Review

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The Effect of Activated Carbon Addition on Membrane Bioreactor Processes for Wastewater Treatment and Reclamation - A Critical Review

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Abstract

This review concentrates on the effect of activated carbon (AC) addition to membrane bioreactors (MBRs) treating wastewaters. Use of AC-assisted MBRs combines adsorption, biodegradation and membrane filtration. This can lead to advanced removal of recalcitrant pollutants and mitigation of membrane fouling. The relative contribution of adsorption and biodegradation to overall removal achieved by an AC-assisted MBR process can vary, and "biological AC" may not fully develop due to competition of target pollutants with bulk organics in wastewater. Thus periodic replenishment of spent AC is necessary. Sludge retention time (SRT) governs the frequency of spent AC withdrawal and addition of fresh AC, and is an important parameter that significantly influences the performance of AC-assisted MBRs. Of utmost importance is AC dosage because AC overdose may aggravate membrane fouling, increase sludge viscosity, impair mass transfer and reduce sludge dewaterability.

Keywords: Wastewater Treatment; Membrane Bioreactors; Membrane Fouling; Powdered Activated Carbon, Granular Activated Carbon

1. Introduction

Membrane bioreactor (MBR) technology integrates biodegradation by activated sludge with direct solid-liquid separation by membrane filtration. Nowadays, MBRs are considered an attractive alternative to conventional activated sludge process (CASP) for the treatment and reuse/recycle of industrial and municipal wastewaters (Judd S., 2011; Jamal Khan *et al.,* 2012; Hai *et al.,* 2014). The application of MBR systems for wastewater treatment is favored over conventional treatment methods due to considerable advantages including excellent and stable effluent quality, less excess sludge production, operation at high volumetric loadings, and smaller footprint (Li *et al.,* 2005; Ng *et al.,* 2006). However, their widespread application is still restricted by a phenomenon called membrane fouling (Chang *et al.,* 2002; Li *et al.,* 2005; Ying and Ping, 2006; Feng *et al.,* 2006). Uncontrolled membrane fouling leads to rapid reduction in membrane permeate flux (MPF) and/or increase in transmembrane pressure (TMP), resulting in high energy consumption and operating cost (Liu *et al.,* 2007; Tian *et al.,* 2010). A number of techniques have been explored for fouling control: these techniques either target at adopting suitable aeration strategies (*e.g.,* high-shear slug flow aeration in submerged configuration) or optimization of other operating conditions such as sub-critical flux operation, periodic air/permeate back-flushing and/or intermittent suction allowing a relaxation period for back diffusion of loosely attached foulants from membrane surface. A notable membrane fouling mitigation strategy is the addition of "membrane fouling reducers" (*e.g.,* flocculants or adsorbents) to MBRs (Chang *et al.,* 2002; Li *et al.,* 2005; Ng *et al.,* 2006; Tian *et al.,* 2010; Skouteris *et al.,* 2012; Yang *et al.,* 2012a).

The use of adsorbents such as activated carbon (AC) in conjunction with biological wastewater treatment processes such as CASPs or MBRs can be also beneficial in terms of stable treatment of recalcitrant wastewater. According to the available literature, potential advantages of dosing ACs such as powdered activated carbon (PAC) to CASPs include: (i) Protection of autotrophic and heterotrophic microorganisms from peak loads of inhibiting compounds, (ii) Biodegradation of refractory organic compounds, (iii) Increase in AC adsorption capacity due to the presence of a biofilm, (iv) Increase in sludge settleability and dewaterability, and finally, (v) Bioregeneration of AC. Because the AC added into CASPs can be washed out along with the treated effluent, frequent replenishment of AC becomes necessary. This significant maintenance cost restricts their widespread use (Munz *et al.,* 2007; Meng *et al.,* 2009; Ng *et al.,* 2012). Nevertheless, to date, ACs, in particular PAC, have been used in conjunction with CASPs to treat recalcitrant wastewater streams including industrial effluent (with inhibitory materials such as phenol, aniline or dye), landfill leachate, and high salinity oil-field brine (Ng *et al.,* 2006).

Unlike CASPs, owing to the complete retention of sludge by the membrane, in MBRs, a decoupling of hydraulic retention time (HRT) and sludge retention time (SRT) is possible. This allows operation of MBRs at a longer SRT. The reduced frequency of sludge removal reduces loss of PAC, simultaneously reducing the maintenance cost. Thus MBRs appear more suitable than CASPs to couple with AC adsorption. Furthermore, AC dosing to MBRs can potentially reduce the operating cost for membrane cleaning and/or membrane replacement by about 25% (Yang *et al.,* 2010). In this way the operating cost for PAC dosing can be potentially offset by the reduction in the cost for membrane maintenance, thus making the addition of ACs to MBRs highly attractive.

As noted above, the use of adsorbents in combination with MBR technology integrates adsorption and biodegradation of organic matter with membrane filtration (Figure 1). It has been proven to be an alternative approach to modify the characteristics of the mixed-liquor in order to remove recalcitrant compounds from wastewater efficiently, enhance MPF and control membrane fouling (Li *et al.,* 2005; Tsai *et al.,* 2005; Feng *et al.,* 2006; Iversen *et al.,* 2009a). For example, Li *et al.,* (2005) mentioned that the near critical-flux for an AC-assisted MBR was 32% higher than that of a conventional MBR. To date, the beneficial aspects of AC dosing to MBRs such as membrane fouling mitigation and efficient treatment of resistant wastewater have been separately and only briefly covered in relevant available reviews which focused either on membrane fouling (Le-Clech *et al.,* 2006; Drews, 2010) or treatment of recalcitrant wastewater (Hai *et al.,* 2014). However, a comprehensive understanding of the phenomena involved, particularly the interrelated impacts of AC on membrane performance and biodegradation, are yet to be critically analyzed. Thus this paper aims to provide an in-depth discussion on AC-assisted

MBR systems. AC-assisted MBRs have also been tested in relation to drinking water treatment (Tian *et al.,* 2009; Williams and Pirbazari, 2007); however, this work will focus mainly on the effect of ACs on MBRs treating different kinds of wastewaters. A notable originality of this review paper is that it covers a critical assessment of integration of AC adsorption with both aerobic and anaerobic MBR (AnMBR) technology.

[Figure 1]

2. Coupling membrane technology with adsorption and biodegradation

2.1 Pollutant removal by activated carbon adsorption

Wastewater-borne pollutant removal takes place through their diffusion onto the surface and/or into the pores of the AC (Tsai *et al.,* 2005; Vyrides *et al.,* 2010). However, some organic pollutants show greater adsorption than others. For example, organics such as toluene and chlorinated organics that have a low solubility in waters can be adsorbed by ACs more easily than the organics that are polar (Vyrides *et al.,* 2010).

ACs have been widely explored for the removal of a large number of pollutants including persistent xenobiotics and trace organic contaminants (TrOCs) such as pharmaceutically active compounds and endocrine disrupting compounds (EDCs), residual organic matter (ROM) and other refractory organics (Snyder *et al.,* 2007; Nguyen *et al.,* 2012; Whang *et al.,* 2014) from different kinds of wastewaters (Ng *et al.,* 2006; Munz *et al.,* 2007; Liu *et al.,* 2007, Remy *et al.,* 2010; Lin *et al.,* 2011). In general, they are successful in removing all compounds that can cause undesirable colour, odor or taste in water - details can be found in Table 1. However, the contaminant removal efficiency of ACs is subject to, among other factors, their particle size. Vyrides *et al.,* (2010) reported that small AC particles (≤ 0.25 mm) adsorb better (98% COD removal) than larger particles (≤ 0.75 mm) (50% COD removal) due to the fact that smaller particles have higher diffusion transfer and larger surface area. Also, Ng *et al.,* (2013) showed that fine PAC particles can control membrane fouling better than coarser ones provided that the applied MPF does not induce severe PAC deposition on the membrane.

[Table 1]

The efficiency of ACs in the removal of pollutants is also subject to the size of the molecules of the pollutants, with the addition of PAC achieving greater removal of high molecular weight compounds (Aquino *et al.,* 2006). Large-molecular weight pollutants adsorbed in large AC pores reduce the effective pore diameter, so the rate of adsorption of smaller molecules, that have no option but to pass through these pores to reach smaller pores, is reduced. This unavoidably leads to a decrease in adsorption over time, particularly when there is a diversity of high-molecular weight compounds in the wastewater and a large range in the pore size distribution. Vyrides *et al.,* (2010) studied adsorption of organic effluent from an AnMBR treating saline sewage. They observed that 53% of the pollutants had molecular weights lower than 1 kDa, and the adsorption of high-molecular weight solutes on the PAC during the first 15 minutes of adsorption was able to block a significant fraction of pores, making the adsorption of lower-molecular weight compounds more difficult as it takes place slowly. They concluded that both fractions of organics in wastewaters, namely fulvic acid-like low-molecular weight pollutants (molecular weight: 0.1 - 10 kDa) and humic-carbohydrate-like higher-molecular weight pollutants (molecular weight: >50 kDa) were inadequately adsorbed by ACs, mainly granular activated carbon (GAC). However, the case of lower-molecular weight organics seem to be rather worse.

Even though adsorption on ACs may lead to high removal of organics initially, it is inevitable that their adsorption capacity will be exhausted over time (Magic-Knezev and van der Kooij, 2004; Nguyen *et al.,* 2012). Spent AC needs to be either safely disposed or can be regenerated. To date, a wide variety of methods have been reported for AC regeneration. Common regeneration techniques are based either on thermal methods (exposure to steam, carbon dioxide or inert atmosphere) or chemical methods (exposure to pH-swing or extraction with solvents). When combined with a biological process, "bioregeneration" may also occur. However, all these techniques often fall short of a "practical" solution either from a technical or an economical point of view (Chen et al., 2013; Li et al. 2015). Further coverage of regeneration of ACs is beyond the scope of this review.

2.2 Rationale of combining membrane technology, adsorption and biodegradation

Membrane processes such as microfiltration (MF) and ultrafiltration (UF) are widely used to separate particles, colloids and microorganisms both in water and wastewater treatment. However, neither MF nor UF is effective in removing soluble organic matter. Organic matter removal can be enhanced by a combined adsorption/membrane filtration process, which can be further improved by degradation by microorganisms (Lesage *et al.,* 2008; Tian *et al.,* 2008; Tian *et al.,* 2010).

As noted earlier, the addition of adsorbents such as ACs to CASPs has been found to lead to improved chemical oxygen demand (COD) reduction due to the coupling of biodegradation with adsorption. In recent years, AC-assisted MBR systems have been explored based on the experience with CASPs. Notably, AC addition within the tanks housing the membranes also mitigates membrane fouling (Ahmed and Lan, 2012). The modification of traditional MBRs with the addition of adsorbents has been found to have an effect both on the biodegradation process (combined adsorption/biodegradation for efficient removal of organic pollutants from wastewaters) and on membrane filtration. In general, AC-assisted MBR technology has been found to be highly attractive when advanced treatment of wastewaters is required, as, in this case, combination of different treatment methods is considered more efficient than individual technologies. AC increases the surface available for liquid-solid contact and makes adsorption easier, providing at the same time a better environment for microbial metabolism which leads to efficient removal even at low organic rates affecting the microbial community. However, the interaction between ACs and biomass needs to be studied further (Lin *et al.,* 2011).

2.3 Activated carbon-assisted membrane bioreactors

To date, PAC-assisted MBRs have been studied mainly in relation to membrane fouling mitigation. However, several recent studies demonstrate the potential of integrating AC adsorption, in particular that of PAC, with MBR technology for the removal of recalcitrant pollutants from wastewater (Snyder *et al.,* 2005; Li *et al.,* 2011a; Nguyen *et al.,* 2012; Nguyen *et al.,* 2014). Majority of the available studies have been conducted on aerobic MBRs treating municipal and synthetic wastewaters, although studies focusing on industrial wastewaters, with COD values in the range of 4000 - 5000 mg L^{-1}, and anaerobic MBRs are also available (Hu and Stuckey, 2007; Satyawali and Balakrishnan, 2009). In addition to controlling MPF deterioration due to membrane fouling and concentration polarization, AC addition has been found to improve the MBR treatment process as follows: 1. ACs adsorb biologically resistant compounds that may be toxic to the microbial community, 2. They provide an excellent surface for the attachment of microorganisms, 3. They have high affinity for molecular oxygen and consequently enhance the driving force to bring oxygen actively into the biofilms, and 4. They reduce the effects of fluctuating loading of hazardous chemical compounds (Lesage *et al.,* 2005; Li *et al.,* 2005; Tsai *et al.,* 2005; Lesage *et al.*, 2008; Satyawali and Balakrishnan, 2009; Xing *et al.,* 2011; Baêta *et al.,* 2014). It needs to be noted that the addition of ACs directly into an MBR tank may prevent the necessity of potentially more expensive polishing steps; however it is not yet fully known whether a sufficient degree of adsorption, together with biological regeneration of the PAC, can be accomplished (Remy *et al.,* 2009). The performance of activated carbon-assisted MBRs is critically analyzed in Section 3 onward.

2.4 Post-treatment of membrane bioreactor permeate with activated carbon

AC adsorption has been traditionally used as a post-treatment method for secondary treated wastewater. MBR permeate is free of suspended solids with very low total organic carbon (TOC) content; therefore post treatment of MBR permeate with ACs appears to be highly suitable. Particularly this can be an efficient process to target residual TrOC from MBR permeate as there would be reduced interference from the bulk organics (Vyrides *et al.,* 2010; Nguyen *et al.,* 2012).

In general, MBRs are known to be very effective for the removal of hydrophobic and readily-biodegradable pollutants, but may fail to remove hydrophilic compounds, particularly those which are also resistant to biodegradation (Nguyen *et al.,* 2013a; Nguyen *et al.,* 2013b) Coupling AC adsorption with MBR treatment can ensure removal of the hydrophilic compounds (and remaining hydrophobic compounds) from MBR permeate. For example, post-treatment of MBR permeate with PAC achieved removal of compounds such as m-aminophenylacetylene (C_8H_7N), cyclohexane 1,2,4 trimethyl (C_9H_{18}) and cholestan 3-one ($C_{27}H_{46}O$) that were resistant to MBR treatment (Vyrides *et al.,* 2010). Similarly, GAC post-treatment was found to remove hydrophilic and persistent TrOCs such as carbamazepine, diclofenac and fenoprop, ensuring high removal of a broad spectrum of investigated TrOCs by combined MBR-GAC treatment (Nguyen *et al.,* 2012, Nguyen *et al.,* 2013b).

A few studies have systematically demonstrated the superiority of AC adsorption as a polishing step when they are combined with biological treatment (aerobic or anaerobic). A comparison among several techniques, namely, aerobic biological oxidation, coagulation and flocculation with aluminium sulphate and adsorption with the aid of ACs for treatment of an effluent coming from anaerobic digestion of simulated coffee wastewater showed that only AC could significantly reduce the effluent COD (Vyrides *et al.,* 2010). Trzcinski *et al.,* (2011) tested several options and reported AC as the best performing post-treatment option for stabilized leachate from a submerged AnMBR. PAC and GAC resulted in an overall COD removal of 84% and 80%, respectively, although both had a similar adsorption efficiency of about 60% for the fractions of organics with molecular weights of lower than 1 kDa. Among the other tested treatment techniques, a UF membrane of 1 kDa resulted in a removal percentage of 75%, a coagulation-flocculation process with $FeCl_3$/polyelectrolyte in 45%, only $FeCl_3$ in 32%, and polymeric adsorbents such as XAD7HP in 46% and XAD4 in 32% (Trzcinski *et al.,* 2011). While post-treating the effluent from an AnMBR with various methods, Vyrides *et al.,* (2010) observed that the order of the treatment methods, starting from the most efficient one, was: aerobic biomass coupled with PAC > only PAC > aerobic biomass > anaerobic biomass coupled with PAC > anaerobic biomass. This observation indicates the added advantage of a combined adsorption - biodegradation process in comparison to adsorption only.

It is worth mentioning that occasionally it may be more preferable to treat wastewaters with an MBR and then apply AC post-treatment, because direct AC addition within the MBR tanks may reduce the adsorption of the target compounds due to their competition with bulk organic matter for adsorptive sites. Only a few studies have systematically compared the application of AC as a polishing step vs. direct dosing of AC to MBR. In a study by Nguyen *et al.,* (2013b), both direct PAC addition into an MBR (PAC-MBR) or GAC post-treatment (MBR-GAC) was observed to significantly complement MBR treatment to obtain high overall removal of less hydrophobic and biologically resistant TrOCs. In both systems, however, gradual breakthrough of resistant compounds occurred over an extended operation period. Based on a simple comparison from the long-term performance stability and AC usage points of view, PAC-MBR appeared to be a better option than MBR-GAC treatment (Nguyen *et al.,* 2013b). While treating an oily wastewater, William, (2009) also reported that PAC-MBR was better than MBR-GAC system in terms of effluent quality, less frequent membrane cleaning, tolerance to upsets and immediate acclimation.

3. Pollutant removal by powdered activated carbon dosing to aerobic membrane bioreactors

3.1 Membrane configuration and MBR format

In almost all investigations reported in the literature, AC-assisted MBRs are of submerged configuration (Figure 1), integrating adsorption and biodegradation of organic matter with membrane filtration in one unit (Li *et al.,* 2005; Lin *et al.,* 2011; Kim and Lee, 2003). However, a few examples of side stream AC-assisted MBR-related investigations can be found. For example, Tsai *et al.,* (2005) modelled a side stream PAC-assisted MBR fitted with a tubular membrane for treatment of water. Regarding the configuration of membranes, the majority of the available studies operated hollow fibre membranes; however membranes in flat sheet configuration have also been tested. More details can be found in Table 2 which shows combination of different membrane configurations with ACs.

[Table 2]

[Table 3]

3.2 Improved removal performance

Through selected references, Table 3 demonstrates better performance of PAC-assisted MBRs over conventional MBRs. For example, addition of PAC into a pilot-scale MBR treating dye wastewater improved its performance in terms of COD removal by 12% (Fang *et al.,* 2012). Some studies point out that COD removal/improvement due to PAC addition to MBR may at times be marginal, particularly for wastewater containing greater proportion of biodegradable compounds (Ying and Ping, 2006; Munz *et al.,* 2007; Liu *et al.,* 2007; Lesage *et al.,* 2008; Remy *et al.,* 2010). However, often it has been observed that the PAC-amended MBRs can lead to satisfactory effluent COD values even for wastewaters bearing very high initial COD and salinity and predominantly containing hardly biodegradable compounds (Vyrides and Stuckey, 2009; Tzcinski and Stuckey, 2010; Nguyen *et al.,* 2013a; Nguyen *et al.,* 2013b). Examples, where conventional biological treatment methods followed by post-treatment methods such as coagulation and electrochemical oxidation failed but PAC-assisted MBR efficiently treated the wastewater, also exist. For example, bactericide wastewater as reported by Han *et al.,* (2008): they noticed that an increase in PAC concentration from 0.1 g L^{-1} to 2 g L^{-1} led to a further increase in the COD removal efficiency, from 90% at 0.1 g L^{-1} of PAC concentration to 99% at 2 g L^{-1}. Although in this case, significantly higher dosage was required to achieve a COD removal exceeding 90%, its importance cannot be overlooked (Ying and Ping, 2006; Liu *et al.,* 2007 and Munz *et al.,* 2007; Hai *et al.,* 2008). Lin *et al.,* (2011) noted that the addition of PAC in MBRs may not have any significant effect on nitrification. They observed that both PAC-amended MBRs and conventional MBRs led to a NH_4^+-N removal efficiency of higher than 95%. Munz *et al.,* (2007) also reported that the addition of PAC may not affect nitrogen removal. However, nutrient (nitrogen and phosphorous) removal necessitates sequential exposure of sludge to aerobic and anoxic/anaerobic conditions, and conceptually such contrasting environment may prevail at different depths of biofilm formed on PAC. Further research on clarifying this aspect is necessary.

Recent studies have convincingly shown that PAC-assisted MBRs (either aerobic or anaerobic) can efficiently remove colour from wastewater (Table 3). With PAC addition, excellent stable decolouration of wastewater containing dyes can be achieved by an MBR (Abbeglen *et al.,* 2009; Fang *et al.,* 2012; Baêta *et al.,* 2014). For example, Hai *et al.,* (2008) reported excellent (>99%) stable removal of two tested dyes, as well as stable enzymatic activity following addition of PAC to a fungal MBR. Interestingly, comparison of the reactor-supernatant and the membrane-permeate qualities revealed the significant contribution of the membrane to the overall removal (biosorption/PAC-adsorption, cake layer filtration, and biodegradation) of the dyes. Also, the great potential of PAC-assisted MRBs in regard to treatment of highly recalcitrant landfill leachate must be highlighted. In general, high physicochemical adsorption of leachates on PAC has been shown (Vyrides *et al.,* 2010).

Finally, as already noted in Section 2.4, ACs have been found to efficiently remove TrOCs from water. TrOCs such as pharmaceutically active compounds, pesticides, industrial chemicals and steroid hormones are detected in concentrations of ng L^{-1} to µg L^{-1} in wastewaters. TrOC removal is an emerging challenge in the wastewater treatment sector given that treated water should now meet stricter environmental quality standards set by new regulations such as the Water Framework Directive (Choubert *et al.,* 2011; Nguyen *et al.,* 2013).

A significant variation in the removal of TrOCs by MBRs, ranging from near-complete removal for some compounds (*e.g.,* ibuprofen and bezafibrate) to almost no removal for several others (*e.g.,* carbamazepine and diclofenac), can be observed in the literature. However, adsorption onto sludge has been reported to be an important means of aqueous phase removal of TrOCs. The addition of ACs can increase this adsorption, thereby significantly increasing the retention of soluble TrOCs. This can potentially enhance their subsequent biodegradation (Li *et al.,* 2011a; Nguyen *et al.,* 2014). Indeed, PAC-amended biosolids have been reported to provide a better surface to adsorb more TrOCs and achieve better aqueous phase removal (Nguyen *et al.,* 2013a). Periodic withdrawal and replenishment of PAC is required for stable removal, and in this case, frequent and smaller dose PAC addition may be preferable. The extent of biodegradation (transformation) following adsorption onto PAC-sludge has been reported to be compound-specific (Nguyen *et al.,* 2013a). Pollutant removal is mainly dependent on their hydrophobicity and loading as well as on the PAC dosage. For example, Li *et al.,* (2011a) reported that when the concentration of PAC increased from 0.1 g L^{-1} to 1.0 g L^{-1}, significant removal of carbamazepine (from negligible to 92±15%) occurred. The high removal efficiency at the higher PAC dosage was attributed to the fact that carbamazepine is quite hydrophobic, which subsequently resulted in its higher adsorption affinity. Similarly, Yang *et al.,* (2012b) demonstrated stable removal of the hydrophobic steroid hormones 17β-estradiol and 17α-ethynylestradiol by a PAC-assisted MBR. However, negatively-charged compounds such as fenoprop and diclofenac were observed to show high resistance (Nguyen *et al.,* 2013a).

3.3 Factors influencing removal performance

In a PAC-assisted MBR system, not only does the use of PAC increase the removal of low-molecular weight organics by adsorption but also PAC acts as a supporting medium for attached bacterial growth: it influences the population of microorganisms and it affects the concentration of extracellular polymeric substances (EPS) (Li *et al.,* 2005). As PAC decreases the compressibility of sludge flocs and increases the porosity of the cake layer, enhanced MPFs are also achieved. Other benefits of PAC include decrease in sludge production and increase in the resistance to toxic substances (Lesage *et al.,* 2008; Satyawali and Balakrishnan, 2009). In general, PAC increases adsorption of organics in bulk liquor and improves the filterability of the latter.

When added intermittently to an MBR, PAC can form biological powdered activated carbon (BPAC) (Li *et al.,* 2005, Vyrides *et al.,* 2010). The stable microbial film that tends to form on the PAC surface, transforming it into BPAC-sludge, enhances the pollutant removal as the microorganisms on the biofilm can biodegrade the pollutants that had previously been adsorbed by PAC. The advantages of BPAC can be summarized as follows: (i) it increases the efficiency of substrate removal, (ii) it improves the activated sludge filterability, and finally, (iii) it reduces the adverse effects of toxic chemical species on biomass through adsorption and leads to better performance by withstanding shock loads (Ng *et al.,* 2006). To facilitate formation of a stable microbial film specifically on PAC particles, it has been recommended that the membrane filtration be started later (*e.g.,* 40 days later). In addition, fresh PAC must be added any time sludge wasting takes place in order to maintain its stable concentration in the MBR (Li *et al.,* 2005).

The selection of the correct type of PAC is critical and depends on the pollutant type. Iversen *et al.,* (2009b) tested two types of PAC -the NORIT SA SUPER and the PICA PICAHYDRO LP27- and in general both PACs managed to remove successfully humic and low molecular weight contaminants. At the highest PAC concentration tested, about 80% of all these contaminants were eliminated due to their adsorption on PACs. However, the removal efficiency of biopolymers, which have higher molecular weights, depended on the type of PAC. NORIT SA SUPER removed biopolymers with greater efficiency (maximum removal of 69%). The difference in the elimination efficiency of high molecular weight pollutants of the two tested PACs mentioned above was explained by the different structure of the pores or by their different chemical properties, such as hydrophobicity and electric charge. For instance, NORIT SA SUPER has more micropores which allow larger molecules like biopolymers to penetrate the PAC without blocking the micropores. Although both PACs achieved high removal of polysaccharides and proteins from the supernatant, higher dosages of PICA PICAHYDRO LP27 were required. For example, the highest protein elimination efficiencies of 75% and 81% were achieved by NORIT SA SUPER at 450 mg L^{-1} and PICA PICAHYDRO LP27 at 5000 mg L^{-1}. Nonetheless as polysaccharides and proteins are the main constituents of EPS and soluble microbial products (SMP), which have adverse impact on membrane fouling, it can be concluded that PAC does help mitigate membrane fouling conditions (Iversen *et al.,* 2009b), with proteins being retained more efficiently than polysaccharides (Chu *et al.,* 2013).

Apart from the competitive adsorption of large and smaller molecular weight compounds in general, with respect to PAC addition to an MBR, it has been found that pollutants with molecular weight cut-off below 1kDa are removed by direct adsorption and biodegradation, whereas those with molecular weight above 1kDa are rejected by the cake layer on the membrane and gradually degraded by microorganisms due to their extended contact (Seo *et al.,* 2004, Chu *et al.,* 2013).

The solids residence time (SRT) is an important parameter that significantly influences the performance of PAC-assisted MBRs. Ng *et al.,* (2013) observed more severe membrane fouling when operating an MBR at an SRT of 10 d compared to at 30 d. This may be attributed to the higher fouling propensity of the mixed liquor at an SRT of as low as 10 d (Ng and Hermanowicz, 2005). However, when PAC was added, fouling control appeared to be more efficient in case of the shorter SRT (10 d), possibly due to the fact that at shorter SRTs fresh PAC has to be added more often, so "active" PAC is always present in the mixed liquor in adequate amount. Ma *et al.,* (2014) operated two identical PAC-assisted MBRs, one at an SRT of 30 d and another at a longer SRT of 180 d. They reported that at the shorter SRT, the frequent replacement of PAC led to higher removal of low molecule weight (< 5 kDa) effluent dissolved organic matter (DOM), which also exhibited higher hydrophobicity. Prolonging the SRT, the effluent DOM became more hydrophilic which favored the removal of high molecular weight (> 5 kDa) DOM mainly due to biodegradation by well-acclimatized microorganisms (Ma *et al.,* 2014).

3.4 Shock-load buffering

The addition of PAC has been proven to be effective under various stress conditions. For instance, good removal efficiencies of organics was observed even for organic loading rates in the range of 1/20 - 1/3 of that suggested by Water Environment Federation (Lin *et al.,* 2011). Similarly, PAC addition may allow operation at higher organic loading rates than usual. This can be explained by the fact that PAC dosing to MBR decreases stress or toxic shocks, allowing better biodegradation of toxic compounds (Lesage *et al.,* 2004; Lesage *et al.,* 2008; Satyawali and Balakrishnan, 2009). The rough surface provided by the adsorbent for microbial growth can protect the bacteria from toxicity caused by the lipophilic nature of some pollutants and enhance the buffering capacity of the microbial system by adsorbing acid and alkaline compounds, as well as enriching dissolved oxygen (Satyawali and Balakrishnan, 2009). However, PAC dosages must be selected as such to avoid any detrimental impact on microbes. When PICA PICAHYDRO LP27 PAC of 5 g L^{-1} was added as an MPF enhancer, the pH of the mixed liquor decreased from 7.1 to 5.7, which adversely impacted oxygen uptake rate (-28%), nitrification (-90%) and denitrification rate (-43%) (Iversen *et al.*, 2009a). Specific oxygen uptake rate (SOUR) is monitored to probe any stress or toxic impact on the microbial community. SOUR rates decrease with increasing SRT, as expected, due to accumulation of inert matter during prolonged operation periods, and PAC may not contribute to increase SOUR rate in such a situation (Satyawali and Balakrishnan, 2009).

3.5 Relative contribution of adsorption and biodegradation

Available studies report varying extents of relative contribution of adsorption and biodegradation to overall removal achieved by an AC-assisted MBR process. Whang *et al.*, (2004) reported that during polishing the biologically-treated swine wastewater by an MBR with PAC, a total of 81% removal of ROM was achieved. Of the total removal of 81%, the largest removal occurred via adsorption (46.5%), while activated sludge and membrane separation accounted for 20.8% and 4.4% removal, respectively. Notably, PAC addition to the MBR further increased the removal efficiency of activated sludge by 9.3% (30.1% in total), a percentage that was attributed to enhanced microbial activity. In another study, Lin *et al.,* (2011) reported a total ROM removal of 62.7% from secondary effluents by a PAC-packed MBR. Out of this total removal efficiency, 46.6% of the total amount of organics was removed by PAC adsorption, 13.3% by biological degradation, and 2.8% by membrane separation. These examples demonstrate that for hardly degradable compounds, such as the ROM remaining following secondary treatment, adsorption may account for a major part of the removal. Nguyen *et al.*, (2014), while investigating removal of a set of 22 micropollutants by a PAC-assisted MBR, observed that "biologically activated carbon" wherein adsorption, biodegradation and PAC regeneration may occur simultaneously, was not fully established. They attributed this to two likely reasons: (i) under competition with other organic compounds in the synthetic wastewater, only a small fraction of the PAC added to the MBR can be effectively utilized for adsorption of the micropollutants, (ii) pore blockage by bulk organic matter including products of microbial degradation and dead microbial cells reduces the adsorption capacity of target compounds on PAC.

4. Pollutant removal by granular activated carbon dosing to aerobic membrane bioreactors

GAC has both good capacity to adsorb contaminants and excellent biomass retention capacity as a biological attachment medium due to its extensive surface area and shear force-sheltering capabilities (Xing *et al.,* 2011). However, given the higher specific gravity of GAC compared to PAC, it is more difficult to prevent its settling and keep it in suspension. Thus GAC has been mostly used in the form of fixed bed column as a polishing step for MBR effluent. However, when GAC is directly dosed into MBRs, in addition to efficient removal of recalcitrant pollutants, biofouling can be prevented (Cecen and Aktas, 2011). Thus GAC-assisted MBRs have indeed been tested in recent years, particularly in relation to treatment of recalcitrant wastewater such as phenolic wastewater and ethanol-based wastewater (Johir *et al.,* 2011).

Reported studies on phenolic wastewater removal indicate that addition of GAC to MBR may enhance biodegradation of phenolic compounds, possibly by reducing the toxic and inhibitory impact of phenol via adsorption. For example, Thuy and Visvanathan, (2006) tested the ability of a GAC-assisted MBR to remove phenolic compounds, namely, phenol and 2,4-dichlorophenol. Relatively low values of phenol adsorption on GAC and biomass, and high maximum substrate removal rates obtained from a biokinetic experiment, proved that the removals were mainly due to biodegradation. Li and Wang, (2008) observed improved removal of phenols by incorporating GAC into hollow fiber membranes on which *Pseudomonas putida* was immobilized. The GAC-bioreactor showed its superiority over the GAC free-bioreactor during start-up and elevated loading phases. The performance improvement was attributed to enhanced biodegradation following adsorption of phenol onto GAC-filled hollow fiber which increased phenol tolerance of bacteria. Hai *et al.,* (2012) reported that additional GAC layers on the membrane module within a whole-cell fungal MBR set up to treat dye wastewater was effective in minimizing enzyme washout and in improvement of decolouration (degradation of the dye) (Hai *et al.,* 2012). In another study, Hai *et al.,* (2011) demonstrated the importance of including a GAC-amended anaerobic zone within an aerobic MBR. Stable decolouration, along with significant TOC removal during a period of over 7 months under extremely high dye-loadings, demonstrated the superiority of the proposed hybrid process.

In addition to the laboratory or pilot scale studies discussed above, it is interesting to note that there is a patented Biological Membrane-Assisted Carbon Filtration (BioMAC) technology which uses a combination of GAC filtration and MF/UF for enhanced treatment of concentrated streams (van Hege *et al.,* 2002). It has been reported to be effective, for example, in the removal of TrOCs from wastewater. Noteworthy also is the EcoRight MBR System, provided by Siemens, which incorporates the use of GAC in the aeration tanks of MBRs. EcoRight MBR is designed to meet the needs of the Oil and Gas Industry as it is able to meet the increasingly strict effluent requirements, and offer economical reuse of water and wastewater through a combination of biological treatment and carbon adsorption, (www.energy.siemens.com, last accessed Nov. 2014).

5. Membrane fouling mitigation by powdered activated carbon or granular activated carbon dosing

5.1 Underlying concept

As noted earlier, membrane fouling, which can be either reversible (fouling that can be removed by physical cleanings) or irreversible (fouling that can be removed by chemical cleaning only) still restricts a wide-spread commercialization of MBRs.

Membrane fouling, which is attributed to the interaction between the membrane and the components in the mixed liquor, not only decreases the MPF (reduced productivity), but it also requires frequent cleaning procedures or even their replacement, increasing maintenance and operating costs (Chang *et al.,* 2002, Li *et al.,* 2005).

Nowadays, it is commonly accepted that air bubbling close to the membrane is one of the most efficient means of minimizing reversible membrane fouling in MBRs and ensuring their long-term sustainable operation. Bubbling induces local shear stress, which controls fouling of membranes and creates a favorable hydraulic distribution over the membrane. However, due to the presence of irreversible interactions between soluble compounds or bacteria and membrane material, membrane fouling may not be fully controlled only by aeration. Incorporation of supporting media/adsorbents may be relevant to scour a part of the foulants on the membrane surface and capture some of the fouling-causing substances prior to their contact with membrane material. Addition of adsorbents such as ACs into the biological treatment tank can potentially remove organic compounds like EPS and SMP that cause irreversible membrane fouling (Ying and Ping, 2006; Johir *et al.,* 2011).

Several studies have explored direct dosing of AC, particularly that of PAC, to MBR for mitigation of membrane fouling. It has been reported that addition of ACs improves the filtration characteristics of the mixed liquor as well as increasing the beneficial effect of slug flow on membrane fouling. These together lead to a decrease in the membrane fouling rates and energy and chemicals consumption (Yang *et al.,* 2011; Ng *et al.,* 2013). ACs may also have a scouring effect for removing the cake layer from the membrane surface. MPF improvement can then be achieved as ACs manage to reduce the biomass cake resistance and change the overall particle distribution to a larger size range (Munz *et al.,* 2007; Yang *et al.,* 2006).

As mentioned before, PAC not only incorporates bioflocs (forming BPAC), it also adsorbs biopolymers from the sludge suspension. Its addition to MBRs provides a solid support for biomass growth, hence reducing floc breakage. Moreover, the BPAC flocs in MBRs are very strong and dense, preventing particle accumulation on the membranes (Meng *et al.,* 2009). BPAC flocs are more resistant to shear force, so higher MPFs can be sustained. For example, Remy *et al.,* (2009) reported a 19% increase in the MPF. This rugged floc structure is very important, particularly for flocs located close to the membrane surface which are exposed to high shear owing to strong air bubbling applied to control membrane fouling. It can therefore be expected that the strong sludge flocs incorporating PAC will release less amount of foulants and will cause less gel layer formation than the weaker conventional flocs (Remy *et al.,* 2009).

The addition of PAC in MBRs also affects the capillary suction time - a parameter that indicates any change in the floc structure, and which relates to sludge dewaterability and filterability. Experiments conducted with different types of PAC showed that the capillary suction time is also subject to the type of the PAC used. Testing two different PACs, Iversen *et al.,* (2009b) concluded that the capillary suction time for NORIT SA SUPER at 1000 mg L^{-1} was improved by 40% (maximum percentage), whereas in terms of PICA PICAHYDRO LP27, the maximum improvement was lower, only 21% at 5000 mg L^{-1}. The improvement of capillary suction times when PAC is added to an MBR tank can be attributed both to the removal of large amounts of dissolved organic carbon (DOC) and to the change of the flocs due to the incorporation of PAC particles into them (Iversen *et al.,* 2009b).

5.2. Impact on membrane filtration resistance

A number of studies have shown that the addition of PAC in MBRs can enhance membrane performance in terms of filtration resistances and sustainable operation time (Johir *et al.,* 2011; Li and Gao, 2011; Lin *et al.,* 2011). During the membrane fouling observation of a conventional MBR and a PAC-amended MBR, TMP profiles for both MBRs showed an initial period of slow TMP rise followed by a rapid TMP rise. However, when comparing the two TMP profiles, the initial "slow TMP rise period" for the PAC-assisted MBR was two times longer. This practically means that the addition of PAC delayed significantly the requirement of membrane cleaning (Lin *et al.,* 2011). Fang *et al.,* (2011) also studied the way TMP-rise can be delayed by the addition of PAC in an MBR. They observed that TMP increased steadily and peaked at 0.016 MPa on day 6 of MBR operation without PAC, while it increased and peaked at 0.015 MPa on day 10 when PAC was added to the MBR.

The fact that the addition of ACs such as PAC or GAC does alleviate membrane fouling can also be demonstrated by the analysis of filtration resistances of membranes. Lin *et al.,* (2011) demonstrated that the total membrane resistance of an MBR amended with PAC was smaller than that of a conventional MBR operating without addition of ACs. In addition, three out of the four individual resistances that constitute the total membrane resistance, namely the intrinsic membrane resistance, the concentration polarization layer resistance and the internal membrane resistance were also smaller for the PAC-assisted MBR. The only exception was the fourth partial resistance, namely the external membrane resistance, which was found to be larger for the PAC-assisted MBR which can be attributed to the increase in the concentration of solids in the bulk liquid due to the addition of PAC particles. However, the cake resistance, that is the sum of concentration polarization layer resistance and the external membrane resistance, is once again smaller for PAC-assisted MBR (Lin *et al.,* 2011).

The addition of PAC into an MBR decreases the mixed liquor viscosity and increases the sludge particle size leading to lower TMP values. As its addition decreases the concentration of EPS, it slows down the development of the cake resistance mitigating membrane fouling (Li and Gao, 2011). In addition, it also reduces the concentration of SMP, another major foulant, consequently slowing down the development of concentration polarization layer resistance and preventing further membrane fouling.

The addition of PAC to MBRs can adsorb EPS, SMP and colloids existing in the mixed liquor (main membrane foulants), hence successfully mitigating membrane fouling. For example, colloidal TOC concentration in the sludge of a PAC-assisted MBR has been found to be 62% lower than that in the sludge of an MBR operating without addition of PAC (Yang *et al.,* 2010). Furthermore, the addition of ACs in mixed liquor leads to the formation of larger flocs. Lin *et al.,* (2011) showed that the mean floc size in a PAC-amended MBR was 84 µm, whereas in a conventional MBR operating without any AC was 56 µm. As floc size does affect membrane fouling development, with larger flocs leading to better operating conditions in terms of membrane fouling, it can be concluded once again that the addition of ACs significantly contributes to membrane fouling control (Lin *et al.,* 2011).

In addition to PAC, the effect of GAC on membrane fouling has been studied and identified. Johir *et al.,* (2011) operated an MBR without any adsorbent at an aeration rate of 1.0 m^3 $m^{-2}_{\text{membrane area}}$ h^{-1} and achieved a maximum MPF of 25 L m^{-2} h^{-1} – any attempt to increase the MPF further led to sudden TMP increase indicating that membranes had become fouled. However, sudden rise of total membrane resistance could be prevented when GAC of particle size of 300 - 600 µm was used in suspension (ranging from 0.5 g L^{-1} to 2 g L^{-1}). Similar to PAC, this may be attributed to the ability of GAC to prevent foulants from being deposited on the membrane or to scour foulants off the membrane surface due to extra shearing stress. In addition, a significant reduction in the aeration rate to 0.5 m^3 m^{-2} h^{-1} at an even higher MPF was achieved. Membrane resistance analysis following GAC dosing was also in line with that carried out for PAC: the total membrane resistance increased when the concentration of GAC decreased, indicating that the amount of the suspended medium is a key issue regarding membrane fouling reduction at a certain set of operating conditions. The membrane resistance analysis finally showed that the addition of GAC maintained pore blocking at a low level regardless of the aeration rate and the MPF that were applied and also reduced the cake deposition on the membrane surface.

5.3 Effect of activated carbon dosage on membrane fouling

Available studies highlight the importance of AC dosage for membrane fouling control. As noted earlier, "optimum" dosages of ACs result in a significant reduction in membrane fouling via the following possible mechanisms: (i) through improved scouring of the membrane surface by the PAC, (ii) through adsorption of membrane foulants by the PAC and their subsequent biodegradation, and finally (iii) through the positive effect that PAC has on the strength of sludge flocs. However, overdosing with PAC may fail to reduce membrane fouling because of its potential to become a foulant itself, either through the formation of a cake layer over the membrane and/or by blocking membrane pores (Yang *et al.,* 2012a). Up to an "optimum" dose, PAC addition can control membrane fouling formation and the extent of concentration polarization, which can be attributed to the high porosity and loose compaction of the cake layer formed by PAC on the membrane. PAC particles on the membrane surface is believed to reduce the thickness of hydrodynamic boundary layer, hence, permeate can flow more easily through the membrane. However, under an excessive dose, PAC will start depositing on the membrane surface and will start plugging the membrane pores, thus triggering steep TMP-rise (Ying and Ping, 2006). The "optimum" dose, however, appears to vary with wastewater strength and operational parameters such as SRT.

PAC dosages ranging between 0.5 - 3 g L^{-1} (Iversen *et al.,* 2009b) have been commonly explored for MBR applications; however very high dosages of 50 - 80 g L^{-1} have also been tested (Seo *et al.,* 2004; Ma *et al.,* 2012). In the current review, PAC dosages up to 0.5 g L^{-1} will be considered as low dosages. Even though the addition of PAC does manage to increase the MPF that can be maintained under certain operating conditions, there seems to be an optimum dosage above which the MPF starts deteriorating. This can be attributed to increased sludge viscosity at high PAC concentrations. On top of that, the addition of PAC in MBRs operating without sludge removal shows unstable performance, indicating that regular replacement of aged BPAC with fresh PAC is necessary (Meng *et al.,* 2009). In general, it has been found that more frequent addition in smaller dosage may be preferable to higher dosage but less frequent PAC addition mode. Nguyen *et al.,* (2014) tested two PAC dosages, namely 0.1 g L^{-1} and 0.5 g L^{-1} for the removal of a range of resistant TrOCs, and observed stable performance for a longer period under the higher PAC concentration. However, comparison of the removal efficiency while taking into consideration the treated volume per unit weight of PAC confirmed the suitability of the smaller dose PAC addition up to a certain level of loading on PAC.

Even though most of the available studies explored the use of PAC in relatively high concentrations, low PAC dosage (in the range of 0.5 g L^{-1}) coupled with short SRTs can be beneficial for fouling mitigation in MBRs (Remy *et al.,* 2010). Such an operation regime can improve the critical flux of the MBR system, increase the filtration period at higher MPFs, decrease the gel deposition on the membrane surface during prolonged operation, and make the removal of the membrane gel layer easy. In addition, application of low PAC dosages may bring about additional advantages with regard to oxygen transfer and dewaterability, which may provide savings on operational costs. However further research on this aspect is necessary (Remy *et al.,* 2009, Remy *et al.,* 2012).

Finally, in an attempt to estimate the optimum PAC dosage, Iversen *et al.,* (2009a) observed no impact of PAC dosing on the turbidity of the MBR mixed liquor at PAC concentrations lower than 0.1 g L^{-1}. On the other hand, at higher concentrations of PAC (7 g L^{-1}), turbidity increased by 26%, showing that at these very high concentrations the PAC particles (8 - 35 µm) may remain in suspension and not be embedded into the flocs, which can increase membrane blockage or abrasion.

6. Activated carbon-assisted anaerobic membrane bioreactors

ACs, both PAC and GAC, have also been tested with AnMBRs as shown in Table 3. In terms of pollutant removal, PAC has been found to be more efficient than GAC (Hu and Stuckey, 2007). Following operation of three laboratory scale AnMBRs including one control MBR without AC, one MBR dosed with 1.7 g L^{-1} of PAC and a third one dosed with 1.7 g L^{-1} of GAC, Hu and Stuckey, (2007) reported >90% COD removal by all MBRs. However, the average COD removal in the PAC-assisted MBR was higher than the average COD removal in the GAC-assisted MBR, which was not significantly better than that of the control MBR. As PAC has a significantly greater surface area per mass than GAC, it is probable that this difference was primarily due to the greater adsorption of fine colloidal particles and high molecular weight organics onto PAC (Hu and Stuckey, 2007). In general, comparison between PAC-assisted and conventional AnMBRs confirm that AC enhances the stability of MBR operation and the removal efficiency of COD, volatiles fatty acids (VFA), turbidity and colour (Baêta *et al.,* 2012). For example, Trzcinski and Stuckey, (2010) reported efficient removal of bisphenol A and bis(2-ethylhexyl)phthalate.

Similar to aerobic MBRs, AC dosing has been explored for mitigation of membrane fouling in AnMBRs (Kim *et al.,* 2011). Membrane fouling is more critical for AnMBRs as anaerobic sludge has worse filterability. Fine colloids existing in the anaerobic broth are indeed responsible for the increase in the cake layer resistance (Yang *et al.,* 2012a), and PAC addition may reduce their (foulants) direct deposition onto membrane. Increased salinity leads to higher SMP concentrations in bioreactors. Thus, in AnMBRs treating saline wastewater, the advantages of PAC addition may not be as pronounced; however, the performance is still expected to be better than conventional AnMBRs (Vyrides and Stuckey, 2009). It is worth noting here that the sooner the PAC is added to an AnMBR, the more efficient it can be. SMP and inorganics that have already been attached to a membrane below the biofilm layer cannot be removed easily, even after the addition of PAC. Therefore, PAC addition can be more efficient if added during the start-up period (Vyrides and Stuckey, 2009).

The addition of PAC to an AnMBR results in the efficient reduction in high molecular weight compounds, whereas the low molecular weight compounds are not adsorbed by PAC as efficiently. Vyrides and Stuckey, (2009) reported that the amount of high molecular weight compounds reduced by 69% during the first 72 h and by

74% in the following 144 h, whereas the amount of low molecular weight compounds reduced only by 31% after 210 h of operation. It is likely that the high molecular weight compounds released by the biomass have a higher charge compared with the lower molecular weight ones that are substrate intermediates. As a result, the charged compounds are adsorbed more effectively by PAC (Vyrides and Stuckey, 2009).

The fraction of fine particles deposited on the membrane surface is remarkably reduced when PAC addition takes place. PAC also restricts membrane attachment of flocs with high concentrations of SMP on their outer surface. This may be attributed to membrane scouring that PAC particles may perform. PAC scours the membrane resulting in the removal of the flocs and colloids from the gel layer and adsorbs the high molecular weight SMP and colloids from the mixed liquor of AnMBR, facilitating higher DOC removal efficiencies and stable membrane flux (Vyrides and Stuckey, 2009).

In general, PAC addition has been found to alleviate membrane fouling, and this is mainly attributed to the adsorption of fine particles and their further biodegradation. However, in the longer run, it may have a negative effect on membrane fouling mitigation as optimal PAC dose is usually case-specific and it is always possible that its addition may deteriorate the filterability of the suspension as PAC in excess can become a membrane foulant. Regular replacement of exhausted PAC also puts an additional constraint to its use. An important aspect that needs to be taken into account is the fact that the removal of used or inactive ACs inevitably leads to active biomass loss, hence reducing sludge concentration. This is critical in regard to anaerobic bioreactors in which microbes grow slowly and any biomass loss must be prevented. Therefore the application of AC addition to an AnMBR must be carefully considered (Yang *et al.,* 2012a).

7. Future research priorities

The improved performance of MBRs due to the addition of ACs is subject to several parameters: among them of the utmost importance is PAC dosage. PAC overdosing may in fact aggravate membrane fouling. There is no clear consensus about the concentration range in which AC usually acts as a flux enhancer and when it in fact acts as an additional pollutant. To date attempts to optimize AC dosage have generally returned case-specific results, and further systematic studies are deemed imperative. It is thus recommended that, during start-up of an MBR, the addition of high quantities of AC should be avoided - ideally, initial addition should be restricted to about 0.5 g L^{-1}, and, then, depending on the performance of the MBR, dosage of AC can be gradually increased.

More research also needs to be carried out regarding the applied SRTs in AC-assisted MBRs. It is well known that maintenance of adequate SRT is vital for removal of both TOC and nutrients (nitrogen and phosphorous), and in general longer SRT may facilitate proliferation of special degrading microbes. However, it is also noted that excessively long SRT (and consequently maintenance of very high mixed-liquor suspended solids (MLSS) concentration) may be counterproductive in terms of membrane fouling control. Thus in conventional MBRs the SRT is fixed based on both removal performance and membrane fouling control considerations. In this context, it is interesting to note that compared to conventional MBRs it may be worth working at shorter SRTs when PAC is added to an MBR. This is to facilitate timely replenishment of saturated PAC and thus avoid the adverse impacts of aged PAC on membrane fouling. However, too short SRTs may lead to inefficient biodegradation as microbes need time to acclimatize and biodegrade. Thus further research is necessary to elucidate the effect of SRT and, if possible, determine a range of optimum SRT values for PAC-amended MBRs. This will also allow optimization of AC dosage, thus improving also the process from an economical point of view.

The elucidation of the mechanisms involved in mitigation of fouling in AC-amended MBR as well as clarification of the "AC bioregeneration" concept are important research gaps impeding the widespread application of this process. Furthermore, majority of the studies involving direct dosing of AC have reported the use of PAC, while the application of GAC has been mainly restricted in post treatment of MBR effluent in the form of a packed column. Compared to PAC, GAC may act differently as a biocarrier owing to its larger particle size. On the other hand, carbon based nanoparticles may perform better than PAC or GAC because of their high surface area. Systematic studies on the impact of particle size of carbon-based adsorbents are highly necessary.

8. Conclusion

This review concentrated on the effect of AC addition to MBRs treating wastewater. There is a strong consensus in the literature that coupling AC adsorption with biodegradation and membrane filtration leads to better permeate quality, and facilitates membrane fouling control. It is, however, notable that AC addition may only slightly improve the final permeate quality in terms of bulk effluent quality parameters *e.g.,* COD. The advantage of AC-assisted MBRs compared to conventional MBRs is strongly-related to their ability to remove resistant pollutants. Factors such as SRT and AC dosage are important determinants of performance of AC-assisted MBRs.

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Figures

Figure 1

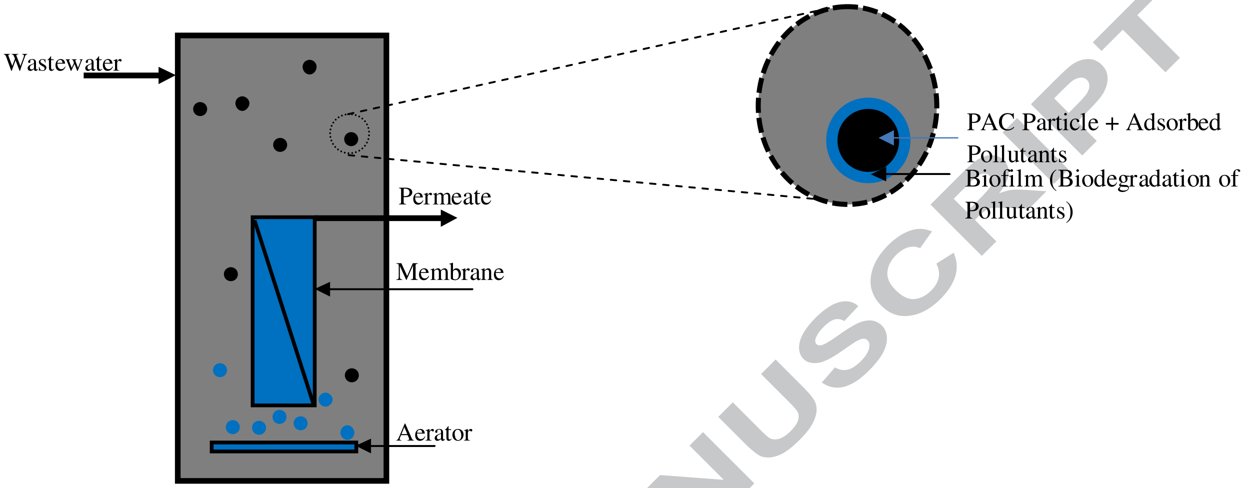

Figure 1: Submerged PAC-amended Membrane Bioreactor

Tables

Table 1: Pollutant removal and reduction of toxicity/stress on microbes with the aid of ACs

Pollutant/Stress	**Selected References**
Phenol and Phenolic Compounds (PAC)	Thuy and Visvanathan (2006), Li and Wang (2008)
Dyes (PAC)	Hai *et al.* (2008), Baêta *et al.* (2014)
ROM (PAC)	Whang *et al.* (2004)
Natural Colour (PAC/GAC)	Abegglen *et al.* (2009)
Micropollutants: EDCs/Pharmaceuticals/Hormones/Bactericides, *etc.* (PAC/GAC)	Yang *et al.* (2012), Snyder *et al.* (2007), Nguyen *et al.* (2012), Li *et al.* (2011a), Li *et al.* (2 011-2), Lipp *et al.* (2007), Han *et al.,* (2008)
Toxicity	Lesage *et al.* (2005), Lesage et al. (2008), Baêta *et al.* (2014)
DOM (PAC)	Tian *et al.* (2008)

ROM: Residual Organic Matter, EDCs: Endocrine Disrupting Compounds, DOM: Dissolved Organic Matter

Table 2: MBR and membrane configurations combined with activated carbons

MBR Configuration	**Type of Activated Carbon**	**Membrane Configuration**	**Area (m^2)**	**Pore Size (µm)**	**Reference**
Sub	PAC	FS	0.2	0.14	Johir *et al.* (2011)
Sub	PAC	HF	0.1	0.2	Lesage *et al.* (2008)
Sub	GAC	HF	-	0.2 - 1 (mm)	Li and Wang (2008)
Sub	PAC	FS	0.4	140 (kDa)	Lin *et al.* (2011)
Sub	PAC	HF	1	0.1	Liu *et al.* (2007)
Sub	PAC	HF	0.06	0.1	Ma et al. (2012)
Sub	PAC	HF	6	0.4	Munz *et al.* (2007)
Sub	PAC	HF	-	50 (kDa)	Ng *et al.* (2009)
Sub	PAC	Mesh Filter	0.05	30	Satyawali and Balakrishnan (2009)
Sub	PAC	HF	-	0.1	Seo *et al.* (2004)
Sub	PAC	HF	0.25	0.2	Ying and Ping (2011)
Sub	PAC	HF	0.4	0.01	Tian *et al.* (2008)
Sub	PAC	HF	0.074	0.01	Li *et al.* (2011-1)
Sub	PAC	HF	0.42	0.1	Jamal Khan *et al.* (2012)
Sub	PAC	FS	0.1	0.4	Vyrides and Stuckey (2009)
Sub	PAC	FS	0.1	0.4	Aquino *et al.* (2006)
Sub	PAC	Mesh Filter	0.05	58	Chu *et al.* (2013)
Sub	PAC	HF	0.04	0.4	Whang *et al.* (2004)

FS: Flat Sheet, GAC: Granular Activated Carbon, HF: Hollow Fibre, PAC: Powdered Activated Carbon, Sub: Submerged

Table 3: Case Studies of Aerobic Membrane Bioreactors and Activated Carbons

Scale	**Type of Wastewater**	**Volume (L)**	**MLSS (g L^{-1})**	**SRT (d)**	**HRT (h)**	**Dosage (g L^{-1})**	**Initial COD (Average) (mg L^{-1})**	**Final COD (Average) (mg L^{-1})**	**COD Reduction (%)**	**Reference**
Lab/A	Secondary Effluent	37.5	4.5	-	4	0.75	10.7± 1.32 (TOC)	8.98 ± 0.62 (no PAC) 4.0 ± 0.84 (with PAC)	16.1 ± 4.6 (no PAC) 62.7 ± 7.7 (with PAC)	Lin *et al.* (2011)
Pilot/A	Tannery Wastewater	0.52 m^3	12 - 18 (TSS)	32 - 95	50 - 100	0 - 3	4051	-	Low (but not Negligible Improvement) with PAC	Munz *et al.* (2007)
Pilot/A	Municipal Wastewater	85	9.6 (no PAC) 10.1 (with PAC)	25 - 50	10	0.5 (in terms of sludge)	300	33 (no PAC) 30 (with PAC)	-	Remy *et al.* (2010)
Lab/A	Synthetic Wastewater	2	12 ± 1 (no PAC) 17 ± 1 (with PAC)	30	4	0 - 5	370 ± 10 (TOC)	15 (TOC) (no PAC) 5 (TOC) (with PAC)	-	Ng *et al.* (2006)
Lab/A	Medicine Wastewater	35	-	-	~6	0.25	575 - 3201	-	89.27 (no PAC) 89.79 (with PAC)	Liu et al. (2007)
Lab/A	Domestic Wastewater	24	-	30	-	0 - 1.5	271.44 - 575.24	-	29.15 - 47.26 (no PAC) 24.36 - 47.52 (with PAC)	Ying and Ping (2006)
Lab/A	Low-Strength Synthetic Wastewater	2 (Effective)	-	-	2	0 - 75	5 - 6.5 (DOC)	4.64 (DOC) (no PAC) 1.16 (DOC) (with PAC)	80% (with PAC)	Ma *et al.* (2012)
Lab/A	Synthetic Wastewater	-	9	20	24	0.2 g d^{-1}	-	-	94 ± 2 (no PAC) 96 ± 2 (with PAC)	Lesage *et al.* (2008)

Lab/A	Sewage-contaminated Surface Water	2 (Effective)	-	20	0.5	8 (mg L^{-1})	4.13 ± 0.37	2.66 ± 0.29 (no PAC) 1.66 ± 0.21 (with PAC)	35.3±7.6 (no PAC) 59.5±6.8 (with PAC)	Tian *et al.* (2008)
Lab/An	High Salinity Synthetic Sewage	3 (Effective)	-	250	8	1.7	145 ± 10 (DOC)	-	93% (no PAC) 98% (with PAC)	Vyrides and Stuckey (2009)
Lab/An	Municipal Solid Wastes Lechate	10	4.4	30	1.5	2	5000	2380 (SCOD) (no PAC) 1550 (with PAC)	-	Tzcinski and Stuckey (2010)
Lab/An	Low-strength Synthetic Wastewater	3	2.6 ± 01.3 (no PAC) 3.7 ± 0.19 (with PAC)	150	6	1.7	450 ± 20	18 ± 11 (no PAC) 18 ± 9 (with PAC)	-	Aquino *et al.* (2006)
Lab/A	Biologically Treated Swine Wastewater	22.1	-	-	2.5	1 - 10	217	172 (no PAC) 32 (with PAC)	88.7 (with PAC)	Whang *et al.* (2004)

A: Aerobic, An: Anaerobic, COD: Chemical Oxygen Demand, DOC: Dissolved Organic Carbon, HRT: Hydraulic Residence Time, MLSS: Mixed Liquor Suspended Solids, PAC: Powdered Activated Carbon, SCOD: Soluble Chemical Oxygen Demand, SRT: Solids Residence Time, TOC: Total Organic Carbon

HIGHLIGHTS

1. AC addition to MBRs fortify adsorption, potentially enhancing biodegradation
2. AC-assisted MBRs more effectively remove resistant pollutants than usual MBRs
3. AC addition to MBRs can retard membrane fouling and improve membrane flux
4. For AC-assisted MBRs, AC dosage and retention time must be carefully controlled
5. Frequent but low-dose AC addition may facilitate timely replenishment of spent AC