Demonstration of suburban sewage pre-concentration by enhanced membrane coagulation reactor (E-MCR) for organic matter recovery: a pilot study

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Abstract:
Aiming at semi-decentralized suburban sewage resource recovery, pilot-scale Enhanced membrane coagulation reactor (E-MCR) was set up for the first time for preliminary demonstration of sewage pre-concentration. During around 2400h continuous operation with different working scenarios, a relative stable average permeability of 30-50 L/m^2h·bar was achieved through the combination of enhanced coagulation process and air backflushing for raw sewage severe fouling control. The average COD concentration of the concentrate with an SRT of 2.5d could reach around 18000mg/L, which is close to blackwater and benefit for anaerobic digestion for energy recovery. By energy balance estimation, the net energy production of E-MCR system was anticipated as 0.0557 kWh/m^3 at optimal conditions. The application of E-MCR for semi-decentralized sewage resource recovery was demonstrated promising.

Keywords:
Enhanced membrane coagulation reactor (E-MCR); Pilot study; semi-decentralized sewage pre-concentration;

INTRODUCTION

The potential to pre-concentrate sewage organics and nutrients and economically recover valuable resources coupled with anaerobic digestion (AD) has been recognized(McCarty *et al.*, 2011; Verstraete *et al.*, 2009). Consequently, the significance of sustainable sewage pre-concentration processes has been extensively emphasized than ever before, not only because of its key role in minimizing energy requirement for mainstream wastewater treatment, but also in maximizing the comprehensive resource recovery from organics and nutrients in wastewater in terms of methane and digestate etc.(Shoener *et al.*, 2014; van Lier, 2008) In achieving sustainable pre-concentration, efforts have been accentuated on the improvement of concentration efficiency, the mitigation of the subsequent side effect, the maximization of preserved recovery and the minimization of process consumption(Verstraete and Vlaeminck, 2011).

As such, an effective physical concentration process, e.g. membrane process, would surpass chemical ones on matter preservation and process consumption(Odegaard, 1998; Diamantis *et al.*, 2013), and biological ones on concentration efficiency and recovery rate(Hernández Leal *et al.*, 2010; Meerburg *et al.*, 2015). Microfiltration (MF) have been preferred as a result of its low process consumption. Direct membrane pre-concentration can suffer great severe fouling problem, alleviation of which by enhanced chemically cleaning would be carried out at the expense of more chemicals consumption and more organics loss in terms of mineralization(Lateef *et al.*, 2013). The combination of bioflocculation and membrane was accordingly set up as high-loaded membrane bioreactor (MBR) to prevent the chemicals addition, whereas the sewage organics recovery and membrane fouling control along with the concentration factor are still not satisfactory, on account of investable biological mineralization and biofouling matters excretion(Akanyeti *et al.*, 2010; Faust *et al.*, 2014). Dynamic membrane was also applied to limit biological side effect, but the permeate quality and preserved organics recovery would be unfavorably influenced(Gong *et al.*, 2014). Consequently, moderate coagulation and membrane was brought together aiming at simultaneously limiting organics mineralization and severe fouling impact, with enhanced organics recovery and fouling control by coupling air backflushing (AB)(Jin *et al.*, 2016; Jin *et al.*, 2015). Yet none of above processes have managed to achieve long-term and pilot-scale operation, to theoretically and practically address the fouling problem in an optimized way which can be regarded as a sustainable sewage pre-concentration, and to evaluate the performance of the concentrate in AD. Accordingly, the feasibility of a sustainable sewage pre-concentration process demands demonstration and illustration.

In this study, a pilot-scale enhanced membrane coagulation reactor (E-MCR) is therefore proposed to demonstrate the feasibility of one sustainable sewage pre-concentration option. Ultrafiltration (UF) other than MF in previous studies is applied to explore whether the pre-concentration can be more efficient and attain permeate with better water quality. The detailed roles of enhanced coagulation process (ECP), UF membrane and fouling control strategy in achieving sustainable sewage pre-concentration are discriminated respectively. The main fouling causes and their roles in pre-concentration are also analyzed. With further evaluation of the performance of the long-term concentrate in AD, a sound understanding and a practical vision of sustainable sewage pre-concentration can be provided.

MATERIAL AND METHODS

Raw sewage

The raw sewage for pilot-scale test was picked from a 20,000 t/d Xiaojiahe municipal wastewater treatment plant (after the screening unit) in Beijing suburbs, P.R China. Sewage from surrounding communities is collected in this plant without intrusion of industrial wastewater. Therefore, less toxicity of the sewage would lead to the pilot-scale concentrate more practical for resource recovery.

System configuration and operation

The configuration and schematic diagram of the pilot-scale UF E-MCR system are presented in Figure 1. A set of submerged commercial polyvinyl chloride (PVC) reinforced hollow fibre UF membrane (Litree Co. Ltd., China) with an effective surface area of 24 m^2 and a nominal average pore-size of 0.02μm was applied. The inner and outer diameters of the hollow-fibre were 1.00 mm and 2.00 mm. A Perspex tank with a working volume of 280 L was set as the concentration vessel. For introduction of ECP, polyaluminium chloride (PACl) as the optimal coagulant in previous study(Wang *et al.*, 2015), and powdered activated carbon (PAC) were dosed into sewage simultaneously in connection tubes by peristaltic pumps (Longer, USA). The concentrations of PACl and PAC in the reagent containers were both 10 g/L with continuous stirring in PAC tank to prevent settlement. The flow ratios of the influent pump to the dosing pumps determined the dosage of PACl and PAC. A piston pump (Seko, Italy) was applied to intermittently draw permeate from the UF modules. A combination of a pressure sensor, a metal tube rotameter and a paperless recorder was set up to monitor and record the flow rate and transmembrane pressure (TMP) data. Intermittent air backflushing (AB) was implemented according to previous study(Jin *et al.*, 2015) and optimized to prevent severe cake-layer-induced membrane fouling and to minimize organic matter mineralization. In order to minimize the impact of the compressed air inside membrane on water filtration, an air ejector (Jingwei, China) was connected to the air tube and released the air at the end of backflushing period. All solenoid valves (Changjiang, China) and the ejector was controlled by a time delay relay (Symore, China) to keep the ordered intermittent filtration and AB operation.

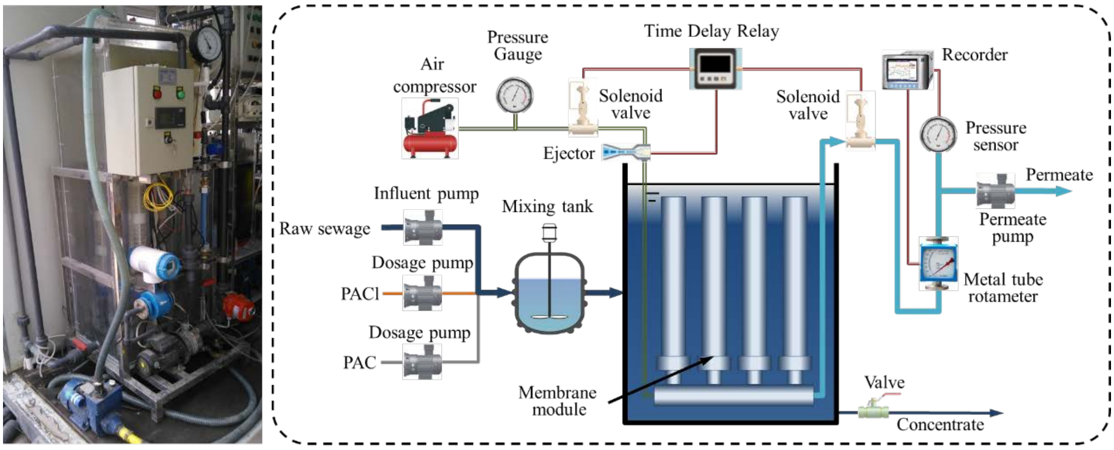

Figure 1 Schematic diagram of pilot-scale UF E-MCR for long-term continuous sewage pre-concentration

Working scenarios

The details of key parameters in different working scenarios are shown in Table 1. Direct sewage pre-concentration and common UF with coagulation were included in scenarios I and II for comparison to clarify the role of ECP for pilot-scale long-term UF operation. Normal intermittent aeration (IA) was also applied in scenarios II and III to testify the performance of AB. Impacts of coagulant and adsorbent dosage, initial flux, AB pressure and concentrate retention time (CRT) were integrally investigated in scenarios IV~VI. The adjustment of CRT, as a way to control the degree of concentration and mineralization of organic matters, was implemented by periodically manual discharge of the concentrate through the valve at the side-bottom of the reactor after AB mixing. The change of scenario started when TMP reached 70kPa or the concentrate and filtration became relative stable for more than 500h. Physical cleaning rather than the dominant and costly chemical cleaning for membrane flux recovery was applied between the scenarios.

Table 1 Lists of different working scenarios for the whole pilot-scale long-term UF E-MCR operation

Scenarios	Dosage	Fouling control	Initial Flux (L/m^2h)	Relaxation ratio (min:min)	AB pressure/ IA rate	AB ratio /IA ratio	CRT (d)
I	/	None	5	5:1	/	/	/
II	PACl 60mg/L	IA	5	5:1	0.5 m^3/m^2h	1min:5min	/

III	PACl 60mg/L+ PAC 40mg/L	IA	5	5:1	0.5 m^3/m^2h	1min:5min	/
IV	PACl 60mg/L+ PAC 40mg/L	AB	5	5:1	50 kPa	30s:5min30s	5
V	PACl 60mg/L+ PAC 40mg/L	AB	10	5:1	50 kPa	30s:5min30s	5
VI	PACl 30mg/L+ PAC 20mg/L	AB	20	5:1	100 kPa	30s:5min30s	2.5

IA: Intermittent aeration
AB: air backflushing
CRT: concentrate retention time

Analytical procedures

For better E-MCR filtration performance evaluation with respect to the comprehensive impact of ECP, reflecting the impact both on the TMP and on the flux at the same time, the permeability L_p is used and computed as in Eq. (1) (Belafi-Bako *et al.*, 2006):

$$L_p = J/\Delta P \tag{1}$$

where J is the flux of the filtration (L/m^2h), and ΔP is TMP pressure(bar).

Chemical oxygen demand (COD) was determined according to the Chinese National Environmental Protection Agency (NEPA) Standard Methods (CNEPA, 2002). Biochemical methane potential (BMP) tests were used to determine the anaerobic biodegradability of the concentrate (Angelidaki *et al.*, 2009). Details are described in our previous study (Jin *et al.*, 2016). The energy balance of the pilot-scale UF E-MCR coupled with the performance of the CSTR demonstration was also implemented. The method was described in previous work(Jin *et al.*, 2016).

RESULTS AND CONCLUSIONS

Filtration performance

Figure 2 shows the permeability profiles of all six working scenarios in the long-term pilot-scale UF E-MCR sewage pre-concentration. Significant differences in the permeability profiles could be observed among first three working scenarios. In scenario I that lasted for about 250h, the permeability of the direct sewage UF system dropped dramatically from 112 $L/m^2{\cdot}h{\cdot}bar$ to below 10 $L/m^2{\cdot}h{\cdot}bar$ within 48h. This corresponds to fast dense cake layer formation on UF membrane like microfiltration as mentioned in previous study(Jin *et al.*, 2016). During the next around 200h, the permeability fluctuated between 4 $L/m^2{\cdot}h{\cdot}bar$ and 9 $L/m^2{\cdot}h{\cdot}bar$, indicating that the balance between the cake layer formation and system filterability was reached. While this was far from the requirement of practical application and modification was accordingly in demand. It is also worth mentioned that physical cleaning (rinse the membrane using tap water) removed almost all cake layer deposits on the membrane and the initial TMP recovered to 1 kPa at the beginning of the next working scenario. Therefore, biofouling is no more the major issue in membrane-based sewage pre-concentration like MBR, whereas the filterability optimization of the cake layer deserves detailed investigation.

When PACl (a dosage of 60 mg/L) and IA (5 min of aeration every 6 min) was applied in scenario II, the decline of the permeability from 118 $L/m^2 \cdot h \cdot bar$ to 10 $L/m^2 \cdot h \cdot bar$ costed around 100h and turned out to be much slower than that in scenario I. This demonstrates the performance of the coagulation process and IA in preventing fast membrane fouling in direct sewage UF. However, the permeability further decreased to 6 $L/m^2 \cdot h \cdot bar$ within the next 30h, indicating that the cake layer with poor filterability formed at the end and common coagulation process (CCP) with IA was not efficient enough to reduce the vulnerability of the UF membrane to severe fouling arisen from dense cake layer formation. As such, the ECP which incorporated PAC into CCP was implemented with IA in scenario III. As shown in Figure 2, the initial permeability was improved to 187 $L/m^2 \cdot h \cdot bar$ and the end permeability was around 30 $L/m^2 \cdot h \cdot bar$ for the 120h UF filtration, which was much higher than previous two scenarios. This demonstrates the role of PAC in improving the filterability of the cake layer. The better filtration performance can be also attributed to the adsorption of soluble microbial products (SMP) and extracellular polymeric substances (EPS) by PAC which can prevent the formation of poor permeable gel layer on the membrane (Yu *et al.*, 2006).

In saving aeration energy consumption, which is regarded to comprise the majority of energy cost in commercial MBR, IA was replaced by AB in scenario IV. The initial permeability was further improved to 217 $L/m^2 \cdot h \cdot bar$ compared to scenario III. This can be ascribed to the enhanced back transport forces induced by AB on the cake layer(Viero *et al.*, 2007). Moreover, the permeability decreased to 50 $L/m^2 \cdot h \cdot bar$ after 120h operation. This reveals the better filtration sustainability when AB was applied. For next 540h operation, which was set for the demonstration of pilot-scale reactor feasibility after the investigation of different roles of ECP and AB in sewage UF pre-concentration, the permeability mainly ranged between 20~40 $L/m^2 \cdot h \cdot bar$, indicating a pseudo-steady UF concentration operation was achieved. However, for the last 100h the permeability reduced below 20 $L/m^2 \cdot h \cdot bar$, though higher than 15 $L/m^2 \cdot h \cdot bar$ and still better than scenario I and II. This shows that the thickening and compression of the cake layer is inevitable during long-term operation and optimization of the cake layer deserve further examination.

Consequently, the initial flux was doubled in scenario V for efficiency improvement. The enhancement of the initial flux arose the initial permeability up to 224 $L/m^2 \cdot h \cdot bar$, while the permeability decline to 30 $L/m^2 \cdot h \cdot bar$ was also accelerated within 110h, followed by relative stable fluctuation between 20 $L/m^2 \cdot h \cdot bar$ and 30 $L/m^2 \cdot h \cdot bar$ for the rest 520h in scenario V. As such, the improvement of initial flux promoted the achievement of E-MCR pseudo-steady state which was similar to scenario IV. This implies the faster formation of the cake layer on membrane in scenario V which can limit the formation of gel layer and expand the sustainability of the UF filtration in a more stable state. Accordingly, AB pressure was improved in scenario VI to enhance the AB effect in fouling control coupled with the higher initial flux and lower dosage for efficiency optimization. The permeability at the beginning of scenario VI reached 355 $L/m^2 \cdot h \cdot bar$ and dramatically dropped to around 50 $L/m^2 \cdot h \cdot bar$ within 50h. During the rest 570h pseudo-steady operation, the permeability decreased slowly from 50 $L/m^2 \cdot h \cdot bar$ to 20 $L/m^2 \cdot h \cdot bar$, which was slightly higher than the average permeability in the pseudo-steady state of scenario V. The higher initial permeability introduced faster cake layer formation along with the less dosage of PACl and PAC, but the higher AB pressure managed to effectively alleviate the thickening and compression of the cake layer and enhanced the filterability of the UF membrane. This illustrates the key role of AB in preventing severe membrane fouling and improving the sustainability of the E-MCR operation in sewage pre-concentration. The life span of the UF membrane can be also prolonged with minimized chemical cleaning in E-MCR system.

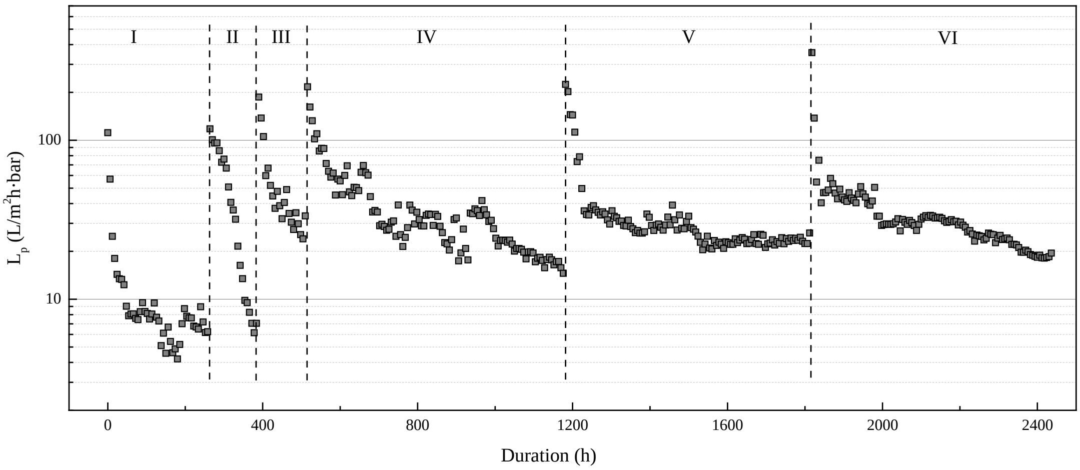

Figure 2 Permeability (L_p) profiles of pilot-scale UF E-MCR for different working scenarios

Concentration Performance

Figure 3 shows the COD concentration profiles of the concentrates for different working scenarios. In scenario I, the COD concentration of the concentrate rose up to around 5400mg/L without discharge. The failure of further concentration increment might be attributed to the decline of the flux and the deposition of organics on the membrane surface and reactor walls, which formed the cake layer only collectable by physical cleaning. When CCP was applied in Scenario II, the COD concentration of the concentrate increased to 7454 mg/L within 120 hours. The faster concentration rate demonstrated the role of coagulation in improving concentration efficiency. ECP introduced even greater enhancement in concentration efficiency, which was supported by the concentrate COD of around 15000 mg/L after 120h concentration. On account of strengthened interaction of particles within the cake layer caused by PAC, the concentration efficiency improvement may be ascribed to the subsequent improved detachability of the cake layer, which would be susceptible to the IA induced disturbance and drawn back into the concentrate. The ECP was demonstrated more effective in improving concentration efficiency.

Scenario IV~VI investigated the impact of AB and CRT on long-term sewage pre-concentration efficiency. With a CRT of 5d (120h), the concentrate in scenario IV obtained a COD concentration of around 19500 mg/L by the end of the first 120h, with the maximum value of around 30000 mg/L at 170h, and then mainly varied between 20000 mg/L and 28000 mg/L along with an average of around 23300 mg/L. It can be inferred that the pushing force induced by AB managed to drive more part of the cake layer back into the concentrate, leading to further enhanced concentration efficiency than scenario III. Considering the heterogeneous structure of the cake layer, the amount of organics accumulated on the cake layer may seldom be same as those detached from caused by AB, thus leading to the fluctuation of the concentrate COD like a pseudo-steady operation. The concentrate in scenario V reached around 28000 mg/L within 120h and mainly fluctuated between 22000 mg/L and 30000 mg/L for pseudo-steady period. The enhanced initial flux and more stable filterability may account for such further increment in the concentrate COD. To minimize the mineralization of organics, the CRT was reduced to 2.5d in scenario VI. As such, the concentrate COD stayed around 18000 mg/L in a more stable state. The initial flux of 20 $L/m^2 \cdot h$ further increased the concentration rate by achieving the maximal level within around 60h. The optimized AB was also proven to promote stabilizing the concentration operation.

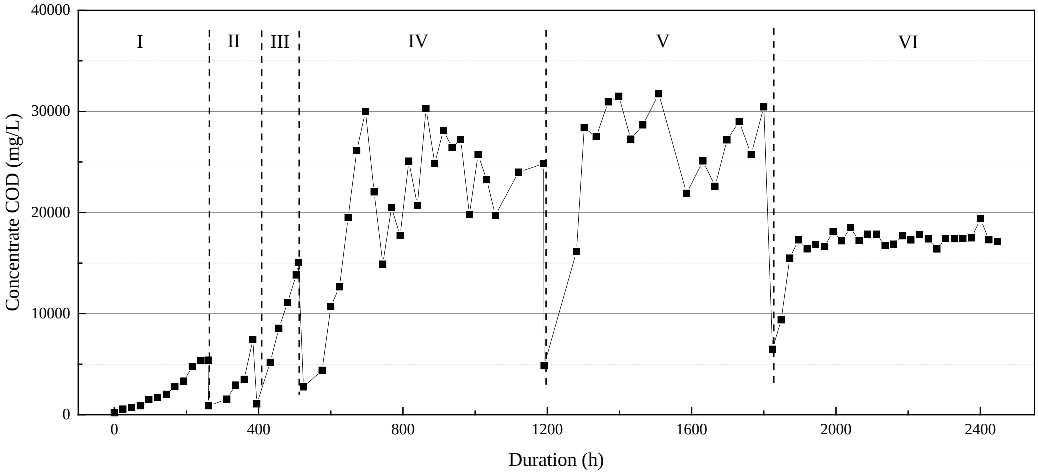

Figure 3 COD concentration profiles of pilot-scale UF E-MCR during the long-term operation

Energy consumption

A summary of the energy balance for the pilot-scale UF E-MCR in scenario VI is shown in Table 4. The electrical energy requirement for lifting the influent was found to be 0.011 kWh/m^3. The energy requirement for dosing reagents was not taken into account because of its negligible head loss. The average energy loss of permeate flow through the membrane was calculated based on the average TMP of 46.8kPa, equivalent to a hydraulic head loss of 4.68 m, giving the permeate energy requirement of 0.013 kWh/m^3. As such, including the pumping energy of 0.0006 kWh/m^3required for the influent passing through the tube, the total pump power requirement in scenario VI was 0.2416 kWh/m^3. The AB energy requirement is calculated by Eq. (5). For a normal hollow fiber system, the value of λ is often 1.4 and the δ has a value of 54(Verrecht *et al.*, 2008). In the pilot-scale UF E-MCR, the depth of the middle point of the hollow fiber membrane (1.0 m) was set as the blower depth assuming that air bubbles equally came out of the membrane during the AB. The normalized average air flow rate was 0.135 m^3/h after calibration by air compressor pump curve and the AB ratio. This makes the AB energy requirement being 0.012 kWh/m^3. In general, the main total energy requirement for E-MCR in scenario IV was 0.0366 kWh/m^3, which is only a small fraction of the reported energy consumption (0.7-1.1 kWh/m^3) in commercial aerobic MBRs for sewage treatment(Krzeminski *et al.*, 2012).

Based on the anaerobic biodegradability of 60.0% derived from the BMP tests and about 90% of influent COD recovered, the expected methane produced from the E-MCR concentrate in scenario VI was about 2.34 mol/m^3 influent. On account of the energy conversion efficiency of 21% of the total gaseous methane energy(McCarty *et al.*, 2011), the energy production from methane could reach to 0.1102 kWh/m^3. Thus the E-MCR in scenario VI would produce 0.0736 kWh/m^3 of energy. By further optimization, more net energy production based on sewage organic matter pre-concentration process can be anticipated. The application of E-MCR for semi-decentralized sewage resource recovery was demonstrated promising.

Table 2 Energy balance of pilot-scale UF E-MCR for scenario VI

	Pilot-scale E-MCR
Electrical Energy Requirement	
Energy for Influent	
Influent flow rate, m^3/d	7.83
Lifting head, m H_2O	4
Lifting pump energy, kWh/m^3	-0.011
Head loss of influent tube, m H_2O	0.2
Required pumping energy, kWh/m^3	-0.0006
Energy for permeate extraction	
Average TMP, m H_2O	4.68
Permeate flow rate, m^3/d	7.83
Required pumping energy, kWh/m^3	-0.013
Total required pumping energy, kWh/m^3	-0.0246
Energy for air backflushing	
Average membrane air blowing depth, m H_2O	1.0
Average air flow rate, m^3/h	0.135
Required air compressor energy, kWh/m^3	-0.012
Total energy consumption, kWh/m^3	-0.0366
Electrical Energy Production Potential	
Average influent COD, mg/L	278.1
Recovery efficiency, %	90
Anaerobic biodegradability, %	60.0
Methane production, mol/m^3 influent	2.34
Electrical energy production from methane, kWh/m^3	+0.1102
Electrical energy produced, kWh/m^3	**+0.0736**

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