Municipal wastewater reclamation and reuse using membrane-based technologies: a review

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ABSTRACT

Human well-being and socio-economic development of the society depend notably on two invaluable resources water and energy. Water scarcity and increasing water demand have made water supply a challenge to the world. Thus, further efforts should be made to develop and improve technologies for wastewater treatment and reuse'which can provide an alternative water supply. Membrane technology is the most efficient technology in wastewater treatment, and thus this paper mainly reviews the recent advances of membrane-based technologies applied to wastewater treatment and reuse. Firstly, the potent pollutants in wastewater and the related traditional treatment methods were discussed. Then, the development, applications and challenges in membrane technology for wastewater treatment were reviewed. Furthermore, the membrane-based integrated technologies and the prospects of these technologies were discussed, including membrane filtration combining with pre-treatments, membrane filtration combining with activated sludge process and membrane filtration combining with the advanced oxidation process.

Keywords: Wastewater reuse; Potent pollutants; Membrane-based technology; Advanced oxidation processes; Integrated technology

1. Introduction

Water is essential for human survival and social development [1]. The increasing water demand contrasts sharply with diminishing water supplies especially in arid regions [2]. Scarce water resources and uneven water distribution are regional problems all over the world, for example, North Africa, the Middle East, southern Europe, Australia, and the southern states of the USA [3]. Water scarcity in China is more severe than before with no sufficient water resources to meet the increasing water demands [4]. Not only the uneven distribution, but also the pollution of water resources made two-thirds of China's 669 cities have the problem of water shortages [5]. Overuse of natural formation water such as groundwater exploitation and seawater desalination is consuming the existing water

resources. And the city's groundwater overexploitation of underground voids will cause land subsidence, resulting in significant impacts on the geological and ecological environment [6]. Meanwhile, due to human activities and climate change, land cover change, the water ecosystem problems have been increasingly prominent [7,8]. In order to deal with the problems mentioned above, reclaimed water is an ongoing area of focus that is developed as the alternative water sources [1].

It is estimated that approximately 12 billion gallons of municipal wastewater effluent are discharged to the ocean in the USA daily. While, only under 10% of the effluent is reused in the USA (Fig. 1) $[9-12]$. If such huge amounts of wastewater were treated meeting specific water quality criteria, it would become a valuable resource and could be reused for a range of purposes. In 2018, the total amount of

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Fig. 1. Total water withdrawal (a) of different countries (Africa: Egypt, Morocco and Tunisia; America: Argentina, Brazil, Mexico, Peru and USA; Asia: China, Japan and Saudi Arabia; Europe: France; Oceania: Australia), and the produced municipal wastewater, treated municipal wastewater and direct use of treated municipal wastewater as a percentage of the treated municipal wastewater (b) [9–12].

domestic water in China is 85.99 billion m^3 , and the sewage discharge treatment rate has reached 92%, but the recycling rate is only about 10%. China's total industrial water consumption is 126.16 billion $m³$. According to the requirements of environmental protection and clean production, most of the industrial wastewater needs to be recycled, especially the high salinity industrial wastewater must reach zero discharge or recycling [13]. Governments of China have released policies intending to promote wastewater recycling with the advancement of a national strategy for wastewater reclamation and reuse. It is widely known that industrial wastewater especially from textile mills, tanneries and refineries contains many pollutants. The typical textile dyeing process produces wastewater characterized by a perceptible content of surfactants [14,15]. Wastewater generated from leather industries contains a complex mixture of nitrogen, trivalent chromium, tannin, sulfate, and other ions [16]. In refinery process, notable constituent contains phenols, ammonia, H₂S and BTEX (benzene, toluene, ethylbenzene and xylenes) [17]. At present, a large majority of sewage treatment technologies are utilized to meet the pollutant emission standards. Progress should be made to improve the existing technologies transforming from "discharge" to "recycle" and achieve the standards of water reuse.

Industrial wastewater and urban wastewater can be reused in a variety of ways including process water and boiler feed water in factories, irrigation and landscape water and even potable water (Fig. 2) [18–20]. A lot of researches have demonstrated the feasibility of wastewater reuse.

Fig. 2. Proportion of the different applications of water reuse in global (a) [18], Japan (b) [12], Australia (c) [19], and USA (d) [20].

With further attention and researches on wastewater reuse paid by scholars around the world, membrane separation processes have made a remarkable development in recent years. The use of the membrane-based technologies has been adapted successfully, gradually replacing the conventional treatment methods which consume a large amount of energy [21,22]. More than that, the processes are highly efficient, easy to operate and occupy less space compared with conventional technologies such as sedimentation process, anaerobic reactors [23,24]. While the traditional single technology has been strengthened further, combined processes, the integration of different technologies, grow up gradually and then play key roles in wastewater reclamation areas [2].

With the rapid development of science and technology, numerous researches on innovation technology based on membrane filtration and membrane separation have been reported within a few years. All the efforts and progress of the development of technology aim at achieving mild synthesis, quick reaction and high performance. Moreover, several problems during the operation process including membrane fouling, toxic by-products should be solved. Therefore, this paper summarized several potent pollutants in municipal wastewater, and reviewed the innovation and developments of wastewater reuse and reclamation technologies within the last 7 y (2014–2020), especially removal of trace emerging organic compounds and the integrated process. Also, it revealed the prospect and trends of wastewater reuse.

2. Potent pollutants in municipal wastewater treatment and reuse

2.1. Refractory organics

With the growing type and discharge of sewage, the constituents have been becoming complicated which contain many refractory organics such as phenol, benzene sulfonic acid, chlorine phenols, pesticides, polychlorinated biphenyls, polycyclic aromatic hydrocarbons, nitro aromatic hydrocarbon compounds, dyes and humic acid and so on [25]. They are called the refractory organics since it is difficult to be degraded by microorganisms and its decomposition is incomplete, which may be cleaved to toxic metabolites. Although there are no specific regulations covering these compounds, they are being deeply investigated because their presence was found as a potential cause of damage to the quality of natural water [24,26]. Such pollutants easily accumulate in the organisms and become a possible source of water pollution. Some of these organics have carcinogenic, teratogenic, mutagenic and other effects causing enormous harm to the environment and humans. The common features of these substances are toxic, composition-complex, high chemical oxygen demand. It's not surprising that general microbial degradation has limited effect on them. Consequently, the treatment of refractory organics has attracted the attention of domestic and foreign experts and it has become the hot and difficult point of the prevention and treatment of water pollution.

At present, the traditional treatments of refractory organics mainly include physical, biological and chemical methods. Physical methods include coagulation technology and adsorption technology. However, the former will lead to a significant increase in the salt content of the effluent, which will increase the conductivity of the effluent and reduce the reuse rate of wastewater, and the high cost of the latter limits its wide application [27–29]. Advanced oxidation is a common chemical method, which has a high removing efficiency, however, the operation of this process is complex and high-cost [30]. The traditional biological treatment is ineffective sometimes, and needs to be improved. Efficient strain theory shows that any organic material can be biodegradable [31,32]. The petrochemical and oil industrial wastewater contains high organics concentration [33]. So, the bacterium with special decomposition ability is required to isolate to deal with the industrial wastewaters [34]. Haddadi and Shavandi [35] found that when exposed to 100 mg/L of phenol as the sole source of carbon and energy, the selected strains made the degradation of phenol up to 1,100 mg/L. Despite that, all microbial can adapt to the degradation of organic pollutants in theory, when faced with endless challenges of new compound today, only relying on the adaption capacity of microorganisms is clearly lagging behind. Co-metabolism technology, according to the theory of co-metabolism of microorganisms, many hard-biodegradable organics can be degraded when they are in company with easy-degradable organic matters, such as glucose, ethanol, etc. Lu et al. [36] employed primary substance in starch wastewater treated by photosynthetic bacteria, which improved the efficiency greatly. As a method of biodegradation of refractory organics, the co-metabolism technology is relatively low in operation cost and does not bring secondary pollution, but it is still in the laboratory research stage, there are many technical bottlenecks to be solved, and it is still a certain distance from the application of practical engineering.

2.2. Pathogenic microorganisms

Waterborne infectious diseases have been identified as the main source of high morbidity and mortality worldwide that cause about 2.2 million deaths per year [37]. There is only one index of fecal coliform bacteria in the discharge standard of China's sewage treatment plant, which does not require the content of other pathogenic microorganisms. Therefore, there is a possibility that pathogenic microorganisms can be transmitted through the reuse of effluent from sewage treatment plant [38]. Reported outbreaks of viral infectious diseases caused by insufficiently treated wastewater emphasizes the importance of wastewater treatment as a barrier for the virus transmission, especially for reclamation and reuse [39]. This consist of two categories relating to the path of transmission. First, diarrhea, dysentery and gastroenteritis, caused by Salmonella, *Shigella*, pathogenic *E. coli* and *Vibrio*, occur because of the ingestion of water contaminated by pathogenic microorganisms [40]. Then, when exposed to contaminated water bodies in labor, swimming or other processes, people may be infected with the diseases infecting through the skin and mucous membrane, such as schistosomiasis and leptospirosis [41]. Reclaimed water containing pathogenic microorganisms above will cause harm to human health. There is no doubt that removal of these pathogenic microorganisms by wastewater treatment processes is required if treated wastewater is reclaimed.

The most commonly used methods to remove pathogenic microorganisms are chlorine disinfection and ozone disinfection, comparatively simple chemical-adding oxidation methods. Since the 1840s, to prevent the spread of infectious diseases which infect through the water medium, chlorine and ozone sprung up as disinfectant. Chlorine is the primary means of disinfection in many countries for treating wastewater containing microbial pathogens [42,43]. Melilla Constance, a French scientist, noted that even thin ozone air can also make sewage disinfection and sterilization effect in 1886. Ozone has attracted growing interests since that it does not produce harmful halogenated organic compounds in the water treatment process. Bacteria are sensitive to disinfectant, so low concentrations and short-term ozone treatment can achieve significant effect. However, viruses are more resistant to water disinfection treatments [44], meanwhile the resistance of spores is about 10 times more than the virus. So, the removal efficiency is unsatisfactory. When under conditions of chlorine 6.6 mg/L and contact time 15 min, only 50% of polio virus was inactivated [45]. Therefore, there is a need to increase the concentration of ozone and the adequate contact time to inactivate virus and spores.

2.3. Nitrogen and phosphorus

The heavy use of fertilizers and pesticides increased the nitrogen and phosphorus content in wastewater, which is the main cause of eutrophication [46,47]. Wastewater treatment plants discharge effluents that normally contain significant amounts of dissolved organic nitrogen into surface water. In recent years, the ammonia emissions of domestic sewage and industrial wastewater is increasing rapidly and most of which comes from organic nitrogen. Organic nitrogen will produce ammonia when decomposed by ammoniated bacteria. When wastewater containing ammonia-nitrogen is discharged into the water, especially the slow-flowing rivers and lakes, it could prone to algae and other microorganisms multiply resulting in eutrophication phenomenon [48]. In addition, nitrogen and phosphorus in reuse water should be eliminated to prevent excessive proliferation of biological mucous membrane in water pipelines and water equipment surface which may cause blockage or inefficiency. Ammonia generates

chloramine after reaction with chlorine [49], thus increasing the amount of chlorine needed for disinfection and improving water treatment costs. Given that the municipal sewage is developed as second water source of a city, we must control the content of nitrogen and phosphorus.

With the in-depth study of biological nutrient removal technologies, two traditional technologies including anaerobic ammonium oxidation (Anammox) and denitrification phosphorus removal are widely utilized, which give an enormous impetus to the development of wastewater nitrogen and phosphorus removal. First, Anammox: under anaerobic conditions, ammonia is oxidized to nitrogen directly in the process when nitrite conducts as an electron acceptor. The reaction equation is: $NH_4^* + NO_2^- \rightarrow N_2 + 2H_2O$ [50,51]. Then, denitrification and phosphorus removal mean that under hypoxia (oxygen-free, but the presence of nitrate nitrogen) conditions, denitrifying phosphorus-removing bacteria, using nitrate as an electron acceptor, can produce biological phosphorus uptake role. Meanwhile, the nitrate is reduced to nitrogen [52,53].The common methods for chemical phosphorus removal include chemical precipitation [54], adsorption [55] and electrocoagulation [56].

Existing traditional wastewater treatments would be decided on achieving discharge limits set by national or international environmental regulations [57]. Nevertheless, produced water which satisfies environmental discharge regulations is not in accordance with the standard of water reuse. Stricter water quality regulations cannot be effectively met by conventional treatment processes [58]. To date the most stringent regulations have been issued in the United States by the California Department of Public Health, which relate to indirect reuse of wastewaters as a source of raw drinking water through groundwater [59]. As a consequence of the increase in water quality regulations, we need to add advanced treatment technologies to traditional treatment processes and give intensive treatments to various pollutants in sewage, making it a better return for use of the reclaimed water.

3. Membrane separation technology

3.1. Development of membrane separation technology

The first microfiltration (MF) began to appear in 1930s and it was used initially in the medical science to remove the micro-aggregates like fibrin particles and blood cells in stored blood. Its technical characteristics including uniform pore size, high filtration precision, less absorption and no media loss drive the incredible progress of microfiltration. It is widely used in the sterilization and decontamination of beverage and pharmaceutical products in food and pharmaceutical industries, and also used in the removal of particles of ultrapure water disposal process in semiconductor industry, and in the concentration and separation of biological products in fermentation broth in the field of biotechnology. Electrodialysis (ED) was introduced in the 1950s. ED is an electrochemical separation process in which charged ions and uncharged component in the solution is separated through the ion-exchange membrane under the influence of an electrical field [60,61]. It is established and investigated for producing fresh water from saline

water. The main advantages of ED are less water pre-treatment, higher selectivity and the option of ED reversal for membrane fouling control [62].

In 1960, the first asymmetric membrane produced by Loeb and Souriraian was made of cellulose acetate [63]. It is capable of providing practical levels of water flux while maintaining high levels of salt rejection, and it is the basis for discovery of reverse osmosis (RO) and ultrafiltration (UF). RO membrane is very hydrophilic. Therefore, water will be able to readily diffuse into and out of the membrane polymer structure [64]. Add a higher pressure than osmotic pressure to the side of the liquid to be separated, then liquid solvent is pressed through the semi-permeable membrane into the other side. RO process is the reverse process of forward osmosis (FO). In some cases, FO can be used as an advanced pre-treatment process for RO [65,66]. The features of reverse osmosis technology are no phase change, low energy consumption, high membrane selectivity, compact device structure, easy operation, easy maintenance, and no pollution to the environment.

In 1980s, the high operating pressure of the traditional the RO process caused the increase of energy loss. Also, the effluent from RO system was unable to meet water requirements. Therefore, membranes with lower solute retention and higher osmotic flux as the emerging field had come to the attention of researches. Nanofiltration (NF) is a nanoscale-aperture reverse osmosis technology with a porous membrane and has a high permeability under low pressure. NF membranes, whose characteristics fall between UF and RO, operate with no phase transition, high-energy efficiency and typically have high rejections of multivalent inorganic salts and small organic molecules at modest applied pressure [67]. The major mechanism of NF interception is size exclusion and hydrophobic adsorption [68]. The wastewater treated with baker's yeast by the two-step NF system met the agricultural irrigation water quality according to the Turkish Standard [69]. The pressure difference provides the driving force for the separation. UF gives a good rejection performance in producing solid-liquid separation or the classification of different molecular weight substance using the high-precision ultrafiltration membrane. Its technical feature is that it can have the concentration and separation process of macromolecules or colloidal substances at the same time. With lower operating pressure and energy consumption, higher selectivity and no phase change compared with RO, UF has a wide application in medicine, industrial wastewater treatment, ultra-pure water preparation and bio-technology industries.

Although membrane separation technology has been found wide applications in wastewater treatment, several pressing issues limit widespread implementation of wastewater reuse including membrane fouling, selectivity and trade-off between permeability and selectivity, low removal efficiency and energy consumption. Advancements in membrane technology and innovation design in membrane process have been developed to improve the above problems [70]. The novel technologies include new membrane materials, modification, membrane cleaning, as well as emerging combined technology based on membranes.

3.2. Improvements of membrane fouling, selectivity and removal efficiency

3.2.1. Membrane materials and membrane modification

Membranes have been fabricated from a variety of different materials with different membrane characteristics. Surface charge, hydrophobicity and roughness are three main surface characters of the membrane and play the important roles in the membrane separation process. Surface charge value can be obtained according to zeta potential measurements. Membranes with high negative surface charge accumulate lower amounts of foulants [71]. Hydrophobic membranes tend to get dirty more quickly and therefore have a more pronounced decline of the permeate flux and a reduced lifespan. Usually, UF membranes are less hydrophilic than NF [72]. Two UF (OT050, GR60PP) and four NF (DK, CK, TFC-SR3, MPF-34) commercial polymeric flat membranes were tested and compared in a study. Several indexes showed that NF membranes had better characteristics than those of UF. Hydrophilicity is determined through dynamic contact angle measurements. Not only can membrane modification enhance the hydrophilicity of the substrates, but also smoothen the top surface which could be examined through an atomic force microscope [73]. Another requirement of the membrane is to be tolerant to chemical cleaning processes [74]. For nanofiltration membranes, different membranes include different materials for the active layer and the support layer. The most popular material for the active layer is polyamide and for the support layer is polyether sulfone. Hydrophilic and biologically persistent compounds could be removed more completely with polyamide thin film composite membranes compared to cellulose triacetate membranes [75]. In recent years, some emerging carbon-based materials have been introduced into reverse osmosis desalination processes and membrane bioreactors [76]. Carbon nanotube [77] and graphene oxide [78] depending on high antibacterial activity [79,80] and hydrophilicity have attracted growing interests and exhibited high antifouling performance. The functional group of graphene oxide making the membrane a large negative zeta potential also contributes to impeding biofouling process [81].

The hydrophilic polysulfone or polyethersulfone are expensive and not commercially. So, in addition to constructing composites with better characteristics, we can now define the ideal characteristics of clean membranes known as membrane modification [82]. Subsequent modification of membranes after the manufacture is studied to enhance separation performance and reduce cost. Membranes modified by dopamine or polydopamine have been widely used in FO, MF membranes. The polymerization process of dopamine to form polydopamine (PDA) makes it stable to coat on any substrate [83]. The PDA-decorated surfaces are always hydrophilic with a static water contact angle of about 40°–60° [84]. With the increase of dopamine coating time, the water contact angle initially reduced, indicating a significant increase in surface hydrophilicity and further increase of coating time had a minimal effect on the water contact angles [85]. The smaller the water contact angle is, the higher the hydrophilic is. A superhydrophilic surface's water contact angle is less than 5°. A polypropylene microfiltration membrane hydrophobicity was transformed into superhydrophilic amazingly through the two-pot coating process which demonstrated high oil/water emulsion separation efficiency and excellent antifouling performance [86].

3.2.2. Membrane cleaning

Membrane cleaning including physical cleaning, chemical cleaning, and back washing is one of the fouling preventing measures. Periodical cleaning can minimize both membrane fouling and scaling. The cleaning frequency can vary from as much as once per day to once per month in potable reuse applications depending on the quality of wastewater treated and the membrane type. Membrane fouling including deposition, adsorption and biological fouling severely limits the separation performance of membrane, leading to a reduction in membrane permeability, significant loss of productivity and increase of operational cost [87,88]. In practice, several fouling can occur simultaneously or one of those is the dominant foulants. Besides they would affect each other. Xiao et al. found the major component of the foulants was loose inorganic deposits while biofouling and organic fouling could be ignored through RO membrane autopsy analysis [89]. In another research [90], high concentrations of organic compounds severely fouled the FO membrane and substantially reduced water flux by 68% within 21 d. Not only the hydrophobicity of the membrane but also the hydrophobicity of the natural organic matter (NOM) affects membrane filtration performance. The fouling caused by the tannins could be attributed to its hydrophobicity and the tendency to absorption on the surface of the membrane [91]. Chemical tolerance of the membrane should be considered according to the type of chemical used. Inorganic UF and MF membranes like ceramic are more tolerant to chemical cleaning than the polymeric counterparts. Besides, Air scouring can help to remove loose foulants from the membrane active layer, thus helping to recover up to 89.5% of the original flux. Chemical cleaning of the fouled active layer of the FO membrane was not as effective as air scouring [92]. For ED, ED reversal may be an efficient alternative for preventing fouling caused by large anions. It consists in a periodical inversion of the electric field to reduce fouling and thus achieving high water recovery [93,94].

Membranes are replaced as their performance declines due to membrane fouling caused by organic compounds while chemical cleaning cannot effectively recover its performance. Complete removal is not possible and fouling has to be tolerated up to a decrease of mass flux down to 75% of the original flux. Good operating practice calls for the chemical cleaning of the membranes if normalized permeate flow decreases by 10%, feed channel pressure loss increases by 15% or normalized salt rejection decreases by 10% from the initial conditions during the first 48 h of plant operation.

4. Membrane-based combined technologies

4.1. Membrane filtration combining with pre-treatments

Currently, a major trend of membrane filtration technology is to combine membrane filtration with other pretreatment processes, to improve the filtration properties of the membrane. With selecting the appropriate pre-treatments,

fouling of membranes and removal efficiency would be improved. The pre-treatment methods include the use of prefilters, screens, precipitation, coagulation or flocculation to reduce the amount of foulants in the feed [95].

Coagulation is reported as the most effective anti-fouling method to remove particle and colloidal organics. Al³⁺ at 5 mg/L could increase the end flux from 21 to 313 LMH. When Al^{3+} at 15 mg/L was used, the membrane flux substantially increased to 1321 LMH. 30 mg/L $Fe³⁺$ exhibited similar fouling mitigation performance [96]. So, coagulation is effective to improve the membrane flux which is also demonstrated by Yu. In Yu's research, humic acid was used to interact with hydrophobic organic contaminants PFOS and form precipitation which resulted in membrane fouling. Adding the coagulant AlCl₃ not only enhanced the PFOS rejection (>95%), but also improve the permeate flux in the NF process [97]. Ultrasound applied on RO/FO membranes reduced the formation of biofouling by suppressing algal growth and biofilm. Experiments conducted by Koh et al. showed that sonication alone had a small but significant effect on membrane fouling, however, the use of heat pre-treatment in combination with ultrasound reduced the membrane pore blockage and foulant cake growth significantly especially at higher solid concentrations [98]. FO, MF or UF is also used as a pre-treatment technique to provide high quality filtrate for the following process. The foulants from permeating into the draw solution was prevented and fouling of the downstream osmotic membrane bioreactor (OMBR)-RO operation was significantly reduced attributed to the FO pre-treatment process. A pre-treatment UF process before the NF process could remove more than 50% of the organic substances (0.97– 1.10 mg/L) from the raw seawater (2.21–2.54 mg/L) [99].

4.2. Membrane filtration combining with activated sludge as dominate treatment

Conventional activated sludge (CAS) process needs for secondary clarification and tertiary steps like sand filtration. Combining the bioreactor with CAS and a low ultrafiltration or microfiltration is used to separate effluent from activated sludge. Membrane bioreactor (MBR) can be operated in aerobic or anaerobic conditions [100]. Floc sludge can be adapted to treat wastewater and generate reuse water. While recent studies have focused mostly on granular sludge, the substitute of floc sludge, owe to its benefits such as huge biomass (up to 20 g TSS/L), rich microbial diversity, low sludge generation and ability to withstand high organic load [101].

Organics with high biodegradability have strong electron donating functional groups (e.g., amine and hydroxyl) [102]. These compounds were effectively removed (>90%) by OMBR-RO operation. While other hydrophilic trace organic compounds (TrOCs) were poorly removed because they were biologically resistant substrates. The pharmaceuticals (PhACs) removal efficiencies of the MBR-RO and MBR-NF treatment were tested [103]. The two treatments with membranes both showed high removal performance than membrane process alone (>80%) [104]. While the MBR-RO showed near complete removals (>99%) better than MBR-NF (90%). The synergy between the anaerobic membrane bioreactor (AnMBR) and the membrane distillation (MD) unit contributed to 76% to complete removal of all 26 selected TrOCs by the integrated AnMBR-MD system. In the course of the integrated system, MD played an important role in rejecting bulk organic matter and phosphate [105]. MD can utilize low-grade waste heat and solar thermal that is otherwise unusable by other means, and it has been recognized as an emerging technology in wastewater reuse treatment [106]. Aerobic granular sludge, UF, NF process were utilized successively as primary, secondary and tertiary treatment of a viable integrated system for treating municipal wastewater. High removal efficiency for organic matter and nutrients was achieved: chemical oxygen demand (COD) 99.26%, TN 98.06% and TP 98.73%. The biologically active carbon (BAC) is an ecosystem equipped with simultaneous processes of adsorption and biodegradation because of the addition of powdered activated carbon (PAC) into the MBR system. The BAC is to remove organic matter through the synergistic effect of activated carbon adsorption, ozone oxidation and biodegradation. The organic matter in water is continuously adsorbed on the surface of activated carbon, and the contact time between organic matter and biofilm is fully guaranteed, so that the efficiency of biochemical organic matter is greatly improved, and the organic matter adsorbed on the activated carbon is biodegraded. At the same time, its adsorption capacity was also restored. The removal rate of aromatic organic matters was improved from 34% to 83% and the membrane biofouling was relieved. The good performance of PAC attributed to its strong adsorption of organics matters and tendency to form BAC [107,108].

High removal of bacteriophages in membrane bioreactor can also be observed partially due to retention by the membrane and adsorption by activated sludge. Viruses are typically smaller than the nominal membrane pore sizes, while studies [109,110], have demonstrated high log removals of pathogenic viruses, which suggests the preferable performance of membrane cake layer. In the multiple-barrier concept, each unit process of wastewater treatment is assigned a credit value of pathogen reduction efficiency. Several studies have shown that different operation conditions in MBR systems have obvious effects on LRVs. The mean LRV of human adenovirus varied between 2–5.5 [111,112]. For human enteric viruses (EV), MBR treatment achieved 0.5 and 5.1 log reduction [113]. In the Lv et al. study, a LRV of 4.59 was obtained for T4 phage using a 0.22 mm membrane while the LRV of T4 phage was improved to 6.05 by using a membrane with nominal pore size of 0.1 mm [114]. The number of hydrophobic amino acids in the external capsid surface is one of the influencing factors relating to removal of the virus. The hydrophobic amino acid groups can interact more freely with the hydrophobic portions of the bacterial flocs and extracellular polymeric substance (EPS) in sludge which in turn leads to a higher LRV. A summary of recent studies of the MBR processes in wastewater treatment for reuse is presented in Table 1 [92,100,101,103,107,110,115–126].

4.3. Membrane filtration combining with advanced oxidation processes as post-treatment

Advanced oxidation processes (AOPs) are based on the in-situ production of highly reactive radical species, and have been proved to be more effective technology to remove recalcitrant molecules from wastewater. Most

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Abbreviations: "pore size; "surface area; "working volume; "bioelectrochemical; "thin film composite; [/]anaerobic membrane bioreactor; "polyethylene; "polypropylene; 'membrane distillation; ^jaerobic granular sludge;
"mo Abbreviations: "pore size; "surface area; 'working volume; 'bioelectrochemical; 'thin film composite; 'anaerobic membrane bioreactor; 'polyethylene; 'polypropylene; 'membrane distillation; 'aerobic granular sludge; *k*molecular weight cut-off; *l*polytetrafluorethylene; *m*granular activated carbon; *n*not available; SBR – sequencing batch reactor.

organics including pesticides, chlorinated compounds, phenolics and cyanides can be degraded through AOPs from a few thousand mg/L to less than 1 mg/L [127]. In general, advanced oxidation processes can be divided into two categories: chemical oxidation such as ozone, $H_2O_{2'}$, $H_2O_{2'}$ UV, Fenton, and photo-Fenton; and oxidation without the addition of chemicals including UV, electrochemical, solar photocatalysis using $TiO₂$, ultrasound and wet air. A list of mechanisms and application processes of the AOPs is provided in Table 2 [128–137].

It is reported that hydrogen peroxide photolysis with ultraviolet C radiation (H_2O_2/UVC) is widely utilized as a polishing step for the remediation of a textile wastewater, regarding its discharge into the environment and its reuse in the textile industry [138,139]. pH was identified as a key operating parameter in H₂O₂/UV process. Under acidic conditions (pH 3.5), resistance genes (*sul*1, *tetX*, and *tetG*) were removed efficiently [140]. However, organic contaminations such as 1,4-dioxane, benzoate and carbamazepine were demonstrated to be removed better across pH 5.5–8.3. For the past few years, boron doped diamond (BDD) has gained great popularity as an ideal anode [141]. Mass transfer and electrical current density are two key factors that affect the efficiency of pollutant removal and electricity use. Oxidation process may occur only in a thin reaction layer adjacent to the anodic surface, so the organic removal is often a mass transfer-controlled process.

Some novel structures of electro-oxidation reactors are emerging to enhance the efficiency such as plunger flow [142] and fluid-bed electrochemical reactors [143]. Using higher current density and stronger conditions induces the decrease of operating time and improvement of removal

Table 2 Mechanisms and application processes of the AOPs

AOP processes	Elementary reactions	Target pollutant	Reuse purpose	References
Ozone	$O_3 + OH^- \rightarrow HO_2^{\bullet} + O_2^{\bullet-}$ $O_3 + OH^- \rightarrow HO_2^- + O_2$ $2O3 + 2H2O \rightarrow 2*OH + O2 + 2HO*$	Pharmaceutical residues (Iopromide, Primidone, Acesulfame, Bezafibrate, Metoprolol, Venlafaxine, Carbamazepine, Diclofenac, Sulfamethoxazole)	Agricultural irrigation	$[128]$
H_2O_2/UV	$H_2O_2 + hv \rightarrow 2^{\bullet}OH$ $H_2O_2 + O'H \rightarrow H_2O + HO^*$	Dissolved organic carbon (DOC) in textile industry	Specific textile manufacturing process including souring, bleaching, dyeing	[129]
Fenton process	Elementary reaction: $Fe^{2+} + H_2O_2 \leftrightarrow Fe^{3+} + OH^- + {}^{\bullet}OH$ $Fe^{3+} + H_2O_2 \leftrightarrow Fe^{2+} + H^+ + HO^{\bullet}$ $H_2O_2 + O'H \rightarrow H_2O + HO_2^*$ $HO^{\bullet}_{2} \leftrightarrow O^{\bullet-}_{2} + H^{\circ}$ Fe ⁰ -H ₂ O ₂ Fenton-like process $\text{Fe}^0 + 2\text{H}^+ \leftrightarrow \text{Fe}^{2+} + \text{H}$, $Fe^{0} + H_{2}O_{2} + H^{+} \leftrightarrow Fe^{2+} + OH^{-} + H_{2}O$ $\text{Fe}^0 + 2\text{Fe}^{3+} \leftrightarrow 3\text{Fe}^{2+}$ $Fe^{3+} + 3OH^{-} \leftrightarrow Fe(OH)$	Decolorization of dyes (azo methyl orange, orange G)		[130, 131]
Electro-Fenton	$M + H_2O \leftrightarrow M(^{\circ}OH) + H^+ + e^-$ $O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$ $Fe^{2+} + H_2O_2 \leftrightarrow Fe^{3+} + OH^- + {}^{\bullet}OH$ $Fe^{3+} + e^{-} \rightarrow Fe^{2+}$	Degradation of bisphenol A (BPA) and COD removal		[132]
Photo-Fenton	$Fe^{2+} + H_2O_2 \leftrightarrow Fe^{3+} + OH^- + {}^{\bullet}OH$ $\text{Fe}(\text{OH})^{2+} + h\nu \rightarrow \text{Fe}^{2+} + {}^{\bullet}\text{OH}$	Pathogens including E. coli, F-specific RNA bacteriophages (FRNA), somatic coliphages (SOMCPH), sulphite- reducing clostridia (SRC)	Water reuse according to different reclaimed water guidelines	[133]

(Continued)

Table 2 Continued

efficiency because the more highly-oxidized species are observed [144]. Microorganisms such as *E. coli* and *Artemia salina* could be removed completely with the current density between 12 and 25 mA/cm² [145]. Photocatalytic process has been utilized widely to degradation of emerging organic contaminants and dyes [146–149].

Combining membrane technology and advanced oxidation process can be utilized to deal with recalcitrant organic pollutants. It has become a feasible strategy. AOPs are usually used in the final stage after the membrane filtration step of wastewater reuse treatments. There are several advantages for this approach, which attract the interest of researchers. First and foremost, the integrated technologies combine the merits of both processes. AOPs, as the post-treatment step, can be applied for volatile low-molecular-weight compounds decomposition and non-biodegradable pollutants destruction, which cannot be removed through membrane technologies or MBR [150]. H_2O_2 /UV had a good performance on the removal of residual organic compounds. So, it was always installed after membrane processes removing the soluble microbial products and extracellular polymeric substances [120]. A combination of microwave and H_2O_2 resulted in effectively reducing amounts of sludge solids from the membrane-enhanced biological phosphorus removal process [151]. MD which has been mentioned before is recognized as a useful technology for desalination. Nowadays, some other applications of MD show that MD offers the possibility of concentrating emerging contaminants and pathogens present in wastewater. For both pathogens *Bacillus* sp. and *Clostridium* sp. spores, the photo-Fenton process did not affect the concentration of spores while the combination of MD and photo-Fenton process achieved a significant reduction [152]. Combination of biological treatment and AOPs had been considered as a method of industrial wastewater decontamination [153,154]. Add membrane technology to the combination, and the MBR-AOP hybrid treatment would be obtained. The proposed process would achieve lower AOP treatment time, more efficient reagent consumption and removal of pollutants and it would be utilized as a polishing step for wastewater reuse [155].

Next, membrane fouling, which affects the performance of membrane processes can be improved through AOPs supplement. Chloramines were introduced to prevent membrane biofouling. In this research, chloramines also played a part in degradation of 1,4-dioxane via the generation of $Cl_2^{\bullet-}$ and \bullet OH) [156]. UV/H₂O₂ treatment combining a brackish water RO desalination system were operated, and RO biofouling caused by indigenous microorganisms was controlled at a low level [157]. After 46 d operation of coupled electron beam radiation and MBR treatment, only 2% membrane flux decreased was observed, which indicated that the membrane fouling was improved by AOPs [158].

In addition, the hybrid systems can reduce the formation of toxic intermediates. The noticeable problem should be considered during AOPs is that complete mineralization may not be obtained for some organic contaminants resulting in generating toxic intermediates. The dosage of O_3 exceeding its demand in wastewater could lead to the formation of bromate [159]. The maximum contaminant level for bromate in drinking water has been set to 10 µg/L both in the United States and in the European Union [160]. Italian regulation on harmful disinfection by-product indicates a maximum concentration for total trihalomethanes in the effluent of 30 mg/L in case of agricultural reuse [161]. Addition, production of halogenated nitrogenous disinfection by-products (N-DBPs), such as N-nitrosodimethylamine (NDMA) and other nitrosamines was also observed [162,163], which was difficult to prevent by controlling operating conditions. And, H_2O_2 process has the same problem as ozone process, formation of organic disinfection by product. Study by Wert et al. [159] showed that O_3/H_2O_2 produced greater concentrations of assimilable organic carbon (5%–52%), aldehydes (31%–47%), and carboxylic acids (12%–43%) compared to O_3 alone which mainly contributed to hydroxyl radical exposure. By using a ceramic membrane contacting, ozone diffusion through membrane pores could bring the uniform addition of ozone. Dissolved ozone concentration was controlled in a low range, which reduced bromate formation successfully. Also, the simultaneous addition of H_2O_2 helped ozone to be converted into •OH more quickly and reduced bromate formation [164].

Finally, the hybrid systems provide better quality of reclaimed water. As we all known, a sequence of processes is available to remove microbiological, inorganic and trace organic contaminants in wastewater reuse treatments [165]. Membrane technologies are acceptable for non-potable water reuse application, such as agricultural irrigation landscaping and road washing [125,166]. While, more and more installations combining membrane technology followed by AOPs are constructed for potable reuse, which meet the drinking water standards. There were several related researches emphasizing the significance of AOPs in hybrid membrane-AOPs treatments for potable reuse [130,167].

Several additional treatment processes are also utilized after the dominate treatment to ensure that residual pollutants are further removed. So that the safety of reclaimed water can be improved. 3.0 g/L methanol addition in the later period of the MBR operation improved the performance of nitrogen removal from 45% to 90%. Methanol could act as an external carbon and replenish the alkalinity required for nitrification [118]. When the reclaimed water from MBR treatment entered the landscape lake, a significant decrease of PPCPs was observed from 85.85% to 39.54%. Adsorption, biodegradation, photolysis, and ecologically mediated processes (via aquatic plants and animals) might have worked for PPCPs removal.

5. Conclusions

Although wastewater reclamation and reuse technologies started late, they have been developing rapidly. Topics involving water reuse and water reuse technologies have at least doubled in the number of articles published in the last 5 y suggesting researchers' growing interests in it [59]. There have been many achievements on treating effluents for reuse which cannot only save a lot of existing fresh water, but also reduce wastewater pollution [17]. This review offers a discussion of the progress associated with the membrane-based systems as well as future prospects. Membrane separation technology is mainly used to remove nutrients and pathogens. The removal efficiency depends on the size of compounds, hydrophilic of both compounds and membrane. Investigations are also concentrated on the combination of membrane filtration and other technologies including pre-treatments, MBRs and AOPs. Advanced oxidation process shows good performance on the removal of recalcitrant organic compounds and inorganic nitrogen, which often follows the membrane filtration to enhance the removal efficiency for potable reuse. In addition, membrane fouling and toxic by-products associated with the operations highlight recent advancements and future needs in reuse technology.

From the view of technological development, new membrane materials and high activity catalytic materials should be vigorously developed. As for AOPs, the depth study of mechanism and various interactions between oxidants and organic matter is significant. The current status mentioned demonstrates that there are potentially more researches still to be completed in this area and more researches are encouraged to get started in order to provide recycled water of expected quality. Except for studies of the treatment process itself, methods and ideas of investigations should also be improved. Firstly, it should be flexible for us to choose the treatment process depending on the source of sewage, the main pollutants, the standard of recycling water quality and local conditions. Secondly, the vast majority of researches were conducted at bench and pilot scale indicating that further investigations can be focused on scaling up the operation for industrial or commercial use. Meanwhile, monitoring the performance of treatment regularly is necessary. Thirdly, public perception and acceptance can be a big challenge. People are concerned about the risk of the recycled water from wastewater. So, a modelling tool should be established to investigate long time environmental

risk and economic benefit to make sure that the reclaimed water can be widely accepted and implemented.

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