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Advances in Granular Growth Anaerobic Membrane Bioreactor (G-AnMBR) for Low Strength Wastewater Treatment

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ABSTRACT

The concept of sustainability has been evolved in the recent years and widely used in reassessing the feasibility of various wastewater treatment technologies. Nowadays, anaerobic bioprocesses are considered as a sustainable technology which has no requirement of oxygen (low cost) while producing bioenergy with low sludge yield. Currently, granular growth anaerobic membrane bioreactor (G-AnMBR), a newly discovered hybrid anaerobic biotechnology, which integrates the granular technology and membrane based separation, has attracted increasing number of studies due to its competitive advantages of less fouling and high energy efficiency. In face of the significance of this hybrid technology, this paper presents an up-to-date review on the performance enhancements of G-AnMBR in low strength wastewater treatment over the last decade while highlighting future research direction in the conclusion.

1. Introduction

Anaerobic biotechnology has long been considered as a sustainable approach, which incorporates waste management with the recovery of useful byproducts and renewable bioenergy. The worldwide application of anaerobic processes would not only alleviate environmental pollution but also ease the stress on energy insecurity, global demand on fossil fuels, continuous exploitation of limited natural resources and the emission of toxic air pollutants, particularly greenhouse gases to the atmosphere [Khanal, 2008]. Moreover, compared to the aerobic counterparts, the operational costs of aeration and sludge dewatering/disposal in anaerobic bioprocesses are distinctly reduced as no oxygen is required and sludge yield is much lower. Additionally, anaerobic treatment has the storage capability unfed for several months without serious deterioration and still generates biogas [Lim and Kim, 2014].

Anaerobic granulation is an autoimmobilization in which fluffy biosolids assemble and agglomerate as dense and compact granules under controlled operational conditions. Compared to the conventional bioflocs, anaerobic granules have a regular and well-defined shape, strong structure, and good settling velocities. They enable high biomass retention and withstand high strength wastewater and shock loadings. This means that the formation of anaerobic granular sludge allows the decoupling of hydraulic retention time (HRT) and solid retention time (SRT), and therefore the efficient treatment of wastewater can be carried out at much higher organic loading rates (OLRs) with a significantly reduced reactor footprint. Anaerobic granular sludge bioreactor (AnGSB) technology has been extensively employed in industrial and municipal wastewater treatment practices since 1980 [Lettinga et al., 1984]. However, AnGSB technology has to overcome some drawbacks which includes: (i) the requirement of extremely long start-up period, (ii) a relatively high operation temperature, (iii) unsuitability for low strength organic wastewater such as municipal wastewater, (iv) poor capability of removing nutrients, and (v) post treatment required to meet discharge standards [Liu and Tay, 2004; Liu et al., 2004; Lim and Kim, 2014]. Recent research efforts have been mainly directed to the discovery of specific high rate AnGSB technology that could resolve the above-mentioned disadvantages. Theoretically, the sufficient inoculation of seed granular sludge in operating AnGSB has the advantage of achieving high organics removal within an accelerated startup period. Some researchers also added additives such as natural polymers, cationic polymers and hybrid polymers to promote particle agglomeration, in order to realize shortening of startup time and enhancement of granulation [Show et al., 2004; Wang et al., 2004; Jeong et al., 2005; Tiwari et al., 2005; Cao et al., 2010]. Zhang et al. [2009] showed that rapid startup could be successfully accomplished by using a hybrid upflow anaerobic blanket (UASB) – anaerobic fixing filter (AFF) reactor with internal hydraulic circulation and external sludge circulation. Jung et al. [2013] also adopted high rate circulation to

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accelerate the formation of hydrogen-producing granules in a UASB. Many investigators have also devoted themselves into modifying the reactor design and developing hybrid anaerobic systems for treating various types of wastewaters. Ikuo et al. [2010] reported that an expand granular sludge bed (EGSB) bioreactor, a modified UASB configuration, has the capability of treating low strength wastewaters at low temperature. Li et al. (2007) studied the performance of an integrated EGSB-Zeolite bed filtration (EGSB-ZBF) hybrid system for the removal of carbon and nutrients from low strength wastewater at 35°C for 7 month and the combined system could effectively reduce the COD concentration by 71.58%, and completely remove Ammonia and phosphate. Anaerobic Ammonium Oxidation (ANAMMOX) - EGSB combined system is a novel nitrogen removal process that achieved the nitrogen remove efficiency up to 94.68% [Chen et al., 2011]. As advanced membrane-based separations are well suited to water recycling and reuse, membrane coupled AnGSB technology (so called granular growth anaerobic membrane bioreactor G-AnMBR) is now experiencing a rapid growth as a tertiary treatment process for treating municipal wastewater as compared to suspended growth anaerobic membrane bioreactors (S-AnMBRs) [Salazar-Pelaez et al., 2011a; Salazar-Pelaez et al., 2011b; Herrera Robledo et al., 2010; Herrera-Robledo et al., 2011; Liu et al., 2013].

There are several reviews papers which focused on the specific factors affecting granulation in UASB [Abbasi and Abbasi, 2012], the applicability of UASB, EGSB and static granular bed reactor (SGBR) [Lim and Kim, 2014], the application of anaerobic hydrogen-producing granules [Li and Yu, 2013], the feasibility of granule-based anaerobic baffled bioreactor (GAnBR) [Hassan and Dahlan, 2013], and performance enhancement of upflow anaerobic sludge blanket (UASB) reactors [Chong et al. 2012], theories on anaerobic sludge granulation [Hulshoff Pol et al., 2004]. However, there is still a lack of reviews and documentation on the enhancements of G-AnMBR with collective information. With the rapid development of G-AnMBR technology, a comprehensive analysis of recent research progress would be useful.

Thus, the main objective of this review is to provide an in-depth literature review to the recent advances made in developing sustainable G-AnMBR, with emphasis on G-AnMBR performance for municipal wastewater treatment, to serve as a guideline for rapid granulation process, as well as to provide a solid platform for the development of novel G-AnMBR.

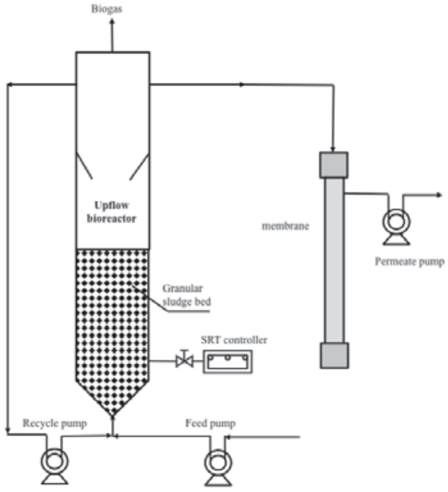
2. Granular growth anaerobic membrane bioreactor (G-AnMBR)

2.1. G-AnMBR reactors

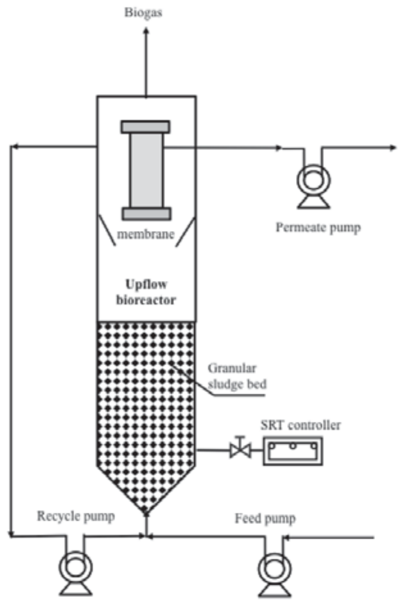
In the recent years, G-AnMBR has experienced increasing applications due to its superior performance such as high effluent quality, complete biomass retention, high biomass content, less sludge bulking problem, relatively low-rate sludge production, higher loading capacity, compact design and rapid start-up period. The G-AnMBR consists of an anaerobic granular growth bed reactor such as upflow anaerobic sludge blanket (UASB) and a membrane module. Membrane unit could be externally connected to the granule bed system as the side-stream mode (Fig. 1a), submerged in the granular sludge bed reactor (Fig. 1b) or immersed in a separated bioreactor (Fig. 1c). Polymeric membranes such as polyvinylidene fluoride (PVDF) and polyethersulfone (PES) were predominantly used in G-AnMBR mainly due to economic concerns. In regards to filtration, both microfiltration (MF) and ultrafiltration (UF) membranes are the most common ones, with membrane pores ranging from 0.4 ìm [Diez et al. 2012], 0.2 ìm [Lew et al., 2009] or 0.1 ìm [Liu et al., 2013] in the MF region to values as low as 30kDa [Gao et al., 2010] in the UF region. Hollow fiber membrane configuration gained the most popularity as compared to flat sheet (plate or frame) and tubular modes in G-AnMBR studies.

2.2. Granular growth AnMBR versus Suspended growth AnMBR

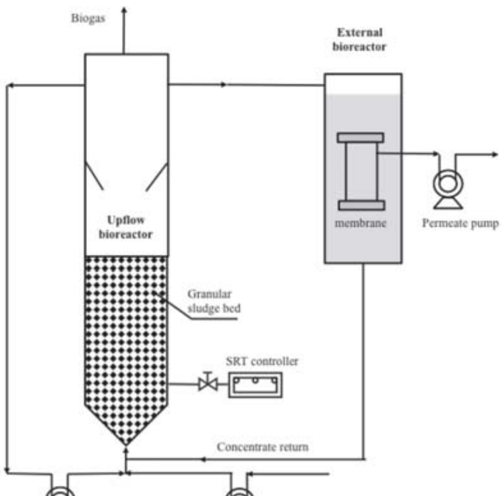
G-AnMBR offers a promising alternative approach to the traditional suspended growth AnMBR (S-AnMBR). These conventional AnMBRs generally consist of completely stirred tank reactors (CSTRs) with either internal or external membrane separation devices, and they are operated based on suspended growth pattern. AnMBRs studies by far were predominantly S-AnMBRs [Huang et al., 2011; Martinez-Sosa et al., 2011; Martinez-Sosa et al., 2012], because of the ease of use and construction. However, critical obstacles including membrane fouling, low flux and high operational costs still exist and limit the wider application of S-AnMBR. Traditional AnMBR was usually operated at a lower biomass concentration compared to high rate anaerobic reactors (HRARs)



(a) External crossflow G-AnMBR (membrane as solo polishing step)



(b) G-AnMBR with membrane place directly in the reactor (Membrane as part of G-AnMBR)



(c) SAnMBR with the membrane immersed in a separated bioreactor (Membrane as part of G-AnMBR)

Figure 1. Schematic of G-AnMBR configurations

mainly due to fouling issues, corresponding to a lower OLR (<10 kg COD/m^3/d) [Lin et al., 2013]. To maintain a well mixed flow regime and sufficient mass transfer, rigorous mechanic mixing is required, which is energy intensive. In addition, the membrane unit is directly exposed to the bulk sludge, and the high suspended solid concentration subjected to MF/UF membrane filtration worsen cake deposition and compaction in all CSTR configurations [Liao et al., 2006, Ozgun et al., 2013]. More rapid and dense cake layer build-up results in heavy membrane fouling and low flux, as under sufficient mixing, effluent solids concentration of CSTRs remains the same as the bulk solids concentration. To resolve this issue, frequent physical and chemical cleaning, interval operation, and likely sub-critical flux operation to sustain the flux should be in place. Moreover, a dramatic decrease in the sludge floc size owing to sludge recirculation through the membrane feed pump can result in sever membrane fouling (Ozgun et al., 2013), particularly in the side-stream membrane configurations. The high shear stress may also impact the biological activities of anaerobic microbes due to negatively impacted juxtapositioning of acetogens and methanogens, restricting the essential hydrogen transport for acquiring a superior specific methanogenic activity (SMA).

Table 1 Comparison of conventional aerobic treatment, anaerobic treatment, S-AnMBR and G-AnMBR

Feature aerobic treatment	Conventional anaerobic treatment	Conventional	S-AnMBR	G-AnMBR
Organic removal efficiency	High	High	High	High
Effluent quality	High	Moderate to poor	High	High
Organic loading rate	Moderate	Moderate	High	High
Sludge production	High	Low	Low	Low
Footprint	High	High	Low	Low
Biomass Retention	Low to moderate	Low	High	Excellent
Nutrient requirement	High	Low	Low	Low
Alkalinity requirement	Low	High	High to moderate	High to moderate
Energy requirement	High	Moderate to low	Moderate to low	Low
Bioenergy recovery	No	Yes	Yes	Yes
Mode of treatment	Total	Essentially pretreatment	Total or pretreatment	Total or pretreatment
Mode of Operation	Continuous	Batch/continuous	Continous	Continuous
Start-up time	2-4 weeks	2-4 months	< 2 weeks	<1 weeks or 2-4 month
Types of Wastewaters	Low to moderate	High to moderate	High to low	High to low
Membrane fouling	-	-	High to moderate	Moderate to low
Mode of Operation	Continuous	Batch/continuous	Continuous	Continuous

Table 1 represents a comparision of conventional aerobic treatment, anaerobic treatment, S-AnMBR and G-AnMBR. It is apparent that as a hybrid system, G-AnMBR combines the advantages of granular technology and MBR technology, yielding maximum joint benefits. Firstly, biomass retention is achieved by the spontaneous formation of granular sludge, and the granule bed systems are characterized by total suspended solids (TSS) concentrations ranging between 20 and 40 g/L reactor volume; whereas significantly lower effluent total solids (TS) concentration at 50 mg/L was possible [An et al., 2009] and this makes them feasible for high organic and hydraulic loadings. Since biomass is not directly exposed to a membrane module in this reactor design, less apparent dense cake layer formation and consolidation will occur in comparison with conventional S-AnMBR when coupling these reactors with a membrane module. Secondly, the natural occurring turbulence caused by the rising biogas bubbles and liquid upflow force, which buoy the granular sludge, provides sufficient substrate and microorganism contact so as mechanical mixing is no longer required and relevant operational cost would be greatly decreased [Chong et al., 2012]. In addition, less severe fouling is found in G-AnMBR configuration, thus allowing enhanced operation with reduced gas sparging demand and increased fluxes [Martin-Garcia, 2010]. Mathioudakis et al. [2012] reported that the net specific operational energy demand of a G-AnMBR based flowsheet treating 10,000 m^3d^{-1} domestic sewage was around 0.14kWhm^{-3} whereas the values for immerse AnMBR configurations could be as high as 3.57 kWhm^{-3}. The potential of the proposed GAnMBR offers high treatment efficiency with significantly reduced energy demand as compared to traditional sewage treatment. For example, for the typical specific net energy demand of a typical activated sludge plant utilizing anaerobic sludge digestion can be more than 0.6kWhm^{-3} [Martin et al., 2011]. Most importantly, the granule structure offers ideal conditions for syntrophic associations such as those between H_2-accumulating acetogenic bacteria and H_2-consuming methanogens, allowing high chemical oxygen demand (COD) removal efficiencies even under presence of toxicity or hydraulic loading events [Ozgun et al., 2013]. The compactness of the granules also renders a more compact reactor design, resulting in a much smaller footprint to apply this technology. Apart from the above-mentioned benefits, the capital cost of UASB reactors can also be reduced when coupling a membrane unit and it is by eliminating the necessity for a gas–liquid–solids (GLS) separator in a UASB [Liao et al., 2006]. However, the high sludge carry over to the effluent can occur with increased biogas production when operating in the absence of GLS. Last but not least, the granulation process and start-up period can be greatly reduced due to the membrane absolute barrier to provide the complete retention of methanogens.

2.3. G-AnMBR development

Recent studies on G-AnMBRs were predominantly applied to low strength municipal wastewater treatment rather than high strength organic industrial wastewaters such as brewery and alcohol-distillery wastewater. Municipal wastewater has long been categorized into complex wastewater due to its high fraction of particulate organic material, moderate biodegradability and its low strength. For this reason, domestic sewage treatment by anaerobic means is still challenging due to the kinetic limitations of anaerobic metabolism. Low substrate affinity of anaerobic biomass compared to aerobic bacteria and the rate-limiting step of hydrolysis of particulate matter into dissolved molecules particularly under low temperature (<20 °C) conditions have made it hardly practical to achieve low effluent chemical oxygen demand (COD) concentrations and to fulfill more stringent legislation for wastewater reclamation and reuse [Ozgun et al., 2013; Lin et al., 2013]. G-AnMBRs have offered the most sustainable options of municipal wastewater treatment and reuse essential in all the land-scarce, water-short and energy-poor countries. **Table 2** presents a number of studies that have used G-AnMBRs for treating domestic wastewater under various operating conditions. G-AnMBRs have typically shown high COD/TOC removal efficiencies with 77%-97% and this high efficiency could be sustained even at psychrophilic temperature and high hydraulic loading rate.

2.3.1. Membrane as solo polishing step

A pilot scale UASB reactor followed by a post-treatment external UF membrane operating at different HRTs (12-4 h) was able to achieve permeate free of total suspended solids (TSS), and with COD concentration less than 120 mg/L for the treatment of real domestic wastewater with a high variability in its characteristics [Salazar-Pelaez et al., 2011a]. In spite of high variations in COD, total solids (TS), volatile total solids (VTS) and total suspended solids (TSS) concentrations by sediments washout from pipelines in the rainy seasons, the combined system consistently produced permeate fulfilling the Mexican standards established for wastewater reclamation in public services at all times. Similarly, such a system of lab scale yielded a slightly better total COD removal (89 and 82% respectively at HRT of 8 and 12h) than that of a UASB reactor alone at the steady state in treating synthetic wastewater with average COD concentration at 350 mg/L at ambient temperature [Salazar-Pelaez et al., 2011b]. However, as suggested by the authors, the transition to lower HRT (12 to 4 h) deteriorated the AnMBR performance by inducing a higher production of soluble microbial products (SMP) and extracellular polymeric substances (EPS) and particle release in UASB

Table 2 Summary of G-AnMBR performance for municipal wastewater treatment

Type of wastewater	Innoculum	Scale	Volume (L)	Reactor configuration	Characteristics of membrane	Operating condition	Removal efficiency (%)	Reference
Domestic wastewater (COD=100-2600 mg/L)	Digested sludge	L	17.7	Hybrid upflow anaerobic bioreactor + submerged membrane	PE Hollow fibre UF pore size: 0.03µm Surface area: 0.3 m^2	OLR= 0.5 - 12.5 kg COD/m^3/d HRT=6 and 4 h Ambient temperature SRT=150 d Flux= 5 LMH MLSS=16-22.5 g/L	COD= 97%	Wen et al. (1999)
Synthetic wastewater (COD=383-849 mg/L)	Granular sludge	L	4.7	EGSB with submerged membrane	PE Hollow fibre MF pore size: 0.1µm Surface area: 0.1 m^2	UV^b= 2-8 m/h OLR= 1.6- 4.5 kg COD/m^3/d HRT=3.5, 4.6 and 5.7 h Temp= 25, 20 and 15°C	COD= 85-96%	Chu et al. (2005)
Raw municipal wastewater (COD=58-348 mg/L)	Digested sludge	P	34	UASB+ external membrane	Polyacrlonitrile Module 1: ID/OD of 1.2/2.1 mm, 0.2m^2 Module 2: ID/OD of 1.9/2.9 mm, 0.2 m^2 Module 3: ID/OD of 3.0/3.9 mm, 0.2 m^2	OLR= 0.3-0.9 kg OD/m^3/d Temp= 27-30 ° CCHRT= 5.5-10 h MLSS=12-32 g/L SRT=α	COD= 77-81%	An et al. (2009)
Pre-settled domestic wastewater (COD=540 mg/L)	Granular sludge	L	180	Upflow anaerobic bioreactor + External membrane	PVDF Hollow fibre MF Pore size: 0.20µm Surface area: 4 m^2	OLR= 1.08, 2.16, 4.32 kg COD/m^3/d Temp= 25 °C SRT=α HRT= 12, 6 and 4.5 h Flux=3.75, 7.5, 11.25 LMHMLSS=14-80 g/L	COD> 88%	Lew et al. (2009)
Synthetic wastewater (COD=500 mg/L)	Granular sludge	L	10	UASB + External membrane	PVDF and PEI Flat sheetUF, 100 kDa MWCO and 30 kDa MWCO Surface area: 0.052 m^2	OLR= 5 kg COD/m^3/d Temp= 30 °C HRT= 24 h MLSS=12-32 g/L SRT= 50 d	COD= 96%	Gao et al. (2010)
Real municipal wastewter (COD=646 mg/L)	-	L	3.5	UASB+ external membrane	TubularUF, 40 kDa MWCO Surface area: 81 cm^2	Temp= 20-25 °C HRT= 3 hSRT= 60, 100 d	COD= 86-87%	Herrera-Robledo et al. (2010)
Raw sewage (COD=445 ± 138mg/L)	-	L	849	UASB+ external membrane	PVDF Tubular UF, 100 kDa MWCO Surface area: 5.10 m^2	Uv=0.64 m/h Temp=22±3°C Flux=6LMHHRT=6 h SRT=180 d	COD= 93%	Herrera-Robledo et al. (2011)
Synthetic wastewater (COD=350±10 mg/L)	Granular sludge	L	12.5	UASB+ external membrane	PVDF Tubular UF, 100 kDa MWCO	UV^b= 0.122, 0.061 and 0.041 m/h Temp= 27-30 ° CHRT= 4, 8 and 12 hpH= 7, Flux=5 LMH	COD= 81-89%	Salazar-Pelaez et al. (2011b)
Real domestic wastewater (COD=285-2,088 mg/L)	Granular sludge	P	700	UASB + External membrane	PVDF TubularUF, 100 kDa MWCO Surface area: 0.84 m^2	SRT=150 dAmbient temperature HRT= 4, 8, 12 h	COD= 86-96%	Salazar-Pelaez et al. (2011a)
Settled primary wastewater (COD=338±74 mg/L)	Granular sludge	P	125	Upflow anaerobic tank+ external membrane	PVDF Hollow fibre UF pore size: 0.08µm Surface area: 0.93 m^2	Uv=0.7-1 m/hTemp=8-22°C Flux=6LMHHRT=16h	COD= 84-86%	Garcia et al. (2013)
Synthetic wastewater (COD=500±10 mg/L)	Granular sludge	L	10	UASB with submerged membrane	PVDF Hollow fibre MF pore size: 0.1µm Surface area: 1 m^2	UV^b= 2.5m/hOLR= 1 and 6 kg COD/m^3/dTemp= 27-30 ° CHRT= 2 and 12 hFlux=5 LM HMLSS= 1.6 and 10.1 g/L	COD= 97%	Liu et al.(2012) Liu et al.(2013)
Synthetic wastewater (530±40mg/L)	Flocculent anaerobic sludge	L	7	UASB+ external MBR	PES TubularUF pore size: 30nm Surface area: 0.0038 m^2	UV^b= 0.6 m/hOLR= 2 kg COD/m^3/dTemp= 25 °C HRT= 6 h Flux= 12.3LMH	COD= 92%	Ozgun et al. (2015)

effluent, thus eventually increased COD and solids concentrations in both, UASB effluent and permeate (total COD removal 81% vs 89% in the case of Salazar-Pelaez et al., 2011b). The authors attributed declined removal efficiency to the following reasons. Firstly, the operation of UASB reactor at a low HRT consequently resulted in higher upflow velocity and OLR, therefore increasing shear forces inside the reactor and biogas production. Both facts caused solids washout as well as particle disaggregation and stress in microorganisms and promote biopolymer release in its effluent [Wang et al., 2009]. Secondly, the lower removal efficiency was mainly due to the limited contact time between microorganisms and substrate for the physical and biological processes at low HRTs. The last but not least, the system was incapable of retaining UASB sludge, and therefore had the decreased filtration capacity of the sludge bed at higher upflow velocities [Leitão et al., 2005]. The enhanced release of biopolymeric substances in the effluent at lowest HRT also worsened the fouling propensity in the UASB effluent by increasing the fouling rate and the specific cake resistance and decreasing particle sizes. Therefore, both studies recommended to avoid operating AnMBR (UASB + external UF arrangement) at HRTs lower that 4 hours in order to control SMP and EPS fouling potential, and maintain better COD removal performance.

On the other hand, An et al. [2009] showed that with gradually decreasing HRT from 10 to 5.5 h, the application of membrane filtration as a polishing unit of UASB effluent could achieve better TOC removal efficiency from 80% to 85%, and much higher biogas yield from 61.8 to 120.7 mL/g $COD_{removed}$ in treating raw municipal wastewater at ambient temperature in Singapore. Such a behavior at lower HRT was attributed to the increased upflow velocity which improved water distribution and provided more even contact between substrate and microorganism, and higher sludge loading rate in a low-strength wastewater treatment system, which favored the anaerobic microorganisms activity. Their investigation also indicated that periodic 20 seconds backwash every 10 min suction presented the best result to reduce fouling and membrane cleaning frequency and prolonged membrane longevity.

Herrera-Robledo et al. [2010] showed that with HRTs (3h) more than three times lower than those in full-scale UASB applications for municipal wastewater treatment, the operation of UASB+MBR process at ambient temperature in southern Mexico City was still feasible. They compared the performance of this combined system operated at two different SRTs (100 and 60 d) at local temperatures of 20-25°C, and found there was no difference on COD removal efficiency (COD_t and COD_s removal efficiencies of over 85% and 73% were achieved in the parallel systems, compared to 50% in the UASB reactors alone, suggesting that such a hybrid system operated at relatively low HRT and SRT is viable at the Mexican climate conditions. Their investigation also indicated that longer SRTs operation during long-term operation of 500 h resulted in more repetitive sudden TMP and flux changes which might be explained by a fouling lay collapse and compression hypothesis for cross-flow membrane ultrafiltration, suggesting a stronger fouling layer structure. Furthermore, Herrera-Robledo et al. [2011] demonstrated the efficacy of a high rate UASB-MBR for raw sewage treatment at HRT and SRT of 6 h and 180 d in producing an effluent free of suspended solids, pathogens (fecal coliforms) and parasite ova, with 93% COD removal and 73% phosphorous reduction (sorption by the biofouling or even chemical precipitation through biomineralization), that met official Mexican regulation for direct urban water reclamation. Based on fouling analysis, SMP with size lower than membrane pores (89 mg/L) tended to absorb on membrane surface or inside the pores, and were considered significant for fouling development. It was also suggested that a mild cleaning procedure using chlorine (NaClO at 300 mg/L, for 30 min) accomplished a limited removal of fouling mass per unit area (13%), and the biofouling remnants was partly resulted from biologically-induced mineralization materials that were synthesized (massive EPS secretion from colonizing cells) in response to cleaning procedure, and may be the basis of irreversible membrane fouling.

The studies mentioned above have used the membrane unit as a solo polishing step after the UASB reactor. In such a configuration, the concentrate streams were not recycled back to the bioreactors, and therefore the hydraulics and biogranulation in the UASB reactors remained undisturbed from the membrane incorporation, resulting in continuous selection of stable granules with good settling properties for efficient anaerobic digestion. Nevertheless, Ozgun et al. [2015] elucidated that the addition of membrane as an absolute barrier could cause a detrimental effect on the suspended solids (SS) accumulation in the membrane tank situated after UASB reactors, contributing to an intensifying increase in the SS loading on the membrane unit and subsequent high tendency to foul.

2.3.2. Membrane as part of G-AnMBR

A great number of studies have suggested employing membranes as an element of a G-AnMBR system to provide nearly absolute biomass retention and allow for operation at high SRTs via concentrate flow recycle to the granular reactor, thus leading to the enhanced reactor performance subject to psychrophilic methanogenesis, high loading events, loading shocks and climate temperature fluctuations [Chu et al., 2005; Liu et al., 2012; Liu et al., 2013]. Membranes in these systems are either located as an external side-stream [Lew et al., 2009; Garcia et al., 2013; Ozgun et al., 2015] in which the concentrate is recycled back to the granular bioreactor or submerged at the top of the reactor [Chu et al., 2005; Liu et al., 2013; Wen et al., 1999]. In such a setup, the membrane is not simply regarded as a physical barrier, but also facilitates a general cultural adaptation to the prevailing loading conditions of the reactor environment.

Liu et al. [2012] utilized a granule based AnMBR system in which a MF hollow fiber membrane was immersed in the expanded section to evaluate the impact of food-to-microorganisms (F/M) ratio on the system performance and fouling treating low strength wastewater at 27-30°C. They reported impressive reclamation with the TOC removal efficiency of more than 96% in the high load AnMBR. However, they observed more severe fouling in the high load G-AnMBR as compared to the low load system, with cake resistance responsible for over 98% of the total fouling in both systems. This higher cake resistance was attributed to higher amounts of SMP and higher tightly-bound to loosely-bound EPS ratio in the cake layer whereas the greater amount of fine particles in the high loading system was also responsible for more serious fouling. The authors pointed out that membrane filtration deteriorate sludge bioflocculation, which in turn exacerbated membrane fouling, and a lower F/M ratio and strategies for cake layer elimination were preferred for the long term sustainable operation of AnMBRs. Thereafter, Liu et al. [2013] used the same system to evaluate specific enhancement of the UASB performance by incorporating submerged membrane to the overloaded reactor at mesophilic temperatures, and obtained similar TOC removal of 97% at HRT of 2 h and specific organic loading rate (SOLR) of 3.8 kg COD/kg MLSS/d. Linear increased TOC removal from 55% to 91% by bulk sludge mainly accounted for the enhanced performance, implying that submerged membrane addition overcame the upper SOLR limit in the UASB reactor treating low strength wastewater and significantly enhanced biological activity of the suspended sludge when encountering the high loading events. One of the major advantages of this system was the membrane retention of biomass with sufficiently high SMA and an especially beneficial and diverse microbial community structure predominated by Methanothrix-like bamboo shaped rods when the loading rate is too high for sufficient sludge retention and when the high quality effluent is required for reclamation. The author also suggested that the enhancement by the membrane incorporation to the UASB was featured by two phenomenal behaviors: (1) the comparatively higher removal by the membrane at the early transition stage when the process was adapting to the introduced membrane, and (2) dramatic increase in the bulk removal during the adaption stage as well as gradual decrease in the membrane removal to the steady state.

On the other hand, Ozgun et al. [2015] reported that the introduction of membrane to a UASB reactor significantly affects the system in both biological and physical perspectives. Membrane incorporation induced the deterioration of sludge settleability and more frequent sludge washout, due to the decreasing of particle size distribution (PSD) caused by an accumulation of fine particles and decreasing EPS, thus causing a resultant increase in COD and total suspended solids (TSS) and SMP concentrations in the UASB effluent. The authors justified that, despite the SMA and stability of the UASB sludge deteriorated after membrane incorporation, the increase in microbial community index in both richness and evenness was found, and hence the enhancement in overall system performance was observed in terms of higher COD removal efficiency of 92% (72% COD reduction before membrane addition) and more methane production at HRT of 6 h, due to the complete retention of all particulate and colloidal matter and biomass inside the reactor by the membrane. In addition, the stable transmembrane pressure (TMP) was observed at 85 mbar in average during AnMBR operation, indicating no severe membrane fouling propensity was encountered.

Lew et al. [2009] on the other hand, suggested that at temperature of 25°C, using an HRT and OLR ranging from 4.5 to 12 h, and 1.08 to 4.32 kg $COD/m^3/d$, an innovative external G-AnMBR, in which the traditional cross-flow external membrane unit was replaced by a microfiltration, hollow fiber, dead end external unit placed below the bioreactor effluent exit, would provide the most energy efficient measure of municipal wastewater treatment than other AnMBR configurations, due to the enough transmembrane pressure provided by the height difference between the bioreactor and the membrane which allowed no pump used for recirculation or transmembrane pressure enhancement, promoting energy savings. Furthermore, as suggested by the authors, intermittent backwash was adopted for fouling amelioration instead of gas bubbling scouring most often found in other studies [Garcia et al., 2013; Huang et al., 2011; Lin et al., 2009], and the best backwash frequency for energy savings and fouling mitigation of 30-60 min. EPS was found as the fouling agent during slow linear increase of fouling rate, according to the observation of the sulfate and aliphatic accumulation on the membrane.

In order to widely apply G-AnMBR technology, anaerobic reclamation of low strength municipal sewage should be maintained under ambient temperature (7-20°C) due to the excessive energy cost for heating [Martin et al., 2011]. However, problems still remain in terms of psychophilic methane fermentation. For instance, the anaerobic process requires a long start-up period, its performance may be unstable, and the kind of wastewater that can be treated is limited due to the activity of methanogen [Ikuo et al., 2010]. Lettinga et al. [2001] suggested that AnMBR operation at psychrophilic temperature is technically feasible, although sludge retention time (SRT) must be maintained twice those commonly applied in mesophilic operation, causing SRTs of 120-160 d. Nevertheless, in light of the limitation of anaerobic metabolism below 20°C, only partial solid hydrolysis and incomplete digestion of volatile fatty acids into methane are achieved, resulting in the increase in colloidal and soluble solids content in anaerobic effluents and on membrane fouling propensity [Kashyap et al. 2003]. Garcia et al. (2013) compared the treatment efficiency and membrane performance of a granular and suspended growth anaerobic membrane bioreactor (G-AnMBR and S-AnMBR respectively) for 250 days treating settled sewage under UK weather conditions, and concluded the impact of configuration was negligible with COD and BOD removal of 80-95% and >90%. As temperature dropped from 20°C to 10°C, COD removal efficiency experienced noticeable reduction from 97% to 78%, due to the production of non-biodegradable organics rather than the accumulation of VFAs at lower temperatures. This study also confirmed the lower fouling potential in the G-AnMBR as compared to the S-AnMBR, due to the reduced solid and colloidal load (by a factor of 10 and 3) to the membrane which was allowed by the enhanced interception of solids in the granule bed of the G-AnMBR as a result of mixed liquid recycle from the membrane tank to the bioreactor at a low upflow velocity. Similarly, van Voorthuizen et al. [2008] reported the much severer fouling in an anaerobic MBR as compared to an UASB coupled to membrane unit due to the accumulation of higher amount of colloidal matter. They suggested the reduced colloidal matter was mainly due to G-AnMBR biodegradation of dissolved organics taken place predominately within the granules, and colloidal particles arising from the influent solids [Lant and Hartley, 2007] physically adsorbed and retained in the granule bed protecting the membrane from their influence on fouling. Therefore, the granular system would require lower gas sparging intensity and lower energy requirements for fouling control [van Voorthuizen et al., 2008]. Moreover, energy efficiency could be further enhanced especially when backwashing was implemented within the granular AnMBR [Garcia et al. 2013].

Singh et al. [2006] reported that the dead space of the UASB reactor was 10%-11% and it is dependent upon the operating temperature, which meant the smaller volume of mixing zone the more by-pass flow can occur in the reactor when low temperature is applied. Therefore, UASB reactors treating municipal wastewater at low and moderate temperatures are sometimes characterized by a poor mixing regime, which causes a decrease in soluble COD treatment efficiency. To resolve this issue, tall reactors with a higher ratio of height to width and external or internal effluent recirculation, so-called EGSB reactors, are increasingly applied to provide a very high mixing intensity and efficient wastewater-biomass contact induced by the high upflow velocity. In the study of Chu et al., [2005], a U shape hollow fiber membrane submerged in the upper part of EGSB, was utilized for treating domestic wastewater during 7-month period in the range 11–25°C, and at HRT of 3.5 to 5.7 h. At temperatures above 15°C, the combined system had the capability of removing 85–96% of total COD and 83–94% of TOC despite HRT variations. However, at 11°C, increasing HRT from 3.5 to 5.7 h contributed to the enhanced COD removal from 76 to 81%, which indicated the significance of HRT for low strength wastewater treatment at lower temperature. Upflow velocity, as the other important parameter governing hydraulic mixing, was found significant in achieving better effluent removal efficiency, and a higher membrane permeability due to a rinsing effect on the membrane at the low temperature. Nevertheless, the granule segregation was also induced by the high upward velocity, which was found from granule size distribution andd SMA test along the sludge bed. This study showed that psychrophilic sewage treatment using EGSB reactor coupled with membrane technology is feasible, due to the intensified mass transfer between substrate and microbes and viable retention of granule sludge comprised predominantly of filamentous Methanothrix-like species.

Although many studies have proved the competitive advantages of such G-AnMBRs configurations in municipal wastewater treatment, hydraulic selection pressure required for granulation is minimized by the membrane barrier in these cases, through the avoidance of the washout of flocculent sludge with poor immobilization characteristics [Ozgun et al., 2013], thus resulting in a sludge bed with poor settling properties in granule reactors in the long term. Furthermore, the high fraction of partly degradable particulate matter in domestic wastewater, which can be entrapped and gradually accumulated in the granular sludge bed, may further impede the applicability of granular reactors in an G-AnMBR system configuration in the long-term operation.

3. Conclusion

G-AnMBR can be seen as a promising technology for low strength wastewater treatment (e.g. municipal wastewater) compared to traditional S-AnMBR due to its better treatment efficiency with significantly low energy consumption. G-AnMBR has superior quality of granular sludge, which leads to less fouling propensity, enhanced operation with no mechanical mixing, reduced gas sparging demand, and increased membrane flux. G-AnMBR has strong potential to cope with high hydraulic loading events and high organic loading events despite the need of further investigation on granular deterioration. In light of the inherent eco-friendly nature of psychrophilic G-AnMBR without heating, and so as to overcome the temperature constraints on anaerobic bioprocesses, further studies are necessary to investigate microbial community structure of granules at lower temperature and their influence on process stability towards accomplish kinetic perspective-based improvement of biogas production.

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