Integration of aerobic granular sludge and membrane filtration for tapioca processing wastewater treatment: fouling mechanism and granular stability

Hong Thi Bich Truong, Phuong Thi Thanh Nguyen and Ha Manh Bui

ABSTRACT

In this study, a novel aerobic granular sludge membrane bioreactor (AGMBR) was developed and its performance was compared with that of a traditional membrane bioreactor (MBR). The findings of membrane filtration at the flux of 12 L/m^2 h showed that the fouling rate of the AGMBR was 0.490 kPa/day, half that of the MBR. Resistance analysis implied that the cake resistance was the major fouling factor in the MBR, up to 67.2% of total resistance, while pore blocking was the key fouling factor in the AGMBR, accounting for 50.3%. Aerobic granules maintained stability in the AGMBR with a size of 1.2–1.5 mm and a sludge volume index (SVI) of 60–70 mg/L. It was therefore possible to reduce cake resistance and fouling rate. In addition, the concentration of soluble extracellular polymeric substances (sEPS) in the AGMBR was 33.1 mg/L, three times lower than that of the MBR, which resulted in a reduction in membrane fouling for the AGMBR compared with the MBR.

Key words | aerobic granular membrane bioreactor, aerobic granular sludge, filtration performance, granular stability, membrane fouling, tapioca processing wastewater

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INTRODUCTION

Membrane bioreactor (MBR) technology combining an activated sludge process with membrane filtration has exhibited advantages compared with a conventional activated sludge process, such as producing effluent without suspended solids (SS) and microorganisms, high biomass concentration, low excess sludge production, and a small footprint. With the above advantages, MBR technology has attracted attention from many researchers and has been commercialised since the 1960s. In spite of high treatment performance, frequent membrane fouling in MBRs decreases permeability and membrane life. That is because the small size of the activated sludge used in a MBR easily adheres to the membrane surface and causes serious fouling.

Aerobic granular sludge has exhibited advantages compared with conventional activated sludge, such as good settleability, the ability to withstand high organic loading rates and tolerance to toxicity (Wang *et al.* 2017), and the capacity for simultaneous nitrogen and phosphorus removal (Li *et al.* 2017). In addition, using aerobic granules with compact structure and large particle size has been the accepted approach for restricting membrane fouling (Wang *et al.* 2013; Li *et al.* 2017). However, it has been reported that the aerobic granular sludge process may not meet SS discharge standards (Thanh *et al.* 2008; Nguyen *et al.* 2016). Therefore, the integrated operation of aerobic granular sludge and membrane filtration could be a way of combining the advantages and addressing the problems associated with both processes.

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The membrane was used to separate SS from the effluents, while granular sludge was used to reduce fouling.

An aerobic granular sludge membrane bioreactor (AGMBR) integrating aerobic granular sludge with membrane filtration was first developed by Li *et al.* (2005) and attracted considerable attention from many researchers. Studies determined removal and filtration performance as well as the membrane foulants. The findings showed that the chemical oxygen demand (COD) removal efficiency of the AGMBR was higher than 90% (Tay *et al.* 2007; Tu *et al.* 2010). The simultaneous occurrence of nitrification and denitrification in aerobic granules improved NH_4^+-N and total nitrogen (TN) removal efficiency to more than 90 and 70%, respectively (Tu *et al.* 2010; Jang *et al.* 2012). Comparing filtration performance between activated sludge and granular sludge, most of the studies suggested that aerobic granular sludge maintained higher permeability and reduced fouling rates (Tu *et al.* 2010; Wang *et al.* 2013). However, a major challenge is to maintain the stability of aerobic granules during continuous membrane operation. It has been reported that the AGMBR with operational mode as a sequencing batch reactor (SBR) created suitable conditions for the stability of aerobic granules (Tay *et al.* 2007; Jang *et al.* 2012; Wang *et al.* 2013). However, aerobic granules stabilised in the reactors at a rather small size, lower than 1 mm. It has been reported that the cake layer resistance of aerobic granular sludge decreased as particle size increased. Thus, the granules of larger size and more compact structure should be maintained in the reactor in order to improve the filtration performance and mitigate membrane fouling. At present, the main foulants and fouling mechanism have not been clarified. Both the short-term batch filtration (Zhou *et al.* 2007) and long-term continuous filtration in a batch reactor (Wang *et al.* 2013) revealed that the pore blocking level of aerobic granular sludge was higher than that of activated sludge. Moreover, Sajjad & Kim (2015) conducted long-term continuous filtration in a reactor performing the same operation and showed the reduction of both pore and cake layer blocking levels in an aerobic granular sludge system compared with an activated sludge system. Therefore, further investigation is necessary to determine clearly the fouling mechanism and suitable operational conditions for the stability of aerobic granular sludge.

In addition, the AGMBR has been studied mainly in terms of synthetic wastewater from glucose, protein (Li *et al.* 2005; Sajjad & Kim 2015) and acetate (Tay *et al.* 2007; Wang *et al.* 2013). Information associated with real wastewater is still limited, with the exception of the study of Li *et al.* (2017) regarding municipal wastewater treatment. It has been believed that the filtration performance increased with increasing granular size and strength. Meanwhile, tapioca processing wastewater, which has high concentrations of organic compounds and nutrients, is appropriate for the formation of aerobic granules. Aerobic granules formed in tapioca processing wastewater had large particle size and compact structure (Truong *et al.* 2018), and this method would therefore be an efficient approach for fouling control and filtration performance improvement.

In this study, a novel AGMBR was developed and its performance was compared with that of the traditional MBR. The main objective of this work was to investigate membrane filterability and the stability of aerobic granules in the AGMBR. The membrane fouling mechanism was also revealed through filtration resistance analysis and scanning electron microscopy (SEM) images of fouled membrane surfaces. In addition, treatment performances in terms of COD and NH_4^+-N removal were included. The findings obtained are expected to provide useful information for further development of AGMBR technology.

MATERIALS AND METHODS

Reactor setup

This study was performed in a cylindrical-column reactor with a working volume of 3.3 L (Figure 1). The reactor was 6.5 cm in internal diameter and 100 cm in height. Air was supplied by an air pump (ACO-003, Hailea, Taiwan) through the pumice stone at the reactor bottom in order to provide mixing as well as shear force through the membrane at an airflow rate of 5 L/min (surface air velocity 2.5 cm/s). Wastewater was fed into the reactor with a dosing pump (PZD-500-VTCF, Tacmina, Japan). The effluent was discharged through the membrane module by a dosing pump (DULCO®flex, ProMinent, Germany). Transmembrane pressure (TMP) was recorded by a pressure

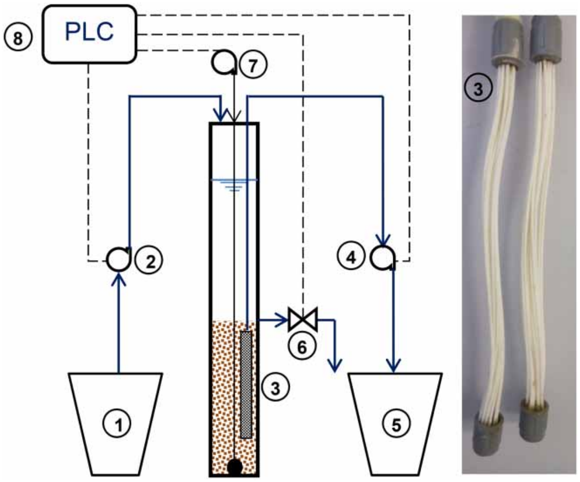

Figure 1 | Schematic diagram of MBR and AGMBR: (1) feeding tank, (2) feeding pump, (3) membrane module, (4) membrane pump, (5) effluent tank, (6) discharged valve, (7) air pump, (8) PLC controller.

sensor (PSAN-LV01CV-R(PT)1/8, Autonics, Korea) installed in the inlet pipe. This system was automatically controlled by a programmable logic controller (PLC) (12/24RC, Siemens, Germany). A hollow fiber membrane module (Mitsubishi Rayon, Japan) was used, as specified in Table 1.

Operational procedure and conditions

The MBR and AGMBR had the same operational scheme and parameters but different seed sludge. The reactors were operated as SBRs with a 3-h cycle consisting of 5 min of influent filling, 60 min of aeration, 110 min of simultaneous aeration and membrane filtration, and 5 min of idling. A membrane pump ran intermittently, 10 min on and 1 min off, to restrict fouling. Mixed liquor equal to 3/8 of the reactor's working volume was discharged through the membrane module in proportion to a volumetric exchange ratio of 37.5%. Poorly settling sludge was withdrawn periodically in order to maintain a stable mixed liquor suspended solids (MLSS) concentration in the reactors. Operational parameters of the reactors are presented in Table 2. All experiments were carried out in a laboratory and were operated at an ambient temperature range of 28–32 °C.

Table 1 | Parameters of hollow fiber membrane module

Parameter	Unit	Value
Material	–	PVDF (polyvinylidene fluoride)
Module size (D × H)	cm	2.5 × 35
Membrane pore size	μm	0.4
Outer diameter of hollow fiber	mm	2.6
Membrane surface area	m^2	0.05
Operating temperature	°C	0–40
Operating pressure	kPa	0.5–15
Filtration direction	–	Cross-flow

Table 2 | Operational parameters and conditions of MBR and AGMBR

Parameter	Unit	Value
Organic loading rate (OLR)	kg COD/m^3 day	2.5
Chemical oxygen demand (COD)	mg/L	787
Hydraulic retention time (HRT)	h	7.56
MLSS	g/L	5–6
Food/microorganism (F/M)	kg COD/kg MLSS day	0.4–0.5
Airflow rate	L/min	5
Filtration flow rate	L/h	0.6
Flux	L/m^2 h	12.0

Membrane cleaning

When TMP exceeded 15 kPa, membrane modules were taken out and cleaned. Membranes were first flushed with tap water to remove sludge cake on the membrane surface, then submerged in 1 L 0.1% sodium hypochlorite (NaClO) solution for 8 h to remove the pore foulants.

Materials

Wastewater

Tapioca processing wastewater was produced every month by the traditional method in the laboratory and stored in a refrigerator at 5 °C. The properties of the raw wastewater were as follows: pH 3.9–4.5, COD 4,800–14,000 mg/L, biological oxygen demand 2,500–11,550 mg/L, CN^- 2–75 mg/L, SS 350–1,000 mg/L, NH_4^+-N 95–182 mg/L, total nitrogen (TN) 145–470 mg/L, total phosphorus (TP) 127–432 mg/L. Prior to being fed into the reactors, the raw wastewater was diluted with tap water until it attained the influent COD values listed in Table 2. The pH was maintained at 7.0 ± 0.5 using 0.1 N NaOH and 0.1 N HCl. The influent was supplemented by nutrient ingredients (NH_4^+-N, PO_4^{3-}-P) to ensure a COD:N:P ratio of 100:5:1 and macronutrient, micronutrient dosage as suggested by Nguyen *et al.* (2016).

Seed sludge

The activated sludge and aerobic granular sludge were used as seed sludge in the MBR and AGMBR, respectively. The properties of the seed sludge were as follows: mean size lower than 100 μm, the ratio of mixed liquor volatile suspended solids to mixed liquor suspended solids (MLVSS/MLSS) 85.81%, SVI 148 mL/g for activated sludge and mean size 1.5 mm, MLVSS/MLSS 94.15%, SVI 58 mL/g for granular sludge.

Analytical methods

EPS extraction and analysis

Soluble extracellular polymeric substances (sEPS) were obtained by mixed liquor filtration using a membrane with a pore size of 0.45 μm. Bound EPS (bEPS) in sludge were extracted using the formaldehyde–sodium hydroxide method (Liu & Fang 2002). The extracted soluble and bound EPS were analyzed for proteins and polysaccharides. Proteins were determined by the modified Folin–Ciocalteu phenol method (Lowry *et al.* 1951), in which bovine serum albumin was used as the protein standard. Polysaccharides were determined by the phenol-sulfuric method using glucose as the standard (Dubois *et al.* 1956).

Resistance analysis

Filtration resistances were determined by Darcy law:

$$R_t = R_m + R_f = R_m + R_c + R_p = P/(\mu \times J) \quad (1)$$

$$R_f = R_c + R_p \quad (2)$$

where R_t is total resistance, R_m is membrane resistance, R_f is fouling resistance, R_c is cake resistance, R_p is pore resistance, P is the transmembrane pressure, μ is the dynamic viscosity, and J is the flux.

The experimental procedure to determine resistances was as follows (Zhou *et al.* 2007): (1) R_m was evaluated by clean water filtration through new membrane, (2) R_t was evaluated by clean water filtration through fouling membrane, and (3) $(R_m + R_p)$ was evaluated by clean water filtration through fouling membrane after being removed from the cake layer. The pore resistance (R_p) was calculated from steps (1) and (3) and the cake resistance (R_c) was calculated from steps (2) and (3): $R_p = (3) - (1)$ and $R_c = (2) - (3)$.

Other analytical methods

Granules were observed with an optical microscope (BX 51, Olympus, Japan) with an attached camera (DP 71, Olympus, Japan) and image analysis software (ImageJ). Roundness was determined by ImageJ analysis software (0 = line, 1 = circle). The particle size distribution (PSD) was measured by the wet sieve method as reported by Truong *et al.* (2018). The granule settling velocity was determined in the reactors. The time for sludge settling to half the reactor working volume was used to determine the granule settling velocity. The strength of granules was defined as the capacity of the granules to resist disintegration during operation and determined according to the method of Wang *et al.* (2004). The sample sludge was diluted 10 times with tap water, and 25 mL of the diluted sample was taken for the test. This sample was subjected to abrasion by placing it on a shaker at 200 rpm for 5 min. It was then allowed to settle within 1 min. The two portions including the settled granules and supernatant with unsettled and disintegrated particles were collected for the volatile suspended solids (VSS) test. The granule strength is expressed in terms of integrity coefficient (%), which is defined as (residual granule VSS: total VSS) × 100.

The environmental parameters (COD, NH_4^+-N, TP, SS) and sludge parameters (MLSS, MLVSS, SVI) were analyzed according to standard methods (APHA 2005). The membrane structure was determined by SEM (S-4800, Hitachi, Japan). Membranes were dehydrated by drying at 60 °C for 24 h before the SEM analysis.

Data analysis

The experiments were performed in triplicate and the average values ± standard deviations (SD) were reported. The one-way analysis of variance was performed to determine the statistical heterogeneity of TMP data between the MBR and the AGMBR with a confidence limit of 95% ($P < 0.05$).

RESULTS AND DISCUSSION

Filtration performance and fouling mechanism

Membrane fouling

The variation in TMP with time during the 64-day operation showed TMP increased more slowly in the AGMBR than in the MBR (Figure 2). The average time to maintain permeability of the AGMBR was 22 days, twice that of the MBR. The TMP profile may be divided into two stages, including a slow increase in approximately the first half of the operational time and a rapid increase during the remainder of the time. The fouling rates of the MBR and the AGMBR were 0.986 and 0.490 kPa/day, respectively. It can be seen that the fouling rate of the granular sludge reactor was half that of the activated sludge reactor. Statistical analysis results also showed the heterogeneity of the TMP data in the MBR and the AGMBR ($P = 0.03$; details can be found in the supplementary data, available with the online version of this paper). With the same operational mode, other studies also revealed that fouling decreased with increasing granular size (Wang *et al.* 2013; Sajjad *et al.* 2016). It was reported that the granular sludge tended to remain suspended rather than settle on the membrane surface, while the floc sludge settled easily on the membrane and increased the density of the sludge cake layer (Tay *et al.* 2007; Wang *et al.* 2012). The Carman–Kozeny equation also provides the important implication that the specific resistance of the cake layer has an inverse relationship with particle size (Zhou *et al.* 2007). In addition, sEPS, which were considered to be key fouling factors, were synthesized into granules for stable granulation and that resulted in the reduction of membrane foulant amount in the AGMBR of three times compared with the MBR.

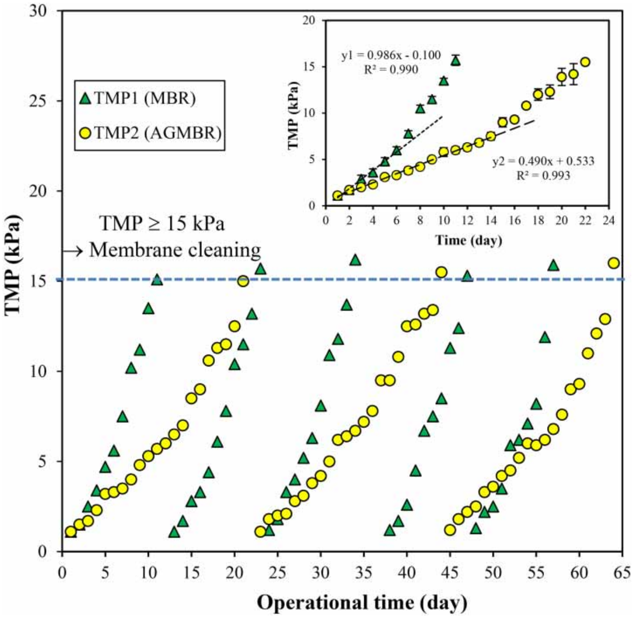

Figure 2 | The variation in TMP with time during membrane filtration operation.

Filtration resistance

To understand the mechanism of membrane fouling, the contribution of different filtration resistances was determined. Resistance analysis results in Table 3 show that large and compact aerobic granules had low compressibility on membrane surfaces and that reduced cake resistance. The cake layer resistance in the MBR contributed to 67.2% of total resistance, while pore blocking was the main resistance in the AGMBR, accounting for 50.3%. The continuous filtration in the SBR showed that the main resistance of the aerobic granular system was pore blocking, which contributed to 76.21% of total resistance, while the main resistance of the flocculent system was cake fouling, up to 61.23% (Wang *et al.* 2013). Under the same operational conditions, Li *et al.* (2017) also found that the irreversible fouling resistance of the aerobic granular system was

Table 3 | Filtration resistance of MBR and AGMBR

Reactor	R_m ($\times 10^{11}$/m)	R_c ($\times 10^{12}$/m)	R_p ($\times 10^{12}$/m)	R_t ($\times 10^{12}$/m)	R_c/R_t (%)	R_p/R_t (%)
MBR	3.88	3.90	1.51	5.81	67.2 ± 0.4	26.1 ± 0.2
AGMBR	4.00	2.45	2.89	5.74	42.7 ± 1.6	50.3 ± 1.5

R_m membrane resistance, R_c cake resistance, R_p pore resistance, R_t total resistance.

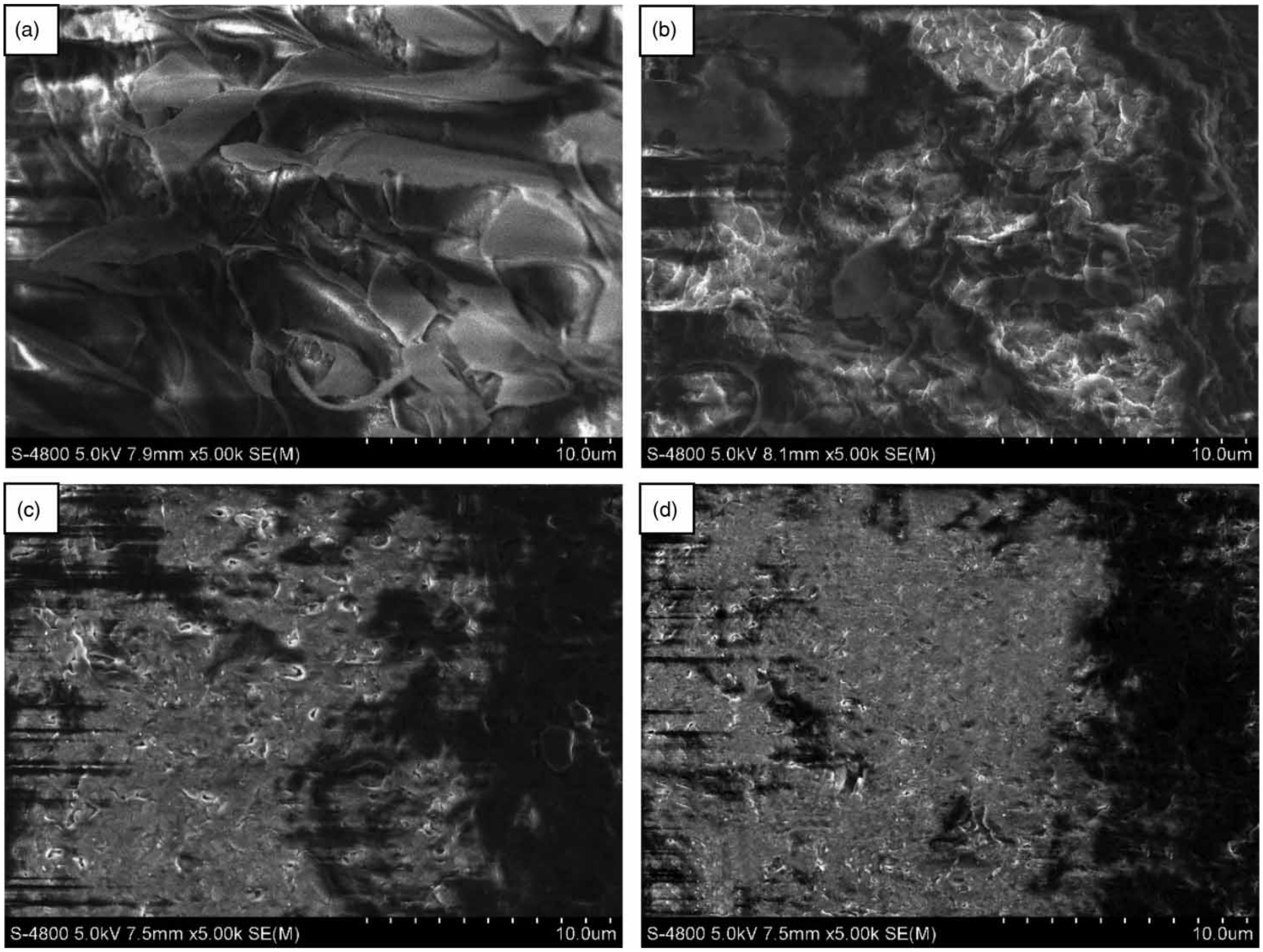

Figure 3 | SEM images of membrane surfaces: (a) and (b) fouled membranes for MBR and AGMBR, (c) and (d) hydraulic cleaned membranes for MBR and AGMBR, respectively.

higher than 85%. The study conducted on the batch filtration test also demonstrated that pore resistance contributed 44.2% of total resistance in the aerobic granular system and cake resistance contributed 72.68% in the flocculent sludge system (Zhou *et al.* 2007). This can be explained by the tendency of the aerobic granules to remain suspended rather than to settle on the membrane surface, which resulted in the reduction of cake resistance to 42.7%.

Figure 3 shows the SEM images of fouled and hydraulic cleaned membranes. In the AGMBR fouled membrane, the cake layer formed with more porosity compared with the MBR (Figure 3(a) and 3(b)). The aerobic granules had a large size and compact structure and did not easily attach to or clog on the membrane surface. Thus, the formed cake layer had a loose structure, and that was beneficial for maintaining permeability. The dense image of the MBR may be induced by the deposition of fine particles in activated sludge on the membrane surface. Under high suction force of membrane filtration, the fine particles would easily and quickly accumulate on the membrane surface to form a compact cake layer. The substances which were smaller than the pore size of the membrane entered into the pore and caused blocking. The photographs of cleaned membranes revealed more opened pores in the MBR than the AGMBR (Figure 3(c) and 3(d)). This result showed that the thick cake layer which developed on the membrane surface in the MBR could serve as a barrier to restrict the deposit of pore-blocking substances, such as dissolved substances and colloids, into the membrane pores. This is why the MBR had lower pore resistance than the

Table 4 | Membrane recycled efficiencies for MBR and AGMBR

Reactor	New membrane resistance R_{nm} ($\times 10^{11}$/m)	Recycled membrane resistance R_{rm} ($\times 10^{11}$/m)	Recycled efficiency (%)
MBR	3.88	4.00	96.7 ± 5.8
AGMBR	4.00	4.25	93.6 ± 5.5

AGMBR. SEM images of the membrane surface in the short-term batch filtration test of Zhou *et al.* (2007) also revealed more opened pores of the cleaned membrane in the floc system than the granular system. Indeed, it can be disclosed from the findings of resistance and SEM analysis that the key fouling mechanism of the MBR was cake layer fouling, while that of the AGMBR was pore blocking.

When TMP exceeded the critical value of 15 kPa, membrane modules were taken out and cleaned with 0.1% NaClO solution. The results in Table 4 show that membrane recycled efficiencies were high for both the MBR and the AGMBR, with values of 96.7 and 93.6%, respectively. In the literature, NaClO solution was usually used as an effective membrane cleaning agent to obtain permeability recovery of more than 90% (Zhou *et al.* 2007; Sajjad & Kim 2015; Wang *et al.* 2018). NaClO solution has been used generally to clean membrane in practical wastewater treatment.

Membrane foulants

It has been reported that membrane foulants included suspended solids, colloids and dissolved molecules in mixed liquor. Among such foulants, EPS-microbial products excreted by cells have been identified as the most significant factor responsible for membrane fouling (Tay *et al.* 2007; Wang *et al.* 2012; Vijayalayan *et al.* 2014). Depending on the location of the EPS compared with the cell, the EPS consist of soluble EPS (sEPS) and bound EPS (bEPS). The components of EPS include polysaccharides, proteins, nucleic acid, lipids, humic acid, uronic acid, amino acid and inorganic matter. Among them, proteins (PN) and polysaccharides (PS) are the main matters in EPS. It is necessary to evaluate the components and amounts of EPS in mixed liquor to determine the cause of fouling.

It has been believed that appropriate bEPS amounts of approximately 100–150 mg/g SS and a proteins/polysaccharides ratio of higher than 1 would increase the compact structure of granules. In contrast, EPS concentration higher than 200 mg/g SS resulted in the occlusion of granule porosity and the limitation of nutrient transport from the bulk into granules and consequently the death of bacteria (Li *et al.* 2008). When granular sludge was disintegrated, a large number of broken cells, EPS and soluble substances were released into mixed liquor. These small particles and soluble substances adhered to the surface or covered the pores, which resulted in membrane fouling (Tay *et al.* 2007). Therefore, appropriate EPS amounts should be maintained in order to ensure the stability of aerobic granules for fouling control.

The results in Table 5 show that the bEPS amount in aerobic granules was at an appropriate level of 132.5 mg/g SS, 2.24 times higher than that of flocculent sludge. In addition, there was a large difference in the bound proteins/polysaccharides (bPN/bPS) ratio between granular sludge and flocculent sludge. The bPN/bPS ratio of granular sludge was 2.9, which was 3.4 times higher than that of flocculent sludge. Proteins were identified to be hydrophobic matters, while polysaccharides were hydrophilic matters. In a thermodynamic sense, increasing the hydrophobicity of the cell surface causes a decrease in negative surface charge, which creates favourable conditions for solid (cells)–liquid phase separation and the formation of microbial aggregates. Consequently, a higher hydrophobicity of the

Table 5 | EPS amount in the bulk phase and sludge

	Bulk phase (sEPS)				Sludge (bEPS)			
Reactor	sEPS (mg/L)	sPN (mg/L)	sPS (mg/L)	sPN/sPS	bEPS (mg/g SS)	bPN (mg/g SS)	bPS (mg/g SS)	bPN/bPS
MBR	99.4 ± 2.6	43.1 ± 2.2	56.3 ± 2.8	0.8 ± 0.07	59.2 ± 2.8	26.9 ± 1.3	32.3 ± 1.6	0.8 ± 0.01
AGMBR	33.1 ± 1.0	11.1 ± 0.6	22.0 ± 1.1	0.5 ± 0.04	132.5 ± 3.0	98.2 ± 3.5	34.3 ± 1.3	2.9 ± 0.20

sEPS, soluble EPS; bEPS, bound EPS; sPN, soluble proteins; bPN, bound proteins; sPS, soluble polysaccharides; bPS, bound polysaccharides.

cell surface would result in a stronger cell-to-cell interaction and a dense and stable granular structure (Tay *et al.* 2002; Thanh *et al.* 2008; Liu & Sun 2010). Therefore, a high bPN/bPS ratio would be beneficial for granulation and fouling control. In contrast, the sEPS amount of the MBR was 99.4 mg/L, three times higher than that of the AGMBR. This explained why the MBR had a higher fouling rate compared with the AGMBR. Obviously, most proteins and polysaccharides accumulated in the granules and the sEPS amount was reduced three times compared with that in the MBR, which resulted in the reduction of the fouling rate in the AGMBR. Similar results were also reported by other studies, which found that the aerobic granular system had a suitable bEPS amount of 85–100 mg/g VSS and a high bPN/bPS ratio of 2.6–3.9, as well as a low sEPS amount, and that resulted in fouling reduction (Sajjad & Kim 2015; Sajjad *et al.* 2016).

Stability of aerobic granular sludge in AGMBR

It has been reported that the performance of membrane filtration was determined by the stability of the granules. The variation in size and morphology of the aerobic granules with time in Figure 4(a) shows that granules were still stable in the AGMBR. The granules reached steady size in the range of 1.2–1.5 mm, lower than the size of 1.5 mm on the first day of membrane operation. The morphology of the granules was rather circular, with a roundness of 0.8–0.9. The stabilised granules had a compact structure, including the inner sludge layer with nematodes and the outer sludge layer surrounded by stalked ciliates (Figure 5). The stability of the granules was due to the batch operational mode of the AGMBR, which created stressful culture conditions for the production and consumption of EPS. Therefore, the structure of the granules was improved and EPS content in the reactor was always controlled at an

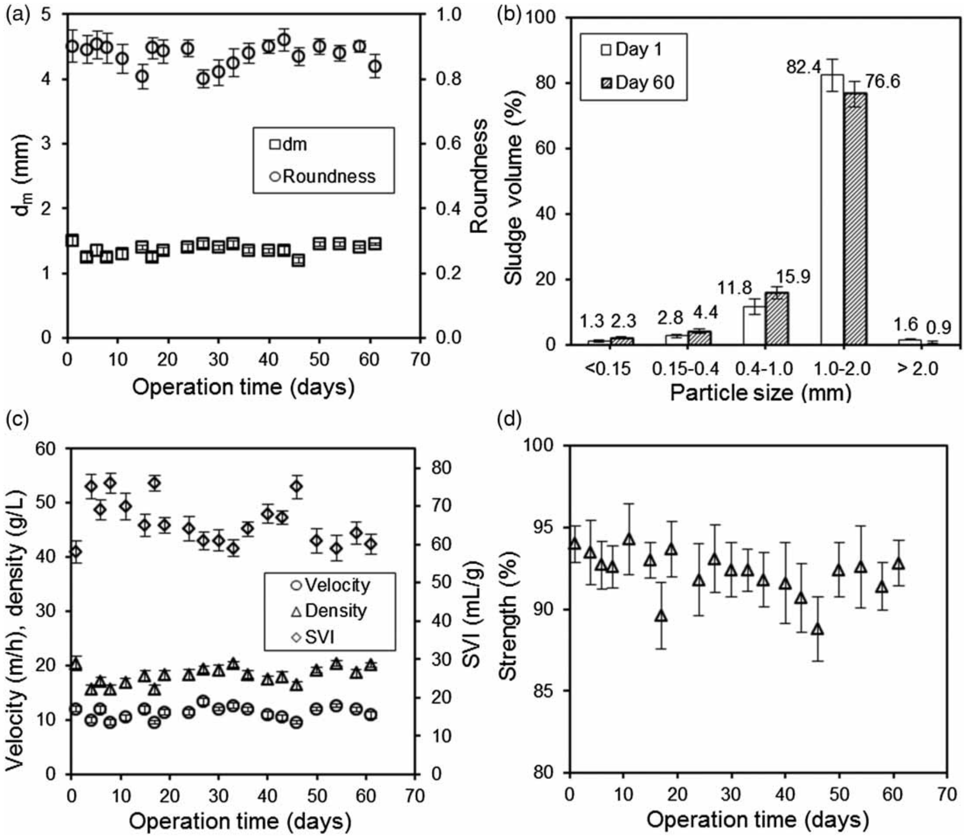

Figure 4 | The properties of granular sludge in AGMBR: (a) the variation in mean diameter (d_m) and roundness with time, (b) PSD, (c) the variation in settling velocity, density and SVI, and (d) strength with time.

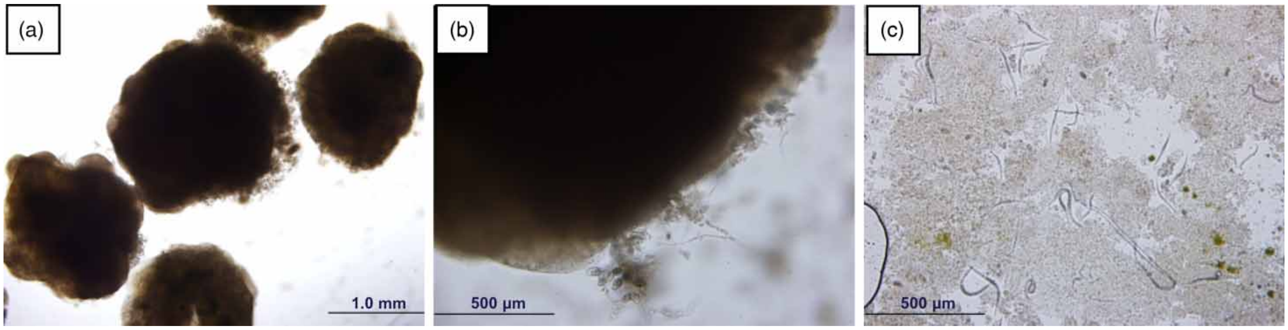

Figure 5 | The granular sludge in AGMBR: (a) granules, (b) ciliates surrounding granule surface, (c) nematodes inner granule.

appropriate level. The frequent discharge of poorly settling floc in the reactor also played an important role. The flocculent sludge includes mainly heterotrophic aerobic bacteria which have quick growth rates. Meanwhile, the bacteria inside the granules are more difficult to contact with the substrate so that they have lower growth rates and are easily competed with by bacteria in the floc. Therefore, the flocculent sludge in the AGMBR should be withdrawn frequently to restrict the competition between flocculent and granular sludge for the long-term stabilisation of aerobic granular sludge. Obviously, the batch operation and floc removal maintained the stability of aerobic granules in the reactor. This is confirmed by the results of other studies which found that the granules were disintegrated in continuous operational reactors (Zhou *et al.* 2007; Li *et al.* 2008). Meanwhile, the studies carried out in batch reactors still maintained the stability of the granules. However, the size of the granules was rather small, less than 1 mm, and the morphology was not uniform (Tay *et al.* 2007; Wang *et al.* 2013; Li *et al.* 2017). The experimental conditions disadvantaged for the granulation in these literatures, such as low OLR of 0.25 kg TOC/m^3 day (Li *et al.* 2017), low surface air velocity 0.1 cm/s (Wang *et al.* 2013) and high HRT 1 day (Jang *et al.* 2012), resulted in a decrease in granule size. The PSD in Figure 4(b) also shows that the diameter of the granules in the AGMBR decreased slightly compared with the seed granules. The granules then stabilised, with the granular particles larger than 1 mm accounting for 77.5%, while flocculent sludge accounted for only 2.3%. The suitable operational conditions in this study, such as high air velocity of 2.5 cm/s, high OLR of 2.5 kg COD/m^3.day and short HRT of 7.56 h, favoured the stability of aerobic granules for fouling control.

The settleability of granules was evaluated through SVI, settling velocity and density (Figure 4(c)). In the first two weeks of operation, the settleability of granules dropped slightly and stabilised in the SVI range of 60–70 mg/L. This is due to the effect of the membrane module on the disturbance and oxygen diffusion in the reactor. In addition, the granules collided easily with the membrane module, leading to partial disintegration. The density and settling velocity of granules were in the range of 16.0–20.0 g/L and 9.0–12.0 m/h, respectively. Strength is one of the important parameters for evaluating the stability of aerobic granules. As presented in Figure 4(d), granules had high strength of 90–94%, which confirmed the stability of granules in AGMBR. The strength in this study was higher than that of granules formed in glucose synthetic wastewater with a value in the range of 70–88% (Wang *et al.* 2004). It can be seen that tapioca processing wastewater has more suitable properties for the formation of compact and strong granules compared with synthetic wastewater.

Removal performance

When the membrane filtration was operated after 60 min of aeration, only COD and NH_4^+-N were removed efficiently because substrate consumption happened quickly in the first 20–30 min of aeration. Nitrogen and phosphor removal had not yet occurred completely, so only COD and NH_4^+-N removal efficiencies were determined.

Figure 6(a) shows that COD removal efficiencies of both granules and floc were as high as 89.3–92.4% for the

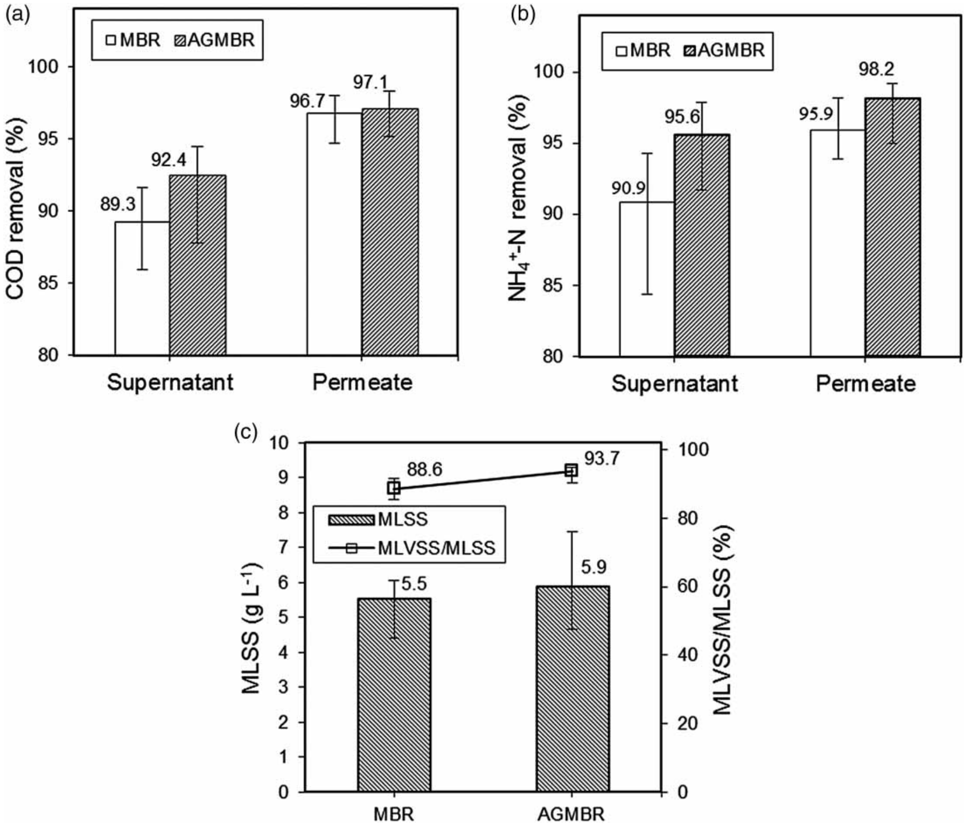

Figure 6 | Removal efficiencies and biomass properties in MBR and AGMBR: (a) COD and (b) NH_4^+-N removal efficiencies, (c) biomass concentration and MLVSS/MLSS ratio.

supernatant and higher than 96% for the permeate. Microfiltration membrane with the pore of 0.4 μm removed SS completely, leading to a decrease in the concentration of dissoluble organic compounds and an increase in COD removal efficiency. SS removal efficiency of permeate was 100%, which aligns with the result of Tay *et al.* (2007), in which 0.1 μm micro-filtration was used. Figure 6(b) shows that NH_4^+-N removal efficiency of granular and flocculent sludge was rather high, reaching 90.9–95.6% for the supernatant and 95.9–98.2% for the permeate. NH_4^+-N was removed mainly by cell synthesis and partially by nitrification. The high substrate removal efficiencies of both the MBR and the AGMBR were due to a high biomass concentration of 5.5–5.9 g/L and an MLVSS/MLSS ratio of 88.6–93.7% (Figure 6(c)). These results are similar to the findings of studies for synthetic wastewater treatment in the same reactors in which COD and NH_4^+-N removal efficiencies were more than 90% (Jang *et al.* 2012; Li *et al.* 2017; Iorhemen *et al.* 2018).

PRACTICAL APPLICATIONS AND FUTURE PERSPECTIVES

This study demonstrated the stability of aerobic granules in an AGMBR with a large size of 1.2–1.5 mm and a compact structure. This was therefore effective to reduce membrane fouling and increase substrate removal efficiency. The filtration time of the AGMBR was twice as high as that of the MBR which reduced membrane cleaning frequency. Thus, operational cost was reduced and membrane life span was increased. For treatment performance, the concentrations of effluent COD and NH_4^+-N were rather low, 13.7–41.3 and 0.3–1.6 mg/L, respectively, so they satisfied the wastewater reuse requirement. In addition, the AGMBR could reduce the required construction area remarkably due to its capacity to withstand high loading rates, good settleability, simultaneous COD and nitrogen removal of aerobic granular sludge, as well as its capacity as an alternative for secondary sedimentation and disinfection tanks of membrane filtration. Indeed, the AGMBR exhibited practical application potential for wastewater reuse and increasing cost efficiency.

The performance of the AGMBR depends on the stability of aerobic granules. To sustain the stability of aerobic granules, the operational scheme and conditions should be controlled well. Thus, further studies on real wastewater treatment as well as full-scale investigations should be carried out in order to determine the optimal operational conditions and suitable design parameters. In this study, the reactors were operated at a low organic loading rate (OLR) of 2.5 kg COD/m^3 day for the suitability of both activated and granular sludge. However, the low OLR did not create appropriate conditions for the development of large granules. Therefore, operation at optimal OLR for the stability of mature granules should be conducted to improve the performance of the AGMBR. In addition, to make membrane cleaning easier, the membrane module should be installed in a separate tank following the aerobic granular SBR. At that time, total nitrogen and phosphorus removal performances should also be investigated.

CONCLUSIONS

Aerobic granular sludge maintained stability in the AGMBR with the average size of 1.2–1.5 mm and good settleability with an SVI of 60–70 mL/g. Granules having compact structure and large size improved the permeability. The fouling rate of the AGMBR was 0.490 kPa/day, half that of the MBR. The main fouling factor in the AGMBR was pore blocking resistance, which accounted for 50.3% of total resistance, while cake blocking was the main fouling factor resistance in the MBR, up to 67.2%. This is because large and compact granules had low compressibility on the membrane surface, which reduced the cake layer resistance. In addition, EPS were concentrated into granules and sEPS concentration in the bulk was decreased, leading to the fouling reduction in the AGMBR. The findings demonstrated the potential applicability of the aerobic granular sludge MBR for the improvement of substrate removal efficiency and the reduction of membrane fouling.

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