Biomass properties and permeability in an immersed hollow fibre membrane bioreactor at high sludge concentrations

Z. Z. Wang, T. Zsirai, K. Connery, M. Fabiyi, A. Larrea, J. Li and S. J. Judd

ABSTRACT

This study aimed to investigate the influence of biomass properties and high mixed liquor suspended solids (MLSS) concentrations on membrane permeability in a pilot-scale hollow fibre membrane bioreactor treating domestic wastewater. Auxiliary molasses solution was added to maintain system operation at constant food-to-microorganisms ratio (F/M = 0.13). Various physicochemical and biological biomass parameters were measured throughout the trial, comprising pre-thickening, thickening and post-thickening periods with reference to the sludge concentration and with aerobic biotreatment continuing throughout. Correlations between dynamic changes in biomass characteristics and membrane permeability decline as well as permeability recovery were further assessed by statistical analyses. Results showed the MLSS concentration to exert the greatest influence on sustainable membrane permeability, with a weaker correlation with particle size distribution. The strong dependence of absolute recovered permeability on wet accumulated solids (WACS) concentration, or clogging propensity, revealed clogging to deleteriously affect membrane permeability decline and recovery (from mechanical declogging and chemical cleaning), with WACS levels increasing with increasing MLSS. Evidence from the study indicated clogging may permanently reduce membrane permeability post declogging and chemical cleaning, corroborating previously reported findings.

Key words | cleaning, clogging, hollow fibre membrane bioreactor, membrane permeability, MLSS concentration

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INTRODUCTION

Whilst immersed membrane bioreactor (iMBR) technology has become increasingly popular for treating municipal and industrial wastewater, its operation remains challenged by permeability decline from surface fouling and channel clogging. Generally, fouling constituents are divided into microbial floc gross solids, colloidal matter and solutes, each apparently playing a different role in membrane permeability decline. It is thought that 'permanent' membrane surface fouling and pore plugging results primarily from solutes and colloidal particles blocking the membrane pores (Chang *et al.* 2002; Drews 2010), whereas sludge solids can accumulate in the membrane channels to cause clogging (Buzatu *et al.* 2012). Both membrane fouling and clogging are manifested as permeability decline, incurring downtime for cleaning and so decreasing the net flux and increasing operational and maintenance costs.

The use of membranes for thickening (Reed *et al.* 1993) and combined thickening and digesting (Wang *et al.* 2008, 2009; Wu *et al.* 2009) of activated sludge has received little attention in the past, despite membrane thickening operations being employed at full scale at some MBR installations (Judd 2010). Whilst foulant concentrations are reported to decrease slightly with increasing sludge retention time, and thus with mixed liquor suspended solids (MLSS) concentration (Li *et al.* 2008; Domínguez *et al.* 2012), it is also recognised, and is intuitive, that excessive MLSS concentrations deleteriously affect flux (Wang *et al.* 2008).

The current study aims to quantify impacts of increased sludge concentration on the operation of a hollow fibre (HF) iMBR under normal and thickening operating conditions, with aerobic biotreatment sustained at all sludge

doi: 10.2166/wst.2014.142

concentrations. Parameters explored included those relating to both bulk sludge quality and classical surface fouling. Sludge quality was assessed according to its settleability, dewaterability and microbial ecology over a range of concentrations between 8 and 35 g/L MLSS. Statistical analysis was employed to elucidate relationships between membrane permeability and sludge characteristics under operating and maintenance conditions identical to those applied at full scale.

MATERIALS AND METHODS

Pilot MBR plant

The pilot-scale membrane bioreactor (Figure 1, total volume 6.75 m^3) comprised a biotank (74% of total volume) and a membrane tank (26%), both being open to the atmosphere. The biotank base was fitted with a fine bubble aerator with the air flow rate manually controlled to provide a dissolved oxygen (DO) concentration of 1–2 mg/L throughout. The two tanks were connected by a recirculation pump (operating at four times the permeate flow) and pipework. A peristaltic pump was used to pump diluted molasses into the biotank, aimed at maintaining a target food-to-microorganisms (F/M) ratio of 0.13 d^{-1} during operation. DO, suspended solids (SS) and temperature were all monitored using dedicated sensors installed in the biotank.

The membrane tank was fitted with two vertically oriented 0.04 μm-pore-size polyvinylidene difluoride HF membrane modules providing a total area of 46.4 m^2. A second peristaltic pump fitted to the permeate line provided both filtration and backflushing. The membrane surfaces were air-scoured intermittently (10 s on, 10 s off) using coarse-bubble aerators, placed 100 mm below the membrane element channels, operating at an overall specific aeration demand (SAD_m) of 0.25 $Nm^3/(m^2\,h)$. The membrane was backflushed for 30 s every 10 min at a flux of 15 $L/(m^2h)$ (LMH). This was supplemented by periodic chemically enhanced backflushing (CEB) as required.

A programmable logic controller and supervisory control and data acquisition (SCADA) system were employed for control of feeding, recirculation, permeation, membrane aeration and waste sludge discharge. A pressure sensor was fitted in the biotank for controlling the sludge level, with another fitted at the membrane module outlet. Membrane outlet pressure, and permeate and backflush flows, were recorded every 30 s by the outlet pressure transducer and an ultrasonic flowmeter connected to the SCADA system. The plant underwent three campaigns based on steady-state operation of aerobic biotreatment at high and low MLSS concentration, with an intervening thickening period (Table 1).

The plant was fed with settled municipal wastewater (tCOD = 483.2 ± 290.5 mg/L; sCOD = 117 ± 60.5 mg/L; NH_4^+-N = 28.9 ± 8.2 mg/L, TN = 44.3 ± 10.6 mg/L, TP = 8.2 ± 2.8 mg/L, SS = 230 ± 190 mg/L, volatile suspended solids (VSS) = 190 ± 160 mg/L, pH = 8.2 ± 1.1) from the primary settlement tank at Cranfield University Wastewater Treatment Works. This was supplemented with a solution of molasses (Algerian refined cane sugar, British sugar, 1,142 gCOD/L), the flow rate being adjusted so as to maintain the appropriate organic load.

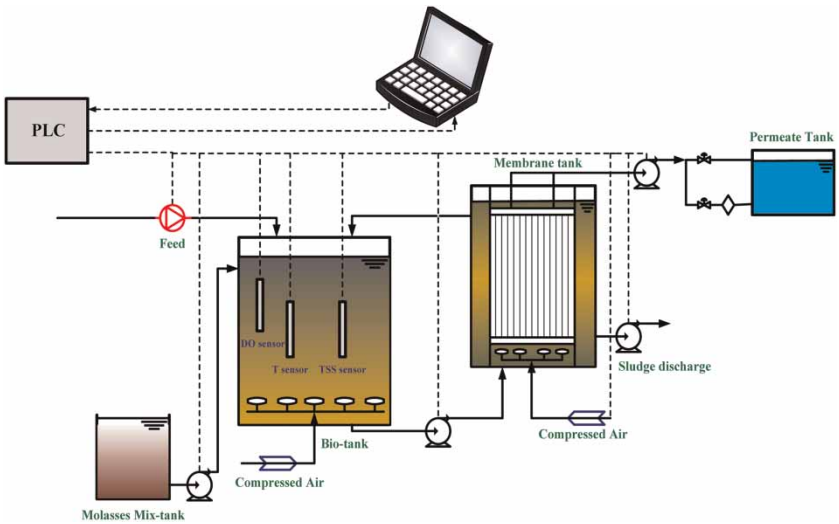

Figure 1 | Schematic diagram of the pilot MBR plant (PLC = programmable logic controller).

Table 1 | MBR operational conditions

		Operational periods		
Parameters	**Unit**	**Pre-thickening**	**Thickening**	**Post-thickening**
SRT	Days	38 ± 4	87–103	47 ± 1
HRT	Hours	7.9 ± 0.5	7.8–22.7	21.8 ± 0.9
VLR	kgCOD/(m^3·day)	1.18 ± 0.48	0.80–5.59	4.23 ± 0.43
F/M	g COD/(gMLSS·day)	0.13	0.13	0.13
Y_H	kgMLVSS/(kgCOD·day)	0.26 ± 0.15	0.03–0.18	0.13 ± 0.02
SAD_m	N m^3/m^2 h	0.25	0.25	0.25
Air flow rate	m^3/h	16	16–220	220
Flux	L/(m^2 h) or LMH	20.7 ± 1.8	22–6.2	6.9 ± 0.44
Net flux	L/(m^2 h) or LMH	17.6 ± 1.7	18.2–4.7	5.9 ± 0.44
Permeability	LMH/bar	367.4 ± 5.7	364.8–77.1	126 ± 31

Analytical methods

Analysis of influent sewage, molasses and permeation

The influent was monitored for total chemical oxygen demand (tCOD), soluble chemical oxygen demand (sCOD), total nitrogen (TN), ammonia nitrogen (NH_4^+-N), total phosphorus (TP), total suspended solids, VSS and turbidity. The measured effluent quality parameters included sCOD, TN, TP and turbidity, all in accordance with *Standard Methods* (APHA 1998). COD, TN, NH_4^+-N and TP were determined colorimetrically using a Merck Spectroquant Photometer (Merck, Germany).

Analysis of biomass properties

Sludge samples were collected from the biotank top and base to assess the mixing efficiency via the measured mixed liquor quality parameters MLSS, mixed liquor volatile suspended solids (MLVSS) and capillary suction time (CST), all according to *Standard Methods* (APHA 1998). Samples were taken three times a week. Sludge settleability was evaluated by measuring the diluted sludge volume index (DSVI, the SVI value of the sludge sample diluted to ~3 g/L). Particle size diameter (PSD) was determined using a Malvern Mastersizer instrument (Malvern 2000, Worcestershire, UK) with a measurement range of 0.1–1,000 μm. Soluble microbial product (SMP) and extracellular polymeric substances (EPS) were extracted according to the method of Morgan *et al.* (1990). Both were measured as total organic carbon (TOC, mg/L), EPS being normalized against MLSS concentration since it is associated with the solids. The pH was measured using a pH-conductivity meter (EC500, EXTECH, UK).

Supplementary weekly sludge-quality measurements were conducted to determine the microbial ecology, and specifically to appraise the filamentous bacteria population. A direct microbial observation method was used, recognised as being highly subjective and simplistic but providing a measure of filamentous matter according to an arbitrary index of 1–5 (Eikelboom *et al.* 1998).

Assessment of permeability recovery

Retained sludge solids were determined gravimetrically by difference using a bespoke 100 g-resolution load cell (Straightpoint Miniweighter, UK) fitted to the membrane cassette. The module was removed following each test and the residual liquid allowed to drain for 1 hour. Declogging was then implemented by washing with low-pressure water combined with gentle agitation. M_c, defined as the specific mass concentration of the wet accumulated clogged solids per unit membrane area (WACS, g/m^2), was determined gravimetrically following removal and draining of the module from the tank. CEB was performed with 500 mg/L sodium hypochlorite applied at 25 LMH based on 10 pulses of 30 s duration with 2 min relaxation between pulses.

ΔK, the ratio of accumulated solids to mixed liquor solids loading, was determined as a measure of membrane clogging propensity

$$\Delta K = \frac{M_c \cdot f}{J_{\text{net}} \cdot T \cdot \text{MLSS}} \times 100\% \quad (1)$$

where J_{net} is the net flux, T the filtration period, and f the weight ratio of dry to wet solids in the WACS, estimated as being 10% (Buzatu *et al.* 2012).

The absolute recovered permeability ΔL in LMH/bar from declogging followed by a CEB (declogging and CEB) is defined as

$$\Delta L = L_{clean} - L_{end} \quad (2)$$

where L_{clean} is the permeability immediately after cleaning and L_{end} the permeability at the end of the previous test recorded immediately before the clean.

Statistical analysis

Statistical analysis was used to characterize the significance of the biomass properties for membrane permeability. The relationships within biomass properties were also evaluated using univariate linear correlations. The Pearson correlation coefficient (r_p) was calculated to determine the strength and direction of (pseudo-)linear correlations between two parameters. Correlations were considered statistically significant at a 95% confidence interval.

RESULTS AND DISCUSSION

Foulant and bulk properties of the sludge

The evolution of MLSS concentration and membrane permeability (Figures 2(a) and 2(b)) revealed a dramatic decrease in sustainable permeability from around 18–20 g/L MLSS, falling from 370 to ~70 LMH/bar between 18 and ~32 g/L MLSS. The sustainable flux declined from 22 to 6.2 LMH over the same period. Negligible membrane permeability reduction and flux decline was observed between 8 and 18 g/L (pre-thickening), and between 32 and 35 g/L MLSS (post-thickening), the permeability and flux stabilizing at approximately 120 LMH/bar and 6.9 LMH respectively. The observed threshold value of 18 g/L would appear to be in reasonable agreement with the value of 15 g/L identified by Rosenberger *et al.* (2005) in their similar study of two parallel bench-scale HF iMBRs.

Permeate water quality during thickening deteriorated, such that post-thickening the COD had risen to a mean concentration of more than 250 mg/L compared with ~70 mg/L during pre-thickening. This was attributed to increased SMP levels (Table 2, Figure 2(c)) associated with floc breakage

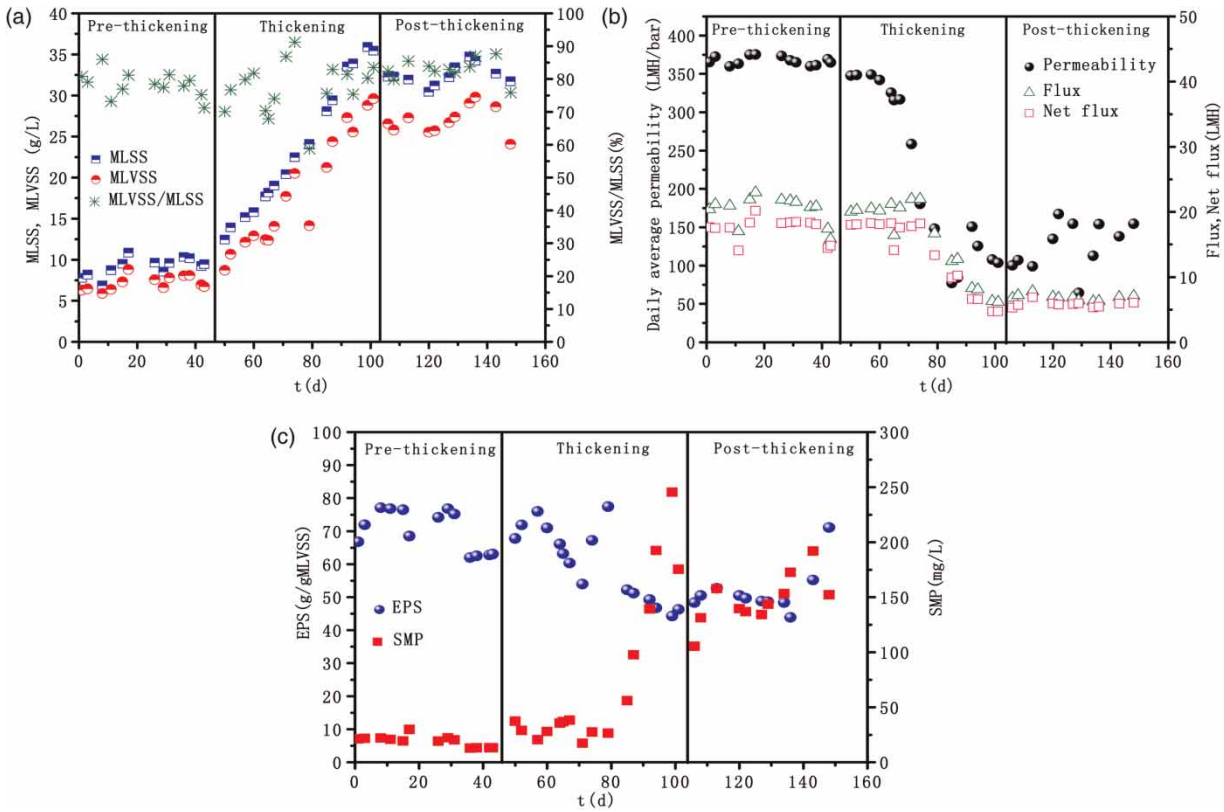

Figure 2 | Evolution of (a) membrane permeability, flux and net flux, (b) MLSS and MLVSS concentrations, (c) EPS and SMP concentrations.

Table 2 | Biomass properties during MBR operations

		Operational periods		
Sludge properties	**Unit**	**Pre-thickening**	**Thickening**	**Post-thickening**
MLSS	g/L	9.1 ± 1.2	9.4–35.9	32.5 ± 1.3
MLVSS	g/L	7.2 ± 0.9	6.7–29.6	27.0 ± 1.7
EPS_n	g/g MLVSS	70.9 ± 6.1	76–44.2	51.6 ± 7.1
SMP	mg/L	19.6 ± 4.8	13.1–245.4	147.1 ± 22.7
sCOD	mg/L	74.7 ± 19.6	57.5–458	462.1 ± 93
DSVI	mL/g	349.6 ± 63.8	302.5–462.5	418.0 ± 88.9
PSD	μm	93.4 ± 14.1	100.2–35.7	32.4 ± 3.3
CST	seconds	11.3 ± 2.8	10.6–191.1	123.9 ± 11.8

MLSS: Mixed liquor suspended solids concentration; MLVSS: Mixed liquor volatile suspended solids concentration; EPS_n: Extracellular polymeric substances concentration normalized against MLSS; SMP: Soluble microbial products concentration; sCOD: soluble chemical oxygen demand concentration; DSVI: Diluted sludge volume index; PSD: Particle size diameter; CST: Capillary suction time.

from the elevated aeration rate demanded by the higher MLSS concentrations (Lim & Bai 2003; Meng & Yang 2007), which then also impacted on the floc particle size. On the other hand, the effluent phosphorus levels were significantly lower (~1 mg/L cf. 4–5 during pre-thickening and thickening), indicating increased P uptake by the biomass at the high biomass solids levels. Ammonia removal was maintained at ~100% throughout.

Calculated Pearson coefficient values for permeability vs the various sludge quality parameters (Table 3) confirmed the strong negative linear correlation ($r_p = -0.958$) with MLSS concentration during thickening. Correlation with all parameters other than MLVSS was weaker, although coefficient values for SMP, sCOD, PSD and CST were all between 0.82 and 0.85. Correlation of permeability decline with recognized fouling parameters such as SMP and sCOD has been widely reported (Bae & Tak 2005; Sabia *et al.* 2013), and specifically when associated with floc breakage (Wang *et al.* 2009), whilst correlation with CST and PSD respectively relate directly to the increased sludge solids concentration (i.e. the MLSS, for which $r_p = -0.901$) and reduced bulk fouling layer permeability. The strong correlation ($r_p = 0.957$) between CST and SMP obtained in this current study reflects the fouling propensity of the latter.

Microscopic examination of the sludge for filamentous bacteria indicated a general increase in microbial diversity with time, with higher organisms becoming increasingly evident. Whilst filamentous bacteria apparently increased in concentration with time overall, yielding filamentous index (FI) values of 4 from values of 1–2 at the low initial MLSS concentrations, no excessive, uncontrollable foaming resulted (Bella & Torregrossa 2013). Thus, whilst providing an indication of the sludge quality, the FI appeared not to relate to sludge foaming propensity, nor to its filterability and/or MBR membrane permeability.

Table 3 | Pearson correlation coefficients, membrane permeability vs biomass properties during thickening

	Permeability	**MLSS**	**MLVSS**	**EPS_n**	**SMP**	**sCOD**	**DSVI**	**PSD**	**CST**
Permeability	1.000								
MLSS	−0.958	1.000							
MLVSS	−0.944	0.989	1.000						
EPS_n	0.782	−0.828	−0.848	1.000					
SMP	−0.822	0.900	0.906	−0.784	1.000				
sCOD	−0.850	0.899	0.897	−0.652	0.864	1.000			
DSVI	−0.469	0.494	0.457	−0.210	0.541	0.530	1.000		
PSD	0.831	−0.879	−0.869	0.657	−0.765	−0.879	−0.568	1.000	
CST	−0.830	0.901	0.911	−0.802	0.957	0.853	0.524	−0.770	1.000

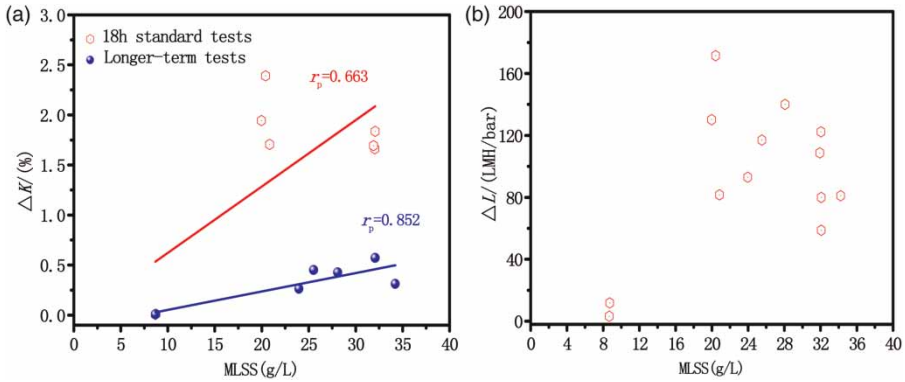

Figure 3 | (a) The ratio of accumulated solids and (b) absolute recovered permeability from declogging and chemically enhanced backflushing vs MLSS concentration.

Fouling and clogging

The WACS mass varied from 0.1 to 64 kg over the whole trial. Gravimetric determination of WACS revealed no significant clogging ($M_c < 2$ g/m^2) below an MLSS concentration of 18 g/L, whereas M_c reached approximately 1,080 g/m^2 at an MLSS concentration of 32 g/L. Solids accumulation varied with time, with the standard run-time of 18 h generating M_c levels of up to 650 g/m^2. For more prolonged periods of 7–10 days, levels varied between 1,030 and 1,380 g/m^2. Data generated from the 18h standard test runs indicated no correlation ($r_p = 0.663$) between ΔK and MLSS (Figure 3(a)), whereas for the longer-term test periods a stronger correlation, but nonetheless weak, was evident ($r_p = 0.852$). The absolute permeability recovery achieved by declogging followed by a CEB varied from 70 to 170 LMH/bar between 20 and 35 g/L (Figure 3(b)).

No other significant relationship between M_c and any of the measured parameters was evident, other than with particle size and absolute permeability recovery. In both cases the correlation was weak ($r_p = 0.8$–0.82), but nonetheless indicative of possible impacts of particle size on clogging.

The application of a CEB alone was found to achieve a permeability recovery close to that of the virgin membrane when the membrane was operated at low MLSS concentration (~8 g/L), with no incipient clogging evident. Under these conditions, absolute permeability recovery was negligible since little or no loss of permeability took place during operation: backflushing alone was found to adequately sustain operation in the absence of clogging. However, operation during and post clogging was significantly compromised. Permeability recovery achieved from applying a CEB to a clogged membrane was found to be ephemeral, with membrane permeability decreasing to the pre-cleaned value within 5 hours (Figure 4). On the other hand, the recovered

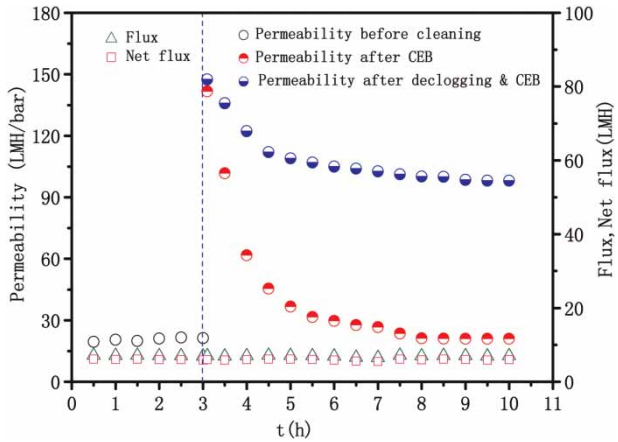

Figure 4 | Typical permeability evolution after cleaning at high MLSS concentration (28–32 g/L).

permeability from declogging followed by a CEB was sustained for up to 100 hours, provided subsequent operation continued without clogging. At the highest MLSS concentrations (32–35 g/L), this implied an operational flux of 5–6 LMH. Results corroborate previously reported findings (Buzatu *et al.* 2012) of permanently compromised permeability of HF membranes post clogging.

CONCLUSIONS

A study of the impact of high sludge concentrations on the operation of an aerobic/sludge thickening immersed HF membrane bioreactor has revealed that:

- MLSS concentration exhibits a statistically more significant impact on membrane permeability than any other sludge parameter, with a dramatic decrease in permeability taking place at MLSS concentrations above 18 g/L;

- the efficacy of chemical cleaning – instigated as a chemically enhanced backflush (CEB) – on permeability recovery was strongly dependent on the level of membrane clogging, the propensity for which depended on MLSS concentration;
- flux and permeability levels decreased by a factor of 3–3.5 between 18 and 32 g/L MLSS;
- declogging combined with a CEB, demanded at the higher MLSS concentrations, successfully recovered sufficient membrane permeability to allow continued operation, but only at very low fluxes of 5–6 L/(m^2h) and at accompanying low permeabilities;
- evidence from the study suggests clogging may permanently reduce membrane permeability, notwithstanding mechanical declogging and chemical cleaning, supporting previous findings.

ACKNOWLEDGEMENTS

The authors are grateful to Praxair International, which funded the work. The financial support of the Chinese Scholarship Council (201206540011) is also gratefully acknowledged.

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First received 21 November 2013; accepted in revised form 4 March 2014. Available online 28 March 2014