Development and evaluation of a two-dimensional microscale transport and biofilm process model
by Ernest Jay Visser

A thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Civil Engineering
Montana State University

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DEVELOPMENT AND EVALUATION OF A TWO-DIMENSIONAL MICROSCALE TRANSPORT AND BIOFILM PROCESS MODEL

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A thesis submitted in partial fulfillment
of the requirements for the degree

of

Doctor of Philosophy

in

Civil Engineering

MONTANA STATE UNIVERSITY-BOZEMAN
Bozeman, Montana

July 1998

APPROVAL

of a thesis submitted by

Ernest Jay Visser

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

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ACKNOWLEDGEMENTS

I would like to say thank you to my committee members who contributed advice and insight into my research. Special thanks for patience and advice goes to my thesis adviser Dr. Al Cunningham and Dr. Benito Chen and Dr. Marty Hamilton for their patience and perseverance in helping me complete my graduate work. Thanks also to Brian Goldstein for computer related support and putting up with my computational requirements. I would like to say thank you to all the people at the Center for Biofilm Engineering who made my stay a pleasant time and also supplied many interesting discussions of related research.

A special thanks also to my parents for their support while I was a student.

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ABSTRACT

A mathematical model describing the interactions of fluid dynamics, substrate transport, and biofilm reaction is formulated. Equations modeling the processes of fluid dynamics, substrate transport, and biofilm reaction are solved. The Finite Difference method is used to solve coupled partial differential equations. A perturbation scheme is formulated that can account for the influence of thin biofilms on the boundaries. This model is compared with available experimental dissolved oxygen data. Application of the model demonstrates the influence that biofilm distribution can have on biofilm system performance. Simulation of capillary tubes demonstrates the influence of domain geometry on local velocity and concentration distributions.

CHAPTER 1

INTRODUCTION

Biofilms can be found in most natural and industrial aquatic systems and can account for significant microbial activity in these systems (Characklis and Marshall, 1990). Biofilms are becoming recognized as a prominent mode of growth for bacteria (Potera, 1996). Biofilms can be beneficial or detrimental in environmental systems (Mattila-Sandholm and Wirtanen, 1992; Jass and Lappin-Scott, 1997). For example, biofilm play a substantial role in degrading xenobiotic compounds in groundwater and soil. Significant problems caused by biofilms include: biofouling in cooling towers which costs more than $7 billion U.S. Dollars for biocides to control (Lappin-Scott et al., 1992); biofilms found on medical implants, poses serious health risks (Lappin-Scott et al., 1992); biofilm plaque on teeth, which can increase risk of cavities (Marsh, 1995); and biofilm growth in drinking water systems, which can present a health risk (Camper, 1994; Camper et al., 1994; Mittleman, 1995; Gagnon et al., 1997). Biofilms can cause significant pressure losses in industrial pipeline processes (Picologlou et al., 1980; Butterworth, 1981; Lira and Sengupta, 1990) and increased heat transfer resistance (Lira and Sengupta, 1990). Biofilms may be found in harsh environments, including those contaminated by acid mine drainage (Boult et al., 1997).

In porous media biofilm growth can cause permeability reduction through pore blocking. The growth of sulfate reducing bacteria in oil bearing formations may cause souring of petroleum reservoirs through sulfide production. Conversely, beneficial aspects of biofilms in porous media include bioremediation of contaminant plumes.

Biofilm Processes in Uniform Flow Systems

Processes governing fluid dynamics, mass transport, biofilm accumulation, and the biotransformation of organic constituents are intrinsically interrelated. Suspended cells are transported to the substratum where they may adsorb. Some fraction of these adsorbed cells desorb and return into suspension perhaps through some diffusion-like process. If environmental conditions are favorable, the adsorbed cells grow, replicate, and form a matrix composed of cells and extracellular polymeric substance (EPS), which binds the cells together. The aggregate of attached cells, EPS, and other particulate matter is termed a biofilm (Characklis and Marshall, 1990; Costerton and Lappin-Scott, 1995). Once a biofilm is established, additional cells and particulate matter may attach to and detach from the biofilm surface.

Biofilm Processes in Porous Media

In porous media, biofilm accumulation may cause bridging across the pore channel, thereby further enhancing accumulation by the filtration of particulate matter from suspension. The net accumulation of biofilm in porous media is defined by the difference between biomass added to the surface from the processes of adsorption, growth, attachment, and filtration and the amount of mass removed by the processes of desorption and detachment.

Porous media biofilms may develop as patchy discontinuous colonies, or the colonies may grow together and form a continuous film. The nature of the accumulation pattern may have a strong impact on behavior of microbial biofilm processes in porous media (Baveye and Valocchi, 1989). As film thickness increases, the effective pore space of the media will decrease, thereby causing a corresponding decrease in media porosity and permeability (Cunningham et al., 1991; Taylor et al., 1990). If the piezometric gradient remains constant, pore velocity will also decrease, thereby

reducing both advective and dispersive transport of nutrients through the system. Decreased nutrient transport results in a corresponding reduction in biofilm cell specific growth rate, and thus, in the rate of biotransformation of organic material. Decreased pore velocities reduce the shear rate, which could reduce the rate of detachment. Both biofilm growth rate and detachment rate will continue to change, unless a steady state biofilm thickness is achieved.

If the flow rate is held constant, the shear stress will increase as the biofilm grows and restricts the cross sectional area available for fluid flow. This increase in effective pore velocity will increase the shear stress, which can increase the detachment rate. Shear stress and biofilm detachment loss rates have together been the subjects of biofilm research, with both strong and weak correlations being reported (Peyton, 1996; Peyton and Characklis, 1993; Rittmann, 1982; Lau, 1995; Cao and Alaerts, 1995). A biofilm may slough and adjust to higher shear with a stronger biofilm matrix. In the case of porous media, the change in shear rate with time is small; and the biofilm could dynamically adapt.

Review of Biofilm System Models

Biofilm modeling started with trickling filters for which empirical models, experimental investigations, and conceptual models were applied to the attached biofilms (Atkinson and Daoud, 1968; Atkinson et al., 1968; Atkinson and Daoud, 1970; Atkinson and Davies, 1974; Atkinson and How, 1974; Atkinson et al., 1967).

Atkinson et al. (1967) were some of the early researchers to attempt modeling of microbial films. In this work, a *pseudo-homogeneous* and a *heterogeneous* model were analyzed. Conclusions resulting from this work were: 1) mathematical models derived on a heterogeneous basis are applicable to the biological film reactor, and 2) liquid phase diffusion resistance has considerable influence on the performance of a

continuous flow biological reactor. This work was extended, and diffusion limitations within the biofilm was found to be important. Atkinson and Daoud (1968) suggested that a model based on the catalytic nature of microbial biomass might help to clarify the importance of diffusional limitation. Atkinson and Daoud (1968) went on to develop model equations for biofilms and "flocs" in terms of a characteristic thickness and concentration of reactant in adjacent solution.

Williamson and McCarty (1976a) presented a model of substrate utilization by bacterial films using Monod kinetics (Monod, 1949) for a single substrate. This model was limited to a single substrate and it was necessary to determine which substrate limited overall biofilm uptake. This could be performed using model and stoichiometric coefficients in the biochemical transformation equation representing the metabolic reaction within a biofilm. An allowance for mass transfer was included using a defined liquid film thickness outside the biofilm as a model parameter. In experimental, work Williamson and McCarty (1976b) found that the liquid layer could not be completely removed and attributed this to the uneven or sponge like nature of the liquid-biofilm interface. Solutions were found for thick biofilms, which are limited by mass transfer (diffusion). In subsequent experimental verification studies of their biofilm model (Williamson and McCarty, 1976b), the model was extended to include thin biofilms. In their experimental work they found that diffusion coefficients for substrates (ammonia, nitrite, nitrate, and oxygen) moving through the biofilms of nitrifying bacteria were approximately 80 to 100 percent of corresponding values in water (Williamson and McCarty, 1976b).

Rittmann and McCarty (1978) developed expressions to provide a more convenient and easily used biofilm model based on the biofilm substrate removal model provided by Williamson and McCarty (1976a). This work provided a variable-order model that yields an approximate analytical solution for the flux of limiting substrate

into a fixed bacterial film, that is "deep". This worked to simplify application of the model of Williamson and McCarty (1976a) and aided application of fundamental, fixed-film kinetics to practical reactor problems.

Rittmann and McCarty (1980b) further extended the biofilm modeling of Williamson and McCarty (1976a) to include the mass of the biofilm, coupling the steady-state flux of substrate into a biofilm to the mass (or thickness) of the biofilm for a given bulk substrate concentration. In this analysis, a first-order decay term was used, which defined a minimum substrate concentration below which no significant biofilm activity should occur. This model provided substrate flux and biofilm thickness for bulk substrate concentrations greater than the minimum substrate concentration. Biofilm thickness was based on the total amount of biofilm mass which which could be supported by the substrate flux. Experimental evaluation of this steady-state biofilm model was performed (Rittmann and McCarty, 1980a), which confirmed the hypothesis that, for a single substrate limitation there exists a predictable steady-state concentration, below which no significant biofilm activity occurs. It was also noted that the steady-state model of biofilm kinetics successfully predicted substrate utilization in a steady-state reactor at maximum utilization rates; however, predicted biofilm thickness tended to be overestimated relative to those achieved experimentally.

A model for estimating substrate flux into a biofilm of any thickness was developed by Rittmann and McCarty (1981) as an extension of previous work (Atkinson and Daoud, 1968; Atkinson and Davies, 1974) by including external mass transfer resistance. The external mass transfer resistance was lumped into an effective stagnant layer through which all diffusional resistance takes place. With this model, the flux became a direct function of the bulk substrate concentration, which is typically the measured concentration and is often the concentration of interest. An explicit solution was available for thick biofilms, and a simple iterative solution was used for

thin biofilms. This model had the ability to predict the flux of a single, rate-limiting substrate when the biofilm thickness was independently known.

The steady-state biofilm model of Rittmann and McCarty (1980b) was criticized by Arcuri and Donaldson (1981) for only including decay as a loss mechanism when decaying cells would be trapped under the young cells. Arcuri and Donaldson (1981) argue that substrate uptake rate predicted by steady-state biofilm model (Rittmann and McCarty, 1980b) may not be realistic since the major mechanism for loss of biomass is probably sloughing of outer cells or cell aggregates as film becomes large and unstable. In response to this criticism, Rittmann (1982) presented a procedure to implement shear loss from biofilm by incorporating loss into a first order decay term in the steady-state biofilm model.

The steady-state biofilm model of Rittmann and McCarty (1980b) later was improved by correcting an error in the pseudo-analytical solution (Sáez and Rittmann, 1988). Additional work was performed to estimate error associated with limiting cases of the steady-state biofilm model (Sáez and Rittmann, 1990). Heath et al. (1990) provided a procedure for design of biofilm processes using normalized loading curves based on the steady-state biofilm model (Rittmann and McCarty, 1980b) and the improved solution technique (Sáez and Rittmann, 1988). This process was based on families of normalized loading curves allowing simple and rapid computation of required design volume for biofilm reactors. An improved pseudo-analytical solution (Sáez and Rittmann, 1992) to the steady-state biofilm model (Rittmann and McCarty, 1980b) was used to eliminate some inaccuracies (Cannon, 1991).

Benefield and Molz (1985) developed a mathematical model for fixed-film biological process (film flow over a flat plate) which described bulk liquid transport, diffusional transport of oxygen and organics across a stagnant film, diffusional transport of oxygen and organics into the biofilm, biochemical reactions by the individual

cells within the biofilm, biofilm growth, and cell density change within the biofilm due to cellular decay. Kinetics were modeled as double Monod for oxygen and one organic (electron donor and carbon source). This model was solved numerically with a grid of points in the flow direction and a grid of points within the biofilm. Some shortcomings of this model included lack of a general detachment expression accounting for erosion and sloughing type losses and a relationship between flow velocity and diffusion layer thickness used in the film type diffusional resistance from bulk model.

Kissel et al. (1984) started work on numerical simulation of mixed culture biofilms. Some of their assumptions prompted a discussion (Gujer and Wanner, 1985; Kissel et al., 1985). Wanner and Gujer (1986) developed a multispecies biofilm model to model biofilm processes. . This model formulation and background also appears in a later publication (Gujer and Wanner, 1990). This model was adopted in a technical report on biofilm modeling (Characklis et al., 1989) and has been used to model spatial dynamics (Wanner, 1989; Wanner, 1994). A computer software package BIOSIM (Ruchti et al., 1991; Reichert et al., 1990) utilized this biofilm model in conjunction with CSTR's for modeling biofilm systems. With this model the significance of spatial distributions in mixed culture biofilm was simulated (Fruhen et al., 1991). BIOSIM has been used in modeling of biofilms systems (Drury et al., 1993; van Loosdrecht et al., 1994; Arcangeli and Arvin, 1995; Okabe et al., 1995; Okabe et al., 1997) The BAM (Camper et al., 1994) computer software package also uses this mathematical biofilm model. The BAM computer software package was developed at the Center for Biofilm Engineering and has been used for simulation of biofilm systems (Stewart, 1994).

Rittmann and Manem (1992) looked at steady-state solutions to the multispecies biofilm model. In this approach, detachment was lumped into a first order decay parameter.

Szego et al. (1993a) developed and solved a model for simulation of biofilm processes in pipelines. This model was restricted to circular pipes of constant cross section and allowed for both laminar and turbulent flow (Szego et al., 1993b). Wend (1994) extended the use of biofilm modeling by including a gas phase for use in modeling vapor-phase bioreactors.

Computer models have already been utilized very effectively to help in developing an understanding of the basic biofilm growth process. The BIOSIM code has been used extensively to study various biofilm systems with fairly simple fluid flow regimes. For example, this model has been used to model flow and growth in pipes or conduits, along river beds, and on the surface of tanks or large containers. BIOSIM has been used effectively to describe and explain experimental data (Okabe et al., 1995; Okabe et al., 1997; Drury et al., 1993; van Loosdrecht et al., 1994; Arcangeli and Arvin, 1995).

A newer version of this biofilm system model called AQUASIM (Reichert, 1994b; Reichert, 1994a) has recently been developed. It models mixed culture biofilms and has some added features over BIOSIM (Wanner and Reichert, 1996). For example, it allows for movement of solids in biofilm in the liquid phase (Reichert and Wanner, 1997). AQUASIM has been used to model biofilm in porous media (Wanner et al., 1995), biofilm systems (Janning et al., 1995; Arcangeli and Arvin, 1997; Horn and Hempel, 1997; Beaudoin et al., 1998), and activated sludge (Reichert et al., 1995).

This review of biofilm system modeling has shown that modeling of systems is typically one dimensional steady uniform flow. In the case of porous media, the biofilm models are typically based on bulk media properties. These models do not consider microscale flow/transport effects. Experimental research into biofilm systems shows that in some situations they have some heterogeneity associated with them. Experimental research into the structure of the biofilm community and local transport

variations in mass and momentum transfer provide new insight into the biofilm processes (de Beer, Stoodley, Roe and Lewandowski, 1994; de Beer and Stoodley, 1995; de Beer, Stoodley and Lewandowski, 1994; Lewandowski et al., 1993; Lewandowski and Stoodley, 1995; Lewandowski et al., 1995; Lewandowski et al., 1994; Bishop et al., 1995) This is a local phenomenon and biofilm systems modeling should reflect the local flow/transport conditions. While not all processes are well understood at this point, some of these processes can now be simulated. In some situations it is not necessary to model all processes associated with biofilm system (Bishop and Rittmann, 1995).

Research Goals and Objectives

At the microscale, biofilm formation is a complex problem involving fluid dynamics, mass transfer, chemical reactions and biochemical reactions (Brading, Jass and Lappin-Scott, 1995). To understand the interaction of these different processes, it is useful to use a mathematical model to describe these processes. The goal of this research is to couple equations describing fluid flow, mass transport, and biofilm processes at the microscale to more accurately describes bulk fluid transport processes at the micro scale and the subsequent effect on the associated biofilm processes at the microscale. The resulting model is applied to different distributions of biomass and porous media capillary model. Results of these simulations demonstrate the coupled behavior of fluid dynamics, substrate transport, and biofilm reaction.

This research couples the continuity equation for incompressible steady-state fluid flow

$$\nabla \cdot \mathbf{u} = 0 \tag{1.1}$$

with the steady-state momentum equations for an incompressible isothermal Newto-

nian fluid

$$\mathbf{u}\cdot\nabla\mathbf{u} = -\frac{\nabla p}{\rho} + \nu\nabla^2\mathbf{u} + \frac{\mathbf{f}}{\rho} \tag{1.2}$$

and steady-state species balance equation for a dissolved constituent such as oxygen

$$\mathbf{u}\cdot\nabla O = \nabla\cdot(D(\mathrm{x})\nabla O) + r_O \tag{1.3}$$

for the bulk fluid region. Equations (1.1)–(1.3) are solved in two dimensions with the boundary conditions arising from solution of the equations for the biofilm.

For the application and model evaluation a single species biofilm model is used for steady-state oxygen consumption

$$0 = D_f\frac{\partial^2 O}{\partial z^2} - V_{\max}\frac{O}{K_O + O} \tag{1.4}$$

which is applied for biofilm located on walls of simulation domain.

Due to nonlinearities and the multidimensional nature of the resulting mathematical model, it was necessary to use numerical techniques to obtain a solution. The resulting model allows simulations to be performed with flow tubes as shown in Figure 1. Tubes such as this one can be used to form *capillary* models of porous media (Dullien, 1991; Kaviany, 1995; Tiwari, 1997). This approach can determine the effect that system geometry and flow conditions have on biofilm accumulation and biotransformation in tortuous, laminar flow systems. In addition, the general flow and transport around patches of biofilm and subsequent development of the mass transfer boundary layer can be investigated with this computational mathematical modeling approach.

Bàveye and Valocchi (1989) discuss the concept of a uniform biofilm vs. colonies in porous media. Using the above model, simulations can be performed describing local fluid velocity and transport variation to effects on various types of biofilm distributions (Figure 2). This allows different simulations to be performed, which can then be compared with laboratory experiments using biofilm reactors of similar geometry.

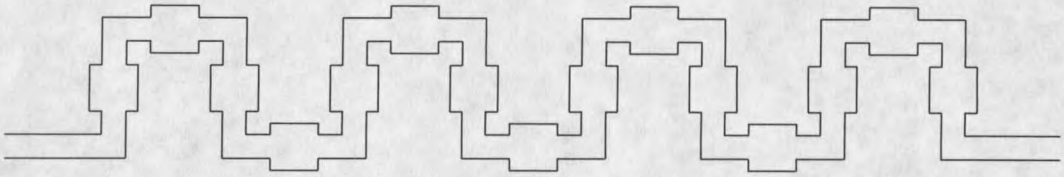

Figure 1. Two-dimensional model of a porous medium pore channel.

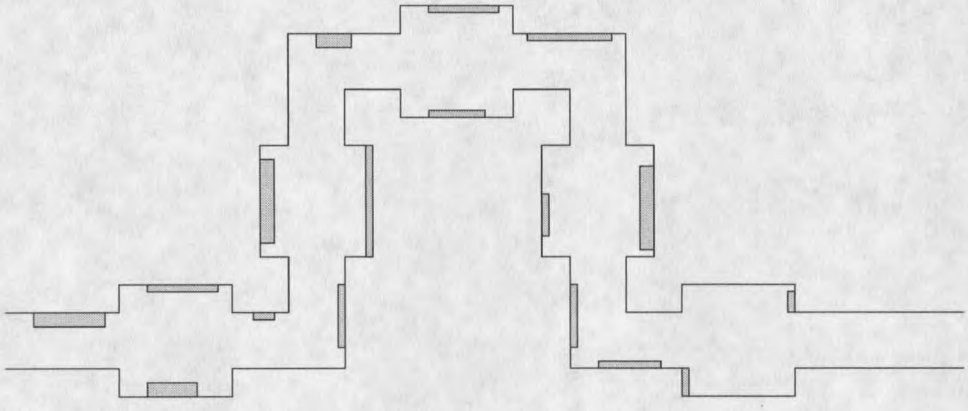

Figure 2. Tortuous tube with patchy biofilm.

The specific objectives of this thesis are to

- Formulate a mathematical microscale model.
- Solve the microscale mathematical model.
- Evaluate the model against experimental data.
- Apply the microscale model to biofilm systems.

Model Development Approach

Although some of the growth mechanisms in pipes are beginning to be fairly well understood (Picologlou et al., 1980), the growth process in a tortuous porous media is considerably more complicated. Due to this complexity, it is important to try to understand and model the basic processes occurring and their rates at the pore level. If the basic processes are understood and can be modeled at a small scale it is possible to scale up this nonlinear information to the field scale.

When physical processes are nonlinear and are coupled to other nonlinear processes, the ability to understand the coupled phenomena at a local level and then to scale this knowledge up to longer length scales is extremely important. For example, typical length scale for pore scale flow governed by Navier-Stokes ($10^{-4} - -10^{-3}$ meters) is quite different from the scale required by field-scale simulations ($10 - -10^{3}$ meters) (Ewing, 1997). Numerical simulations hold enormous promise for this difficult problem. First numerical simulations can be used to separate the processes so that we can understand each specific phenomena by itself. Then the processes can be coupled in various ways to build the intuition of the modeler as to the effects of the various couplings. This separation of processes is difficult if not totally impossible in the laboratory. However the "computational laboratory" is the basis of a third new methodology, taking its place along side analysis and laboratory experimentation as as powerful new scientific tool (Chen et al., 1994; Ewing, 1997). Simulations can

be compared with laboratory experiments to support important phenomenological models and to determine reaction rates. Once a model is chosen, computer simulations can predict sensitivities of various parameters and coupled interactions in the complex set of partial differential equations used in simulators for porous media can then be scaled up (Ewing, 1997).

Biofilm modeling can be a useful aid in designing experimental systems and scaling-up of biofilm processes and reactors. There has been some discussion in the literature on the scale up issues associated with of biofilm reactors and their nonlinear processes (Manem and Rittmann, 1990). Scale up issues in porous media applications are inherently nonlinear. When biological processes are added, the scale up process becomes much harder.

One component of biofilm modeling is the mass transfer to the biofilm from the bulk fluid to the biofilm. This process is often modeled using mass transfer coefficients. Average mass transfer coefficients are typically found in the literature for simplified geometries. Mass transfer correlation equations for common flow system geometries such as circular tube, flat duct, and flat plate are available (Cussler, 1984; Weber and DiGiano, 1996). These correlations typically have some simplifying assumptions related to uniform geometry and a uniform flux or concentration boundary conditions. In this thesis, the approach is to formulate equations describing the flow and transport of dissolved substrates and consumption by biofilm, and solve them using established computational fluid dynamics (CFD) techniques. The use of CFD techniques allows different geometries to be simulated where changes in reactor geometry lead to changing of local flow characteristics which can influence mass transfer to the biofilm surface. This approach will allow one dimensional patches of biofilm to be coupled with a two dimensional transport model for fluid velocity and mass transport outside the biofilm region, thereby facilitating a investigation of biofilm patchiness

and biomass distribution for conditions of interest.

The outline of this thesis is as follows. Chapter 2 contains the conceptual view of biofilm and bulk fluid processes with assumptions made to obtain a mathematical model describing the biofilm system. In Chapter 2, I discuss the Navier-Stokes equations and advective-diffusion-reaction equation describing transport in the bulk fluid region and the biofilm process model. Coupling of the mathematical equations is also discussed. In Chapter 3, I present a numerical scheme for solving the model equations presented in Chapter 2 in two spatial dimensions for the fluid dynamics and one dimension for the biofilm. Discretization of mathematical equations and resolution of nonlinear equations are examined. Chapter 4 contains an model evaluation using experimental data taken from an open channel reactor using an artificial biofilm. Application of the model to relevant biofilm systems is presented in Chapter 5. Conclusion are contained in Chapter 6 and future work in Chapter 7.

CHAPTER 2

MATHEMATICAL MODEL DEVELOPMENT

It is convenient to separate the domain of interest into a bulk fluid region and a biofilm region. These regions are depicted in Figure 3.

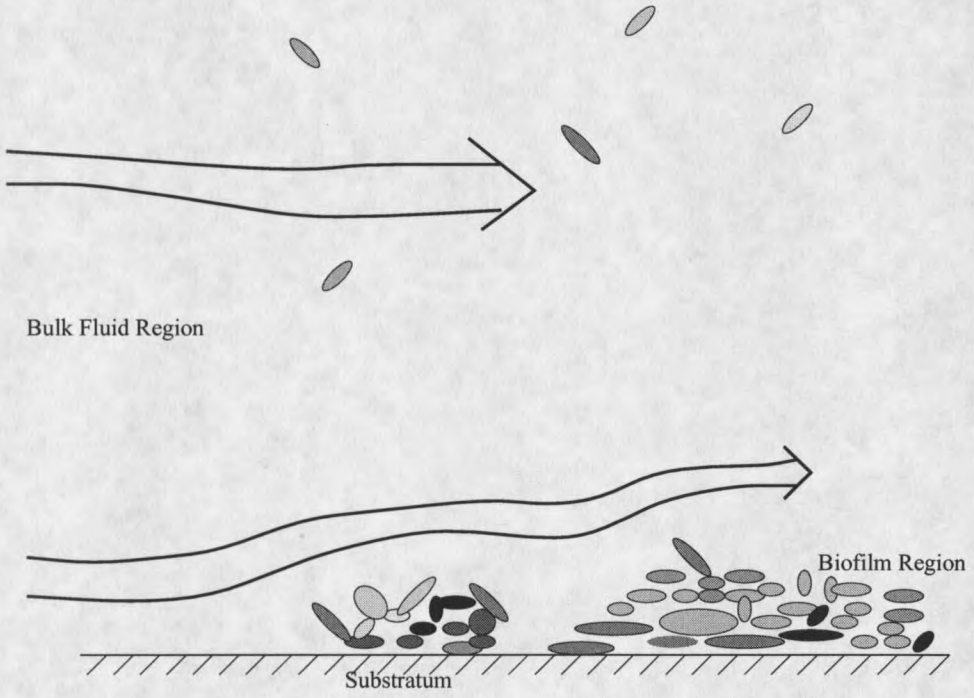

Figure 3. Sketch of bulk fluid and biofilm regions.

The bulk fluid region is characterized by dilute concentrations of suspended microorganisms and other dissolved constituents. The concentrations of these species is assumed to be small enough that their influence on the flow is negligible. The concentration of suspended microorganisms is typically small, so that the resulting reaction rate is small compared to reaction in the biofilm region. Conversely, the

biofilm region is characterized by high concentrations of biomass attached to a solid substratum. The biofilm region has a high concentration of biomass which leads to a higher substrate consumption than in the bulk.

These two regions are connected by an interface across which the dissolved and particulate components move. This interface region will have mathematical equations describing the transport of constituents across the interface and movement of the interface itself. Due to the high concentrations of biomass in the biofilm relative to the bulk fluid, sharp concentration gradients can develop near the biofilm bulk fluid interface.

Bulk Fluid

The bulk fluid region is defined as the region above the biofilm region as depicted in in Figure 3. Physical processes in the bulk fluid provide transport of nutrients and removal of biochemical transformation products from the biofilm. In addition, biomass can be transported to and from the substratum and biofilm surface. For dissolved species, transport processes are by fluid convection and diffusion/dispersion. Diffusion from high concentration to low concentration is typically modelled by Fick's law: the diffusive flux of a substance is proportional to the gradient of the concentration of that substance. Fick's law is mainly used for describing diffusion of soluble components, but can also be used for large molecules or small particles, such as microbial cells diffusing in water (Characklis, 1990a).

Microorganisms can be motile or nonmotile and thus different behavior is expected in bulk fluid for microorganisms possessing motility (Characklis and Marshall, 1990). Motile microorganisms can respond to concentrations of a substrate in which they move in increasing direction of substrate concentration which is termed *chemotaxis*. Movement away from an undesirable constituent would also be possible. Some

research has been done on determining chemotaxis coefficients (Ford et al., 1991) and single cell modeling with chemotaxis (Ford and Lauffenburger, 1991). Cellular dynamics simulations have been used to model large population of individual chemotactic bacteria in three dimensions (Frymier et al., 1993). In the case of a bare substratum, the physical process of interest is the transport of microorganisms to the surface and subsequent attachment of some microorganisms. The influence of hydrodynamics in movement of microorganisms in the vicinity of a boundary, between boundaries, and vicinity of another microorganism has been modeled by Ramia et al. (1993). Pedley and Kessler (1992) investigated hydrodynamics of suspensions of swimming microorganisms.

Dillon et al. (1995) developed a model including bacterial swimming, chemotaxis, and substrate consumption. This model was solved in two dimensions for a limited number of bacteria in the fluid. Fluid dynamic interaction of swimming bacteria was accounted for along with fluid force on bacteria. The ability to include a attachment term when bacteria come in proximity to a surface or other bacteria suggests this model might have potential for some initial events work. This model was extended in this way to model biofilm processes under laminar flow conditions (Dillon et al., 1996).

Quite often fluid dynamics of biofilm reactors are modeled with dispersed flow models (Muslu, 1990). Dispersed flow models such as continuously stirred tank reactor (CSTR) and plug flow reactor (PFR) represent average values in the bulk fluid. If mixing is high, dispersed flow models like CSTR and PFR are used although some diffusional resistance is encountered at the wall which is sometimes modeled with an effective liquid layer thickness across which all diffusional resistance effects are accounted for.

Simulation of fluid dynamics depends on the type of flow, whether it is laminar,

turbulent, or in the transition stage. If the fluid dynamics to be simulated is in the transitional or turbulent region it is necessary to include an additional model for the turbulence. The ability to solve the flow equations at a fine enough scale to resolve turbulent fluctuations requires massive computational resources and is not feasible for most practical applications (Calmet and Magnaudet, 1997). In view of this limitation turbulence models have been formulated and are being developed to allow simulations of turbulence to be performed. For the purposes of this modeling effort, it is assumed that the fluid flow is in the laminar region.

The concentration of dissolved substrates is assumed to be sufficiently small that it does not affect the viscosity or density of the bulk fluid. Another assumption is that biomass in the bulk does not affect the viscosity or density of the bulk fluid. This assumption may not be valid near the biofilm/bulk fluid interface where flow velocity is lowest and biomass concentration could be the highest. In this region, suspended biomass, which could be biofilm aggregates, may influence the local behavior of the bulk fluid by increasing the effective viscosity.

External mass transfer is often dealt with by use of a layer of effective thickness of fluid that is undisturbed for the purpose of including diffusional effects from bulk fluid to wall (Vogel, 1994). Note that this is not really undisturbed fluid but a region where diffusion is the dominant transport mechanism. This is an approximation to the real diffusive boundary layer which is used to account for the mass transfer resistance. Biofilm models with dispersed flow bulk fluid models have used an effective liquid layer thickness for diffusional resistance (Reichert et al., 1990; Reichert, 1994b). This dissertation improves on the prevailing mass transfer model component by solving mass transport in two dimensions around biofilm patches, thereby providing an analysis of mass transport in complicated geometries such as that shown in Figure 1. To solve problems such as that shown in Figure 1 with current models, would

be cumbersome. It would be necessary to model individual sections such as tubes of varying size and hence different residence times utilizing CSTR or PFR reactor models and allow for some external mass transfer to walls with biofilm.

The bulk fluid region provides the transport of liquid, dissolved constituents (chemical and biomass). Bulk fluid flow in the laminar flow region can be modeled with the Navier-Stokes equations to obtain the fluid velocity. A mass balance equation for the dissolved substrates, biomass species, and inert species is used to model their behavior.

Navier-Stokes Equations

The bulk fluid dynamics can be modeled by the Navier-Stokes equations for an isothermal incompressible Newtonian fluid with constant viscosity (Bird et al., 1960; Probstein, 1989). The continuity equation obtained from a mass balance on the fluid gives

$$\nabla \cdot \mathbf{u} = 0 \tag{2.1}$$

while the balance of momentum equations give

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{\nabla p}{\rho} + \nu \nabla^2 \mathbf{u} + \frac{\mathbf{f}}{\rho} \tag{2.2}$$

where $\mathbf{u} = \mathbf{u}(\mathbf{x}, t) = (u(\mathbf{x}, t), v(\mathbf{x}, t), w(\mathbf{x}, t))$ $(\mathrm{L\,t^{-1}})$ is the bulk fluid velocity, $p = p(\mathbf{x}, t)$ $(\mathrm{M\,L^{-1}\,t^{-2}})$ is the fluid pressure, ρ $(\mathrm{M\,L^{-3}})$ is the fluid density, (assumed constant) ν $(\mathrm{L^2\,t^{-1}})$ is the fluid kinematic viscosity (assumed constant), $\mathbf{x} = (x, y, z)$ (L) are spatial coordinates, and $\mathbf{f}$ $(\mathrm{M\,L\,t^{-2}})$ is sum of body forces acting on fluid (i.e., gravity forces etc.).

Recent formulations for swimming microorganisms use an immersed boundary condition to include the influence of swimming microorganisms on fluid dynamics (Dillon et al., 1995; Dillon et al., 1996; Fauci, 1993; Fauci and Peskin, 1988; Fauci,

1996). In these formulations individual tracking of microorganisms is used. Alternatively the effects of microbial suspensions could be lumped into variable viscosity which depends on the concentration of the microorganisms similar to way suspensions are dealt with. In this conceptual model, it is assumed that concentrations of microbial biomass are dilute and the biomass particles are small enough not to influence the local transport properties significantly.

The flow of liquid in the biofilm region could be modeled using higher viscosity in biofilm region. Stolzenbach (1989) looked at biofilm as a collection of momentum capturing particles for modeling flow in the biofilm. This resulted in a body force applied to the fluid by the biofilm proportional to fluid velocity in biofilm. A similar approach could have been applied in this study, but once flow is considered in the biofilm region, the modeling complexity becomes much greater.

Equation (2.2) could be modified to include a term similar to Darcy's law for porous media flow which would have a non zero permeability only in the biofilm and could be used to allow flow in the biofilm. These modifications could be performed on this model to more accurately model the behavior in biofilm once sufficient data becomes available to quantify flow in biofilms.

Species Balance Equation

The mass balance equation of dilute concentrations of dissolved species in the isothermal incompressible bulk fluid under laminar flow is given by the species balance equation (Bird et al., 1960; Probstein, 1989).

$$\frac{\partial c_i}{\partial t} = -\nabla \cdot \mathbf{J}_{T_i} + r_i \tag{2.3}$$

In equation (2.3) $c_i = c_i(\mathbf{x}, t)$ (M L^{-3}) is the substrate mass concentration, $r_i = r_i(\mathbf{x}, t, c_1, ..., c_N)$ (M $L^{-3}t^{-1}$), is the production rate of species i which can be dependent on the other species in the bulk, and $\mathbf{J}_{T_i}$ (M L^{-2} t^{-1}), is the total mass flux of

species i and is given by

$$\mathbf{J}_{T_i} = \mathbf{u}\cdot\nabla c_i - D_i(\mathbf{x})\nabla c_i. \tag{2.4}$$

The first term on the right hand side of equation (2.4) represents the convective flux due to bulk fluid flow and the second term represents the diffusive flux which is assumed to obey Fick's first law and $D_i(\mathbf{x})$ ($\mathrm{L}^2\ \mathrm{t}^{-1}$), is the diffusion coefficient for species i. If the flow is not resolved at the actual scale it occurs, D_i typically becomes larger than molecular diffusion and is called a dispersion coefficient. In some cases it becomes a function of the velocity field. For the unsteady state velocity case, the dispersion coefficient would then be implicitly a function of time.

Other types of flux related to thermal or pressure influences are assumed to be negligible for this modeling effort. It could be possible to add a chemotaxis flux term to equation (2.4) to include chemotaxis phenomena.

Insertion of (2.4) into (2.3) and recalling (2.1) gives a model equation for balance of dissolved species in the bulk fluid region

$$\frac{\partial c_i}{\partial t} + \mathbf{u}\cdot\nabla c_i = \nabla\cdot(D(\mathbf{x})\nabla c_i) + r_i \tag{2.5}$$

The species balance equation (2.5) holds for all the dissolved species in the bulk. It is assumed that equation (2.5) also holds for particulate and inert species in the bulk.

In the case of inert or particulate species with no motility, $D_{X_i}(\mathbf{x})$ is a Brownian motion coefficient. In the case where particulate species possesses motility, it may represent motility speed. It may be better to use a chemotaxis expression for motile species where they are are attracted or repelled by one or more of the dissolved species. For example, motile particulate species may be attracted by higher concentrations of a substrate and swim in the direction of increasing concentrations. Chemotaxis modeling could lead to other flux term for a particulate species to reflect the movement of motile species toward the increasing nutrient concentration.

In order that mass is conserved, the sum of the reaction terms must be equal to zero, i.e,:

$$\sum_{i=1}^{N} r_i = 0 \tag{2.6}$$

where N is the total number of species in the bulk fluid. The total number of species in the bulk fluid includes the number of dissolved constituents, N_D, the number of particulate species, N_X, the number of inert species, N_I, and the water phase.

$$N = N_D + N_X + N_I + 1 \tag{2.7}$$

In general it is not feasible to solve for all the dissolved constituents due to computing resource limitations and also the fact that not all of the coefficients for the reaction terms are known or even measurable. Some of these coefficients may need to be obtained in an indirect manner from other measurable parameters. A common assumption is that some constituents are not limiting and the species balance equation (2.5) is solved only for the relevant limiting species of interest. In some case the reaction rate may be influenced by other reactants or products which would require accounting for these other species to properly model the reaction rate.

As an example, the reaction of a microbial species with glucose as the primary substrate and oxygen as the electron acceptor would require the solution of (2.5) for substrate, oxygen, and biomass in the bulk. These three equations would be solved in conjunction with biofilm equations for the substrate, oxygen, and biomass in the biofilm. This could be fairly complicated if multiple microorganisms are modeled along with multiple states for a microorganism such as active and inactive. Other variations include cell death and lysis with a portion of cell lysis products yielding soluble nutrients for cell growth. Some reactions may be influenced by products which may influence the overall reaction rate. The microbial reaction kinetics may determine the number of constituents that must be modeled in a biofilm system.

Interfacial Processes

Interfacial processes occur at the biofilm/bulk fluid interface. The interface is defined as the surface location where a cell is attached to biofilm or substratum and no longer suspended in the bulk fluid. This is a mathematical interface definition which allows different regions to be defined and material balances written for each region with different assumptions. Some of the processes deemed important in biofilms (Characklis and Marshall, 1990) are described below. This general approach follows the process analysis approach (Characklis, 1990b; Characklis and Cooksey, 1983; Characklis, 1984; Characklis, 1990c).

Adsorption

Adsorption is the interphase accumulation or concentration of molecules or cells on a substratum or interface. In three dimensions, adsorption is a two dimensional process normal to the surface or interface. This process is influenced by the fluid flow in the region near the surface or interface (Escher and Characklis, 1990). Interest in the initial event leading to colonization of surfaces has been the subject of other researchers (Brannan and Caldwell, 1982; Malone and Caldwell, 1982; Caldwell et al., 1982; Kieft and Caldwell, 1982; Powell and Slater, 1983; Escher and Characklis, 1990; Mueller et al., 1992; Mueller, 1996).

Desorption

Desorption is the reverse of adsorption and refers to the movement of molecules or cells from the substratum back into the bulk liquid compartment. In one study it was noted that a significant number of cells contacting the surface do not become irreversibly attached; assuming so can introduce significant error (Powell and Slater, 1983).