Evaluation of intermittent air sparging in an anoxic denitrification membrane bioreactor

E. J. McAdam, A. L. Eusebi and S. J. Judd

ABSTRACT

The impact of intermittent air sparging on the operation of an anoxic (dissolved oxygen $< 0.1\,mg\,l^{-1}$) immersed membrane bioreactor (iMBR) applied to potable water denitrification is discussed. Air sparge length and specific aeration demand per unit membrane area (SAD_m) were varied to determine impact on oxygen transfer and membrane fouling. For $SAD_m > 0.39\,m^3\,m^{-2}\,h^{-1}$ with sparge lengths of 10 to 60 seconds, a low dissolved oxygen residual of 0.05 to $0.90\,mg\,O_2\,l^{-1}$ was formed which typically inhibited denitrification; oxygen transfer efficiency increased with increasing sparge time. Residual oxygen was rapidly consumed at a rate, r_{O_2}, of $0.35\,mg\,O_2\,l^{-1}\,min^{-1}$. Once oxygen was depleted, denitrification proceeded. When intermittently sparging at a $SAD_m < 0.39\,m^3\,m^{-2}\,h^{-1}$ for 30 seconds (following 10 minute dead-end filtration cycles in the iMBR), no dissolved oxygen residual was observed and a flux of $21\,l\,m^{-2}\,h^{-1}$ was sustained with fouling rates $< 0.001\,m\,bar\,min^{-1}$ recorded. This method provides for effective integration of air sparging into anoxic/anaerobic iMBR environments to simplify process design and delivers a tangible reduction in specific energy demand from $0.19\,kWh\,m^{-3}$ (for constant sparging) to $0.007\,kWh\,m^{-3}$.

Key words | air, anoxic, denitrification, fouling, membrane bioreactor, sparging

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INTRODUCTION

There is a current interest in adapting membrane bioreactor (MBR) technology to replace ion exchange for the removal of nitrate from potable water; the key driver is the reduction of waste volume as up to 2% of product flow in ion exchange is converted into a nitrate concentrated brine requiring disposal (McAdam & Judd 2008*a*,*b*). To date studies of this application both at laboratory scale (Chang *et al.* 1993) and full scale (Urbain *et al.* 1996) have principally focussed on externally configured (sidestream) MBR as fouling can be controlled by adjusting fluid velocity rather than air flow (as for immersed MBR) which permits simpler maintenance of the anoxic conditions required to facilitate effective denitrification (dissolved oxygen $< 0.1\,mg\,l^{-1}$). However, immersed MBR offers significant reductions in specific energy demand versus pumped sidestream; consequently several authors have recently attempted to integrate immersed membranes by using novel gas strategies. Rezania *et al.* (2007) successfully used recycled headspace gas (principally comprising nitrogen gas) for sparging of an immersed membrane. Intermittent nitrogen gas sparging (c. 30 seconds every 10 minutes) has also been demonstrated to promote sustained permeability of immersed membranes at relatively high fluxes in anoxic conditions (McAdam & Judd 2008*a*,*b*); this protocol offers further 'aeration' savings versus standard immersed MBR applications operated either under constant or cyclical (10 s on/10 s off) gas sparging conditions. Whilst these studies have proven effective, implementation of headspace recirculation at full scale requires considerable process adaptation (McAdam & Judd 2008*a*,*b*); additionally, pressure regulation of the headspace is required as research suggests a significant increase in nitrogen partial pressure

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can inhibit microbial growth (Arao *et al.* 2005). Lee *et al.* (2001) opted to circumvent headspace recirculation and identified that by integrating an air backwash through the immersed membrane with an interval <5 seconds every 10 minutes (in conjunction with a 63 μm pre-filter), long-term membrane operation could be maintained without recording a dissolved O_2 residual. The current paper examines the integration of air (to replace nitrogen) into an intermittent gas sparging protocol (McAdam & Judd 2008*a*,*b*) to deliver a simplified anoxic MBR process design with a minimum specific energy demand. The study specifically addresses: (a) oxygen mass transfer in anoxic conditions; (b) impact of air on anoxic biological denitrification; and (c) identification of limits for sustained fouling control.

MATERIAL AND METHODS

Experimental unit

The anoxic MBR comprised a 75 L bioreactor with an impeller mixer that provided low energy mixing at a speed of 30 rpm. The shear imparted to the membrane from this mixer was demonstrated to be negligible (McAdam & Judd 2008*a*,*b*). A hollow fibre out-to-in PVDF membrane module was used with a total area of 0.93 m^2 and nominal pore size 0.04 μm. Trans-membrane pressure was monitored using a calibrated -0.5 to 0.5 bar g pressure transducer (Gem Sensors, UK) sited on the permeate line with data recorded on a 16-bit data logger (ADC-16, Pico-technology). Air was supplied through an integrated aerator sited 0.70 m below the liquid surface and was controlled with a 0 to 25 l min^{-1} needle gauge (RS components, UK). Solids retention time (SRT) was 25 days during this study resulting in an MLSS concentration of 1.1 ± 0.2 g l^{-1}. Three SRT were reached prior to analysis. Ethanol was supplied as the exogenous substrate (C:N ratio 1.45 g g^{-1}) to support denitrification. The analogue feed comprised an influent nitrate concentration of 22.6 mg NO_3^--N l^{-1} to simulate groundwater. The membrane was operated in dead-end mode. Under this protocol, permeate withdrawal was undertaken in the absence of gas sparging up to a set filtered volume (V_f); upon reaching V_f, permeate withdrawal was ceased and gas sparging of the surface undertaken. Further experimental detail is available in McAdam & Judd (2008*a*,*b*).

Analysis

Dissolved oxygen measurements were undertaken using a calibrated meter (WTW Oxi340I); the probe was sited at 0.30 m below the liquid surface and 0.40 m above the aerator. Batch respirometry was conducted in triplicate on 250 ml sludge samples extracted from the anoxic MBR to determine ethanol uptake under aerobic conditions. The respirometer used an electrolytic multi cell differential pressure sensor for measurement and absorption of CO_2 and simultaneous supply of O_2 (CES Ltd., UK). Bubble size measurement was undertaken in clean water conditions at identical hydrostatic pressure to the MBR for classification of the aerator (coarse or fine) by using a clear perspex tank to enhance camera resolution; as this was an approximation only, process variables that may influence bubble dynamics including salinity, bubble compression, viscosity and solids concentration were temporarily neglected.

Mass transfer analysis

The oxygen transfer coefficient was based on two-film theory where diffusion through the liquid film is rate limiting and is given by k_L. Due to the difficulty associated with specific measurement of the interfacial area, k_La and the overall mass transfer coefficient (s^{-1}) is given by:

$$\frac{dC}{dt} = -k_La(C^* - C) \quad (1)$$

where C is the gas concentration in the bulk phase (ML^{-3}) and C^* is the gaseous saturation concentration in the liquid phase (ML^{-3}) which reflects salinity concentration, temperature and pressure of the field tests. k_La was corrected for temperature to 20°C ($\theta = 1.024$). Where biomass exerts an oxygen demand, the rate of microbial uptake of oxygen (r_{O_2}, $ML^{-1}T^{-1}$) can be incorporated:

$$\frac{dC}{dt} = -k_La(C^* - C) - r_{O_2} \quad (2)$$

The volumetric oxygen transfer rate (OTR) was calculated using:

$$OTR = k_La \cdot C^* \cdot V \quad (3)$$

where V is tank volume.

RESULTS

Intermittent aeration

At a specific aeration demand per unit membrane area (SAD_m) of 0.39 $m^3 m^{-2} h^{-1}$ (volumetric air flow rate, Q_a 4.8 $m^3 m^{-3} h^{-1}$), the bulk liquid phase dissolved oxygen (O_2) concentration did not increase from a background concentration of 0.03 $mg l^{-1}$ for sparge times ranging 10 to 60 seconds (Figure 1). However, on increasing $SAD_m >$ 0.39 $m^3 m^{-2} h^{-1}$, dissolved O_2 concentration increased, recording a maximum of 0.9 $mg l^{-1}$ corresponding to an SAD_m of 1.94 $m^3 m^{-2} h^{-1}$ and sparge time of 60 seconds. Lee *et al.* (2001) similarly observed an increased dissolved O_2 concentration when air backwash time was set to 10 seconds in their immersed anaerobic MBR; an air backwash of 5 seconds was therefore retrospectively adopted to avoid oxygen inhibition and maximise methanogenesis.

After normalising SAD_m for sparge time and membrane area, a near linear correlation ($r^2 = 0.95$) between air volume and dissolved O_2 concentration was found (Figure 2). In anoxic conditions, inhibition of denitrification has been cited to occur when dissolved O_2 increases above 0.09 $mg l^{-1}$ (Oh & Silverstein 1999). Based on data from this study, approximately 7.5 l of air can be applied before reaching this limit which corresponds to sparge times of 74.5 and 15 seconds for SAD_m of 0.39 and 1.94 $m^3 m^{-2} h^{-1}$ respectively. Below 3 l of supplied air, dissolved O_2 remained at background concentration implying that sparge times of 30 and 6 seconds could be implemented at a SAD_m of 0.39 and 1.94 $m^3 m^{-2} h^{-1}$ respectively without inducing residual dissolved O_2.

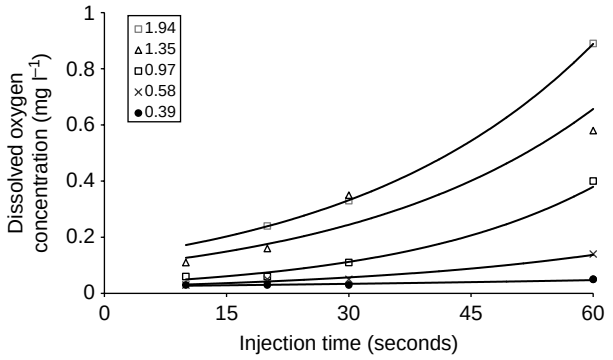

Figure 1 | Intermittent aeration trials conducted in-situ. SAD_m range 0.39–1.94 $m^3 m^{-2} h^{-1}$ (6 to 30 l min^{-1}). Sparge times of 10–60 seconds adopted.

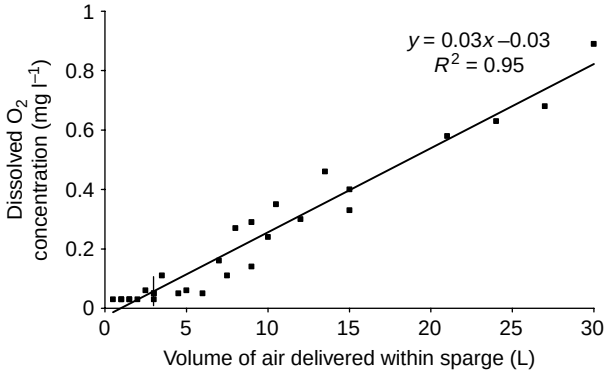

Figure 2 | SAD_m data range re-plotted to demonstrate a relationship between air supply volume and residual dissolved oxygen (O_2) concentration.

Dissolved O_2 recovery and biological uptake

With increasing SAD_m, the time taken to recover to the initial dissolved O_2 concentration (O_2 concentration pre-sparge) increased; a maximum recovery time of 150 seconds was recorded for a 60 second sparge at a SAD_m of 1.94 $m^3 m^{-2} h^{-1}$ (Figure 3). Recovery time was dependent upon biological activity; whilst several authors have reported simultaneous metabolism of electron acceptors (Patureau *et al.* 1996), with an increase in dissolved O_2 concentration, an inhibition of nitrate reduction was observed as microorganisms preferentially utilised O_2 as the electron acceptor due to the higher available energy yield. Once oxygen was consumed, denitrification proceeded. Based on recovery data within the reactor, r_{O_2} was approximated at 0.35 mg O_2 l^{-1} min^{-1} or 19.2 mg O_2 g SS h^{-1}. Batch respirometric analysis validated r_{O_2}

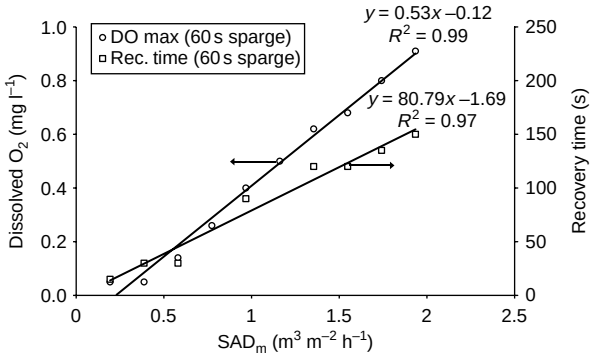

Figure 3 | Residual dissolved oxygen concentration post 60 second air sparge at various SAD_m. Time to recover to initial dissolved oxygen concentration (c. 0.03 mg l^{-1}) also recorded as a function of SAD_m.

data recording $0.34 \pm 0.1\,mg\,O_2\,l^{-1}\,min^{-1}$ ($17.7 \pm 1.7\,mg\,O_2\,g\,SS\,h^{-1}$) and demonstrated an ethanol to oxygen consumption ratio of $1.7 \pm 0.1\,mgEtOH\,mg\,O_2^{-1}$ (Figure 4).

Mass transfer analysis

Under clean water conditions at an air flow rate of $7.5\,l\,min^{-1}$ (SAD_m $0.48\,m^3\,m^{-2}\,h^{-1}$), bubbles created by the integrated membrane module diffuser were principally developed in the coarse range (>2 mm), however, fine bubbles were observed to shear from coarse bubbles as has been described previously (Hodkinson *et al.* 1998). In the MBR, under static conditions, the global k_La was $0.2 \times 10^{-3}\,s^{-1}$ (k_La $0.5 \times 10^{-3}\,s^{-1}$ corrected for r_{O_2}) for a SAD_m of $0.58\,m^3\,m^{-2}\,h^{-1}$ (Q_a $7.2\,m^3\,m^{-3}\,h^{-1}$, MLSS $1.1\,g\,l^{-1}$; viscosity c. $1 \times 10^{-3}\,m\,Pa\,s^{-1}$). This value is broadly in agreement with other coarse bubble aeration studies (Chern & Yang 2003) and is below that observed by Germain *et al.* (2007) who recorded k_La values in the range of 1.4×10^{-3} to $6.9 \times 10^{-3}\,s^{-1}$ at a Q_a of $6.1\,m^3\,m^{-3}\,h^{-1}$ using a fine bubble ceramic diffuser in concentrated and viscous MBR sludges (7.2–30.2 g MLSS l^{-1}; 10.8–213 m Pa s^{-1}). In this study, K_La increased linearly to 1.9×10^{-3} s^{-1} corresponding to an SAD_m of $1.94\,m^3\,m^{-2}\,h^{-1}$ (Figure 5); as diffused airflow increases, bubble number increases enhancing interfacial area and therefore global mass transfer (Chern & Yang 2003). Oxygen transfer efficiency (OTE) was examined to quantify the impact of sparge length rather than specific diffuser efficiency; for sparge lengths 10, 20 and 60 seconds, OTE remained

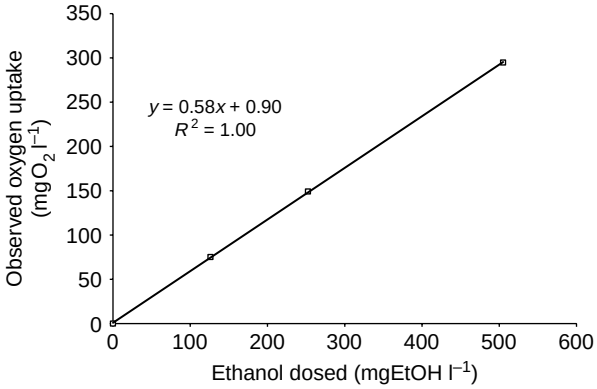

Figure 4 | Cumulative oxygen uptake observed during respirometric trials on 250 ml MBR sludge samples; ethanol dosed at 0, 125, 250 and 500 mg l^{-1}.

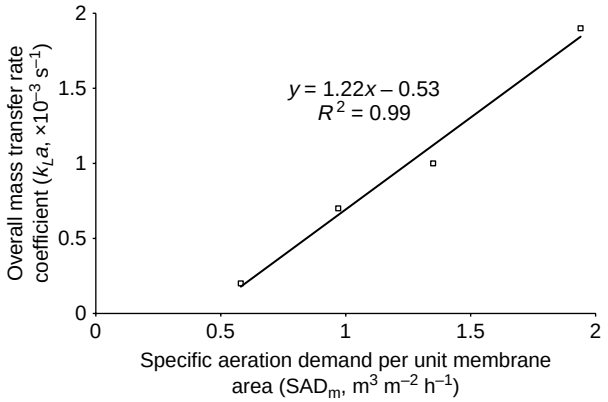

Figure 5 | Overall O_2 mass transfer coefficient (k_La, $\times 10^{-3}\,s^{-1}$) calculated as a function of specific aeration demand per unit membrane area (SAD_m 0.58–$1.94\,m^3\,m^{-2}\,h^{-1}$).

<0.85% (Figure 6) with a plateau reached corresponding to a SAD_m of c. $1.5\,m^3\,m^{-2}\,h^{-1}$ (Q_a $18.6\,m^3\,m^{-3}\,h^{-1}$). At a SAD_m of $1.94\,m^3\,m^{-2}\,h^{-1}$ (K_La $1.9 \times 10^{-3}\,s^{-1}$), a volumetric OTR of $4.8\,g\,m^{-3}\,h^{-1}$ was determined which is substantially below the range of c. 20 to $250\,g\,m^{-3}\,h^{-1}$ reported by Cornel *et al.* (2003) for full-scale MBR aeration; the higher OTR observed by the authors can be ascribed to the integration of both fine and coarse aerators to achieve sufficient residual O_2 for aerobic respiration in addition to the consistency in sparging. However, with increasing sparge length in this study, oxygen mass transfer efficiency also increased, implying sparge length should be limited to minimise oxygen residual.

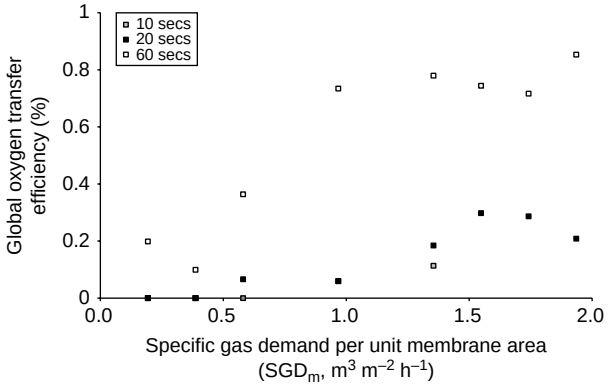

Figure 6 | Global oxygen mass transfer efficiency (%) measured to observe the influence of sparge length (cf. diffuser capacity) ranging 10 to 60 seconds versus specific air demand per unit membrane area (SAD_m 0.39 and $1.94\,m^3\,m^{-2}\,h^{-1}$) and sparge time (10–60 seconds).

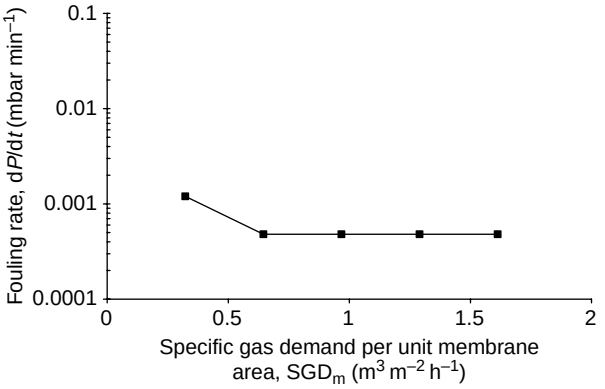

Figure 7 | Impact of SGD_m on fouling rate (dP/dt) over 24 hours for J 21 L m^{-2} h^{-1} and filtered volume (V_f) of 3.5 L m^{-2}. Backflush J set at × 2 forward J; 30 second gas sparge/backflush per cycle.

Fouling rate

Experiments were performed at a flux (J) of 21 l m^{-2} h^{-1} with an adopted filtered volume (V_f) of 3.5 l m^{-2} between gas sparging (approximates to c. 10 minutes filtration in the absence of gas). For a SAD_m of 0.32 m^3 m^{-2} h^{-1} (sparge length 30 seconds), fouling rate (dP/dt) was 0.001 m bar min^{-1} over 24 hours (Figure 7). Once SAD_m was increased to > 0.65 m^3 m^{-2} h^{-1} (sparge length 30 seconds), dP/dt was reduced to 0.00048 m bar min^{-1} and a hydrodynamic limit was reached; this observation has been made in other MBR studies (Le-Clech *et al.* 2005; McAdam *et al.* 2005). Under this intermittent gas sparging mode, long-term sustained permeabilities have been previously reported for anoxic immersed MBR sparged with nitrogen (McAdam & Judd 2008*a*,*b*). Accordingly, a SAD_m of 0.65 m^3 m^{-2} h^{-1} incorporated on a 30 second sparge/10 minute filtration cycle basis equates to a SAD_{mnet} of c. 0.062 m^3 m^{-2} h^{-1}. At the lowest SAD_m studied, gas sparge length was also examined (V_f set at 14 l m^{-2}). Increasing sparge length from 30 to 120 seconds decreased dP/dt from 0.00547 m bar min^{-1} to 0.00074 m bar min^{-1} (c. 7 times reduction).

DISCUSSION

Oxygen mass transfer

In standard MBR operation, coarse bubble membrane aeration is principally opted for as wider diameter bubbles induce higher velocities and thus exert higher shear (Judd 2006). In this study, the key finding was that almost no residual O_2 was evident for sparge times up to 60 seconds at the lowest SAD_m studied. This contrasts the observations of Lee *et al.* (2001) in which a residual O_2 concentration sufficient to impede anaerobic degradation was recorded after only 5 to 10 seconds. In their investigation, air was delivered through back-washing the immersed 0.5 μm membrane thus the initial bubble diameter (approximated using Tate's law, Equation (4)) could have been around 0.28 mm (Devatine & Mietton-Peuchot 2009).

$$d_i = \left[\frac{6d_o\sigma}{g(\rho_L - \rho_g)}\right]^{1/3} \quad (4)$$

Air backwashing therefore apparently supports micro bubble formation in the liquid phase resulting in higher mass transfer efficiency and shorter attainable sparge times. At an identical airflow rate, as bubble size halves, number density increases by c. 8 (Wicaksana *et al.* 2006) creating enhanced interfacial area and improved gas-liquid mass transfer. In contrast, as this current study demonstrates, extended sparge times (at higher air flows) can be achieved with low O_2 residuals by adoption of immersed coarse aeration. Hodkinson *et al.* (1998) observed that whilst OTR increased linearly with Q_a, poor O_2 transfer efficiency occurred with increasing Q_a due to the increase in bubble size reducing specific surface area and therefore oxygen transfer efficiency. However, low oxygen transfer efficiencies, as observed herein, have been previously reported for low water depths (0.70 m in this study) due to rate limiting liquid side mass transfer based either on insufficient turbulence at the gas-liquid interface (Deront *et al.* 1998) or low bubble residence time (Hodkinson *et al.* 1998). Using the median bubble size determined at a SAD_m of 0.48 m^3 m^{-2} h^{-1} (Figure 5), approximate bubble rise velocity (Equation (5)) was c. 0.05 m s^{-1} which provides a bubble residence time of c. 14 seconds (Wicaksana *et al.* 2006).

$$\bar{U} = \frac{2}{3}(gr_c)^{1/2} \quad (5)$$

Thus at full scale, where process depths of 3 m are practicable, extended bubble residence times of c. 60 seconds may be attained which will improve oxygen transfer efficiency over that reported in this study.

Implications for full-scale

Adoption of a low V_f affords the application of a sufficiently low SAD_m ($0.65\,m^3\,m^{-2}\,h^{-1}$ for 30 seconds) to maintain permeability and incur a limited dissolved O_2 residual ($0.05\,mg\,l^{-1}$); at full-scale, this residual may be higher due to improved oxygen transfer efficiency with increased fluid depth. The financial implications of an O_2 residual are significant: assuming an order of magnitude increase in O_2 residual to $0.5\,mg\,l^{-1}$ and no simultaneous electron acceptor reduction (Patureau *et al.* 1996), 120 seconds of recovery time will be required per sparge corresponding to c. 11.5 minutes h^{-1} without denitrification thus greater bioreactor capacity (19% capacity loss) may be needed; an additional ethanol cost of c. 1.5 €cents per m^3 treated will also be required for oxygen removal (cf. 5 €cents per m^3 for nitrate). However, such an increase in transfer efficiency is unlikely to transpire provided sufficiently coarse aeration is selected.

Based on the findings of this study, although oxygen mass transfer efficiency was observed to increase with sparge length, sustained permeability is possible by extending sparge length at lower SAD_m ($< 0.39\,m^3\,m^{-2}\,h^{-1}$). This is qualitatively similar to Kennedy *et al.* (1998) who identified that backflush effectiveness was more strongly correlated to time than pressure thus minimising mass transfer rate and O_2 residual; validation is required at full-scale process depths. Integration of an intermittent sparging protocol has the potential to deliver a substantial reduction in specific energy demand from $0.19\,kWh\,m^{-3}$ permeate for constant gas sparging to $0.007\,kWh\,m^{-3}$ for this intermittent strategy, constituting a net energy reduction of $0.183\,kWh\,m^{-3}$ treated (96.3%) regardless of the sparge gas introduced. This technique may be extended to anaerobic MBR particularly to promote controlled micro-aeration (Hartley & Lant 2006) for anaerobic process augmentation.

CONCLUSIONS

In this paper, the first application of aeration for membrane sparging in an anoxic MBR has been undertaken. The key criteria to sustain low dissolved oxygen concentrations under this protocol is to minimise oxygen mass transfer; immersed coarse bubble aeration is recommended as it provides both effective shear and poor oxygen mass transfer efficiency. When low SAD_m were implemented at 30 second sparge lengths during the 24 hour membrane fouling trials, stable permeability was observed. However, to reach the optimum hydrodynamic limit, a low residual dissolved oxygen concentration was recorded. Further minimisation of the oxygen residual can be achieved by adopting longer sparge lengths at lower SAD_m as this combination delivers more effective permeability maintenance and poorer overall mass transfer. Intermittent aeration provides standard MBR design to be adapted to anoxic conditions with the simultaneous advantage of an order of magnitude reduction in specific energy demand for membrane sparging.

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