Journal of Membrane Science 319 (2008) 76–81

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Ammonium removal in advective-flow membrane-aerated biofilm reactors (AF-MABRs)

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ARTICLE INFO

Article history:
Received 2 October 2007
Received in revised form 12 March 2008
Accepted 14 March 2008
Available online 22 March 2008

Keywords:
Hollow-fiber membrane fabric
Advective flow
Ammonium removal
Bioreactor
Model

ABSTRACT

A novel membrane-aerated biofilm reactor configuration was tested in which substrate was delivered to the biofilm via advective as well as diffusive transport. Ammonium-rich synthetic wastewater was pumped across a fabric of hollow-fiber membranes and biofilm grew around and between the membrane fibers to form a continuous structure. High ammonium removal rates (10–24 g N m^{-2} d^{-1}) were achieved in our reactors compared to the conventional biofilm processes, which rely mostly on diffusive transport for substrate delivery. The removal rates were in good agreement with our mathematical model predictions. Low pressure drops (<0.05 atm) were maintained across the biofilm throughout the course of study. The major problem with this novel configuration appeared to be the short-circuiting due to non-uniform structure of the biofilm formed on membrane fabric. Further research is needed to address the problem of short-circuiting by studying the effect of operational parameters and membrane design on the behavior of system.

1. Introduction

In recent years gas permeable membranes have been used for gas transfer during water and wastewater treatment applications [1–6]. Whenever these membranes are used for wastewater treatment, they become rapidly covered with biofilm, which decreases the gas transfer efficiency to the water. The development of an active biofilm, however, may actually increase the rate of gas transfer across the membrane to the biofilm, which can play a beneficial role in wastewater treatment [7,8].

Membrane-aerated biofilms (MABs) differ from conventional biofilms because the oxygen from the membrane diffuses into the biofilm, while contaminants in the wastewater (e.g., COD or NH_4^+) diffuse into the biofilm from the liquid side. This counter-diffusion of electron acceptor and electron donor, respectively, creates a unique microbial stratification that ranges from oxygen-rich to anoxic [9,10]. As a result, several investigators have noted that MABs can simultaneously achieve COD and complete nitrogen removal within the same biofilm [3–5,11].

Unfortunately, as MABs become excessively thick, the electron donor and electron acceptor have to diffuse over greater distances. This increased resistance to mass transfer reduces the biofilm performance substantially [7,9,12–14]. Previous attempts to address this problem by increasing oxygen partial pressures and creating higher shear conditions have been largely ineffective [9,15].

We believe that the decline in the performance of MABs has roots in the mechanism of mass transport within the biofilm (i.e., diffusive transport) and that the resistance to the mass transfer within the MABs might be overcome by forcing wastewater to flow through the biofilm instead of flowing over it. This configuration would take advantage of advective and diffusive transport simultaneously. As a result, the performance of MABs would be expected to increase due to the increased rate of substrate delivery. We have previously developed a computer model to predict the performance of this configuration [16]. This steady-state model was used to predict the removal of a single substrate (e.g., COD) by a single population of microorganisms (e.g., aerobic heterotrophs) assuming a two-dimensional domain, no detachment, and a biofilm of constant porosity and density. The model suggested that higher COD removal rates were feasible compared to the conventional configuration.

In the present study, we experimentally investigated the performance of advective-flow membrane-aerated biofilm reactors (AF-MABRs). Sheets of stitched hollow-fiber membrane fabric were used to support the biofilm. The fabric was comprised of parallel, hollow-fiber membranes that were stitched to provide an even spacing between adjacent fibers. Air or oxygen was supplied to the membranes and a synthetic wastewater was pumped through the fabric. The biofilm grew around the individual fibers, became thicker, and eventually grew together to form a continuous structure around and between the fibers. Using this experimental design,

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doi:10.1016/j.memsci.2008.03.020

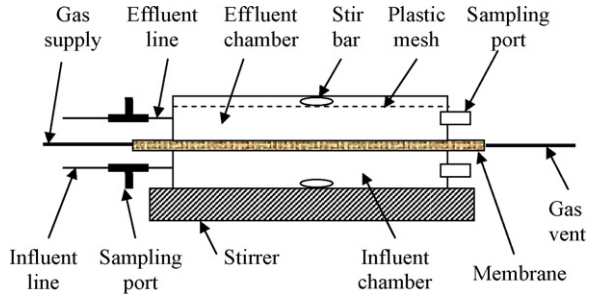

Fig. 1. Schematic of the AF-MABR.

we tested our hypothesis that higher contaminant removal rates can be achieved using AF-MABRs with a relatively low pressure drop across the biofilm. A secondary goal of these experiments was to calibrate/evaluate the mathematical model developed previously.

2. Materials and methods

2.1. Experimental design

Two 230 mL square reactors were constructed (Fig. 1), each housing a fabric of hollow-fiber membrane that divided the reactor into influent and effluent chambers. Synthetic wastewater was pumped into the influent chamber, through the membrane fiber fabric, and into the effluent chamber before exiting the reactor. The membrane fabric in contact with the wastewater was 7.3 cm × 7.3 cm (53 cm^2). Removal rates were calculated based upon the above surface area (53 cm^2), which is the effective surface area of the biofilm, rather than the membrane surface area (102 cm^2). The liquid in the influent and effluent chambers was mixed using magnetic stirrers and stir bars (length: 1.3 cm; diameter: 0.3 cm). To make the mixing more effective, however, the small stir bars were replaced with larger ones (influent chamber: length: 6.5 cm, diameter: 0.8 cm; effluent chamber: length: 3.8 cm, diameter: 0.8 cm) on day 119 of the reactors operation (see below). The stir bar in the upper effluent chamber was contained by plastic mesh to prevent it from contacting the membrane.

The fabric of hollow-fiber membrane (specifications in Table 1) was potted into two plastic manifolds. One manifold was connected to the gas supply and the other one was left open to the atmosphere for venting. The gas flow rate was adjusted to provide a residence time of less than 1 s within the membrane fibers. The gas pressure drop across the membrane fibers was negligible and therefore gas pressure inside the fibers was close to atmospheric pressure.

For the purpose of this study, we decided to study the nitrification performance of AF-MABRs. A nitrifying biofilm was encouraged to grow on the membrane fabric by supplying an ammonium-rich wastewater that contained no organic substrates. The reactor was initially inoculated with a mixed culture of nitrifying bacteria that had been enriched from wastewater collected from the Metropolitan Wastewater Treatment Plant (St. Paul, MN, USA). The temperature of the reactors was maintained at 23 ± 2 °C. The synthetic wastewater consisted of (per liter of deoxygenated and deionized water): 12 mg magnesium sulfate, 1.1 mg calcium chloride, 1 mL of SL7 trace elements solution [17], 136 mg monobasic potassium phosphate, 142 mg dibasic sodium phosphate, 85 mg sodium carbonate, 500–750 mg sodium bicarbonate, and ammonium chloride. The pH of the feed was typically between 8.0–8.4 and the effluent pH varied between 6.7 and 7.9. The feed reservoir headspace was flushed with nitrogen gas to prevent re-oxygenation.

Table 1
Membrane specifications

Manufacturer	Celgard
Type	Composite (a non-porous skin on the fibers outer surface)
Material	Polyolefin
Oxygen permeance (stdcc s^{-1} cm^{-2} $cmHg^{-1}$)	3×10^{-5}
Inside diameter (μm)	200
Outside diameter (μm)	380
# of fibers (cm)	16
Fibers center-to-center spacing (μm)	625

The reactors were set up in parallel with an enrichment culture of nitrifying bacteria and initially operated at 10 L m^{-2} h^{-1} (Lmh). During the study, the loading rates, oxygen partial pressure and the mixing conditions within the reactors were manipulated (Table 2). The performance of the reactors was monitored by measuring the concentrations of ammonium, nitrate, nitrite, and dissolved oxygen. The pressure drop across the biofilm was also measured.

2.2. Analytical methods

Ammonium was measured according to Nessler method [18]. Nitrate and nitrite were measured according to Standard Methods [19] using a UV-spectrophotometer (DU®530, Beckman Coulter Inc., CA, USA) and an ion chromatograph (761 Compact IC, Metrohm-Peak Inc., TX, USA), respectively. Nitrite measurements were performed only occasionally as no nitrite was typically detected in the reactors. Samples were filtered through a 0.2 μm membrane filter prior to the above analyses. Samples for dissolved oxygen measurement were taken from the influent and effluent chambers and analyzed immediately using a DO meter (OM-4 Oxygen Meter, Microelectrodes Inc., NH, USA) equipped with an oxygen electrode (MI-730, Microelectrodes Inc., NH, USA). The permeability of the biofilm was measured by pumping dechlorinated tap water into the reactor and through the biofilm at different flow rates and measuring the pressure drop across the biofilm using a differential pressure gauge (0–5 psig, CeComp Electronics, IL, USA). A dye test was performed during the course of study by pumping dechlorinated tap water containing methylene blue into the reactors and the dye flow pattern was visually observed.

Table 2
Operational conditions used for the reactors throughout the course of study

Condition	Days of operation	NH_4^+-N (mg L^{-1})	Flux (Lmh)	Oxygen partial pressure (atm)	Mixing	HRT (min)
Start-up	1–40	20–35	10	0.21	Slow	260
1	40–48	40–50	10	0.21	Slow	260
2	48–58	40–50	20	0.21	Slow	130
3	58–72	40–50	20	0.6	Slow	130
4	72–119	40–50	30	0.6	Slow	85
5	119–146	40–50	30	0.6	Fast	95
6	146–165	40–50	30	0.6	Intermittent	95