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# MODELING OF SUBSURFACE BIOBARRIER FORMATION

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## ABSTRACT

Biofilm-forming microbes can form biobarriers to inhibit contaminant migration in groundwater and potentially biotransform organic contaminants to less harmful forms. Biofilm-forming microbes thereby provide an *in situ* method for treatment of contaminated groundwater. A mathematical and numerical model to describe the population distribution and growth of bacteria in porous media is presented here. The model is based on the convection-dispersion equation with nonlinear reaction terms. Accurate numerical simulations are crucial to the development of contaminant remediation strategies. We use the nonstandard numerical approach that is based on non-local treatment of nonlinear reactions and modified characteristic derivatives. This approach leads to significant qualitative improvements in the behavior of the numerical solution. Numerical results for a simple biobarrier formation model are presented to demonstrate the performance of the proposed new method. Comparisons of simulated results with experimental results obtained from the Montana State Center for Biofilm Engineering are also presented.

**Key words:** biobarriers, models, biofilms, simulations

## INTRODUCTION

Controlling pollution in underground water is a very important and difficult problem. There are bacteria that will destroy many organic contaminants in subsurface regions (Characklis and Marshall, 1990). But for most pollutants, including heavy metals, a more promising concept is the creation of biobarriers for containment and remediation of contaminated soil and groundwater (James et al., 1995). Biobarriers are in situ barriers that are formed by stimulating the growth of biofilm-forming microbes that are either already present or introduced into the aquifer. As the microbial biomass increases, it plugs the free-pore-space flow paths through porous media, thereby reducing the hydraulic conductivity and mass transport properties (Cunningham et al., 1991). By adequately choosing where to plug the porous medium, it is possible to prevent the migration of groundwater contaminants from hazardous waste sites. An even better scenario is to have biobarriers that will not only contain the contaminant plume but will also degrade it.

Mathematical models are needed to complement experimental work in the use of biofilms to form biobarriers. Mathematical models help to understand the mechanisms for flow, solute transport, biological and chemical reactions, biofilm accumulation, and natural biodegradation in porous media. The equations describing the mathematical models generally lead to strongly coupled systems of nonlinear ordinary and partial differential equations that are difficult to solve (Chen et al., 1994). Analytical solutions for the full, coupled problem are non existent and numerical methods have problems such as instabilities and artificial diffusion (Morton, 1996).

In this article, we use new methods that are reliable, accurate, and efficient for the model describing subsurface biobarrier formation (Kojouharov and Chen, 1999). Without these methods, results of numerical simulations are of unstable nature. We compare the results obtained from using our numerical simulator with some of the experimental results for short cores presented in Cunningham et al. (1991). The results compare well, which is a good validation of the model. The simulator can now be used as a predictive tool to determine values of parameters that are difficult or impossible to measure, and to help design experiments, field studies, and actual biobarriers.

The outline of the paper is as follows. In the next section, the governing system of differential equations is formulated for a three-phase, four-species mixture. In Section 3, the non-standard numerical method for solving the reactive solute transport problem in porous media is given. To demonstrate the performance of the proposed method of solution for the model and the effective-ness of biobarriers for reducing the hydraulic conductivity, numerical results and comparisons with experiments are presented in Section 4. In the last section, a summary of results is presented.

## GOVERNING SYSTEM OF EQUATIONS

Consider a three-phase mixture consisting of a liquid phase, a solid rock phase, and a biofilm phase. Even though the biofilm can be considered to be part of the solid phase, it is simpler to take it as a separate phase. The four molecular species present in the porous medium are the biofilm-forming microbes, labeled *M*; the soluble contaminants or nutrients, labeled *N*; and the water and rock species. We assume that interactions in the system occur only between the microbial and nutrients species. Furthermore, we assume that the microbes are immobile, i.e., they are attached to the rock as biofilm.

The fundamental equation for saturated transient groundwater flow of constant density, in horizontal direction, can be written in the form (Allen, 1988):

$$S_s \frac{\partial h}{\partial t} - \frac{\partial}{\partial x}\left( K \frac{\partial h}{\partial x} \right) = f. \qquad \text{(fluid flow)} \qquad (1)$$

The single fluid-flow equation (1) arises from the mass balance law

$$S_s \frac{\partial h}{\partial t} + \frac{\partial}{\partial x} = f, \qquad (2)$$

when we substitute for the specific discharge vector *v* using the Darcy's law

$$v = -K \frac{\partial h}{\partial x}. \qquad (3)$$

Here $h$ denotes the hydraulic head, $S_s$ is the specific storage, $K$ is the saturated hydraulic conductivity, and $f$ represent sources or sinks. The specific discharge vector $v$, called Darcy velocity, represents the speed of the water.

The transport and reaction of nutrients and the growth of microbes are governed by a system of partial differential equations (Allen, 1988). Since the rock phase doesn't change, we assume that the solid rock matrix is stationary and that the diffusion of microbial and nutrient species in the solid phase is negligible. Therefore, we can work only with the liquid and biofilm phases:

$$\frac{\partial}{\partial t}\left(\phi^B \rho_M\right) = r_M\left(\rho_M, \rho_N\right) \qquad \text{(microbes)}$$

$$(4)$$

$$\frac{\partial}{\partial t}\left(\phi^L \rho_N\right) + \frac{\partial}{\partial x}\left(v\rho_N\right) - \frac{\partial}{\partial x}\left(D_N \frac{\partial \rho_N}{\partial x}\right) = r_N\left(\rho_M, \rho_N\right). \qquad \text{(nurtients)}$$

Here $\rho_i$ $(i = M, N)$ represents the intrinsic mass density of microbes and nutrients, respectively. For a single-fluid flow, the quantity $\phi^L = V_L/(V_L + V_B)$ and the quantity $\phi^B = V_B/(V_L + V_B)$ where $V_L$ and $V_B$ represent the volumes occupied by the liquid and by the biofilmk, repectively, $D_N$ is the hydrodynamic dispersion coefficient for the nutrients, and $r_i$ represents the total rate at which species $i$ is produced via reactions and sources.

The microbial death rate is assumed to be proportional to the size of the biofilm population. The rate of biofilm growth is given by the Monod model

$$\mu(S) = \frac{\mu_{max} S}{K_S + S}, \qquad (5)$$

where $\mu_{max}$ is the maximum specific growth rate, and $K_S$ is the value of the concentration of nutrients $S$ where the specific growth rate $\mu(S)$ has half its maximum value (Bailey and Ollis, 1986). We assume that only the growth and accumulation of biofilm in the pore spaces cause changes in the porous media properties. Let $X_f$ be the current biofilm concentration, then $\tilde{X}_f = X_f/r_M$ is the normalized biofilm concentrations (Clement et al., 1996), is given by

$$\phi = \phi_0\left(1 - \tilde{X}_f\right), \qquad (6)$$

where $\phi_0$ is the clean surface porosity. For the saturated hydraulic donductivity K, we assume the following form

$$K = K_0\left(1 - \tilde{X}_f\right)^{n_k}, \qquad (7)$$

where $K_0$ is the initial hydraulic conductivity and $n_k$ is an experimentally determined parameter titiwhich takes values around 3 (Clement et al., 1996). For simplicity, from now on we will drop the

tilde from the normalized biofilm concentration. We assume there are no sources and sinks for the fluid, therefore *f=0* in Equation (1). We also assume a piecewise steady, state fluid flow, due to the relatively slow changes in the porous media properties (Cunningham et al., 1991). Also, we are modeling very short cores with uniform biofilm distribution so we can take the velocity to be independent of *x* (Cunningham et al., 1991).

Invoking all simplifying assumptions to Equations (1) and (4) and using concentrations as the unknows gives the final form of the governing system of differential equations:

$$-\frac{\partial}{\partial x}\left(K(X_f)\frac{\partial h}{\partial x}\right)=0,$$

$$\frac{\partial X_f}{\partial t}=\frac{\mu_{max}S}{K_S+S}X_f-k_r X_f, \tag{8}$$

$$\frac{\partial S}{\partial t}+v\frac{\partial S}{\partial x}-\frac{\partial}{\partial x}\left(D_N\frac{\partial S}{\partial x}\right)=-\frac{1}{Y}\frac{\mu_{max}S}{K_S+S}X_f,$$

where $k_r$ is the first-order endogenous decay rate and *Y* is the yield rate coefficient (Bailey and Ollis, 1986).

## NUMERICAL METHODS FOR BIOFILM GROWTH

Equation (8) represents a coupled system of nonlinear, time-dependent partial differential equations and ordinary differential equations that is very difficult to solve numerically. A key objective of the numerical simulation is to develop time-stepping procedures that are accurate and computationally stable. Different time-stepping ideas can be applied to solve the governing system of equations (Russell and Wheeler, 1983). One possible time-stepping approach is the sequential solution technique (see Figure 1).

The sequential method first solves implicitly for the Darcy velocity *v* at the current time level by solving the fluid-flow equation (1). Then the species transport system (4) is solved implicitly for the concentrations *S* and $X_f$, in a decoupled fashion (Ewing and Russell, 1982). New values of porosity and permeability are then calculated and the cycle is repeated by calculating the new velocities.

For the solution of the ordinary differential equation in System (8), modeling the fluid flow, we use a standard finite-difference method to calculate *h*. Then we numerically differentiate using Equation (3) to get the velocity field *v*.

Unfortunately, there are only few cases for which analytic solutions to the solute transport equation in System (8) exist. The form of the convection-dispersion-reaction equation ranges from
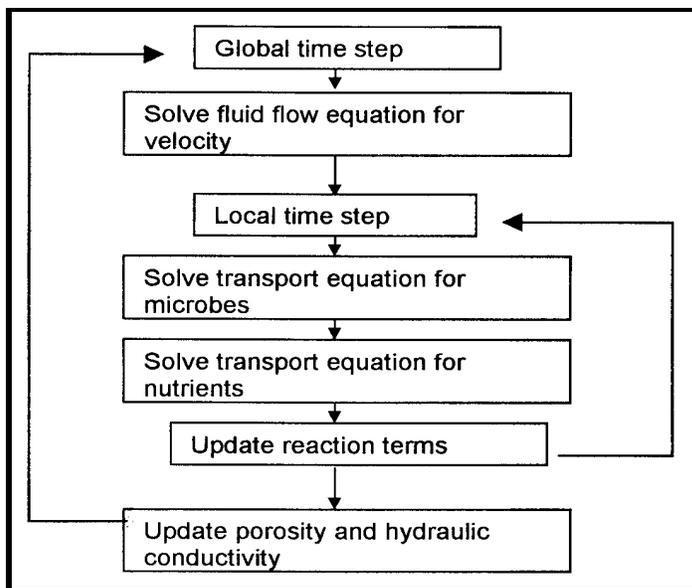
**Figure 1.** Sequential time-stepping procedure.

parabolic to almost hyperbolic, depending on the ratio of convection to dispersion (Chiang et al., 1989). One can measure the degree of convection dominance via the dimensionless Peclet number $Pe = vL/D_N$ , where $L$ is the length of the domain (Liu et al., 1996). When $Pe<<1$, the equation resembles the heat equation, which rapidly smooths sharp fronts. When $Pe>>1$, sharp fronts and plumes remain sharp and cause numerical difficulties. Typically, the criterion for oscillation-free solution requires that the grid Peclet number $Pe = Pe\mathbf{D}x/L = O(1)$ (Jensen and Finlayson, 1980). However, in underground flows with field-scale pressure gradients applied by pumping wells, Peclet numbers greater than $10^2$ are common (Lake and Hirasaki, 1981), so the near-hyperbolic regime is important in engineering applications. While classical numerical techniques, such as the standard finite-differences or Galerkin finite-elements, work well for problems of solute transport that are dominated by dispersive movement, they suffer from severe nonphysical oscillations and excessive numerical dispersion when convection dominates the dispersive effects (Morton, 1996). Solutions of hyperbolic-type equations can be represented from the initial data propagating over characteristic paths in the surface and can be viewed as dispersing away from these paths, along which the concentration is a smooth function (Douglas and Russell, 1982). Therefore, it is logical to design numerical procedures that recognize the hyperbolic nature of the convection-dominated solute transport problems, such as the Eulerian-Lagrangian methods (Celia et al., 1990). In recent years, many such schemes have been developed (Healy and Russell, 1993; Allen and Liu, 1995), but still little has been done to improve the numerical solutions of problems in which nonlinear reactions are present. Nonlinear reaction terms play a significant role in applications involving bacterial growth and contaminant biodegradation in subsurface regions (Chiang et al., 1991; Liu et al., 1996).

In Kojouharov and Chen (1999), we proposed a new Eulerian-Lagrangian numerical method for solving the reactive solute transport equation that works very well for Peclet numbers large and small. The numerical solution of the convection-reaction part is defined using an "exact" time-stepping scheme (Kojouharov and Chen, 1998). This enables us to follow the transport and track sharp fronts much more accurately than with the standard numerical schemes. Having dealt with the most difficult part of the transport problem, only the smoothing property of the dispersion term remains. Then, standard finite differences or finite elements are well suited for solving the dispersion part.

We now apply the new method to the following dispersion-free system of differential equations:

$$\frac{\partial X_f}{\partial t} = \frac{\mu_{max} S}{K_S + S} X_f - k_r X_f, \qquad \text{(microbes)}$$

$$\tag{9}$$

$$\frac{\partial S}{\partial t} + v \frac{\partial S}{\partial x} = -\frac{1}{Y} \frac{\mu_{max} S}{K_S + S} X_f. \qquad \text{(nutrients)}$$

The microbes' equation is a linear, first-order ordinary differential equation whose "exact solution" is given by

$$\frac{X_f^{m+1}(x) - X_f^m(x^m)}{\dfrac{e^{\lambda^m \Delta t} - 1}{\lambda^m}} = \lambda^m X_f^m(x^m), \tag{10}$$

where $\lambda^m = \dfrac{\mu_{max} S^m(x)}{(K_s + S^m(x))} - k_r,$ and the bactrack point $\overline{x}^m$ has the expression

$$\overline{x}^m = x - \left[ P_n((m+1)\Delta t) - P_n(m\Delta t) \right],$$

for constant in space, time-dependent velocity fields: $v(t) = P_{n-1}(t)$.

The "exact" time-stepping scheme for solving the nutrients transport equation from System (9) is given by the expression

$$\frac{S^{m+1}(x) - S^m(\overline{x}^m)}{\Delta t} = \lambda^m - \frac{K_s}{\Delta t} \ln\left( \frac{S^{m+1}(x)}{S^m(\overline{x}^m)} \right), \tag{11}$$

where $\lambda^m = -\dfrac{\mu_{max} X_f^{m+1}(x)}{Y}.$

Applying the "exact" time-stepping scheme (11) to the dispersive nutrients transport equation from System (8) yields the following implicit-in-nature, semi-discrete procedure

$$\frac{S^{m+1}(x) - S^m(\overline{x}^m)}{\Delta t} - \frac{\partial}{\partial x}\left( D_M^{m+1} \frac{\partial S^{m+1}(x)}{\partial x} \right) = \lambda^m - \frac{K_S}{\Delta t} \ln\left( \frac{S^{m+1}(x)}{S^m(\overline{x}^m)} \right). \tag{12}$$

To complete the construction of the new Eulerian-Lagrangian method we need to introduce an approximation technique for discretizing the spatial derivatives involved in the dispersion term from Equation (12). Let us consider the centered, weighted second difference approximation (Huyakorn and Pinder, 1983):

$$\frac{\partial}{\partial x}\left( D_M^{m+1} \frac{\partial S^{m+1}(x)}{\partial x} \right)_i \approx \delta_{\overline{x}}\left( D_N^{m+1} \delta_x S^{m+1} \right) = \frac{D_{M_{i+\frac{1}{2}}}^{m+1}\left( S_{i+1}^{m+1} - S_i^{m+1} \right) - D_{M_{i-\frac{1}{2}}}^{m+1}\left( S_i^{m+1} - S_{i-1}^{m+1} \right)}{\Delta x^2}, \tag{13}$$

where

$$D_{M_{i+\frac{1}{2}}}^{m+1} = D_M\left( \left( \frac{x_i + x_{i+1}}{2} \right), (m+1)\Delta t \right) \tag{14}$$

is the hydrodynamic dispersion coefficient located at the center of a space increment.

Combining the semi-discrete procedure (12) with the above spatial approximation of the dispersion term yields the non-standard difference method for solving the nutrients transport equation:

$$\frac{S_i^{m+1} - S^m(\overline{x}_i^m)}{\Delta t} - \delta_{\overline{x}}(D_N^{m+1}\delta_x S^{m+1})_i = \lambda_i^m - \frac{K_S}{\Delta t} \ln\left( \frac{S_i^{m+1}}{S^m(\overline{x}_i^m)} \right), \tag{15}$$

where $\lambda_i^m = -\dfrac{\mu_{max} X_{fi}^{m+1}}{Y}$ and the backtrack point $\overline{x}_i^m$ has the expression

$$\overline{x}_i^m = x_i - \left[ P_n\left( (m+1)\Delta t \right) - P_n\left( m\Delta t \right) \right].$$

**Remark.** In general, the *"backtrack"* point $\overline{x}_i^m$ does not lie at a grid point. If the approximates solution $S$ is being determined by a finite-difference procedure, the convective concentration $S^m(\overline{x}_i^m)$ must be evaluated by an interpolation of the approximate solution values $\{ S_i^m \}$ at the grid points $x_i$.

## NUMERICAL RESULTS

We now turn to a set of numerical experiments to demonstrate the performance of the proposed new method and the effectiveness of microbial barriers for reducing the hydraulic conductivity property of porous media. The governing system of equations examined here has the following form:

$$-\frac{\partial}{\partial x}\left(K\frac{\partial h}{\partial x}\right)=0, \qquad v=-K\frac{\partial h}{\partial x} \qquad\qquad \text{(fluid flow)}$$

$$\frac{\partial x_f}{\partial t}=\frac{\mu_{max}S}{K_S+S}G(X_f)X_f-k_rX_f, \qquad\qquad \text{(microbes)} \qquad (16)$$

$$\frac{\partial S}{\partial t}+v\frac{\partial S}{\partial x}-D_N\frac{\partial^2 S}{\partial x^2}=-\frac{1}{Y}\frac{\mu_{max}S}{K_S+S}G(X_f)X_f, \qquad\qquad \text{(nutrients)}$$

where $\quad G(X_f)=\dfrac{1-X_f}{1-X_f+\gamma},$

with $\boldsymbol{g}$ typically small, is introduced to restrict the growth of the microbes as the pores are being plugged (Freter et al., 1986; Jones and Smith, 2000); $h$ is the hydraulic head; $X_f$ is the normalized biofilm concentration; and $S$ is the nutrients concentration. Assumptions made in the above mathematical model (16) are that all bacteria are attached to the solid rock surface, as a part of the biofilm structure, and that the concentration of nutrients present in the solid phase is negligible. Changes in the hydraulic conductivity $K$ are caused by the accumulation of soild-phase biomass in the pore spaces. The biofilm concentration-porosity relation used is given by Equation (6). The conductivity-reduction relationship examined here is given by Equation (7) with $n_k=3$.

**Table 1.** Parameters used in the mathematical model.

| | Columns | |
| --- | --- | --- |
| *Parameters* | **0.70 mm sand** | **0.54 mm sand** |
| Initial (clean surface) permeability, $k_0$ | $3.19 \times 10^{-6}$ cm$^2$ | $2.17 \times 10^{-6}$ cm$^2$ |
| Hydraulic conductivity, $K_0$, for water at 15°C | 0.2404 cm/s | 0.1635 cm/s |
| Initial porosity, $\boldsymbol{f}_0$ | 0.35 | 0.35 |
| Maximum specific growth rate, $\boldsymbol{m}_{max}$ | $1.041 \times 10^{-4}$ /s | $1.041 \times 10^{-4}$ /s |
| Saturation constant, $K_S$ | 0.799 mg/L | 0.799 mg/L |
| Yield coefficient, $Y$ | 0.0975 | 0.0975 |
| Endogenous decay coefficient, $k_r$ | $7.161 \times 10^{-5}$ /s | $7.161 \times 10^{-5}$ /s |
| Dispersion coefficient, $D_N$ | $5 \times 10^{-4}$ cm$^2$/s | $5 \times 10^{-4}$ cm$^2$/s |
| Gamma, $\boldsymbol{g}$ | 0.1 | 0.1 |
| *Initial conditions at t=0* | | |
| Nutrients concentration, $S(x,0)$ | 0.5 mg/L | 0.5 mg/L |
| Normalized biofilm concentration, $X_f(x,0)$ | 0.02 | 0.02 |
| *Boundary conditions* | | |
| Hydraulic head at $x=0$, $h(0,t)$ | 0.5 cm | 0.5 cm |
| Hydraulic head at $x=1$, $h(1,t)$ | 0 cm | 0 cm |
| Nutrients concentration at $x=0$, $S(0,t)$ | 0.5 mg/L | 0.5 mg/L |
| Nutrients concentration gradient at $x=1$, $\frac{\partial S}{\partial x}(1,t)$ | 0 mg/LT | 0 mg/LT |
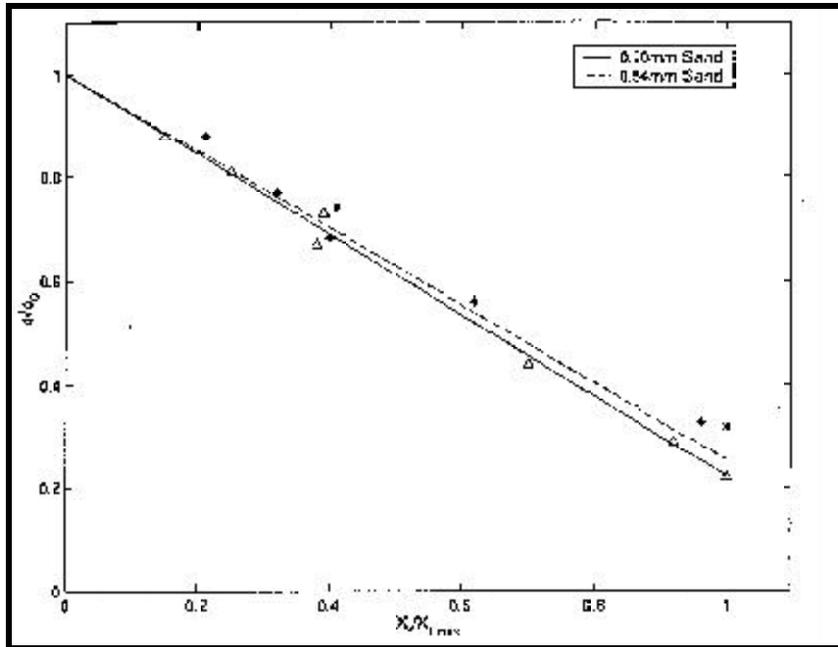
**Figure 2.** Variation in simulated normalized media porosity with normalized biofilm concentration. The triangles and the stars represent the experimental values for .70 mm and .54 sands, respectively, from Cunningham et al., (1991).

We stimulate two of the experiments done by Cunningham et al. (1991) for a 5-cm-long reactor packed with 0.70 mm and 0.54 mm (in diameter) sands. For ease of calculations, the reactor's lenght was scaled to 1, i.e., spatial domain $\Omega=[0,1]$, and the nutrients concentration was scaled by a factor of 50 for graphing purposes. The parameters used in the mathematical model, for both types of porous media, are summarized in Table 1 on the previous page.

The boundary and initial conditions considered in the model are in agreement with Cunningham et al. (1991); the reaction parameters are taken from Taylor and Jaffe, (1990); and the gamma parameter in the function G is taken from Jones and Smith, (2000).

The figures present the results of our calculation, together with some of the experimental values shown in Figures 5 and 8 from Cunningham et al. (1991). We use concentrations instead of biofilm thickness since we cannot calculate the thickness without making assumptions on the distribution of microbes. But, it is reasonable to assume that there is a linear relation between biofilm thickness and microbial concentration.

Figure 2 shows the variation of the normalized porosity with the normalized biofilm concentration. $X_{fmax}$ is the maximum value of the microbial concentration and symbols represent some experimental results.

Figure 3 is a plot of the permeability decrease and the increase in the microbial concentration with time. In our results the normalized biomass goes to 1 in about two days, the same time it takes the normalized permeability to decrease to about 0.1. In Figure 5 of Cunningham et al. (1991), the permeability also decays to 0.1 in about two days, but the normalized biofilm thickness takes about

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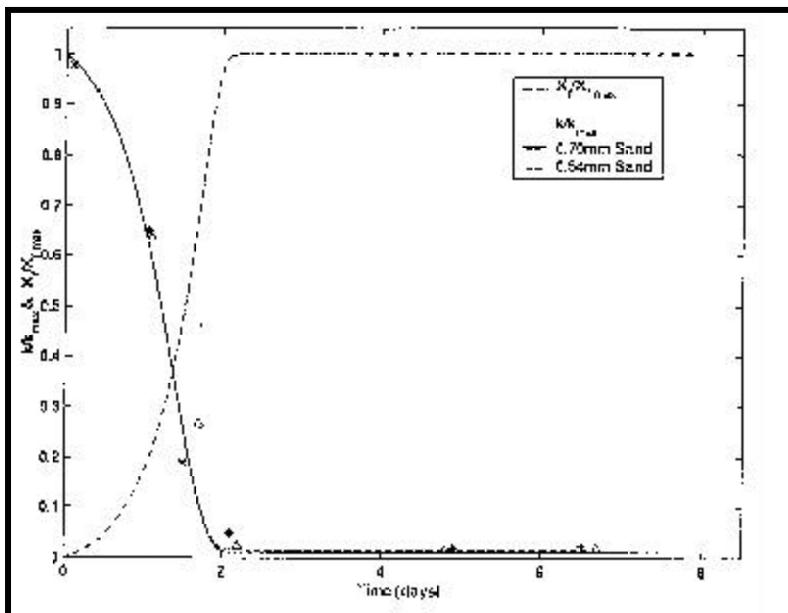
**Figure 3.** Normalized porous media permeability decrease corresponding to increased normalized microbial concentration versus time. The microbial concentration curve is the average for both types of sand. The triangles and the stars represent experimental permeability values for .70 mm sands and .54 mm sands, respectively, from Cunningham et al., (1991).

six days to tend to 1. The difference is due to the averaging of the biofilm thickness done by Cunningham et al. (1991), where the dominant component is for 1mm glass spheres (experiment not modeled in this article). Note that Cunningham et al. (1991) use permeability $k$ instead of the more widely used hydraulic conductivity $K$. The relation between the two is $K=k\,\textbf{r}g/\textbf{m}$, with $\textbf{r}$ and $\textbf{m}$ the density and viscosity of water, respectively, and $g$ the acceleration of gravity.

Figure 4 shows the growth of biomass together with the decrease in nutrients. The amount of biomass reaches a maximum steady state at about two days, which coincides with the time it takes for the nutrients to reach their minimum.

The agreement is very good and shows that the model can reproduce experimental results and that in the future can be used as a predictive tool. However, the curves in Figure 2 are closer together than the corresponding experimental ones. One reason is that we are plotting biomass concentrations instead of biofilm thickness. Another possible reason is that we took all the bacteria to be in biofilm form with no significant detachment, so all the biomass reduces the porosity and permeability. In practice there is detachment and the free-floating microbes will not change the physical properties of the medium. Also, for the 0.54 mm sand, the pore channels are smaller and the velocities higher, which would increase the detachment in this case and add to the separation of the curves.
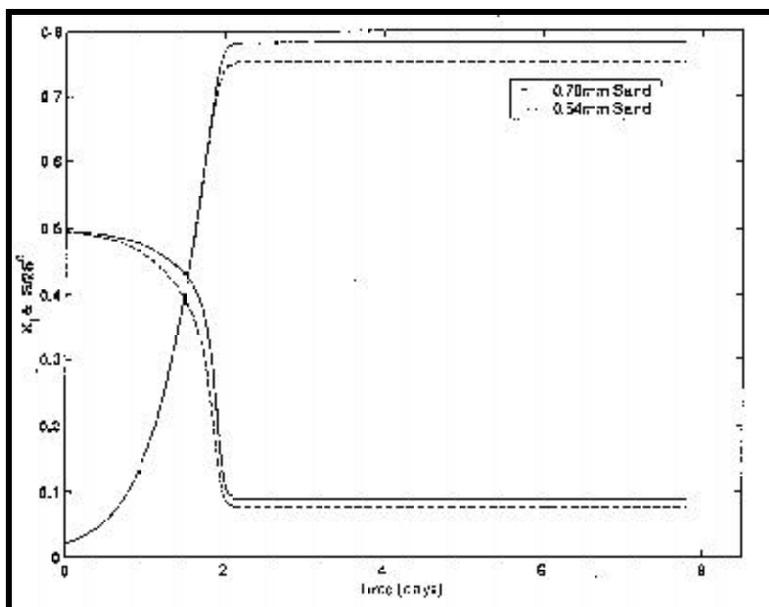
**Figure 4.** Increase in microbial concentration and decrease of normalized nutrient concentration with time.

## CONCLUSIONS

A new class of numerical methods has been developed for solving one-dimensional, transient convective-dispersive transport equations with nonlinear reactions. Large time steps can be taken without affecting the accuracy of the numerical solution. The appropriate time step size for a particular model problem can be determined by physical considerations, rather than stability, convergence, or consistency reasons.

The proposed new methods have been successfully applied to biobarrier formation models incorporating Monod kinetics. Numerical results confirmed the theoretical and experimental predictions that microbial barriers are effective for manipulating the porous media properties in general, and for reducing the hydraulic conductivity in particular.

## NOMENCLATURE

The symbols L, M, and T denote the dimensions of length, mass, and time, respectively. (See Table 2.)

## ACKNOWLEDGEMENTS

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**Table 2.** The symbols L, M, and T denote the dimensions of length, mass, and time, respectively.

| Parameter | Symbol | Dimensions |
|---|---|---|
| Hydraulic head | $h$ | $L$ |
| Specific discharge | $v$ | $L/T$ |
| Hydraulic conductivity | $K$ | $L/T$ |
| Specific storage | $S_s$ | $L/T$ |
| Point source or sink | $f$ | $1/T$ |
| Volume fractions | $f^L$, $f^B$ | dimensionless |
| Intrinsic mass densities | $r_M$, $r_N$ | $M/L$ |
| Dispersion coefficient | $D_N$ | $L^2/T$ |
| Maximum specific growth rate | $m_{max}$ | $1/T$ |
| Saturation constant | $K_S$ | $M/L$ |
| Yield coefficient | $Y$ | dimensionless |
| Endogenous decay coefficient | $k_r$ | $1/T$ |
| Nutrients' concentration | $S$ | $M/L$ |
| Biofilm concentration | $X_f$ | $M/L$ |
| Normalized biofilm concentration | $\widetilde{X}_f$ | dimensionless |
| Porosity | $f$ | dimensionless |

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