



Membrane Technology for Water Pollution Control: A Review of Recent Hybrid Mechanism

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Abstract

Current development of membrane technology for purifying polluted water and wastewater is discussed in this review in accordance with its hybrid application with other types of water treatment methods such as adsorption, advanced oxidation processes (AOPs), and biological activated sludge. The hybrid implementation aims are to lengthen membrane lifespan and elude severe fouling on the surface and pore of the membrane. Membrane material and fabrication technology were concisely included. Two prominent materials to fabricate membrane namely polymeric and ceramic are reviewed well along with the exploration of biopolymers based-materials such as starch and alginate. Several fabrication methods available to be implemented including interfacial polymerization technique, non-solvent induced phase separation, temperature induced phase separation, electrospinning, and sintering are deliberately discussed. The next section includes fouling analysis involving reliable fouling mechanism namely Hermia's models to help with the explanation of fouling occurrence during filtration namely complete blocking, standard blocking, intermediate blocking, and cake filtration models. Ultimately, recent research reporting the outcome of the hybridization of membrane and adsorption, coagulation-flocculation, AOPs, and biological treatment using microorganism are properly reviewed. Overall, this review relates to the findings of relevant works which mostly revealed positive outcome of the hybrid membrane system contributing to stronger foundation for future research.

Keywords: Fabrication, flux decline, hybrid system, membrane fouling

1. Introduction

There is an increasing demand for clean water worldwide as the impact of climate change, population and industrialization growth. Therefore, water reclamation has been a great loophole to improve clean water sustainability (Wu, 2019, Martini and Yuliwati, 2020). Municipal and industrial wastewater may contain various pollutants including dyes, heavy metals, oil, and grease compounds (Bhattacharjee et al., 2020, Mishra et al., 2021). Industries such as petroleum refinery, pharmaceuticals, cosmetics, textile, and paints could be part of carcinogenic and non-biodegradable polluted water sources (Afroze and Sen, 2018, Chakraborty et al., 2020, Pavithra and Jaikumar, 2019). Therefore, the quality of wastewater must be improved before final discharge to the environment through selected treatment methods including coagulation/flocculation, flotation, biological ways, adsorption process, and membrane filtration (Putatunda et al., 2019).

Membrane separation then can be regarded as a reliable option offering smaller carbon

emission, great selectivity, and lower defect rate despite its costly installation and maintenance fund. The benefits then have led to an increasing interest of applying technology in various water treatment plants (Yadav et al., 2021, Martini and Ang, 2019, Asif et al., 2019). However, membrane fouling can bring harmful effect on membrane performance due to permeate flux reduction leading to shorter membrane usage. In accordance with this matter, fouling mitigation efforts like design improvement, regular maintenance, pre-treatment or hybrid mechanism, and surface modification could be dependable alternatives (Tan et al., 2018, Al Aani et al., 2020).

Membrane technology applied for filtering raw industrial wastewater containing high concentrated pollutants needs complex cleaning process chemically and physically resulting in higher maintenance cost. Regarding this issue, some relevant works have tried to find the best options for reducing the burden including through applying hybrid mechanism.

Despite some studies already reviewing integrated research in wastewater treatment area, the development of the recent hybridization of membrane with other techniques such as adsorption, coagulation-flocculation, AOPs, and biological treatment using microorganism including several related aspects still needs more discussion. To conclude, this work will critically review the findings of current works regarding hybrid membrane system for lower fouling and better permeate level.

2. Methodology

This review was written by following particular sequence regarding relevant issues to recent development of fouling mitigation on ceramic and polymer based-membrane through pre-treatment. Firstly, it was started by introductory part discussing wastewater effect and its sources followed by various available wastewater treatment technologies. The second part covered methodology explaining the structure of the paper in relation to the topics of each part. The next section then reviewed membrane materials and fabrication methods. The fourth section resumed fouling related issues like its mechanisms and types. Further section then focused on the critical discussion of the latest studies investigating various hybrid mechanisms using pre-treatment such as adsorption, coagulation-flocculation and biological stage as well as its effect on fouling mitigation and membrane performance as a whole. Ultimately, conclusion section was presented by pointing important aspect of each previous section.

3. Membrane Materials and Fabrication

3.1. Membrane Materials

Membrane engineering plays significant roles in varying industrial, medical, biotechnological, and water management fields. It is linked to the idea for membrane processes to substitute conventional energy intensive separation techniques enabling sustainable industrial growth. The compatibility between various membrane operations in hybrid systems, the good stability under operative conditions, and the environmentally compatible aspect are crucial for better rational utilization and feed recovery (Drioli and Giorno, 2009, Martini and Setiawati, 2021).

In general, there are three basic materials used to fabricate membrane including polymeric, ceramic, and metallic based membranes. Polymeric-based membranes

have been popular due to comparatively smaller footprints, being flexible to operate in various conditions, cost-effectiveness, and having better resistance to organic solvent (Barambu et al., 2020, Dong et al., 2021). Synthetic polymers-based membranes including polysulfone (PS), polyethersulfone (PES), polyvinylidene fluoride (PVDF), polyamide (PA), polycarbonate resins, and polyelectrolyte complexes have also been widely applied (Martini and Yuliwati, 2020). Other than those aforementioned, the exploration of biopolymers based-membranes such as starch, alginate, poly (lactic acid), silk and poly (butylene succinate) have also been reported (Mansoori et al., 2020). Furthermore, ceramic membranes like Al_2O_3 , ZrO_2 , SiO_2 , and TiO_2 -based membrane have great performance and better stability related to thermal, chemical, and mechanical challenges despite being more costly than that of polymeric membrane (Suresh et al., 2017). The observation of cheaper materials like mineral-based substances made from zeolite, fly-ash, and kaolin for fabricating ceramic membranes has attracted more interest as those materials could produce lower-cost ceramic membranes having comparable filtration performance (Li et al., 2020).

The next type regarding membrane material construction is metallic mesh-based membrane representing its potentiality related to ultra-low pressure difference involving hydrostatic pressure and certain additive substances such as TiO_2 , ZnO, HNTs, CNTs, SiO_2 , BiVO_4 , UiO-66 (Barambu et al., 2020). Their ability is competitive. The application of modified stainless-steel mesh membrane purifying oily contaminated water, for example, has showed satisfying outcome in terms of the percentage of oil removal (Gunatilake and Bandara, 2017). This metallic membrane could be modified using modest method like dip or spray coating as well as layer by layer technique.

Eventually, there is a new membrane material called mixed matrix membranes (MMMs) featuring polymers and inorganic or organic materials. This can also relate to incorporated nanomaterials in solid-liquid-phase dispersed in a continuous phase. The advancement of membrane technology has enabled to orchestrate efficient process in fabricating mixed matrix membrane in the field of water and wastewater treatment (Vatanpour et al., 2012). This membrane type offers stable physicochemical characters of the inorganic or organic materials supported by basic behavior of the polymers itself and, at the same time, improves morphological aspect of the

membrane leading to better permeate selectivity and fouling resistance (Qadir et al., 2017). Filler addition would perform on the morphological feature influencing transport phenomenon where the interfacial void formation, aggregation and pore blockage could be the resultant of MMMs (Vatanpour et al., 2012). While interfacial voids add some additional sites allowing solvent to pass through membrane, membrane density would concern on the mechanical strength and rejection rate. There are some studies reported their finding on MMMs research such as a study proposing various types of MMMs, namely ordinary nanocomposite, thin film composite with nano thin film, nano composite, thin film composite with nanocomposite substrate and surface located nano composite (Goh and Ismail, 2015). These differences corresponded to the filler used in the fabrication stage such as inorganic filler, organic filler, biofiller and hybrid filler. Additionally, organic filler like cyclodextrin, chitosan beads, wheat straw, yellow birch, pine, and rice husk have been claimed reliable to be used in substrate matrix especially via blending and phase inversion method (Wang et al., 2015).

3.2. Membrane Fabrication

Membrane can be fabricated via several methods that can be properly conducted by considering main membrane materials and operating conditions needed. There are various prominent methods that have been widely implemented. Firstly, interfacial polymerization technique. This way has been widely applied to produce many types of membranes like reverse osmosis, ultrafiltration, thin film composite, and desalination membranes. The amine diffusion into decided organic solution and reactive monomers will lead to compact polymer based-membrane with a thin layer strengthening membrane internal structure during pressure-based filtration (Dong et al., 2021). Several accountable monomers utilised in membrane fabrication via interfacial polymerization are piperazine and trimesoyl chloride (Lau and Ismail, 2011, Dong et al., 2021).

Secondly, phase separation techniques such as non-solvent induced phase separation (NIPS) and temperature induced phase separation (TIPS). For NIPS, a selected polymer or the mixture of polymers are dissolved in the solvent until forming homogeneous dope solution along with other additives or coating particles for better membrane characters (Hausman et al., 2010).

The dope solution is then cast resulting in a liquid film on a prepared glass plate or the substrate to be further immersed in a coagulation non-solvent bath. Eventually, phase inversion would occur when the solvent in the formed film exchanged with the non-solvent resulting in asymmetric membranes having dense selective layer with supportive sub-layer. Furthermore, TIPS method refers to a prepared dope solution comprising polymer and solvent processed at certain temperature near the polymer melting point followed by casting stage into a film and cooling down process at room temperature. In this way, phase separation would be processing within the change of temperature that needs higher energy intensive leading to the formation of a dense film (Dong et al., 2021). Apart from polymeric membrane, phase inversion method has also been applied in creating ceramic-based membrane especially for porous ceramic membranes having hollow fibre geometry (Zhu et al., 2015).

Other type of well-known membrane fabrication is electrospinning. In this process, high electric terrain is used to generate nanofibres derived from the charged polymer solutions. The formation of fibre structures is supported by the repulsive electrostatic force given during electrospinning process. Different types of membrane morphology and surface topography could be obtained by modifying parameters and polymer properties in solution during electrospinning (Valizadeh and Mussa Farkhani, 2014, Ismail et al., 2020). A study then revealed the ramification and bending process of applied charged polymer jet via differing the voltage of electrical field to have columned tissue of the polymer nanofibres to form closed loops modes (Xin and Reneker, 2012).

Eventually, sintering is another emerging method of membrane fabrication especially for ceramic based-membrane. Ceramic membranes which could be the asymmetric membranes consist of several parts namely alumina support, immediate layer, and filtration layer. In this case, alumina support is sintered at high temperature mainly around 1600 °C or above to enable the existence of bending strength for final support followed by coating stage of some immediate layers for penetration prevention. Lastly, the membrane layer should be coated for getting great accuracy during filtration process (Zou and Fan, 2021).

Apart from fabrication method, the use of solvent has also an important role in order to create membrane having great functionality

as solvent could effect on membrane morphology along with other properties. Some organic solvents such as N-methylpyrrolidone (NMP), N,N-dimethylacetamide (DMAc), N,N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), acetone, dimethyl phthalate (DMP), triethyl phosphate (TEP), and tetrahydrofuran (THF) could be used and adjusted especially for fabricating a membrane involving polymer material (Dong et al., 2021, Zhao et al., 2021). However, those solvents have been considered harmful for human health and environment quality, both for the long term usage and massive amount aspects. Therefore, concern on more environmentally friendly solvent types has led to more investigation on the discovery of green solvents having much lower toxic impact. Several relatively newly developed green solvents have been then investigated such as organic carbonates and polarClean (Rasool et al., 2019, Dong et al., 2021).

4. Membrane Fouling

Membrane fouling is still a significant concern on membrane technology, and this is indicated by the rate profile of permeation flux during separation process. Overcoming severe and early fouling could help membrane reaching longer lifespan. In general, the measurement of permeate flux could be rated using the following equation (Eq. 1) (Martini and Ang, 2019):

$$J = \frac{V}{A t} \quad (1)$$

Remark:

- J : permeate flux rate (L/m² h)
- V : permeate volume at certain time interval (L)
- A : active membrane area (m²)
- t : permeate collection time (h)

For measuring membrane flux rate, the Eq. 1 can also be expressed as following formula (Eq. 2):

$$\frac{dJ}{dt} = -K (J - J_{ss})J^{2-n} \quad (2)$$

Remark:

- K : constant permeation flux rate
- J_{ss} : steady state permeation flux rate

Where the values of n are 0, 1, 1.5, and 2 for cake filtration, intermediate blocking, standard blocking, and complete blocking model, respectively.

Hermia's models are sufficiently reliable to give possible explanation of fouling mechanism during filtration based on the experimental data. These models covers

complete blocking, standard blocking, intermediate blocking, and cake filtration models and can be explored by following general Hermia's equation (Eq. 3) (Martini and Ang, 2019, Khan et al., 2020).

$$\frac{d^2t}{dv^2} = K \left(\frac{dt}{dv}\right)^n \quad (3)$$

Cake formation theorizes particular phenomenon where pollutants may have averagely a bigger size than membrane pore size leading to pollutants accumulation building up cake layer on the surface. In this case, each pollutant particle would locate on other particles already blocked the pore. The intermediate fouling then could be defined as fouling caused by pollutant particles having an equivalent size to the pores settling on the existed particles previously blocked some pores. Standard blocking explains that there could be no uniform particles adsorbed the pores leading to lower pores diameter as every particle arriving on the surface would be deposited onto the internal pore walls decreasing the volume of the pores. Eventually, complete blocking model refers to blocked pores due to pollutant particles which averagely have bigger size than the pores and they settle on the surface reducing permeate flux rate in accordance with time and no superposition of particles (Martini and Ang, 2019). These models can be stated and illustrated in Table 1 and figure 1, respectively.

Table 1. Hermia's blocking models

| Blocking mechanism | n | Formula |
|-----------------------|-----|---|
| Cake layer | 0 | $\frac{1}{J^2} = \frac{1}{J_0^2} + K t$ (4) |
| Intermediate blocking | 1 | $\frac{1}{J} = \frac{1}{J_0} + K A t$ (5) |
| Standard blocking | 1.5 | $\frac{1}{J^{0.5}} = \frac{1}{J_0^{0.5}} + K t$ (6) |
| Complete blocking | 2 | $\ln(J) = \ln(J_0) - K t$ (7) |

Membrane reusability is also beneficial to predict its lifespan and resistance. Therefore, flux recovery ratio (FRR) can be measured by following formula (Eq. 8) (Martini and Ang, 2019):

$$FRR = \left(\frac{J_{f,c}}{J_{f,i}}\right) \times 100 \quad (8)$$

Remark:

- $J_{f,i}$: feed flux rate using a fresh membrane
- $J_{f,c}$: feed flux rate using a cleaned membrane

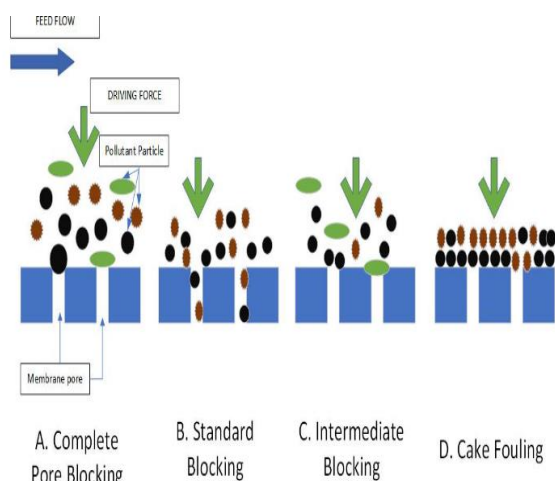


Figure 1. The illustration of Hermia's blocking models

Apart from fouling mechanism, the dominant type of pollutants causing fouling should also be taken into consideration for fouling mitigation. Several fouling classifications including inorganic fouling, organic fouling, biological fouling, and colloidal fouling may happen individually or simultaneously depending on the content of the feed.

Inorganic fouling usually refers to the inorganic solid materials like calcium carbonate, silicate, aluminium oxide, and calcium phosphate accumulating on the pores or surface during separation process leading to crystallization and transport process (Choudhury et al., 2019, Goh et al., 2018). The existence of some salt could restrict their solubility in water along with the alteration of temperature, solvent evaporation, and concentration polarization (Goh et al., 2018).

Furthermore, organic fouling links to the deposition of natural organic matters (NOM) such as bovine serum albumin, humic acid, proteins, lipids, amino acids, and organic acids on membrane surface or inside the pores. High content of NOM adsorbed in the surface or pores will alleviate the hydrophobicity of membrane material as the adhesion of the organic matter influence both hydrophobic and electro-static interactions yielding severe fouling with time due to a dense gel layer having low permeability on the surface (Guo et al., 2020, Ying Shi et al., 2020, Ricceri et al., 2021).

In terms of biological fouling, the availability of microorganisms in the feed accumulating on the surface and forming biofilm is the main factor. This biofilm has certain component named extracellular polymeric substances consisting of living, inactive, or decomposed microorganisms accumulated in multiple

stubborn layers possibly leading to irreversible membrane blockage in some parts of the membrane.

Ultimately, colloidal fouling happens due to suspended colloids leverage having nano or micrometres in size settled on the pores or surface. Colloidal compounds can be linked to both inorganic particles like silica, clay, and aluminium silicate minerals; and organic pollutants including proteins and polysaccharides (Flemming et al., 1994, Tang et al., 2011).

5. Hybrid Mechanism in Membrane Application

There are several efforts that can be conducted to avoid earlier severe fouling on membrane as well as optimize its hydrodynamics such as pre-treatment process, periodic cleaning, and physical or chemical surface modification. However, pre-treatment process that could be also defined as a hybrid membrane mechanism has been popular since it supports membrane performance and, at the same time, increases permeate quality. Various investigations have been reported in the literature regarding this hybrid mechanism. While some mostly conducted the additional treatment before feed entering membrane system, others put them as following stage of the membrane system in order to gain some additional purpose.

5.1. Adsorption – Membrane

The hybrid of adsorption and membrane systems has been considered effective to improve membrane performance by reducing pollutants contained in the feed as adsorption is mostly put at the first stage as pre-treatment as displayed in figure 2. For different membrane types like reverse osmosis, nanofiltration (NF), ultrafiltration (UF), and micro-filtration (MF), their integration to various inorganic and organic adsorbents like activated carbon (AC), zeolite, graphene, carbon nanotubes based-sorbents or iron oxy/ hydroxide based-biochars, and metal-organic frameworks have shown positive outcome lowering membrane workloads leading to lower flux profile decrease (Xu et al., 2017, Martini and Setiawati, 2021).

The transport of inorganic and organic pollutants by a hybrid membrane system is influenced by several aspects such as the properties of both adsorbent and membrane along with particular behaviour of the

pollutants contained in the water like their type, size, charge, and hydrophilicity. The AC made through partial combustion of biomass, biowaste and other byproducts has contributed significantly in the field of wastewater purification including coal-based AC and oxygen- and nitrogen-based activated biochar to remove humic acid (Chu et al., 2017).

In a comparative study, pollutant retention of hybrid UF and adsorption using AC, oxygen-based activated biochar, and nitrogen-based activated biochar having the same dosage of 20 mg/ L was around 2.4%, 12%, and 13% higher than that of the sole UF system, respectively (Chu et al., 2017). Furthermore, other study reported the removal of ibuprofen, 17 α -ethinyl estradiol, and carbamazepine pharmaceutical pollutants using both sole and membrane systems involving biochar based-adsorbent (Kim et al., 2019). This work resulted in the average removal efficiency for those pollutants where sole UF membrane and hybrid activated biochar/UF membrane system yielded, at neutral pH 7, 42% and 53%, respectively.

In terms of dye contaminants, other study also proved that adsorption could help increasing membrane performance along with higher

quality of permeate. By using MXenes, a newly improved multilayered two-dimensional inorganic graphene-based material, a study applied hybrid titanium carbide ($Ti_3C_2T_x$) MXene and a polyamide composite membrane having MWCO of 3000 Da to separate methylene blue and methyl orange dyes from solution (Kim et al., 2020). The evaluation of permeate flux rate modelling permeate flux was done for three different system comprising the sole UF membrane, hybrid MXene/ UF, and PAC/ UF membrane. It was indicated that hybrid MXene-UF led to thinner cake layer on membrane surface.

In contrast, this study found that the hybrid AC-UF membrane could increase the thickness of cake layer on membrane surface indicating the unexpected role of AC as foulant. Apart from that, cake filtration model was also considered well-fitted for ethylene blue removal using hybrid MXene-UF membrane compared with other Hermia's blocking models. When it was utilised to analyse hybrid PAC/ UF system, cake filtration model also strongly linked to the main fouling mechanism indicating the deposited PAC particles forming a thicker cake layer on the surface (Kim et al., 2020).

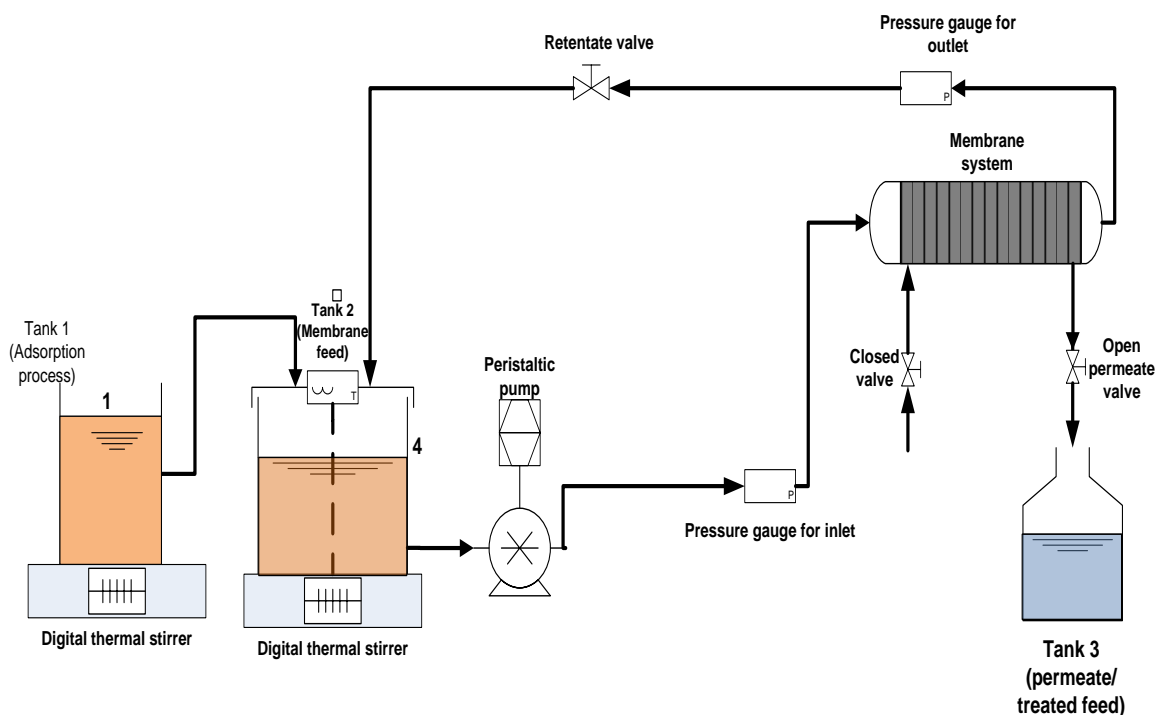


Figure 2. Schematic illustration of hybrid adsorption and membrane process in the lab-scale research.

5.2. Coagulation/ Flocculation Membrane

The terms of coagulation and flocculation seem to happen in sequence. Both of these processes utilise specific chemical agents namely coagulant or flocculants such as aluminium and ferric chloride (Zhao et al., 2020). Certain dominant parameters could decide their efficiency including initial concentration of pollutants, coagulants/flocculants dosage, and initial pH. The initial aggregation of particles could occur via destabilization during coagulation process in which charge neutralization, interparticle bridging, and sweep flocculation are taken into account. Once particles in natural water are mostly negatively charged, the charge neutralization is then possible when sufficient cationic metal hydroxides are adsorbed leading to reduced zeta potential. Furthermore, sweep flocculation may happen as the concentration of coagulants exceeds the solubility limit yielding precipitating and enmeshing the particles. It could also be influenced by the initial quality of wastewater before entering membrane system (Malkoske et al., 2020).

In this process, the collision among pollutant particles lead to floc formation. Specifically, fluid shear induced through the mixing acting as dominant flocculation mechanism when two colliding particles have more than 1 μm in diameter, while Brownian motion would dominate when one of them has less than 1 μm in diameter along with differential sedimentation (Youn and Lawler, 2019). The aggregation phenomenon would be higher when particles are completely destabilized by coagulation. The hydrodynamic conditions of coagulation/ flocculation process can be described by mean velocity gradient, \bar{G} , and contact time, t . There are three particular categories of coagulation/ flocculation that may happen during this process.

Firstly, for coagulation process with no or incidental flocculation, a coagulant is typically implemented on a continuous basis followed by the rapid or static mixing without involving flocculation stage with contact time less than one minute. The coagulant dose, solution pH, hydrodynamic condition and contact time have an impact on the floc properties such as floc size, growth value, surface charge, strength, and structure. Secondly, coagulation and flocculation type which was typically conducted using mechanical mixing followed by membrane filtration. The other type is conventional coagulation which involves

sedimentation. In this style, operating condition should be optimized for better floc growth and turbidity and organic matter reduction during settling (Malkoske et al., 2020).

A study utilised inorganic and organic coagulant types namely alum ($\text{Al}_2(\text{SO}_4)_3$), iron(III) chloride (FeCl_3), polyaluminium chloride (PAC), and processed Moringa Oleifera powder (MOP) to support nanofiltration (NF) membrane separation for purifying wood processing wastewater (Bouchareb et al., 2020). It then revealed the role of coagulation/flocculation process as pre-stage regarding the turbidity, COD, and colour removal. To conclude, this hybrid chemical coagulation and NF could result in up to 99.9%, 99.5%, and 99.4% of removal efficiency of those parameters, while the use of MOP (2000 mg/L) acting as biocoagulant in the hybrid NF membrane could also drastically reduce wastewater turbidity, COD, and colour for up to 99.6%, 93.0%, and 99.5%, respectively, along with the least membrane fouling by 48.9 %. Those results confirmed the effectiveness both chemical and green coagulants as pre-treatment stage to help reduce membrane feed burden.

Other work utilised polyaluminum chloride as coagulant followed by flat-sheet cross-flow ultrafiltration (UF) membrane for treating polluted effluent from biological treatment process in a paper mill (Wu et al., 2019). It was found that the increasing coagulant used from 0 to 2,000 mg/L led to decreasing total membrane resistance and increasing membrane flux, however, the concentration polarization resistance was also increasing. In accordance with the concentration polarization resistance, it was assumed that cross flow velocity mode might have more leverage than the trans-membrane pressure. Three factors namely coagulant dose, cross-flow velocity (CFV), and trans-membrane pressure (TMP) mostly affected the fouling and water flux. Overall, this finding could control membrane fouling especially for tertiary treatment using flat-sheet cross-flow membrane process.

For dye removal scenario, a study of the hybrid coagulation/flocculation and MF membrane reported that the solophenyl blue (SB) dye could be significantly removed using bio-coagulant namely potato starch (PS) in the pre-treatment stage (Januário et al., 2021). As particles and colloids had been previously removed during coagulation-flocculation process, the feed contained much lower pollutant before entering membrane system. Furthermore, unlike unmodified membrane

material, the addition of TiO₂ when fabricating membrane has led to exceptional outcome where a 100% colour removal could be achieved with the considerable decrease in fouling rate at around 63.65%.

Ultimately, raw textile wastewater that was treated previously using AC and aluminium sulphate acting as coagulant has been confirmed of having better quality enabling lower pollutant loading for the subsequent UF stage (Alibeigi-Beni et al., 2021). This work added chitosan as coagulant aid. It revealed that the use of those coagulant and chitosan by 100 mg/L and 1 mg/L, respectively could give maximum COD and turbidity removal. Other than that, it also noticed the phenomenon that the increasing TMP and cross velocity would cause flux enhancement. In contrast, the removal efficiency of both turbidity and COD decreased. When considering membrane material, PVDF polymer-based membrane has found to have better flux rate and turbidity removal, while PES polymer-based membrane hold better COD removal. Despite its efficacy related to significant decrease in pollutant content in the wastewater prior to entering membrane system, costly reagent and the generation of toxic sludge could be concerns that need be further addressed.

5.3. AOPs – Membrane

AOPs have been widely chosen as part of reliable methods for treating various contaminated solution. There are assorted types of AOPs like Fenton, photo-Fenton, Fenton like, ion exchange, and photo-catalysis reaction using metal oxides or catalysts or metal oxides such as TiO₂, ZnO, MgO, ZnS, and CdS that can be implemented in the treatment plant (Aljuboury and Shaik, 2021, Martini and Ang, 2019, Al-Mamun et al., 2019). Research on AOPs sustainability has been focused on the lower energy requirement, circular economy and waste minimization along with the improvement of catalytic performance.

Solar photo-catalysis is one of AOPs having great connection to the environmental benefits and cost saving due to the usage of natural resource. In the life cycle assessment, solar photocatalysis related to photovoltaic (PV) cells could work at lower waste condition (Dominguez et al., 2018), The emerging methods for nanocatalysts preparation has attracted an increasing interest in the use of AOPs for water and wastewater treatment. They are targeted to create more surface basic sites and coordination number of surface

atoms for higher organic pollutant degradation. These methods could also improve solution quality by reducing some targeted pollutants or parameters like chemical oxygen demand (COD) and total organic carbon (TOC) (Rao and Shrivastava, 2020, Guo et al., 2018, Al-Mamun et al., 2019). Furthermore, Fenton-based AOPs are also promising towards the optimum degradation of recalcitrant organic matters in which they can be considered competitive with other counterparts such as UV-based process, microwave and electrochemical-based AOPs (Miklos et al., 2018).

When it comes to the hybrid AOPs system with membrane technology, several studies have acknowledged their positive outcome. A study conducted comparative analysis on the effectiveness of hybrid TiO₂/UV-UF, Fenton/UV-UF and sole UF membrane for treating raw canola oil effluent along with membrane fouling assessment (Martini and Ang, 2019). It then confirmed that hybrid TiO₂/UV-UF showed best outcome by obtaining around 82 % and 86 % of COD and oil removal, respectively, where flux rate decline was 32 %. On the contrary, sole UF membrane system faced more than 50 % decline.

Other study investigated the effectiveness of a membrane oxidation reactor involving Fenton and photo Fenton along with submerged UF membrane employing the Taguchi experimental method (Dogan et al., 2021). It then revealed the enhancement of UF treatment efficiency with the implementation of Fenton and photo Fenton (UVC₂₅₄ and UVA₃₆₅) in an effort to treat raw pulp and paper wastewater. The minimal consumable chemicals could be achieved by optimizing the synergistic link among specific reactor operation. The reuse of iron ions was kept inside the reactor by membrane reducing sludge production. It was also informed that membrane fouling could be minimised by the role of air pumped into the reactor enabling oxidation process at the membrane surface.

The relationship between AOPs and membrane system in the form of hybrid mechanism has also come to the new perspective by directly adding photo-catalytic agent namely TiO₂ when fabricating membrane. This emerging trend has showed positive reports as stated by several studies. Fabricating catalytic membrane by adapting a built-in system in membrane material added TiO₂ could generate hydroxyl radical to decrease adsorbed foulant on the surface. An investigation on the flat sheet membrane to purify humic acid solution

through in-situ colloidal precipitation was revealed (Teow et al., 2021). In this case, the effect of TiO₂ nanoparticles sizes having around 8 nm, 20 nm, and 21 nm adding in PVDF/DMAc casting solution with the ratio of 18:82 was investigated. The study then stated that the TiO₂ size of 21 nm was superior regarding FRR to other size, however, all photocatalytic agents could help membrane with anti-fouling property due to higher reactive surface area. Further work also revealed that TiO₂ incorporation could give the membrane better porosity degree and selectivity for removing metal ions (Abba et al., 2021). AOPs could be reliable options to reduce the severity of membrane fouling as it will reduce the level of organic and non organic contaminants contained in the feed quickly, however this method could be relatively costly regarding chemical usage.

5.4. Biological Process – Membrane

Since decades ago, biological treatment has been one of the most common techniques in treating wastewater due to their environmentally friendly characteristics, great removal efficiency, and lower operational cost. The conventional biological processes include activated sludge process, trickling filters, moving bed biofilm reactors, nitrification, microalgae/fungi based treatments, biological activated carbon, and other microbiological treatments involving bacteria (Xu et al., 2021). The microorganism can be aerobic (with oxygen), anaerobic (without oxygen), or facultative microorganisms in nature (Pang and Abdullah, 2013, Mishra and Maiti, 2020, Gómez-Ramírez and Tenorio-Sánchez, 2020, Venegas et al., 2021). They are widely employed for the excessive sludge removal from the sewage treatment plant, working in bioreactors, lagoons, ponds or digesters. One of the most widely adopted biological process in industrial is activated sludge process involving biodegradation mechanism in the aeration tank (Ahmed et al., 2017). However, the increasing number of industries in many sectors generating more effluent containing complex and recalcitrant pollutants has been a challenge to implement it as a sole method due to its longer reaction time and wider area needed.

Therefore, there have been various efforts taken to increase the outcome of this method regarding time and efficiency. When it comes to its coordination with modern membrane separation, several works have reported the benefit of combining these biological and membrane system. To begin with, a hybrid microfiltration forward osmosis membrane

bioreactor operated to purify raw municipal wastewater containing various antibiotics like enrofloxacin, sulfamethazine and cefalexin, amoxicillin, lomefloxacin and ampicillin showed satisfying outcome (Qiu et al., 2021). This study informed that around 58.9–100% overall removal of those antibiotics. All the antibiotics were mostly rejected by the membrane while biological treatment has lower efficiency.

Furthermore, a study worked on a sequencing batch reactor (SBR) and NF membrane process for treating textile effluent containing reactive blue dye and sodium dodecyl sulfate (SDS) surfactant (Khosravi et al., 2020). The experimental data then gave a clue that the biological way support the existence of surfactant to better reduce the concentration of the dye and chemical oxygen demand. This collaborative treatment resulted in the removal efficiency values of dye, COD, and SDS by 98%, 98.5%, and 99%, respectively. Despite biological treatment benefits such as more economical and environmentally friendly reasons for integrating with membrane, it could take longer time and need bigger ponds or area for treatment plant.

6. Conclusion

This review critically covers the development of membrane technology focussing on hybrid mechanism for fouling mitigation. Several associated aspects such as prominent membrane materials, fabrication technologies and solvents, fouling models and analysis have also been selectively referred. Furthermore, different types of membrane fouling namely organic fouling, bio-fouling, inorganic fouling, and colloidal fouling which could happen solely or simultaneously are also analysed along with their fouling mechanisms called Hermia's model consisting of cake layer formation, intermediate pore blocking, standard pore blocking, and complete pore blocking model. The main emphasis is given to the hybridization of membrane separation and other technology whether it acted as pre-treatment, collaborative or additive mechanism including adsorption, biosorption, coagulation-flocculation, biological treatments, and AOPs. To conclude, obtaining better membrane performance and fouling mitigation could be achieved via conducting appropriate hybrid or pre-treatment mechanism.

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