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Review

Bioflocculants in anaerobic membrane bioreactors: A review on membrane fouling mitigation strategies

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ABSTRACT

Anaerobic membrane bioreactors hold immense promise for wastewater treatment owing to their elevated organic load tolerance, cost-effectiveness, and potential for energy recovery. Nevertheless, membrane fouling is a significant impediment to its widespread implementation. This review focuses on utilizing eco-friendly bioflocculants on membrane fouling control in anaerobic membrane bioreactors. Initially, it provides a comprehensive overview of bioflocculants and the mechanisms of mitigating membrane fouling. Subsequently, it examines the performance, mechanisms, and influencing factors of reducing membrane fouling by bioflocculants in AnMBRs. Charge density, molecular weight, bioflocculant dosage, feed water properties, and operational parameters can affect their efficacy. The primary mechanism is helpful reduce the concentrations of Extracellular Polymeric Substances (EPS) and Soluble Microbial Products (SMP) in AnMBRs. Additionally, conventional methods for membrane fouling control are introduced and compared with bioflocculants, emphasizing their notable advantages, such as biodegradability, low toxicity, widespread availability, and potential cost-effectiveness. In conclusion, the review outlines the challenges associated with membrane fouling control in anaerobic membrane bioreactors and elucidates potential directions for future development.

1. Introduction

Membrane bioreactors (MBRs) combine bioreactors with membrane separation, with the advantages of biological treatment and membrane technology [1]. In comparison with aerobic membrane bioreactors (AeMBRs), AnMBRs have better solid–liquid separation, higher organic loading and lower operating cost, which can treat high-strength wastewater such as landfill leachate, industrial distillery wastewater and pharmaceutical wastewater [2–5]. In addition, it has the potential for energy recovery, which can convert organics in sewage into biogas [6]. However, membrane fouling remains the bottleneck of AnMBRs, which limits its wide application [7–10]. The energy required for fouling control accounts for more than half of the energy consumption of AnMBRs [4]. Therefore, it is essential to review feasible measures to mitigate membrane fouling for AnMBRs.

Researchers have carried out many strategies to control membrane fouling, including addition of adsorbents/flocculants, alkaline/acid pretreatment, optimization of operational conditions, and membrane modification [11,12]. Some studies have demonstrated that enhancers can significantly alleviate membrane fouling by modifying the characteristics of sludge mixture and feedwater [12,13]. Additionally, it is much easier and cheaper to introduce enhancers for fouling control [13,14]. Nevertheless, the potential secondary pollution, toxicity, recycling and high dose requirements are some barriers to their application [15,16]. Notably, bioflocculants are economical and green enhancers that are biodegradable and induce lower levels of toxicity

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[15,17]. Koseoglu et al. (2008) applied chitosan and starch to control membrane fouling, and the results showed that both can significantly increase membrane permeability and prevent fouling [18]. Besides, bioflocculants such as algae, fungi and bacteria demonstrated the capability to inhibit fouling [19,20]. For instance, algae can change the properties of the sludge mixture, reducing the concentration of extracellular polymeric substances (EPS) and soluble microbial products (SMP) in MBR while alleviating membrane fouling [19]. Therefore, a comprehensive analysis is needed to identify the potential of bioflocculants to control membrane fouling in AnMBRs and provide a sustainable solution.

This is the first review on eco-friendly bioflocculants in AnMBRs for fouling mitigation. Existing literature covers some reviews of membrane fouling control methods, including different additives and improvement of operational conditions [2,6,12]. Nevertheless, green materials have yet to be discussed. The main objective of this review is to unlock the full potential of green bioflocculants in tackling membrane fouling challenges. Additionally, many explorations are conducted and analyzed in AeMBRs, but more research needs to be studied on the fouling control strategies in AnMBRs [1,2,12,13]. This review focuses on AnMBRs, aiming to provide a feasible approach. Based on this, the application of bioflocculants for controlling membrane fouling in AnMBRs was assessed in terms of their performance, mechanisms, and influencing factors. Furthermore, the advantages of bioflocculants were illustrated in comparison with conventional methods. Finally, the review outlines the challenges of alleviating membrane fouling in AnMBRs using bioflocculants and provides prospective strategies for addressing these challenges.

2. Bioflocculants: Types and characteristics

2.1. Types of bioflocculants

Flocculants are always used for pollutant removal in wastewater, which can be divided into chemical-based flocculants and bio-based flocculants according to their sources [15,21]. Chemical-based flocculants have developed faster than bioflocculants due to their low production costs and high efficiency [6]. However, as they cause equipment corrosion and produce secondary environmental pollution, their application is limited to a certain extent [15]. In addition, chemical-based flocculants have pH sensitivity, toxicity and high dosage requirements [16]. Therefore, bioflocculants have attracted the attention of researchers due to their biodegradability, low toxicity, and potential for cost-effectiveness, which makes them have great potential to become a substitute for chemical flocculants [21–23]. In some articles, researchers only consider flocculants produced by microorganisms as bioflocculants [17]. In this review, bioflocculants include flocculants prepared from all biological sources, such as plants, animals and microorganisms.

Bioflocculants are classified as animal-based, microbial-based and plant-based depending on their sources (Fig.S1) [24]. According to their chemical structure, they can be divided into cationic, anionic and non-ionic bioflocculants [25]. As non-ionic bioflocculants perform poorly in many cases, it is necessary to modify them for enhancing their flocculation performances [16].

2.2. Characteristics of bioflocculants

Bioflocculants can be obtained from natural resources or wastes. Most of them are similar in components (polysaccharides (Ps), proteins (Pr), etc.) and possess some functional groups (e.g., hydroxyl, carboxyl) that allow them to flocculate in wastewater. Most still need to be commercially available, in lab-scale trials, or only used for specific water bodies. Currently, commercial bioflocculants are mainly produced from chitosan (animal-based), moringa (plant-based) and tannins (plant-based) [21].

Bioflocculants have the following advantages (Fig.S2): i) widely available, rich in variety and source; ii) low-toxic, biodegradable, and environmentally friendly; iii) low cost, especially when utilizing waste as feedstock, which can reduce waste generation and utilize local resources [21,26]. Therefore, they are expected to achieve sustainable development from multiple perspectives and become a promising approach in wastewater treatment.

2.2.1. Plant-based bioflocculants

Many studies have focused on plant-based bioflocculants, covering their applications, extraction and modifications [27–29]. As shown in Table 1, they are obtained from plants, readily available, low toxic, biodegradable and highly effective in wastewater treatment [24,28,30]. It was reported that bioflocculants produced from okra could effectively purify actual palm oil mill effluent mainly through particle bridging, and the maximum removal efficiency of turbidity and total suspended solids (TSS) reached 94.97 % and 92.7 %, respectively [31]. Yang et al. (2022) developed and evaluated a tannin-based bioflocculant by co-grafting acrylamide and diallyl dimethyl ammonium chloride. Results showed it could effectively remove algae within the pH range of 3–11, with a maximum removal rate of 98.8 % [32]. They found that increasing the charge density and molecular weights (MW) of tannins improved their flocculation performance, and introducing polymers as side chains through grafting significantly boosted their charge density and MW [32].

In addition, starch has also become one of the sources of bioflocculants. Still, it cannot be directly used as a flocculant in water purification due to its poor solubility, absence of charge characteristics, and low MW [54]. Posada-Velez et al. (2023) modified starch by acetylation, resulting in a rapid reduction of turbidity in the actual wastewater (taken from a canyon in Colombia) from 121.58 NTU to about 2.95 NTU and colour from 269.5Pt/Co to about 73Pt/Co [35]. Liu et al. (2017) prepared a starch-based bioflocculant through co-grafting polymerization of acrylamide and [(2-methacryloyloxyethyl) trimethyl ammonium chloride]. The removal rate of kaolin suspension (200 mg/L) and sodium humate (NaHA) aqueous solution (50 mg/L) could reach nearly 100 % at the optimal dosage [55]. They also found that the flocculation properties enhanced with increasing charge density [55]. Bendoraitiene et al. (2018) modified potato starch and found that the biodegradability decreased when the cationic groups exceeded 0.1 mol/mol_{AGU} and became non-biodegradable when the degree of substitution was higher than 0.54. Similarly, the biodegradability of the starch derivatives was greatly affected when the cross-linking degree was higher than 92.5 % [56]. They found that modified starch's biodegradability decreased with increased modification degree [56].

Table 1 also displays that cellulose has attracted much attention and is widely found in agricultural wastes (e.g., banana peels, sawdust, and corn cobs) [16,21,24,57]. Due to the free-OH groups in the structure of cellulose, it can chelate with metals and some dissolved organic matter in water [16]. Still, its chemical reactivity and water solubility are poor. Introducing desired functional groups into cellulose increases its hydrophilicity and surface polarity, which is a viable way to improve its flocculant properties [58–60]. Li et al. (2018) introduced the cationic ammonium chloride group onto cellulose, which obtained a lower dosage and better pH adaptability than commercial polymeric aluminium chloride [58]. Mohamed Noor et al. (2020) prepared a polyacrylamide magnetic cellulose microwave-assisted, which had better flocculation performance than polyacrylamide and alum [60]. Furthermore, Zhang et al. (2023) formulated the ternary bioflocculants from carboxymethyl cellulose, sodium alginate and itaconic acid, which showed superior flocculation performance and lower operating costs than commercial anionic polyacrylamide (PAM) [59].

In summary, plant-based bioflocculants are available from various sources, and their application in wastewater treatment is developing rapidly. However, most plant-based bioflocculants have weak reactivity that can be improved by chemical modification. This may trigger problems such as secondary pollution, reduced biodegradability, and

Table 1
Plant-based bioflocculants for wastewater treatment.

Origin	Wastewater types	Optimal flocculation efficiency (%)	References
Okra	Palm oil mill effluent	Turbidity: 94.97 % TSS: 92.70 %	[31]
Acacia mearnsii tree	Municipal wastewater	Ammonia: 70 %	[33]
Acacia mearnsii tree	Lake with algal blooms	Algae: 98.8 %	[32]
Acacia mearnsii tree	Synthetic wastewater	Turbidity: 100.00 % Color: 89.9 %	[34]
Potato	Domestic wastewater (85 %) and industrial wastewater (15 %)	Turbidity: 97.27 % Color: 72.91 %	[35]
Corn	Domestic wastewater (85 %) and industrial wastewater (15 %)	Turbidity: 97.57 % Color: 72.28 %	[35]
Cocoyam tuber	Tanning industries wastewater	TSS: 85.50 % Biochemical oxygen demand (BOD): 74.00 % Lead (Pb): 65.00 %	[36]
Cactus	Oil sands process-affected water	Turbidity: 98.00 %	[37]
Alyssum seeds	Bilge water	Turbidity: 96.25 % Chemical oxygen demand (COD): 84.63 % Surfactant: 99.00 %	[38]
Picralima nitida seeds	Landfill leachate	Turbidity: 86.64 % COD: 75.25 % Color: 66.49 %	[39]
Origin	Wastewater types	Optimal Flocculation efficiency (%)	References
Orchis mascula tuber	Bilge water	Turbidity: 92.21 % COD: 90.63 %	[40]
Aloe vera	Palm oil mill effluent	Turbidity: 82.78 % COD: 32.95 % TSS: 83.40 %	[41]
Buttonwood leaves	Synthetic turbid water	Turbidity: 75.50 %	[42]
moringa oleifera seeds	Surface water	Turbidity: 90.00 %	[43]
Moringa oleifera	Olive oil mill wastewater	Turbidity: 83.00 % COD: 57.60 %	[44]
Cactus and banana peels	Turbid water from lake	Turbidity: 87.13 % TSS: 82.15 %	[45]
Grape seeds	Synthetic wastewater	Chromium (VI) ions: 99.97 %	[46]
Pomegranate seeds	Pulp and paper wastewater	Turbidity: 87.00 % COD: 60.00 %	[47]
Dillenia indica seeds	Landfill leachate	Bisphenol A: 60 %	[48]
Ipomoea batatas leaves	Synthetic turbid wastewater	Turbidity: 96.00 %	[49]
Rice husk	Turbid water from river	Turbidity: 95.17 % COD: 74.23 % TSS: 89.82 %	[50]
Moringa seeds	Oily steelworks wastewater	Turbidity: 90.00 %	[51]
Origin	Wastewater types	Optimal Flocculation efficiency (%)	References
cassava peel starch	Institutional wastewater	Turbidity: 60.19 % COD: 30.19 % TSS: 57.79 %	[52]
Acacia dealbata pollen	Winery wastewater	Turbidity: 83.00 % COD: 38.10 % TSS: 85.50 %	[27]
acorn skin	Winery wastewater	Turbidity: 88.80 % COD: 43.50 % TSS: 87.60 %	[27]
acorn flour	Winery wastewater	Turbidity: 87.60 % COD: 32.60 % TSS: 87.10 %	[27]
Platanus acerifólia seeds	Winery wastewater	Turbidity: 88.40 % COD: 40.40 % TSS: 88.40 %	[27]
Tanacetum vulgare seeds	Winery wastewater	Turbidity: 89.00 % COD: 45.60 % TSS: 89.80 %	[27]
Aleppo pine seeds	Synthetic dyes water	Congo red dye: 81.00 %	[53]

complex synthesis processes. Therefore, green plant-based bioflocculants have to be further explored.

2.2.2. Microbial-based bioflocculants

Microbial-based bioflocculants are produced by microorganisms with flocculation ability (such as bacteria, yeast, fungi, algae, and actinomycetes), which are low toxicity, biodegradation and high flocculation efficiency [16]. Moreover, microbial flocculants can adapt to wastewater with a broader pH range than chemical flocculants [61]. According to statistics, about 67 % of research on bioflocculants is microbial-based [15].

Microbial flocculants can be produced by different strains (Gram positive, Gram negative, etc.) and have variable properties accordingly [62]. Table 2 summarizes the chemical composition and flocculation effect in wastewater treatment of microbial flocculants isolated from different strains. As shown in Table 2, Pu et al. (2020) extracted polysaccharide-based microbial flocculants BM2 from *Bacillus megaterium*, with removal rates of 90.23 %, 88.14 %, and 82.64 % for kaolin, Congo red, and Pb^{2+}, respectively [24,63]. Microbial flocculant (MBF-5) produced by the *Klebsiella pneumonia* strain removed *Acanthamoeba cysts* efficiently with optimal conditions. The flocculation rates of kaolin suspension and *Acanthamoeba cysts* were 98 % and 84 %, respectively [17]. The bioflocculant produced by *Azotobacter chroococcum* could effectively flocculate waste coal slurry, approximately 83 % [64]. However, their production was influenced by various factors such as substrate content and type, sources of carbon and nitrogen, C/N ratio, pH, temperature, metal ions, and stirring [65]. The long cultivation process and expensive production costs greatly limit the large-scale application of microbial-based bioflocculants. Therefore, using cheap substrates such as wastewater and agricultural waste is feasible to reduce production costs. Researchers demonstrated that wastes are expected to replace nutrients such as carbon, nitrogen, and inorganic salts to cultivate microbial flocculants because they are rich in organics [16]. Wang et al. (2007) used dairy wastewater as the substrate and extracted bioflocculants from *Klebsiella mobilis*, with a removal rate of 91 % for dispersed Violet HFRL [66]. In another study, *B. agaradhaerens* C9 could convert kitchen wastes into microbial flocculants, effectively treating a pilot-scale (30 L) mineral processing wastewater. When 9 mg/L bioflocculants were added, the removal rate could reach 92.35 % [67]. It was reported that waste reduced production costs by about 60 %, making it an essential resource for large-scale production and application of bioflocculants [68]. However, it has yet to develop fully. In addition, most studies are based on lab-scale using synthetic wastewater, so the practical applications need further systematic study.

2.2.3. Animal-based bioflocculants

Most current studies focus on chitosan with a high positive charge density [16,77]. It was reported that the flocculation efficiency of chitosan on microalgae could be up to 90 %, and the required dosage was one-tenth of aluminium sulphate [78]. Hadiyanto et al. (2022) optimized the operating conditions by response surface methodology and increased the flocculation efficiency of chitosan on microalgae to 96.12 % [79]. However, extraction methods of chitosan for large-scale production mainly involve chemical processes, which need a long time and

Table 2
Characteristics and performance in wastewater treatment of microbial-based bioflocculants.

Origin	Strain	Chemical composition	Wastewater types	Optimal flocculation efficiency (%)	References
Camellia assamica leaves	*Bacillus megaterium* PL8	Ps: 78.50 % Pr: 9.20 % Others: 12.30 %	Synthetic wastewater	Kaolin: 90.23 % Congo red: 88.14 % Pb $^{2+}$: 82.64 %	[63]
Soil	*Bacillus* sp.	Ps: 97.13 % Pr: 1.30 %	Textile dyeing wastewater	Turbidity: 99.60 % COD: 92.54 % TSS: 73.59 % Dye color: 82.78 %	[69]
Ruditapes philippinarum conglutination mud	*Vibrio and Bacillus*	Ps: ~ 100 %	Synthetic wastewater	Kaolin: 98.92 % Methylene blue: 98.78 % Crystal violet: 89.37 % Malachite green: 99.11 %	[70]
Ramie degumming wastewater	*Alcaligenes faecalis.*	Ps: 75.30 % Pr: 20.60 %	Synthetic wastewater	Kaolin: 95.44 % Disperse blue-2BLN: 84.70 %	[71]
Water treatment plant	*Pseudomonas monteilii*	Ps: 3.60 % Pr: 84.00 % Lipid: 0.80 %	Synthetic wastewater	Kaolin: 91.60 % Congo red: 78.90 % Rhodamine-B: 54.50 %	[72]
Water treatment plant	*Paenibacillus xylanilyticus*	Ps: 8.20 % Pr: 85.50 % Lipid: 1.60 %	Synthetic wastewater	Kaolin: 97.80 % Congo red: 97.80 % Rhodamine-B: 95.67 %	[72]
Water treatment plant	*Bacillus pumilus*	Ps: 4.30 % Pr: 83.50 % Lipid: 1.25 %	Synthetic wastewater	Kaolin: 87.5 % Congo red: 90.50 % Rhodamine-B: 77.20 %	[72]
Water treatment plant	*Pseudomonas putida*	Ps: 7.40 % Pr: 82.00 % Lipid: 1.50 %	Synthetic wastewater	Kaolin: 91.80 % Congo red: 68.80 % Rhodamine-B: 88.30 %	[72]
Origin	Strain	Chemical composition	Wastewater types	Optimal flocculation efficiency (%)	References
Brewing industry	yeast	Ps: 29.00 % Pr: 46.00 %	Textile industry wastewater	Color: 72 %	[73]
Municipal sludge	*Bacillus subtilis*	Ps: 77.20 % Pr: 14.80 %	Synthetic wastewater	Kaolin: 95.50 % Methylene blue: 76.3 % Crystal violet: 89.8 %	[74]
Laboratory strain	*Aspergillus oryzae*	Ps: 37.30 % Pr: 62.70 %	Synthetic turbid water	Turbidity: 91.09 %	[75]
Laboratory strain	*Phanerochaete chrysosporium*	Ps: 98.59 % Pr: 1.41 %	Municipal wastewater	COD: 91.80 %	[76]

have limited extraction rates, resulting in high production costs and secondary contamination. Physically assisted extraction methods (e.g., ultrasound, microwave, etc.) are immature now and more complex [21]. Additionally, drawbacks such as low water solubility and limited pH range restrict the broad application of chitosan [21,80]. Many researchers focused on modifying their structure and functional groups to overcome these weaknesses. The novel chitosan-based bioflocculant prepared by Jin et al. (2023) could treat dye wastewater with a broader pH range of 2 ~ 9, which optimal removal rates were 85.76 %~99.4 % [81]. Ma et al. (2023) prepared the magnetic chitosan-based bioflocculant can be even more effective than commercial flocculants in treating real steel rolling oily wastewater, with an oil content of 10.3 ~ 14.6 mg/L and turbidity of 26.1 ~ 37.6 NTU. When the dosage was 6 mL/L, the oil and turbidity removal rates were 95.79 % and 87.85 %, respectively [82].

Additionally, researchers identified that wastes such as eggshells and fish bones also have flocculation properties [83]. Hadiyanto et al. (2021) indicated that the flocculation efficiency of eggshells in harvesting *Chlorella pyrenoidosa*, reached 90.78 % after optimising the process parameters through response surface methodology [83]. Another study suggested that bioflocculants prepared from waste fish bones could also have a flocculation efficiency of 97.65 % on microalgal biomass [84]. Animal-based bioflocculants extracted from waste or by-products had the potential to achieve sustainable development.

3. Mechanisms of membrane fouling mitigation in anaerobic membrane bioreactors

Membrane fouling is mainly caused by the deposition of contaminants on the surface and in the membrane's pores [1]. Generally, EPS and SMP directly contribute to membrane fouling in the AnMBRs[6], and the main components are polysaccharides and proteins. Most can be effectively degraded in the AnMBRs. Still, some can persist in the mixture and form gel/cake layers or block membrane pores, leading to reversible and irreversible membrane fouling [6]. Wang et al. (2022) elaborated the mechanism of membrane fouling in AnMBRs based on temporal, which divided the fouling process into three stages: pore narrowing, pore blocking, and gel/cake layer formation. As shown in Fig.1, the first stage (irreversible) was mainly attributed to medium-sized SMP, soluble EPS and small-sized colloids. Macro-molecular SMP, cellular debris, and colloidal EPS dominated the second stage (irreversible). The gel/cake layer formation stage (partially or

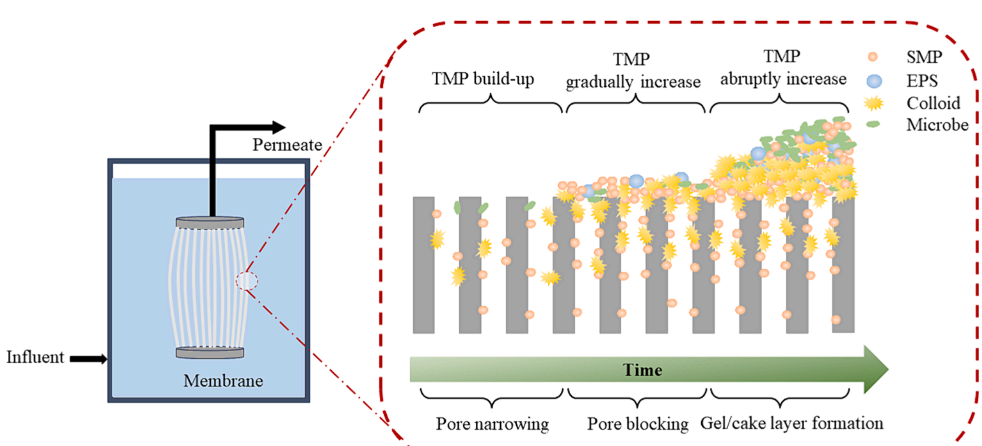

Fig. 1. The configuration of AnMBRs and membrane fouling development.

reversible) was principally caused by EPS, macro-molecular SMP, metal ions, and microbial colloids [85].

The primary strategy to control membrane fouling is reducing the concentration of SMP and EPS, and specific mechanisms include adsorption, mechanical scouring, flocculation, microbial colonization etc. [6,87]. Table 3 summarizes the methods and mechanisms of membrane fouling mitigation in AnMBRs. As shown in Table 3, numerous studies demonstrated that adsorbents such as activated carbon and biochar could effectively mitigate membrane fouling in AnMBRs [6,88,89]. Due to the large specific surface area and high porosity, they reduced the concentration of SMP and EPS by adsorption and are conducive to microbial colonization [85]. It was reported that powdered activated carbon could enrich specialized bacteria (such as *Acinetobacter*, *Comamonas*, *Flavobacterium* and *Pseudomonas*), which were conducive to increasing sludge floc size and reducing EPS production [96].

Similarly, Chen et al. (2023) revealed that biochar effectively alleviated membrane fouling through multiple pathways during a half-year operation [88]. On one side, the scouring effects on the membrane surface can effectively alleviate fouling. On the other hand, biochar enriched many functional microorganisms, which were beneficial for the metabolism of organics [88]. For example, Chen et al. (2020) revealed that adding biochar resulted in the metabolism of EPS-proteins by the Mesotoga genus while inhibiting the growth of the significant biofoulant Arcobacter genus [87]. In addition, Jiang et al. (2022) indicated that polyurethane sponges could work as carriers to alleviate pore clogging by 3.3 %~9 % and reduce filtration resistance by around 52 %. Introducing carriers effectively colonized microorganisms and reduced the sludge's solids, EPS and SMP concentration [90]. As for mechanical scouring, Düppenbecker et al. (2017) showed that fluidized glass beads worked as turbulence enhancers to effectively mitigate membrane fouling in AnMBRs, reducing the fouling rate by over 95 % [8]. The mitigation mechanism mainly included two aspects: (i) particle mixing decreases the concentration polarization, and (ii) some deposited fouling is removed by mechanical scouring.

Table 3
The methods and mechanisms of membrane fouling mitigation in AnMBR.

Wastewater types	Methods	Effect	Mechanisms	References
Synthetic swine wastewater	Biochar	Total fouling resistance (R_t) decreased by 12.33 %	Adsorption Biodegradation	[86]
Pharmaceutical wastewater	Biochar	Trans-membrane pressure (TMP) rising rate decreased by 56.00 %	Adsorption Microbial colonization	[87]
Municipal wastewater	Biochar	TMP decreased by over 90.00 %	Mechanical scouring Microbial colonization	[88]
Synthetic domestic wastewater	Powdered activated carbon (PAC)	TMP rising rate decreased by 53.57 %	Mechanical scouring	[89]
Synthetic domestic wastewater	Granular activated carbon (GAC)	TMP rising rate decreased by 56.43 %	Mechanical scouring	[89]
Municipal wastewater	Fluidized glass beads	TMP rising rate decreased by over 95.00 %	Mechanical scouring Particle mixing	[8]
Food waste streams	Polyurethane sponge	Filtration resistance was reduced by around 52 %	Microbial colonization	[90]
Synthetic wastewater	$FeCl_3$	TMP rising rate decreased by over 90.00 %	Flocculation (charge neutralisation)	[13]
Waste activated sludge	Polymeric aluminium chloride	TMP decreased by 28.57 %	Flocculation (charge neutralisation)	[91]
Wastewater types	Methods	Effect	Mechanisms	References
Waste activated sludge	PAM	Poor effect	Flocculation (absorbing and bridging)	[91]
Municipal wastewater	Vibrating	TMP rising rate decreased by over 89.40 %	Shear enhancement	[92]
Synthetic municipal wastewater	Zero valent iron (ZVI)	TMP rising rate decreased by 20.00 %	Complexation Co-precipitation Physical enmeshment Flocculation (charge neutralisation)	[93]
Synthetic swine wastewater	Nano-zero-valent iron (nZVI)	TMP decreased by 83.00 %	Flocculation	[94]
Landfill leachate	Graphite	TMP decreased by over 95.00 %	Microbial communities changing	[95]

4. Bioflocculants and membrane fouling reduction

4.1. Performance of membrane fouling reduction by bioflocculants

Flocculation is generally used as a pretreatment to mitigate membrane fouling, developed with positive results [21]. Table 4 shows the performance of membrane fouling mitigation by bioflocculants. As illustrated in Table 4, the modified starch-bioflocculant (CGMS) effectively reduced the concentration of macromolecules (MW ≥ 100 kDa) in the MBR supernatant and increase the porosity of the fouling layer. Results showed that the favourable formation of larger-sized flocs made it easy to detach from the membrane surface, thus alleviating membrane fouling [97]. Similarly, Deng et al. (2020) prepared a starch-based bioflocculant (GemFloc™) that significantly mitigated membrane fouling in MBRs, with an increase of *trans*-membrane pressure (TMP) by only 2.5 kPa after 70 days and performed well in natural municipal wastewater [11,98]. Another study found that bioflocculant (Arthrobacter) derived from marine microorganisms exhibited high resistance to membrane fouling in the treatment of simulated saline wastewater (salinity was 0 ~ 30 g/L), with a flocculation efficiency of 80 %. The results demonstrated that Arthrobacter was more likely to capture larger molecules (30 % ~50 %), while smaller molecules (tryptophan) showed a relatively low decrease (20 %) [20]. Studies confirmed that bioflocculants could perform well in mitigating membrane fouling, but most were conducted in the laboratory. The situation in natural wastewater is more complex and variable. Katalog et al. (2018) found that Moringa oleifera significantly mitigated membrane fouling when added to the actual river water but slightly increased the concentration of total organic carbon (TOC) [99]. Besides, most studies are based on AeMBRs systems, and the microbial communities in AnMBRs are more complex

Table 4
The performance of membrane fouling mitigation by bioflocculants.

Origin	Classification	Configuration	Wastewater types	performance	References
Starch	Plant-based	AeMBR	Synthetic municipal wastewater	TMP decreased by over 48.71 %	[97]
Starch	Plant-based	Submerged MBR	Municipal wastewater	R_t decreased by 39.15 % TMP rising rate decreased by 46.85 % Operation time extended by 28 days	[11]
Marine bacteria	Microbial-based	AeMBR	Synthetic saline wastewater	TMP rising rate decreased by 26.09 % Operation time extended by 18 days	[20]
Algae	Microbial-based	Aerobic granular sludge membrane bioreactors	Synthetic domestic wastewater	TMP rising rate decreased by 17.76 % Operation time extended by 27 days	[100]
Algae	Microbial-based	AeMBR	Synthetic municipal wastewater	TMP rising rate decreased by 57.30 %	[101]
Laboratory strain	Microbial-based	Aerobic granular sludge membrane bioreactors	Synthetic municipal wastewater	R_t decreased by 90.3 % Membrane flux increased by 79.4 %	[102]
Algae	Microbial-based	AeMBR	Synthetic wastewater	TMP rising rate decreased by 50.00 %	[19]

and variable. Ensuring the performance of bioflocculants for membrane fouling mitigation in AnMBRs is challenging. Therefore, their effectiveness in natural wastewater and compatibility with AnMBRs should be investigated.

4.2. Mechanisms of membrane fouling reduction by bioflocculants

The commonly accepted flocculation mechanisms include charge neutralisation, sweeping, bridging, and patching. Moreover, adsorption can be regarded as an intermediate mechanism that induces the suspension to attach to the surface of bioflocculants and continue to charge neutralisation or bridging [15]. Theoretically, the mechanisms of bioflocculants remain the same [15]. However, the bioflocculation process is relatively complex and closely related to the properties of bioflocculants and targeted pollutants. In addition, operation conditions such as flocculant dosing, pH, temperature, etc., affect the flocculation performance and mechanism. Thus, the mechanisms must be analysed case-by-case, combined with the AnMBRs, when choosing bioflocculants to control membrane fouling.

Zhang et al. (2022) modified microbial-based bioflocculants to control membrane fouling by aggregating pollutants into larger flocs through charge neutralisation and adsorption bridging. They found that the fouling mechanism changed from intermediate clogging to a loose porous cake layer (from irreversible to reversible) after introducing bioflocculants [102]. Conversely, Sun et al. (2018) found that the floc size in the system without flocculants was more significant than in the bioflocculants-MBR system, but the membrane fouling rate was faster. It was because of the overgrowth of filamentous bacteria, which led to the flocs becoming more difficult to aggregate and thus increasing their size [19]. Deng et al. (2020) showed that the addition of GemFloc™ successfully controlled membrane fouling through the synergistic effect of multiple factors. Firstly, GemFloc™ effectively reduced the concentration of SMP and loosely bound EPS. In addition, it increased the zeta potential and relative hydrophobicity of sludge flocs, facilitating the formation of larger flocs. Finally, further observations of the microbial communities revealed that GemFloc™ successfully enriched the microorganisms favourable for fouling reduction (*Arenimonas* and *Flavihumibacter*) and reduced the microorganisms that may exacerbate fouling (*Sphaerotilus* and *Povalibacter*) [11]. Furthermore, the MBR-*Arthrobacter* system was primarily designed to control membrane fouling through flocculation. Specifically, EPS and SMP were captured by the bioflocculant and enriched on the sludge surface. Partial of them were degraded by the microorganisms in the sludge, thus mitigating the fouling on the membrane. In addition, bioflocculants increased the hydrophobicity of the sludge through charge neutralisation, which enhanced the permeability of the cake layer, thus reducing the fouling rate [20]. However, the situation of another research study was quite different [19]. Results showed that algae addition increased the proteins/polysaccharides (Pr/Ps) value in SMP and decreased the concentration of tightly bound EPS and its Pr/Ps value, weakening the hydrophobicity, which improved the filtration of the flocs and reduced the formation capacity of the filter cake layer, playing a positive role in membrane fouling mitigation. Moreover, inhibiting filamentous bacteria by algae helped improve the stability of flocs in the reactor, thus alleviating membrane fouling [19].

5. Factors influencing bioflocculants performance

The performance of membrane fouling reduction by bioflocculants is not only related to the bioflocculant, but the AnMBRs also play an essential role. As shown in Fig.2, parameters such as flocculant type and dosage, feedwater properties and mixing mode can affect the performance and mechanism of the flocculation process in AnMBRs and impact the mitigation of membrane fouling [65]. As for the AnMBRs, the operating parameters inevitably need to be optimized to achieve the optimal removal rate while obtaining better hydrogen production. This may indirectly affect the bioflocculant performance. Meanwhile, wastewater type and membrane material also impact the fouling degree and the bioflocculation process in the AnMBRs. Therefore, it should be considered comprehensively when applying bioflocculants to alleviate membrane fouling in the AnMBRs.

5.1. Types of bioflocculants

The structural properties of bioflocculants are closely related to their origin, which can dramatically affect their performance. It was reported that removing humic acid by chitosan could reach 90 %, but tends to cause severe membrane fouling. This is because chitosan produces some slight and neutral particles that manage to accumulate on the membrane surface [103]. Yang et al. (2020) prepared chitosan-based flocculants with moderate hydrophobicity, which enhanced the treatment performance in surface water (e.g., the removal rates of turbidity, UV_{254}, norfloxacin and tylosin about 95 %, 90 %, 90 % and 80 %, respectively). Moreover, it can effectively control membrane fouling in the flocculation-ultrafiltration membrane system. The TMP in the system with bioflocculant was lower than 6 kPa after 40 days, whereas the comparison system with aluminium chloride was 14 kPa [104]. It was due to the modified effect of residual bioflocculant on the membrane, which means the hydrophilic chitosan chains formed a hydrophilic layer on the membrane surface with fouling resistance. In addition, studies found that microbial flocculants were rich in functional groups, which could be used for various types of wastewater treatment [96,102,105]. However, they required increased dosage to achieve satisfactory results due to their low charge density and poor flocculation performance [16]. Some monomers are introduced by graft copolymerisation to enhance their charge density, improve their flocculation performance and indirectly control membrane fouling [102]. Therefore, wise selection and optimisation of bioflocculants are essential to obtaining a low

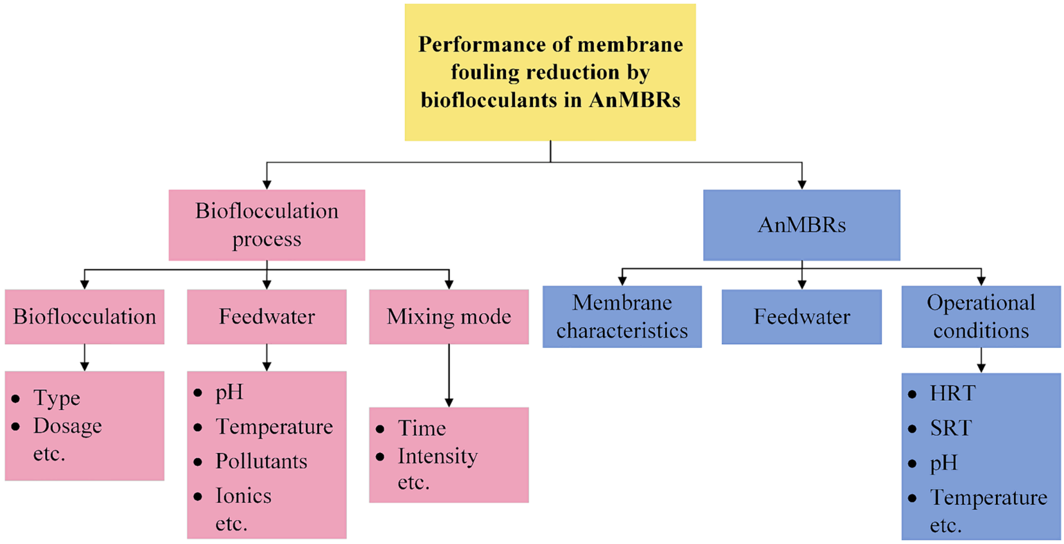

Fig. 2. Factors influencing membrane fouling reduction by bioflocculants in AnMBRs.

contamination tendency in AnMBRs.

5.2. Dosage of bioflocculants

Dosage is a critical factor that directly affects the performance during the flocculation process. It was reported that the optimal dosage for treating aquaculture wastewater (rich in microalgae) by chitosan-based bioflocculants was 30 mg/L. As the dosage increased, the removal efficiency of microalgae gradually increased. When the dosage was 30 mg/L, the removal rate could reach 98.31 %. Subsequently, increasing the dosage had a weak impact on flocculation efficiency and may even restabilise microalgae suspension [106]. Subramaniam et al. (2020) prepared an eggshell-based bioflocculant to achieve the maximum microalgae flocculation efficiency at 60 mg/L.

Similarly, a further increase in dosage would cause a decrease in flocculation efficiency because the exceeding dosage restabilised the system [107]. In addition to microalgae flocculation, Pu et al. (2020) investigated microbial flocculants' performance on kaolin and showed the same tendency [63]. They indicated that the bioflocculant had a poor flocculation effect on kaolin particles due to insufficient action sites at a low dosage. Conversely, excessive dosage increases repulsive between particles, inhibiting the flocculation rate and resulting in stabilisation [63]. Besides, Zhang et al. (2022) investigated the effect of different dosages of modified microbial-based bioflocculants on membrane fouling in aerobic granular sludge membrane bioreactors. Results showed that the pore-clogging pollution (irreversible) was successfully transformed into the cake layer fouling (reversible) when the dosage was 10 mg/L, significantly improving the membrane flux and alleviating the membrane fouling. Pollutants could not be fully flocculated when the dosage was insufficient, but excessive bioflocculants would inhibit their bridging ability and could become a new source of membrane fouling [102]. Currently, limited research is focused on applying bioflocculants for membrane fouling mitigation, and further research is required. As the flocculation process is a synergistic effect of multiple mechanisms, the optimal dosage may differ even if the same bioflocculant is applied in different systems. Therefore, optimising the dosage of bioflocculants is crucial for controlling membrane fouling in AnMBRs.

5.3. Types of wastewater

The different types of wastewater vary in composition, affecting the working conditions, performance, and mechanisms of the bioflocculant process [62]. For example, the microbial flocculant isolated from soil had excellent effects on electroplating wastewater containing large amounts of metal ions. The removal rates of copper (Cu), zinc (Zn), lead (Pb), and cadmium (Cd) were 250 mg/g, 96.7 mg/g, 551.1 mg/g, and 233.3 mg/g, respectively [105]. Tannery wastewater contains heavy metal ions such as chromium (Cr), Pb and Cd [36]. Shende et al. (2023) found that the plant-based bioflocculants prepared from cocoyam exhibited excellent outcomes in removing metal ions from tannery wastewater, and the removal efficiencies of Pb, Cr and nickel (Ni) were 65 %, 60 % and 57.9 %, respectively [36]. Further exploration revealed that the removal of metal ions is mainly achieved through electrostatic interactions between metal ions and functional groups of bioflocculants. Nie et al. (2021) found that the fungal bioflocculant (*aspergillus oryzae*) can promote the flocculation process through the adsorption and bridging effects generated by the surface function groups (such as amino, carboxyl and hydroxyl). Turbidity was removed by over 91 % with the optimal conditions in drinking water treatment [75]. The performance of bioflocculants is directly related to the types of wastewater, so it is necessary to choose an appropriate bioflocculant according to the actual situation.

In addition, the degree of membrane fouling in AnMBRs is highly related to the types of wastewater, thus indirectly influencing the performance of the bioflocculant-AnMBRs. High-strength wastewater generally contains fat, oil, organics or inorganics, the strength of which can be indicated by the biological oxygen demand (BOD_5)/chemical oxygen demand (COD) value [108]. The lower the value, the more slowly biodegradable or contains some non-biodegradable material [108]. For example, the BOD_5/COD values of textile, dyeing, and pharmaceutical wastewater are around 0.1, 0.2, and 0.5, respectively [108,109]. It indicates that they should have physical or chemical treatment before biological treatment. It was confirmed that AnMBRs exhibited excellent performance in high-strength wastewater treatment, such as livestock wastewater, food waste wastewater and leachate [109]. However, the membrane fouling can be accelerated by the high concentration of organics in the feedwater [6,110,111]. Besides, some extreme conditions of feedwater also affect the lifetime of the membrane. Sanguanpak et al. (2015) demonstrated that the degree of membrane fouling (0.91 kPa/d) was severe during the leachate treatment because of low pH [112].

5.4. Membrane characteristics

The membrane is the core component in the AnMBRs. The physicochemical properties (e.g., hydrophobicity, surface charge and roughness) and spatial properties (e.g., pore size and morphology) of membrane materials significantly influence the performance of fouling control [4,113]. Polymeric membranes are widely used due to their cost-effective properties [114]. PVDF membranes are commonly used in the market (57 %), with better chemical resistance and lower fouling tendency than cellulose membranes and polysulfone (PSF) membranes [115]. Similarly, Gao et al. (2010) found that polyetherimide (PEI) membranes were more prone to scaling than PVDF membranes through long-term monitoring of membrane flux in the AnMBRs [116]. Generally, various modifiers are introduced into the matrix or on the membrane surface to improve the antifouling ability [117]. For example, Sethunga et al. (2018) prepared a modified PVDF hollow fibre membrane with perfluorinated polyether (PFPE), effectively mitigating the membrane fouling in the AnMBR. The membrane flux decreased by 7 % after 8 days [118]. Dastbaz et al. (2017) obtained a novel super-hydrophobic PVDF membrane with excellent antifouling ability by dry/wet jet spinning, displaying outstanding performance after 10 days in seawater desalination [119].

In addition, some studies demonstrated that inorganic membranes such as stainless steel and ceramic were more resistant to fouling than polymer membranes [6]. Aslam et al. (2017) applied ceramic membranes in the AnMBRs to treat wastewater with an average COD of 260 mg/L. They found that high membrane fluxes (14.5 ~ 17 L/m^{2}h) could be maintained after long-term operation (395 days) by regular cleaning [14]. Liu et al. (2019) compared the fouling degree of different membrane materials in the AnMBR system. Results showed that the fouling degree of the PVDF membranes was higher than that of ceramic membranes. The pollutants on PVDF membranes were mainly organics and biomass, while ceramic membranes contained many inorganics [120]. However, the expensive preparation cost limits the practical application of inorganic membranes. Therefore, selecting optimal membranes with high filtration efficiency, anti-fouling solid ability, and economics is vital for the bioflocculant-AnMBRs. At present, membrane separation is widely used in various industries, and selecting a suitable membrane dramatically impacts the application performance. The specific selection refers to many factors, such as the membrane's pore size and distribution, the selective layer's thickness, and the membrane material's physicochemical properties [121].

5.5. Operational conditions of anaerobic membrane bioreactors

Operating conditions such as hydraulic retention time (HRT), solid retention time (SRT), temperature, and pH can indirectly affect membrane fouling mitigation by influencing the concentration of EPS and SMP [122]. In addition, temperature and pH can also affect the flocculation performance [62]. Therefore, considering the influence of operating conditions is necessary to achieve optimal performance of bioflocculants in the AnMBRs.

5.5.1. Hydraulic retention time (HRT)

Short HRT may be preferred in practical applications due to the lower operational costs, but being too fast may cause a higher organic loading rate (OLR) and SMP concentration. Several studies demonstrated that the TMP increased when the HRT decreased [123,124]. Kudisi et al. (2022) investigated the influence of HRT in the AnMBR system when treating terephthalic acid-containing wastewater. They found that the TMP significantly increased when the HRT decreased. The TMP rising rates were 0.02, 0.15, and 0.1 kPa/d when HRTs were 48, 36, and 24 h, respectively [124]. Deng et al. (2016) found that short HRT caused the bound-EPS and biopolymer clusters to accumulate on the membrane surface in the sponge-submerged membrane bioreactor, which exacerbated the generation of the cake layer and blockage of membrane pores. The total fouling resistance (R_t) increased from 2.50 × 10^{12} to 4.50 × 10^{12} m^{-1} when the HRT decreased from 6.67 to 4.00 h [125]. It was inferred that increasing the HRT appropriately could mitigate the cake layer formation and prevent membrane pore clogging [125]. Furthermore, Deng et al. (2016) added Gemfloc™ at the optimal HRT (6.67 h) and further mitigated the membrane fouling, with the R_t reducing to 2.07 × 10^{12} m^{-1} [125].

5.5.2. Solid retention time (SRT)

SRT is another critical factor that affects the performance of AnMBRs. Generally, long SRT reduce the production of SMP and EPS and mitigate membrane fouling. However, an excessive SRT will cause high sludge concentration and viscosity, affecting mass transfer and increasing membrane fouling [6]. Huang et al. (2011) supposed that the influence of SRT was related to the HRT. Long HRT would prolong the SRT, which could decrease the microbial metabolic rate and increase the concentration of SMP. As a result, TMP showed an exponential increase. In addition, short HRT would lead to an infinite SRT, which resulted in a higher concentration of mixed liquor-suspended solids (MLSS) and SMP, exacerbating the membrane pore clogging and biofilm formation [123]. Zhang et al. (2022) explored the influence of SRT in the microalgal-bacterial membrane photobioreactor when treating industrial wastewater. Results showed a non-linear relationship between SRT and membrane fouling degree, with the most severe fouling when the SRT was 20 days. The R_t were 13.06 × 10^{12} m^{-1}, 28.19 × 10^{12} m^{-1} and 4.562 × 10^{12} m^{-1} when SRTs were 10, 20 and 30 days, respectively. It was mainly due to the high concentration of SMP and EPS, especially SMP. The growth of microalgae and bacteria was dominated by the competition mechanism when the SRT was 20 days, which caused an unstable environment and promoted SMP and EPS release [126]. Therefore, SRT optimisation is an effective method to control membrane fouling in AnMBRs.

5.5.3. pH value

It was reported that pH could significantly affect the degree of membrane fouling during the operation. Sweity et al. (2011) found that the membrane fouling rate at pH 8.3 was significantly higher than at pH 6.3 when applying the MBR reactor to treat synthetic domestic wastewater. It was because high pH stretched the structure of the EPS into linear, which was more likely to enter the membrane pores and lead to clogging [127]. Another study concluded that the effect of pH was mainly because of the concentration polarisation formed by colloids on the membrane surface. They found that larger colloids (1 ~ 5 μm) were more likely to cause membrane fouling than smaller colloids (100 kDa ~ 0.2 μm). The colloids (0.2 ~ 5 μm) produced at pH 11 caused the TMP to exceed 30 kPa within 100 min [128].

In addition, pH can directly affect the flocculation efficiency. Pandey et al. (2019) reported that eggshell bioflocculants performed better in acidic conditions. The flocculation efficiency for *Chlorella pyrenoidosa* could reach 99 % at the pH 4. They found that the isoelectric point was pH 4, which indicated the surface charge was neutralized entirely. The colloidal system was the most unstable at this point and achieved the maximum flocculation efficiency. When the pH was further reduced to 2, the efficiency could still be maintained at 97 %. However, the flocculation efficiency gradually decreased with increasing pH and fell to 89 % at 10 [129]. Zhang et al. (2023) prepared a ternary bioflocculant, which could remove more than 70 % of crystal violet in the synthetic dye wastewater at pH 4 ~ 10. The overall trend showed that the highest removal efficiency of 88.9 % was at pH 7 [59]. It was because the change of pH could cause an excess of H^+ or OH^- in the solution, resulting in a competitive effect that weakened the flocculation performance. Therefore, the pH value can directly affect the surface charge distribution on the bioflocculants and pollutants, which can influence the flocculation effect directly [62]. Besides, Yang et al. (2022) suggested that the primary flocculation mechanism under acidic conditions might be charge neutralisation, but bridging could dominate more as pH increases [32]. They demonstrated that charge neutralisation played a crucial role in the algal removal of a novel tannin-based bioflocculant at pH 3. When the pH increases to 11, it can achieve a high flocculation effect through bridging because of its high MW [32]. The flocculation mechanism might change at different pHs, and the optimal pH values are various in other flocculation processes. Therefore, exploring the influence of pH in the bioflocculants-AnMBRs is crucial.

5.5.4. Temperature

The temperature in AnMBRs is also an essential influence that cannot be ignored. Gao et al. (2014) found that lower temperature significantly accelerated the fouling process, with a rapid TMP rise when the temperature was 15 ℃ [130]. Similarly, Watanabe et al. (2017) observed the TMP increased from 0.1701 kPa/d to 0.5720 kPa/d with decreasing temperature from 25 °C to 10 °C. This was mainly due to the self-protection mechanism of microorganisms at low temperatures, which means the release of SMP and EPS [131]. On the contrary, high temperature would cause the release of slight particles, which clog membrane pores and exacerbate membrane fouling [115]. Therefore, unsuitable temperatures could exacerbate membrane fouling in AnMBRs, which influences the performance of bioflocculants indirectly.

Moreover, the temperature can influence the performance of the bioflocculants directly. It was noted that higher temperatures could accelerate molecular motion and improve flocculation [30]. Zhang et al. (2023) found that the flocculation efficiency of bioflocculants towards synthetic dye wastewater increased gradually with increasing temperature (10 °C ~ 40 °C), from about 70 % to 94 %. They concluded that higher temperatures enhanced the collision between the bioflocculants and pollutants, improving flocculation performance [59]. However, high temperatures may affect the properties of bioflocculants, altering their spatial structure and inactivating them entirely or partially, thus weakening their performance. A novel microbial flocculant obtained from *Bacillus subtilis* ZHX3 can maintain high flocculation rates at 25 °C ~ 40 °C. The flocculation efficiencies were 85.3 %, 91.2 % and 88.6 % when the temperature was 25, 30, and 40, respectively. It was because the temperature can change the enzyme activities of microorganisms [74]. However, Pu et al. (2020) found that the polysaccharide-based bioflocculant BM2 can flocculate kaolin more than 87 % from 20 ℃ to 100 ℃ due to the negligible impact of temperature on polysaccharides [62,63]. In addition, it was reported that the growth of some microbial flocculants was inevitably affected by temperature [132,133]. For example, the optimal growth temperature for *Aspergillus niger* was 30 °C [132] and for sludge bacterium *Bacillus subtilis* was 40 °C [133].

6. Comparative analysis: Bioflocculants vs. Conventional methods

6.1. Conventional methods

6.1.1. Feedwater pretreatment

Conventional methods for membrane fouling control in AnMBRs include feedwater pretreatment, optimization of operational conditions and membrane modification (Fig.S3). In detail, feedwater pretreatment contains adding adsorbents, flocculants, carriers, etc. The optimization of operating conditions includes HRT, SRT, temperature, and membrane modification, which refers to developing or modifying some new membrane materials. It was demonstrated that adding low-cost and readily available additives to the feedwater as pretreatment is practical and feasible [6,12]. Zhang et al. (2017) observed that powdered activated carbon effectively alleviated pore clogging and irreversible fouling, with the TMP rising rate decreasing from 0.94 to 0.06 kPa/h when the dosage was 400 mg/L [13]. However, Charfi et al. (2017) found that small-sized granular activated carbon (GAC) (0.18–0.5 mm) might deposit on the membrane surface, exacerbating the formation of filter cake layers and negatively affecting membrane fouling control [134]. Compared to activated carbon, biochar can reduce the production cost significantly. The production cost of biochar through pyrolysis is 0.2 ~ 0.5 USD/kg compared to 0.6 ~ 20 USD/kg for activated carbon [135]. Chen et al. (2020) evaluated the potential of biochar for membrane fouling control in the AnMBR during pharmaceutical wastewater treatment. Results showed that biochar could significantly reduce the concentration of SMP and EPS in the reactor (especially proteins), with the TMP rising rate decreasing by 56 % [87]. Another study indicated that biochar mitigated membrane fouling in the AnMBR at a low temperature (10 °C) by adsorping dissolved organic matter (DOM) in sewage [136]. Meanwhile, the scouring effect inhibited the accumulation of large particles on the membrane surface, dramatically increasing the membranes' operating time (200 %) [136]. Chen et al. (2023) added 5 g/L biochar in the AnMBR and found the TMP was reduced by more than 90 % (2.60 kPa) after long-term operation (TMP ≥ 40.00 kPa). They inferred that biochar could control fouling by regulating the metabolism of microorganisms [88]. Therefore, biochar is expected to be an effective alternative to activated carbon. However, the diversity of its inherent properties and the complexity of the AnMBRs make their descaling mechanism not systematic and clear enough, and in-depth discussion is required.

Adding flocculants to alleviate membrane fouling in the AnMBRs is also a current research hotspot. Zhang et al. (2020) investigated the performance of Nano zero-valent iron (nZVI) in the AnMBR during municipal wastewater treatment. Results showed that nZVI increased the floc size and decreased the concentration of the soluble and colloidal organic materials, significantly reducing the TMP rising rate by 20 % [93]. Dong et al. (2015) evaluated the membrane fouling mitigation in the AnMBR system by ferric chloride ($FeCl_3$) in natural municipal wastewater. They found that the TMP was maintained under 12 kPa after 90 days when the dosage was 26 mg/L [137].

Similarly, Zhang et al. (2017) found that the fouling rate could be reduced by more than 90 % when the $FeCl_3$ dosage was 80 mg/L. This was because $FeCl_3$ increased the sludge floc size and colloid particle size, significantly reducing the concentration of colloid and SMP, thus mitigating the fouling layer [13]. Differently, Dong et al. (2015) produced a thicker but porous fouling layer after adding $FeCl_3$ due to the different cross-flow velocities [13]. Due to the wide variety of flocculants, the performance and mechanisms of mitigating membrane fouling in AnMBRs should be assessed case by case.

In addition, some studies reported the descaling effect of other additives. For example, Düppenbecker et al. (2017) found that 1.5 mm glass beads significantly alleviated membrane fouling at lower cross-flow velocities (0.053 ~ 0.073 m/s) [8]. Jiang et al. (2022) mitigated pore clogging by 3.3 % ~9 % by adding polyurethane sponges as carriers, and the filtration resistance was reduced by about 52 %. This was because adding carriers inhibited the retention of microorganisms and decreased the accumulation of EPS and SMP in the membrane pores [90].

6.1.2. Optimization of operational conditions

Optimizing the operating conditions to change the mixing fluid characteristics in AnMBRs indirectly improve the activated sludge properties and thus reduce membrane fouling [4]. Different operational conditions may be required due to the diversity of influent characteristics, system hydraulics, reactor configurations and biomass properties [138]. As mentioned, HRT and SRT are vital factors affecting the degree of membrane fouling. In general, high-concentration wastewater requires a longer HRT since anaerobic digestion needs to maintain an appropriate OLR within the system [139]. For example, when treating wastewater containing synthetic terephthalic acid through the AnMBRs, the system could reach stability at the HRT of 24 h. The average COD removal was 65.8 ± 4.1 %, producing the most minor membrane fouling [124]. The best HRT was 5 days when treating simulated dairy wastewater, with a COD removal rate of over 98 %, and the membrane flux was 9.6 ~ 12.6 L/ (m^2•h) [139]. The HRT should also be ensured for more than 4 h when treating municipal wastewater because too short would increase filtration resistance, accumulate volatile fatty acids (VFAs), and decrease biogas production [138,140]. In addition, factors such as temperature and pH are also related to the degree of membrane fouling [141]. Therefore, adjusting the operating conditions according to the actual situation is crucial for membrane fouling control in the AnMBRs.

In addition, optimizing the reactor design can also prevent membrane fouling in AnMBRs. For example, biogas sparging is an essential operation through cyclic aeration with biogas generated in the reactor, producing a shear effect that can scour the membrane surface [142,143]. Studies have shown that biogas sparging reduced the fouling rate from 1.16 kPa/d to 0.25 kPa/d [141]. However, low gas sparging intensity is insufficient to remove fouling from the membrane. In contrast, high gas sparging intensity can lead to the fragmentation of sludge flocs, producing colloid, polysaccharides and small-sized sludge particles, thus increasing the fouling tendency [138,143]. Ruigómez et al. (2016) proposed a rotating membrane configuration can significantly alleviate membrane fouling (93 ~ 96 %) by enhancing the shear force through membrane rotation, which was considerably better than gas sparging (41 ~ 44 %) [144]. Another study found that installing baffles around the membrane modules could promote turbulence to control fouling, mainly through the enhanced scouring effect on the membrane surface [145]. Gouveia et al. (2015) configured an ultrafiltration unit in AnMBR to treat municipal wastewater at around 18 °C. Results showed that it could be operated continuously for three years under optimal operations (e.g., HRT was 12.8 ~ 14.2 h, continuous biogas sparging (9 ~ 16 m/h)) [146]. Some studies revealed that numerous novel improved module designs, such as anaerobic dynamic membrane bioreactor (AnDMBR), anaerobic electrochemical membrane bioreactor (AnEMBR), anaerobic vibrating membrane bioreactor (AnVMBR), anaerobic osmotic membrane bioreactor (AnOMBR) enabled to elevate the performance and control membrane fouling [4].

6.1.3. Membrane modification

Currently, researchers are focusing on modifying membrane materials and developing new materials in the AnMBRs to mitigate membrane fouling. As most foulants in feed streams are hydrophobic, most organic membranes are highly hydrophobic and prone to fouling. This guides the membrane modification strategies that focus on changing the hydrophobicity and surface roughness [147]. Modifying the membrane will form more hydrophilic surfaces, minimizing the attractive interaction between the membrane surface and the foulants in the feedwater and thus reducing the membrane fouling [147]. To date, reported modification methods involve blending, surface coating, gamma ray, plasma treatment, thermal-induced grafting, etc. [148].

Nevertheless, a specific modification method generally applies to certain particular materials, which cannot be successful between different types of membranes [149]. Therefore, selecting membrane materials and modifying methods is crucial for improving the anti-fouling ability of membranes. Maneewan et al. (2021) changed the PVDF membrane with tannic acid and Cu(II), which effectively reduced irreversible fouling on membranes [150]. Deowan et al. (2016) modified the PES membranes through polymerizable bicontinuous microemulsion technology, effectively overcoming the fouling problem when treating textile wastewater. It demonstrated that the modified membranes had a lower fouling tendency and showed an antimicrobial effect after 105 days [7].

Moreover, Lv et al. (2018) prepared a novel graphene oxide-cellulose nanocrystal composite and modified the membrane to improve the fouling resistance in the AnMBR. The EPS concentration on the modified membrane surface and Rt were reduced by 45.7 % and 46.8 %, respectively [151]. However, the durability of membrane modification needs to be considered in practical applications.

6.2. Comparative analysis

As shown in Fig. 3, conventional strategies for alleviating membrane fouling have some disadvantages compared to bioflocculants, limiting their practical application. As mentioned, additives such as adsorbents, chemical flocculants, and carriers can work effectively as pretreatment to control membrane fouling in AnMBRs. However, most additives involve chemistry and are non-biodegradable, which may exacerbate membrane fouling and even negatively affect human health and the environment [6]. Bioflocculants are biodegradable and low toxicity, generally considered green and safe. They can alleviate membrane fouling without increasing system toxicity, providing a new strategy for replacing traditional chemical additives. Optimizing conditions to mitigate membrane fouling is another effective strategy. However, due to the complexity of wastewater, they are challenging in practical operation [152]. In addition, high energy expenditure, high operating costs, and complicated operations still need to be optimized to obtain an efficient and energy-saving measure [141].

In contrast, bioflocculants are simple to apply and non-energy

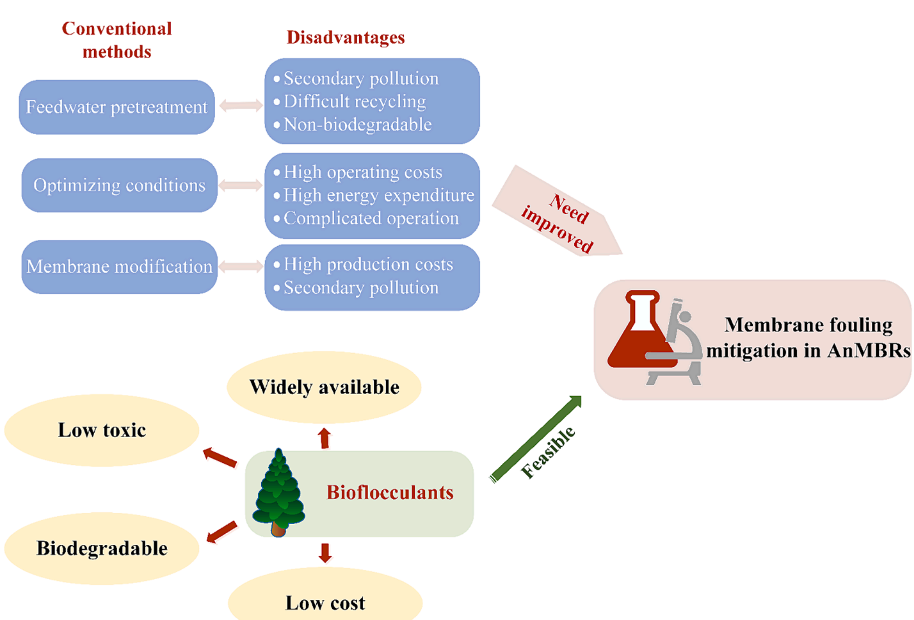

Fig. 3. Comparative between traditional methods and bioflocculants for membrane fouling control in AnMBRs.

supplied, effectively reducing operating costs. Besides, developing and modifying membrane materials require high production costs, making it difficult to promote in practical applications. Bioflocculants can use waste as raw materials, which is expected to reduce production costs and achieve sustainable development.

7. Challenges and future directions

7.1. Identification of current challenges in the application of bioflocculants for membrane fouling reduction in AnMBRs

While the bioflocculant-AnMBRs show promising potential for controlling membrane fouling, numerous challenges need resolution given the current limited research.

1. Most studies addressing the alleviation of membrane fouling by bioflocculants have primarily concentrated on the AeMBR systems. However, AnMBRs are more susceptible to membrane fouling than AeMBRs because of the complex characteristics of anaerobic sludge. Due to the intricate nature of the AnMBRs, it is imperative to ascertain whether bioflocculants can yield similar effects in anaerobic systems. The flocculation mechanism, fouling compositions, and fouling inhibition mechanisms specific to the bioflocculant-AnMBRs remain unclear.
2. Regarding environmental impact, bioflocculants are generally considered low toxicity and safe due to their natural origin. However, unmodified bioflocculants often exhibit suboptimal flocculation properties. Current research has predominantly focused on chemical modifications, introducing potential new pollutants. The toxicity of modified bioflocculants and their impact on organisms remains unknown. Additionally, there needs to be more research concerning the recovery and degradation of bioflocculants. It remains uncertain whether residual bioflocculants are fully biodegradable after use. Moreover, the emphasis has been on enhancing bioflocculant performance, with limited attention to sustainable disposal practices.
3. Regarding the economy, the sources of bioflocculants (plants, animals, and microorganisms) are diverse, resulting in varied extraction and purification methods. However, all these methods entail multiple intricate stages, leading to time consumption, low output, and elevated production costs. Consequently, these factors pose constraints on large-scale application.
4. Regarding practical applications, most research is conducted in laboratories, diverging from real-world industrial conditions. The confirmation of the bioflocculant-AnMBR system's applicability in practical industry remains to be discovered. The diversity and complexity of actual wastewater may impact the stability or compromise the performance of bioflocculants, thereby impeding the achievement of expected goals.

7.2. Potential strategies to overcome these challenges and highlight future research directions

In addressing the current challenges, several strategies and potential avenues for future research are proposed:

i) Conduct comprehensive research on bioflocculants within the context of AnMBRs from multiple perspectives. Investigate and consolidate the mechanisms and influencing factors of the bioflocculant-AnMBRs to establish a robust theoretical foundation for developing and optimizing novel bioflocculants.
ii) Given the multifaceted nature of the flocculation process, optimizing operational parameters is imperative to achieve optimal performance in mitigating membrane fouling while minimizing dosage requirements.
iii) Minimize chemical modification whenever possible. Explore the feasibility of synergistically controlling membrane fouling in AnMBRs by combining operational optimization, membrane modification, and reactor configuration improvements with bioflocculants. This may involve selecting optimal membrane modules and configuring reactors before the operation, followed by adjusting operating conditions post-bioflocculant addition.
iv) Before practical application, rigorously assess the toxicity, stability, and biodegradability of bioflocculants (both modified and unmodified) to confirm their safety and environmental friendliness. Additionally, it emphasizes the recycling of industrial and agricultural wastes for bioflocculant production, contributing significantly to sustainable development.
v) Shift the research focus from laboratory-scale experiments to large-scale commercial production. Develop novel extraction methods and utilize cost-effective raw materials to reduce production expenses, aiming for large-scale commercial bioflocculant production at the lowest possible cost.
vi) Undertake an in-depth analysis of actual wastewater to enhance the system's applicability. This approach facilitates a more comprehensive understanding of real-world scenarios, enabling timely and appropriate adjustments for optimal system performance. In addition, further study on the modelling and simulation of bioflocculant-AnMBRs is essential for optimizing fouling alleviation, including their performance, operational parameters, influence factors, etc.

8. Conclusion

This review explores the efficacy and mechanisms of membrane fouling mitigation by bioflocculants in Anaerobic Membrane Bioreactors (AnMBRs). The key conclusions are as follows:

1. Compared to conventional methods, bioflocculants demonstrate biodegradability, low toxicity, widespread availability, and potential cost-effectiveness. These attributes position them as a promising strategy for alleviating membrane fouling.
2. Bioflocculants-AnMBRs are crucial in mitigating membrane fouling by reducing the concentrations of Extracellular Polymeric Substances (EPS) and Soluble Microbial Products (SMP) in the AnMBRs. Several factors influence their efficacy, including charge density, molecular weight (MW), bioflocculant dosage, feedwater properties, and operational parameters.
3. Future research endeavors should prioritize AnMBRs-specific investigations, optimization of operational parameters, integration with complementary methods, assessment of toxicity, stability, and biodegradability during utilization, utilization of waste-derived bioflocculants, development of innovative extraction methods, and exploration of real wastewater applications. This comprehensive approach will advance the field and unlock the full potential of bioflocculants in tackling membrane fouling challenges.

CRediT authorship contribution statement

Yuanying Yang: Writing – original draft, Resources, Methodology, Formal analysis. **Wenshan Guo:** Writing – review & editing, Supervision, Formal analysis. **Huu Hao Ngo:** Writing – review & editing, Supervision, Project administration, Conceptualization. **Xinbo Zhang:** Resources, Methodology, Formal analysis. **Shuang Liang:** Writing – review & editing, Resources. **Lijuan Deng:** Resources, Methodology, Formal analysis. **Dongle Cheng:** Resources, Methodology, Data curation. **Huiying Zhang:** Writing – review & editing, Resources, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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