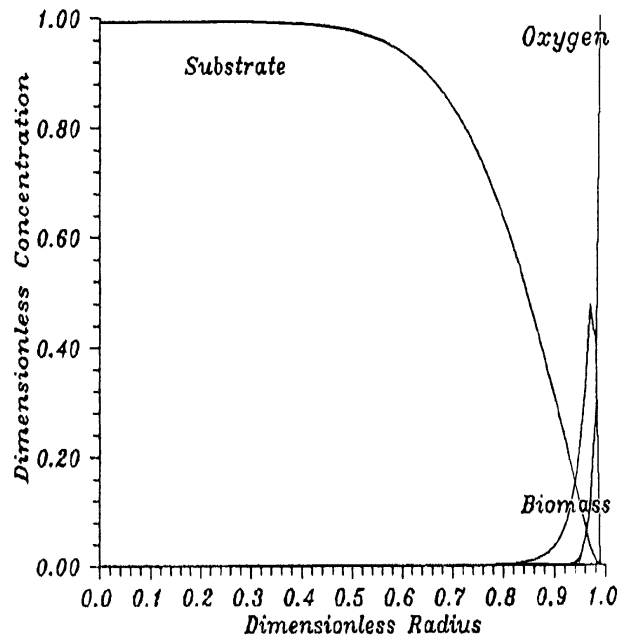4. Rifai, H. S., Bedient, P. B., Wilson, J. T., Miller, K. M. and Armstrong, J. M., "Biodegradation Modeling at Aviation Fuel Spill Site", *J. Envir. Engrg.*, 114, (1988), 1007-1029.

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