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Quantification of particulate matter attached to the bulk-biofilm interface and its influence on local mass transfer

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ABSTRACT

A large fraction of the chemical oxygen demand (COD) in municipal wastewater is associated with the particulate matter. The presence of these particles might impose a negative impact on the mass transfer in biofilm systems when they attach to the bulk-biofilm interface. We thus investigated the impact of real wastewater particles by combining optical coherence tomography (OCT) and oxygen microsensor measurements. The deposition of wastewater particles of different size classes onto the biofilm surface was captured in 3D by means of OCT in a lab-scale flume. The thickness of the particle layer was calculated from OCT images. The influence of the particle deposition with respect to oxygen mass transfer into the biofilm was monitored by measuring oxygen profiles before and after particle deposition. By employing OCT, the formation of the particle layer on the biofilm surface was monitored *in situ* and non-invasively over time. Results from oxygen microsensor measurements concluded 20–70% reduction of the dissolved oxygen flux from the biofilm surface into the biofilm matrix. This decrease in oxygen availability lowered substrate conversion in biofilms exposed to high loadings of organic wastewater particles.

1. Introduction

A large part of the chemical oxygen demand (COD) in municipal wastewater is associated with particulate matter [15,26]. The size of these wastewater particles ranges from submicron to several hundred of microns in diameter. However, substrate uptake by bacteria is limited to molecules of a molecular weight < 1000 Da [13], meaning these organic particles cannot be used by microorganism directly. In most wastewater treatment systems, the removal of carbon and nutrients is limited by the hydrolysis of organic particles [20]. Many researchers have focused on the hydrolysis of organic particles, defined as the sum of all the processes that make the particulate organic matter available for microbial growth [12,20]. Due to the complexity of the particles contained in wastewater with respect to their chemical composition [19] and broad range of particle size, model compounds with simplified composition and well defined particle characteristics are often used to investigate the mechanism of hydrolysis as well as the interaction between particles and biofilms. The model compounds often used include bovine serum albumin [5], starch [21], dextran [5,13], soy protein [21] and egg protein [7]. Confer and Logan [5] used bovine serum albumin and dextran as model compounds to investigate protein and polysaccharide degradation, respectively. They speculated that the hydrolysis of macromolecules repeated a loop of cell-associated hydrolysis followed by the release of hydrolytic fragments, which diffuse back into the bulk liquid until these fragments are small enough to be assimilated by bacteria. Very low hydrolysis rate coefficients were determined by Eliosov and Argaman [10] for both raw primary suspended solids and for non-settable solids in an activated sludge system. Using artificially produced protein particles from hard boiled eggs, Dimock and Morgenroth [7] clearly showed that with their particle break-up model hydrolysis was influenced by the size of the organic particles. By incorporating the particle break-up process from large into small particles, the increase of the specific surface area was captured, which successfully correlated the physical characteristics of the particles to the hydrolysis kinetics.

Boltz and La Motta [1] suggested to divide the removal of organic particles in biofilm systems into four steps: (1) transport of the particles to the biofilm surface, (2) attachment of the particles, (3) hydrolysis of the particles, followed by the release of hydrolytic products and (4) biochemical transformation of these products. As the aforementioned studies mostly focused on the latter two steps, the removal of dissolved substances is well understood. However, there is still a lack of

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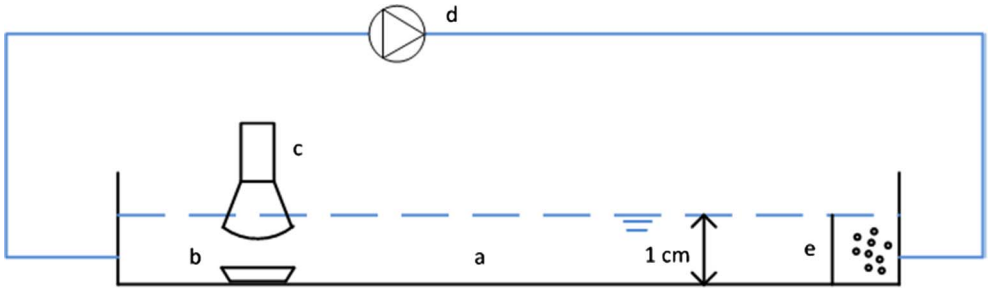

Fig. 1. Schematic drawing of the experimental setup (side view) composed of: (a) flume, (b) biofilm carrier, (c) OCT or oxygen electrode, (d) pump and (e) a weir to stabilize the flow. The dimension of the flume is 60 (length) × 8 (width) × 6.1 (height) cm^3. Aeration was provided near the inlet.

knowledge on the transport of organic particles and their interaction with biofilms. Direct contact between bacteria and organic particles is a prerequisite for hydrolysis [5]. In the theoretical speculation of Bouwer [2], the authors separated the deposition of particles in biofilm systems into two steps. Particles are firstly transported from the bulk liquid to the bulk-biofilm interface, which is a physical process governed by hydrodynamics. Then the particles attach to the biofilm surface, which is a physicochemical process depending on the properties of the biofilm surface, particles and the chemistry of the surrounding solution. Drury et al. [9] demonstrated that microbeads with a diameter of ≤1 μm can readily penetrate biofilms. However, increasing particle sizes lead to a reduced mass transport into the biofilm, as large particles being retained at the biofilm surface [3,4,20]. To date, the attachment of larger particles to the biofilm surface has not been assessed. Further, it is still not elucidated how the particles would organize themselves at the bulk-biofilm interface.

The physiology and morphology of biofilms may be altered by the attachment of particles at the bulk-biofilm interface, thereby influencing the mass transfer of dissolved substances. With a lab-scale rotating drum reactor, Särner [23] concluded that the attachment of organic particles onto the biofilm surface lowered the removal of dissolved substances at high temperature and high glucose concentration. It was speculated that such a negative effect might be caused by a local oxygen shortage in the biofilm matrix as a result of the degradation of organic particles attached [24]. On the contrary, de Kreuk et al. [6] reported increased oxygen consumption in aerobic granular five hours after addition of particulate starch. These examples show that there is still a lack of understanding how the attachment of particles at bulk-biofilm interface would affects transport as well as transfer of dissolved substances to and into biofilms.

Based on the analysis above, we defined the objectives of this study as to (i) visualize and (ii) quantify the attachment of raw wastewater particles of different particle size classes onto the bulk-biofilm interface by means of optical coherence tomography (OCT) as well as (iii) to evaluate the impact of the particle deposition on the substrate transport into the biofilm performing oxygen microsensor measurements.

2. Materials and methods

2.1. Characterization of the organic particles

Municipal wastewater was collected after the grid chamber at the sewage treatment plant in Neureut (Karlsruhe, Germany) and served as the particle source. By immediately sieving the raw wastewater, the wastewater particles were separated into different size classes: $d_p \geq 500$ μm, $250 \leq d_{p1} < 500$ μm, $100 \leq d_{p2} < 250$ μm, $45 \leq d_{p3} < 100$ μm and $28 \leq d_{p4} < 45$ μm [28]. Particles were resuspended in tap water and stored at 4 °C before usage. Total solids (TS) was measured from the resuspension to determine the particle concentration in the municipal wastewater.

The densities of the different size fractions were quantified with a pycnometer (Duran, Mainz, Germany). The particle solutions were firstly filtered to remove the bulk liquid. The particle density (ρ_s) was calculated according to

$$\rho_s = \frac{m_2 - m_0}{(m_1 - m_0) - (m_3 - m_2)} \cdot \rho_w \quad (1)$$

where m_0 and m_1 are the mass of the empty pycnometer and the mass of the pycnometer filled with water, respectively. m_2 is the mass of the pycnometer filled with the drained particles to be determined. m_3 is the mass of the pycnometer filled with drained particles and water. ρ_w is the density of water.

2.2. Experimental procedure

The experiments were conducted in a lab-scale flume. The experimental setup is illustrated in Fig. 1. The flume had a dimension of 60 × 8 × 6.1 cm^3, the total water volume was 0.8 L. A pump was used to recirculate the flow. A Chip M carrier (AnoxKaldness, Lund, Schweden) cultivated with heterotrophic biofilms was taken from a lab-scale moving bed biofilm reactor fed with glucose and fixed inside the carrier compartments of the flume. Biofilm together with deposited particles were visualized using a GANYMEDE-I spectral domain OCT (Thorlabs GmbH, Dachau, Germany). Oxygen microsensor measurements were performed by means of a fully automated setup (Unisense A/S, Aarhus, Denmark). OCT and microsensor measurements were performed in sequence.

The volumetric flow rate in the flume was adjusted to 30 $mL \cdot s^{-1}$. The water depth in the flow cell was adjusted to 1 cm by the weir (see Fig. 1), which led to an average flow velocity of 5 $cm \cdot s^{-1}$.

The carrier was placed 50 cm away from the inlet so that laminar flow (Re = 400) could develop before reaching the carrier. Before any measurement, the system was aerated for 1 h to achieve a DO (dissolved oxygen) concentration between 8 and 9 mg/L. COD (glucose) concentration in the bulk was 400 $mg \cdot L^{-1}$ at the beginning of the experiment.

Before adding particles to the system, a 3D OCT image was acquired to serve as blank for the image analysis. Particles were added at the end of the flume. The particle concentration was set to a value, which was ten times higher compared to raw wastewater. These high particle concentrations were chosen to enhance the effect of the particles on the biofilms, which otherwise could have been difficult to observe if too few particles deposited on the biofilm surface. To monitor the particle deposition onto the biofilm surface, OCT images were acquired 20 min, 1 h, 2 h, 3 h and 4 h after particle addition.

2.3. Microsensor measurements

The microsensor measurements were conducted with a 1D motorized MicroProfiling system (Unisense A/S, Aarhus, Denmark). SensorTrace PRO was used to log the data and control the microsensor