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Pilot-scale Submerged Hollow Fibre Membrane Bioreactor Thickening Operation: Membrane Clogging, Sustainable Flux and Sludge Characteristics Assessment

Zhaozhao Wang*, Kai Zhang*, Lina Yan, Fengbing Tang*, Wei Zhang* and Simin Li *†**
*College of Energy and Environmental Engineering, Hebei University of Engineering, Handan, 056038, China
**Graduate School, Hebei University of Engineering, Handan, 056038, China
†Corresponding author: Simin Li

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ABSTRACT

The sludge characteristics and their impact on membrane clogging propensity and flux behaviours by statistical analyses were investigated in a pilot-scale hollow fiber membrane bioreactor during thickening operation at constant F/M ratio of 0.13. The cleaning efficiencies by chemical enhanced backflushing (CEB) and mechanical declogging at different mixed liquor suspended solids (MLSS) concentration and flux levels were further assessed through (post-cleaning) permeability decline rates. The results showed that MLSS concentration exerted the greatest impacts on the ratio of accumulated solids to mixed liquor solids loading (ΔK) and sustainable flux (J_{SUS}), and its measurement could be used to predict both ΔK and J_{SUS}, with a strong correlation between ΔK and J_{SUS} observed. Moreover, membrane permeability recovery exhibited strong dependence on MLSS concentration and operational flux. On the premise of sub-critical flux operation, the recovered permeability can be sustained with a standard chemical enhanced backflushing (CEB) or declogging combined CEB at low (~8 g/L) or high (20-32 g/L) MLSS concentrations. However, the recovered permeability could only be obtained instantaneously regardless of cleaning modes when operation is exposed to super-critical flux condition, indicating the significance of the operational flux on membrane permeability sustainability.

INTRODUCTION

Membrane bioreactor processes have been increasingly popular to be employed treating municipal and industrial wastewater for water reuse or reclamation owing to its excellent effluent quality (Judd 2016). Several advantages of this technology over conventional activated sludge (CAS) have been verified through decades' applications (i.e. allowing higher MLSS concentration, smaller footprint, lower sludge production, decoupling HRT and SRT, physical disinfection, etc.) (Santos et al. 2011, Judd 2016). Nevertheless, membrane surface fouling is still the major drawback for MBR processes (Chang et al. 2002, Le-Clech et al. 2006, Meng et al. 2017). Generally sludge bulking is the mixed liquor of three fractions: microbial floc, colloids and solutes. Each fraction plays a different role in membrane permeability decline. The liquid fraction is separated from the sludge bulk by the suction drag force, which induced the solutes and colloidal particles to block the membrane pores, followed by sludge flocs depositing on the membrane surface.

Whilst, membrane clogging, referred to as 'sludging' (filling of membrane channels with gross solids) or 'ragging' (aggregation of solids at the channel inlet and outlet), is identified as a more serious obstacle limiting the wider applications of MBRs (Buzatu et al. 2012, Gabarrón et al. 2014). Both of the membrane fouling and clogging behaviours lead to the permeability reduction, and thus result in the increasing operational and maintenance costs. Over past decades, many researchers have extensively investigated the effects of the mixed liquor properties on membrane surface fouling behaviour through the assessments of correlations between biomass properties with membrane fouling rates (Li et al. 2008, Sabia et al. 2013). However, there is still a lack of comprehensive knowledge on the assessment of sludge characteristics, sustainable flux and membrane clogging propensity in a thickening membrane bioreactor at the constant F/M ratio.

Scope of this research aims to: (a) elucidate the interrelationships between sludge properties and sustaninable flux as well as membrane clogging propensity during the thickening process; (b) assess the relative efficacy of different cleaning modes with reference to permeability recovery under different sludge concentrations and operational fluxes conditions.

Statistical analyses were employed to process the experimental data and reveal the relationships within the research results obtained from the pilot-scale hollow fibre MBR under O&M conditions identical to those applies at full-scale.

MATERIALS AND METHODS

Reactor set-up and operation: The pilot-scale membrane bioreactor installation (total volume 6.75 m^3), as illustrated schematically in Fig. 1, comprised of a biotank (74%) and a membrane tank (26%). The biotank base was fitted with a fine bubble distributor to provide compressed air with the dissolved oxygen (DO) value manually controlled between 1-2 mg/L. The biotank and membrane tank were connected by a recirculation pump and tubing. Two other pumps were cascade-connected to pump the diluted molasses into the biotank. The DO data were monitored online through a DO sensor equipped in the biotank. Furthermore, total suspended solids (TSS) and a temperature sensor were also installed in the biotank to provide MLSS concentration and temperature values during the operational periods.

The membrane tank was fitted with two vertically-oriented HF membrane elements (membrane material: PVDF; pore size: 0.04 µm; membrane area: 46.4 m^2), which were operated via a dedicated peristaltic pump offering both filtration and backflush fluxes. The membrane surfaces were soured intermittently (10s-on/10s-off) by using coarse-bubble aerators placed 100 mm below the element channels. The membrane was maintained through backflushing for 30 s in 10 min filtration period. Besides, declogging followed by chemically enhanced backflushing (declogging & CEB) was performed to maintain the membrane permeability.

A programmable logic controller (PLC) equipped with supervisory control and data acquisition (SCADA) system were employed to control the whole loops of the plant running: feeding, recirculation, permeate, membrane aeration and waste sludge discharge. A pressure sensor was fitted in the biotank for the sludge level control. The transmembrane pressure (TMP), permeate and backflush flows were recorded every 30 s by the immersed pressure transducer and ultrasonic flowmeter connected to the SCADA system. The pilot-MBR plant operational parameters and periods are summarized in Table 1.

The MBR plant was fed with the sewage (tCOD, 487.2±190.1 mg/L; sCOD, 120±59.1 mg/L; NH_4^+-N, 30.1±7.5 mg/L, TN, 45.3±11.6 mg/L, TP, 6.2±2.5 mg/L, SS, 220.5±80.6 mg/L, pH, 7.1-8.2) from the primary settlement tank at a wastewater treatment plant. The molasses (1142 gCOD/L) was diluted by tap water and pumped into the biotank.

Chemicals and sludge quality analysis: Feed and permeate water quality parameters including chemical oxygen demand (COD), total nitrogen (TN), ammonia nitrogen (NH_4^+-N), total phosphorus (TP), total suspended solids (TSS), volatile suspended solids (VSS) and turbidity were measured in all in accordance with the standard methods (APHA 1998). Besides, sludge samples were collected from the biotank top and base to assess the mixing efficiency and mixed liquor quality parameters were monitored three times a week with reference to MLSS, MLVSS, capillary suction time (CST), all according to the same standard methods. Sludge settleability and flocculability were evaluated by measuring the diluted sludge volume index (DSVI, the SVI value of the sludge sample diluted to 3 g/L). The particle size diameter (PSD) was also determined using a Malvern Mastersizer instrument (Malvern 2000, Worcestershire, UK) with the measurement range of 0.1-1000 µm. Extracellular polymeric substances (EPS) and soluble microbial productions (SMP) were extracted according to the heating methods (Morgan et al. 1990), evaluated as total organic carbon (TOC, mg/L). pH was measured by the pH-conductivity meter (EC500, EXTECH).

Flux-step trails: The short-term flux-step trial comprised four one-hour flux steps incorporating six backflush steps whose values were adjusted according to the sludge concentration. The one-hour flux step tests were followed by four extended (18-hour) overnight flux steps (Zsirai et al. 2012, Zsirai et al. 2013). All tests were performed under standard conditions of intermittent aeration and with an

Table 1: MBR thickening and steady operational conditions

Parameters	Unit	Operational periods		
		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

intermediate CEB (Zsirai et al. 2012, Zsirai et al. 2013). The TMP and flow response was continuously monitored at a rate of two readings per minute. Flux was corrected from operational (instantaneous) to net flux from a consideration of backflushing and for temperature using the standard viscosity correction. The sustainable flux was identified from the critical flux values through flux-step trials during the thickening process.

Cleaning and clogging assessment: Retained sludge solids were determined gravimetrically by difference using a bespoke 100 g-resolution load cell fitted to the membrane cassette. The module was removed following each test and the residual liquid allowed to drain for one hour. Declogging was then implemented by washing with low-pressure water combined with gentle agitation. M_w, defined as the specific mass concentration of the wet accumulated clogged solids per unit membrane area (g/m^2), was determined gravimetrically following removal and draining of the module from the tank. Chemically-enhanced backflushing (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 (clogged) solids rate (M_c) to mixed liquor solids loading rate (M_L), was determined as a measure of membrane clogging propensity:

$$M_c = \frac{M_w \cdot f}{t \cdot A_f} \qquad ...(1)$$

$$M_L = \text{MLSS} \cdot J_{net} \qquad ...(2)$$

$$\Delta K = \frac{M_C}{M_L} \times 100\ \% \qquad ...(3)$$

Where, t is the filtration period, A_f the filtration area, J_{net} is the net flux, and f the weight ratio of dry to wet solids in the wet accumulated clogged solids, estimated as being 10% (Buzatu et al. 2012).

Statistical analysis: Statistical analysis was used to characterize the significance of the sludge characteristics and sustainable flux on membrane clogging propensity. The relationships within sludge characteristics and sustainable flux 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. In addition, the coefficient of partial correlation (r_p') and regression analyses were also performed to determine the critical factors affecting the sustainable flux as well as clogging propensity and to obtain mathematic representation of the relationships. All statistical analyses were carried out using SPSS 20.0 software.

RESULTS AND DISCUSSION

Process overall performance and sludge characteristics: The HF-MBR system was operated at the constant F/M ratio by increasing the influent organic loading rate from 1.18 kg/(m^3·d) to 4.23 kg/(m^3·d), with the COD removal efficiency remaining above 93%. Moreover, TN and TP removal efficiencies increased from 32%, 40.6% to 78.9% and 90% respectively resulted from the increased MLSS concentration for microbial demands. The complete nitrification could be achieved as the DO concentration in the biotank maintained at 1-2 mg/L, with NH_4^+-N removal efficiency kept above 99.5%. The turbidity in the effluent was kept below 0.5 NTU, verifying the membrane integrity throughout the whole thickening process.

The sludge characteristic matrix at different MLSS concentration levels during the thickening process is given in

Table 2: Sludge characteristics during thickening operations.

MLSS (g/L)	MLVSS (g/L)	EPS_n (g/g MLVSS)	SMP (mg/L)	sCOD (mg/L)	DSVI (mL/g)	PSD (µm)	CST (seconds)
6.8	5.9	77.1	22.1	120.5	405.0	81.9	12.1
8.7	6.4	76.8	20.7	81.0	400.0	96.8	12.8
10.3	8.0	62.0	12.8	73.5	287.5	105.9	9.3
12.4	8.7	67.8	37.4	201.0	337.5	53.2	16.4
13.9	10.7	71.9	28.9	180.0	350.0	53.0	20.9
15.8	12.9	71.0	28.0	230.0	375.0	45.2	16.4
18.2	12.3	63.2	36.9	203.0	350.0	69.2	15.0
20.4	17.7	54.0	17.3	132.5	319.0	61.3	17.6
22.5	20.5	67.2	27.4	210.0	320.0	54.3	17.8
25.5	20.1	59.7	66.25	254.0	450.0	31.4	51.6
28.1	21.2	69.1	55.94	276.0	405.0	43.6	38.6
32.1	28.9	62.0	125	356.0	425.0	43.5	83.1

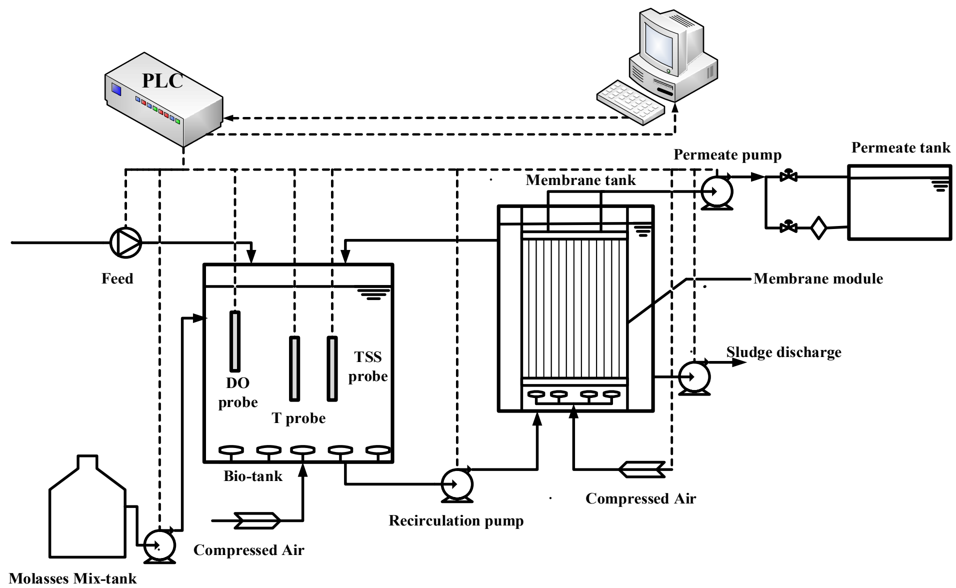

Fig.1: Schematic diagram of the pilot MBR plant.

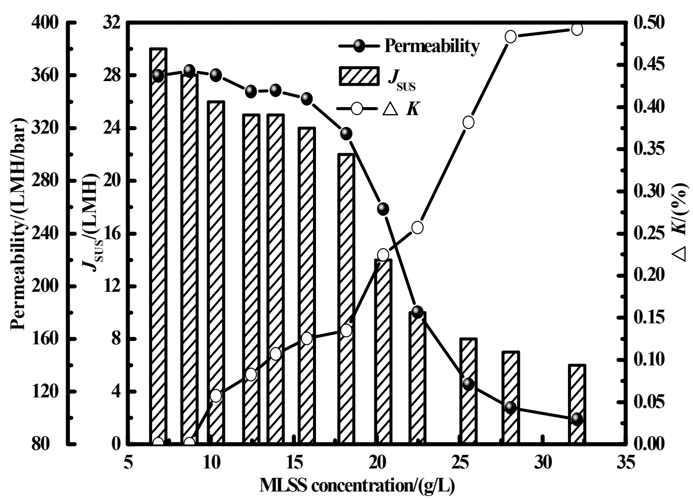

Fig. 2: Evolution of membrane perability, J_{SUS}, and ΔK during thickening operation.

Table 2. The SMP and sCOD concentrations showed obvious increases (SMP from 22.1 to 125 mg/L, sCOD from 73.5 to 356 mg/L) due to the increased microbial metabolic products with the increasing organic loading rate (Domínguez et al. 2012). However, the EPS specific concentration exhibited a decreasing trend (from 77.1 down to 62.0 mg/g). Wang et al. (2008) also found reverse changing trends of SMP and EPS contents when the MBR plant was operated for simultaneous thickening and digestion of waste activated sludge. The PSD also decreased (D_{50} from 81.9 to 43.5 μm) resulting from the high shear stress induced by the increased aeration intensity (from 16 to 220 m^3/h) to keep the DO concentration at 1~2 mg/L through the whole operational periods. Moreover, the sludge settleability presented little variations with the fluctuations of DSVI values, whereas the sludge filterability tend to deteriorate from the CST measurements.

Permeability, sustainable flux and clogging trends: The variations of membrane permeability, sustainable flux (J_{SUS}) and ΔK through the thickening operation are shown in Fig. 2. The membrane permeability and sustainable flux decreased from 359.6 L/(m^2·h·bar) and 30 L/(m^2·h) to 98.87 L/(m^2·h·bar)

Table 3: Pearson correlation coefficients matrix, ΔK, J_{SUS} vs. sludge properties.

	ΔK	J_{SUS}	MLSS	MLVSS	EPS_n	SMP	sCOD	DSVI	PSD	CST
ΔK	1.000									
J_{SUS}	-0.963	1.000								
MLSS	0.977	-0.966	1.000							
MLVSS	0.951	-0.960	0.979	1.000						
EPS_n	-0.478	0.566	-0.547	-0.537	1.000					
SMP	0.784	-0.673	0.776	0.776	-0.261	1.000				
sCOD	0.837	-0.744	0.856	0.826	-0.234	0.864	1.000			
PSD	-0.714	0.681	-0.734	-0.696	0.265	-0.577	-0.851	1.000		
DSVI	0.443	-0.325	0.368	0.336	0.239	0.625	0.514	-0.434	1.000	
CST	0.841	-0.750	0.818	0.830	-0.334	0.974	0.826	-0.599	0.643	1.000

Table 4: Partial correlation analyses between sludge characteristics and ΔK, J_{SUS}.

Sludge characteristics	Partial correlation (ΔK)		Partial correlation (J_{SUS})	
	r_p'	p'	r_p'	p'
MLSS	0.977	0.000	-0.966	0.000
MLVSS	-0.143	0.675	-0.269	0.423
EPS_n	0.315	0.345	0.606	0.175
SMP	0.190	0.575	0.470	0.145
sCOD	0.002	0.996	0.623	0.080
PSD	0.023	0.947	-0.158	0.643
DSVI	0.425	0.192	0.125	0.714
CST	0.341	0.305	0.276	0.411

and 6 L/(m^2·h) respectively. Nevertheless, ΔK increased from 0.0005% to 0.49%, indicating the aggravated membrane clogging propensity with the increasing MLSS concentrations. In addition, it can also be seen from Fig. 2 that clogging behaviour appeared markedly at the MLSS concentration of 18 g/L where sharp decreases of membrane permeability and sustainable flux occurred.

The Pearson correlations of the membrane clogging propensity (ΔK), sustainable flux (J_{SUS}), and sludge properties are given in Table 3. ΔK showed a strong negative linear correlation with J_{SUS} (r_p= -0.963, $p<0.05$) and a positive linear correlation with MLSS concentration (r_p= 0.977, $p<0.05$), respectively. Furthermore, the MLSS concentration exerted the greatest effect on J_{SUS} and exhibited a strong negative linear correlation (r_p= -0.963, $p<0.05$).

CST showed a positive linear correlation with ΔK (r_p= 0.841, $p<0.05$), revealing that the worsened sludge filterability would lead to the occurrence of membrane clogging behaviour. Moreover, CST presented a negative linear correlation with J_{SUS} (r_p= -0.750, $p<0.05$), inconsistent with Sabia' (2013) research results reporting a non-liner correlation within the two parameters. In addition, CST exhibited a strong positive linear correlation with SMP concentration (r_p= 0.974, $p<0.05$), suggesting a close relationship with membrane fouling behaviour.

MLSS concentration was identified as the major sludge property affecting both ΔK and J_{SUS} by using the partial correlation analyses (Table 4) and it could be used as the predictors of ΔK and J_{SUS}, with the fitting formulas as the following equations (4)-(5):

$$\Delta K = 0.021\text{MLSS} - 0.185 \quad ...(4)$$

$$J_{SUS} = 38.211 - 1.09\text{MLSS} \quad ...(5)$$

Clogging propensity and permeability recovery: The 18-h permeability evolution trends at different MLSS concentrations and cleaning modes are showed in Fig. 3. The slight differences between declogging and CEB were found to recover the membrane permeability at low MLSS concentration (-8 g/L) and sustainable flux conditions (Fig. 3-(1)). However, the membrane permeability decline rate under sustainable flux condition was faster than that of supercritical flux conditions.

The membrane permeability decline rate increased after declogging and CEB and CEB implemented with the increased MLSS concentrations (20-32 g/L). Moreover, the membrane permeability decline rate after declogging and CEB was smaller than that of CEB, which was more apparent under sustainable flux conditions (Fig. 3-(3), Fig. 3-(5)), indicating that the sustainable permeability recovery could be achieved when declogging and CEB was demonstrated at medium and high MLSS concentrations. Similarly, it was

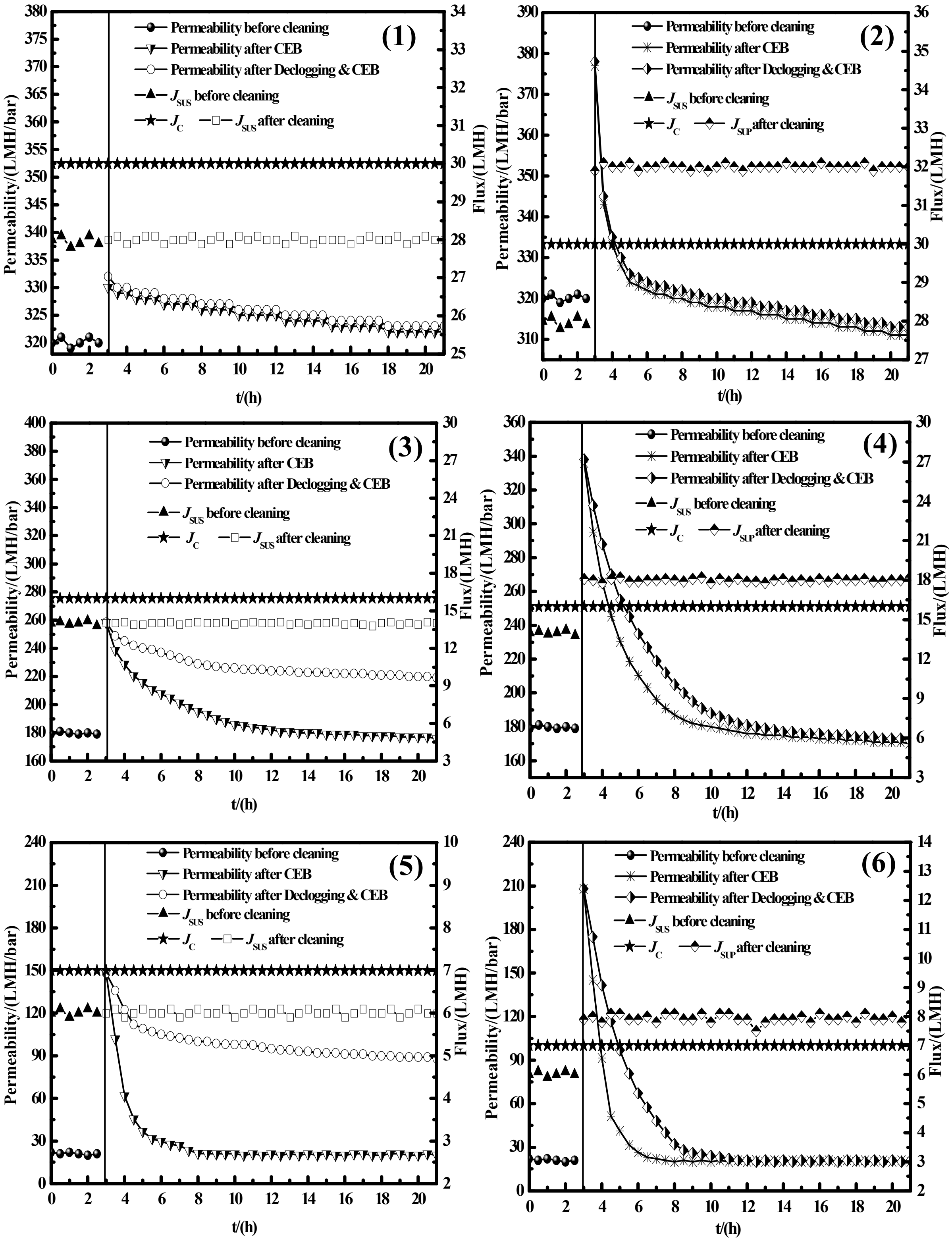

Fig. 3: Permeability evolution after cleaning at (1)-(2) low MLSS concentration (~8 g/L); (3)-(4) medium MLSS concentration (~20 g/L); (5)-(6) high MLSS concentration (~32 g/L).

Table 5: Clogging behaviour parameters (post-declogging & CEB) within 18 h standard testing.

J_{net} (LMH)	MLSS (g/L)	M_L g/(m²·h)	M_C g/(m²·h)	ΔK %
28(J_{SUS})	8	224	0.119	0.005
32(J_{SUP})	8	256	0.242	0.009
14(J_{SUS})	20	280	0.396	0.141
18(J_{SUP})	20	360	0.571	0.159
6(J_{SUS})	32	192	0.644	0.336
8(J_{SUP})	32	256	0.900	0.352

observed that the recovered instantaneous membrane permeability increased after membrane cleaning under the supercritical flux and corresponding MLSS concentration conditions, whereas the membrane permeability decline rate increased significantly as well, implying the unsustain-ability of membrane permeability recovery which was obvious in high MLSS concentration conditions (Figs. 3-(5, 6)). Therefore, the sustainable flux was of critical importance for the sustainability of membrane permeability recovery after membrane cleaning.

In addition, the membrane clogging parameters were investigated to elucidate the membrane permeability decline characteristics after declogging and CEB implementation (seen from Table 5). The lower membrane clogging propensity (ΔK) was observed at low MLSS concentration (-8 g/L) and high operational fluxes (J_{SUS} of 28 LMH, J_{SUP} of 32 LMH). However, higher membrane clogging propensity were obtained at medium/high MLSS concentration (20-32 g/L) and low operational fluxes (6-18 LMH). Moreover, the clogging propensity was more serious at supercritical flux condition than that of sustainable flux condition, which could interpret the membrane decline rate characteristics after cleaning at corresponding operational conditions. Therefore, membrane clogging propensity was confirmed as the key factor for membrane permeability decline after membrane cleaning.

CONCLUSIONS

The assessment of membrane clogging, sustainable flux and sludge characteristics in a thickening immersed hollow fibre membrane bioreactor have revealed that MLSS concentration exhibited the strongest correlations with membrane clogging propensity and sustainable flux. The prediction models were further established through SPSS analysis (ΔK=0.021MLSS-0.185, J_{SUS}=38.211-1.09MLSS). The membrane permeability recovery was related directly to cleaning modes and operational flux, suggesting sustainable flux operation at high MLSS concentration to alleviate membrane clogging propensity.

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