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Simultaneous use of a crossflow filtration membrane as microbial fuel cell anode – Permeate flow leads to 4-fold increased current densities

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ABSTRACT

A new concept for the combination of membrane bioreactors and microbial fuel cells is introduced, that aims at the production of electricity for reducing the overall energy consumption of wastewater treatment. In contrast to previous approaches, the anode is integrated as microfiltration membrane in sidestream crossflow configuration. Using a stainless steel filtration membrane with *G. sulfurreducens* and an acetate-based synthetic medium, up to 4-fold higher current densities are achieved. In a standard setup without filtration, a membrane of filter grade 1 µm shows current densities of $5.8\,A\,m^{-2} \pm 0.5\,A\,m^{-2}$ compared to $> 11\,A\,m^{-2}$ when it is used simultaneously as membrane filter. With smaller pore sizes of filter grade 0.5 µm, $4.4\,A\,m^{-2} \pm 0.5\,A\,m^{-2}$ in a standard setup and $> 15\,A\,m^{-2}$ in a filtration setup are achieved. The permeate flow was identified as the main parameter leading to increased current densities.

1. Introduction

The combination of membrane bioreactors (MBRs) and bioelectrochemical systems such as microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) is a promising approach to reduce the net energy consumption of wastewater treatment while obtaining a high quality effluent.

Membrane bioreactors (MBRs) are an established wastewater treatment technology with a high yearly global market growth rate of 10.5% (Kraume and Drews, 2010). Their main advantage is the complete decoupling of solid retention time (SRT) from hydraulic retention time (HRT). This way, high quality effluents free of solids and pathogens regardless of granulation or settling properties can be obtained. The operation of MBRs requires active pumping or gas sparging on the surface of the filtration membrane to induce turbulences and thus prevent fouling. This results in a high energy demand leading to high operational costs. Depending on the implementation (anaerobic vs. aerobic), configuration (submerged vs. sidestream), module geometry, wastewater strength and loading rates, temperature, sludge age and mixed liquor suspended solids, the energy demand can range from 0.03 to $16.52\,kWh\,m^{-3}$ (Martin et al., 2011). For comparison, the energy demand of conventional activated sludge treatment is usually lower (e.g. between 0.3 and $1.89\,kWh\,m^{-3}$ in the case of municipal wastewater treatment plants in Japan (Mizuta and Shimada, 2010)).

Microbial fuel cells (MFCs) and microbial electrolysis cells (MECs), are an emerging new technology to directly convert the organic fraction of wastewaters into electricity (Scott and Yu, 2016). In these systems, electroactive bacteria oxidize organic carbon under anoxic conditions and transfer the released electrons to an anode. At the cathode, either oxygen (MFC) or protons (MEC) are reduced as terminal electron acceptors, yielding electrical energy or hydrogen as products, respectively. At present, low current densities and thus high investment costs are considered to be among the main hurdles for successful commercialization of this new technology. In literature, three main concepts to combine MFCs/MECs and MBRs are reported (Yuan and He, 2015) (for details the reader is referred to the on-line supplementary material accompanying this article):

1. Serial arrangement of MFCs/MECs and an anaerobic fluidized membrane bioreactor (Ren et al., 2014).
2. Placement of the MFCs/MECs and a filtration module in a single reactor (Ge et al., 2013; Tian et al., 2015).
3. Simultaneous use of the MFCs/MECs cathode or separator as the filtration membrane (Katuri et al., 2014; Malaeb et al., 2013; Su et al., 2013; Wang et al., 2013).

In all these approaches, the membrane filtration modules are operated in submerged mode, which generally results in lower permeate fluxes compared to sidestream configurations. Furthermore, in the published configurations microbial anodes and filtration modules are

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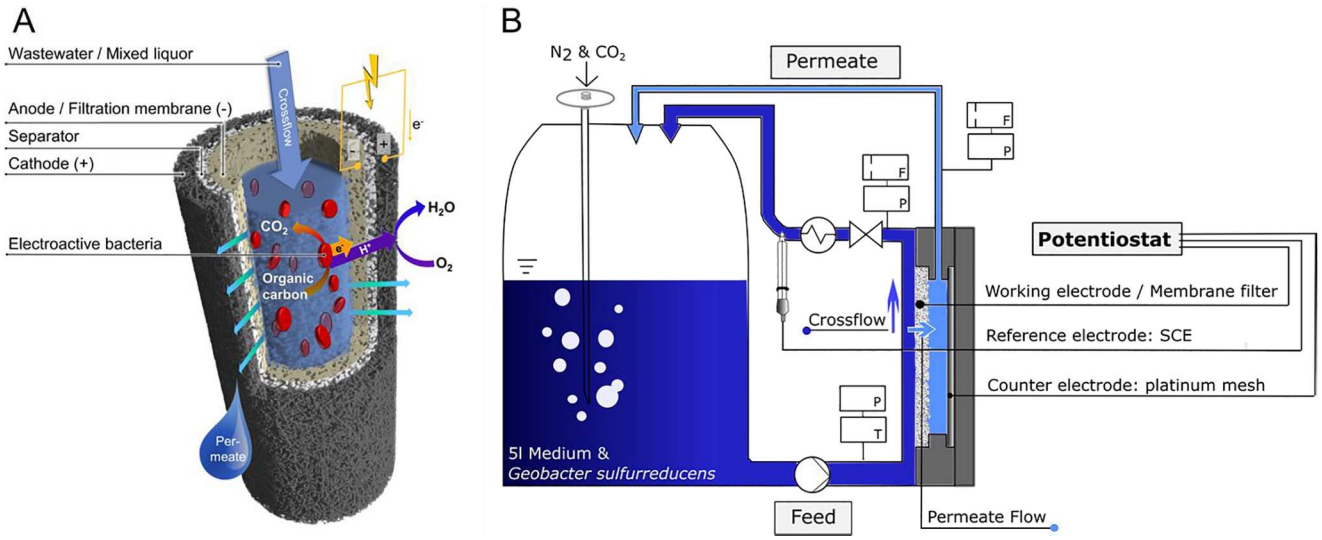

Fig. 1. New principle of using the anode as membrane filter to combine MFCs/MECs and MBRs. **A**: Vision of the implementation as a combined tubular membrane and MFC. The figure shows one examplary tube with the filtration membrane serving as anode in an MFC. The mixed liquor is pumped through the tube perpendicular to the membrane surface (crossflow configuration). The separator ensures the electrical insulation between anode/membrane and cathode. On the cathode oxygen is reduced. An overpressure in the inner part of the tube leads to a permeate flow through the membrane, enhancing proton transport from anode to cathode. **B**: Experimental setup of the lab-scale tests with a 5 L reactor and 10 cm^2 flat plate membrane configured as electrochemical half cell.

integrated as two separate functional units (examples 1 & 2), which likely results in high investment cost. This is not the case when the MFC/MEC cathode or separator membrane and the filtration membrane form a single functional unit (example 3). However, to enable filtration at the cathode the microbial anode must be arranged at a certain distance. This significantly increases ohmic resistance and negatively affects current density and performance of an MFC or MEC.

To overcome these issues, we developed a new concept in which the microbial anode and the MBR filtration membrane form an integrated functional unit operated in crossflow mode. In Fig. 1A, the principle of our new concept is exemplarily shown for a tubular filtration module in MFC configuration. At its centre an electrically conductive filtration active layer is situated, which doubly functions as microbial anode. Its envisioned application is the treatment of wastewater in an anaerobic MBR (AnMBR).

Prerequiste for the realization of this new concept is an efficient electron transfer from the electroactive bacteria to the anode under the conditions of crossflow filtration. In particular, high shear rates caused by the crossflow may be potentially hindering biofilm formation and thus impede direct electron transfer (Pham et al., 2008). With respect to our new concept for the combination of MFC/MEC and MBR this leads to the determining question if significant current on a microbial anode can be generated in a crossflow regime typical for AnMBRs.

In the present work, we demonstrate that simultaneous crossflow filtration and microbial current generation at an electrically conductive filtration membrane is not only possible but actually advantageous as the anode current densities increase with the permeate flow. These experiments were performed as a half-cell setup (Fig. 1B) with an acetate-based synthetic medium, using commercially available stainless steel filtration membranes together with the electroactive model bacterium *Geobacter sulfurreducens* as microbial anode. The synthetic medium and the single strain have deliberately been chosen to ensure reproducible conditions and allow a conclusive comparison of different experiment batches with and without filtration.

2. Materials and methods

2.1. Experimental setup

The use of a half cell system enables us to specifically study anode performance under crossflow conditions without any limitations due to the cathode reaction. The chosen crossflow velocities are typical or higher compared to state-of-the-art AnMBRs to ensure similar fouling conditions. As different research groups use different reactor configurations, materials, and electrochemical testing environments, classification and comparison of results with literature values is challenging. Therefore, the comparability of filtration and non-filtration conditions of the microbial anodes is ensured by conducting half cell experiments in a state-of-the-art setup prior to the filtration experiments. Within these experiments, the suitability of the sintered metal filters as microbial fuel cell anodes together with *Geobacter sulfurreducens* under non-filtrating conditions were characterized using the 6-electrode half-cell setup reported elsewhere (Kipf et al., 2014). To this end, chronoamperometry experiments at two subsequent potentials were performed in triplicates in the same reactor in fed-batch mode. Initially, a potential of −0.159 V vs. SHE was applied for 9 days. Subsequently, the potential was switched to 0.000 V vs. SHE for another 6 days. For these experiments MM 510 type potentiostat systems (Material Mates, Milano, Italy) were used. To ensure sterility, the reactor was autoclaved at 121 °C for 20 min prior to use. The reference electrode was inserted after being sterilized in H_2O_2 for two hours.

For electrochemical experiments with simultaneous filtration a commercial crossflow filtration setup (Sartoflow Study, Sartorius, Germany) was used. This device comprises a Sartojet 4-piston membrane pump (maximum flow rate of 0.7 $m^3 h^{-1}$ at 4 bars), pressure sensors, flowmeters, and automated data acquisition. It was fitted with a custom filter cassette made from polypropylene to accommodate the electrically conductive filtration membrane used as anode, and a counter electrode operating as hydrogen evolution cathode (for details the reader is referred to the on-line supplementary material accompanying this article). The filtration membranes were 10 mm × 100 mm in size, and the feed channel in front of the membrane was 10 mm × 5.5 mm to achieve the desired crossflow velocities of 0.9–2.7 m s^{-1}. The transmembrane pressure (TMP; difference between the pressure on the feed and the permeate side) was adjusted manually to values between 1 and 3 bars with a valve on the retentate line. As there are pressure losses along the feed channel, the TMP was calculated based on the mean value of the pressure before and after the membrane cassette. The system was operated in fed-batch mode in which the permeate was recirculated to a 10 L reactor made from