Membrane technology for rainwater treatment and reuse: A mini review

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ABSTRACT

Because of the current situation of global water shortage, finding strategies that can effectively guarantee water safety and sustainable use has become an urgent problem that needs to be solved at present. Rainwater is a type of clean energy and the method of the treatment and reuse of rainwater has become a pivotal problem that is worthy of consideration. Membrane technology has become the preferred method in the field of wastewater treatment due to its small footprint, good treatment effect, and low cost, and has also received increasing attention in rainwater treatment field. This review aims to retrospect the existing research technology of rainwater treatment with membrane technology and seek out the most critical research gaps to meet future research needs and technological exploration. The characteristics of different types of membrane technologies in rainwater treatment were summarized, the water quality after treatment and the feasibility in practical applications was analyzed. Membrane fouling has been identified as the main challenge. Nowadays, the research on membrane surface modification and membrane process optimization is gradually deepening, and the exploration and synthesis of new membrane materials and the process of treating rainwater with various technology combinations are still under research. The future application prospects are worth looking forward to.

1. Introduction

It is generally accepted that there is an increasing demand and a tougher criterion for quality and quantity of potable and non-potable water in recent years on account of the current status of global water scarcity, climate change and the growing of population [1]. According to the prediction of International Water Association, under the business-as-usual scenario, the world is projected to face a 40% global water deficit by 2030. This challenge has promoted the search of new methods for the sustainable use of water [2]. Surface water and groundwater have been generally used as a source of drinking or non-drinking water all over the world nowadays. However, the pollution sources of surface water are diverse, difficult and costly to deal with, and the exploitation of groundwater is affected by topographical conditions, and it is easy to cause unfavorable natural phenomena such as swamping, salinization, landslides, and land subsidence during the change process [3,4]. Groundwater may also be contaminated by refractory pollutants such as arsenic on account of human economic activities and planned development in different regions, making it more difficult to degrade and unsuitable for drinking [5–7]. With the development of modern technology of water treatment, rainwater regarded as clean energy has gradually been recognized by many people, and rainwater harvesting (RWH) and reuse systems have gradually been noticed by the public [2, 8]. Proper treatment of rainwater can not only save water sources but also reduce the burden on urban drainage facilities [9]. In recent decades, rainwater harvesting technologies have been supported and implemented in many countries, such as domestic rainwater harvesting (DRWH) and greywater (GW) systems in Ireland, for which rainwater harvested and treated even replace about 94% of public water, significantly lessening demand [10]. Amos et al. have investigated the potential that roof rainwater harvesting systems can supply water to urban agriculture in Australia, demonstrating integrated use of RWH with urban agriculture may promote health, related to social activities, well-being and overall sustainable development [11].

For non-potable or potable water applications, one of the main concerning points for rainwater use is water quality [12]. As a kind of clean energy, rainwater always has low hardness and turbidity below the World Health Organization (WHO) standard for drinking water, with no disinfection by-products and man-made pollutants [13]. But, the composition of rainwater will be changed after contact with the rainwater container or catchment surface. Research in recent years has shown that the pH of rainwater harvested on the roof tank is in the near-neutral range (6.0–8.0) [14,15], and the pH value obtained by the surface runoff rainwater is generally in the neutral and weak alkaline range (7.0–8.5) [16], which both meet the WHO standard. In terms of roof rainwater, it may be contaminated by contact with particulate matter and weathered roofs during the fall [17]. Zhang et al. investigated the effect on the quality of harvested rainwater of conventional roofing materials (concrete, asphalt and ceramic tile roofs) and green roof, suggesting the mean Total Suspended Solid (TSS) from the concrete roof (120 mg/L) was significantly higher than other roofs (< 80 mg/L) owing to flushing [18]. Generally speaking, TSS in rainwater is lower than the drinking water standard because the TSS originated from accumulated materials on the roof rather than rainwater [19].

For chemical parameters, the median value of the NO_3^- concentration in the rainwater tank is usually around 1 mg/L (far less than 50 mg/L), and the NO_2^- concentration is less than 3 mg/L, both of which meet the WHO standard [13,14,20]. However, Mao et al. found that the average NH_4^+ concentration of rainwater samples collected from different roofing materials did not meet Chinese drinking water quality standards, thus roof rainwater could not be directly used as drinking water [15]. In addition, rainwater in roof runoff is usually affected by the excrement of birds and rodents, which will increase the content of nitrogen, phosphorus and pathogenic microorganisms in the rainwater [19]. Lee et al. used the wooden shingle tile roof to assess the quality of harvested rainwater, finding that the concentrations of Total Organic Carbon (TOC), NO_3^- and SO_4^{2-} in the water samples taken from the first flush tank of the wooden shingle tile roof were relatively high, which was due to the weathering of the roof material and the growth of lichen moss [14]. Untreated rainwater from the rainwater collection system on the roof can be detected coliforms while fecal coliform is found in the collected samples of atmospheric rain [21]. According to the WHO standards, coliforms and *Escherichia. Coli* should not be detected in drinking water, so the collected rainwater needs to be further processed. Moreover, heavy metals and other organic ions are likely to be prevalent in rainwater due to human-made pollution in urban areas, and higher concentrations of pesticide residues and fertilizers are likely to occur in rural areas [17]. The quality of surface runoff rainwater is greatly affected by road pavement materials. Motor vehicles can affect the concentration of certain metals on the road surface, and road materials play a key role in the quality of runoff water [16]. In general, the quality of rainwater is relatively good, but it is inevitable to be contaminated by organic matters (such as polycyclic aromatic hydrocarbons), heavy metals, inorganic salt ions (such as NH_4^+), and pathogenic microorganisms (such as *Pseudomonas aeruginosa and Cryptosporidium*) during the landing and harvesting process [22]. Appropriate treatment and disinfection strategies should be adopted to further reduce pollution and facilitate the use of rainwater.

The present research has shown that the rainwater of roofs and roads is poor in biodegradability, hence physical and chemical treatment should be adopted, that the purification process is determined by the quality of the effluent and the purpose of use [7,23]. Currently, several methods have been proposed for rainwater treatment, such as chlorination, pasteurization by solar technology, UV treatment and membrane separation, etc [24]. Collecting rainwater on the roof and using it for non-drinking water after chlorination can reduce consumption and cost, but it is inconvenient for the distributed domestic use, and it can only be used as a means of disinfection like ultraviolet disinfection. They should be used in conjunction with filtration and other technologies to remove particles [25]. Membrane processes are techniques widely known in the field of water treatment, which has a small footprint, high treatment efficiency, and a large amount of permeated water capacity, showing strong advantages in the application of distributed rainwater collection systems [26]. Membrane technology can maximize the treatment of rainwater to reach the water quality reuse standard. In addition, its decentralized and convenient features facilitate household rainwater purification. Types and characteristics of different membrane technologies are summarized in Table 1. Membrane technology has been put into use in some areas with significant effects. For instance, during the Sydney Olympic Games held in 2000, membrane technology was adopted in the rainwater recycling system, utilizing polypropylene hollow fiber microfiltration (MF) membrane as a pretreatment to remove suspended pollutants and pathogens in rainwater and then desalting by RO technique to make salinity of outlet water up-to-standard. After that, the effluent was chlorinated and used to flush toilets. Membrane technology that has excellent processing efficacy, simple apparatus and easy integration in combination with other processing facilities may be hopeful for emergency water supply. It proved that the quality of water treated by membrane technology can reach or even exceed the criterion of domestic or potable water in contrast with traditional methods of treatment, rainwater was fully utilized [27,28]. Peter-Varbanets et al. reviewed the research on decentralized systems used to purify drinking water [26]. Dispersed membrane systems are mainly based on microfiltration (MF), ultrafiltration (UF) or reverse osmosis (RO), and UF-based systems have been put on the market. The large pore size of the MF membrane cannot completely remove viruses, and RO technology requires higher membrane pressure and additional energy to improve membrane efficiency, so it has not been widely put on the market for rainwater treatment research. Membrane technology that has excellent processing efficacy, simple apparatus and easy integration in combination with other processing facilities is suitable for domestic and emergency supplies. Thus, membrane technology has promising application prospects and high research value for water treatment. Porous membranes lose their hydraulic performance as materials accumulate on their surfaces and within their pores, which can be called membrane fouling, the main limitation of the membrane system [29]. Measures just like chemical cleaning, pretreatment, etc. can efficiently mitigate membrane fouling, which will be introduced in detail later.

The author conducted a statistical review on the field of rainwater treatment and membrane technology to highlight and track the research trends completed in recent years, and to predict research trends and future research prospects. The Web of Science database has been harnessed as a scientific platform for research statistics detection and academic papers gathering across the continuum of journals available. The search was specified to involve academic articles and reviews that in effect related to the term “rainwater treatment” and “rainwater treatment and membrane” in Fig. 1. In the meanwhile, the authors reviewed the characteristics of other rainwater treatment technologies and the number of articles published in related fields in the past 20 years in Table 2. Obviously, since 2000, the trend in rainwater treatment and membrane technology has been generally increasing. With the development of polymer chemistry, chemical technology, material engineering and many other fields, membrane technology has a prominent position in the field of water treatment. At the time of writing, there have been 201 journal articles published on Web of Science related to the topic of “rainwater treatment and membrane”, showing that membrane technology is an innovative technology with a widespread range of applications. And there is a continuous and relatively steady growth year after year, foreboding that membrane technology has become a topic of great research interest. It should be noted that only relying on keyword search cannot determine whether the overall object of the article is rainwater reuse or membrane technology in the survey, and there are still certain errors in the data. Several reviews have been published concerning membrane technologies in wastewater treatment [30] and rainwater treatment technologies [24], etc. However, the application of membrane technology in the field of rainwater treatment and reuse has not been comprehensively discussed. Therefore, this review aims to highlight the novel ways in the field of existing membrane technology and the need for the systems, expound the advantages and disadvantages and future research

Table 1
Types and characteristics of different membrane technologies.

Types of membrane technology	Membrane pore diameter	Applicable raw water	Advantages	Disadvantages	Reference
Ultrafiltration	5–100 nm	Fractionation of macromolecular substances; biopharmaceutical; sewage treatment; part process of water purifier.	Low energy consumption and costs; high tolerance of acid, alkali and high temperature; high physical separation capacity.	Difficulties in handling grease; susceptibility to heavy metals.	[30,31]
Microfiltration	0.1–5 μm	Treatment of sewage with a high concentration of suspended particles.	Low hydrostatic pressure; high contaminant rejection and solvent flux.	Low removal rate of organics and pathogens with single microfiltration; need to combine with other water treatment processes.	[32]
Nanofiltration	1–2 nm	Water treatment; pharmaceuticals; food; etc.	High solute retention rate; low energy consumption	Limitation of membrane materials development.	[33]
Reverse osmosis	0.1–0.7 nm	Desalination of seawater and brackish water; preparation of pure water and ultrapure water; industrial/heavy metal/printing and dyeing wastewater treatment.	High removal rate of soluble organic pollutants;	Membrane is susceptible to contamination; need to combine with other water treatment process; high energy cost.	[34]

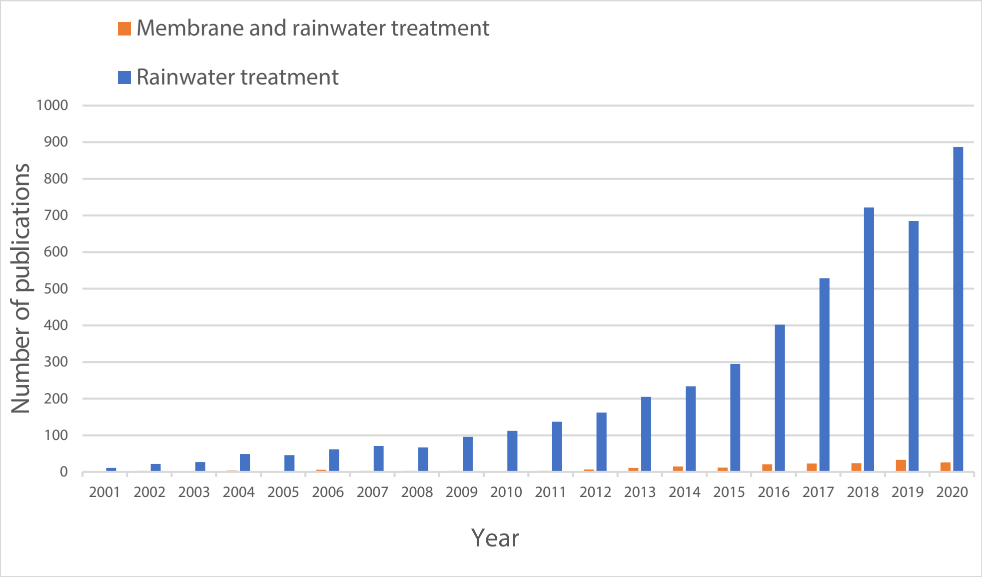

Fig. 1. Numbers of publications every year and cumulative numbers of publications on four terms over the past 20 years.

Table 2
Characteristics and trends of other rainwater treatment technologies.

Types of treatment technologies	Advantages	Disadvantages	References	Number of published articles
Slow sand filtration	Economically feasible and contributing to the protection of water source; microbiologically safe after chlorination.	Fecal coliforms exceed the recommendations; not efficient at reducing turbidity.	[25]	19
Solar disinfection (SODIS)	Simple, green and low-cost; effective in inactivating waterborne pathogens; able to enhance the antimicrobial effectiveness of chlorine.	Low volume of treated water and possible genotoxicity of PET reactors; existing resistant microorganisms; intermittent nature of sunlight availability; turbidity level affects its effectiveness.	[31,32]	21
Solar pasteurization	Free, natural source of energy; low cost and high efficiency; inactivating microorganisms at a temperature of at least 70 °C without radiation.	Persistence of high temperature is affected by many factors; rainfall variables may affect the management of the treatment system; existing resistant microorganisms.	[33]	95

directions, and emphasize technical performance evaluation and social demand. Analyze future research trends by reviewing the effect of membrane treatment and the influence of membrane fouling.

2. Characteristics of efficient membrane technology field

This part presents traditional and novel membrane technologies and their application in rainwater treatment. Characteristics of novel membrane technologies for rainwater treatment are summarized in Table 3.

2.1. *Application of traditional membrane technology*

With the rapid development in the field of materials and chemical engineering, the application field of membrane technology has extended from conventional sewage treatment to special or unconventional

Table 3

Characteristics of novel membrane technologies for rainwater treatment.

Types of membrane technologies	Advantages	Main obstacles	Reference
Membrane surface modification	Good flexibility and larger specific surface area of polymer hollow fiber membranes; high tolerance of higher pressures and temperatures; high removal rate of microorganisms and particles in rainwater.	High costs; short-term stability of materials; anti-fouling properties of membrane.	[31–33]
Gravity-driven membrane (GDM) processes	Backwash-free; High removal rate of turbidity and bacteria.	Low removal rate of low molecular-weight organics.	[34,35]
Bio-Reactor (MBR) process	Stability and good quality of effluent; simplified process; small footprint.	Severe membrane fouling; high aeration operation costs; limited application in rainwater treatment.	[36]

contaminated sewage treatment, unceasing continued to advance at the technical level and played a constructive role in ensuring water security [31–33]. Traditional membrane technologies are familiar to the public including reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF) [23]. The technique category of membrane is defined by the pore diameter. Determine distinct membrane pore diameters according to the types of pollutants, just as MF membranes have pore diameters in the range of 0.1–5 μm, and UF is a proven technology featuring membranes with a pore size in the range of 5–100 nm [34,35]. The appropriate treatment process should be selected according to the permeability of the specific membrane, and remove particles based on the principle of physical separation. These processes present significant advantages to the treatment of drinking water such as smaller footprint requirements, lower operation control requirements and lower demand for chemicals [36]. The filtration performance of membrane separation technology depends on the quality of raw water and the purpose of its application. A practical and simple separation process can meet the required requirements when the effluent quality demand is not high, and on the contrary, when the effluent quality demand is high, advanced wastewater treatment is required.

2.1.1. Microfiltration (MF)

Traditional membrane separation technology has been widely utilized in the field of rainwater collection and treatment. Microfiltration is capable of treating sewage with a high concentration of suspended particles and intercepting most of the suspended solids, protozoa, bacteria, large-scale organic matter, etc., and such membranes require low hydrostatic pressure to obtain high foulants rejection and solvent flux [32, 37]. Single MF can only remove part of large-diameter organics and pathogens, etc., which has some limitations in rainwater reuse application. Thus, MF often needs membrane surface modification or conjunction with other water treatment processes [38]. Dobrowsky et al. constructed a polyvinyl (alcohol) (PVA) nanofiber membrane/activated carbon column MF water treatment system to evaluate the efficiency of MF systems for the treatment of harvested rainwater and surface water [39]. Results indicated that the number of *E. coli*, total coliforms and heterotrophic bacteria in the first 3 L of filter water has been reduced above 99%, meeting the drinking water guidelines of the Department of Water Affairs and Forestry (DWAF, 1996). It would be possible for the combination of MF and coating and electrospinning technology to improve the sanitation and safety of decentralized rainwater treatment. However, the water sample filtered by the membrane system still contains potentially pathogens like the negatively charged adenovirus and potentially pathogenic bacteria, which may cause health risks when used for drinking. Therefore, MF technology adopted in decentralized households as a treatment method for collecting rainwater is not safe enough, and the system needs to be further optimized nonetheless [39]. Nowadays, the direct membrane filtration system using MF for rainwater treatment is still at the laboratory scale, but the development of this field is on the rise [31]. The cost of MF membrane components continues to decrease, and the application prospect will be broader with the rapid development of membrane industry [40]. Future research can focus on the improvement of MF membrane filtration performance and anti-biological pollution performance. At the same time, it can be used as a pretreatment method for microorganisms and macromolecular substances to develop rainwater treatment technology [37].

2.1.2. Ultrafiltration (UF)

Ultrafiltration is a special kind of filtration technology with higher accuracy in comparison with MF. Bacteria, suspended solids and colloids and even harmful substances in water can be effectively removed by UF, which means UF membrane is highly repellent to macromolecular substances. UF technology has been widely used in rainwater management. Oosterom et al. found that rainwater contains low mineral elements and does not require desalination, and the use of MF or UF technology can remove colloids and suspended solids in rainwater, and disinfect at the same time, which is considered to be a new method of producing demineralized water [41]. Compared with the use of RO technology to produce demineralized water, this alternative technology has a lower cleaning frequency, which greatly reduces energy consumption and costs. UF has been used in lab-scale and pilot-scale research and plays a key role in rainwater management. Faragò et al. confirmed the technical feasibility of UF plants (UF-UV and UF-H_2O_2) for pilot-scale rainwater management to replace non-drinking water supplies. UF plants have little impact on the environment while generating added value, which also characterizes the environmental friendliness of membrane technology [42]. Ortega et al. designed a treatment system combining MF and UF membranes for rainwater in the parking lot of the Faculty of Law at the University of Passo Fundo, simulating the worst pollutant load scenario [43]. Both membranes show high removal efficiency for TSS about 100%, MF for biochemical oxygen demand [44] about 74.21% and UF for 65.56%, etc., but low removal efficiency for heavy metals. UF technology can withstand high temperatures that would be used for high-temperature steam disinfection while resisting both acid and alkali, hence owning higher cost performance. Nevertheless, there are deficiencies such as difficulties in handling grease and susceptibility to heavy metals, so technology research strengthening and development are still needed for application in the field of rainwater harvest and reuse. In the meanwhile, unit processes such as coagulation, sedimentation or any other membrane processes are needed introducing as pretreatment steps to overcome the high risk of membrane fouling [30,45]. Following research activities need exceptional efforts for exploiting high-performance UF membranes combining high permeability and maximum selectivity [46]. Furthermore, the UF and additional functions can be strengthened by modifying and coupling [47].

2.1.3. Nanofiltration (NF)

With properties between those of UF and RO membranes, NF membrane has the advantages of high solute retention rate and low energy consumption [48]. Purification of the surface water and underground water employing NF makes it possible to obtain higher purification intensity or even meeting potable water standards [49]. Existing studies have applied NF technology to rainwater treatment due to its efficient processing performance. Kose-Mutlu investigated the methods of NF to treat rainwater stored in a cistern [50]. Three polymeric membranes were used and the average natural organic matter (NOM) removal efficiencies of NP010, NP030, and NF90 were 97.27%, 98.06%, and 99.20%, respectively, and the removal rate of NOM and sulfate even reached over 99% under the most beneficial conditions. The results show that the NF membrane can effectively remove organics and inorganic salt ions in rainwater, which provides us with new evidence of using rainwater to prepare drinking water. Yu et al. explored the feasibility of using Granular Activated Carbon (GAC) filtration, NF membrane filtration and low-dose chlorination to produce rainwater from isolated islands into drinking water. The proportion of conditional pathogens after NF decreased from 23.40% to 7.77%. Moreover, the treated rainwater has a lower potential for disinfection by-products, realizing sustainable drinking water production on a pilot scale [51]. It is worth mentioning that rainwater from isolated islands usually contains fewer types and lower content of pollutants than other rainwater sources due to the simple biological activities and the small impact of human-made pollution. Therefore, for NF technology we still need to further explore to obtain sufficient theoretical support. Nowadays, more and more polymer materials have been utilized to prepare NF membranes. The highly crosslinked and constrained polymer chains resist the improvement of membrane properties, so there is a lot of room for improvement [52]. In recent years, a great process has been made in the development of high-performance membranes based on nanomaterials, and membrane modification will be discussed in detail in the next section.

2.1.4. *Reverse osmosis (RO) and forward osmosis (FO)*

RO technology drives water molecules through a dense RO membrane with pressure to achieve solution separation, effectively removing dissolved salt, colloids, organic matter and microorganisms in the water up to 99.5% [53], which has been highly accounted in the field of rainwater treatment. RO technology has been officially put into production in the field of rainwater treatment and has achieved significant benefits. Taking Singapore as an example, the rainwater collection system takes RO as the core, adopting a highly centralized approach with a treatment train consisting of MF, UF, RO, ultraviolet (UV) and so on to ensure penetrating fluid free of any microbial content all the time. Nowadays, on average 30% of domestic water demand is supplied by four NEWater plants, a number which is expected to rise to 55% by 2060 [54]. Soluble organic pollutants can be effectively removed by RO technology. Pervov and Matveev designed a device that can handle high suspended solids content and maintain a high recovery rate of the membrane unit [55]. Using technology based on RO systems, the feedwater is repeatedly concentrated to remove synthetic surfactants and petroleum products from rainwater, with two times shorter than the time required to remove the same amount by sedimentation. RO technology has high-efficiency treatment effects and provides the possibility for rainwater reuse and re-production of potable water. RO also has nonnegligible shortcomings that inadequate maintenance can easily bring about fouling and corrosion, causing damage to the membrane element, and also affect the effect of sewage treatment [56]. Due to the compact structure and small free volume of the RO membrane, the strong interaction between water and ions and membrane molecules occurs during the rainwater permeation process. Therefore, more energy is required to overcome the osmotic pressure difference generated by the penetration process, which will lead to additional energy costs. At the same time, membrane fouling is the major challenge during RO membrane treatment.

In contrast to RO, forward osmosis [25] is a filtration process driven by osmotic pressure, avoiding using energy drives [57]. Wang et al. evaluated the feasibility of FO in the treatment and reuse of rainwater as the makeup water source technology for the cooling water [27]. The water flux of the FO membrane was about 1.75 L/(m^2·h) at 23 °C, and reduced to about 0.65 L/(m^2·h) when the total dissolved solids (TDS) concentration in the draw solution was diluted 4 times. The flux obtained when the water temperature rose to 50 °C was even 10 times higher than that obtained at 3 °C and no decreased flux was observed, so FO rainwater treatment is a potential technology for cooling water dilution and reuse, which has sufficient flux stability. However, FO technology requires further optimizing modification methods and performance testing to overcome the challenges of reverse solute diffusion, concentration polarization and membrane fouling [58,59]. In the future, the application of RO technology in the field of rainwater will still focus on more reasonable membrane modules and process development and design.

In general, the main disadvantage of these membrane processes is their high energy cost, especially the osmotic pressure change caused by the concentration gradient difference before and after the pressure-driven membrane, which requires additional external pressure to overcome. At the same time, membrane fouling will inevitably affect the flux of the membrane, so pretreatment or backwashing is needed to eliminate it [27,60]. The research in the future will focus on developing new materials of membrane, improving the efficiency of membrane process, reducing energy consumption and mitigating membrane fouling, and this technology field will gain broad space for future development.

2.2. *Membrane fabrication and surface modification*

In the field of water treatment, membrane technology has been recognized as an effective separation process for water treatment, and its effectiveness depends on its pore structure and physical or chemical properties of the surface. The surface properties play a key role in controlling selectivity, flux and anti-fouling performance of membrane and surface modification is mainly used to adjust the surface properties. The processing performance of the membrane is highly dependent on its hydrophilicity, and hydrophobicity is its main disadvantage, causing membrane fouling to often occur on hydrophobic surfaces. Therefore, membrane modification is often based on providing higher hydrophilicity on the membrane surface and more efficient performance. Mamah et al. reviewed the application of polysulfone membrane modification in water treatment that nanomaterial/hydrophilic macromolecule modified PSF membrane was confirmed to improve water treatment capacity, which could promote the development of innovative nanocomposite membranes [44]. Recent research attempts have focused on developing polymer-based nanocomposite membranes for water purification [61], such as carbonaceous materials that have been applied to prepare membranes, and as an additive to polymeric membranes for water purification [62]. However, the large-scale production of cost-effective nanocomposite membranes is still a huge challenge. As a typical membrane separation material, polymer hollow fiber membranes have good flexibility, smaller footprint and larger specific surface area than flat-sheet membranes and inorganic membranes, and are considered to be more effective than flat-sheet membranes. However, the complex structure and form of polymer hollow fiber has certain technical limitations for surface modification. As far as preparation is concerned, the preparation of flat-sheet membranes is more convenient and suitable for situations with ample land. Reinforced hollow fiber membranes are mainly used in UF or MBR processes, which will also have a good application prospect in NF and RO processes [63]. In future research, it is necessary to comprehensively consider the development of low-cost, compatible and long-term stability materials in the membrane matrix, and the need to monitor the actual application of anti-fouling properties, etc. and more research is still needed for the relevant results consolidation in this field.

It has been indicated now that metal membrane filtration is an effective new technology for rainwater clarification [64]. Metal membranes can withstand higher pressures and temperatures, resist outer shock power and chemical oxidation, and has longer service life compared to polymer membranes. Kim et al. have developed a novel technology to treat contaminated rainwater by metal membranes, and ozone was attempted to inject for powerful oxidation and preventing membrane fouling [65]. The experimental results showed that total coliform was reduced above 98% by 0.1um polymeric membrane filter and 1 μm metal membrane filter (MMF) whereas 5 μm MMF resulted in about 78% of removal efficiency. Combinations of ozone treatment for 1min, metal membranes lead to almost complete inactivation of coliforms. And the volume-based particle rejections for 1 μm and 5 μm MMF were 0.95 and 0.80 respectively, strongly proving that metal membranes can effectively reduce microorganisms and particles in rainwater. Beyond that, Kim et al. also used metal membranes to investigate the feasibility of reuse and treatment of greywater and rainwater. The study was completed on a lab-scale and proved that metal membranes could effectively remove particles and the treated rainwater was enough to produce water for non-potable use inside buildings [66]. There has been rare research on metal membranes in recent years, which is probably owing to the high costs of materials and operation, and the relatively limited space for technological development. However, it is still necessary to conduct research in this field in the future to control operating costs, improve rainwater utilization and put it into practice due to its unique characteristic. In addition, ceramic membrane technology has also been widely used in the field of water treatment. Compared with polymer membranes, ceramic membranes have stronger hydrophilicity and lower surface charge, which proves that ceramic membranes have better processing performance and stronger anti-fouling properties [67]. Its resistance to chemical oxidants also means that it is expected to be used in refractory wastewater, such as the field of photocatalytic oxidation. It still has certain limitations in widespread applications due to its high capital cost. As a new resource recycling field, rainwater reuse still needs a period of development. There are very few applications of ceramic membranes in rainwater treatment. Experts still need to study low-cost ceramic materials and membrane fouling mechanisms. In the meanwhile, pilot-scale operation evaluation and technical and economic analysis are required to ensure the long-term effective operation of the technology [68].

In addition to the above technologies, further in-depth research on forwarding osmosis membranes, anion exchange membranes, poly (vinylidene fluoride) membranes, polyether sulfone membranes, etc. are underway in the field of water treatment [69–71]. It's expected for these emerging technologies to be fully utilized in the field of rainwater harvesting and reuse.

2.3. *GDM processes*

In recent years, gravity-driven membrane (GDM) filtration processes are receiving great attention from scholars as a new surface water and wastewater treatment approach [72]. The system often consists of UF and MF membranes, and water flows through the filtration membrane by gravity in the dead-end filtration mode. Compared with conventional membrane separation processes, GDM filtration can obtain a constant permeation volume without needing mitigating membrane fouling by backwashing or chemical cleaning when the hydrostatic pressure reaches about 40–100 mbar [73]. Research shows that the flux stability is attributed to the biological filtering layer formed on the surface of the filter membrane which has a pre-filtering effect that can strengthen the removal of turbidity and microorganisms in the water. Ding et al. have investigated a low-pressure gravity-driven membrane filtration system to simulate the storage of rainwater treatment [74]. Research showed that dissolved organic carbon (DOC) could not be removed by the GDM process while the removal efficiency of bacteria and turbidity was good. Even if affected by the pore size of the UF membrane, the removal rate of bacteria could still reach nearly 95%, and the turbidity of the permeate was much lower than the limitation of the Chinese drinking water quality standards. It is proposed that further research was needed to find new methods of enhancing the removal of low molecular-weight organics and increasing the stable flux value. In order to improve the efficiency of GDM process to remove organic matter, Ding et al. have improved the granular activated carbon (GAC) pretreatment device put forward by Kus et al. set GAC on the surface of membrane combined with the GDM process [75,76]. The system could effectively decrease the content of turbidity, DOC and heavy metal, and most of the organic matter in the rainwater was removed. Due to the adsorption of GAC, the removal efficiency of DOC could reach 37%. Tang et al. compare the performance of fresh GAC/GDM and saturated GAC/GDM. The results showed that the combination of GAC and GDM could effectively remove dissolved organic compounds (~80% in the fresh GAC/GDM and ~40% in the saturated GAC/GDM filtration), and could achieve a higher and more stable permeation flux in the long-term filtration process compared to the traditional GDM control, which proved its feasibility in decentralized and emergency drinking water supply [77]. However, this system also has some drawbacks. For example, Ding et al. found that the flux reduction caused by particle obstruction and adsorption was reduced from 4.5 L/(m^2·h) to 3.2 L/(m^2·h) compared with the control system and the filter cake is denser [75]. Layer resistance and serious membrane fouling can be removed by physical cleaning. Moreover, the current system GAC layer is statically placed on the membrane, which is different from the suspended state in the reported research, so the system still has development prospects and improvement directions. At the same time, no matter how long the backwash lasts, the permeation flux cannot be completely restored by regular backwashing [73]. In the future, researchers should focus on the study of the adsorption mechanism of the GAC layer in the system and membrane fouling cleaning to improve the current status of stable flux reduction and the impact of membrane fouling.

As a new type of membrane material, ceramic membrane has a more stable hydraulic performance. Gravity-driven ceramic membranes (GDCM) systems consist of both gravity-driven devices and excellent quality ceramic membranes and have broad application prospects, which can provide insights for membrane selection and operation optimization of ultra-low pressure driven filtration systems [78]. Chen et al. evaluated the performance of the GDCM microfiltration process for roof rainwater treatment. After treatment, all tested pollutants were reduced below the limit of the Chinese Recycling Water Guidelines (2014), and the average removal rate of DOC even reached 76.9% and The production water flux was stable at 22–45 L/(m^2 ·h), and no backwashing was performed during the experiment, which was an advantage over traditional GDM system [38]. Of course, GDCM technology also has shortcomings that cannot be ignored, such as high material input costs, high technical requirements, and little research in rainwater treatment-related fields. Moreover, there are more research data is demanded to support it because of the differences in the quality of rainwater in current studies. Therefore, the authors infer that the GDM combined with GAC process has more research prospects than the GDCM process.

In the GDM system, the flat membrane has a higher and more stable permeation flux than the hollow fiber membrane, but the hollow fiber membrane can provide higher productivity per unit area, showing a lower potential for filter cake layer contamination. When the space is limited, hollow fiber membrane modules with larger packing density have greater advantages [34].

Furthermore, Du et al. combined a typical RWH system with GDM filtration processes to treat roofing rainwater over 160 days, found GDM filtration could obtain the relatively stable level of permeate flux at ~4.0 L/(m^2·h) and ~2.4 L/(m^2·h) under a setting water head of $\Delta H = 0.4$ m (140 days) and $\Delta H = 0.6$ m (20 days), provided a new method of improving rainwater quality with high feasibility [79]. The resulting biofilm has considerable biological activity. Energy, space and maintenance costs were saved and backwashing was omitted during the process, and this method may be applied to practice after the technology is mature. Regardless of the composition of the rainwater, the GDM system can be easily combined with the RWH system with sustainability and feasibility. Therefore, the GDM process shows more advantages than conventional MF in case of low capacity and is more suitable for decentralized supply. The application potential has been highlighted, and better development is expected in the future [34].

2.4. *Membrane bio-reactor (MBR) process*

Membrane bioreactors (MBR) are usually combined with membrane separation technology and biodegradation and have been actively employed for municipal and industrial wastewater treatment [80]. Compared with the traditional membrane process or activated sludge process, the main problems during MBR process are severe membrane fouling and high aeration operation costs. The MBR process is commonly used in wastewater treatment plants as a centralized treatment technology. On the contrary, the reused rainwater is usually harvested by the roof or courtyard rainwater storage tanks or reservoirs, and rainwater reuse technology is deemed as a decentralized treatment technology to replenish domestic water. Hence, the former is mainly utilized for municipal wastewater treatment [81], while it is still in the experimental stage in terms of rainwater reuse currently.

3. Quality assessment of rainwater after membrane treatment

Rainwater is an economical and high-quality source, and being taken advantage of can effectively alleviate water shortage. The chemical substance content in rainwater is much lower than that of river water or groundwater normally. Nowadays, rainwater captured by roofs has been utilized as potable and non-potable water sources in many countries [82]. Nevertheless, rainwater has not been widely used as a source of drinking water, domestic washing, or irrigation owing to lack of the capacity to assess water quality quantitatively, such as evaluating the content of microorganisms and chemical substances in the water tank [22,83]. Therefore, it's crucial to efficiently evaluate the rainwater quality and examine the frequently detected contaminants in rainwater harvesting systems to ensure the quality of rainwater for future quality guidelines [13]. Pollutants and quality parameters of rainwater are summarized in Table 4.

Table 4
Pollutants and quality parameters of rainwater.

Rainwater source	Catchment materials	Parameters	Reference
In a 30 km radius around the City of Guelph in Ontario, Canada.	1L polypropylene Nalgenew bottles.	pH 5.8 ± 0.9–8.2 ± 0.9 Turbidity 0.9 ± 0.5–2.6 ± 3.1 NTU TN 1.5 ± 0.4–2.0 ± 0.6 mg/L TOC 1.8 ± 1.0–2.0 ± 0.5 mg/L Total coliforms >1 CFU/100 ml.	[12]
A kindergarten and a primary school in Cu khe village in Hanoi, Vietnam.	Sterilised 1L polyethylene bottles.	pH 7.0–8.1, Turbidity 0.1–1.3 NTU, NO2–N 0–1.398 mg/L, N03–N 0.1–8.6 mg/L, NH3–N 0.03–0.86 mg/L, Coliform 10–12000 CFU/100 ml, *E.coli* <3200 CFU/100 ml	[13]
In the Mekong Delta (MD), Vietnam.	Storage basins.	TDS 5.0–113 mg/L, pH 4.3–8.2, Turbidity <10.1 NTU, COD 0.1–23.2 mg/L, Nitrate 0.1–3.9 mg/L, Nitrite 0.004–0.091 mg/L, Total coliforms <102500 CFU/100 ml, *E . coli*<4650 CFU/100 ml	[20]
In Tongji University campus, an urban area of Shanghai, eastern coast of China.	Ceramic tile roofs, pilot-scale.	Turbidity 5.03 ± 1.73 NTU, TOC <22.86 mg/L, TN < 4.23 mg/L, NH4–N <1.85 mg/L, NO3–N <1.56 mg/L, Al<0.01 mg/L, Fe < 0.02 mg/L, Zn No detection, Pb No detection.	[15]
In three governorates (Irbid, Jarash, and Ajloun), located in the northwestern part of Jordan, about 65 km north of the capital Amman.	A bucket placed on the roof and samples collected using polyethylene bottles	Turbidity <50 NTU Hardness <130 mg/L TDS <200 mg/L Decreasing over the time.	[21]
In Dingxi County, Gansu Province, China	Roof-yard system and roads.	Roof yard: Turbidity 2–3.5 NTU, TDS 185.0–750.0 mg/L, COD_{Cr} 8.74–23.83 mg/L, Ions < WHO standard for drinking water Roads: Turbidity 7.5–18.3 NTU, TDS 491.0–1942.0 mg/L, COD_{Cr} 30.18–49.25 mg/L, Al 0.157–1.934 mg/L, Se 0.17–0.21 mg/L,	

According to the water quality standards required for water quality and effluent, the production and preparation of drinking water and household non-potable water usually need to be achieved through multiple treatment processes, such as coagulation, precipitation, filtration, disinfection, etc., and membrane technology is expected to replace the precipitation, filtration and/or disinfection process to simplify the treatment process, improving technical simplicity and treatment efficiency. Nowadays, researches on rainwater usually focus on roof-harvested rainwater and surface runoff rainwater. This part presents the evaluation of rainwater quality in detail and analyzes the quality of rainwater after treatment through membrane technology. The authors will discuss the applicability of membrane technology in rainwater treatment based on the source of rainwater.

3.1. *Physical analysis*

3.1.1. *pH*

As far as roof rainwater is concerned, regardless of the roof material, the average pH value of rainwater collected from all pilot-scale roofs is within the near-neutral range (pH 6.0–9.0), The pH of pure rainwater may be low due to rainwater acidification caused by atmospheric pollution or spoiled plants. Despins et al. found that the pH of rainwater stored in plastic tanks tended to be slightly acidic, with an average pH of 6.5 at all sites and a minimum pH of 4.8 [12]. On the contrary, The average pH of all sites in the concrete rainwater tanks was 7.7, and the maximum pH was 10.2. Under normal circumstances, the natural acidity of rainwater can be neutralized by the alkaline substance from the material of the reservoir, or by adding lime to the plastic container. The pH value of rainwater has no direct effect on drinking and non-drinking water. Therefore, there are few reviews and studies on the pH of membrane technology in the past 20 years. But a low pH value may cause the corrosion of the rainwater collection container, thereby affecting the taste and smell of the effluent after treatment [13].

3.1.2. *Particles*

Pure rainwater is not polluted by environmental factors such as roof materials and atmospheric particles, which is usually clean and has a very low particle content. When rainwater falls on the roof or the ground, the TSS concentration of the collected rainwater rises significantly due to the erosion of the materials and the sediment on the contact surface. Gikas and Tsihrintzis found that the mean TSS concentration of the rainwater samples FS1, FS2, FS3, FS4, and FS5 collected from the roof for the first-flush were respectively 15.2, 6.2, 6.8, 3.0 and 2.4 times of the rainwater samples collected by the rainwater tank [19]. And thus, the rainwater source exerts a tremendous influence on the selection of the subsequent treatment process and reuse purpose. Correspondingly, potable water has higher quality demand. Chen et al. used the GDCM system (0.1 μm) to treat rainwater harvested from a planted roof, suggesting that the GDCM system reduced the turbidity by an average of 92.2%, and reduced the turbidity from an average of 5.86 ± 1.64NTU to 0.46 ± 0.33 NTU, with the effluent of turbidity complied with Chinese Drinking Water Guidelines (2006) [38]. Particles with a diameter more than 0.1 μm are rejected through the MF process, and the turbidity parameters of the effluent have met the drinking water standards, so the cleaner roof rainwater may be able to meet the turbidity purification requirements of domestic water to a certain extent through MF technology.

3.2. Chemical analysis

3.2.1. Total nitrogen (TN) and total phosphorus (TP)

Rainwater is usually clean. Kus et al. have proved that with regards to ammonia, nitrate, nitrite and orthophosphate, all metropolitan and rural rainwater tanks complied with the Australian Guidelines for Water Recycling (2009) (AGWR) limits of 0.5 mg/L, 50 mg/L, 3 mg/L and 1 mg/L respectively [84]. But the growth of moss lichen and animal waste on the roof leading to the deposition of pollutants will cause higher concentrations of nitrate and nitrite when washed by rainwater. Lee et al. found that the mean NO_3^- concentration of rainwater for the first flush on the wooden shingle roof was 3.3 mg/L, which was higher than the average concentration of the other three roofing materials (2.55 mg/L, 1.89 mg/L and 2.8 mg/L for the concrete tile, clay tile and galvanized steel samples, respectively) [14]. This was contributed to the larger pores of the shingle wooden roof, which promoted the growth of moss lichens and led to the growth of microorganisms, thus bringing more total nitrogen content. Phosphorus is the major pollutant in rainwater runoff, typically originating from lawn fertilizers, atmospheric deposition, animal waste, detergents, etc. [70]. Ortega Sandoval et al. used MF and UF technology to treat parking lot runoff rainwater. The average removal rate of TN is 37.01%, UF is 70.83%, and the average removal rate of TP is 64.91%, UF is 98.28%, both relatively high. The removal rate of nitrate and nitrite is very low, only less than 5% [32]. As mentioned above, to achieve the removal of inorganic salt ions in rainwater, NF or even more efficient technology is needed to treat runoff rainwater to potable water standards.

3.2.2. Organics

Generally speaking, the possibility of roof rainwater being exposed to pollutants is very low, so the content of organic matter in rainwater is limited, which even meets the quality standard of potable water. The quality of collected rainwater depends largely on the surrounding environment. The removal rate of organic matter by MF process is lower than 20% [38]. Ding et al. have confirmed that DOC in rainwater is not well removed in the GDM process, and adding a GAC layer can enhance the removal of organic matter in the GDM system during the rainwater cycle [74,75]. Nowadays, the most commonly studied membrane technology in the field of rainwater treatment is the GDM system, so it is necessary to appropriately increase the pretreatment process for the removal of organic matter. Regarding the development of membrane technology to treat rainwater, researchers still need to conduct feasibility analysis based on quality criteria and operating conditions.

3.2.3. Heavy metal

Heavy metals in rainwater are largely derived from human activities, such as the burning of fossil fuels. If surface runoff flows into the receiving water body, it will cause serious ecological risks, so it needs to be effectively treated. The direct leaching of metal components caused by the erosion of the metal plate that collects rainwater is also the reason for the appearance of heavy metals. Lee et al. found that the total aluminum content from the galvanized steel roof was significantly higher than the samples of other roofs in the first flushing tank, and the average concentration of copper in the water samples of the concrete shingle and galvanized steel roof was higher than that of the shingle roof and clay water sample of the tile roof, but neither of them exceeded the recommended level of U.S. Environmental Protection Agency's (USEPA) [14]. The content of heavy metals in water and sediments in urban rainwater tanks is usually relatively high and may exceed the level recommended by the guidelines, and the lead content even reaches 35 times the acceptable level of ADWG (2004). It is known that MF and UF technologies have low removal efficiency for heavy metals, while NF has a high removal rate for inorganic salt ions. Therefore, NF and even higher treatment efficiency rainwater technology can be considered as a heavy metal treatment technology.

3.3. Biological analysis

When rainfall reaches the roof surface, it carries microorganisms and contaminants present in the air, meanwhile absorbs more sedimentary contaminants such as bird droppings heaping up on the roof or man-made fuel emissions, so there are a lot of pathogens, including opportunistic pathogens [85,86]. Opportunistic pathogens have been isolated from drinking water like *Pseudomonas* spp., *Aeromonas* spp., and *legionella spp.*, etc. [87]. Rainwater tanks have been put into use on a large scale and included in government policies in Australia [83]. The quality and microbial content of rainwater tanks have become an increasing concern for people. The *E. coli* content in the water storage tank is much higher than the drinking water standard, and the biofilm on the inner surface of the tank has a huge impact on water quality. Biofilms in the rainwater tanks consisted of a core group of bacteria that are predominantly *Bacillus Spp.* that originate from soils and the environment [88]. Current research has determined that improper design of rainwater tanks will probably increase the risk of sediment and heavy metal binding during rain falling [89]. Kim et al. examined 5 mm and 1 mm metal membrane filters to treat rainwater [65]. The removal efficiency of 5 mm and 1 mm metal membrane filters for coliform bacteria were respectively 78% and >98%, and the retention of particles rates were respectively 80% and 95%, indicating that it is an effective way to improve rainwater quality. Although it is not necessary to eliminate all microorganisms and pollutants in rainwater due to drinking and non-drinking uses of rainwater, its quality should be reduced to a level that avoids human health risk so that rainwater treatment and quality testing methods can be considered effective [24]. In order to ensure the safety of effluent water, adding a disinfection program after membrane technology treatment can ensure the removal of microorganisms in rainwater. The coupling of a membrane system and a disinfection system are commonly used for intensive treatment in Ireland for domestic harvested rainwater, and the obtained water maybe drinkable [10]. Membrane system can remove turbidity while also removing microorganisms in water. The removal effect of organic matter is better, but the removal ability of nitrogen and phosphorus in water is limited. Therefore, various processes are often used in combination.

4. Analysis of viability and operation maintenance

With the advancement of urbanization, the water environment is deteriorating and the ecological environment is destructed, and the problem of urban rainwater accumulation has become increasingly prominent. Till now, the shortage of water resources has caused people to regard rainwater and wastewater as a useable resource instead of as a burden Rainwater reuse is still in a stage of rapid development as a novel research field, especially in water-scarce countries. Rainwater reuse will contribute to strengthening flood resilience and reducing the non-point pollution created by a load of surface pollutants, and at the same time, it weakens the impact of pollutants in rainwater runoff on the ecological environment such as rivers and lakes, revealing remarkable economic, ecological and social effects. However, due to the technical limitation, the true costs and benefits have proven to be difficult to assess. The real implementation of an engineering technology requires comprehensive consideration of the influence of many factors. Here we carry on the brief analysis and discussion.

4.1. Viability

The implementation of rainwater recycling projects conducts an energetic effect on urban water sources supplement and urban water environment improvement. In developing countries, the most affordable infrastructure is needed to meet people's daily needs, and the relationship between technology costs and benefits also needs to be considered. The application of rainwater industry has broad development prospects, which can attract a large number of private capitals, and a new industrial chain may be formed to promote sustainable economic growth and receive more elites to affiliate as time goes on. In the long run, the progress of this field will go a long way towards the development of cities. According to recent researches, economics and lack of funds are indeed the major obstacles to the implementation or expansion of the concept of water recycling, which obstructed the monetization of environmental benefits, and the out of proportion between input costs and the benefits obtained [1]. For now, water resources management in the process of rainwater reuse is often limited by high input and operating costs, and its operational stability and safety are also the focus of consideration. Membrane technology has continuously improved and reduced costs in the development process in recent years. Because of its efficient treatment performance, the water quality can reach the maximum use standard during the operation process, and the safety can be fully guaranteed. Decentralized management systems based on source isolation are often used in developing countries, which are more sustainable than centralized water supply systems in principle, but it is difficult to come into widespread use [90]. Membrane technology, as a decentralized treatment technology, can be convenient for residents to use while maintaining low cost. The innovation and development of rainwater purification equipment promote the large-scale application of this technology. Simultaneously, residents' psychological barriers are also the focus of consideration. Although all microorganisms present in rainwater are not necessary to be eliminated, the presence of pathogens should be reduced to a level that can avoid human health risks [24]. We need to consider the acceptance of residents to membrane technology on rainwater treatment, the use of treated water and the necessity of technology research strengthen, development or publicity. If the concept of rainwater recycling by membrane technology is introduced to residents, it may play a role in the long term, but it may still be restricted by other factors such as drinking water taste preference [9]. In addition, alternative technologies such as desalination are cheaper and easier to be accepted by decision makers in some regions, and then rainwater reuse will be further restricted. Therefore, the reliability and stability of membrane technology in rainwater treatment and reuse still need to be further developed, material costs and operating costs are appropriately reduced, and safety assurance performance evaluation is strengthened. Once the technology is more perfect, it is expected to be popularized and fully utilized.

4.2. *Operation maintenance*

The characteristics of the water market determine the necessity and feasibility of rainwater reuse systems, while economic and technical factors hinder the actual development of rainwater reuse [1]. The regulatory framework and the current applications are still not satisfactory due to the lack of network for economically and effectively water resources allocation and reasonable supervision methods [91]. Garcia-Montoya et al. presented an optimization formulation for synthesizing water networks in residential complexes, incorporating LCA in the design of residential water integration networks, formulated account for two objective functions as total annual cost and freshwater consumption, which solved the water supply problem for a dwelling complex in the city of Morelia in Mexico [92]. Nevertheless, it still has certain geographical limitations and complex implementation process.

With the development of economy and society, membrane technology has been widely used in the field of water treatment, but the low filtration efficiency existing in some membranes and membrane fouling are key issues that restrict the technical field. Compared with the traditional membrane filtration technology, the GDM process has the advantages of avoiding cleaning and maintenance, low energy consumption, decentralized and easy to use, and not requiring technical management, and is expected to be accepted by the public. Research on membrane technologies used in rainwater treatment is still limited, and the technical field needs further development. The future researches of this field focus on the development of membranes with higher permeation flux to reduce the high energy consumption in the process, and the development of fouling-resistant membrane modules to extend their service life. The mechanism of membrane technology is still not thorough enough, and technology, social development, political background and topographical conditions, etc., may have a partial impact on the sustainability of the system. In consequence, continuous exploration and extension are needed to achieve the sustainable development of water resources management.

5. Influence of membrane fouling

Although membrane technology has developed rapidly in the past 20 years and has been gradually applied in the field of rainwater treatment and reuse, membrane fouling has always been a key problem that plagues its development [93]. In membrane-based rainwater treatment, membrane fouling is considered inevitable [65]. The accumulation of pollutants on or inside the membrane matrix can affect membrane flux, reduce or even eliminate the permeability of the membrane, which will affect the subsequent treatment effect. Membrane fouling is classified as reversible fouling and irreversible fouling, according to the method of descaling [29]. Reversible membrane fouling caused by the difference in the concentration of the filter cake layer surface or material can be removed by mechanical removal, backwashing, or chemical cleaning. In the process of direct pressure-driven membrane filtration, the reversibility of fouling can be achieved through physical washing [94]. Nevertheless, membrane fouling in direct membrane filtration operation can probably lead to a decrease of membrane lifetime owing to more frequent physical cleanings [37]. Irreversible membrane fouling cannot be eliminated and will cause the filter membrane permanent damage, which means the membrane must be cleaned or replaced frequently [95].

Biofouling is defined as the bacterial adherence with growth forming a biofilm, and is synergistic with organic fouling [96]. Through direct attack resulting in membrane decomposition or through the formation of a flux inhibiting layer, biofouling causes a membrane performance decline [59]. Therefore, higher operation and maintenance costs are then generated, and the service life of the membrane is shortened, bringing about water quality after treatment is reduced.

To maintain the economic viability of a membrane process, membrane fouling has to be kept to a minimum. Researchers have devised various strategies to prevent or reduce membrane fouling and improve membrane cleaning efficiency for membrane flux recovery [97]. Some scholars use pretreatment of feed water to remove additional pollutants, such as adding coagulants or disinfectants like ozone or chlorine before membrane filtration and removing potential membrane fouling to maintain the stability of membrane performance. However, whether the pretreatment will increase the cost of the membrane filtration system or not requires further research to calculate and the harmful by-product produced by chlorination should be noted. The application of *Bdellovibrio bacterivorous* as a pretreatment to filtration during potable and wastewater treatment could significantly reduce membrane fouling in water treatment plants as the initial microbial load in the water was reduced [24]. In addition, the development of membrane monitoring and cleaning and membrane surface modification has also provided great help in solving the problem of membrane fouling.

MF and UF processes are common membrane technologies for rainwater treatment. Ortega Sandoval et al. used MF and UF hollow fiber membranes to treat rainwater, aeration was used to reduce fouling, achieving high recovery and efficient cleaning by combining with backwashing [43]. In rainwater treatment process, the GDM process as a decentralized water treatment method has received extensive attention from scholars in recent years. Du et al. investigated the performance of GDM for roofing rainwater reuse, the results showed that almost all particles were removed but a small amount of organics and heavy metals were removed by GDM [79].

In the GDM systems, the formation of cake layer is considered to be the main mechanism of membrane fouling. A bio-fouling layer with EPS (polysaccharides and proteins) is formed on the surface of the membrane, and the contents of ATP and EPS determine the permeate flux of the membrane and characterizes the membrane filtration performance [74]. Owing to its special fouling mechanism, the periodic backwash adopted to the traditional pressure-driven membrane filtration process is probably not able to meet its needs. Wu et al. attempted to use short-periodic backwash to recover the membrane permeability of the GDM systems [73]. Research has shown that shorter HRT was considered to be the optimal condition, exactly as HRT was taken for 27 h in this study. The periodic backwash is an effective fouling control strategy in the pressurized UF process, while it is not recommended for GDM systems. Even short-periodic backwash also increases the resistance of the filter cake layer, resulting in worse filtration performance.

The GDM filter with GAC layer added was able to improve the removal efficiency of organic matters that organic matters concentrated on the GAC layer attached to the membrane, as the presence of the GAC layer played a negative role in the membrane flux and deteriorated the development of membrane fouling, leading to lower membrane flux. This reduction in membrane flux can be improved by flushing, and still has an excellent optimization potential [75]. Sabina et al. used PAC adsorption as the pretreatment process of rainwater membrane filtration, investigated the effect of PAC adsorption on SDI and MFI [98]. The MFI values decreased from 1436 s/L^2 to 147 s/L^2, and the SDI values decreased from 5.7 to 3.0 after PAC adsorption, indicating that PAC adsorption as a pretreatment of rainwater membrane filtration could effectively mitigate membrane fouling. Nowadays, methods of membrane fouling mitigation still need to be further studied to achieve strategies optimization and sustainable operation in practice [31].

6. Challenges for future research

As a relatively abundant water resource, rainwater will have great significance if it can be fully utilized to alleviate the current situation of water shortage and improve the urban ecological environment and water environment. However, there are still many challenges of rainwater reuse, such as the seasonal impact of rainfall on the amount of water, the difficulty of rainwater collection and storage, the impact of current membrane technology on rainwater treatment, the psychological barriers of residents to rainwater reuse, etc., and longer-term research and practice are still needed for further optimization. Moreover, the opposition between input costs and actual benefits, the lack of funds, the acceptance of residents to membrane technology are also important obstacles for the application of membrane technology. The main obstacles of membrane technologies in the field of rainwater treatment were summarized in Fig. 2. Modeling is usually used to characterize the treatment effect of the process or to carry out economic or technical feasibility analysis and risk assessment. Nevertheless, in recent years, the research trend of single membrane technology in the field of modeling and simulating is decreasing due to the complexity of numerical models or the failure to apply classical models, even though this topic is the key to improving the level of operation and scientific nature of membrane technology [37,99]. The difficulty of research has caused the development of this field to stagnate, which means researchers need to make greater contributions in the future.

As far as membrane technology is concerned, the main challenge in this field is membrane fouling. The main factors of limiting membrane technology in rainwater treatment are shown in Table 2. In the membrane filtration process, organic matter, inorganic matter, colloids and microorganisms are considered to be the main pollutants that cause membrane fouling. Membrane fouling has a strong impact on filtration flux, desalination rate and membrane properties, which limits the desalination process, increases operating costs and shortens the service life of the membrane. It has been reported that periodical chemical cleaning can effectively reduce irreversible fouling, thereby helping to achieve sustainable operation of membrane filtration [31]. Pretreatment alone is not enough to reduce biofouling. Microorganisms will still adsorb on the membrane surface, accumulate and grow, and the chlorination disinfection process may destroy the membrane, resulting in irreversible damage. In addition, the rainwater characteristics and rainfall intensity will also have a non-negligible impact on the membrane treatment of rainwater. Rainwater characteristics would be influenced by many factors such as human activities, erosion of the materials and the sediment on the contact surface, atmospheric pollution, etc. Therefore, the selection of membrane technology and operating parameters should be adjusted following actual conditions during rainwater treatment. If the rainwater contains more particles and suspended solids, it is more appropriate to choose an MF system while UF and NF systems are more suitable for rainwater with a higher content of macromolecular organic matters. However, the research on the workload of membrane treatment is still insufficient, which will bring obstacles to the practical application of membrane treatment. During the operation of membrane technology, the membrane surface area and transmembrane pressure difference should be improved to adapt to the increase of rainfall intensity. Nowadays, with the continuous improvement of technical level,

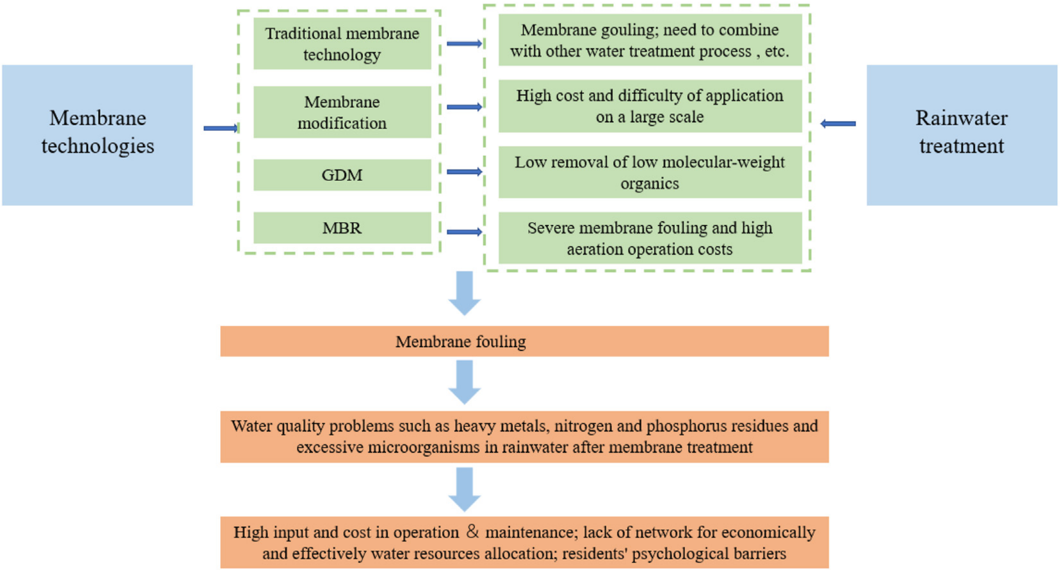

Fig. 2. Main obstacles of membrane technologies in the field of rainwater treatment.

membrane production and operating costs have gradually decreased, performance and stain resistance have also been improved. However, traditional single membrane treatment technology still cannot meet people's needs in some cases. Methods to deal with the challenges of membrane technology in rainwater reuse are as follows:

- Strengthen the research on the modeling process of economic benefit analysis or system feasibility analysis to improve scientific and technical operation level of membrane technology;
- Research and develop new anti-fouling membrane materials or components to extend the service life of membrane components, or strengthen research on pretreatment and chemical cleaning to improve operational stability;
- Optimize the membrane process to improve the efficiency of rainwater treatment and solve the problem that the single membrane technology cannot achieve satisfactory treatment results;
- Develop pressure-driven membranes with higher membrane rejection or permeation flux to improve energy utilization and reduce process energy consumption;
- Strengthen the publicity of membrane technology on rainwater reuse and increase the acceptance of residents.

The research on membrane surface modification is gradually deepening, and the development and utilization of new materials and synthesis processes will open up new methods for the improvement of membrane performance. Future research in this field is expected to produce fruitful discoveries and broad prospects.

7. Conclusion

With the development of rainwater reuse and membrane technology, membrane technology has been gradually applied to rainwater treatment. It is of vital importance to recognize rainwater reuse as a key resource for securing adequate future water supplies and membrane technology will still be in a key position in the future development trend. In this study, we have determined the application status and development direction of membrane technology in rainwater reuse, which is convenient for future technological improvement. Researchers should be fully aware of the need to solve membrane fouling and its high-energy drive requirements. The development of new membrane materials and the improvement of membrane surface properties are still the main research areas in the future. The technical field must be fully considered its cost and benefit analysis, and select the most suitable and reliable technology for further implementation. Researchers should actively explore and exploit applicable membrane treatment components to enhance future treatment efficacy in the field of water treatment.

Declaration of competing interest

The authors declared that they have no conflicts of interest to this work.

We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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