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Membrane aeration bioreactors for wastewater treatment: completely mixed and plug-flow operation

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Abstract

A laboratory scale membrane aeration bioreactor (MABR) with a void volume of 1.35 l was tested for its ability to treat synthetic wastewater. The MABR process couples aerobic biological wastewater treatment with the bubbleless mass transfer of pure oxygen through hollow fibre membranes. The biofilm that was formed on the surface of the hollow fibres utilised oxygen transferred from the fibre lumen for the degradation of organics transferred from the synthetic wastewater flowing over the biofilm. When operated as a completely mixed reactor at an organic loading rate of 24.5 kg chemical oxygen demand (COD)/m^3/day and a hydraulic retention time of 34 min, an 89% COD removal efficiency was achieved. During plug flow operation, 86% COD removal efficiency was obtained at an organic loading rate of 5.8 kg COD/m^3/day and a hydraulic retention time of 47 min. Regular membrane cleaning to control biomass growth was necessary to prevent channeling. The ability of the reactor to operate under plug flow conditions demonstrated that the biofilm plays a significant role in oxygen mass transfer.

Keywords: Wastewater treatment; Hollow fibres; Bioreactor; Bubbleless membrane oxygenation; Pure oxygen; Biofilm; Completely mixed reactor; Plug flow reactor

1. Introduction

Combining membrane technology with biological reactors for the treatment of municipal and industrial wastewaters has led to the development of three generic membrane processes within bioreactors: (i) for the separation and recycling of solids; (ii) for bubbleless aeration of the bioreactor and (iii) for the extraction of priority organic pollutants from hostile industrial wastewaters. Commercial aerobic and anaerobic membrane separation bioreactors already provide a small footprint alternative to conventional biological treatment methods, producing a high quality effluent at high organic loading rates. Both bubbleless aeration and extractive membrane bioreactors are in the development stages [1].

Oxygen mass transfer using a synthetic membrane to provide bubbleless aeration for the biological treatment of wastewaters has been demonstrated on a laboratory scale bioreactor with volumes ranging from 0.5 to 6.0 l [2,3]. Organic removal between 63–91% of influent chemical oxygen demand (COD) at loading rates between 0.06 and 8.94 kg/m^3/day have been reported [4–7]. Since no oxygen bubbles are formed, gas stripping of volatile organic compounds and foaming due to the presence of surfactants can be prevented [8–10]. Bubbleless oxygen mass transfer can be accomplished using gas permeable dense membranes or hydrophobic microporous membranes [11]. Both plate and frame, and hollow fibre membrane configurations have been used to supply oxygen. The specific surface area of such membranes ranges from a 19 m^2/m^3 plate and frame unit to a hollow fibre unit of 5108 m^2/m^3, far greater than in conventional attached-growth bioreactors [6,7]. Research has focused on hollow fibres with the gas phase on the lumen side and wastewater on the shell side of the fibres. These provide a high surface area for oxygen transfer and biofilm formation whilst occupying a relatively small volume within the bioreactor. Since lumen gas partial pressure is independent of tank depth, large transfer driving forces in shallow tanks can be achieved [11]. Fluidised hollow fibre bundles with fibres moving independently are also known to be less susceptible to clogging [12].

Oxygen diffusion through a dense membrane material can be achieved at high gas pressures without bubble formation. In hydrophobic microporous membranes, the pores remain

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gas filled, oxygen is transported to the shell-side of the membrane through the pores by gaseous diffusion or Knudsen flow transport mechanisms. The partial pressure of oxygen is kept below the bubble-point to ensure bubbleless supply of oxygen in clean membrane aeration processes [8,10,12–14]. The conventional bubble-point measurement is used to determine membrane pore size, wherein the gas pressure on one side of the membrane is increased steadily until gas bubbling is observed in the stagnant liquid phase on the other side of the membrane. The bubble-point pressure is calculated from the liquid surface tension, contact angle of the liquid at the membrane and the membrane pore size. However, conventional bubble-point measurement is not directly applicable to the membrane aeration bioreactor (MABR) process in which wastewater flows over a biofilm attached to the membrane surface. Though the physical and chemical characteristics of the wastewater and biofilm will alter the liquid surface tension, contact angle and effective membrane pore size, the wastewater velocity as well as oxygen utilisation within the biofilm will also affect the pressure at which gas bubbles are observed.

Pressurised hollow fibres have been investigated in dead-end and flow-through modes of operation. The evacuation of carbon dioxide from the bioreactor is a benefit in flow-through operation, though no quantitative work to determine removal rates has been undertaken [2,11]. Until recently, dead-end operation was avoided due to significantly decreased performance and condensate formation in the lumen [11]. Treatment of the sealed ends of these hollow fibres provides the means for condensate to escape and makes a 100% oxygen transfer efficiency achievable [7].

In most MABR processes, a biofilm will form on the membrane surface, which is in intimate contact with the oxygen source and protected against abrasion and grazing [2,8]. Thus, oxygen transfer resistance due to the thickness of the porous membrane and the liquid boundary layer are not necessarily decisive limiting factors [2,8,9]. On a laboratory scale MABR xylene no longer moved from the bulk liquid phase to the gas phase once a biofilm had developed on the open-ended silicon tubes, demonstrating that biofilms degrade volatile organic compounds that may otherwise be stripped out of the system via the gas phase [4]. Excessive biofilm accumulation can result in substrate mass transfer limitations, decline in biomass activity, metabolite accumulation within the biofilm, and the channeling of flow in the bioreactor, such that steady-state conditions are not maintained [4,5,7]. To operate at maximum efficiency, occasional membrane cleaning, air scouring backwashes, and high liquid shear regimes have all been employed to control biomass accumulation [5,7–9].

Several models exist that describe oxygen and pollutant mass transfer, and biofilm kinetics within the MABR process [4,15–18]. Mass transfer of oxygen and pollutant substrates has commonly been described by Fickian diffusion. Zero-order, first-order and Monod equations have all been employed to describe kinetic behaviour within the biofilm. The models developed, along with limited collaborative experimental data, have shown that the location of highest activity within the biofilm varies with biofilm thickness, the ratio of oxygen and pollutant concentrations, oxygen and pollutant diffusivities in the biofilm and liquid boundary layer, and their stoichiometric reaction coefficients [4,15–17]. Most investigations concur that for a fixed and sufficient oxygen supply pressure and fixed biofilm thickness, increasing the pollutant concentration results in the active layer moving from the biofilm/liquid interface towards the biofilm/membrane interface [4,15–17].

The supply of pure oxygen and the high-rate recirculation of wastewater needed to maintain a high liquid shear velocity at the biofilm surface and reduce the liquid boundary layer thickness, constitute the energy required during MABR operation. Therefore, any reduction in oxygen or recirculation would reduce the operating cost, making the MABR process a more attractive wastewater treatment option. It has been shown that the MABR process can achieve a 100% oxygen utilisation efficiency during the high rate nitrification of a synthetic ammonia-rich wastewater [19]. This paper investigates the treatment of a synthetic wastewater under both completely mixed (high rate recirculation and energy demand) and plug flow (no recirculation and low energy demand) operating conditions.

2. Materials and methods

The MABR module contained a vertically and centrally mounted fibre module consisting of 1140 microporous, 280 μm diameter polypropylene based hollow fibre membranes (Fig. 1). Pure oxygen was supplied to one end of the hollow fibres via a manifold at the base of the MABR module. The other ends of the fibres were sealed with the fibres moving independently of one another in the module. The fibres had a packing density of 4%, membrane surface area of 0.69 m^2, and when placed in the perspex tube, the reactor had a void volume of 1.35 L.

The bioreactor was operated continuously at a range of organic loading rates under completely mixed and plug-flow conditions. A sterile solids-free, peptone based synthetic wastewater was used as the feed concentrate to ensure consistent influent quality [20]. The concentrate was delivered to the reactor via a syringe needle using a peristaltic pump (Watson Marlow, Model 503S). Tap water was employed to dilute the concentrate. During completely mixed operation, the influent was delivered to the recirculation line. The high upflow recirculation rate of 1.3 l/min ensured that the liquid phase was completely mixed. During plug-flow operation, recirculation was terminated. Oxygen (99.7% pure, BOC) was passed through the lumen of the fibres and regulated using a pressure gauge and flow meter (Cole–Parmar Instruments model L-03293-00). A 500 ml sampling bottle was placed in the recirculation line so that

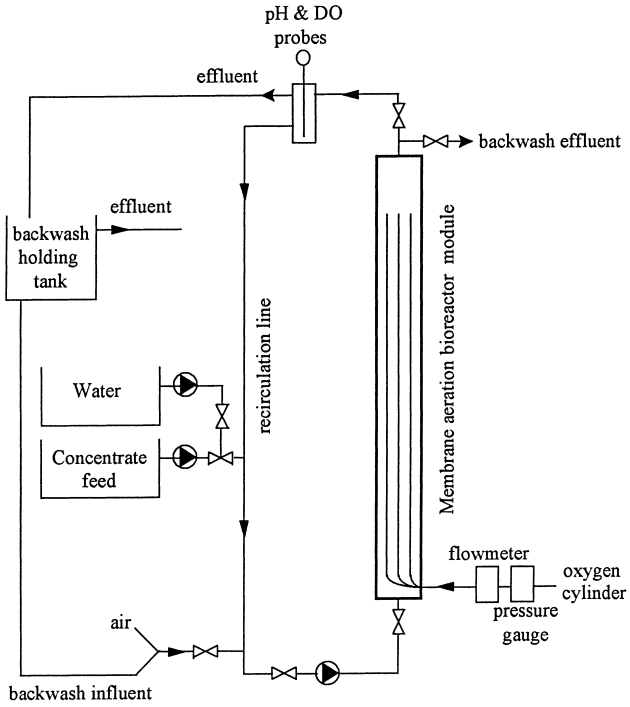

Fig. 1. Schematic of the laboratory scale membrane aeration bioreactor.

in-line dissolved oxygen (DO), pH and temperature measurements could be made.

During start-up, to establish biofilm growth on the fibres, synthetic wastewater seeded with a 40% innoculum of secondary effluent from Cranfield Wastewater Treatment Works was circulated through the reactor for 3 days. The membranes were cleaned daily soon after start-up to remove excess biomass. This consisted of a 10 s air scour using compressed air at a pressure of 108 kPa followed by a 60 s water flush at 1.8 l/min using final effluent. This procedure was repeated three times. At the end of the membrane cleaning process, the water in the reactor was replaced with treated effluent.

Filtered samples for COD, nitrate and nitrite were analysed according to Standard Methods [21]. Ammonia was measured with a probe (pHOX, model 182). Effluent suspended solids (ESS) were vacuum filtered through a glass microfibre filter and dried. Probes in the sampling bottle were used to measure dissolved oxygen (Jenway, model 9070), pH and temperature (Jenway, model 3070).

3. Results

3.1. Completely mixed operation

The MABR was operated as a completely mixed reactor for a total of 206 d at five different organic loading rates (Table 1). The temperature, pH and DO averaged 23.1°C, 6.3 and 7.6 mg/l, respectively. A recirculation rate of 1.3 ± 0.2 l/min ensured that the bulk liquid phase was completely mixed and had a bulk velocity of 72 m/h, equivalent to a modified Reynolds number of <100. This modified Reynolds number takes into account the effect of the wetted perimeter of the hollow fibres [12,14,22].

A thick biofilm developed rapidly during the first week of operation and membrane cleaning commenced on Day 9. Regular membrane cleaning procedures prevented the development of an excessively thick biofilm and channeling of the liquid phase. At an organic loading rate of 3.3 kg/m^3/day, the COD removal efficiency was 70% (1st loading, Table 1). At organic loading rates of 6.6 and 8.6 kg/m^3/day, respective removal efficiencies of 80% and 84% were achieved (2nd and 3rd loading). A maximum COD removal efficiency of 89% was achieved at organic loading rates of 12.4 and 24.5 kg/m^3/day during completely mixed operation of the MABR (4th and 5th loading).

The ESS concentration increased as the organic loading rate increased. As the feed was a solids-free synthetic wastewater, all solids detected in the effluent resulted from biomass removal from the MABR. At an organic loading rate of 3.3 kg/m^3/day, the ESS averaged 8 mg/l (1st loading) compared to 39 mg/l at an organic loading rate of 24.5 kg/m^3/day (5th loading). As the organic loading rate increased, the biomass concentration within the module increased,

Table 1
Steady state performance during completely mixed MABR operation

Loading	1	2	3	4	5
Days	1–97	97–143	143–179	179–192	192–206
Organic loading rate (kg COD/m^3/day)	3.3 ± 0.0	6.6 ± 0.2	8.6 ± 0.0	12.4 ± 0.2	24.5 ± 0
Influent COD (mg/l)	144 ± 1	299 ± 10	395 ± 1	575 ± 9	1135 ± 0
Actual HRT (min)	60 ± 1	52 ± 3	38 ± 2	37 ± 2	34 ± 2
COD removal (%)	70 ± 2	80 ± 1	84 ± 0	89 ± 1	89 ± 2
ESS (mg/l)	8 ± 5	7 ± 6	14 ± 5	21 ± 8	39 ± 11
Influent NH_4-N (mg/l)	6.8 ± 0.1	14.0 ± 0.5	18.6 ± 0.3	27.1 ± 0.4	53.4 ± 0
Effluent NH_4-N (mg/l)	16.6 ± 0.8	34.7 ± 2.2	49.1 ± 3.7	77.5 ± 4.0	147 ± 2.5
NO_3-N (mg/l)	8.2 ± 1.0	3.8 ± 1.8	5.6 ± 2.4	4.2 ± 4.2	0.6 ± 0.3
NO_2-N (mg/l)	1.2 ± 0.7	0.6 ± 0.2	0.5 ± 0.2	0.5 ± 0.3	0 ± 0
pH	6.4 ± 0.1	6.4 ± 0.1	6.2 ± 0.1	6.1 ± 0.1	6.1 ± 0.2
DO (mg/l)	7.8 ± 1.4	9.8 ± 2.3	9.3 ± 2.2	7.1 ± 4.7	2.4 ± 1.3

Table 2
Steady state performance during plug flow MABR operation

Loading	1	2	3	4	5
Days	1–6	6–11	11–19	19–27	27–34
Organic loading rate (kg/m^3/day)	3.1 ± 0.1	5.8 ± 0.6	9.0 ± 0.3	10.5 ± 0.2	1.3 ± 0.3
Influent COD (mg/l)	139.2 ± 5.5	268.5 ± 31.9	410.3 ± 16.1	485.5 ± 22.2	512.4 ± 14.6
COD removal (%)	83 ± 4	86 ± 4	66 ± 1	28 ± 3	25 ± 5
ESS (mg/l)	5 ± 4	22 ± 22	50 ± 35	18 ± 6	7 ± 7
Influent NH_4-N (mg/l)	10 ± 1	18 ± 3	27 ± 1	27 ± 2	26 ± 3
Effluent NH_4-N (mg/l)	18 ± 1	29 ± 2	31 ± 9	32 ± 5	43 ± 11
NO_3-N (mg/l)	7.7 ± 0.0	6.4 ± 0.4	7.1 ± 0.5	5.9 ± 0.5	6.5 ± 0.8
NO_2-N (mg/l)	0.1 ± 0.1	0.3 ± 0.1	0.1 ± 0.0	0.2 ± 0.0	0.3 ± 0.1
pH	6.5 ± 0.3	6.2 ± 0.3	6.4 ± 0.2	6.4 ± 0.1	6.4 ± 0.3
DO (mg/l)	10.9 ± 0.6	16.1 ± 5.0	10.9 ± 6.3	7.8 ± 1.5	15.7 ± 5.1

resulting in an increase in the naturally sloughed biofilm that appeared in the effluent.

At low organic loading rates, nitrite and nitrate were detected in the effluent at concentrations of 1.2 and 8.2 mg/l, respectively (1st loading, Table 1). As the organic loading rate increased, concentrations of nitrite and nitrate decreased to 0.0 and 0.6 mg/l (5th loading), respectively. Effluent ammoniacal-nitrogen (NH_4-N) concentrations exceeded influent NH_4-N concentrations due to the proteinaceous breakdown during biofilm growth releasing ammonia.

3.2. Plug-flow operation

The MABR was operated as a plug-flow reactor at five different organic loading rates over a 34-day period (Table 2, Fig. 2). Plug-flow operation was achieved by terminating the recirculation of wastewater. A thick non-uniform biofilm rapidly developed on the membrane surface and membrane cleaning commenced on Day 4. The average temperature, pH and DO measured in the effluent were 21.0°C, 6.4 and 12.3 mg/l, respectively.

At organic loading rates of 3.1 and 5.8 kg/m^3/day, the COD removal efficiencies were 83% and 86%, respectively (1st and 2nd loadings, Table 2 and Fig. 2). Thereafter, as the organic loading rate was increased progressively, the COD removal efficiency declined, and at organic loading rates of 9.0 and 11.3 kg/m^3/day, the respective COD removal efficiencies were 66% and 25% (3rd and 5th loading). Effluent suspended solid concentrations were comparatively high during plug-flow operation (Table 2). Furthermore, the ESS concentration varied more widely than observed during completely mixed operation, showing no obvious trend as the organic loading rate was increased.

Effluent nitrate concentrations ranged from 5.9 to 7.7 mg/l, whilst effluent nitrite concentrations remained below 0.3 mg/l (Table 2). Effluent ammonia concentrations were again higher than influent concentrations.

4. Discussion

Chemical oxygen demand removal efficiencies reached a maximum of 86% at an organic loading rate of 5.8 kg/m^3/day during plug-flow operation compared to 89% at an organic loading rate of 24.5 kg/m^3/day during completely mixed operation. In the completely mixed MABR, the substrate concentration was the same along the length of the modules and equaled the effluent concentration. The effluent COD concentration ranged from approximately 43

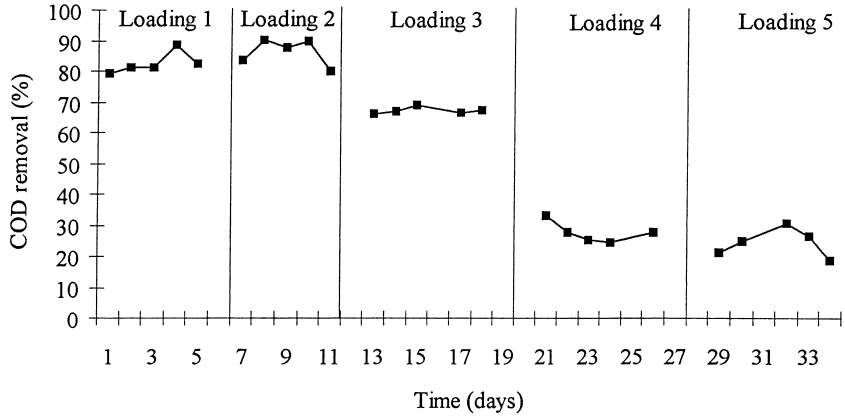

Fig. 2. Percentage COD removal efficiency during plug-flow operation of the MABR at five different organic loading rates.

to 124 mg/l as the organic loading rate was increased. Plug-flow conditions resulted in a substrate concentration gradient along the length of the module, with the highest concentrations at the influent end of the module. The effluent COD concentration, which represents the lowest concentration in the MABR during plug-flow operation, ranged from 24 to 384 mg/l as the organic loading rate was increased. The COD saturation constant for mixed heterotrophic cultures ranges from 5 to 30 mg/l [23]. Therefore, during completely mixed operation, the MABR performance may have been substrate limited at the lower organic loading rates. However, at higher organic loading rates, the COD concentration within the reactor was greater than the saturation concentration and therefore, mass transfer limitation may have occurred. During plug-flow operation, the performance may have been limited by the reduction in the rate of substrate mass transfer to the biofilm due to the increase in the boundary layer thickness. However, Brindle et al. [24] achieved 81% COD removal efficiency at an organic loading rate of 27 kg/m^3/day in a pilot-plant MABR operated under plug-flow conditions, and the negative effect of the increase in liquid boundary layer thickness was countered by the high organic concentrations along the length of the module.

The DO concentration in the bulk liquid throughout the experiment remained significantly greater than 2.0 mg/l, with higher concentrations present in the biofilm matrix than measured in the bulk liquid. The oxygen saturation constant for mixed heterotrophobic bacterial populations ranged from 0.5 to 1.0 mg/l [23]. As oxygen was directly transferred to the biofilm, the rate of oxygen mass transfer and oxygen concentration did not limit the rate or degree of organic removal. Exceptionally high effluent DO concentrations were observed during plug-flow operation (Table 2), with higher levels along the length of the module and at the membrane/biofilm interface. High oxygen partial pressures at the membrane/biofilm interface could lead to oxygen toxicity and microbial lysis within the biofilm. This phenomena has been observed in several biofilm MABR processes where high oxygen concentrations have been suspected [25,26]. However, oxygen toxicity of a biofilm attached to dead-end microporous hollow fibres was not suspected during the treatment of a high oxygen demanding wastewater operated under similar conditions [24].

During completely mixed and plug-flow operation of the MABR, solids were observed in the effluent due to the rapid growth and natural detachment of the biofilm. Under completely mixed conditions, the concentration of biological solids in the effluent increased as the organic loading rate increased. Though membrane cleaning was undertaken daily, the biofilm thickness that developed between each successive cleaning was greater at higher organic loading rates, and naturally, more biomass was detached from the biofilm. During plug-flow operation, the effluent bio-solids concentration showed no clear trend as regards organic loading rate. However, effluent bio-solids concentrations tended to be significantly higher during plug-flow operation compared to completely mixed operation at similar organic loading rates (Tables 1 and 2). Similar observations were made by Brindle et al. [24] during the treatment of wastewater containing suspended solids. These workers speculated that at high recirculation flow rates, the MABR had a greater filtration capacity than during plug-flow operation when the recirculation was terminated. The lower ESS concentrations achieved during completely mixed operation may have also been influenced by the increase in the biofilm density and its adherence to the membrane surface as observed at high liquid velocities by Debus et al. [27] or oxygen toxicity resulting in cell lysis during plug-flow operation.

5. Conclusions

- A membrane aeration bioreactor operated under complete mixed conditions achieved 89% COD removal efficiency at a volumetric loading rate of 24.5 kg/m^3/day and a HRT of 34 min.
- During plug flow operation, a maximum COD removal efficiency of 89% was achieved at a loading rate of 5.8 kg COD/m^3/day and a HRT of 47 min. Operating at higher organic loading rates was limited by either the rate of substrate mass transfer or oxygen toxicity.
- Daily membrane cleaning was necessary to maintain performance and prevent channelling.
- The utilisation of oxygen in the biofilm attached to the membrane surface maintained the oxygen concentration gradient required to drive oxygen mass transfer.

6. Nomenclature

COD	chemical oxygen demand (mg/l)
ESS	effluent suspended solids (mg/l)
MABR	membrane aeration bioreactor
DO	dissolved oxygen
NH_4-N	ammoniacal-nitrogen (mg/l)
NO_2-N	nitrite-nitrogen (mg/l)
NO_3-N	nitrate-nitrogen (mg/l)

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