Modelling three-phase fluidized bed bioreactor for wastewater treatment

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Abstract
Economically and technically interesting, the bioreactor phenomena are studied on both micro (pore- and particle-size) and macro (bioreactor) levels, to describe synergetic between bio-dynamics and physicochemical dynamics. Still, the three-phase bioreactors modelling remains complex, it required taking into account numerous factors: the pollutant biodegradation rate in the biofilm, the reactant interfacial gas-liquid and liquid-solid mass transfer, the biofilm composition and growth, the granular structure, and all the transfer phenomena occurring between phases in presence. The aim of this paper is to present a model based on biofilm composition and growth, the fluidised bed structure and its hydrodynamics. The simulation results are compared with our experimental data obtained for a biological treatment, carried out in three-phase fluidized bed reactor for hydrocarbons removal from refinery wastewaters. The equipment used is laboratory scale and the obtained results showed that we could degrade hydrocarbons efficiently.

Key-words Biological treatment, refinery wastewaters, three-phase fluidized bed G/L/S

1. Introduction
During the last few years, a great interest is conceded to the biologic treatment processes, which prove to be economic and efficient. Application of fluidization in the biotechnology field has considerably increased. These treatments processes are based on the use of microorganisms, fixed or not, to eliminate the principal pollutants, generally metals and organic substances, by decomposing the inorganic and organic pollutants in simple products. The existing wastewater treatment processes can use three kinds of microbial aggregates: static biofilms as the trickling filters, particulate biofilms as in the fluidized bed bioreactors, upflow anaerobic sludge blanket reactors, the biofilm suspension reactors airlift, and the flocs as in activated sludge processes. To identify the range of suitable operating conditions in wastewater treatment processes, Nicolella et al (2000) proposed a configurations-diagram, which indicates flow rate and concentration intervals for each kind of configuration. Among these available different processes to create efficient contacts between phases, fluidized bed reactor seems to be the best one and present many advantages relating to hydrodynamics and mass transfer phenomena. In these kinds of bioreactors, the solid particles covered by biofilm are fluidized by two ascending flows, air, and contaminated water. With favourable operating conditions, from hydrodynamic and mass transfer point of view, the pollutant could be biologically degraded up to 90%. Therefore, successful application of fluidized bed reactors lead several researchers to propose models that could describe the biological mechanisms with realistic accuracy.

Up-to-date, the complexity of various interactions between microorganisms, mass transport phenomena, and hydrodynamics occurring in these kinds of systems, let proposed models questionable since they are established on number of simplifications and assumptions.

2. Theoretical background
Transfer phenomena taking place in fluidized bed bioreactors are numerous and complex to be described. In fact, aerobic wastewaters treatment by biodegradation consists of an oxygen absorption in liquid phase with biological and biochemical reactions in biofilms where substrate degradation occurs. During these operations four stages can be distinguished:

1) oxygen mass transfer in liquid phase (oxygen dissolution)
2) oxygen and substrate mass transfer through liquid-biofilm interface
3) oxygen and substrate mass transfer by diffusion in biofilm followed by oxidation bioreactions which lead to organic matter degradation
4) biofilm detachment which is due to shearing, abrasion and bioparticles collisions.
In these bioreactors, bioparticles are suspended in bulk liquid through which, the air (or oxygen) moves as dispersed bubbles to be dissolved then transferred to biofilm particles and suspended biomass. There, oxygen is consumed by biochemical reactions. At the same time, the present substrates in bulk liquid has also to penetrate biofilms by molecular diffusion, which is a slow process, to reach reacting site and to be degraded. The substrate concentration in biofilm is determined by the substrate conversion process and diffusion. Three characteristic substrate concentration profiles was defined by Rittmann and McCarty (1981), in fully penetrated biofilms substrate the concentration profile is flat, in deep biofilms the substrate is depleted at or before the particle surface and in shallow biofilms, the substrate concentration profile is between the two cited cases (figure 1).

![Figure 1: Substrate concentration profiles within the biofilm (after Gantzer, 1989 and Suidan, 1987)](image)

2.1. Biofilm characteristics

2.1.1 Biofilm structure:
Complex and heterogeneous the natural biofilm consists on cells and EPS (extracellular polymeric substances) mixtures where cells are embedded in matrix gel of extracellular polysaccharides. In the late 1980s, the confocal scanning laser microscopy (CSLM) allows the visualisation of living biofilm samples in three dimensions by optical sectioning, subsequent computer enhancement, and processing. The other improvement was the use of the immunofluorescence staining and species-specific RNA probes. That why, different species are labelled, in situ. Lawrence et al. (1991) observed three single species biofilms by using CSLM. The structures of these biofilms were spatially heterogeneous, with channels and void spaces permeating the film. The structure architecture also varied from species to species. Massol Deya et al. (1995) observed that degrading groundwater contaminants biofilms consist of dense base films of 10 to 75 µm thickness, and much thicker channelled and diffuse sections of film, up to several hundred micrometers. They also found that the film structure is age-dependent. The young films initially colonizing surface showed no structural patterns and as biofilms aged, channels developed, separating the previously continuous film into discrete clusters or lobes. The conventional mathematical models describe biofilms as planar structures with homogenous cell distribution. Mass transfer through the mass layer boundary and within the biofilm is assumed diffusional and perpendicular to the support surface. However, microscopic observations indicate that biofilms are not flat and the microorganisms’ distribution is not uniform. Biofilms form highly complex structures containing voids, channels, cavities, pores, and filaments, with cells arranged in clusters or layers. Such complex structures found in a wide variety of biofilms, because of spatial gradients, microbial species density, the volume fraction of the water phase (porosity), and tortuosity of biofilms they have to change as the depth of biofilm increases. These spatial distributions of biotic and abiotic components in turn affect the mass transfer mechanisms and diffusivities in biofilms (Bishop et al., 1995). Using a continuum approach and observing the conservation principles, an analytical mathematical model of microbial interaction in biofilms was developed by Wanner and Gujer, (1986) which predicts changes in biofilm thickness and describes the dynamics and spatial distribution of microbial species and substrates in the film.

In general, the biofilm can be divided into two zones, the base film, and the surface film. Both contain an assemblage of microorganisms and other particulate material bound together by a matrix of EPS (Leslie Grady et. al., 1999). The base film consists of structured accumulation, with well-defined boundaries. Molecular diffusion processes controls transport in the base film. The surface film provides transition between the base film and the bulk liquid, with transport within the bulk liquid dominated by advection (Henzel et. al., 1997). The relative thickness of the base and surface films depends not only on the hydrodynamic characteristics, substrates
concentration, and environmental factors of the system, but also on the nature of the microorganisms in the biofilm.

2.1.2 Biofilm thickness and activity:
Generally, bioparticles have not the same size, which leads to stratification in fluidized bed bioreactors. Thicker biofilms are found at the top of the bed, whilst bare carriers and thin biofilms remain in the bottom region of the column (Shieh et al., 1981). Particles stratification is attributed to differences in drag and buoyancy that affect particle terminal settling velocity (Di Felice, 1995). Because of stratification, size distribution, biodegradation rate, biofilm composition and biofilm specific activity change along the height of the bed. Typical biofilm thickness in fluidized bed reactors are in the range of 12-500 µm (Hermanovicz, and Cheng, 1990). Concerning the biofilm density Hirata et al., (2004) reported that the biofilm dry density decrease with increasing biofilm thickness and proposed an empirical equation, relating dry density and biofilm thickness. The complex and heterogeneous structure biofilms leads to assume that a fraction of biofilm is inactive, and it is due to the presence of EPS and inactive bacteria (dead cells and cell debris). In addition, the substrate and microbial distributions in biofilms change with the thickness and are closely related to mass transfer phenomena (C. Nicolella, et al, 2000). In fact, the diffusion coefficient of biofilms, which is lower than that of water, is function of their density, porosity, pore size, type of EPSs and minerals (J. Wimpenny, 2000).

2.1.3 Biofilm detachment:
Defined as biomass removal, from attached microbial film to the bulk liquid phase, the biofilm detachment is attributed to four mechanisms, Grazing: the consumption of bacteria from the outer surface of the biofilm by protozoa, Erosion: the continuous loss of individual or small group of cells, Sloughing: the periodic loss of large portions of the biofilm, and Abrasion: analogous to erosion, but it is caused by particle-particle collisions. The biofilm detachment rate is function of: shear stress magnitude, biofilm thickness, biofilm density, biofilm growth rate, stage of biofilm growth, and biological biofilm composition. Based on precedent work giving the relation between biofilm growth and hydraulic shear stress, Rittmann (1982) developed model to correlate biofilm shear loss as function of hydraulic shear stress and attached biomass amount in fixed film biological reactors. Chang et al. (1991) reported that turbulence and attrition were the dominant factors controlling detachment in a biological two phases fluidized bed reactor. They proposed empirical model for specific biofilm detachment rate coefficient, $r_D$ [day$^{-1}$] with glass beads as medium support. Nicolella et al (1996) also showed in a two phase fluidized bed reactor that the specific detachment rate coefficient strongly increases with increasing liquid velocity. Other parameters, such as particle concentration and liquid shear stress, were found to be less significant. Their correlation is based on dimensionless numbers but is developed for narrow range shear stress values (0.14-0.17 N/m²).

2.2. Mathematical Model
We consider a transient model with assumptions cited below.

2.2.1. Model assumptions:
The following assumptions were adopted for modelling the substrate biodegradation:
- contents in the bulk water are well mixed
- particles are assumed to be spherical in shape and homogeneous in size, and the biofilm is homogeneous with respect to thickness, porosity, composition, and density.
- Active fraction of biofilm remains constant.
- Growth kinetics follow a substrate inhibition model (Haldane)
- The substrate in wastewater and oxygen supplied are growth limiting.
- Transport of dissolved components into and out of the biofilm is by molecular diffusion and can be described by Fick’s second diffusion law.
- Concentration gradients are significant only in the direction perpendicular to the support surface; thus, a one-dimensional model is considered.
- Contribution of suspended microorganisms in substrate biodegradation is not neglected.
- The specific rate of detachment of microorganisms from the biofilm is constant under the operating conditions.
2.2.2. Model development:

Considering the above assumptions, the model equations can be developed as follows:

In the liquid phase:

Considering $S_b$, $S_f$ substrate concentration respectively in bulk liquid and within biofilm, $C_b$ oxygen concentration and $X_S$ suspended biomass concentration in bulk liquid, mass balance applied to the reactor gives the following equations:

**For the substrate (pollutant):**

\[
\frac{dS_b}{dt} = D\left[S_0 - S_b(t)\right] - \left(\frac{\mu_b}{Y_g} + m\right)X_S - \frac{4\pi r_f^2 N_p k_s (S_b - S_f)}{\varepsilon_L V_R}
\]  

(1)

Where the term expressed by:

\[
\left(\frac{\mu_b}{Y_g} + m\right)X_S
\]  

(2)

is substrate uptake rate by suspended biomass with $Y_g$ the true growth yield, according to Pirt (1965) and $m$ the maintenance coefficient.

In this model, since the substrate and oxygen supplied are growth limiting, the growth kinetics are assumed to follow Monod kinetics with respect to oxygen and substrate inhibited kinetics with respect to pollutant: Haldane kinetics. Haldane equation has been used widely in the modelling of various wastewater treatment systems and has been recommended for its simplicity.

So specific growth rate equation is:

\[
\mu_b = \frac{\mu_m C_b}{K_O + C_b} \frac{S_b}{K_s + S_b + (S_b^2/K_f)}
\]  

(3)

Where $\mu_m$ is the maximum specific growth rate, $K_O$ is oxygen saturation constant, $K_S$ is substrate saturation constant and $K_f$ is the inhibitory coefficient.

**For oxygen:**

\[
\frac{dC_b}{dt} = D\left[C_0 - C_b(t)\right] - \left(\frac{\mu_b}{Y_o}\right)X_S - \frac{4\pi r_f^2 N_p k_o (C_b - C_f)}{\varepsilon_L V_R}
\]  

(4)

**For suspended biomass:**

\[
\frac{dX_S}{dt} = \mu_b X_S - DX_S + \frac{4\pi N_p \rho_f r_D (r_f^3 - r_p^3)}{3\varepsilon_L V_R}
\]  

(5)

**In the biofilm:**

Mass transfer occurs across the liquid-biofilm interface as well as diffusion and biological reactions within the biofilm. With respectively $S_b$ substrate concentration and $C_f$ oxygen concentration in the biofilm, the non-steady-state relations for biofilm diffusion with reaction can be represented in spherical polar coordinates by the following equations:

\[
\frac{\partial S_f}{\partial t} = D_{sf} \frac{1}{r^2} \left(\frac{\partial}{\partial r} (r^2 \frac{\partial S_f}{\partial r})\right) - \left(\frac{\mu_f}{Y_g} + m\right) f \rho_f
\]  

(6)

\[
\frac{\partial C_f}{\partial t} = D_{cf} \frac{1}{r^2} \left(\frac{\partial}{\partial r} (r^2 \frac{\partial C_f}{\partial r})\right) - \left(\frac{\mu_f}{Y_o}\right) f \rho_f
\]  

(7)

With like boundary conditions:

\[
D_{sf} \frac{\partial S_f}{\partial r} = 0 \quad \text{and} \quad D_{cf} \frac{\partial C_f}{\partial r} = 0 \quad \text{at} \quad r = r_p
\]  

(8)

At liquid-biofilm interface, the flux across the interface from the liquid phase must be balanced by the flux across the interface into biofilm, so that the second boundary equations can be written as follows:
Biofilm growth and detachment: For a bioparticle, the biofilm mass increasing rate is given by the following expression:

\[ \frac{dm_{\text{bf}}}{dt} = r_{\text{croissance}} - r_{\text{détachement}} \]  

With

\[ r_{\text{croissance}} = 4\pi p \int_{r_p}^{r_f} \mu_{\text{f}} r^2 \, dr \]  

The inactive fraction of the biofilm is supposed to be constant. Alike, the detachment rate is the same for the two fractions active and inactive thus the rate of detachment for a bioparticles is:

\[ r_{\text{détachement}} = \frac{4}{3} \pi (r_f^3 - r_p^3) \rho_{\text{f}} r_D \]  

In addition, since the biofilm density is constant the volume variation is equal to the mass variation:

\[ 4\pi r_f^2 \rho_{\text{f}} \frac{dr_f}{dt} = \frac{dm_{\text{bf}}}{dt} \]  

Thus, the rate of biofilm growth is given in the radial direction by the following expression:

\[ \frac{dr_f}{dt} = \frac{1}{r_f^2} \left( \int_{r_p}^{r_f} \mu_{\text{f}} r^2 \, dr - \frac{1}{3} (r_f^3 - r_p^3) r_D \right) \]  

3. Experimental system description

3.1 Experimental setup

The experimental unit (figure 2) is composed by column (ID = 9.4 cm, H = 100 cm), disengagement zone, distributors, calming zone, feeding zone of water and gas, and zone of liquid phase recirculation. Along this column, fourteen pressure taps are located at 3 cm intervals for the pressure profile. The Plexiglas feeding zone is cylindrical part with two entries located at the bottom and side of the column for the water and gas supply. Compressed air is used for aeration (0-90 l/h). Airflow is selected to ensure sufficient dissolved oxygen concentration for the microorganisms breathing, and to have at the same time, dispersed bubbles mode flow. The wastewater circulation was carried out through the bed of bioparticules in closed loop using a peristaltic pump. The Liquid flow recirculation was maintained at 1,5 Q_{mil}. The parameters of refinery wastewaters used are given in Table 1. The column was fitted (10%) by sand particles seeded with refinery wastewaters during 7 days. The identification tests confirmed the presence of bacteria type pseudomonas aeroginosa specific to mediums contaminated by hydrocarbons. The experimental conditions used are listed in Table 2.

### Tableau 1: refinery wastewaters Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PH</th>
<th>T (°C)</th>
<th>COD (mg/L)</th>
<th>BOD (mg/L)</th>
<th>O_{2} (mg/l)</th>
<th>O_{3}^- (mg/l)</th>
<th>NO_{3}^- (mg/l)</th>
<th>PO_{4}^{2-} (mg/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Refinery wastewaters</td>
<td>7.36</td>
<td>23</td>
<td>211.2</td>
<td>175</td>
<td>2.85</td>
<td>0.197</td>
<td>0.0015</td>
<td></td>
</tr>
</tbody>
</table>

### Tableau 2: Experimental data

<table>
<thead>
<tr>
<th>Parameters</th>
<th>( U_1 ) (cm/s)</th>
<th>( U_g ) (cm/s)</th>
<th>( \rho_s ) (kg/m3)</th>
<th>( \rho_l ) (kg/m3)</th>
<th>( d_p ) (µm)</th>
<th>W (kg)</th>
<th>( H_0/D_c )</th>
</tr>
</thead>
<tbody>
<tr>
<td>value</td>
<td>0.03 – 0.6</td>
<td>0.06 – 0.20</td>
<td>2560</td>
<td>1000</td>
<td>283</td>
<td>1</td>
<td>1.06</td>
</tr>
</tbody>
</table>
4. Computation, results and discussion:

The model equations were solved at different time. The substrate and oxygen concentration profiles within the biofilm were approximated as parabola and Leibniz’s Rule was applied to an expansion of equations (6) and (7). A computer program using MATLAB permitted the iterative loop calculations for solving the model equations. The experimental results used are given in Table 3.

**Table 3: Experimental results in three phase fluidized bioreactor**

<table>
<thead>
<tr>
<th>Day</th>
<th>0</th>
<th>1</th>
<th>3</th>
<th>5</th>
<th>7</th>
<th>9</th>
<th>11</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD (mg/L) (refinery Wastewater)</td>
<td>211.2</td>
<td>123</td>
<td>96</td>
<td>76.8</td>
<td>61.4</td>
<td>57.6</td>
<td>38</td>
</tr>
<tr>
<td>BOD₅ (mg/L) (refinery wastewater)</td>
<td>175</td>
<td>100</td>
<td>72</td>
<td>58</td>
<td>45</td>
<td>10</td>
<td>5</td>
</tr>
</tbody>
</table>

Experimental results obtained in a semi batch bioreactor show that hydrocarbon biodegradation requires 5 days. Hydrocarbon concentration falls nearly by the half of its initial value during the first two days and decreases rapidly to the final value close to zero 3 days after. This is confirmed by COD and BOD measurements Table 3. As phenol is a major toxic soluble organic compound in refinery wastewater, simulation tests were achieved by considering transfer, diffusion as well as biokinetics parameters relating to phenol and given by the literature.
The model predicts complete biodegradation after only 14 hours Fig.3a. As regards to biomass concentration; the increasing values are nearly corresponding to the decreasing values of substrate concentration. After maximum value attained during the first 14 hours, the curve decreases slowly until end of treatment. The model shows also that biofilm thickness increases quickly during the fourteen hours of treatment from 4µm to a maximum value of 7 µm and then decrease at end of experiment Fig.3b.

Our experimental results differ from that simulated; the difference is essentially due to the fact that we have:
- Supposed homogeneous the complex and heterogeneous biofilm structure.
- Used mass transfer and biokinetics parameters values of phenol in the model. Phenol does not accurately represent all hydrocarbons dissolved in water. Indeed, among the various dissolved hydrocarbons exist molecules of more complex structure compared with phenol and thus require more significant time for their transfer, diffusion, and assimilation.

The model developed for this study suggests that:
- There is advantage in determining hydrocarbons transfer and biokinetics parameters, which, were supposed to be close to those of phenol.
- In exploring more the variation of some parameters, which, were supposed to be constant.

These allow best understanding of the biodegradation in three phase-fluidized beds reactor and its efficiency for the refinery wastewater.

**Fig. 3. Simulation results: (a) Concentration evolution with time for substrate, suspended biomass and oxygen, (b) biofilm thickness evolution with time.**

**Conclusions**

The aim of this paper was to present model based on biofilm composition and growth, the fluidised bed structure and its hydrodynamics. The simulation results and our experimental data obtained for a biological treatment, carried out in three-phase fluidized bed reactors for hydrocarbons removal from refinery wastewaters shows that the predicted values are less than the data obtained experimentally and that could be due to some hypothesis taken as some important parameters that could be heterogeneous and variable.

**Notation**

- \( C_a \): oxygen concentration in the bulk liquid, [g/L]
- \( C_f \): oxygen concentration within biofilm, [g/L]
- \( D \): liquid dilution rate, \( Q_l/V_l \), [h^{-1}]
- \( D' \): gaz dilution rate, \( Q_g/V_l \), [h^{-1}]
- \( D_{of} \): diffusivity of oxygen in the biofilm, [cm²/s]
- \( D_{sf} \): diffusivity of substrate in the biofilm, [cm²/s]
- \( f \): fraction of active bacteria in the biofilm
- \( k_o \): liquid-biofilm mass transfer coefficient for oxygen, [cm/s]
- \( k_s \): liquid-biofilm mass transfer coefficient for substrate, [cm/s]
- \( L_f \): biofilm thickness [cm]
- \( m \): substrate maintenance coefficient, [g/g.h]
- \( m_bf \): mass of biofilm on a single particle, [g]
- \( N_p \): total number of particles in the reactor at any given time
- \( r \): radial position within biofilm, [cm]
- \( r_{D} \): specific rate of detachment, [h^{-1}]
rf : radial position at biofilm surface, [cm]

rp: radial position at particule surface, [cm]

Sb: substrate concentration in the bulk liquid, [g/L]

Sf: substrate concentration within biofilm, [g/L]

S0: substrate concentration in the feed, [g/L]

VR: reactor liquid volume, [L]

Xf: biofilm biomass concentration in the reactor, [g DW/L]

Xs: suspended biomass concentration in the reactor, [g DW/L]

Yg: true growth yield coefficient, [g DW/g substrate]

Greek symbols:

εL: liquid phase holdup

µ: specific growth rate, [h⁻¹]

µb: specific growth rate in the liquid phase, [h⁻¹]

µf: specific growth rate in the biofilm at a given radial distance, [h⁻¹]

ρf: biofilm dry density, [g DW/cm³ wet volume]

References


