Recent advances in attached growth membrane bioreactor systems for wastewater treatment

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Abstract

To tackle membrane fouling and limited removals of pollutants (nutrients and emerging pollutants) that hinder the wide applications of membrane bioreactor (MBR), attached growth MBR (AGMBR) combining MBR and attached growth process has been developed. This review comprehensively presents the up-to-date developments of media used in both aerobic and anaerobic AGMBRs for treating wastewaters containing conventional and emerging pollutants. It also elaborates the properties of different media, characteristics of attached biomass, and their contributions to AGMBR performance. Conventional media, such as biological activated carbon and polymeric carriers, induce formation of aerobic, anoxic and/or anaerobic microenvironment, increase specific surface area or porous space for biomass retention, improve microbial activities, and enrich diverse microorganisms, thereby enhancing pollutants removal. Meanwhile, new media (i.e. biochar, bioaugmented carriers with selected strain/mixed cultures) do not only eliminate conventional pollutants (i.e. high concentration of nitrogen, etc.), but also effectively remove emerging pollutants (i.e. micropollutants, nonylphenol, adsorbable organic halogens, etc.) by forming thick and dense biofilm, creating anoxic/anaerobic microenvironments inside the media, enriching special functional microorganisms and increasing activity of microorganisms. Additionally, media can improve sludge characteristics (i.e. less extracellular polymeric substances and soluble microbial products, larger floc size, better sludge settleability, etc.), alleviating membrane fouling. Future studies need to focus on the development and applications of more new functional media in removing wider spectrum of emerging pollutants and enhancing biogas generation, as well as scale-up of lab-scale AGMBRs to pilot or full-scale AGMBRs.

Keywords: Attached growth; Membrane bioreactor; Wastewater treatment; Emerging pollutants; Carriers; Media

Abbreviations of emerging pollutants: ACT, acetaminophen; AOX, Adsorbable organic halogens; AZM, Azithromycin; BPA, Bisphenol A; CAF; Caffeine; CAM, Clarithromycin; CBZ, Carbamazepine; CED, Cephradine; CIP, Ciprofloxacin; CTC, Catechol; DCF, Diclofenac; DZP, Diazepam; EB, Ethylbenzene; E1, Estrone; EE2, 17-α ethinylestradiol; E2, 17-β-estradiol; E3, Estriol; ERY, Erythromycin; ERY-H_2O, Erythromycin-H_2O; 2,4,5-TP, fenoprop; FLX, Fluoxetine; Fenoprop; FMQ, Flumequine; GEM, Gemfibrozil; IBP, Ibuprofen; KEP, Ketoprofen;

MTZ, Metronidazole; NA, Nalidixic acid; NPX, Naproxen; OTC, Oxytetracycline; OFL, Ofloxacin; PA, Pipemidic acid; PCM, Paracetamol; PCP, Pentachlorophenol; PHA, Phthalic acid; PHN, Phenanthrene; POF, Pentoxifylline; PRM, Primidone; ROX, Roxithromycin; SA, Salicylic acid; SDZ, Sulfadiazine; SMZ, Sulfamethazine; SMX, Sulfamethoxazole; STY, Styrene; TBP, 4-tert-butylphenol;TCS, Triclosan; TL, Toluene; Tmp, Trimethoprim; TPH, Theophylline; 2-CP, 2-chlorophenol; 4-tOP, 4-tert-octylphenol; 4-NP, 4-n-nonylphenol

1. Introduction

Boosted by more stringent environmental legalizations and intensifying scarcity of fresh water resources, the demand for membrane bioreactor (MBR) technology has experienced rapid growth with many full-scale applications in treating municipal and industrial wastewaters worldwide (Ivanovic and Leiknes, 2012; Skouteris et al., 2015). MBRs have gained popularity due to considerable advantages over conventional process including (1) high quality effluent; (2) excellent microbial separation; (3) absolute control of hydraulic retention time (HRT) and sludge retention time (SRT); (4) lower net sludge production; (5) small footprint requirement; (6) operation at high volumetric loadings; (7) improved nutrient removal; and (8) possibility of a flexible and phased extension for existing wastewater treatment plants (Guo et al., 2010).

Nevertheless, the rapid commercialization of MBR technology is constrained by some challenges, among which membrane fouling is one of the most challenging problems for its widespread application. The literature has revealed that most of MBRs were predominately in the form of suspended growth MBR configuration where pollutants removal was largely dependent on the growth of suspended sludge flocs. However, the deposition of soluble and particulate substances onto/into the membrane can inevitably cause permeate flux decline, resulting in frequent membrane cleaning, necessary membrane replacement, and high energy consumption and operational cost. Other researchers have also reported some issues of conventional MBR (CMBR), including low total nitrogen and phosphorus removals (Jamal Khan et al., 2014) and limited removals of some emerging pollutants (Alvarino et al., 2017; Asif et al., 2020), which hindered its further application as a MBR for advanced wastewater treatment.

To overcome the above-mentioned limitations of CMBRs, several research groups have attempted to integrate attached growth process with CMBR to form a hybrid MBR known as attached growth MBR (AGMBR) (Ivanovic and Leiknes, 2012). Hence, in an AGMBR, pollutants removal can be completed by (1) sorption onto and/or degradation by suspended growth (i.e. activated sludge), (2) sorption onto media, (3) degradation by attached microorganisms on media, (4) rejection by membrane or degradation by attached biofilm. Studies have proven that the use of media in AGMBR could reduce suspended solids concentration, modify the characteristics of sludge suspension and promote physical scouring effect of the suspended carriers, thereby improving membrane filterability and mitigating membrane fouling (Deng et al., 2014; Hu et al., 2014). Improved treatment performance in terms of organic, nutrient and micropollutants removal by AGMBR, as compared to CMBR, has also been reported due to enhanced biomass concentration, enriched microbial biodiversity and activity, and improved simultaneous nitrification and denitrification (Nguyen et al., 2017; Zhang et al., 2017a). Thus, AGMBR has the merits of operating at higher flux, being more compact, achieving higher pollutants removal efficiency, and having efficient membrane fouling control.

Two review papers have been published with regard to the performance of AGMBRs with specific media such as activated carbon (Skouteris et al., 2015), and granular media (Iorhemen et al., 2017). Zhang et al. (2017a) also provided a mini review on recent R&D progress in AGMBRs, but mainly focused on their mechanisms, the degradation of conventional pollutants and membrane fouling control. In addition, the development and performance of AGMBRs were only mentioned in one section or sub-section in some review articles (Du et al., 2020; Goswami et al., 2018; Huang and Lee, 2015). To date, although many researchers have adopted various types of carriers in MBRs for wastewater treatment using different systems, no attempt has been made to provide a comprehensive review of different types of supporting media used in AGMBRs. Since the properties of the growth media determine the efficiency and performance of

AGMBR, an in-depth review on current research progress is of great value. Therefore, this review aims to provide comprehensive discussion on attached growth media used in AGMBRs using both conventional and new media (i.e. activated carbon, polymeric carriers, biochar, bioaugmented carriers containing selected strains/mixed cultures, etc.), and to clearly demonstrate their properties and contributions to the removal of pollutants and fouling control. Moreover, this review covers recent AGMBR advancements in eliminating emerging pollutants. Overall, it is the first review paper for both aerobic and anaerobic AGMBRs.

2. Types of media used in AGMBR systems

2.1. Conventional media

2.1.1. PAC/GAC

Generally, activated carbon (AC) is made from coal, coconut shell or wood. AC, such as powdered activated carbon (PAC) and granular activated carbon (GAC), has high specific surface area (≥ 500-1360 m^2/g), relatively low bulk density (< 1 g/cm^3) and well-developed micropores (≥ 60% microporosity) (Alvarino et al., 2017; Chaiprapat et al., 2016; Zhang et al., 2019a). Particle size of GAC (0.5-2.4 mm) is significantly higher than that of PAC (< 0.15 mm). They have been widely used in AGMBR, i.e. PAC-MBR (Zhang et al., 2019b; Woo et al., 2016), GAC-MBR (Lu et al., 2020; Nguyen et al., 2012), anaerobic fluidized membrane bioreactor (Aslam et al., 2018; Shin et al., 2014), etc. AC addition into MBRs exhibits superior pollutants removal ability though 1) adsorbing biologically persistent or recalcitrant pollutants; 2) offering large surface area for the attachment and growth of microorganisms; 3) showing high affinity for molecular oxygen and further increasing penetration of oxygen into biofilm due to the oxygen concentration gradient from AC surface to inner layer of biofilm; 4) moderating loading of hazardous compounds and minimizing their negative effects on suspended biomass by adsorption onto AC; 5) biodegrading the adsorbed compounds by microorganisms attached on AC; 6) alleviating membrane fouling by scouring membrane surface and adsorbing biopolymers from suspended sludge; 7) improving sludge properties (i.e. floc size, viscosity, filterability, dewaterability) (Du et al., 2017; Remy et al., 2010; Skouteris et al., 2015). PAC and GAC addition also help to enrich special functional microorganisms in AGMBR systems, promoting stable and good performance when treating different types of wastewaters.

Generally, both aerobic and anaerobic AGMBRs can remove > 80-90% of COD and BOD_5 from municipal or domestic wastewater, which are slightly higher than single aerobic MBR or AnMBR. High microbial activities of attached biofilm on AC enable good removal of organic matters. The high specific surface area provided by AC prompts the liquid-solid contact and further encourages the adsorption of microbial cells, enzymes and organic matters as well as enrichment of heterotrophic bacteria onto AC, thereby giving rise to a microenvironment favourable for microbial metabolism. The enzymes facilitate the extracellular biodegradation of adsorbed organic matters on AC (Satyawali et al., 2009; Sirotkin et al., 2001). Anaerobic AGMBR cannot effectively remove nitrogen and phosphorus, whereas good nitrogen and phosphorus removal can be observed in aerobic AGMBR. According to previous studies, although aerobic AGMBR and single aerobic MBR could achieve > 90% of NH_4^+-N removal, aerobic AGMBR had better (about 3-4%) removal compared with aerobic MBR, which was ascribed to the enrichment of nitrifying microorganisms on PAC. Moreover, PAC addition also enhanced total nitrogen (TN) and total phosphorus (TP) removals (up to 75-85%), owing to the formation of anoxic microenvironment inside the attached biofilm on PAC for denitrification and increased abundance of phosphorus accumulating organisms (PAOs) (Asif et al., 2020; Aslam et al., 2018; Lei et al., 2019; Lim et al., 2019; Xiao et al., 2017).

In aerobic AGMBRs, PAC addition enhances abundance and diversity of microbial community in mixed liquor as well as on PAC particles. The dominant genera *Thiothrix* and *Sphingomonas* in mixed liquor grow and adhere onto PAC surface due to their high biofilm-forming potential and release of exopolysaccharides for easy attachment of biomass on carriers. In addition, the certain sulfur content of ~1.0% wt in PAC favors the dominant growth of *Thiothrix* on PAC. PAC addition increases the abundance of phyla *Acidobacteria* and *Bacteroidetes,* class *Gammaproteobacteria* (belonging to phylum *Proteobacteria*) and genus *Sphingomonas*, ensuring good removal of organic matters (i.e. assimilable organic carbon (AOC)). Genera *Nitrospira* and *Nitrosomonas* prefer to accumulate on PAC surface, resulting in effective NH_4^+-N removal. The creation of an oxygen-deficient microenvironment by attached biomass on PAC is also favorable for the growth of anoxic-denitrifying bacteria (e.g. *Methylotenera*, *Haliangium*) and slow-growing aerobic denitrifying bacteria (e.g. *Acinetobacter*, *Zoogloea*), contributing to NO_3^--N reduction. After adding PAC, the increased abundance of phylum *Cyanobacteria* in mixed liquor facilitates nitrification and phylum *Verrucomicrobia* promotes P removal. Additionally, enrichment of genus *Candidatus Accumulibacter* belonging to class *Betaproteobacteria* in mixed liquor is responsible for TP removal (Asif et al., 2020; Jeong 2016; Lindstrom et al., 2004; Zhang et al., 2019b).

In anaerobic AGMBR systems, microorganisms that are responsible for organic removal and methane generation can accumulate on AC surface. Genus *Pseudomonas* in suspended sludge stimulates biofilm formation on GAC surface by releasing exopolysaccharides. The accumulations of phyla *Proteobacteria*, *Bacteroidetes* and *Firmicutes* on GAC particles eliminate organic matters. The enrichment of genera *Geobacter*, *Desulfobulbus* (sulfate reducing bacteria) and *Methanothrix* on GAC particles increases methane generation. More specifically, two groups of syntrophic propionate oxidizing bacteria (SPOB), *Syntrophobacter* and *Smithella*, as well as hydrogenotrophic methanogens (*Methanoregula*, *Methanolinea* and *Methanobacterium*) are highly abundant on GAC. Growth of SPOB can be realized by utilizing/degrading propionate in syntrophic association with hydrogenotrophic methanogens, leading to generation of acetate and hydrogen gas:

$C_3H_5O_2$ (propionate) + $3H_2O \rightarrow C_2H_3O_2^-$ (acetate) + $HCO_3^- + H^+ + 3H_2$ (1) (by *Syntrophobacter)*

$C_3H_5O_2 \rightarrow C_4H_7O_2^-$ (butyrate) (2) (via a dismutating pathway)

$C_4H_7O_2^- + 2H_2O \rightarrow 2C_2H_3O_2^- + H^+ + 2H_2$ (3) (by *Smithella*)

Consequently, genus *Methanothrix* as acetoclastic methanogen becomes dominant in microbial communities as *Methanothrix* has high affinity for acetate. Genera *Geobacter* and *Desulfobulbus*, which have capability of transferring electrons exocellularly without mediators, donate electrons to *Methanothrix* on GAC particles via direct interspecies electron transfer (DIET) as GAC has high electrical conductivity. Then methane is generated by CO_2 reduction pathway for energy recovery (Aslam et al., 2018; Lei et al., 2019; Tian et al., 2019):

$C_2H_3O_2^- + 4H_2O \rightarrow 2HCO_3^- + 4H_2 + H^+$ (4)

$HCO_3^- + 4H_2 + H^+ \rightarrow CH_4 + 3H_2O$ (5)

In addition, the long-term operation of AGMBR systems can lead to biological activated carbon (BAC) formation. When treating wastewater containing refractory compounds with large molecular mass, BAC replacement should be considered. For example, adsorption and biodegradation of non-ionogenic synthetic surfactants with large molecules (i.e. polyoxyethylene, 5×10^6) can only occur in meso- and macropores of BAC through the extracellular enzyme compounds penetrating into meso- and macropores. Then, the biodegradation products (i.e. carboxylic acid) obtained in meso- and macropores are diffused into the micropores and retained in micropores. However, they cannot be biodegraded as the extracellular enzyme compounds are not able to be diffused into the micropores of BAC due to their larger size, resulting in deteriorated adsorption capability. On the other hand, some of non-biodegradable organics (i.e. phenol) are mainly removed by irreversible adsorption onto AC. This is accomplished via an initial formation of strong donor-acceptor complexes between surface carbonyls and lactones of AC and the organics, followed by subsequent surface polymerization of the organics by superoxo radicals that are formed by the oxygen adsorbed on AC. Consequently, the final products and the adsorbed organics gradually reduce the capacity of AC, limiting further adsorption of organics (Satyawali et al., 2009; Sirotkin et al., 2001). Besides, fine colloids, dead cells and microbial products (i.e. extracellular polymeric substances (EPS)) generated by attached biomass might deposit into BAC pores, exhausting adsorption capacity. Attached biomass can detach from aged BAC, forming bioflocs similar like suspended sludge flocs, which could aggravate biofouling by increasing sludge concentration and viscosity in the bioreactor due to presence of the bioflocs (dos Santos and Daniel, 2020; Ng et al., 2013). Thus, it is suggested to implement BAC renewal or replacement to maintain bioactive carbon and provide high surface area for adsorption of pollutants. Furthermore, the replacement ratio should be also taken into account. Zhang et al.

(2019b) pointed out that the optimal PAC replacement ratio (1.67%) favoured microbial diversity compared to other ratios (0%, 1.25%, 2.50%) in terms of COD and NH_4^+-N removals, and effectively mitigated membrane fouling by minimizing soluble microbial products (SMP) in aerobic AGMBR.

2.1.2. Polymeric carriers

Veolia Water Technologies-AnoxKaldnes has developed the first polymeric carriers, namely the K-series AnoxKaldnes™ (e.g. K1, K2, K3), while K1 and K3 have been widely used in moving bed membrane bioreactors. These carriers are made of polyethylene with density of 0.95 g/cm^3 and have a cross in the center which divides them into circular sections by longitudinal ridges (or “fins” on the external surface). They possess different dimensions (diameter and height), available surface areas for biofilm growth and sections defined by rims. K1 carrier has relatively small dimension of length 7 mm and diameter 10 mm (surface area 500 m^2/m^3). The later developed carriers have larger dimension with length and diameter about 15 mm (surface area 350 m^2/m^3) for K2, and length 12 mm and diameter 25 mm (surface area 500 m^2/m^3) for K3. BiofilmChips have also been developed by AnoxKaldnes™, which have larger surface area of 900-1200 m^2/m^3. The increased surface area can control biofilm thickness, thereby retaining active biofilm and increasing mass transfer in biofilm. Nevertheless, the growth and aging of biofilms on BiofilmChips cause pore blocking, which reduces the available surface area and limits mass transfer. To overcome this, Z biocarriers (normal diameter 30 mm, density 0.95 g/m^3, grid height 200-400 µm) were developed with a grid designed on the media, which enables biofilm to grow outside the carrier within a protected environment. As the collision between different carries in the reactor induces lower biofilm height than the grid height, the biofilm thickness can be well controlled (Bassin and Dezotti, 2018; di Biase et al., 2019).

Other manufactures (e.g. Headworks, Nexom, Biowater Technology, Bioprocess H_2O, Warden Biomedia, etc.) have manufactured different types and sizes of biocarriers (i.e. AC450 (PE), Bioportz™, BWT X, BWT S, BioFAS™ B-460, Cylinder Plus, etc.) (di Biase et al., 2019). Table 1 summarizes the properties of common polymeric carriers.

Table 1.

Polymeric carriers can be made into various shapes, such as cylinder carriers, spherical carriers, flat rigid square meshes, fiber bundles and sponge cubes. Generally, carriers mitigate membrane fouling and maintain membrane permeability through 1) reducing the attachment, deposition and accumulation of biomass onto membrane surface; 2) exerting positive effects on the reduction of liquid-phase organic substances because of biofilm's low yield and decay rate as well as less SMP formation; 3) adsorbing or entrapping colloidal and soluble organics (e.g. SMP, biopolymer clusters, strongly and weakly hydrophobic components with small molecular weight (< 10 kDa) in SMP) into biofilm and inside carriers (Rodríguez-Hernández et al., 2014; Sun et al., 2015). The effects of carriers with different shapes on conventional pollutants removal and membrane fouling control in AGMBRs are listed in Table 2.

Table 2.

When the polymeric carriers are prepared in cylindrical shape (generally made from PE and PP), the growth of attached biofilm on carriers demonstrates three stages: 1) the attachment and formation of young biofilm on the carrier (maroon color). During this stage, the gradual attachment of microorganisms on carriers induces increased thickness but uneven distribution of biofilm; 2) the accumulation and combination of biofilm on the carrier (black color), obtaining maximum thickness of biofilm, followed by the growth of microorganisms along the carrier surface and further integrated biofilm formation; 3) biofilm maturation (dark black color) with full coverage of biofilm on carrier surface, leading to continuous and smooth biofilm formation. When biofilm growth reaches a certain degree, a balance is obtained between growth and detachment of biofilm (Tang et al., 2016). The growth of heterotrophic microorganisms in attached biofilm can assimilate readily and slowly biodegradable substrates (Cuevas-Rodríguez et al., 2015). Moreover, the attached biofilm increases hydrolytic activities and enhances the hydrolysis of organic matters (i.e. macromolecules) using extracellular microbial enzymes (the important rate-limiting step for organic degradation). This facilitates subsequent utilization of the hydrolyzed products by bacterial metabolism and thus enhances organic removal (Reboleiro-Rivas et al., 2013).

The attached biofilm on cylindrical carriers also prolongs SRT and creates an environment favorable for enrichment of slow-growing microorganisms (i.e. nitrifying bacteria). Furthermore, the outer layer of biofilm encourages the formation of aerobic micro-environment, while anoxic zone is formed in inner layer of biofilm caused by the oxygen concentration gradient due to the limited oxygen diffusion, which favors simultaneous nitrification and denitrification process (Cuevas-Rodríguez et al., 2015; Palmarin and Young, 2019a; Reboleiro-Rivas et al., 2013). Anoxic and anaerobic microenvironment in the inner layer of attached biofilm might increase anoxic P-uptake activity, leading to higher potential of attached biomass for phosphate removal

(Costa et al., 2019; Xu et al., 2018). When applying the carriers with small dimension (i.e. height 10 mm, diameter 10 mm), decline in floc size of suspended sludge (i.e. about 150 µm) increases the abundance and activity of phosphate-accumulating organisms (PAOs). This is ascribed to that the smaller flocs have higher pore width and larger surface area than larger flocs, which are more favorable for mass transfer (easier penetration of phosphorus and oxygen into flocs) and biological reaction (easier phosphorus removal and higher phosphorus removal activity) (Xu et al., 2018; Wu et al., 2010). However, the small flocs can exacerbate membrane fouling due to pore blocking. On the other hand, the carriers with larger dimension (i.e. height 12 mm, diameter 24 mm) can retard membrane fouling by enriching the floc-forming bacteria in suspended sludge, enlarging floc size, accumulating floc-forming, hydrolytic and fermentative bacteria on membrane surface, as well as enabling hydrolysis and degradation of fouling materials (i.e. proteins, SMP and EPS) (Alresheedi and Basu, 2014; Zheng et al., 2019a and 2019b; Zheng et al., 2020).

It is worth to mention that porous sponge cubes exhibit superiorities over the aforementioned carriers with respect to conventional and emerging pollutants removal as well as fouling control. Sponge cubes ensure higher growth of attached microorganisms, which can utilize more organic substrates and increase COD removal. The immobilization of microorganisms on sponge by entrapment and attachment extends SRT, thus prompting growth of nitrifying bacteria on carriers and further increasing NH_4^+-N removal. The formation of aerobic and anoxic zones at the outer layer of sponge facilitates simultaneous nitrification and denitrification process, which favors NO_3^--N and TN removal. Moreover, anaerobic and/or anoxic condition formed at the inner layer of the sponge cubes improves phosphorus removal.

Generally, PAOs utilize energy which is generated from hydrolysis of intracellular polyphosphate and release of phosphorus to assimilate carbon substrate and further produce poly-β-hydroxybutyprate (PHB) as internal carbon storage under anaerobic condition. However, when NO_3^--N accumulates under anaerobic condition, denitrifying bacteria use carbon substrates for denitrification prior to PAOs, which is unfavorable for phosphorus release. To the contrary, the less residual NO_3^--N results in more carbon source available for PAOs, facilitating phosphorus release. Then more phosphorus is taken up by PAOs under aerobic condition. Additionally, denitrifying phosphate-accumulating bacteria (DPB) as a fraction of PAOs employ NO_3^--N as electron donor in anoxic zone to oxidize PHB, accomplishing denitrification and phosphorus uptake simultaneously (Nguyen et al., 2017; Yang et al., 2010). Sponge cubes also minimize cake layer formation and prevent pore blocking. Moreover, they enlarge sludge flocs by increasing proteins to polysaccharides ratio in bound EPS that positively affect the agglomeration ability of sludge flocs. Foulants (i.e. proteins, carbohydrates, SMP and EPS) can be adsorbed on sponge cubes and subsequently biodegraded by attached microorganisms (Deng et al., 2014; Fu et al., 2016). When treating wastewater containing emerging pollutants (i.e. antibiotics), the attached biomass on sponge encourages the accumulation and growth of slow-growing microorganisms, which provides enough acclimatization time for these microorganisms to adapt to the emerging pollutants and further helps the pollutants removal (Nguyen et al., 2017).

2.2. New media

Biochar, as a carbonaceous material, is obtained through thermochemical conversion of biomass, i.e. agricultural waste (rice husk, corn stalk, pomelo peel, etc.), algal biomass, plants (pine bark, bamboo, apple tree, etc.), manures, activated sludge, energy crops, digestate, etc. It is a promising and cost-effective alternative to activated carbon, which possesses abundant functional groups (e.g. COOH, OH, R-OH), high adsorption capacity, high specific surface area, and high porosity. It not only enables the formation of dense and thick attached biofilm but also allows good COD removal (> 80%) (Xia et al., 2016; Zhang et al., 2017b). In aerobic AGMBR, considerably high NH_4^+-N removal (~ 100%) can be obtained. Additionally, enhanced TN and TP removal are detected via forming the anoxic condition inside the biochar compared to single aerobic MBR (Zhang et al., 2017b). In anaerobic AGMBR, biochar addition also improves NH_4^+-N removal via adsorption. Moreover, methane generation is enhanced by the increased abundance of *Geobacter* and *Methanosarcina* participating in direct interspecies electron transfer (DIET) as well as enrichment of some methanogens (i.e. *Methanosaeta*, *Methanospirillum*, *Methanobacterium*) (Chen et al., 2020; Xia et al., 2016). Biochar can reduce SMP, EPS, colloids and dissolved substances by adsorption and biodegradation by attached biofilm, as well as enhance sludge properties (i.e. settleability) by biomass retention and microbial colonization. Hence, membrane fouling can be effectively alleviated (Chen et al., 2020; Xia et al., 2016).

Bioaugmentation, as a new wastewater treatment technology, artificially immobilizes special functional bacteria or entraps biomass into the bioaugmented carriers. It aims to maximize contributions of bacteria and biomass on carriers to remove pollutants by protecting the microorganisms from washout and adverse conditions (i.e. toxic compounds, high shear stress, etc.). Up to now, bioaugmented carriers have been reported to eliminate targeted pollutants or treat different kinds of wastewaters (Table 3). Although bioaugmented carriers could improve the removal and biodegradation of specific pollutants (e.g. hydrocarbons, nonylphenol, high concentration of nitrogen, etc.) as well as resist salinity shock from wastewater, continuous accumulation of some pollutants (i.e. unremoved petroleum hydrocarbons, recalcitrant intermediate products) during the long-term operation might exhibit their toxicity to the immobilized bacteria. Moreover, the capacity of the immobilized bacteria for biodegradation of targeted pollutants is limited under certain shock loading conditions. For instance, at very high saline levels (salinity expressed as total dissolved solids (TDS), TDS ≥ 150 g/L), COD removal declined significantly (from up to 95% at TDS < 150 g/L to < 65%) because high salinity reduced the ability of the immobilized halophilic microorganisms for hydrocarbon degradation. Besides, the capability of immobilized strain for removal of azo dyes was limited by the excessive ammonium sulfate (> 1.5 g/L) (< 95% vs > 95% at ammonium sulfate < 1.5 g/L) (Azaizeh 2015; Hasanzadeh et al., 2020; Menashe and Kurzbaum, 2016; Tan et al., 2014).

Table 3.

The selected strains/mixed culture immobilized on bioaugmented carriers have also been applied in AGMBRs for treating various types of wastewaters (Table 4). When treating medium-strength domestic wastewater (i.e. soluble COD 550 mg/L), centrifuged (concentrated) anaerobic or aerobic sludge could be selected and entrapped in polyvinyl alcohol (PVA) gel beads in anaerobic AGMBR or aerobic AGMBR. In anaerobic AGMBR, soluble COD removal efficiency was comparable to that in the single AnMBR (about 84%). The entrapped cells could encourage the enlargement of average particle size in cake layer and suspended sludge, reducing cake layer resistance. In aerobic AGMBR, high removal efficiencies of soluble COD and NH_4^+-N were obtained (approximately 95% and 93%, respectively), which were similar with removals in single aerobic MBR. As the particle size of these flocs was larger than membrane pore size, the sludge flocs gave rise to cake layer formation rather than pore blocking, leading to higher cake resistance in aerobic AGMBR (8.80×10^5 m^{-1}) than that in single aerobic MBR (5.03×10^5 m^{-1}).

Nevertheless, considerable reduction of bound EPS and SMP (especially proteins) notably decreased pore blocking resistance in the AGMBR by four times (1.00×10^6 m^{-1} vs 5.03×10^6 m^{-1} in single aerobic MBR). Thus total fouling resistance and overall membrane fouling were lower in the AGMBR, extending operation duration by more than 57% compared to single MBR (Juntawang et al., 2017a and b).

Table 4.

When treating nitrogen-rich wastewater containing high concentrations of NH_4^+-N and NO_2^--N (i.e. 50-160 mg/L), the selected bacteria, anammox bacteria (*Planctomycetes*), was entrapped into macroporous carriers (MPCs, which were prepared by modifying PVA/SA with the pore-forming agents ($CaCO_3$) and post-crosslinked with glutaraldehyde). In aerobic AGMBR, the occurrence of nitrification process at the outer layer of MPCs (Eq. (6)) and anammox process in the anaerobic microenvironment inside MPCs (Eq. (7)) enhanced TN removal rates (314 mg/L·d vs 304 mg/L·d without immobilized bacteria) and reached about 80% of removal efficiency:

$NH_4^+\text{-N} + O_2 \rightarrow NO_2^-\text{-N}$ (6) (by nitrifying bacteria)

$NH_4^+\text{-N} + NO_2^-\text{-N} \rightarrow N_2$ (7) (by the immobilized anammox bacteria)

Moreover, membrane fouling could be alleviated by retaining suspended biomass and anammox bacteria in MPCs, which reduced their deposition on membrane surface and further minimized generation of foulants (i.e. EPS, SMP) by anammox bacteria (Zhang et al., 2016).

To treat high-saline pharmaceutical wastewater, the mixed cultures immobilized in PVC carriers contained marine sediment biomass having halotolerant/halophilic microorganisms taken from coastal shore and conventional activated biomass collected from a local wastewater treatment plant. In aerobic AGMBR, the mixed cultures showed high capability of improving total COD removal as they have adapted to saline conditions by possessing some marine species (i.e. *Dokdonia* sp., *Oleiphilaceac* sp., *Yeosuana* sp., etc.), which could degrade polycyclic aromatic hydrocarbons and recalcitrant compounds. Moreover, the biomass immobilized at the outer and inner layers of carriers maximized the degradation of organic matters. Hence total COD removal was higher (average 80%) compared to single MBR (average 65%). Since large portion of organic matters were removed by the mixed cultures on carriers, less organic matters were available for microorganisms in cake layer, alleviating membrane biofouling. However, NH_4^+-N removal was not satisfactory as nitrifying bacteria are sensitive to refractory compounds and high saline condition (Ng et al., 2016).

Pseudomonas putida cells, which are capable of degrading some toxic compounds (i.e. phenol, p-cresol, quaternary ammonium, etc.), have been entrapped in PVA/SA gel beads (PVA, polyvinyl alcohol; SA, sodium alginate) in order to treat hospital wastewater containing high concentration of CIP at 546 ± 48 μg/L. In an anoxic-oxic AGMBR, the entrapped cells enhanced organic removal via assimilation and CIP (> 90%) removal through releasing degradative enzymes (i.e. oxido-reductase, lyase). The improved TN removal was also partially accomplished due to the entrapped cells. It was noted that nitrite-oxidizing bacteria (NOB) and nitratation process (NO_2^--N → NO_3^--N) could be partially inhibited by CIP, leading to the accumulation of NO_2^--N. This depressed complete nitrification and further inhibited NH_4^+-N removal. Fortunately, the subsequent anoxic unit enabled anammox process, which reduced NO_2^--N and NH_4^+-N to N_2 gas, resulting in better TN removal and good NH_4^+-N removal (45.7% and 87.5%, respectively) compared to those without entrapped cells (16.6% and 90.3%, respectively).

In addition, the PVA/SA beads had porous structure, which facilitated the growth of *P. putida* cells. Moreover, the amount of *P. putida* in beads was maintained at a relatively stable level throughout the experimental period (Hamjinda et al., 2017).

It has been known that bacterial quorum sensing (QS) as a cell-cell communication system is employed by bacteria to coordinate group behaviors (e.g. biofilm formation, population density, motility, SMP and EPS production, exocellular enzyme secretion), which involves the production and release of signal molecules, N-acyl homserine lactone (AHL) as autoinducers. Quorum quenching (QQ) can destroy autoinducer molecules to inhibit QS activities, which effectively mitigates membrane fouling via minimizing biofouling without negative effects on bacterial growth. QQ bacterium (generally *Rhodococcus* sp. BH4) is generally entrapped/immobilized in various types of media, such as cells entrapping beads, hollow cylindrical media, sheets (e.g. glass board), layered beads, vessels (e.g. polyethylene hollow fiber membranes). This combines biological QQ effect with scouring membrane effects and enhances QQ activity to mitigate membrane fouling through improving sludge properties (i.e. declined EPS and SMP levels, increased sludge settleability, decreased apparent viscosity, etc.) (Ham et al., 2018; Iqbal et al., 2020; Köse-Mutlu et al., 2016; Jiang et al., 2013; Lee et al., 2016; Nahm et al., 2017).

3. Performance of AGMBRs with attached growth media

3.1. Conventional pollutants removal and membrane fouling control

Aerobic AGMBR with PAC addition showed better removals of COD (> 92%), NH_4^+-N (> 94%), TN and TP (up to 85%), as well as prolonged filtration time by almost 2 times compared to single aerobic MBR (TN and TP removal efficiencies < 75%) (Asif et al., 2020; Zhang et al., 2019a). Anaerobic AGMBR with GAC or PAC addition demonstrated high COD removal (≥ 87%) and low TMP development. The high retention of biomass on GAC particles induced less suspended growth and further reduced the generation of foulants (e.g. SMP, EPS, etc.) in bioreactor. Moreover, membrane fouling also declined due to the scouring effects of GAC or PAC and less cake layer formation on membrane surface. On the other hand, Aslam and Kim (2019) pointed out that in anaerobic AGMBR, irreversible fouling induced by small colloids could not be effectively removed by GAC fluidization. If proteins and carbohydrates were accumulated on membrane as dominant biofoulants, physical scouring by GAC fluidization was ineffective in removing these biofoulants due to the formation of gel layer on membrane surface. However, combining chemical cleaning (sodium hypochlorite, 25 mg/L) with periodic membrane relaxation was able to minimize biofouling (Aslam et al., 2018).

Aerobic AGMBR with cylindrical polymeric carriers could achieve good organic removal (> 90%) and NH_4^+-N removal (> 95%) comparable to those in aerobic MBR. Moreover, TN (> 68-80%) and TP removals (> 90%) were better than those in aerobic MBR (64-77% and < 60%, respectively). Membrane fouling was also effectively controlled with decreased fouling rate (only 37% of that in the aerobic MBR). Additionally, there was no loss of membrane integrity. When limited nutrients presenting in feed water (i.e. grey wastewater with NH_4^+-N 2.99 ± 0.97 mg/L, TN 6.35 ± 1.18 mg/L, TP 0.534 ± 0.147 mg/L), excessive growth of filamentous bacteria and poor sludge settleability (SVI 295-445 mL/g vs 307-341 mL/g without carriers) were observed, which adversely influenced membrane fouling control (Costa et al., 2019; Palmarin and young, 2019a and 2019b; Xu et al., 2018; Zheng et al., 2020). In anaerobic AGMBR, cylindrical polymeric carriers stimulated high organic removal (around 85% of COD). Under periodic backwashing operation (i.e. 0.5-min backwashing every 60-min filtration), TMP could be maintained at low level for long time (i.e. < 10 kPa for 240 days) without chemical cleaning, owing to physical scouring of the carriers on membrane surface, as well as degradation of EPS and SMP by active microorganisms in attached and suspended biomass (Kim et al., 2020).

Performance of aerobic AGMBRs with other typical shapes of polymeric carriers also demonstrated good performance when removing conventional pollutants. For example, the application of porous spherical carriers ensured high capability of resisting organic shock load (i.e. COD 48-1118 mg/L) and enhanced NH_4^+-N and TN removals (> 90% and > 65%, respectively), compared to aerobic MBR (about 78% and 55%, respectively). Membrane fouling was also lower than that in aerobic MBR (Premarathna et al., 2019; Singh et al., 2016). Improved organic and nutrient removals (84-98%, 97%, 75%, 42% of COD, NH_4^+-N, TN and PO_4-P removals, respectively) could be accomplished in aerobic AGMBR with flat rigid square meshes, compared to single aerobic MBR (80-96%, 93%, 38%, 37% of COD, NH_4^+-N, TN and PO_4-P removals, respectively). The reduction of membrane foulants decreased membrane fouling rate in aerobic AGMBR by approximately 43% of that in aerobic MBR (Rodríguez-Hernández et al., 2014; Sun et al., 2015).

The enhanced treatment performance was observed in aerobic AGMBR with sponge cubes, including comparable COD removal to that in aerobic MBR (85-96%), better NH_4^+-N (> 73%), TN (25-52%) and TP removals (53-61%) than those in aerobic MBR (> 58%, 12-36% and 27-55%, respectively). Compared to single aerobic MBR, better filtration performance was also achieved, while cake layer resistance and fouling rate could be significantly reduced by more than 40% and around 2.8-10 times, respectively. During long-term operation (i.e. > 190 d), the bridging effect between inorganic matters and biopolymers could induce membrane pore blocking, increasing pore blocking resistance (Deng et al., 2014; Fu et al., 2016). For anaerobic AGMBR, the sponge cubes provide anoxic and anaerobic microenvironments inside the sponge, which could not only favor good organic removals (> 90%), but also improve TN and PO_4-P removals (reaching around 32% and 36%, respectively) compared to single AnMBR (around 15% and 17%, respectively). The declined SMP and EPS in mixed liquor reduced cake layer and pore blocking resistances, which decreased total fouling resistance by 50.7% (Chen et al., 2017).

Currently, biochar (i.e. bamboo charcoal) is increasingly used in AGMBRs for removing conventional pollutants. Biochar as support material induced the enrichment of microorganisms, giving rise to the formation of a dense and thick biofilm, and further increasing microbial diversity. In aerobic AGMBR, biochar improved TN and TP removals by 15% and 15-20%, respectively compared to those in aerobic MBR (TN and TP removals < 10%). However, the TN and TP removals were not as high as that in aerobic AGMBRs with other types of carriers (i.e. PAC, polymeric carriers, sponge cubes). After adding biochar, higher enrichment of some functional microorganisms, which are favorable for the degradation of foulants (protein and carbohydrate in EPS and SMP) (i.e. *Aminomonas*, *Anaerofustis*, *Anaerolinea*, etc.), alleviated membrane fouling (Zhang et al., 2017b). In anaerobic AGMBR, better COD removal (94.5% vs 89.1% in AnMBR) could be obtained and biochar could also adsorb a portion of NH_4^+-N. Additionally, the increased biofilm thickness on biochar could be considered as a granulation process through aggregate formation. Thus, larger granular sludge size (higher proportion of particle size distribution over 100 µm) was realized in anaerobic AGMBR, resulting in the formation of a loose and thin cake layer on membrane surface. Moreover, biochar also reduced foulants (i.e. colloids, SMP, proteins, polysaccharides), which led to less pore blocking and total fouling resistance (Xia et al., 2016).

In anaerobic AGMBR, the addition of carriers also improves methane generation and enriches microorganisms contributing to methane generation. After adding GAC, the enriched SPOB (*Syntrophobacter* and *Smithella*), *Geobacter*, *Desulfobulbus* and *Methanothrix* on GAC particles enhanced methane generation via DIET. It was found that the net average total standard methane yield (including gaseous and dissolved methane) was higher in anaerobic AGMBR (0.17 ± 0.05 L/g) than that in single AnMBR (0.14 ± 0.06 L CH_4/g COD). Furthermore, high conversion efficiency of COD to methane (i.e. > 85% with 83% of CH_4 in biogas) was achieved during stable operation (Aslam and Kim, 2019; Chaiprapat et al., 2016; Shin et al., 2014). Net electrical energy obtained from methane could be significantly higher in anaerobic AGMBR with GAC addition. It was reported that if 35% energy transfer efficiency for converting methane to electricity was considered, the net electricity energy yield was about 9.8 times higher than the energy required for operating the anaerobic AGMBR system (Aslam et al., 2018). When operating with low influent COD concentration (i.e. 210 ± 50 mg/L), total energy available from methane combustion was not high (0.10 kWh/m^3). Nevertheless, the obtained energy could meet 70% of total energy consumption need for system operation (0.15 kWh/m^3). Thus, the requirement of net input energy for operating the AGMBR system was equal to 0.05 kWh/m^3 (30% of total energy requirement) compared to that for conventional aerobic systems (> 0.3-0.6 kWh/m^3) (Evans et al., 2019).

In anaerobic AGMBR with cylindrical polymeric carriers, gas production and methane composition in biogas were high (up to 0.216 L CH_4/g $COD_{removed}$ and 75%, respectively). This could be ascribed to the enrichment of propionate-degrading *Syntrophobacter*, sulfate-reducing *Desulfobulbus* and methane producing acetoclastic *Methanothrix*. Therefore, total energy requirement was considerably smaller (0.0109 kWh/m^3), and the electricity obtained from methane production was remarkably higher (0.246 kWh/m^3 considering 33% of pumping efficiency) than the total energy required (Kim et al., 2020). The addition of sponge cubes also encouraged the accumulation and growth of methanogens, leading to higher biogas and methane yield in anaerobic AGMBR (486 ± 12 mL/d and 0.156 ± 0.005 LCH_4 /$COD_{removed}$, respectively) than the single anaerobic MBR (456 ± 9 mL/d and 0.133 ± 0.005 LCH_4/$COD_{removed}$, respectively) (Chen et al., 2017).

Biochar addition enhanced the abundance of functional microorganisms for methane generation, i.e. *Methanosaeta*, *Methanospirillum*, *Methanobacterium*, etc. Moreover, biochar could be considered as an electron transfer conduit similar like GAC involving DIET for enhancement in methane generation. Consequently, biogas production and methane production increased in anaerobic AGMBR (13.2 L/d and 0.25 LCH_4/g COD, respectively) compared to those in AnMBR (10.3 L/d and 0.13 LCH_4/g COD, respectively) (Xia et al., 2016).

3.2. Emerging pollutants removal

Emerging pollutants normally possess high toxicity, bioaccumulation potential, and persistence in aquatic environments. Moreover, some refractory compounds (i.e. polynuclear aromatic hydrocarbons, long-chain hydrocarbons and nitrogenous heterocyclic compounds) in wastewater are carcinogenic and mutative. Thus, these pollutants cause negative effects on the environment and human health (Lim et al., 2019; Wang et al., 2015; Xiao et al., 2017). To date, AGMBRs with different types of carriers, as one of effective strategies, have been increasingly employed in removing emerging pollutants.

3.2.1. Removals of micropollutants

The removal pathways of micropollutants are mainly determined by hydrophobicity (the octanol-water distribution coefficient log D for neutral (uncharged) and ionisable compounds; the octanol-water partition coefficient log K_{ow} for neutral (uncharged compounds)) and molecular structure, such as electron withdrawing groups (EWGs) and electron donor groups (EDGs). Generally, hydrophobic compounds (HOC) (log D (pH= 7) > 3.2, or high log $K_{ow} > 5$) show high sorption tendency, favoring sorption of these compounds onto suspended sludge and attached biomass on media, which facilitates the subsequent biodegradation. On the other hand, hydrophilic compounds and moderately hydrophobic compounds (HIC) are the compounds having lower log D and log K_{ow} compared to HOC (log D (pH= 7) < 3.2, or low log $K_{ow} < 2.5$ for hydrophilic compounds; $2.5 \leq \log K_{ow} \leq 5$ for moderately hydrophobic compounds). In single MBRs, the removals of HIC vary significantly, determining by their biodegradability and molecular structure (EDGs and EWGs). Accordingly, these compounds are divided into two different groups: 1) HIC1 containing EWGs and/or weak EDGs, which show low biodegradation and thus low or moderate removals; and 2) HIC2 containing various EDGs or strong EDGs, which show high biodegradation capacity, demonstrating relatively high removals. Moreover, higher log K_{ow} value could increase adsorption of the compounds (de Almeida Lopes et al., 2020; Tiwari et al., 2017; Zhang et al., 2021).

The HOC (log D > 3.2) with the presence of EDGs (hydroxyl (-OH), methyl ($-CH_3$)) in their molecular structure showed high removals (> 80%) in single aerobic MBR due to their high adsorption onto suspended sludge and the subsequently enhanced biodegradation, including TCS, E1, EE2, E2, β-estradiol 17–acetate, 4-tOP, TBP, 4-NP and BPA (Fig. 1). In aerobic AGMBR, these compounds could be further removed through adsorption onto GAC (Nguyen et al., 2012). Moreover, the hydrophobic compounds were also adsorbed onto attached biomass on GAC by diffusing into the lipophilic cell membrane, which was caused by hydrophobicity of these compounds (Lim et al., 2019; Sipma et al., 2010). Consequently, adsorption of hydrophobic compounds onto GAC and attached biomass promoted biodegradation of these compounds. This led to almost complete removals of these hydrophobic compounds (Lim et al., 2019; Nguyen et al., 2012).

Fig. 1.

HIC1 have one or more strong EWGs (i.e. halides (-Cl, -F, -Br. -I), amide ($–CONH_2$), nitro group ($-NO_2$), esters (-COOR)) and/or weak EDGs (i.e. methyl ($-CH_3$)) in their structures. Thus, HIC1 exhibit low biodegradation (or being recalcitrant to biological treatment), resulting in low removals in single aerobic MBR (< 66%) (Fig. 2). The compounds include DCF with –NH, -Cl and -COOH, CBZ with $-CONH_2$, DZP with $-CH_3$ and –Cl, KEP with $-CH_3$ and –COOH, CAF with only $–CH_3$, PRM with $-CH_3$ and -NHCOR, 2,4,5-TP with $–CH_3$, -COOH and –Cl, and NPX with $-CH_3$ and –COOH. The addition of activated carbon (GAC, PAC) could significantly enhance their removals to almost complete removal in aerobic AGMBR by adsorption, which might be attributed to hydrophobicity-independent mechanisms (e.g. ion exchange, surface complexation, hydrogen bonding, electrostatic interactions, van-der-Waals forces, pore filling, π-π electron-donor acceptor (EDA) interactions, cation-π bonding, Lewis acid-base interaction, etc.) (Alvarino et al., 2017; Dittmann et al., 2020; Liu et al., 2015; Nguyen et al., 2012; Serrano et al., 2011). It should be noted that removals of some compounds (i.e. CBZ, DZP, DCF) declined with time were ascribed to i) pore blocking of PAC as the competition between organic matters and micropollutants in available adsorption sites, leading to saturation of active pores, and ii) insufficient dose of PAC (Alvarino et al., 2017; Serrano et al., 2011).

On the other hand, there are some exceptions for removals of HIC1, which possess EWG (-COOH) and high numbers of the weak EDG methyl group (-CH_3), i.e. GEM with four methyl groups (–CH_3), and IBP with three -CH_3, which showed considerably high removals ranging from 76% to 100% in single aerobic MBR. After adding activated carbon, adsorption dominates the removal of GEM, which might be owing to its relatively high log K_{ow} value (4.30), while the removals of IBP with lower log K_{ow} value (3.5) were mainly due to the biodegradation by attached biomass on activated carbon. As a result, GEM and IBP could accomplish close to 100% of removals in the aerobic AGMBR (Alvarino et al., 2017; Asif et al., 2020; Luo et al., 2015; Monteoliva-García et al., 2020; Nguyen et al., 2012; Serrano et al., 2011).

Fig. 2.

Single aerobic MBR can remove more than 70-80% of HIC2. HIC2 containing various EDGs could be almost completely removed in aerobic AGMBR through the adsorption onto activated carbon and enhanced biodegradation by attached biomass on activated carbon, especially OTC with –OH, -NR_2 and –CH_3, SMX with -NH_2 and -CH_3, TPH with –NH and -CH_3, E3 with –OH and –CH_3, ROX with –OH and –CH_3, and AZM with –OH, -NR_2 and –CH_3.

Besides, the main pathway for the removals of HIC2 with strong EDGs despite the presence of EWGs in aerobic AGMBR is adsorption onto activated carbon (hydrophobicity-independent mechanisms), i.e. FLX with strong EDG (-O-), other EDGs (-NH, $-CH_3$) and EWG ($-CF_3$), PCP with strong EDG (–OH) and weak EWG (–Cl). Two exceptions are removals of Tmp and MTZ in single aerobic MBR. Tmp has more than one EDGs ($-OCH_3$ and $–NH_2$) and MTZ has strong EDG (-OH) and one weak EDGs ($-CH_3$) despite having EWG ($–NO_2$). However, their removals were moderate (< 65%) in single aerobic MBR, which might be due to their very low log K_{ow} (0.9-1.4 and -0.14, respectively). In the aerobic AGMBR, removals of Tmp and MTZ were significantly improved in the AGMBR, reaching almost complete removal owing to adsorption onto activated carbon (hydrophobicity-independent mechanisms) and attached biomass on activated carbon (i.e. electrostatic force interactions) (Alvarino et al., 2017; Luo et al., 2015; Nguyen et al., 2012; Serrano et al., 2011).

Besides, nitrification needs to be taken into account for the removals of some hydrophilic compounds containing EDGs. The removal of ERY with various EDGs (-OH, $-CH_3$, $-NR_2$, and alkoy ($-OCH_3$)) varied in single aerobic MBR at different nitrification efficiencies (i.e. 80% removal with 98% nitrification, 42-64% removal with 70-80% nitrification). The addition of activated carbon enhanced adsorption and biodegradation of ERY, reaching almost 100% of removal in aerobic AGMBR (Serrano et al., 2011; Alvarino et al., 2017). It was found that the enhanced removals in aerobic AGMBR with PAC could be partly ascribed to the high enrichment of special functional microorganisms for recalcitrant micropollutants removal (biodegradation), including nitrifying and denitrifying bacteria (i.e. *Methylotenera* and

Haliangium) and other bacteria (e.g., *Comamonas* and *Methanomethylovorans*) (Asif et al., 2020).

In aerobic AGMBR, sponge cubes removed hydrophobic compounds, most of HIC1 and HIC2 with the presence of EDGs despite of EWGs mainly by biodegradation, i.e. PCM/ACT with –OH and -NHCOR, SA with -OH and -COOH, E3 with –OH and –CH_3, NPX with -CH_3 and -COOH, IBP with -CH_3 and -COOH, GEM with -CH_3 and –OH groups, PRM with -CH_3 and -NHCOR, etc. The attached biofilm on sponge cubes enables the formation of highly diverse bacterial community for effective biodegradation of micropollutants. Moreover, the retention of biomass prolongs contact time between the compounds and microorganisms, which allows microorganisms to obtain sufficient time for acclimatization, thereby enhancing biodegradation of compounds. Besides, sorption onto attached biomass on and inside sponge cubes (i.e. electrostatic force interactions) also contributes to remove some HIC with relatively high log K_{ow} values, including three HIC1 (2,4,5-TP, log K_{ow} 3.45; DCF, log K_{ow} 4.5-4.8; CBZ, log K_{ow} 2.3-2.5) and one HIC2 (PCP, log K_{ow} 5.12), as well as some hydrophobic compounds (TBP and 4-tOP). Although most of compounds could be removed by above 70-80% or even close to 100%, the removals of MTZ, 2,4,5-TP, DCF and CBZ were less than 50% in aerobic AGMBR with sponge cubes due to their hardly biodegradable properties (Luo et al., 2015).

Monteoliva-García et al. (2020) pointed out that cylindrical polymeric carriers could improve removals of CIP with two EWGs (–COOH and –F) by enhanced adsorption onto attached biomass (i.e. electrostatic force interactions, divalent bridges mechanisms), reaching almost 100% in aerobic AMBR. The removal of IBP was also increased to 100% in aerobic AGMBR through the improved biodegradation, which was owing to 1) the attached biomass on carriers providing larger number of microorganisms and higher diversity of microorganisms, and

2) the higher SRT for retention of more pollutants and microorganisms. On the other hand, CBZ was removed by less than 85% as the compound possesses only one EWG (–$CONH_2$) and relatively low log K_{ow} (2.3-2.5).

Higher concentrations of micropollutants favor their removals in aerobic AGMBR with sponge cubes. High and stable CIP removal (78-94%) was achieved with initial CIP concentration of 20-200 µg/L through adsorption onto suspended biomass via electrostatic force interactions and divalent bridges mechanisms. Although high concentration of CIP induced the detachment of biomass from sponge cubes and increase in suspended biomass in the bioreactor, it was interesting that the declined levels of attached biomass on sponge cubes reduced density of the carriers and further increased their velocity in the bioreactor, encouraging stronger scouring effects on membrane surface, which in turn accomplished effective membrane fouling control (Nguyen et al., 2019). In another study, the anoxic-aerobic AGMBR containing *Pseudomonas putida* (*P. putida*) cells entrapped PVA/SA-gel beads could enhance CIP removal (90% vs 58% in single MBR) with influent CIP of 546 ± 48 µg/L. It was ascribed to that *P. putida* attached on beads had high biodegradable capacity for recalcitrant compounds by releasing various degradative enzymes, e.g. oxidoreductase (dioxygenases) and lyase (decarboxylase, C-N lyase), as well as increased amount of active biomass to enhance biodegradation capacity for CIP (Hamjinda et al., 2017).

Compared to AnMBR, PAC addition significantly increased sorption of some micropollutants (Tmp, CBZ, SMX, DCF, and TCS) to solids (mixture of sludge and PAC) in anaerobic AGMBR by more than 3.5 times via hydrophobicity-independent mechanisms. The increased local concentration of micropollutants around adsorptive sites of PAC led to more thermodynamically favorable biodegradation (Fig. 3; Xiao et al., 2017). Anaerobic AGMBR with GAC also effectively removed some selected micropollutants through sorption onto GAC as well as their sorption and biodegradation by attached biofilm on GAC. As a result, the removals were high at 86-100% for SDZ, NA, FMQ, PA, CIP, OFL, CPX, CED, Tmp, CBZ, CAF, POF, ACT, IBP, NPX, KEP, SMX, ERY-H_2O and CAM, although some of them have EWGs (i.e. -COOH, -F). However, DCF was only eliminated by 78.2% as it is recalcitrant to biodegradation (Dutta et al., 2014; Lim et al., 2019).

Fig. 3.

Anaerobic AGMBR with a pomelo peel derived biochar could enhance the removal of sulfonamide antibiotics containing high levels of SMX, SDZ and SMZ (100 μg/L each) in wastewater with pH of approximately 7.5. Biochar slightly affected SMX removal because of easily biodegradable property of SMX, reaching 89.37-97.29% in AGMBR. Both SMZ and SDZ with EDGs (-NH_2, -CH_3) have lower log K_{ow} (-0.09 and 0.62, respectively) than SMX (0.79), facilitating biodegradation. The addition of biochar could improve removals of SMZ and SDZ, reaching up to 80% in anaerobic AGMBR (20-47% in single anaerobic MBR) through encouraging the biofilm formation on the biochar surface, enhancing activity of microorganisms and further biodegrading compounds (Cheng et al., 2021).

3.2.2. Removals of other types of emerging pollutants

It was also found that aerobic AGMBR, which contained an efficient PHN-degrading bacterium *Pseudomonas sp.* strain (strain LZ-Q) entrapped in ceramics, could remove 96% of 20 mg/L PHN and maintain stable degradation ability for 60-day operation for treating PHN-contaminated wastewater with high pH of 8 and salinity of 35 g/L. The removal process was accomplished via two metabolic pathways, namely, SA pathway with SA and CTC as intermediates, and PHA pathway with PHA as intermediates (Jiang et al, 2015).

Anaerobic AGMBR with GAC demonstrated high removal efficiency (92.6 ± 10.4%) for treating low-strength wastewater (COD of 150-300 mg/L) containing high levels of 2-CP (5.0-11.2 mg/L) within 92 days of operation. It should be noted that both 2-CP and COD loading shocks (sudden increase from 5.0 to 9.9 mg/L and from 150 to 157.3 mg/L, respectively) limited the 2-CP removal (only 88.3 ± 15.8%). Nevertheless, the removal efficiency could be recovered to 90.3 ± 14.2% after around 10 days of restoration (Wang et al., 2015). Chen et al. (2020) found that anaerobic AGMBR with biochar at relatively high dosage of 4 g/L had a potential in treating pharmaceutical wastewater containing relatively low AOX level (3.75 ± 1.2 mg/L). The average removal of AOX increased from 56.2% in single AnMBR to 61.5% by adsorption in the AGMBR (Chen et al., 2020).

Compared to the single MBR, addition of sponge cubes could remove more types of intermediate products generated during aromatic compounds biodegradation (including TL, EB, STY and other refractor intermediates) in anoxic/aerobic AGMBR due to the enrichment of special functional bacteria (i.e. *Flavobacterium*, *Holophaga*, and *Geobacter*) in sponge carries (Chen et al., 2021).

4. Future perspectives

The application of AGMBR with conventional media (including PAC, GAC and polymeric carriers in different shapes) in wastewater treatment has been largely investigated in recent years. It has been pointed out that AC mainly suffers from the deterioration of sorption capacity during long-term operation. Some polymeric carriers (i.e. porous spherical carriers, fiber bundles) can neither provide anaerobic condition for phosphorus removal, nor effectively ameliorate membrane fouling. Additionally, sponge cube is subjected to biodegradation over time, shortening its life time. Although current studies have been increasingly developing new media, TN and TP removals are not satisfactory by biochar, while QQ bacterium based carriers are mainly employed for membrane fouling mitigation. When employing bioaugmented carriers, the accumulation of certain pollutants and some shock-loading conditions (i.e. extremely high saline level) are not favorable for activity of the immobilized bacteria and their capacity of biodegradation of specific pollutants. Moreover, some bioaugmented carriers (i.e. PVA/SA-gel beads) may release immobilized bacteria after long operational time. Based on above analyses, future research priorities are given as follows:

1) Development of more new media is highly needed. It would be better if the newly developed media can simultaneously minimize the saturation of adsorption capacity, create diverse microenvironment (aerobic, anoxic and anaerobic conditions) in single carrier, favor enrichment of diverse biocoenosis, improve sludge characteristics, maintain integrity of media during long-term operation, and resist shock loadings.
2) More research needs to be carried out regarding the application of newly developed media in treatment of emerging pollutants as well as enhancement in biogas generation and energy recovery.
3) Fundamental information on microbial community structure in attached biofilm and mixed liquor when treating different types of emerging pollutants is suggested to be updated. More special functional microorganisms for removals of emerging pollutants may be found according to the information, providing references for the development of bioaugmented carriers.

4) Bioaugmented carriers are required to be developed to remove wider spectrum of emerging pollutants.
5) Scale-up of lab scale AGMBRs to pilot or full scale AGMBRs is greatly needed to increase their application at industrial scale.
6) It needs to conduct techno-economic analysis and life cycle assessment for pilot or full scale AGMBRs with new media in circular bioeconomy framework.

5. Conclusions

This review focused on the performance of AGMBRs with conventional and new media for eliminating conventional and emerging pollutants from wastewater. Conventional media, including activated carbon and polymeric carriers, can remove some pollutants (i.e., COD, nitrogen, phosphorus, emerging pollutants) due to the formation of aerobic, anoxic and/or aerobic conditions, retention of more attached biomass and enrichment of diverse microbes. Nevertheless, they still face some problems: porous spherical carriers and fiber bundles are incapable of effective fouling control, AC is not able to keep sorption capacity during long-term operation; sponge cubes cannot maintain integrity over time. The applications of new media (i.e., biochar, walnut shell, GO-PVA gel beads, PVA/SA-gel beads, etc.) have demonstrated efficient removals of COD, nitrogen, some selected emerging pollutants (CIP, SMX, SDZ, SMZ, PHN, AOX) from specific wastewaters (e.g. nitrogen-rich wastewater, high-saline pharmaceutical wastewater, hospital wastewater, PHN-contaminated wastewater, etc.). This could be accomplished via creating anoxic/anaerobic conditions inside media, forming thick biofilm and enriching special functional microorganisms. However, activity and biodegradation ability of immobilized bacteria on the media could be reduced by the accumulation of pollutants and shock loadings. Future studies should focus on the development of more new media to overcome challenges confronted by existing media and extension of range of pollutants treated by new media.

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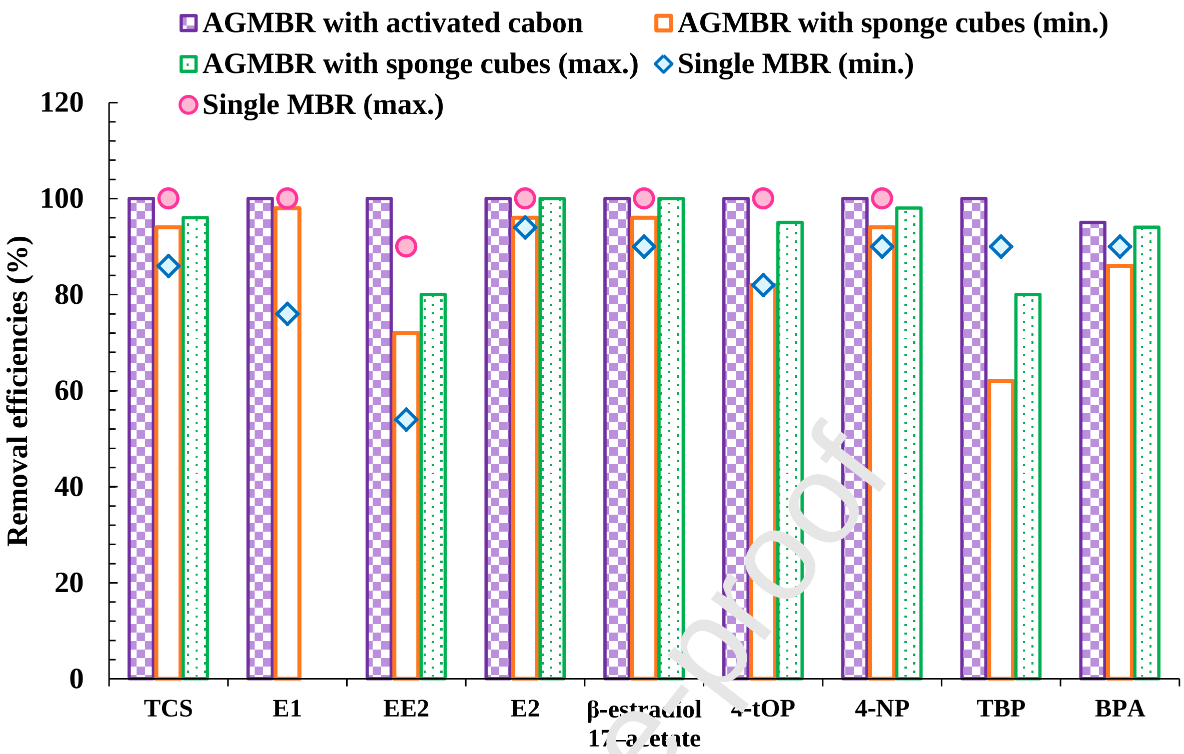
Figure captions

Fig. 1. Removals of hydrophobic compounds (HOC) from micropollutants in aerobic AGMBR with different media and single aerobic MBR (based on information from Nguyen et al. (2012) and Luo et al. (2015); Note: min., minimum removal efficiencies; max., maximum removal efficiencies)

Fig. 2. Removals of hydrophilic or moderately hydrophobic compounds (HIC1 and HIC2) from micropollutants in aerobic AGMBR different media and single aerobic MBR (based on information from Alvarino et al. (2017); Asif et al. (2020); Luo et al. (2015); Nguyen et al. (2012); Serrano et al. (2011); Note: Two exceptions in HIC1, GEM and IBP; Two exceptions in HIC2, MTZ and Tmp; min., minimum removal efficiencies; max., maximum removal efficiencies)

Fig. 3. Removals of selected pharmaceuticals in anaerobic AGMBR with PAC and single AnMBR (Based on the information from Xiao et al. (2017))



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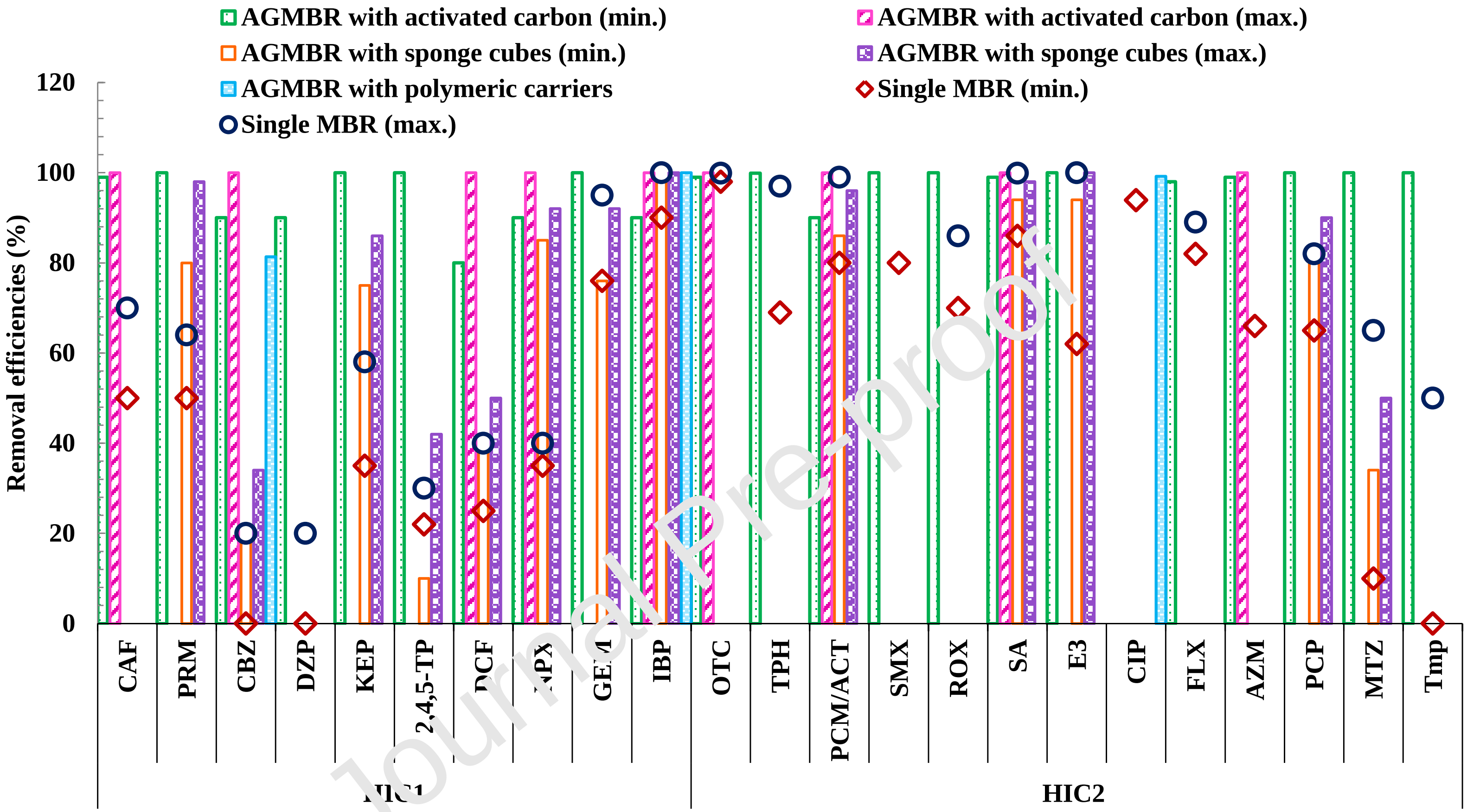

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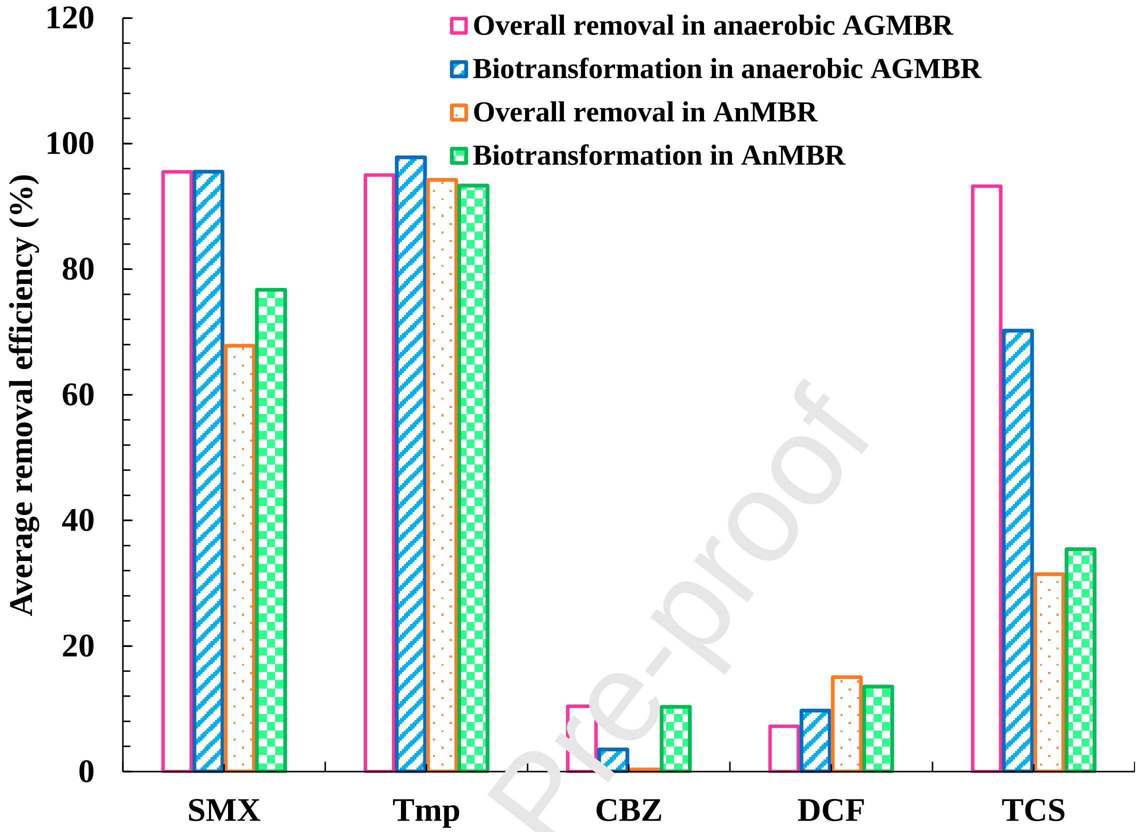

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Table titles

Table 1. Properties of commonly used polymeric carriers

Table 2. The performance of carriers with different shapes on conventional pollutants removal and membrane fouling control

Table 3. The selected strains/mixed cultures employed for the treatment of targeted pollutants or wastewaters and their contributions to the performance during treatment

Table 4. The performance of AGMBRs assisted by bioaugmented carriers to treat different types of wastewaters

Table 1. Properties of commonly used polymeric carriers*

Materials[a]	Density (g/cm^3)	Specific surface area (m^2/m^3)	Porosity	Shapes of carriers	Special properties	Applications[b]
PP	0.5-0.95	400-600	90-97%	• Polyhedron empty ball, cylinder or cuboid with biofilm growth at inner surface of carriers but less biofilm at the outer surface due to collision among carriers	• Coarse texture, giving rise to high surface area and microhabitat heterogeneity	➢ MBR, MBBR-MBR, or MBMBR as suspended carriers
PE	0.85-0.98	500-1530	-	• Cylinder with an internal cross and small longitudinal fins outside the surface	• Rough and porous surface	➢ Aerobic MBR or integrated MBBR-MBR as suspended carriers
		119-210	-	• Flat rigid square meshes		➢ MBR or Bf-MBR as fixed biofilm support media
PVA	1.03	~ 1000	~ 90%	• Ball (PVA gel beads) with a porous structure with continuous passages (diameter of 10-20 µm tunneling) throughout each bead	• Non-toxic and cost-effective material, • Adequate elasticity, • Hydrophilic property, • Resistance to biodegradation, • High mechanical strength to overcome high shear force	➢ Aerobic or anaerobic MBR as suspended carriers
PVC	1.0-1.1	500-600	93-96%	• Ball		➢ Aerobic or anaerobic MBR as suspended carriers
PEG	1.0-1.15	N.G.	-	• Cylinder	• Granular materials originally developed as	➢ Aerobic or anaerobic MBR

					microorganism carriers	as suspended carriers
PU	0.028-0.045	550-3000	~ 98%	• Cube, allowing microorganisms adhering on the surface and at the inner layer of carriers	• Rough surface, • High mechanical strength, • High resistance to organic solvent, • Easy handling, • Cost effectiveness, • High porosity due to the complex pattern of continuous voids, • Strong interaction with most microbial cell surfaces due to its hydrophobic character	➢ MBR or MBBR-MBR as suspended carriers, ➢ Tubular packed bed bioreactor with external membrane module as fixed biofilm carriers

[a]PU, polyurethane; PP, polypropylene; PE, polyethylene; PVA, Polyvinyl Alcohol; PVC, Polyvinyl chloride; PEG, polyethylene glycol

[b]Bf-MBR, Biofilm-membrane bioreactor; MBBR-MBR, moving bed biofilm reactor (MBBR) coupled with MBR; MBMBR, moving bed membrane bioreactor

*References: Alresheedi and Basu, 2014; Chaikasem et al., 2014; Costa et al., 2019; Dai et al., 2020; Deng et al., 2014; Fu et al., 2016; Guo et al., 2013; Hassard et al., 2016; Kurita et al., 2016; Li et al., 2019; Nguyen et al., 2017; Nguyen et al., 2019; Nie et al., 2016; Palmarin and young, 2019b; Premarathna and Visvanathan, 2019; Qaderi et al., 2018; Qiao et al., 2014; Rodríguez-Hernández et al., 2014; Singh et al., 2016; Song et al., 2018; Sun et al., 2015; Tang et al., 2016; Xu et al., 2018; Zheng et al., 2020

Table 2. The effects of carriers with different shapes on conventional pollutants removal and membrane fouling control

Shapes	Materials[a]	Merits[b]	Demerits[c]	References
Cylindrical carriers	PE, PP	✓ Enhanced growth of heterotrophic microorganisms and hydrolytic activities for degradation of organic matters; ✓ Enrichment of nitrifying bacteria for NH_4^+-N removal; ✓ Formation of aerobic and anoxic conditions at outer and inner layers of attached biofilm for enhanced TN removal; ✓ Higher abundance and activity of PAOs in small sludge flocs and increased anoxic P-uptake activity inside the attached biofilm, contributing to higher phosphorus removal	➢ Pore blocking and serious membrane fouling due to formation of small flocs	Costa et al., 2019; Xu et al., 2018
Porous spherical carriers	PP, PVC and PVA	✓ Enhanced NH_4^+-N removal by retaining slow growing nitrifiers; ✓ Formation of anoxic zones at the inner space of carriers for denitrification, enhancing TN removal; ✓ Adsorbing and consuming SMP and EPS by attached biofilm, mitigating membrane fouling	➢ Formation of small flocs (< 90 μm), increasing membrane fouling propensity; ➢ Unfavorable phosphorus removal due to limited anaerobic microenvironment on carriers	Guo et al., 2013; Li et al., 2019
Flat rigid square meshes	PE	✓ High concentration and activity of attached biomass, resulting in better organic matter removal; ✓ Enhanced NH_4^+-N and TN removal; ✓ Phosphorus removal through assimilating by microorganisms in both suspended and attached biomass	➢ The structure of carriers induces increased local shear force, thereby increasing collision among sludge flocs and release of fouling materials (i.e. SMP and BPC)	Rodríguez-Hernández et al., 2014; Sun et al., 2015
Fiber bundles	PE	✓ Growth of attached biomass on the surface of fibers and space between fibers, resulting in high biomass abundance; ✓ High NH_4^+-N and TN removal ability; ✓ Interception, adsorption and degradation of dissolved organic matters, EPS and particulates	➢ Limited anaerobic condition on carriers and low phosphorus removal	Song et al., 2018 and 2020

		by attached biofilm, thus moderating fouling		
Porous sponge cubes	PU, PP	✓ More porous space (including outer and inner spaces) for retention of attached biomass; ✓ Enhanced NH_4^+-N, NO_3^--N and TN by the formation of aerobic/anoxic condition around the surface of sponge for SND process; ✓ Improved phosphorus removal owing to less NO_3^--N in anoxic zone and formation of anaerobic condition inside the sponge for enhanced PO_4-P release; ✓ Improved sludge properties (i.e. enlarged floc size, reduced SMP and EPS levels) to effectively ameliorate membrane fouling; ✓ High potential for eliminating emerging pollutants by retaining slow-growing microorganisms in attached biomass	➢ Being susceptible to biodegradation owing to utilization of sponge as carbon and/or nitrogen source by microorganisms or co-metabolic biodegradation with other nutrients and substrates, which shortens lifespan of sponge; ➢ Reduced membrane surface scouring after a long-term operation due to biodegradation of sponge	Deng et al., 2014; Fu et al., 2016; Ham et al., 2018 Nguyen et al., 2017 and 2019; Urgun-Demirtas et al., 2007

[a] PU, polyurethane; PP, polypropylene; PE, polyethylene; PVA, Polyvinyl Alcohol; PVC, Polyvinyl chloride; PEG, polyethylene glycol

[b] GAOs, glycogen-accumulating organisms; PAOs, phosphate-accumulating organisms; SND, simultaneous nitrification and denitrification; SMP, soluble microbial products; EPS, extracellular polymeric substances; TN, total nitrogen

[c] BPC, biopolymer clusters

Table 3. The selected strains/mixed cultures employed for the treatment of targeted pollutants or wastewaters and their contributions to the performance during treatment

Targeted pollutants /wastewaters treated[a]	**Selected strains /mixed cultures**	**Bioaugmented carriers**[b]	**Performance enhanced by the immobilized cells on carriers**[c]	**References**
Marine weathered oil or crude oil	• Concentrated bacterial consortia containing hydrocarbon degrading strains (e.g. *Actinomycetales*, *Bacillus*, *Delftia* , *Planomicrobium alkanoclasticum* and *Pseudomonas*) • Crude oil-degrading bacterial consortium (e.g. *Bacillus subtilis* ZF3-1)	○ Shell grit, ○ *Eichhornia crassipes* dried straw	✓ Gradual increase in microbial attachment and cell viability during the operation ✓ Improving hydrocarbon removal by the immobilized oil-degrading bacteria ✓ High removal of > C_{32} hydrocabons after long time of operation (61-89% for 84 days)	Simons et al., 2013; Tao et al., 2019
Hypersaline organic wastewater	• Halotolerant bacteria (i.e. *Pseudomonas*)	○ GO–PVA gel beads	✓ High microbial diversity in gel beads, including the halotolerant bacteria and other anaerobic bacteria favorable for metabolizing carbon source (i.e. glucose) ✓ High tolerance to high saline condition	Zhou et al., 2015
Synthetic oilfeld produced water with high salinity	• Halophilic microorganisms (i.e. *Halomonas neptunia* Eplumel (T), *Yarrowia lipolytica*, etc.)	○ Walnut shell	✓ Easy digestion of lighter compounds of petroleum hydrocarbons (C10-C13) by the immobilized microorganisms ✓ Biodegradation of heavier compounds of petroleum hydrocabons (C14-C34, pristine, and phytane) at slow degradation rate, which could be realized at higher retention time ✓ No remarkable accumulation of undigested compounds in the sludge	Hasanzadeh et al., 2020
Wastewater	• Nonylphenol (NP)	○ Bamboo charcoal	✓ Biodegradation of NP adsorbed onto	Lou et al.,

polluted by NP (an environmental pollutant with estrogenic activity)	degrading bacteria (i.e. *Pseudomonas*, *Achromobacter*, *Ochrobactrum*, *Stenotrophomonas*)		immobilized cells ✓ Adsorption regeneration of carriers during long-term operation due to the release of some adsorption sites by biodegradation	2019
Azo dyes	• A newly isolated yeast strain LH-F1 (*Magnusiomyces ingens* which has capability of aerobically decolorizing various azo dyes)	○ Calcium alginate	✓ High microbial activity, stability and adaptability to a broad spectrum of temperature (25-45 ºC) and pH (3-9) as well as good reusability of immobilized strain ✓ Higher initial dye concentration (< 800 mg/L) slightly inhibited the growth rate of the immobilized strain ✓ Being capable of tolerating high concentration of dyes (i.e. acid Red B up to 1200 mg/L) and effectively removing the dyes (> 60%) with less requirements of external carbon and nitrogen sources, leading to generation of decolorization intermediates (i.e. aromatic amines, phenolics) which were highly biodegradable and low toxic compounds	Tan et al., 2014
Nitrogen (NH_4^+-N, NO_3^--N)-rich wastewater (NH_4^+-N ~ 100 mg/L, NO_3^--N ~ 100 mg/L)	• A heterotrophic nitrification–aerobic denitrification bacteria (*Pseudomonas fluorescens* Z03)	○ GO/PVA–SA gel beads	✓ Transferring NH_4^+-N to N_2 gas through oxidizing NH_4^+-N to NO_3^--N and N_2O, which were subsequently reducing to N_2 gas by denitrification, resulting in high removals of NH_4^+-N and NO_3^--N (> 96%) and less accumulation of NO_2^--N (< 0.1 mg/L)	Tang et al., 2020
Coking wastewater containing refractory organic compounds	• Newly isolated *Comamonas* sp. ZF-3	○ Polystyrene solid particles as microorganism-coated filters	✓ Gradual dominance of functional genera (i.e. *Comamonas* (strain ZF-3), *Thiobacillus*, *Pseudomonas* and *Thauera* contributing to removal of phenolic compounds, heterocyclic compounds and polycyclic aromatic hydrocarbons (COD removal of 93%) ✓ Being capable of living in a wild range of temperature (20-35 ºC) and pH (6-10)	Yuan et al., 2020
Sanitary wastewater	• Culturable heterotrophic bacteria	○ SBP, including $\text{NatiCap}_{\text{Petroleum}}$ capsule	✓ Encapsulated culture which was physically separated from suspended sludge in the	Azaizeh 2015;

enriched with a local industrial wastewater, periodic high concentrations of proteins, fats and oil from slaughter-houses as well as olive mill wastewater		for biodegration of petroleum hydrocarbons and other components (i.e. phenols, poly-phenols, and organic solvents) and $\text{NatiCap}_{\text{Municipal}}$ capsule for the encased microbial mixture to adapt to domestic wastewater containing high levels of grease fats and oils)	bioreactor, as well as protected from dilution and being grazed by protozoa ✓ Keeping optimal growth and encasing special functional microorganisms, which could effectively resist the shock loading and enhance the removal of emerging pollutants (i.e. autochthonous bacterial culture for degradation of phenolic compounds (tannic, gallic and caffeic acid) ✓ Offering an additional metabolic process (extracellular biodegradation process) with extracellular enzyme (i.e. lipases), leading to higher biological process stability and effectiveness under stress events (i.e. toxic substances, high OLR) ✓ The encapsulated cultures promoted long-term biodegradation activity	Menashe and Kurzbaum, 2016

[a] NP, nonylphenol

[b] GO/PVA–SA, graphene oxide (GO)-modified polyvinyl-alcohol (PVA) and sodium alginate (SA); SBP, Small bioreactor platform capsule, which is prepared by encapsulating exogenous bacterial culture in a confined environment employing microfiltration membrane as a protective barrier

[c] OLR, organic loading rate

Table 4. The performance of AGMBRs assisted by bioaugmented carriers to treat different types of wastewaters

Wastewaters treated	**Selected strains /mixed cultures**[a]	**Bioaugmented carriers**[b]	**Merits**[c]	**Demerits**[d]	**References**
Medium-strength domestic wastewater (average SCOD ~ 550 mg/L, OLR ~ 0.5 kg COD/m^3·d)	Concentrated anaerobic sludge*	PVA gel beads	✓ Declined membrane fouling by reducing bound EPS and SMP (proteins and polysaccharides) and enlarging floc size	• Slight effects on organic matter (SCOD) removal • Lower methane production and methane yield than theoretical values due to the low OLR operation • Limited NH_4^+-N removal due to lack of aerobic condition for nitrification process	Juntawang et al., 2017a
	Concentrated aerobic activated sludge*		✓ Reduced membrane fouling by decline in EPS and SMP levels	• Slight effects on SCOD and NH_4^+-N removal • Smaller floc size due to the collision between sludge flocs and the gel matrix by shear force from aeration	Juntawang et al., 2017b
Nitrogen-rich wastewater (NH_4^+-N 50-160 mg/L, NO_2^--N 50-160 mg/L)	Anammox bacteria (*Planctomycetes*)	MPCs	Enhanced TN removal rate via anammox process by the immobilized bacteria inside the carriers and nitrification at outer layer of carriers ✓ Reducing deposition of anammox bacteria on membrane surface, which limited their generation of EPS and SMP, controlling membrane fouling	• Only a slight enhancement in TN removal rate with carriers	Zhang et al., 2016
High-saline pharmaceutical wastewater containing high toxicity and	Mixture of conventional activated biomass* and marine sediment	PVC carriers	✓ Enhanced organic matters removal as marine species in biomasses could degrade emerging pollutants in wastewater and the entrapped	• Limited NH_4^+-N removal by the adverse effects of high salinity and emerging pollutants on activity and growth of nitrifying	Ng et al., 2016

refractory organic compounds	biomass* containing halotolerant/halophilic microorganisms		biomass had high affinity for organic matters for degradation ✓ Less membrane fouling by minimizing the accumulation of organic matters on membrane surface, further limiting microorganism growth on membrane surface for cake layer thickening	microorganisms as well as nitrification process (inhibiting effects)	
Hospital wastewater containing high concentration of selected recalcitrant antibiotics (CIP 546 ± 48 µg/L)	*Pseudomonas putida* cells	PVA/SA-gel beads	✓ High organic compound removal (> 90%) ✓ High CIP removal (90%) through releasing degradative enzymes ✓ Enhanced TN removal by the entrapped cells as heterotrophic nitrifier and aerobic denitrifier ✓ Stable growth of *P. putida* in the gel beads	• Release of some immobilized bacteria over time from the gel-beads due to dissolution of SA into aqueous solution • Accumulation of NO_2^--N due to the inhibition effect of recalcitrant compounds on NOB and nitratation process	Hamjinda et al., 2017
Phenanthrene-contaminated wastewater with high pH (8) and salinity (35 g/L)	*Pseudomonas* sp. strain LZ-Q	Ceramics	✓ Growth of the strain on the surface and in the ostioles of carriers ✓ Effective COD removal (94%) by the immobilized strain	• Higher energy requirement caused by increased aeration for suspending ceramic carriers due to their higher density than density of water	Jiang et al., 2015

[*] Suspended sludge was collected and centrifuged at 7000 rpm for 15 min to obtain concentrated sludge; Conventional activated biomass, which is inoculated from activated sludge of a local wastewater treatment plant; conventional activated biomass, which is collected from a local wastewater treatment plant; marine sediment biomass, which is collected from costal shore

[a] CIP, ciprofloxacin; OLR, organic loading rate; SCOD, soluble chemical oxygen demand

[b] MPCs, macroporous carriers; PVA, polyvinyl alcohol; PVC, polyvinyl chloride; SA, sodium alginate

[c] SMP,soluble microbial products; EPS, extracellular polymeric substances; AOB, ammonium oxidizing bacteria

[d] NOB, nitrite oxidizing bacteria; TEA, trimethylamine

Highlights

- Different types of media used in MBRs applications are reviewed.
- Mechanisms of pollutants removal by attached biomass on media are discussed.
- Addition of carriers could enhance performance of both aerobic and anaerobic MBRs.
- Carriers can reduce membrane fouling as well as enhance biogas generation.
- Future research should focus on development of new functional carriers.