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KAJIAN KETERBARUAN PENGARUH SINTESIS ADITIF TERHADAP KINERJA MEMBRAN POLIMER UNTUK PENGOLAHAN LIMBAH CAIR

A-STATE-OF-ART REVIEW ON ADDITIVES FUNCTION ON POLYMERIC MEMBRANE PERFORMANCE FOR WASTEWATER TREATMENT

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Abstrak

Artikel ini secara komprehensif mengkaji pengaruh penambahan zat aditif terhadap kinerja membran polimer dalam mengolah limbah cair. Pemanfaatan zat aditif yang disintesis dan diikutsertakan pada proses pembuatan membran merupakan metode yang dapat diandalkan untuk meningkatkan performa membran dalam mengatasi fenomena fouling dan scaling, terutama saat membran tersebut akan digunakan untuk mengolah limbah cair industri yang mengandung polutan dengan konsentrasi tinggi. Selain itu, pengaruh teknik modifikasi pada penambahan zat aditif di larutan polimer dasar dan beberapa aspek yang relevan seperti mekanisme fouling dan teknologi pengolahan limbah lainnya turut dikaji secara berkesinambungan. Meskipun saat ini telah ada beberapa literatur yang mendiskusikan strategi untuk meningkatkan kinerja membran, namun kajian khusus terhadap manfaat sintesis aditif dengan menggunakan metode dan komposisi sintesis terbaru, khususnya pada membran berbahan dasar polimer masih sangat terbatas. Dengan demikian, diperlukan kajian pembaharuan yang dapat menjadi fondasi untuk pengembangan penelitian selanjutnya terkait tema tersebut. Berdasarkan kajian literatur, dapat disimpulkan bahwa kesesuaian zat aditif yang digunakan berikut komposisi dan metode yang diterapkan serta kondisi operasi selama proses fabrikasi membran, memiliki pengaruh yang besar terhadap peningkatan kinerja membran, khususnya yang berbahan dasar polimer, terutama terkait sifat anti-fouling dan hidrofilisitas membran.

Keywords: Membran polimer, Polutan, Fouling, Hidrofilik, Limbah cair

Abstract

In this article, the recent development of polymeric membrane fabrication using additive for wastewater treatment is presented. The application of this substance has been recognized reliable to increase membrane performance against fouling phenomenon, especially for purifying industrial wastewaters that mostly have high loading of hazardous pollutants. The effects of modification techniques through additives addition on membrane casting solution are considerably included. This paper also discusses membrane fouling mechanism and other existing technologies available for treating contaminated water. Despite the existence of review paper discussing membrane fouling mitigation on literature, there is still the need of comprehensive review related to the novel technology regarding additive blending on membrane, especially on polymer-based membrane for water pollution control. Eventually, clear conclusion can be drawn that the suitability of additive substances and its composition as well as suitable operating conditions have great leverage on polymeric membrane performance regarding its anti-fouling and hydrophilic level.

Keywords: Polymeric membrane, Pollutant, Fouling, Hydrophilicity, Wastewater.

INTRODUCTION

One of the most influential issues of industrialization is the decrease in the availability of clean water for both human needs and other living creatures. (Antonopoulou et al., 2021; Barambu et al., 2020). Industrial manufacturing processes could generate a huge amount of harmful wastewater contaminating open water and soil should there be less or no mitigation effort conducted in the first place. (Martini et al., 2021; Mishra, S. et al., 2021). Contaminants contained in industrial wastewater such as petroleum hydrocarbons, phenols, and poly aromatic hydrocarbons can stall the growth in animals and plants. For human, they would risk some mutations and cancers due to carcinogenic and non-biodegradable characters in nature. Consequently, important mitigation step to prevent further impacts must be properly conducted (Martini et al., 2020).

There are several preferred methods for treating contaminated water including advanced oxidation processes (AOPs), coagulation and flocculation, adsorption, biological activated sludge, and membrane technology. AOPs, for example, have proved their ability to mineralize manifolds organic pollutant types by exploiting produced hydroxyl radical responsible for removing those organic contaminants. However, most AOPs processes also have several concerning issues related to the cost of chemical, sludge and other by-products formation (Garrido-Cardenas et al., 2020).

Furthermore, biological techniques involving particular microorganisms for degrading pollutants concentration need longer reaction time along with bigger wastewater treatment ponds area despite being environmentally friendly way (Xu et al., 2021).

Adsorption mode can also be implemented for treating industrial effluents (Afroze and Sen, 2018; Martini et al., 2020). This technique can catch pollutants to be trapped on the adsorbent pores (Medhat et al., 2021). Adsorption is simple and reliable with less energy consumption (Bilal et al., 2013). However, like other types, this method also suffers from some challenging issues such as limited separation efficiency and the generation of secondary pollutants (Agarwal et al., 2020).

Therefore, the usage of membrane technology has been the preferred mode. Compared to other treatment methods, membranes filtration would result in both great pollutants removal and less by-product generation (Liu et al., 2020; Yang et al., 2020). Membrane separation offers several beneficial points such as higher purity level of treated water due to its great rejection to undesirable pollutant particles based on either pore size or molecular weight (Barambu et al., 2020). As its usage to remove many variants of pollutants has been proven reliable, there are more and more industries utilizing this technology as the main part of their wastewater treatment plant (Liu et al., 2020; Yang et al., 2020). However, the advantageous feature of membrane usage still has challenging issue named membrane fouling that needs high maintenance cost regarding periodic membrane cleaning and changing materials. This review then would focus on the recent development of polymeric membrane regarding the breakthrough of additive synthesis or additive blending and its effect on fouling mitigation and membrane performance as a whole. Other than that, other strongly related aspects such as fouling mechanisms and prominent polymer-based membrane materials would also be critically discussed.

METHODOLOGY

This review was written by selecting relevant issues to recent development of polymer based-membrane regarding the usage of additive substances. Firstly, it is started by introduction part discussing the effect of industrial wastewater disposal on both human and environment, followed by various available wastewater treatment technologies. The second part covers methodology explaining the structure of the paper in relation to the topics of each part. The next section then discloses fouling phenomenon as the main issue in membrane application. In this section, fouling definition and its mechanisms are critically reviewed. The fourth section resumes polymeric membrane materials and relevant recent works related to the membrane fabrication. Further section then focuses on the comparative discussion of the latest studies investigating the usage of newly improved additive on the fabricated membrane as well as their effect on fouling mitigation and membrane performance. Ultimately, conclusion section could be written by pointing important clues of each sections aforementioned.

FOULING PHENOMENON

Complex fouling mechanism is influenced by various parameters connecting to each other like membrane and feed properties (Alkhatib et al., 2021; Dong et al., 2021; Martini and Yuliwati, 2020). The roughness of the surface could impact fouling generation regarding hydrophobic pattern. A higher roughness would lead to hydrophobic tendency as higher trapped air particles would be able to stay in the pores causing faster clogging (Choudhury et al., 2019; Han et al., 2017). The fouling layer on the surface then would cause flux decline. The severity of fouling is mainly affected by membrane surface property, controlling the surface could then be a priority. This may be overcome by surface modification through additive addition blended when fabricating membrane for better energy consumption, surface wettability, and surface texture (He et al., 2021; Maan et al., 2020).

Membrane pore size influences the selection process of membrane type for deciding the most suitable membrane to eliminate particular wastewater. Related to this issue, membrane can be divided into four categories namely microfiltration, ultrafiltration, nanofiltration, and reverse osmosis, while its material can also be categorized into polymeric, metallic, liquid, ceramic, and ion-exchange (Samaei et al., 2018). Among these materials, polymeric and ceramic membranes are mostly used in both industries and research exploration.

Fouling phenomenon is caused by various reasons including concentration polarization, pore blocking, solute adsorption, and gel-layer formation that plugged membrane pores. To understand how fouling could happen on membrane, Hermia's models consisting of four types of mechanisms namely complete blocking, standard blocking, intermediate blocking, and cake filtration models then can be employed (Khan et al., 2020; Martini and Ang, 2019). The package of fouling models developed by Hermia enables good understanding of fouling mechanisms occurred on the membrane surface and/or inside the pores during filtration. Standard blocking happens when the pollutant molecules have a smaller size than the pores, so they can easily enter in the pores, then attaching to the membrane walls leading to diminishing the internal diameter of such pores, while intermediate blocking would come when pollutants have relatively the same size molecules as membrane pores, so they would be able to seal the pore accumulating on each other. When these pollutants form a monolayer, it is called complete blocking type, while cake formation would happen if the solute cannot pass through the membrane pores due to the existence of former pollutants with bigger size clogging the pores and forming cake-like layer formation on the membrane surface (Jepsen et al., 2018; Martini and Ang, 2019; Martini and Yuliwati, 2020). On the other end of the spectrum, the mechanism of membrane fouling could be predicted using Hermia's models by measuring the value of coefficient correlation of each model calculated based on the experimental data.

POLYMERIC MEMBRANE MATERIAL

Some significant improvement in membrane technology like better module design along with more cost-effective type has resulted in the increasing demand of membrane application for water pollution control (Al Aani et al., 2020). Pressure-driven membrane is useful for treating contaminated water and wastewater, while effective pollutant rejection can also be achieved via size selection, adsorptive force and electrostatic repulsion (Asif et al., 2019).

Polymeric membranes have been popular as they have relatively small footprints, flexible operating condition, lower cost, superior organic solvent resistance, and ease of operating (Barambu et al., 2020). There are a variety of synthetic polymers available such as polyvinyl chloride (PVC), polysulphone (PS), polyethersulphone (PES), polyvinylidene fluoride (PVDF), and polyacrylonitrile (PAN) (Bolto et al., 2020). Furthermore, among some well-known methods for polymeric membrane fabrication including sintering, track-etching, stretching, and phase inversion, phase inversion method has been chosen widely for the fabrication of polymer-based membrane (Dong et al., 2021). In this method, a homogeneous polymer solution is made by dissolving the selected polymer in a compatible solvent followed by inducing stage using an external or internal factor like nonsolvent-induced precipitation and thermally-induced phase separation, or evaporation-induced phase inversion (Ismail et al., 2020). Due to various fabrication method available, high removal efficiency, and lower material cost, the organic polymeric membrane then has been used widely in modern wastewater treatment plants (Galiano et al., 2018; Yadav et al., 2021).

However, the enormous development of polymeric materials is still possible to experience some challenging issues such as brittle and fouling-prone membrane surface, along with rigorous preparation methods (Mazinani et al., 2019; Zuo et al., 2018). To overcome this issue, several studies then have investigated the effectiveness of their strategies to increase membrane performance by modifying feed characteristics or developing membrane structure by synthesizing and adding additives substances (Ding et al., 2019; ElSherbiny et al., 2019).

In terms of modifying feed characteristics, it refers to pre-treatment process application to increase the quality of wastewater or contaminated water before entering membrane system (Jepsen et al., 2018). Pre-treatment or hybrid membrane system has been an efficient way to increase the purity of membrane feed (Martini and Setiawati, 2021).

Moreover, developing the internal and external structure of membrane materials relates to the implementation of several modification to increase their strength for facing future fouling and scaling on surface (Hai et al., 2019). This option can be conducted by adding particular additives having ability to increase membrane resistance on pollutants exposure (Barambu et al., 2020).

POLYMERIC MEMBRANE FABRICATION WITH ADDITIVES

The utilization of novel additive has open more chance for better polymer-based membrane performance for treating complex industrial wastewater. Several current studies then reported positive outcome in accordance with compatible synthesized additive added during fabrication process (Table 1).

To begin with, a novel emulsion polyvinyl chloride (EPVC) microporous nanocomposite ultrafiltration membrane has been reported (Farjami et al., 2020). In this work, the combination of EPVC/ (*para*-hydroxy benzoate alumoxane) PHBA ultrafiltration membrane was conducted using the non-solvent induced phase separation technique. It concluded that high hydrophilicity of PHBA particles could significantly improve membranes performance. This happened due to the availability of hydrophilic PHBA additive in membrane casting solution. The modified membranes also demonstrated better percentage of flux recovery ratio (FRR). Specifically, the addition of 0.5 wt.% of PHBA in the casting solution showed the highest water flux growth by more than 47 % along with much better FRR rate by more than 65 %. To conclude, hydrophilic nanosphere additives could reduce the membrane surface roughness related to hydrogen bounding between polymer and functional groups.

Other study then tried to improve the characteristic of organic polymeric PVDF-based membrane by synthesized 3.4-ethylene dioxythiophene (EDOT) additive through chemical vapor deposition methods (Yuan et al., 2021). After multiple filtration tests, it was found that the sheet resistant degree of modified membrane could visibly increase hydrophilic behavior of the membrane which can be regarded to the presence of alkoxy group. Compared to other analysis related to membrane adsorption ability, this modified version showed lower performance on treating three membrane feed streams namely BSA, sodium humate and sodium alginate solution. This has been linked to more blocked pores on membrane due to PEDOT particle. However, it has better chemical stability during alkali cleaning even after the six filtration cycles. In other part of this study, a modified membrane with 1 V/cm of electric field showed better stability on anti-fouling properties due to lower cake formation on the surface. PEDOT polymerization process on the membrane surface might cause higher values of zeta potential and membrane roughness.

A group of study also reported an excellent innovation regarding fouling mitigation on polymer-based membrane through a novel membrane composite consisting of PVDF and Ag@TiO_2 components by adding synthesized additive namely 3-Aminopropyl triethoxysilane (APTES) that could act as a cross linker for holding nanoparticles through phase inversion mechanism (Mishra, J.R. et al., 2021). Regarding the treatment of solution containing Coli bacteria and BSA, synthesized membrane could reach high removal efficiency by more than 90% even for the next cycles after cleaning process. From this point of view, membrane composition can be considered cost-effective to scale-up usage.

The selection of additives that would be synthesized or added to membrane dope solution can be influenced by considering some aspects such as the type of membrane feed, previous research reports, and operating condition. It has been shown by a study developing a new type of graphene–based additive for fabricating asymmetric microporous polyethersulfone (PES)/ polyvinyl alcohol- graphene oxide-sodium alginate (PVA-GO-NaAlg) nanocomposite hydrogel (HG) blended with nanofiltration membranes (Amiri et al., 2020). This complex components were put accordingly to fabricate membrane that can have special characteristics in fighting severe fouling. By using phase inversion method supported by immersion precipitation technique, the hydrophilic PVA-GO-NaAlg nanocomposite hydrogel was processed in situ by chemical crosslinking in the presence of the saturated boric acid and $CaCl_2$. They then claimed that, compared with pristine PES and PES/polyvinyl pyrrolidone (PVP) membranes, the modified membrane containing PVA-GO-NaAlg additive showed much better performance. This study supported its statement by conducting a set of filtration tests on Lanasol blue 3R dye that resulted in more than 83% of dye removal efficiency

Table 1. The breakthrough of membrane fabrication in terms of additive addition

Membrane fabrication components	A state-of-art technique	Feed	Condition and experimental data	In comparison with its unmodified version	Ref
PVC, *para*-hydroxybenzoate alumoxane (PHBA) as additive, PEG, and NMP as solvent	Dope solution consisting of a newly developed emulsion PVC and PHBA for fabricating membrane through phase inversion procedure	BSA protein solution	A dead-end ultrafiltration cell, effective area 19.6 cm^2, pressure 0.2 Mpa, ambient temperature. PHBA 0.5 wt.%, *FRR:* 65.3%	Being more hydrophilic and permeable, the increase in water flux by 47.1% despite having the same BSA rejection value for both pristine and modified membranes by (>98%	(Farjami et al., 2020)
PVDF, polyvinyl pyrrolidone (PVP), ethylene-dioxythiophene (EDOT) as additive, iron trichloride, N-dimethylacetamide (DMAc) as a solvent DMAc as solvent	The combination of the immersion precipitation technique induced-phase inversion and polymerization of EDOT monomer in the vapour phase with co-polymer PVP	BSA, sodium humate, sodium alginate	Anaerobic MBR, external pressure 200 mbar, membrane gap height 100 μm, continuous filtration for 8 h, membrane regeneration using HCl followed by NaOH/ NaClO mixed solution	Having lower pollutants adsorption, the increase in membrane resistance from $0.12x10^{12}m^{-1}$ to $3.69x10^{12}m^{-1}$, lower initial water flux by >50%	Yuan, et al., 2021
PVDF, PVP, (3-Amino-propyl) triethoxysilan, silver nitrate ($AgNO_3$) and TiO_2 as additive, and NMP as solvent	Phase inversion technique followed by Ag@TiO_2 decoration, hydroxylation, and APTES addition	BSA protein solution	Room temperature, pressure 0.1 MPa, BSA solution 1 g/L, membrane active area 19.5 cm^2, rejection: 96.3% FRR: 93.7%, membrane regeneration using de-ionised water and UV radiation	The increase in BSA rejection and FRR from 67.5% to 96.3%, and from 61.4% to 93.7%, respectively	Mishra, et al., 2021
PES, polyvinyl alcohol-graphene oxide-sodium alginate (PVA-GO-NaAlg), PVP, DMAc as solvent	A novel mixture between polymer PES and PVA-GO-NaAlg nanocomposite hydrogel for producing nanofiltration membranes using phase inversion method supported by immersion precipitation way	Dye solution containing lanasol Blue	A dead-end stirred cell filtration system, a cell capacity 200 mL, effective area 19.6 cm^2, 60 min filtration time, pressure 3 bar, at room temperature	Having higher hydrophilic and porous level leading to higher water flux, increasing dye removal and reducing total fouling from 79.23% to >84%, and from 71.2% to 43.6%, respectively	Amiri et al., 2020
PVDF, PVP, TiO_2 as additive, and DMAc as solvent	The mixture of PVDF polymer and PVP along with various amounts of TiO_2 nano-particles through phase inversion method	landfill leachate con-taining copper ions	A dead-end filtration mode, around 20 membrane pieces having uniform length of 35 cm each, pressure 0.3 MPa, total filtration time 200 min with 50 min for each evaluation interval/ rejection: 98.18% , rmembrane egeneration using tap water	Having the increase in copper removal and flux value from 96.36% to 98.18 %, and from 89 to 157 L/m^2h, respectively, while contact angle reduced from 66.71° to 50.01°	Abba, et al., 2021

Furthermore, a study led by Abba then investigated the effect of photocatalytic substance named titanium dioxide (TiO_2) in various percentage in the fabrication of PVDF-PVP fiber membrane via phase inversion method (Abba et al., 2021). The results were incredible where the fabricated membrane having 1.0 wt% of TiO_2 could record maximum flux by 223 $L/m^2 \cdot h$ and copper ion removal by more than 98 %. Morphological analysis informed that the addition of TiO_2 into membrane casting has produced larger pores on membrane matrix with finger-like pores on membrane pores/ surface. The filtration tests conducted more than 3 h confirmed that modified membrane containing 1.0 wt% TiO_2 could double the flux rate. Eventually, cellulose acetate-based polymeric ultrafiltration membrane was developed by involving synthesized additives namely α-aminophosphonate modified montmorillonite (MMT) and Ag-TiO_2 nanoparticle (Abdel-Karim et al., 2021). This study reported that for filtration test performed using dead-end filtration mode, this composite membranes outperformed its pristine version by achieving nearly six-fold increase.

CONCLUSIONS

Massive industrial development has been the most influential factor of wastewater generation. In order to avoid further negative impact of this unwanted by-product, proper wastewater treatment should be a compulsory approach. Despite relatively higher initial installation and maintenance cost compared to other treatment technologies, membrane separation can be regarded as one of the most reliable and efficient options for purifying wastewaters prior to final disposal places. As membrane fouling phenomenon could significantly lower membrane performance, some efforts have to be applied through physical and chemical modification, including by adding some suitable additives in their casting materials. This review article specifically covers some critical findings related to the application of additives during polymeric membrane fabrication or prior to filtration proses on behalf of fouling mitigation. To conclude, the outcome of comprehensively relevant studies regarding additives blending influence on fouling has mostly confirmed positive. In this case, proper operating condition, polymer and additive characters should be taken into account.

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