Evaluation of a model for simulating biofilm processes in porous media reactors
by Warren Thomas Sharp

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in Chemical Engineering
Montana State University

Abstract:
Biofilms are ubiquitously present in many natural and manmade porous media systems. Natural and engineered biofilm systems are important to bioremediation and biofiltration and include both one-fluid-phase flow and two-fluid-phase flow systems. Complex interactions exist between net biomass accumulation, porous media characteristics, and the rates of biofilm/biological processes. This complexity requires a systematic approach to analyzing experimental results. The dynamic behavior of biofilms in porous media is encapsulated in the rates of the biofilm/biological processes contributing to net biomass accumulation. The key to understanding and predicting biofilm/biological processes in porous media is modeling. The goal of this research is to evaluate a biofilm process model in porous media systems for the prediction of experimental results.

Most porous media biofilm models in the literature pertaining to both one-fluid-phase and two-fluid-phase flow porous media systems utilize insufficient biofilm models. The mixed-culture biofilm model (MCB) overcomes many limitations of the former. Combining the MCB model with the appropriate reactor transport models give a porous media biofilm reactor model that has the capability to simulating many types of porous media biofilm systems. Experimental data from four separate systems, including two one-fluid-phase and two two-fluid-phase, were analyzed in order to evaluate the MCB model coupled with the appropriate reactor transport model.

Predicted results from the model for which experimental data exists give a good fit. However, the model can predict more than what is available experimentally; therefore, a full evaluation of model capabilities is not possible at this time. A simulation-experimentation framework which can elucidate necessary experimental data for further model evaluation is provided.

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APPROVAL

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Warren Thomas Sharp

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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Date 5/17/96

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ABSTRACT

Biofilms are ubiquitously present in many natural and manmade porous media systems. Natural and engineered biofilm systems are important to bioremediation and biofiltration and include both one-fluid-phase flow and two-fluid-phase flow systems. Complex interactions exist between net biomass accumulation, porous media charactersistics, and the rates of biofilm/biological processes. This complexity requires a systematic approach to analyzing experimental results. The dynamic behavior of biofilms in porous media is encapsulated in the rates of the biofilm/biological processes contributing to net biomass accumulation. The key to understanding and predicting biofilm/biological processes in porous media is modeling. The goal of this research is to evaluate a biofilm process model in porous media systems for the prediction of experimental results.

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INTRODUCTION

Relevance of Biofilms in Porous Media

Biofilms are ubiquitously present in many natural and manmade porous media systems. Rittmann maintains that "[biofilms] play crucial roles in the biodegradation of contaminants and the clogging of porous media ..." [8] Many natural and engineered biofilm systems utilize porous media as a support for biofilm formation. Natural biofilms in subsurface porous media formations can be used in the field of bioremediation in order to degrade contaminants. Engineered biofilm systems can be used in industrial processes such as biofiltration and drinking water treatment. Together, these examples constitute two types of porous media systems: one-fluid-phase and two-fluid-phase. The two primary measureable activities of importance in porous media biofilm systems are biomass accumulation and biotransformation.

Complexity of Biofilms and Biofilm/Biological Processes in Porous Media

The complexity of biofilms and biofilm processes in porous media stems from the relationship between net biomass accumulation, changes in porous media characteristics, and rates of biofilm/biological processes. Figure 1 summarizes the complexity of biofilms in porous media systems.

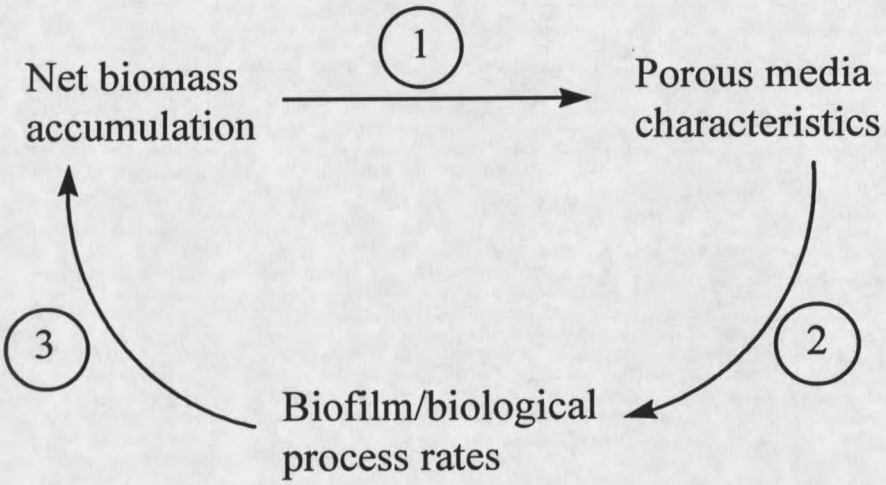

Figure 1. Complexity of biofilms in porous media.

One level of complexity of biofilms in porous media, shown as relationship 1 in Figure 1, is the effect of biomass accumulation on the characteristic properties of a porous medium, such as porosity, permeability, and dispersivity. These effects have been quantified by Taylor, Milly, and Jaffe: increasing biomass accumulation in a porous medium will decrease the porosity and permeability, and increase the dispersivity. [12] Conversely, a second level of complexity, shown as relationship 2 in Figure 1, relates the changes in porous medium characteristics to the transport of substrates to the biofilm. Again, this has been quantified and understood to some extent through computational fluid dynamics. The third level of complexity, shown by relationship 3, explains how the rates of biofilm/biological processes, which depend on the transport of substrates and biomass, contribute to net biomass accumulation. The complexity of this relationship depends on the large variety of potential biological reactions. Rittmann says that the net accumulation of biomass is controlled by 4 biofilm/biological processes: growth and decay (biological), and deposition and detachment (biofilm). [8] The issue of biofilm/biological process complexity becomes very important

when multispecies biofilms are considered. Biological processes of growth alone need to be considered for *m* bacterial species utilizing *n* substrates, the totality of which contributes to net biomass accumulation. One final consideration is that the effect of net biomass accumulation on transport depends on the ratio of overall biofilm thickness to pore volumes. For small ratios that are usually seen for packed beds, the effect on transport will be minimal. However, for sand packed systems, the effect may be significant.

Relevance of Modeling Biofilm/Biological Processes in Porous Media

The dynamic behavior of biofilms in porous media is encapsulated in the rates of all pertinent biofilm/biological processes contributing to net biomass accumulation. Little work has been done with respect to modeling biofilm processes complexity other than the work done by Wanner, Guyer and Reichert [9,10,11,15]. Applications of complex biofilm models to porous media biofilm systems has been sparse. Therefore, the key to quantifying and understanding biofilms in porous media systems to effectively utilize biofilms is modeling the biofilm/biological processes. The potential complexity of the problem requires a systematic approach to analyzing experimental data. The modeling process gives a framework in which educated decisions can be made about both the experimental design and results and the accuracy of the simulations. As a result, the capability to predict biotransformation and biomass accumulation in a porous media will allow for better understanding, control, and design of porous media biofilm systems.

Additionally, regulatory acceptance of a biofilm process used to degrade or transform contaminants is hindered by a lack of understanding on the part of the regulators. Successful

modeling and simulation of biofilms and biofilm processes in porous media may speed acceptance of biofilm processes in industrial applications.

Porous Media Biofilm Reactor Models

In the literature there is a definite distinction between one-fluid-phase and two-fluid-phase models for porous media biofilm systems. Generally, one-fluid-phase models are found in the porous media groundwater modeling literature while two-fluid-phase models are located in the biofilter modeling literature.

Porous Media Groundwater Biofilm Models

Much work was done by Taylor, Milly, and Jaffe [12] to measure and quantify the effect of biofilm growth on the physical properties of a porous medium, which include porosity, permeability, and dispersivity. The strong effort of their biofilm porous media model was to describe transport of biomass and substrates. [13] The biofilm in their model was described by a single Monod growth expression. However, they do acknowledge that more complex kinetics could be incorporated into their model. Baveye and Valocchi [2] provide an evaluation of three different representations of spatial distributions of biomass within a porous medium. Their biofilm is described by a single Monod growth expression. While both of these papers discuss methods to model changes in porous media characteristics due to biomass accumulation, neither provides an evaluation of the importance of the rates at which the processes of biofilm growth and substrate utilization take place. These processes define the dynamic behavior of porous media biofilm systems.

Biofilter Models

Recently, Deshusses, Hamer, and Dunn [3] offered a biofilter model for waste air treatment. Their description of a biofilter included gas and biofilm phases as well as a sorption volume. The biofilm was modeled as a homogeneous, monoculture system. Monod kinetics are used to describe growth and degradation kinetics, but the biofilm is assumed to be at steady state and the degraders are assumed to be evenly distributed throughout the biofilm. Diks and Ottengraf [4] created a model to describe the degradation of dichloromethane in a trickling filter. Their assumptions included steady state thickness, zero order kinetics, and constant volume fraction of the degraders in the biofilm. Baltzis and Shareefdeen [1] give a model for a packed-bed biofilter. The biofilm is modeled as an effective biolayer. More complex kinetics, such as substrate competition, are considered, but only one bacterial species is considered. The drawback to each of these biofilm models is the usage of oversimplified biofilm models, especially when there may be more biological processes than degradation/growth that can affect biofilter degradation performance. Biofilm thickness, volume fraction of cells, and kinetics are not always constant. These models lack the flexibility to describe more complex biofilm processes.

The Extended Mixed-Culture Biofilm Model

Most current groundwater porous media models are of the advection-dispersion-reaction or advection-reaction type. Some models consider the biofilm to be a "sink" for given substrates while others estimate fluxes into the biofilm using Monod expressions and biofilm thicknesses. Biofilter models utilize more complex biofilm models than do the groundwater models, but most still lack the flexibility to consider many important

biological/biofilm processes. The potential complexity of biofilm processes, including detachment, multiculture and multisubstrate biofilms requires a flexible, dynamic biofilm model. The important issue is not only the relationship between biofilm thickness, porous media characteristics, and transport, but also how the rates of biofilm/biological processes are dynamically integrated into the behavior of biofilms in porous media systems.

The most recent biofilm model capable of modeling multiculture, multisubstrate biofilms is the mathematical mixed-culture one-dimensional biofilm (MCB) model discussed by Wanner and Reichert [15]:

> Basically, the MCB model consists of a set of one-dimensional mass balance equations by which the progression of the biofilm thickness and the spatial distribution and development in time of various dissolved components (nutrients, electron donors, and electron acceptors) and particulate components (microbial cells, extracellular polymeric substances, organic and inorganic particles) in a biofilm can be modeled as a function of transport and transformation processes. These mass balance equations are generally valid and can be applied to almost any microbial system if the appropriate stoichiometry and kinetics are provided.

Coupled with the appropriate reactor transport models, the resulting porous media biofilm reactor model has the capability to simulate a large variety of experimental geometries and biofilm/biological processes.

Research Goal

The goal of this research is to evaluate the capability of the extended mixed-culture biofilm process model for modeling porous media biofilm processes in porous media systems for prediction of experimental results. The objectives required to reach this goal are:

- Evaluate the mixed-culture biofilm (MCB) model with the appropriate reactor transport model as a complete porous media biofilm process model with

experimental data from both one-fluid-phase flow and two-fluid-phase flow porous media reactors.

- Determine the limit of behavioral complexity of biofilm/biological processes in porous media reactors that can be modeled with the extended mixed-culture biofilm process model.

MODELING

There are three parts to the porous media biofilm reactor model to be analyzed herein: reactor, biofilm and biological processes. The reactor part of the model is either a one-fluid-phase or two-fluid-phase reactor. The biofilm is represented by the mixed-culture biofilm (MCB) model which can consider an arbitrary number of bacterial and substrate species. Figure 2 shows the conceptual model for the integration of the MCB and reactor models for both one-fluid-phase and two-fluid-phase porous media biofilm reactors. Biological processes can occur in the biofilm, bulk liquid, and bulk gas phases, and area described via appropriate kinetics and stoichiometry.

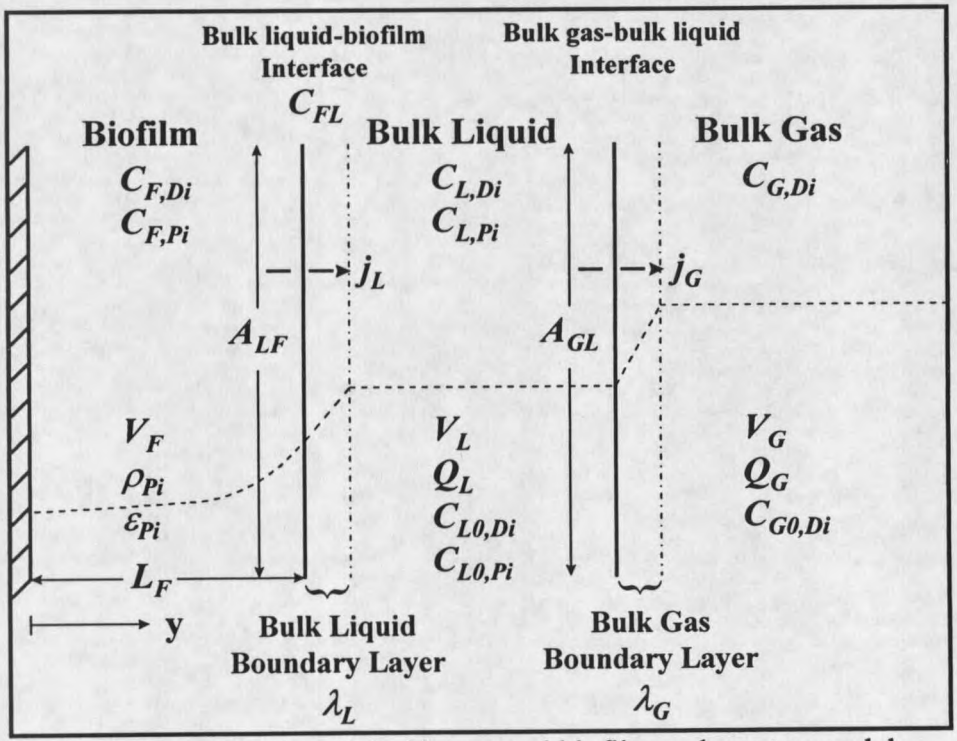

Figure 2. Conceptual model of integrated biofilm and reactor models.

One-fluid-phase Flow Porous Media Reactor

Mass transport is described by a one-dimensional advection-diffusion equation. The reactor is divided into axial segments, and bulk liquid and biofilm sections are distinguished. The bulk liquid phase is well mixed. The mass balance equation for dissolved and particulate components in the bulk phase, respectively, are

$$\frac{d(V_L C_{L,Di})}{dt} = Q_L\left(C_{L0,Di} - C_{L,Di}\right) + A_{LF} j_{LF,Di} + V_L r_{Di} \tag{1}$$

$$\frac{d(V_L C_{L,Pi})}{dt} = Q_L\left(C_{L0,Pi} - C_{L,Pi}\right) + A_{LF} j_{LF,Pi} + V_L r_{Pi} \tag{2}$$

where C_{L0} is the segment influent concentration (ML^{-3}), C_L is the bulk liquid concentration (ML^{-3}), j_L is the mass flux per unit interfacial area across the biofilm-bulk liquid interface ($ML^{-2}t^{-1}$), r is the net production rate ($ML^{-3}t^{-1}$) , V_L is the bulk liquid volume (L^3), Q_L is the liquid volumetric flow rate (L^3t^{-1}), A_{LF} is the biofilm surface area (L^2) of the segment, and t is time. The interfacial mass flux of dissolved and particulate components per unit biofilm surface area is given by

$$j_{L,Di}\lambda_L = \Phi_{L,Di}\left(C_{FL,Di} - C_{L,Di}\right) \tag{3}$$

$$j_{L,Pi}\lambda_L = \Phi_{L,Pi}\left(C_{FL,Pi} - C_{L,Pi}\right) \tag{4}$$

where λ_L is the mass transfer resistance coefficient (L), Φ_L is the effective diffusivity (L^2t^{-1}) of the dissolved or particulate species in the liquid phase, and C_{FL} is the concentration (ML^{-3}) at the biofilm-bulk liquid interface. The bulk fluid volume, V_L is related to the total segment volume and total biofilm volume by

$$V_L = V_C - V_F \tag{5}$$

where V_C is the total volume of the segment, and V_F is the biofilm volume.

Two-fluid phase Flow Porous Media Reactor

Mass transport is described by one-dimensional advection-diffusion equation. The reactor is divided into axial segments. Bulk gas, bulk liquid and biofilm sections are distinguished. The bulk gas and bulk liquid phases are well mixed. The mass balance equation for dissolved and particulate components, respectively, in the bulk liquid is

$$\frac{d(V_L C_{L,Di})}{dt} = Q_L\left(C_{L0,Di} - C_{L,Di}\right) + A_{LF} j_{L,Di} - A_{GL} j_{G,Di} + V_L r_{Di} \tag{6}$$

$$\frac{d(V_L C_{L,Pi})}{dt} = Q_L\left(C_{L0,Pi} - C_{L,Pi}\right) + A_{LF} j_{L,Pi} + V_L r_{Pi} \tag{7}$$

where C_{L0} is the segment influent liquid phase concentrations (ML^{-3}), C_L is the bulk liquid concentrations (ML^{-3}), j_L is the mass flux per unit interfacial area across the biofilm-bulk liquid interface ($ML^{-2}t^{-1}$), r is the net production rate ($ML^{-3}t^{-1}$), V_L is the bulk liquid volume (L^3), Q_L is the liquid volumetric flow rate (L^3t^{-1}), A_{LF} is the biofilm surface area (L^2) of the segment, A_{GL} is the bulk gas-bulk liquid interfacial area (L^2), and t is time. The mass balance equation for dissolved components in the bulk gas is

$$\frac{d(V_G C_{G,Di})}{dt} = Q_G\left(C_{G0,Di} - C_{G,Di}\right) + A_{GL} j_{G,Di} \tag{8}$$

where C_{G0} is the segement influent gas phase concentration (ML^{-3}), C_G is the bulk gas concentration (ML^{-3}), V_G is the bulk gas volume (L^3), j_G is the mass flux per unit interfacial area across the bulk gas-bulk liquid interface ($ML^{-2}t^{-1}$), Q_G is the gas

volumetric flow rate (L^3t^{-1}). The biofilm-bulk liquid interfacial mass flux of dissolved and particulate components, respectively, is

$$j_{L,Di}\lambda_L = \mathcal{D}_{L,Di}\left(C_{FL,Di} - C_{L,Di}\right) \tag{9}$$

$$j_{L,Pi}\lambda_L = \mathcal{D}_{L,Pi}\left(C_{FL,Pi} - C_{L,Pi}\right) \tag{10}$$

where λ_L is the mass transfer resistance coefficient (L) across the biofilm-bulk liquid interface, $\mathcal{D}_L$ is the effective diffusivity (L^2t^{-1}) of the dissolved or particulate species in the liquid phase, and C_{FL} is the concentration (ML^{-3}) at the biofilm-bulk liquid interface. The bulk gas-bulk liquid interfacial mass flux of dissolved components is

$$j_{L,Di}\lambda_G = \mathcal{D}_{G,Di}\left(C_{L,Di} - \frac{C_{G,Di}}{H_{Di}}\right) \tag{11}$$

where λ_G is the mass transfer resistance coefficient (L) across the bulk gas-bulk liquid interface, $\mathcal{D}_G$ is the gas phase molecular diffusivity (L^2t^{-1}), and H is the dimensionless Henry's Law coefficient. The bulk liquid volume is given by

$$V_L = V_C - V_F \tag{12}$$

where V_C is the total volume allowable for biofilm growth in the segment, and V_F is the biofilm volume. The bulk gas volume, V_G, is constant.

Biofilm

Equations (13)-(26) constitute the MCB model [15] with the one exception that the presented formulation assumes that the volume fraction of water in the biofilm does not change in time or space.

The porous media provides support for biofilm growth. The formation of biofilm on the surface of the porous media is described by a one-dimensional mass balance equation for biomass

$$\frac{\partial C_{F,Pi}}{\partial t} = \frac{\partial j_{F,Pi}}{\partial y} + r_{Pi} \tag{13}$$

where $C_{F,Pi}$ is the concentration (ML^{-3}) of particulate i in the biofilm, z is the distance (L) perpendicular to the solid surface, and $j_{F,Pi}$ is mass flux ($ML^{-3}t^{-1}$) of particulate i in the biofilm in the y direction. Biofilm mass flux results from biofilm growth in the biofilm interior and is given as

$$j_{F,Pi} = u_F C_{F,Pi} \tag{14}$$

where u_F is the advective velocity (Lt^{-1}) of the biofilm matrix due to growth. This velocity is calculated as

$$u_F = \frac{1}{1-\varepsilon_l} \int_0^z \left(\sum \frac{r_{Pi}}{\rho_{Pi}} \right) dy \tag{15}$$

where ε_l is the volume fraction of the liquid phase in the biofilm and ρ_{Pi} is the specific density (ML^{-3}) of particulate i. The relationship between density and concentration is given by

$$C_{F,Pi} = \rho_{Pi} \varepsilon_{Pi} \tag{16}$$

where ε_{Pi} is the volume fraction of particulate i in the biofilm. The boundary conditions are

$$j_{F,Pi}|_{y=0} = 0 \tag{17}$$

at the biofilm-substratum interface and

$$j_{F,Pi}|_{y=L_F} = j_{L,Pi} \tag{18}$$

at the biofilm-bulk liquid interface, where j_F and j_L are the interfacial mass fluxes ($ML^{-2}t^{-1}$) of particulate i in the biofilm and bulk liquid phases, respectively, and L_F is the biofilm thickness. The mass flux j_F is given by

$$j_{F,Pi}\Big|_{y=L_F} = u_{de}C_{F,Pi} - u_{at}C_{FL,Pi} \tag{19}$$

where u_{de} is the velocity (Lt^{-1}) at which biomass is detaching from the biofilm and into the bulk liquid and u_{at} is the velocity (Lt^{-1}) at which biomass is attaching to the biofilm from the bulk liquid. Progression of the biofilm thickness is modeled by

$$\frac{dL_F}{dt} = u_F|_{y=L_F} - u_{de} + u_{at} \tag{20}$$

Dissolved components in the biofilm are modeled by the one-dimensional mass balance equation

$$\frac{\partial(\varepsilon_l C_{F,Di})}{\partial t} = -\frac{\partial j_{F,Di}}{\partial y} + r_{Di} \tag{21}$$

The mass flux is given by

$$j_{F,Di} = -u_F(1-\varepsilon_l)C_{F,Di} - f\,\mathcal{D}_{L,Di}\frac{\partial C_{F,Di}}{\partial y} \tag{22}$$

where f is the ratio of diffusivity in the biofilm versus diffusivity in the bulk liquid. The boundary conditions are

$$j_{F,Di}|_{y=0} = 0 \tag{23}$$

$$C_{F,Di}|_{y=L_F} = C_{FL,Di} \tag{24}$$

$$j_{F,Di} = j_{L,Di} \tag{25}$$

with the interfacial mass flux of dissolved component *i* at the biofilm side of the biofilm-bulk liquid interface given as

$$j_{F,Di} = -u_F(1-\varepsilon_l)C_{F,Di} - f\,\mathcal{D}_{L,Di}\left.\frac{\partial C_{F,Di}}{\partial y}\right|_{y=L_F} \tag{26}$$

Biological Reactions

The bacteria utilize an electron donor and an electron acceptor to produce biomass. Biomass is considered as cellular mass and extra polymeric substance (eps). The rate of production of biomass is modeled by either double Monod kinetics or Haldane kinetics which includes inhibition of the overall rate due to electron donor concentration:

$$r_X = \mu_{\max}\frac{C_D}{K_D + C_D}\frac{C_A}{K_A + C_A}X \tag{27}$$

$$r_X = \mu_{\max}\frac{C_D}{K_D + C_D + \frac{C_D^2}{K_I}}\frac{C_A}{K_A + C_A}X \tag{28}$$

where r_X is the rate of production ($ML^{-3}t^{-1}$) μ_{max} is the maxim specific growth rate (t^{-1}), C_D and C_A are the concentrations (ML^{-3}) of the electron donor and electron acceptor, respectively, K_D and K_A and the half-saturation constants (ML^{-3}) for the electron donor and electron acceptor, respectively, X is the concentration (ML^{-3}) of biomass, and K_I is the inhibition constant (ML^{-3}) with respect to the electron donor. The utilization rates of the electron donor and electron acceptor are given by