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# Anaerobic membrane bioreactors for antibiotic wastewater treatment: performance and membrane fouling issues

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#### **Abstract**

Antibiotic wastewater has become a major concern due to the toxicity and recalcitrance of antibiotics. Anaerobic membrane bioreactors (AnMBRs) are considered alternative technology for treating antibiotic wastewater because of their advantages over the conventional anaerobic processes and aerobic MBRs. However, membrane fouling remains the most challenging issue in the AnMBRs' operation and this limits their application. This review critically discusses: (i) antibiotics removal and antibiotic resistance genes (ARGs) in different types of AnMBRs and the impact of antibiotics on membrane fouling and (ii) the integrated AnMBRs systems for fouling control and removal of antibiotics. The presence of antibiotics in AnMBRs could aggravate membrane fouling by influencing fouling-related factors (i.e., sludge particle size, extracellular polymeric substances (EPS), soluble microbial products (SMP), and fouling-related microbial communities). Conclusively, integrated AnMBR systems can be a practical technology for antibiotic wastewater treatment.

Keynotes: Anaerobic membrane bioreactors, antibiotic wastewater treatment, antibiotic resistance genes, membrane fouling

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#### 1. Introduction

Antibiotics are widely used to treat or prevent human and animal diseases, and promote livestock growth. Such behaviour results in high levels of antibiotic residues in municipal wastewater, livestock wastewater and other industrial effluents (Li, 2014, Sabri et al., 2018). It is widely known that the occurrence of antibiotics in the environment could cause serious risks to environmental security and public health due to the emergence and transfer of antibiotic resistance genes (ARGs) and bacteria (ARB) (Berendonk et al., 2015, Sharma et al., 2016). Anaerobic biological treatment is an advisable option for treating high-strength wastewater containing antibiotics in comparison with aerobic treatment. The former treatment method has certain advantages, for example, biogas production, less waste sludge production and lower energy costs (Angelidaki et al., 2003, Chen et al., 2011, Cheng et al., 2018b). Conversely, biodegradation of antibiotics in conventional anaerobic digestion processes - even other modern high-rate anaerobic reactors like up-flow anaerobic sludge blanket (UASB) and anaerobic sequencing batch reactor (ASBR) is limited. This is because the biosorption of antibiotics onto sludge results in high levels of antibiotics and ARGs being released into the environment (Cheng et al., 2018b). The persistence of ARGs through wastewater treatment processes not only presents a serious ecological risk in natural environments, but also has a negative influence on public perceptions of wastewater reuse and its economic viability (Zhu et al., 2018).

Anaerobic membrane bioreactors (AnMBRs) are an available alternative for the treatment of wastewater containing antibiotics owing to their advantages over conventional anaerobic processes, which include a high degradation capacity of anaerobic microorganisms, longer SRT, and better effluent qualities (Cheng et al., 2018b, Ozgun et al., 2013). Previous reports have indicated that AnMBRs are promising

technologies for degrading common organic pollutants and emerging antibiotics in wastewater under optimal conditions. The removal efficiency of COD, tetrahydrofuran dimethylformamide, m-Cresol and isopropyl alcohol in AnMBR systems was more than 90 % (Chen et al., 2018, Hu et al., 2017a, Hu et al., 2017b, Xiao et al., 2017). The membrane of AnMBRs plays a significant role in preventing the escape of antibiotics and microbes with ARGs from the reactor into the environment (Munir et al., 2011).

Nonetheless, the widespread application of AnMBR in wastewater treatment is still restricted by membrane fouling problems. Fouling of the membrane decreases permeate flux and in fact the membrane's lifespan, and this leads to higher operating costs in regards to energy requirements in order to reduce the fouling and membrane replacement (Lin et al., 2011b, Meng et al., 2009). As reported by Pretel et al. (2014), 85-90% of the energy consumption in AnMBR was related to the filtration and membrane fouling control processes. In both aerobic membrane bioreactors (MBRs) and AnMBRs, membrane fouling is caused by the undesirable deposition and accumulation of microorganisms, colloids, solutes, and cell debris in the pores and on the surface of the membrane (Guo et al., 2012, Le-Clech et al., 2006, Lin et al., 2013). Although the same membrane module is used in aerobic MBR and AnMBR systems, the latter system usually encounters more severe membrane fouling problems. Not only are the higher biomass concentrations and longer biomass retention times required in the AnMBRs, they work at lower membrane fluxes than the aerobic MBRs as well (Lin et al., 2013). For example, as reported by Di Bella et al. (2007), the membrane foulants in the AnMBRs are more difficult to remove than that in the aerobic compartment because of the different sludge properties. Lin et al. (2011c) found that the cake thickness in a submerged AnMBR was much higher than was reported in the aerobic MBR systems.

Moreover, the presence of antibiotics in AnMBRs could accelerate the membrane fouling rate and shorten the membrane fouling cycle due to the effect of antibiotics on anaerobic sludge and the microbial communities in AnMBRs (Li et al., 2017, Zhu et al., 2018). For example, Zhu et al. (2018) indicated that the membrane fouling cycle decreased from 25 days to 8 days with the addition of sulfamethoxazole (SMX) and tetracycline (TC) each at 100 μg/L, and further decreased to 4 days when the concentration of SMX and TC rose to 1000 μg/L in the reactor. As similar results have been confirmed by Li et al. (2017), the membrane fouling cycle was obviously short due to the presence of antibiotics (50 mg/L benzothiazole)in the feed wastewater in an integrated anaerobic fluidized-bed membrane bioreactor (AFMBR). In addition, the membrane fouling layer became denser and more compact as the level of antibiotic concentrations increased (Zhu et al., 2018). Therefore, higher concentrations of antibiotics in the AnMBRs possibly result in higher operation and maintenance costs.

In recent years, in order to optimize the performance of AnMBRs for treating antibiotic contaminated wastewater, state-of-the-art technologies have been proposed to remove antibiotics and mitigate membrane fouling simultaneously. The combining of biofilm with conventional AnMBRs has been considered a promising technology to enhance the removal efficiency of antibiotics and reduce membrane fouling. Since the carriers (GAC, PAC, and Sponge) in AnMBRs not only can provide a large surface space for microbial growth, they can also reduce the suspended particles in the reactor (Aun Ng et al., 2006, Chen et al., 2017a, Dutta et al., 2014, Kim et al., 2010). Additionally, bioelectrochemical systems (BES) represent a new and promising technology for wastewater treatment, which is capable of converting organic matter into electricity (microbial fuel cells, MFCs) or hydrogen (microbial electrolysis cells, MECs) via exoelectrogenic microorganisms (Logan & Rabaey, 2012). The integration of BES with

AnMBRs is seen as an alternative technology that can improve antibiotic wastewater treatment, recover energy and reduce membrane fouling (Katuri et al., 2014, Su et al., 2013, Werner et al., 2016).

Currently, antibiotics and ARGs are deemed emerging environmental contaminants. Their occurrence and transfer in the environment pose a great risk to human health and eco-environmental security. The aim of this review is to summarize the performance of AnMBRs for removing antibiotics and ARGs from wastewater in published literature. The influence of antibiotics on membrane fouling in the AnMBRs is critically discussed through the effects of antibiotics on various fouling-related factors, such as sludge particle size, extracellular polymeric substances (EPS), soluble microbial products (SMP), and microbial communities. Finally, new hybrid AnMBR systems for membrane fouling control and antibiotic removal are also introduced in this review. Therefore, this review can help to identify the removal of antibiotics and ARGs in AnMBRs to understand the effects of antibiotics on membrane fouling, and to develop alternative technologies for improving the AnMBRs performance with minimal membrane fouling.

#### 2. Removal of antibiotics and ARGs in AnMBRs

AnMBRs are the combination of membrane separation technology and anaerobic biological wastewater treatment processes. The advantage of anaerobic processes and aerobic membrane bioreactors (MBRs) is that they generate solid-free effluent, total biomass retention, low sludge production and net energy production (Lin et al., 2013). The removal of COD and antibiotics (sulfonamides, macrolides, β-lactams, trimethoprim, etc.) from wastewater by AnMBRs has been summarized Table 1. Biodegradation is the dominant removal mechanism of antibiotics in AnMBRs, although adsorption onto the sludge is the major contributing factor for their initial removal (Harb et al., 2016, Xiao et

al., 2017). Some previous research indicated that the hydrophobicity and molecular features (i.e., functional groups and the presence of nitrogen/sulphur) of micropollutants have a significant relationship with their fate in AnMBRs processes (Wei et al., 2016, Wijekoon et al., 2015). They concluded that hydrophobic antibiotics and the one containing electron donating functional groups (EDG) have high biodegradability in the AnMBRs. The antibiotic like sulfamethoxazole (SMX), which contains both a strong EDG and a strong electron withdrawing group (EWG), the biodegradability might depend on the relative strength of their electron donating and withdrawing capability (Wei et al., 2016).

In the individual AnMBR system, the removal efficiency of SMX and trimethoprim under different operating conditions was 67.8–99.6% and 35.4–97.5%, respectively (Cheng & Hong, 2017, Monsalvo et al., 2014, Wijekoon et al., 2015, Xiao et al., 2017). By contrast, the removal efficiency of β-lactams antibiotics from pharmaceutical wastewater by the AnMBR is relatively low (34.6-79.4%), which might be due to their high initial concentrations in the reactor which inhibited the activity of anaerobic microorganisms (Cheng et al., 2018a, Huang et al., 2018). As shown in Table 1, the integrated AnMBR processes performed better than AnMBR alone for removing antibiotics from wastewater. Specifically, the total removal of micropollutants in the combined AnMBR with nanofiltration membrane (AnMBR-NF) system was better than their removal in the individual AnMBR system, with the removal of SMX being above 98%. In this system, NF played an important role for the removal of micropollutants from wastewater with the average being 87% for all micropollutants (Wei et al., 2016). Liu et al. (2014) and Shah et al. (2012), who indicated that the rejection of NF resulted in a long retention time of antibiotics in the bioreactor to enhance antibiotics removal.

Some previous reports indicate that the addition of powdered activated carbon (PAC) and granular activated carbon (GAC) in AnMBRs was able to enhance the removal of antibiotics by improving their biotransformation (Dutta et al., 2014, Xiao et al., 2017). For example, the removal efficiency of SMX and trimethoprim in the AnMBR with PAC was more than 99% in comparison with  $67.8 \pm 13.9\%$  and  $94.2 \pm 5.5\%$  in the AnMBR without PAC under the same operating conditions. The enhancement of their removal in PAC-AnMBR is perhaps due to their adsorption onto PAC which increased the substrate concentration locally around the adsorptive sites, thus making the biotransformation more thermodynamically favorable (Xiao et al., 2017). In addition, the superior removal of SMX (88-100%) in a staged anaerobic fluidized membrane bioreactor (SAF-MBR) observed, which is attributed to the biodegradation and sorption of GAC and the associated biofilm. McCurry et al. (2014) and Dutta et al. (2014) have demonstrated that a wide range of antibiotics was effectively removed (86–100%) in a two-stage anaerobic fluidized membrane bioreactor (AFMBR) with GAC as the carrier medium in both stages. The combined effects of the biodegradation, sorption onto GAC, and membrane filtration in the AFMBR were responsible for the elimination of antibiotics from wastewater. Without the addition of carriers, AFMBR has shown efficient removal of benzothiazole antibiotic (50 mg/L) with the removal efficiency of  $82.3 \pm 3.7\% - 97.6 \pm 0.5\%$ , but the membrane fouling cycle was relatively short (Li et al., 2018).

Furthermore, anaerobic treatment processes especially in the AnMBRs are efficient for controlling the increased risk of ARGs. For instance, Harb et al. (2016) reported that the average and peak relative abundance of sul1, sul2, intl1, and dfrA5 genes were over one order of magnitude lower in an anaerobic reactor when compared to that in an aerobic reactor. Cheng and Hong (2017) indicated that the AnMBR system is promising for the removal of ARGs (at least  $1.9 - 3.9 \log unit$ ), which correlated positively with the

extent of membrane fouling. The removal of these ARGs (sul1, sul2, tetC, tetX and ereA) and int1 in an anoxic/aerobic membrane bioreactor (A/O-MBR) was stable with the abundance of 0.6 - 5.6 orders of magnitude. This was despite the addition of SMX and tetracycline hydrochloride in the reactor, which increased the ARG abundances by 0.5-1.4 orders of magnitude (Zhu et al., 2018). In the A/O-MBR system, the distribution of ARGs in membrane foulants accounted for 13%–25% of the total absolute abundance. Therefore, it can be expected that fouling layers on the membrane surface contribute to the removal of antibiotics and ARGs from wastewater (Monsalvo et al., 2014, Stewart & Costerton, 2001, Zhu et al., 2018). Two possible reasons have been given for the rejection of antibiotics and the ARGs by membrane foulants. Firstly, the effective pore size of the membrane could be reduced by appropriate membrane fouling through pore blocking as this helps with the retention of smaller size antibiotics and ARGs (Zhu et al., 2018). Secondly, as the principle components of membrane fouling layers, extracellular polymeric substance (EPS) and soluble microbial product (SMP) exhibited significant correlations with the removal of antibiotics and ARGs in the AnMBRs (Cheng & Hong, 2017, Zhu et al., 2018).

These EPS and SMP in the AnMBRs carry charged functional groups (i.e., carboxyl, hydroxyl and phosphoric groups) and possess both structural features of the hydrophilic and hydrophobic sites on their structure. This feature enables them to have cross-linking structure properties, which may contribute to the separation and retention ability of the membrane foulants (Lin et al., 2013, Sheng et al., 2010). In addition, the protein component of EPS and SMP with amino acid compositions and secondary structures may perhaps contribute to their adsorption abilities regarding antibiotics as well (Hou et al., 2015, Xu et al., 2013).

#### 3. Influences of antibiotics on membrane fouling-related factors

Membrane fouling is a complex problem, and the classification and mechanism of fouling in MBRs have been comprehensively reviewed by published literature (Aslam et al., 2018b, Bagheri & Mirbagheri, 2018, Liao et al., 2006, Lin et al., 2013). Different foulant materials such as suspended solids, colloidal materials, attached cells, as well as extracellular carbohydrates, proteins, lipids, and nucleic acids in EPS and SMP can deposit on the membrane surface and/or inside membrane pores, thus reducing flux and increasing Trans Membrane Pressure (TMP) eventually (Gao et al., 2011, Smith et al., 2012). In AnMBR systems, biofouling is the major problem although all forms of fouling occur simultaneously (Flemming et al., 1997, Lin et al., 2009). Biofouling in the AnMBRs is the accumulation of biomaterials on the membrane surface, which is caused by pore clogging, as well as the accumulation of microorganisms and EPS/SMP on the membrane surface (Lin et al., 2013). Illustrated in Fig. 1 are the anaerobic sludge properties including the floc size and production of SMP and EPS, which play an important role in the formation of biofouling (Dvořák et al., 2016, Lin et al., 2009). Additionally, microbial communities and their distributions in the AnMBRs wield significant effects on membrane fouling (Wu & Fane, 2012). Hence, understanding influences of antibiotics on anaerobic sludge properties and microorganisms in AnMBRs is essential to understanding the effects of antibiotics on membrane fouling in AnMBRs.

#### 3.1 Influences of antibiotics on anaerobic sludge properties

The effect of antibiotics on anaerobic sludge properties (particle size, EPS, SMP, etc.) could give valuable information on how antibiotics affect membrane fouling.

Generally, membrane fouling is caused from the initial pore blocking followed by cake formation. The sludge flocs size that is close to or smaller than the size of the membrane pores will penetrate into and block the membrane pores easily (Gao et al., 2011).

Previous studies have concluded that flocs with smaller sized pores contribute more to fouling than larger ones (Lin et al., 2011a, Lin et al., 2009). The formation rate of cake layer correlated with the fraction of smaller sized particles (Lin et al., 2010). One possible reason is that small flocs had a strong tendency to deposit on the membrane surface due to their low back transport force and the compaction of the cake layer.

Another reason is that the smaller flocs have a higher density than the larger flocs with more bridging between biopolymers (Gao et al., 2011, Jeison & van Lier, 2007, Lin et al., 2010).

It is reported that the instabilities such as exposure to toxic conditions, sudden organic load, temperature and pH changes, may cause floc breakage and result in the decrease of particle size in the AnMBRs (Shen et al., 2015). In response to cytostatic drugs presence in an anaerobic osmotic membrane bioreactor, the mean floc size of sludge decreased from 92 to 80 µm leading to higher layer formation rate and membrane fouling (Wang et al., 2018). Aubenneau et al. (2010) and Meng et al. (2015a) also observed the decrease of floc size after the addition of carbamazepine and fluoroquinolone antibiotics in MBRs and further lead to high membrane fouling. As well, the pH in the anaerobic reactor was sensitive to the presence of antibiotics, which decreased sharply after the addition of high concentration of antibiotics in the reactor, leading to the instability of the reactor in a short period of time (Cheng et al., 2018a). This phenomenon might also decrease the particle size and contribute to the problem of membrane fouling in AnMBRs.

The production of EPS by microorganisms is their natural response to the toxic environment, as this plays an important role in protecting microorganisms to cope with the stress, that is, in the presence of heavy metals and or antibiotics (Avella et al., 2010, Sheng et al., 2010). The EPS are believed to be major contributors to membrane fouling,

since they possess complex properties including surface charge,

hydrophobicity/hydrophilicity, and adhesive characteristics, etc., which play roles in flocculation, stability and adhesion behaviors of sludge flocs (Lin et al., 2014). Thus, an increase in EPS will trigger a decline in sludge filterability, and the decrease in flux accompanied by an increase in specific cake resistance in MBRs (Wang et al., 2009). Avella et al. (2010) studied the effect of erythromycin, roxithromycin, amoxicillin, tetracycline and sulfamethoxazole antibiotics on EPS production by ultraviolet-visible spectroscopy, fourier transform infrared microscopy (FTIR) and size exclusion chromatography. The results indicated that these antibiotics at 10 mg/L could increase the production of EPS, erythromycin and roxithromycin in particular. The study by Zheng et al. (2016) and Zhu et al. (2018) also concluded that the addition of antibiotics (SMX, TC and norfloxacin) in the bioreactor stimulated microorganisms to secrete more EPS in response to the inhibition effect of antibiotics. Hence, it is believed that the presence of antibiotics in bioreactors could stimulate the release of EPS and result in membrane fouling.

SMP from cell metabolism and lysis refer to soluble proteins, polysaccharides, and humic-like materials (Jarusutthirak & Amy, 2006, Ma et al., 2015). Numerous studies have confirmed the significance of SMP on membrane fouling and indicated that more SMP would lead to severe membrane fouling (Guo et al., 2012, Lin et al., 2010, Lin et al., 2009). The reason is that SMP not only increase the sludge viscosity and leads to pore blockage, but also serve as the binding sites for cake layer formation, and thus facilitate cake formation on the membrane surface (Lin et al., 2010). Like EPS, the production of SMP can be increased by the addition of toxic compounds in the bioreactor (Aquino & Stuckey, 2004, Li et al., 2018). As reported by Li et al. (2018), the concentrations of EPS and SMP in an AFMBR rose when the concentration of benzothiazole increased from

1.23 mg/L to 12.02 mg/L. Zhu et al. (2018) observed that the mean concentrations of SMP and EPS increased by 340% and 220%, respectively, in a membrane fouling cycle after adding antibiotics. The injection of carbamazepine (90 µg/L) in a MBR induced a decrease of 100–1000 kDa protein-like SMPs and an increase of 10–100 kDa protein-like SMP in the supernatant, which contributed to more severe membrane fouling resulted from pore block or adsorption (Besha et al., 2017, Li et al., 2015). The presence of erythromycin in the anaerobic treatment process also elevated SMP production in terms of the observed increase in effluent COD (Amin et al., 2006). As explained by Zhu et al. (2018) and Aquino and Stuckey (2004), the rising SMP concentration caused by the toxic compounds in the bioreactor was probably due to the overwhelming production of volatile fatty acids (VFAs) and cell lysis products.

Polysaccharides and proteins in EPS and SMP are the primary components, which contribute to the formation of biofouling layers (Kale & Singh, 2016, Wang et al., 2017). As reported earlier, the contribution of proteins to membrane fouling in the AnMBRs was more than that of polysaccharides, because a higher concentration of protein was found in mixed liquor or cake layers (Huang et al., 2011, Juntawang et al., 2017, Ng et al., 2014). For example, the protein concentration was about 1.5 times higher than carbohydrate concentration in a novel staged anaerobic fluidized-bed ceramic membrane bioreactor (Aslam et al., 2018b). Juntawang et al. (2017) have suggested that proteins can form a sticky layer on the membrane surface, which accelerates fouling as cake filtration resistance. This is mainly because hydrolysis is usually considered as a rate-limiting step in the anaerobic processes, as the hydrolysis of proteins can be lower than that of the carbohydrates (Chen et al., 2017b). Furthermore, proteins are more hydrophobic and adhere more easily to the membrane surface (Meng et al., 2006). Zhang et al. (2017a) indicated that the protein was a negatively charged and sticky substance, and so it would

reduce the surface potential of sludge particles and increase the viscosity of the sludge. These sludge particles quickly gathered on the membrane surface and promoted the formation of the gel layer, ultimately leading to serious membrane fouling. Therefore, the aggravation of membrane fouling with the presence of antibiotics in the AnMBRs may result from the positive correlation between the antibiotics and protein concentration (Fang et al., 2002). As reported by Zheng et al. (2016), microorganisms, which were exposed to higher levels of antibiotics, would secrete more protein, probably due to the protein secretion metabolism of microorganisms being more sensitive to antibiotics than that of the polysaccharide secretion metabolism. Xu et al. (2013) stated that although the EPS production was not significantly influenced by sulfamethazine at 500 µg/L in an aerobic activated sludge system, the secondary structure of proteins in EPS altered. Therefore, the presence of antibiotics in the AnMBRs can increase membrane fouling through their effects on anaerobic sludge properties.

#### 3.2 Influences of antibiotics on fouling-related microbial communities

Microbial communities have been regarded as the ultimate factor responsible for the development of the cake layer and thus leading to significant biofouling (Lin et al., 2011b, Takada et al., 2018). The explanation for this lies in the formation of fouling layer being caused by the reproduction and succession process of microbial communities on the membrane surface (Gao et al., 2014b). Previous reports have indicated that the phyla of *Firmicutes*, *Proteobacteria*, *Chloroflexi* and *Bacteroidetes* in AnMBRs made the most contribution to membrane fouling (Juntawang et al., 2017). Thus, the influence of antibiotics on the development of membrane fouling can be reflected through their effects on these microbial communities.

According to previous reports, the phylum *Firmicutes* has the ability to accelerate biofouling in AnMBR and was commonly detected in fouling layers

(Fernández et al., 2008, Gao et al., 2010, Ma et al., 2013, Yu et al., 2012). In particular, Fernández et al. (2008) indicated that the order *Clostridiales* within *Firmicutes* played a significant role in the whole process of fouling formation, because of their preferential growth on the membrane surface and their positive correlation with the production of EPS in anaerobic sludge (Takada et al., 2018). The genus *Carnobacterium* also possesses the ability to attach to the membrane, and behaved as pioneer bacteria in the initial cake layer. Meanwhile *Peptococcaceae* was in high abundance in the cake layers, and this allowed it to contribute to the subsequent development of fouling after the initial fouling stage (Takada et al., 2018). As reported by Rogosa (1971), Peptococcaceae were expected to secret extra EPS, which would help the bacteria adhere to the membrane surface and facilitate the development of colonization. *Bacteroidetes* also represent a key phylum, which contributes to membrane fouling, and is presented in relatively high abundance in the cake layer (Gao et al., 2010, Juntawang et al., 2017). According to Gao et al.'s (2010) report, *Bacteroidetes* were likely EPS-generators with the potential to release more proteinaceous EPS.

Fernández et al. (2008) and Yu et al. (2012) have asserted that members of the *Proteobacteria* were apparently present in the biofouling layer. Specifically, *Sytrophobacteria* in class *Deltaproteobacteria* has been detected in both initial and mature biofilms (Yu et al., 2012). Similarly, *Sphingomonads* and *Arcobacter* genus belonging to the class *Alphaproteobacteria* are also reported as key microorganisms being able to form biofilms causing biofouling under anaerobic conditions (Calderón et al., 2011). *Chloroflexi* is another dominant bacterial phylum in anaerobic processes, although its function is still unclear (Riviere et al., 2009). It is probably the case that *Chloroflexi* was also the EPS-generator, so that high relative abundance of *Chloroflexi* has been observed in the cake layer. As reported in an aerobic MBR, this species in

Family *Caldilineaceae* (belongs to *Chloroflexi*) as filamentous bacteria generates biomass bulking and EPS that cover the membrane surface, thus leading to the biofouling problem in MBR (Choi et al., 2017).

Furthermore, the above-discussed bacteria have been found to be significant microbial groups in anaerobic bioreactors for treating antibiotic wastewaters (Cheng et al., 2018a, Li et al., 2011, Li et al., 2017, Meng et al., 2015b). As reviewed by Cheng et al. (2018a), antibiotics in the anaerobic treatment processes have a positive relationship with the abundance of dominant species in phyla *Bacteroidetes* and *Firmicutes*. The abundance of *Clostiridum* in anaerobic treatment processes could be elevated by increasing the concentration of SMX and TC. As well, the addition of TC in the anaerobic bioreactor could highly enrich *Bacteroidetes* in the sludge (Xiong et al., 2017). *Bacteroidetes* and *Proteobacteria* were identified as the hosts of ARGs in anaerobic digestion, which increased when antibiotic concentrations in the reactor also increased (Sun et al., 2016, Wu et al., 2016, Xiong et al., 2017).

With reference to methanogenic archaea, hydrogenotrophic methanogens emerge as dominant components of biofouling, due to perhaps their accumulation in the cake layer: firstly, making the facilitation of interspecies hydrogen transfer possible; and secondly, maximizing the overall conversion of organic substances (Calderón et al., 2011, Li et al., 2017). For instance, *Methanospirillum*, *Methanobrevibacter*, *Methanocalculus*, *Methanospirillaceae* and *Methanosarcinales* were persistently detected in the fouling biofilm even after chemical cleanings (Calderón et al., 2011). Accordingly, the proportion of hydrogenotrophic methanogens will increase and become the dominant methanogenic group in the anaerobic treatment processes after exposure to antibiotic conditions (Cheng et al., 2018a). It is mainly because hydrogenotrophic methanogens having a higher substrate utilization rate, growth rate and cell yield to expose toxic

substances compared to acetoclastic methanogens (Li et al., 2017). Therefore, the presence of antibiotics in the AnMBRs can potentially accelerate the development of membrane fouling through their influences on the microbial communities in the anaerobic processes.

#### 4. Integrated AnMBRs for fouling mitigation and antibiotics removal

Membrane fouling is a major issue, in that it can seriously affect the membrane's performance and overall longevity (Huang et al., 2011, Mirbagheri et al., 2015). The result increased operating and energy costs in the AnMBR process (Liao et al., 2006). Nevertheless, conventional ways to control membrane fouling in AnMBR, for example, gas-sparging often require high energy costs. The high-energy requirements reduce the potential advantage of the AnMBR over their aerobic counterparts. Thus, finding methods for slowing down cake formation on membrane surface is crucial. Integrating the AnMBR with other technologies has been considered a promising strategy for: (i) improving qualities of effluent; (ii) mitigating membrane fouling; and (iii) enhancing the removal efficiency of antibiotics. The schematic diagram of some integrated AnMBRs is shown in Fig. 2.

#### 4.1 Integration of AnMBRs with biofilm carriers

The introduction of biofilm carriers (e.g., GAC, PAC, and Sponge) into the membrane bioreactor has been considered an effective method for controlling membrane fouling (Aslam et al., 2018a). This is principally because the carrier in the reactor can improve the structure, size, flocculability and settleability of the sludge particles (Kim et al., 2010, Zhang et al., 2017e). The EPS/SMP in the mixed liquor adsorbed by biofilm carriers resulting in lower concentrations than that observed in conventional AnMBR (Aslam et al., 2018a). Moreover, these carriers in MBRs can facilitate the growth of

slow-growing microorganisms which can remove some micropollutants (Guo et al., 2010), and this feature is critically important.

Granular activated carbon (GAC) particles have been widely used in AFMBR as the fluidized medium to support the growth of microorganisms (Vyrides & Stuckey, 2009). The scouring action of the fluidized GAC particles on the membrane surface could essentially eliminate membrane fouling as indicated by low transmembrane pressures (Kim et al., 2010, McCurry et al., 2014, Aslam et al., 2017a). Aslam and Kim (2017b) stated that membrane scouring under GAC fluidization decreased reversible fouling resistance effectively, as the TMP remained less than 0.1 bar without significant membrane fouling during the first 100 days of operation. Unlike the common gassparging fouling mitigation method or cross-flow filtration mode, membrane scouring by fluidized GAC particles under bulk recirculation alone through the reactor consumed relatively low energy (Aslam et al., 2014, Aslam et al., 2018b). As indicated by Kim et al. (2010), the total energy required for fluidization for the AFMBR with GAC was 0.028 kWh/m<sup>3</sup> is less than that in other submerged membrane bioreactors with gas-sparging for fouling control. According to Gao et al. (2014a) and Li et al. (2017), the membrane fouling cycle was obviously longer in AFMBR with the addition of GAC (31d, HRT=8 h) than the one without GAC (3.2 d, HRT=12 h), even with the presence of antibiotics in the reactor. In addition to the scouring effect of GAC on membrane fouling, GAC can not only adsorb fine colloids from the solution, but also decrease the levels of SMP and EPS in bulk from the reactor (Aslam et al., 2017a). The increase of the overall particle size, as well as the decline of SMP and EPS in bulk can help to reduce membrane fouling (Aslam et al., 2017b, Bae et al., 2014).

Similar to GAC, PAC can also provide a solid support for biomass growth in the bioreactor and control membrane fouling. According to previous reports, PAC addition

improved the performance of the AnMBR and reduced membrane fouling in terms of physical scouring at membrane surface, adsorbing fine material in the mixed liquor suspension and maintaining low MLSS concentration due to biofilm growth effective on PAC in the reactor (Aslam et al., 2017a). All these combined effects resulted in a significant flux improvement of the AnMBRs (Akram & Stuckey, 2008, Zhang et al., 2017b). Kim et al. (2010), Chong (2015) and Kaya et al. (2016) observed that both the fouling and cake layer resistances continuously decreased with the addition of PAC due to its scouring effect, and the adsorption of organics and EPS onto PAC. Vyrides & Stuckey (2009) also indicated that the reduced deposition of fine particles on the membrane surface was resulted from the addition of PAC. By contrast, the performance of the AnMBRs with the addition of PAC appears better than that of GAC, because PAC has a greater surface area per mass than GAC. This leads to greater absorbance of the low and high molecular weight of biodegradable matter and fine colloidal particles (Kim et al., 2010, Skouteris et al., 2012). As reported by Hu and Stuckey (2007), the addition of PAC in AnMBR system could achieve a lower TMP value than the system with GAC under the same permeate flux. However, during long-term membrane operation, the adsorption capacity of GAC and PAC should be exhausted (Aslam et al., 2017a). Moreover, GAC and PAC particles are likely to break with the production of small size particles as potential foulants to membrane rather than alleviating membrane fouling (Charfi et al., 2018, Wu et al., 2015). Hence, the addition of PAC and GAC in membrane reactors may accelerate membrane fouling in the long-term operation.

As discussed in section 2, the addition of GAC and PAC in the AnMBR can significantly improve the removal of antibiotics from wastewater, which would further reduce the effect of antibiotics on membrane fouling (Paredes et al., 2018, Xiao et al., 2017). Sorption onto GAC and PAC contributed to the removal of antibiotics from

wastewater to a certain degree, coupled with the enrichment of microorganism species (including the slowing-growing bacteria) in the AnMBR is responsible for antibiotic removal (Skouteris et al., 2015, Yu et al., 2016). In addition, the GAC particles were expected to play a role as promoting interspecies electron transfer in anaerobic reactors due to their conductive properties, which possibly enhanced antibiotic removal (Zhang et al., 2017d). Similarly, the diversity and enrichment of bacteria was improved by adding PAC in MBRs. This phenomenon is related to the highly biodegradation of pharmaceutical compounds (Xiao et al., 2018, Yu et al., 2018).

Using lighter materials rather than GAC or PAC for maintaining the fluidization to reduce the accumulation of foulants on membrane surface is possibly the best available option as it requires less energy (Kim et al., 2010). Sponges with high interior porosity, specific surface area and light weight are considered to be ideal for attached-growth material to be used as a transportation medium for active biomass, and to decrease cake layer accumulation on the membrane surface (Guo et al., 2008, Nguyen et al., 2012). Guo et al. (2008) concluded that sponge additions in a submerged membrane bioreactor (SMBR) decreased the membrane fouling and increased the sustainable flux markedly. The membrane fouling rate could also be reduced after the addition of sponge in a suspended sponge membrane bioreactor (SSMBR) (Nguyen et al., 2012). As explained by Deng et al. (2014), the sponge in the reactor affected the properties of activated sludge, which decreased the production of EPS and SMP through adsorption and biodegradation, lowered sludge viscosity, increased sludge floc size, and then prevented the cake layer formation and pore clogging, thus reducing membrane fouling. Chen et al. (2017a) compared the treatment performance and membrane fouling fate in a conventional granular anaerobic membrane bioreactor (CG-AnMBR) and a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR), and indicated that the SG-AnMBR not

only demonstrated better performance but slower fouling development with 50.7% lower total filtration resistance than the control reactor. Additionally, sponge media with high porosity not only has high sorption capacities for micropollutants but also facilitates the growth of microorganisms, thereby contributing to the removal of micropollutants from wastewater (Luo et al., 2014, Nguyen et al., 2017). For instance, antibiotics, such as norfloxacin, ciprofloxacin, ofloxacin, tetracycline and trimethoprim are effectively removed from wastewater by sponge MBRs (Nguyen et al., 2017).

In summary, the addition of above mentioned carriers in AnMBR has been proved to be an effective method to reduce membrane fouling and enhance the removal of antibiotics from wastewater. The AnMBRs with carriers are more compact, operate with higher fluxes and are energy efficient over the conventional reactor. Nevertheless, as reviewed by Neoh et al. (2016) and Zhang et al. (2017e), membrane fouling may become severe at the later stage of treatment in the BFMBRs, when the breakage of carriers occurs with the overgrowth of filamentous bacteria. Thus, more investigations are necessary for the contribution of carriers in order to control membrane fouling in the AnMBRs.

#### 4.2 Integration of AnMBRs with bioelectrochemical systems (BES)

Bioelectrochemical systems (BES), for example microbial fuel cells (MFCs) and microbial electrolysis cells (MECs), represent emerging technologies for wastewater treatment with recovery of the inherent energy as electricity (MFCs) or hydrogen (MECs) (Du et al., 2007, Escapa et al., 2014). The integration of BES with AnMBR is an alternative technology, which takes advantage of both BES and AnMBR to improve wastewater treatment efficiency, recover energy and reduce membrane fouling simultaneously (Katuri et al., 2014, Su et al., 2013, Tan et al., 2017, Werner et al., 2016).

MFCs utilize the presence of electrochemically-active bacteria as catalysts to convert soluble organic matter in the wastewater into useful electrical energy (Ma et al., 2016). According to the research by Tan et al. (2017), the combination of MFC with AnMBR resulted in higher COD removal efficiencies compared to the AnMBR alone. This is perhaps due to the activities of electroactive bacteria and common bacteria stimulated by electricity, or the effective degradation of the inhibitory compounds (e.g., volatile fatty acids), which enhanced the removal of COD. The author also indicated that using MFC as a pre-treatment prior to AnMBR was beneficial to reduce the finer foulants and EPS production, thereby reducing membrane fouling. As well, Tian et al. (2014) demonstrated that membrane fouling was mitigated largely in an anaerobic membrane bioelectrochemical reactor (AnMBER) in comparison with a control AnMBR by improving the mixed liquor properties. Specifically, the lower particle zeta potential and smaller amount of SMP in the AnMBER attributed to the membrane fouling limitation. Sludge suspension with low zeta potential (absolute value) is expected to have a strong aggregation tendency (Azeredo et al., 1999). Moreover, fine particles (with size near 10 μm), which could cause intense fouling by accelerating the formation of cake layers on membrane surface, were no longer detected in the AnMBER system (Lin et al., 2010, Tian et al., 2014).

MECs are similar to MFCs but can produce hydrogen from the biodegradation of organic matter with added voltage (Wagner et al., 2009). The application of MECs in the AnMBR is a potential method to alleviate membrane fouling, because the applied electric field could enhance the activity of microorganisms and alleviate membrane fouling (Sun et al., 2017). Ding et al. (2018) studied the effects of electrical fields on membrane fouling in a new type of MES-AnMBR reactor, and found that the membrane fouling rate of the MEC-AnMBR reactor gradually slowed down with the increase of

applied voltage (0-1.0 V). A similar result has been reported by Werner et al. (2016), who indicated that the onset of biofouling was delayed and minimized in anaerobic electrochemical membrane bioreactors (AnEMBRs) operating at 0.9 V compared to that of 0.7 V. Lower membrane fouling propensity was obtained in an anaerobic forward osmosis membrane bioreactor coupled with the microbial electrolysis cell (AnOMEBR), wherein the membrane operation cycle was about 1.27 times longer than that in the control reactor (Zhang et al., 2017a). One of the important reasons for the mitigation of membrane fouling in the MEC-AnMBRs was the lower concentration of SMP and EPS in the reactor, as well as the smaller amount of protein in them.

In the MEC-AnMBRs, the increase in applied voltage leads to low cathode potential, which may affect the electron transfer system and oxidative phosphorylation in cells around the membrane, and subsequently influence enzyme activity of ATP synthase and intensity of proton motive force in the extracellular protein secretory pathway (Ding et al., 2018, Katuri et al., 2014). In addition, the electrostatic repulsion interaction between sludge particles and substances in the wastewaters caused by the electric field would abate stability and compactness of the sludge cake layer formatted on the membrane surface (Akamatsu et al., 2010, Chen et al., 2007). The viscosity of sludge would decrease by increasing the applied voltage (Ding et al., 2018). Moreover, the scouring effect of hydrogen gas bubbles on the membrane surface might also contribute to the reduction of fouling in the MEC-AnMBR system (Katuri et al., 2014).

Furthermore, the pretreatment of antibiotic wastewaters by BESs before the AnMBR was able to enhance the degradation of antibiotics and eliminate their antibacterial activity simultaneously. This consequently, reduced their adverse effects on the AnMBR performance and membrane fouling (Wang et al., 2016, Wen et al., 2011, Yan et al., 2018, Zhang et al., 2017c). Yan et al. (2018) found that the removal efficiency

of oxytetracycline (10 mg/L) in MFCs increased to 99.00% in 78 h, which was higher than that in microbial controls (only 58.26%). Wang et al. (2016) investigated the biodegradation of SMX by MFC, and demonstrated that about 85% of SMX (20 mg/L) and its toxic intermediate, 3-amino-5-methylisoxazole (3A5MI), could rapidly degrade in MFC reactors. Zhang et al. (2017c) also observed the improved removal of chloramphenicol (CAP) in MFC reactors with acetate as electrons donor, with 84% of 50 mg/L CAP was degraded within 12 h in the MFC. As well, 91% ceftriaxone (50 mg/L) was degraded within the operation time in the MFC compared with 51% in the anaerobic reactor (Wen et al., 2011).

In addition to the above-explained individual antibiotics, the combination of different types of antibiotics, which creates more serious inhibition to anaerobic microorganisms, still can be removed in great quantities in MFCs. In fact, the efficiency in removing aureomycin, roxithromycin and norfloxacin rose to 100% while that for sulfadimidine reached 99.9% (Cheng et al., 2018b, Zhou et al., 2018). Therefore, anaerobic conditions coupled with electrical stimulation might have a more profound selective effect on specialized groups of functional species than that in traditional anaerobic reactors, which could rapidly enhance the biodegradation of antibiotics (Yan et al., 2018). The existence of electrical fields in reactors could increase the permeability of cell membrane, enhance the absorbance of extracellular substances and alter the microbial metabolism (Yan et al., 2018). Additionally, a long-term acclimation period of antibiotics in the MFCs was important for higher removal efficiency, which led to the enrichment of various functional bacteria and then enhanced their degradation ability (Wang et al., 2016, Zhang et al., 2018). According to the antibacterial activity tests conducted by Wang et al. (2016) and Zhang et al. (2017c), the inhibition effect of antibiotics towards anaerobic microorganisms could be largely eliminated by MFC

treatment. Moreover, Xu et al. (2017) indicated that the pretreatment of antibiotics in BES largely altered the microbial community structure, and the proportion of hydrogenotrophic methanogens could be replaced by the strictly acetoclastic methanogens. Therefore, the integration of the AnMBR with BESs constitutes a promising strategy for the effective treatment of antibiotic wastewater, membrane fouling mitigation and energy production.

#### 4.3 Economic evaluation of AnMBR technologies

Membrane fouling continues to be an important barrier for the application of the AnMBR system due to the costs of fouling control. Based on above discussion, the hybrid AnMBRs system with biofilm carries or BES may be a practical technology for antibiotic wastewater treatment. The reason is not just their potential for removing antibiotics and ARGs effectively, but also their relatively lower electrical energy requirements than conventional AnMBRs. For instance, the energy demands in the anaerobic fluidized membrane bioreactor (mainly for GAC fluidization) ranged from 0.039 to 0.13 kW h/m³ under various operation conditions, which could be satisfied by using the produced methane energy (Aslam et al., 2017b, Kim et al., 2010, Yoo et al., 2012). In contrast, as reviewed by Liao et al. (2006) and Dvořák et al. (2016), to reduce membrane fouling in conventional AnMBRs, 3–7.3 kWh/m³ of energy was used by high cross-flow velocity in external cross-flow membranes, and 0.25–1.0 kWh/m³ was employed through extensive gas scouring in internal submerged membranes.

The integrated system of AnMBRs with BES also has low energy demands during operation. Katuri et al. (2014) stated that the energy required for operating the MES-AnMBR was 0.27 kWh/m<sup>3</sup> even at an applied voltage of 0.7 V. The energy consumption in MES-AnMBR systems could be compensated through the increasing methane production. Moreover, about 0.537 kJ/day positive energy gain could be obtained (Zhang

et al., 2017a). Novel two-stage microbial fuel cell and anaerobic fluidized-bed membrane bioreactors (MFC-AFMBRs) were conducted by Ren et al. (2014) and Kim et al. (2016). They indicated that the MFC-AFMBR could achieve high effluent quality with low energy consumption, as the reactor could operate at a constant high permeate flux without the need or use of any membrane cleaning or backwashing. As reported by Ren et al. (2014), the electrical energy produced by the MFCs (0.0197 kWh/m³) satisfied the total electrical energy required (0.0186 kWh/m³) for the operation of the MFC-AFMBR system. Therefore, the integrated AnMBR system is a promising technology for the treatment of antibiotic wastewater.

#### 5. Future perspectives

As discussed in this paper, the selected antibiotics can be removed in large quantities from wastewater by AnMBRs, especially the integrated AnMBR systems. However, to date, most research has only focused on the synthetic wastewater with a limited range of antibiotics. More studies are required to better understand the removal of more antibiotics by AnMBRs and the responsible mechanisms, because of the different physical-chemical properties of antibiotics and their effects on microbial communities in anaerobic reactors. In addition, transformation products of anaerobically degraded antibiotics and their eco-toxicity or public health risks also need further investigation. Moreover, the full-scale implementation of the AnMBRs for the treatment of antibiotic wastewater is still pending due to membrane fouling problems and their sensitivity to antibiotics.

In this current review, it is obvious that the presence of antibiotics in the AnMBRs might have a positive correlation with membrane fouling through their impacts on fouling related factors. Whereas, the interaction mechanism between different types of antibiotics and the produced EPS and SMP in the AnMBRs still requires further

exploration to consider the complex molecular features of EPS and SMP with these various functional groups, as well as the different physical-chemical properties of different types of antibiotics. The identification of specific microbial communities, which contributed to biofouling, is essential for further investigation of the biofouling mechanism and developing effective membrane fouling control strategies. The effect of antibiotics on microorganisms in AnMBRs requires further analysis to better understand their influences on membrane fouling.

The integration of AnMBRs with other technologies has proven appropriate for slowing down membrane fouling and removing refractory antibiotics from wastewater. The addition of carriers to the AnMBRs is the most practical method for controlling fouling formation, increasing flux and improving removal efficiencies of COD and antibiotics. However, more studies regarding the long-term impact of carriers on membrane structure, strength and effectiveness are necessary in the future, as they may reveal a negative effect on membrane performance under long-term operation conditions. The combination of BES with AnMBR is a highly promising alternative for successfully minimizing membrane fouling and degrading antibiotics simultaneously. Nevertheless, further studies here too are necessary on the presence of different types of antibiotics in wastewaters and examining limitations of removing them under different operating conditions.

In summary, more studies on the behavior of antibiotics in AnMBRs systems are necessary for their elimination and influences on membrane fouling. The hybrid AnMBR systems have been considered as promising alternatives for removal of toxins and controlling membrane fouling with low energy costs, but further tests are required. Essentially, both technically and economically feasible AnMBRs processes should be developed for treating antibiotic wastewaters.

#### 6. Conclusion

The AnMBRs are effective technologies for removing antibiotics and ARGs from wastewater. Yet, antibiotics would aggravate membrane fouling by influencing the floc size, the production of EPS and SMP, and the microbial communities in the AnMBRs. Integrating AnMBRs with carriers demonstrate a higher removal efficiency of antibiotics and a slower membrane fouling rate, but their long-term effects on membrane properties and microbial activities need further investigation. The integrated BES-AnMBRs not only can control fouling, degrade antibiotics and eliminate their antibacterial activity, but also enhance the energy recovery from wastewater. It is therefore a promising technology for antibiotic wastewater treatment.

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#### **Figure Captions**

Fig. 1 Membrane fouling in AnMBRs without and with antibiotics by SMP and EPS attributed to membrane pore clogging and deposition of sludge solids on membranes.

Fig. 2 Schematic diagram of the (a) integration of AnMBRs with biofilm carriers (e.g., AM. GAC, PAC, and Sponge); (b) integrated BES-AnMBRs; (c) combined MFC-AFMBR.

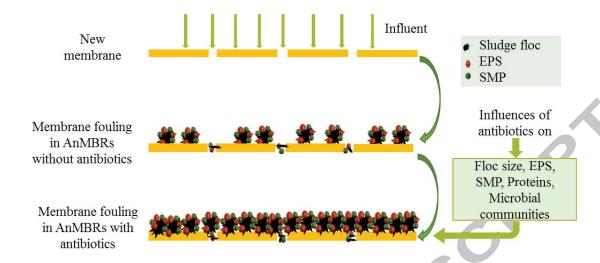


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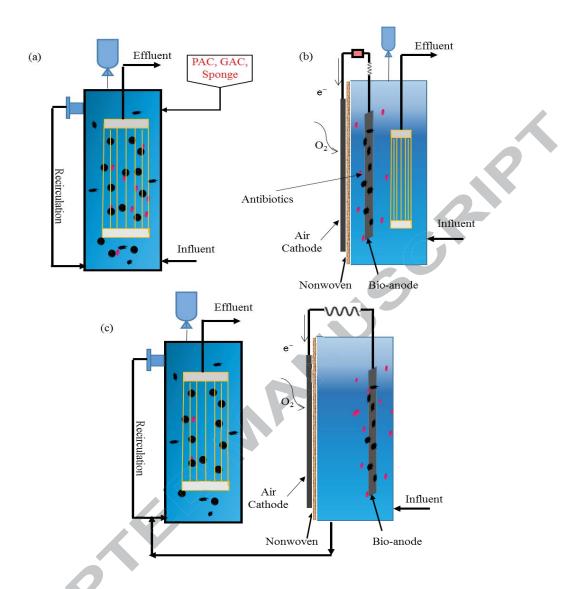


Fig.2 Schematic diagram of the (a) integration of AnMBRs with biofilm carriers (e.g., GAC, PAC, and Sponge); (b) integrated BES-AnMBRs; (c) combined MFC-AFMBR.

#### **Table Captions**

Table 1 Performances of AnMBRs for treating antibiotic wastewater



Table 1 Performances of AnMBRs for treating antibiotic wastewater

-						A4:1.: - 4: - /	_
	T 4 4		Influent	COD	Initial	Antibiotic/	
tic types	Treatment	Operating conditions	COD	removal	antibiotics	ARGs	Re
	strategies		(mg/L)	(%)	(ng/L)	removal	
						(%)	
hoxazole	AnMBR	T=35 °C; HRT=6-12 h; SRT=213	500	$93.9 \pm 1.8$	-	$67.8 \pm 13.9$	(Xi
prim		d; OLRs=2 kgCOD/m³/d; Kubota				$94.2 \pm 5.5$	201
hoxazole	PAC+AnM	microfiltration membrane module		5		>99	
prim	BR	(filtration area 0.11 m <sup>2</sup> )				>99	
prim	AnMBR	$T=30 \pm 1$ °C; HRT=6 h; SRT=30 d;	$240\pm15$	_	$1550\pm238$	35.4	(M
hoxazole		Hollow-fibre membrane ( pore size			$1625\pm263$	95.2	et a
		0.04 μm, total area 0.0245 m <sup>2</sup> )					
prim	AnMBR	T=35± 1 °C, HRT=4 d; SRT=180	_	84%	_	97.5	(W
hoxazole		d; OLR=1.3 gCOD/L/d; Membrane				99.6	et a
		module (pore size and effective					
		surface area of 1 $\mu$ m and 0.09 m <sup>2</sup> )					
hoxazole	AnMBR-	$T=35 \pm 1$ °C; HRT=12 h; PVDF	$400\pm10$	_	10000-	>98	(W
	NF	(nominal pore size 30 nm, filtration			20000		201
		area 310 cm <sup>2</sup> )					
hoxazole	SAF-MBR	HRT =6.8 h; SRT= 36 d, PVDF	_	_	207-276	88-100	(M
	•	membranes (pore size of 30 nm;					al.,
		total surface area of 39.5 m <sup>2</sup> )					
llin	AnMBR	OLRs= $2.37 \pm 0.28$ to	5000	94.00%	3.81-	$73.2 \pm 4.3$	(Hı
		$4.46 \pm 0.87 \text{ kgCOD/m}^3/\text{d}; \text{PVDF}$			10.93 mg/L		al.,

ne		membrane module ( surface area of			2.92-	$47.7 \pm 2.2$	
		1 m <sup>2</sup> and a nominal pore size of			5.73 mg/L		
zone		0.02 μm)			0.35-	$79.4 \pm 4.1$	
					1.02 mg/L		
in					4.81-	$34.6 \pm 3.3$	
					9.52 mg/L		
nides	A1/A2/O-	T=25 °C; Submerged hollow fiber				93.9–97.5	(Zh
	MBR	polythene membrane (nominal pore		5			201
		size: 0.1 μm, membrane area: 0.2					
		$m^2$ )	7				
hoxazole	AnMBR	T=35°C, HRT=12/24h, OLRs= 0.8	400–800	>98		Nearly	(На
prim		g/L/d				complete	201
						removal	
ine	Two-stage	HRT=1.28 h; OLRs=5.65 g/L/d;	250	95.00%	$18.9 \pm 2.1$	93.7	(Dı
hoxazole	AFMBR	PVDF membranes (1.2 mm			$312\pm34.6$	89.1	al.,
ıycin-		diameter, 105 cm long, pore size of			$319 \pm 42.4$	86.3	
		<0.1 μm, membrane surface area of					
mycin		$0.04 \text{ m}^2$ )			$324 \pm 6.4$	89	
ie	O				0.9-417±84	100	
s							
in	▼				$2905 \pm 530$	95.8	
ne					$51.2 \pm 6.0$	93.6	
prim					$15.9 \pm 3.6$	100	
azole	AFMBR	T= 35°C; SRT= 35 d; Hollow fiber	2961-	90.9-93.6	50 mg/L	$82.3 \pm 3.7\%$	(Li
		membrane (surface membrane area	3337			(HRT=12	201