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Development of membrane material for oily wastewater treatment: A review

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ABSTRACT

Our world is facing continued challenges of environmental and ecological pollutions due to human and industrial activities. One of the major threats is oily wastewater mainly discharged from oil fields, refineries, automobile, palm oil industries, and many others. Membrane-based technology offers an almost complete separation of oil from water. However, the technology is facing the challenge of maintaining performance over long periods of operation caused by membrane fouling as a result of interaction between oil droplets and the membrane surface. This attracts research interest mainly on developing customized polymeric, ceramic well as a metallic-based membrane material for improved performance. This paper reviews the recent advances of membrane material developments with the focus on methods of improving the surface chemistry, structure, and hydrodynamics and their implication on the filtration performances.

Abbreviations: ΔP, Trans membrane pressure; SiO_2, Silicon oxide; TiO_2, Titanium oxide; CNTs, Carbon nanotubes; SPEEK, Sulfonated poly (ether ether ketone); PEG, Polyethylene glycol; PVP, Polyvinyl pyrrolidone; LiCl, Lithium chloride; ZrO_2, Zirconium oxide; PSF, Polysulfone; PES, Polyethylene sulfone; PVDF, Polyvinylidene fluoride; PAN, Polyacrylonitrile; SDS, Sodium dodecyl sulfate; PWP, Pure water permeability; CS, Chitosan; GA, Glutaraldehyde; HFP, Hexafluoropropylene; PDA, Polydopamine; PDDA, Poly (diallyl dimethyl ammonium chloride); HNT, Halloysite nanotubes; GO, Graphene oxide; RGO, Reduced graphene oxide; APAN, Aminated polyacrylonitrile; PMTAC, poly[2-(methacryloyoxy)ethyl trimethylammonium chloride]; PEGMA, Poly(poly(ethylene glycol) methyl ether methacrylate); PHFBM, Poly(hexafluorobutyl methacrylate); γ-APTS, γ-aminopropyl triethoxysilane; APT, Aminopropyl triethoxysilane; NiCo- LDH, Nickel cobalt layered double hydroxide; CC, Coprinu comatus; PVC, Polyvinylchloride; Al_2O_3, Aluminum oxide; $BiVO_4$, Bismuth vanadate; SCA, (3-aminopropyl) triethoxysilane; TMC, Trimesoyl chloride; WCA, Water contact angle; UWOCA, Underwater oil contact angle; PVDF–HFP, Polyvinylidene fluoride-co-hexafluoropropylene; G-PANCMI, Graphene attached poly acrylonitrile-co-maleimide; CuAAC, Copper (I) catalyzed azide-alkyne cycloaddition; PPSU, Sulfonated polyphenylene sulfone; CCt, Carbon cloths; PAA, Poly (acrylic acid); ZnO, Zinc oxide; PDH, Poly(dimethylaminoethylmethacrylate-co-2-hydroxyethyl methacrylate); PVC, Polyvinyl chloride; HFO, Hydrous ferric dioxide; PETMP, Pentaerythritol tetrakis (3-mercaptopropionate); PEGDGE, Diethylene glycol diglycidyl ether.

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1. Introduction

Industrial developments open up economic and social opportunities which have contributed tremendously to enhancing human civilization. However, unproperly treated wastes discharged by industries have a detrimental effect on human health, environment, and ecological systems with oily wastewater as one of the emerging issues. These attract global attention for implementation of waste treatment, reuse, and discharge limits imposed by regulations, including for treatment and reuse of oily wastewater [1,2]. Oily wastewater generated by oil and gas industries called produced water alone was estimated to be 71 billion barrels per year [3,4] and has to be treated to comply with discharge limits, let alone when including other sectors such as from food, pharmaceuticals, cosmetics industries, etc. [5,6]. Oily wastewater is difficult to treat mainly because it constitutes of a dispersed phase (the suspended droplets of oil or water) and continuous phase (the medium of suspension) [7,8]. Oily wastewater is classified based on the size (diameter) of the droplets in the dispersed phase, for droplets size >150 μm is defined as free floating, between 20 and 150 μm is defined as dispersed and <20 μm is defined as emulsified [9,10]

Oil-water emulsions are classified as oil-in-water, water-in-oil, and multiple or complex emulsion [11,12] as illustrated in Fig. 1. In

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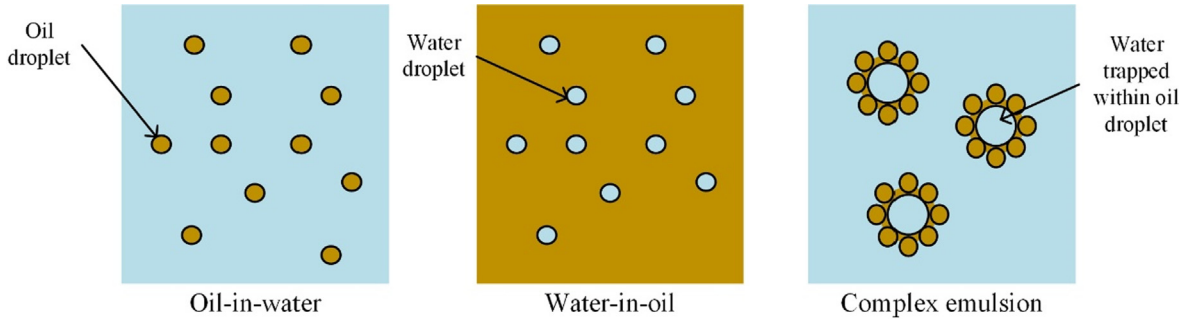

Fig. 1. Classification of oil-water emulsion.

an oil-in-water emulsion, water is the continuous phase while oil is the disperse phase and vice versa for the water-in-oil emulsion. Whereas, complex emulsion consists of tiny droplets suspended in bigger droplets that are suspended in a continuous phase e.g. water in oil in water emulsion consist of water droplets suspended in larger oil droplets that are suspended in a continuous water phase [13,14].

Oil-water emulsions are stable due to the low interfacial tension between the disperse and the continuous phases [15]. Therefore, to accomplish destabilization for easy separation of these phases, the electrostatic barrier that prevents aggregation of the droplets must be removed. It is typically accomplished via one of the following strategies: addition of salt or flocculant, controlling pH or entropy of the emulsion, or by increasing the Van der Waals forces to reach a value higher than the stabilization force via deformation of the droplets [10,16].

Numerous physical and chemical techniques have been developed using various concepts for destabilizing oil-water emulsion for easy separation of oil from water. They include oil skimmers, dissolved air floatation, adsorption, coagulation, centrifuges, electrochemical and photocatalytic treatment, etc. [17]. However, the aforementioned techniques suffer from low separation efficiency, high cost, complex operation (in some cases), and most importantly some generate secondary pollutants and inefficient in the separation of emulsion with <10 μm droplet sizes [18,19]. Therefore, there is a pressing need to develop sustainable, efficient, and reliable technology with a small footprint for continuous industrial development and environmental sustainability [20–22].

Membrane-based technology has emerged as a promising option [22–24]. The technology is in a state of rapid development and is believed to be the leading technology for oily wastewater treatment shortly. Its developments are strongly supported by research on customized membrane material development [25,26].

Several review studies have been carried out on oily wastewater treatment using membrane technology due to rapid technological improvement. Dickhout et al. [4] reviewed produced water treatment using membranes from a colloidal perspective with emphasis on the operating parameters and membrane surface wetting modification. Ahmad et al. [27] reviewed the performance and advancement of thin film composite membranes for oily wastewater treatment. While Tanudjaja et al. [22] review focused on membrane operation modes, membrane module, and economics of membrane operations. There are also recent studies that addressed advancement in polymeric membrane materials and surface modification for oily wastewater treatment [1,28], while some include ceramic membranes as well [29]. However, a study on membrane material development with an emphasis on membrane surface chemistry, membrane surface patterning, and hydrodynamics is still limited. Such an overview is important in other to rationalize the approach for research in this field.

In this review, the challenges on the application of membrane-based technology for oily wastewater treatment are first detailed. Later, membrane fouling as the major issue is highlighted, and the main approaches to cater to the issue via membrane material development are detailed. The current status of materials development including polymeric, ceramic, and metallic-based membrane materials is then reviewed in detail. The discussion is focused on the fabrication approaches and their impacts on filtration performances and limited to pressure driven membranes. Lastly, a brief perspective on future research direction is also provided.

2. Membrane filtration for oily wastewater treatment

Membranes for oily wastewater treatments are classified based on their material of formation as organic (from polymeric) and inorganic (such as ceramic, metal and glass, etc.) [30–32]. Such a wide range of available materials provides options customized to the process conditions/requirements (i.e., high temperature is for ceramic/metallic membranes). This widens up the opportunities for membrane technology to compete with other processes [33,34].

Implementation of membrane-based technology for oily wastewater treatment is facing a challenge in form of membrane fouling. It leads to loss of performance over time which necessitates complex operation, including the application of chemicals for cleaning that eventually increases both operational and capital expenditures [35]. Membrane fouling became very severe for plain membranes especially for the organic membranes due to the hydrophobic nature of polymers that interact well with oil [36]. Therefore, a great number of reports are available in pursuit to turn the surface hydrophilic [35].

Generally, membrane fouling is a result of partial or complete blocking of pores through adsorption of continuous or disperse phases of the emulsion or build-up of one or two phases on the membrane surface promoted by concentration polarization [36,37] as illustrated in Fig. 2. The phenomenon induces a rapid decrease in flux and deteriorates the overall hydraulic throughput [38]. Acidic and alkaline based chemicals are often applied to restore membrane performance. However, frequent use of these chemicals possess not only detrimental health and environmental effects from the secondary pollutants (i.e., halogenated byproducts) but also shortens the membrane lifespan by imposing material degradation [39]. Therefore, the use of these chemicals should be minimized or avoided [40].

To develop effective and sustainable strategies for fouling management, recent researches demonstrated that membrane fouling can be managed by manipulating 1) properties of the feed, 2) the hydrodynamic conditions, and 3) developing fouling-resistant membrane (membrane material development) [41,42]. Implementation of the first approach involves pretreatment using conven-

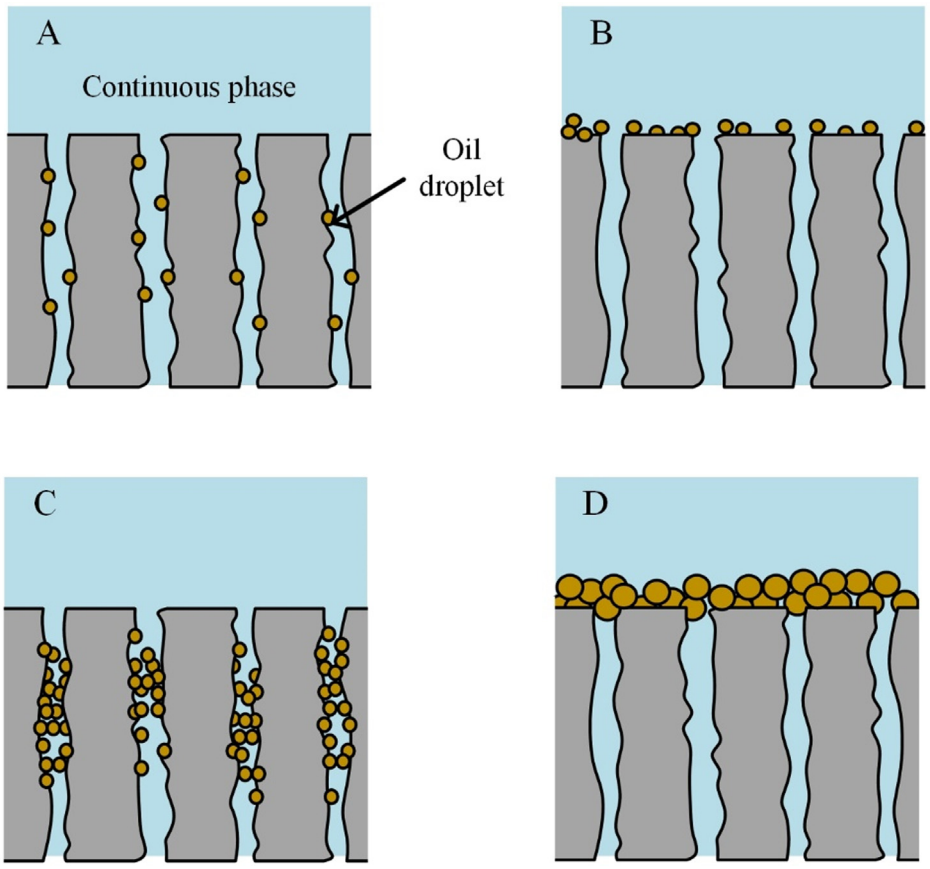

Fig. 2. Membrane fouling models in the treatment of oily wastewater. Partial blocking of membrane pores by droplets of the dispersed phase within membrane pores (A) and on the membrane surface (B). Complete pore blockage by cake layer formation within membrane pores (C) and on the membrane surface (D).

tional techniques to reduce the droplet concentration before membrane filtration. However, this strategy suffers from environmental and economic concerns [19]. While the second approach is limited by the high energy demand [27,35]. The third approach mainly involves the development of fouling resistant membranes via membrane material development [43–45]. This approach is in a state of rapid development and gaining more attention due to its economic and environmental advantages [46]. Fig. 3 illustrates the membrane material development strategies for fouling management.

3. Membrane material development strategies

The basic concept of membrane material development focuses on fine-tuning the surface morphology and/or chemistry by either in-situ or ex-situ modification strategy as depicted in Fig. 3 [47,48]. The former involves direct membrane material development in which the membrane material modification will be done simultaneously with membrane formation. While the latter involves modifying the membrane after its formation [46,49].

Surface chemistry modification is mainly done by improving the membrane wettability, measured through the contact angle of liquid (water or oil) droplet dropped on the membrane surface. The wettability of a solid surface is governed by its ability to repel or absorb water molecules and also its roughness as explained by Wenzel's theory [50]. Membrane surface wettability is classified into four regimes based on the value of water contact angle (θ_{water}): superhydrophobic if $\theta_{water} > 150°$, hydrophobic if $\theta_{water} > 90°$, hydrophilic if $\theta_{water} < 90°$, and superhydrophilic if $\theta_{water} = \sim 0°$. Similarly, for a low surface tension liquid such as oil, membrane surface is considered superoleophobic if underwater oil contact angle (θ_{oil}) is > 150°, oleophobic if $\theta_{oil} > 90°$, oleophilic if $\theta_{oil} < 90°$, and superoleophilic if $\theta_{oil} = \sim 0°$ [50,51].

Several materials prove to enhance surface wettability [52]. They are classified into organic and inorganic additives. The organic additives include polyethylene glycol (PEG), polyvinylpyrrolidone (PVP), and polyetherimide (PEI), etc. [53]. While inorganic additives (nanoparticles) includes silicon oxide (SiO_2), titanium oxide (TiO_2), zirconium oxide (ZrO_2), graphene oxide, and carbon nanotubes (CNTs), chitosan (CS), etc. They are often employed due to the presence of polar functional groups such as hydroxyl, amino, sulfonic, and carboxyl groups and many others on their surfaces [54–56]. Additives are either blend in the dope solution (in-situ modification) or coated on the membrane surface (ex-situ modification) [52]. Other post-treatment techniques (ex-situ modification strategy) such as interfacial polymerization, molecular layer-by-layer, UV-assisted graft polymerization, plasma polymerization, spray coating, immersion, and hydrothermal oxidation to include hydrophilic groups onto the membrane surface have been practiced [57,58].

3.1. In-situ membrane material development strategy

This involves modifying the membrane material simultaneously with the membrane formation. Is a facile and economic strategy that involves loading additives in the dope solution or coagulation bath aimed to preserve/trap the additive within the membrane matrix [49,59].

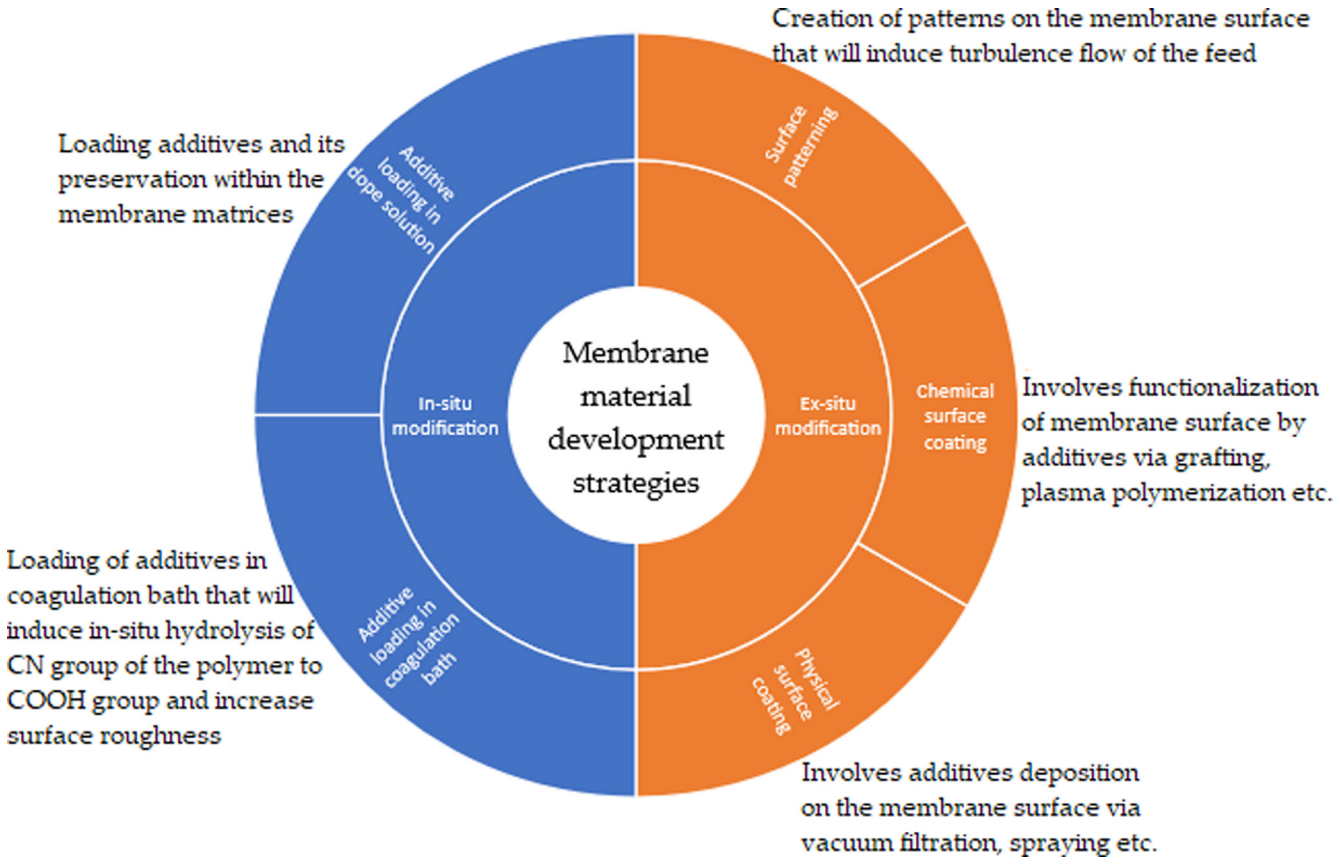

Fig. 3. Overall membrane development strategies.

3.1.1. Additive loading in dope solution

Additive loading in the dope solution technique involves modifying the dope solution preparation in which organic or/and inorganic additives can be dispersed into the dope solution. This technique involves either physical or chemical interaction between the membrane material and the additive [60]. The former involves trapping of the additive within the membrane matrix. This facile technique is gaining acceptance due to its economic and environmental advantages, unlike the latter that involves transforming the chemical structure of the membrane material via chemical reaction [58]. However, controlling the additive density and dispersion remains its major challenge [60,61]. However, recent researches reported improvements and will be detailed herein.

Recently some researches demonstrated that by manipulating the phase separation process to achieve delay demixing, high organic additive density can be preserved. The combination of nonsolvent induced phase separation (NIPS) and vapor induced phase separation (VIPS) termed V-NIPS is commonly practiced for controlling membrane formation [60]. The former involves immediate immersion of the cast film into the nonsolvent bath for instantaneous membrane formation. Therefore, leads to the formation of a highly porous substructure with microvoids [62]. While the latter involves exposing the cast film to a controlled humid air environment, the slow penetration of the nonsolvent into the cast film leads to its precipitation, this resulted in the formation of a symmetric porous membrane with top skin layer [63].

V-NIPS approach recently gained more acceptance in the development of PEGylated membranes due to its ability in controlling both membrane structure and surface chemistry [64]. The slow nonsolvent penetration at the early stage of V-NIPS leads to rapid migration of additive chains towards nonsolvent and thus trapped within the matrix of the top immobile skin layer [64]. Therefore, improving its preservation within the membrane matrix and leads to an increase in surface hydrophilicity. Since membrane formation starts from the top skin layer as it is initially exposed to the nonsolvent. It further controls the kinetic property of the bottom sublayers [49].

Zhu et al. [64] developed PEGylated PSF membranes via the V-NIPS approach from PSF/PEGMA/DMAc solution. They found that the PEGMA density on the surface of the resulted membranes shifted regularly by adjusting the humid air exposure time of the liquid film. Water flux (WF) increased from 110 to 512 (L/m^2 h) as the exposure time raised from 0 to 5 mins. They attributed the performance to the slow demixing ability of VIPS that facilitated PEGMA chains migration and trappement onto the membrane surface.

Dehban et al. [65] studied the effect of humid air exposure time on the performance of polyphenyl sulfone (PPSU) ultrafiltration membranes via the V-NIPS approach. They reported that pore size and permeate flux of the resulted membranes shifted regularly by adjusting liquid film exposure time. By increasing VIPS time from 0 to 15 s, water flux and mean pore radius increased from 17.12 to 20.79 L/m^2.h and 0.96 to 1.44 nm respectively.

The recent advent and exploring of nanoparticles in membrane material development open up many opportunities in membrane filtration such as modification of high flux electrospun membranes and also the development of mixed matrix membranes etc. [2,66].

Recently, there is rapid research interest in electrospun membranes derived from their advantage of high flux due to large porosity with interconnected pore structures over the other types of membranes. However, their major drawbacks are poor mechanical properties, separation efficiency, and the vulnerability to fouling [2,66]. To overcome these challenges additive blending has been employed and found to be effective [2,67]. These efforts

brought about a facile method for efficient modification of electrospun membranes, in which the additive (nanoparticles) is blended with the polymer to form a completely dispersed blended dope solution for the electrospinning process. The fouling control mechanism of the membrane is depicted in Fig. 4.

Naseeb et al. [61] fabricated a nanofiber membrane using PAN, GO, and SiO_2. They used a different mass ratio of nanofillers (GO and SiO_2) and determined the optimum mass ratio that resulted in higher permeability and better fouling management. The nanofillers in the membrane matrix improved the wetting property of the hybrid membrane due to oxygen containing functional groups of the nanofillers and also the formation of the hierarchical structure caused by knots of GO and SiO_2 embedded inside the PAN nanofibers. The modification resulted in an improvement in flux from 2600 Lm^{-2} h^{-1} (pristine: pure PAN) to 3151 Lm^{-2} h^{-1} while maintaining 99.8% oil rejection.

Luo et al. [68] fabricated triangle-shape tri-bore hollow fiber (TBF) polyphenylene sulfone (PPSU) ultrafiltration membrane with different degrees of sulfonation via a dry-jet wet-spinning process using dual-layer tri-needle spinneret. Membrane with 2.5 mol% degrees of sulfonation demonstrate up to 84.1% flux recovery while for membrane with 1.5 mol% degrees of sulfonation was just 60.3% for the treatment of Tween 80 stabilized 50,000 ppm petroleum in water emulsion. They attributed the performance of the former to the improvement of surface hydrophilicity induced by the higher sulfonic group in its matrix.

3.1.2. Additive loading in the coagulation bath

A facile method for membrane surface wetting modification via a modified phase inversion process was explored by many researchers (Fig. 5). They reported improvement in membrane surface wetting properties for better fouling management and improved permeability [59]. This approach involves introducing additives into the nonsolvent coagulation bath that can induce micro-scale segregation of CN group (hydrophobic) of the polymer which resulted in its in-situ hydrolysis to COOH group (hydrophilic) and also increase the membrane surface roughness. Both of which play a vital role in fouling management [59,69].

Zhang et al. [59] modified the surface wettability of the PAN ultrafiltration membrane by introducing alkaline into the coagulation bath. Different concentrations of alkaline additive (NaOH) were added to the coagulation bath and found to play two important roles 1) inducing localize microphase inversion of PAN which leads to the formation of a rough surface and 2) inducing in-situ hydrolysis of CN group of PAN to a superwetting group of COOH. The resulted membrane from the highest alkaline concentration (10% wt./v) exhibited high permeability of 2270 Lm^{-2} h^{-1} bar^{-1} and a flux recovery ratio of 85%. A similar trend was reported by Peng et al. [70] they fabricated superwetting PAN ultrafiltration membrane via hydroxylamine-induced (hydroxylamine hydrochloride) phase inversion process. The resulted membrane exhibited up to 3806 Lm^{-2} h^{-1} bar^{-1} permeability.

Zhang et al. [69] grafted PVDF membrane surface with poly (acrylic acid) (PAA) using salt additive in the coagulation bath. It nucleates at the interface of the two liquids during the liquid-liquid demixing and induces the assembly of PAA-g-PVDF micelle aggregates around the salt seeds (nucleates). The micelle aggregates formed a micro/nanoscale hierarchical structured skin-layer onto the membrane surface which endowed it a superwetting property. The resulted membrane exhibited 1140 Lm^{-2} h^{-1} and 99.99% flux and oil rejection respectively.

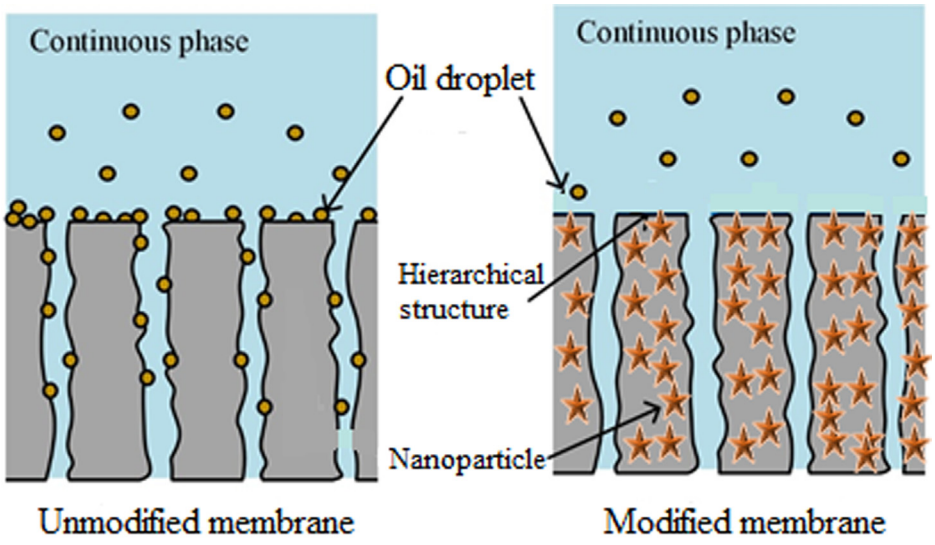

Fig. 4. Illustration of fouling mechanism for modified and unmodified membrane.

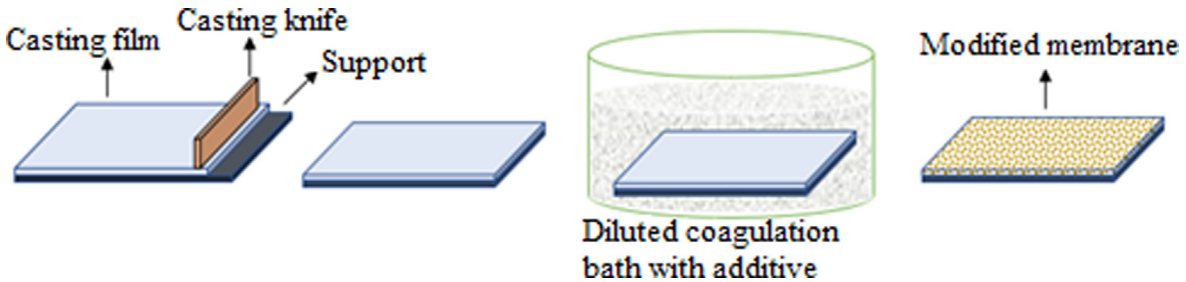

Fig. 5. Illustration of the general procedure of membrane surface hydrophilization via the introduction of additive in the coagulation bath.

3.2. Ex-situ membrane material development strategy

Organic and inorganic additives have been used in polymeric membrane development for fouling management via either physical or chemical coating [71,72]. The former involves the direct coating of the membrane surface with additive while the latter involves disrupting the polymer structure by introducing a functional group via techniques such as grafting and polymerization etc.

Membrane surface patterning is another technique of ex-situ membrane material development. It involves the creation of 3D patterns on the membrane surface that induces turbulence flow of the oily feed through the formation of fluid eddies and thus drive away oil droplets from accumulating on the membrane surface or within the membrane pores [37,46].

3.2.1. Physical surface coating modification

Additives are often coated on the membrane surface for improvement of fouling management as well as rejection performance. The coated layer dominates the membrane surface thereby reducing its interaction with oil droplets [24,54]. An illustration of the role of the coated layer in controlling membrane fouling is shown in Fig. 6.

Physical coating of membrane surface for fouling management has been widely reported by many researchers. Y. Du et al. [54] coated cellulose acetate membrane surface with chitosan (CS) and subsequently with TiO_2 nanoparticles (CST). The CS coating was achieved by vacuum filtration of the CS solution. The solution contained a copolymer network formed from the reaction of the amino group of CS with the aldehyde group of glutaraldehyde. The CST membrane was formed by vacuum filtration of TiO_2 nanoparticles solution with different concentrations (0.1, 0.4, and 0.7 mg/mL) onto the CS coated membrane. As the TiO_2 nanoparticles concentration increased, the TiO_2 nanoparticles layer continuously covered the CS surface resulted in decreased surface pore size and rendered surface roughness due to aggregation of TiO_2 nanoparticles. The TiO_2 nanoparticles completely covered the CS surface and filled the membrane pores at 0.4 mg/mL concentration beyond which no apparent change was exhibited. The membrane surface was found to be superhydrophilic and underwater superoleophobic (WCA = ~0° and UWOCA = 165.7°). Thus, exhibited permeability of ~6700 Lm^{-2} $h^{-1}bar^{-1}$ and 99.5% oil rejection. The performance was attributed to the surface roughness and improved wetting property induced by the TiO_2 coated layer.

Li et al. [73] achieved homogeneous dispersion of polydopamine (PDA) in tetrahydrofuran (THF)-Tris buffer mixture. The carboxyl functional groups are generated on the PDA structure due to the oxidization of THF-peroxide. The hydrophilic carboxyl functional groups in the mixture were immobilized onto polytetrafluoroethylene (PTFE) membrane surface via simply pouring it onto the surface, which also induced the formation of nanomicrospheres hierarchical structures. And transformed the membrane surface to superhydrophilic. Thus, exhibited 1461 Lm^{-2} h^{-1} water flux and 99% oil rejection.

Recently permeability and mechanical stability of graphene-based membranes have been improved via intercalating the graphene oxide (GO) sheets with nanoparticles. Thanks to the nanochannels formed as the result of the intercalation [74].

Peng et al. [74] used vacuum filtration technique to coat PVDF membrane surface with SiO_2 intercalated reduced GO (RGO) nanosheets and further improved the membrane surface property and stability by the introduction of DA onto the membrane surface. They reported improvement in permeability and stability of the membranes due to the nanochannels and interconnection of RGO sheets induced by SiO_2 intercalation and also the presence of DA. The different mass ratio of RGO to SiO_2 was employed. The resulted from 2 mg GO to 2.67 mg SiO_2 mass ratio demonstrated the highest performance with permeate flux of 118 Lm^{-2} h^{-1} and 98.5% oil rejection.

The surface wetting property of electrospun membranes can also be modified via direct coating with additives, as illustrated in Fig. 7. It can be done via immersion of the mat into the additive solution or via vacuum filtration of the additive solution onto the mat. This opens up the advent of Janus membranes with switchable wettability. One side hydrophobic while the other hydrophilic as it was covered by the additive [75].

Ahmed et al. [76] modified electrospun PVDF-co-hexafluoropropylene (PVDF–HFP) membrane by direct coating with cellulose that was regenerated from an ionic liquid of 1-ethyl-3-methimidazolium acetate ([EMIM]Ac). Cellulose solution (5 wt%) was stirred into the ionic liquid for 30 min at 85 °C, the solution was directly coated onto the PVDF–HFP mat and allowed

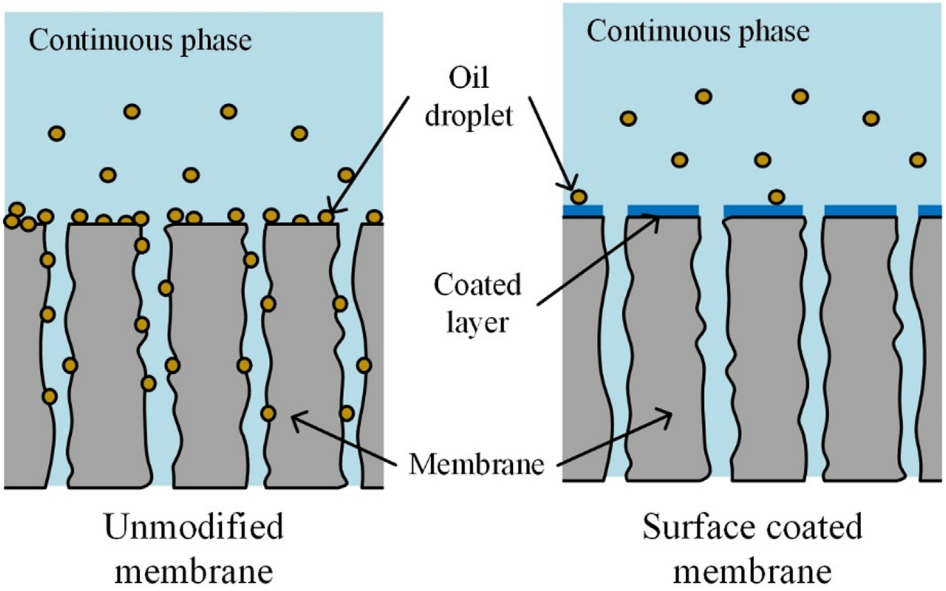

Fig. 6. Illustration of membrane fouling mechanisms for modified (via surface coating) and unmodified membranes during filtration of oily wastewater.

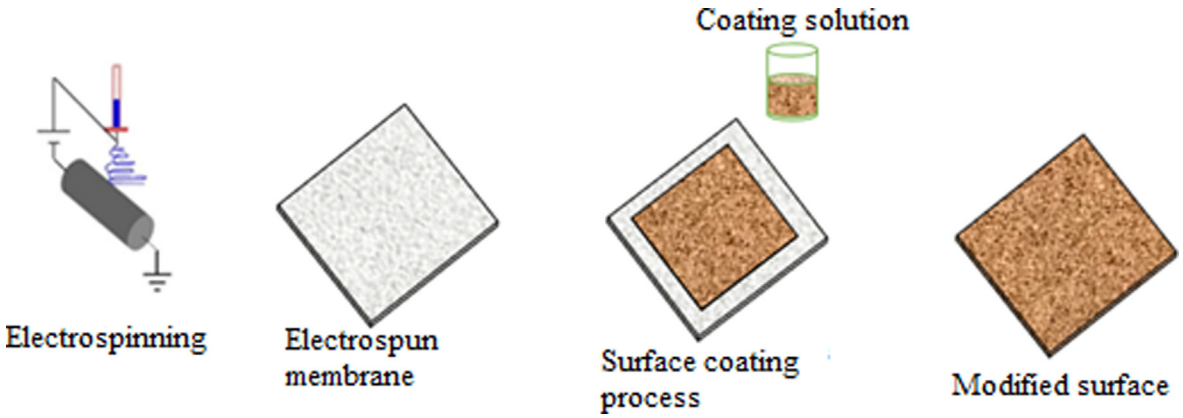

Fig. 7. Illustration of surface coating process of an electrospun nanofiber membrane.

to penetrate for 3 h at room temperature. This enabled the cellulose layer to have control over the porosity, pore size, wettability, mechanical and thermal properties of the membrane. The [EMIM] Ac was recovered from the membrane surface through overnight immersion in water, then 2 h stirred in boiled water and multiple deionized water washing. They reported permeability of 1780 Lm^{-2} h^{-1} bar^{-1} and 99.98% oil rejection. The performance was attributed to the surface wetting property induced by the hydrophilic cellulose and control of the pore size by the coated layer that lowered the pore size, thus enhanced the rejection of the oil droplets.

Recently Janus membranes with switchable wettability have been reported. The membrane demonstrated enhanced water flux at the hydrophilic side and oil flux at the oleophilic side. Therefore, no energy loss during backwashing/flushing for fouling management. Instead, the filtration will be reverse for flushing the adsorption droplets [76].

Jiang et al. [75] fabricated Janus PAN membrane through the following steps: PAN mat was first fabricated via electrospinning, then an ultrathin layer of carbon nanotubes (CNTs) was deposited onto one side of the PAN nanofiber mat via vacuum filtration at ΔP of 0.07 MPa. The membrane exhibited up to 12,000 Lm^{-2} h^{-1} bar^{-1} oil permeability and 80,000 Lm^{-2} h^{-1} bar^{-1} water permeability on the other side. Both sides maintained up to 99% droplets rejection. The performance of Janus membranes is an economic breakthrough for membrane technology. It also gives the impression of the development of fouling free membranes in near future.

3.2.2. Chemical surface coating modification

Unlike physical surface coating where disintegration and leaching of the coated layer remain the major challenge, while the chemical surface coating remains permanent [77]. Recent researches demonstrated improvement in its major challenges of environmental and economic sustainability [78].

Yuan et al. [77] used copper (I) catalyzed azide-alkyne cycloaddition (CuAAC) reaction to graft different molecular weights of propargyl PEG (pro-PEG) onto azide-functionalized Polysulfone (PSF) membrane surfaces. $PSF_{0.45}$-g-PEG_{120} (0.45 degree of azide group functionalization and 120 g/mol PEG grafting density) membrane shows the best performance for the treatment of Dagang oilfield oil in water emulsion with a flux of 120 Lm^{-2} h^{-1}, 99.9% oil rejection, and 95% flux recovery ratio. They attributed the enhanced performance to pro-PEG layer grafted and surface roughness rendered by the grafting process.

Prince et al. [78] modified the wetting property of polyethersulfone (PES) hollow fiber membrane using carboxyl, hydroxyl, and amine modified graphene attached PAN-co-maleimide (G-PANCMI). WCA and UWOCA were modified from 63.7° to 22.6° and from 43.6° to 112° respectively. Thus, resulted in 43% improvement in water permeability while maintaining up to 99% oil rejection. They attributed the improved performance to the presence of amine (—NH_2) and acid (—COOH) groups attached to the nanographene sheets in the G-PANCMI matrix that was coated onto the PES surface.

Yuan et al. [79] coated alkaline treated PVDF membrane surface with hydrogel using interfacial polymerization based on the thiol-epoxy ring-opening reaction between pentaerythritol tetrakis (3-mercaptopropionate) (PETMP) and diethylene glycol diglycidyl ether (PEGDGE). The performance of the membrane was evaluated by treating SDS stabilized n-dodecane oil in water emulsion in a cross-flow filtration setup at ΔP of 0.02 MPa. The membrane maintained up to 2000 $Lm^{-2}h^{-1}bar^{-1}$ permeability even after the 4th filtration cycle (18 h). The hydraulic performance was attributed to the hydrogel layer that completely covered the hydrophobic PVDF surface and induced the surface with superwetting property (UWOCA = 152°, and WCA = < 3°).

Recently, some researches demonstrated the effectiveness of combining two or more surface wetting modifiers for membrane fouling management. Liu et al. [80] turned hydrophobic polyvinylidene fluoride (PVDF) membrane to hydrophilic by coating with CS–SiO_2–glutaraldehyde (GA) layer. It was modified by simple immersion into a CS–SiO_2–GA solution and sonicated for 5 min, air dried and 30 min treatment with 0.1 M of NaOH solution, followed by washing with deionized water. The superwetting solution was made by preparing a CS–SiO_2 solution in which the tetraethoxysilane (TEOS) was hydrolyzed to form silanol groups of SiO_2 that further reacted with hydroxyl groups of CS. And further exposed to GA which resulted in cross-linking of CS with GA through the reaction of aldehyde groups of GA with the amino groups of CS. The membrane exhibited superwetting property with UWOCA = 150°. The improvement was attributed to the CS–SiO_2–GA coated layer due to the presence of silanol and amino groups in SiO_2 and CS respectively, as well as the hierarchical structure rendered by the coated layer.

Assembly of dopamine (DA) onto the membrane surface for improvement of surface wetting property is in a state of rapid development. It can be deposited under both acidic and alkaline conditions, thanks to the presence of functional groups such as catechol and amine in the dopamine matrix [81].

Xiang et al. [81] fabricated PVDF membrane using non-solvent induced phase separation and modified its wetting property via immersion into 2 g/L DA solution to allow it self-polymerization. They studied the effect of the immersion period on the degree of polymerization. The highest performance was achieved for an immersion period of 24 h with 1991 $Lm^{-2}h^{-1}$ and 96.1% water flux and flux recovery respectively. The performance was attributed to

the higher assembly of catechol and amine functional groups induced by DA assembly onto the membrane surface.

Luo and Liu [82] recently developed a facile method of depositing PDA onto a polymeric membrane in an acidic environment (pH = 5.0) via sodium periodate oxidation instead of traditional O_2 autoxidized in a basic buffer solution. They modified the PVDF surface by depositing a PDA layer oxidized by sodium periodate in an acidic condition (pH = 5.0). And thus exhibited 8,649 Lm^{-2} h^{-1} and 100% flux and oil rejection respectively.

Ding et al. [40] systematically accelerated and controlled PDA onto polypropylene (PP) membrane surface via the Schiff base reaction between amine groups of γ-aminopropyl triethoxysilane (γ-APTS) and benzoquinone groups of PDA. The membrane surface wetting property was improved due to surface roughness and the presence of amino and hydroxyl groups in the coated layer. The membrane maintained a water flux of 120 $Lm^{-2}h^{-1}$ against ~40 Lm^{-2} h^{-1} for the pristine.

The chemical surface coating has been practiced also for the development of electrospun membranes and found to be promising [83].

Zhang et al. [83] functionalized electrospun aminated PAN fiber by GO and reported a high flux of 10,000 $Lm^{-2}h^{-1}$ while maintaining oil rejection of 98%. The membrane was obtained by the introduction of NH_2 groups onto the electrospun PAN fiber surface through reaction with diethylenetriamine. Acylation and nucleophilic reactions were used to modified APAN with GO. High flux performance of GO/APAN membrane was attributed to the large porosity of electrospun membranes while high rejection and good antifouling performance were attributed to the carboxyl and hydroxyl groups induced by GO sheets that are connected to the APAN fibers.

3.2.3. Membrane surface patterning modification

Membrane surface patterning involves the creation of 3D patterns onto the membrane surface that induces turbulence flow of the oily feed through the formation of fluid eddies that drive away oil droplets from accumulating on the membrane surface or within the membrane pores as illustrated in Fig. 8 [37,46]. Membrane surface patterning is a "green approach" because it does not involve the use of any harmful chemicals [46]. It also increases the membrane effective area and thus enhances the hydraulic throughput [37,39].

Template based micromolding and direct printing are the two main techniques of membrane surface patterning. The former involves replication of pattern features of a master mold onto the membrane surface via either solution-based or embossing-based micromolding while the latter is a structured layer by layer deposition of materials to form a 3D object based on a computer design model via either inkjet or 3D techniques [46,84]. Membrane surface patterning is gaining more attention due to its proven economic and environmental sustainability [85,86].

Al-Shimmery et al. [37] fabricated membrane selective layer separately using nonsolvent induced phase separation and subsequently deposited it onto a 3D printed patterned and flat porous support via vacuum filtration. The membrane selective layer follows the pattern features of the 3D printed patterned support and thus, render patterns onto its surface. The patterned membrane outperformed the flat by 30% in cross flow filtration of synthesized sunflower oil in water emulsion. It also maintained up to 52% of its initial permeability even after the 5th filtration cycles using only water as the cleaning agent while the flat completely fouled after the first filtration cycle. The performance was attributed to the turbulence flow of the oily feed induced by the surface patterns through the formation of fluid eddies which continuously detach and drive away oil droplets from accumulating on the membrane surface.

Sun et al. [87] patterned hydrophobic and oleophilic isotactic polypropylene (iPP) membrane surface via a facile method that combined embossing micromolding with thermal induced phase separation process (TIPS). The iPP membrane was fabricated on a hot steel plate with a controlled temperature of 180 °C, the liquid membrane was pre-evaporated to reach a semi-gelation state, thereafter a modified patterned roller that was pre-heated to a controlled temperature of 180 °C was rolled on the liquid membrane to replicate the patterned features on its surface. The patterned membrane was quenched in a cooling bath containing deionized water and further treated with ethyl acetate for 48 h and ethanol for 24 h followed by air dried at room temperature. The performance of the patterned membrane was evaluated in the treatment of Span 80 stabilized water in oil emulsion, the patterned membrane showed a high oil flux of 1108 Lm^{-2} h^{-1}.

The performance of various strategies of polymeric membrane development for fouling management and performance improvement have been fully discussed in the aforementioned subsections 1 and 2 of section 3, while Table 1 presents the summary of their performances.

4. Ceramic membrane material development

Ceramic membranes possess some advantages that make them favorable for many challenging applications that require excellent thermal, chemical, and mechanical stability [98]. However, the high cost of materials such as Al_2O_3, ZrO_2, SiO_2, and TiO_2 for its formation hinders their widespread application in industries, including for oily wastewater treatment, despite their advantages. Preparing low-cost ceramic membranes is then the main future challenge [99].

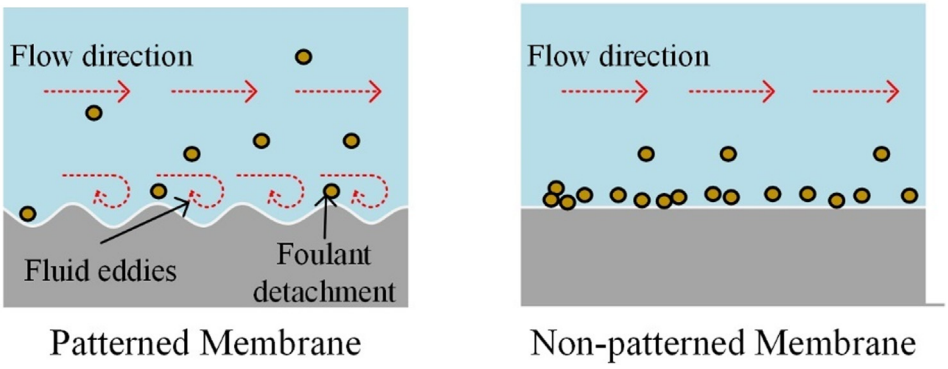

Fig. 8. Comparison between flat and patterned membranes in terms of fouling management.

Table 1
Performance of modified polymeric membrane for oily wastewater treatment.

Membrane	Treatment	Oil concentration in the Feed (ppm)	WCA (°)	UWOCA (°)	Rejection (%)	Permeability (Lm^{-2} h^{-1} bar^{-1})	Pressure (bar)	Ref.
CS-SiO_2-GA@PVDF	Surface coating and NaOH treatment	1000	0	150	99	–	0.3	[80]
PES-G-PANCMI	Surface grafting	200	23	112	99.7	730	1	[78]
PPSU-TBF	Sulfonation and dry-jet wet spinning method	50,000	68	–	97.3	160	1	[68]
NiCo-LDH/PVDF	Polymerization and hydrothermal method	10,000	0	157	99.7	–	–	[88]
CNH/CC	Hydrothermal process	–	–	152.5	98.5	–	–	[72]
PP/DA/γ-APTS	Schiff base reaction and polymerization	20,000	6	154	99.9	–	–	[40]
PVDF@PDA@SiO_2	Phase inversion and polymerization	990,100	0	154	94	572	0.8	[71]
CC@PVDF	Leaching of CC suspension onto PVDF	500,000	Switchable		99.2 99.3	89.4 100	0.85	[89]
PVDF/PHFBM-PEGMA-PMTAC	Polymer blending and radical polymerization	901.03	85.2	129.9	100	198	0.5	[90]
PVC-Bentonite	Mixed matrix	35,000	55.1	–	97	93	2	[91]
PES - PVP - HNT-HFO	Mixed matrix	1000	5	–	99.7	650	1	[92]
3D printed patterned PES	3D printing and vacuum filtration deposition	5000	63	–	96	16	1	[37]
PVDF/RGO@SiO_2/PDA	Intercalation and surface coating	9900	0	134.3	98.5	132.6	0.89	[74]
PVDF-PAA-ZnO	Cold plasma-induced graft-polymerization	–	0	–	85	832.52	1	[93]
$PSF_{0.45}$-g-PEG_{120}	Surface grafting and CuAAC reaction	901	20	–	99.9	1,200	0.1	[77]
iPP	Embossing micromolding and thermal induced phase separation	991,300	154	–	–	1244.9	0.89	[87]
PEN/HNTs@GO_1-PDA	Intercalation and surface coating	9900	0	142.5	99.5	1270.3	0.89	[94]
Cellulose @ PVDF-HFP_{EN}	Surface coating	100,000	0	169	99.98	1780.8	0.65	[76]
PVDF/PDA	Polymerization	10,000	0	170	99.82	1862.5	0.8	[95]
PVDF@PDA	thermal induced phase separation and polymerization	3980	86	111.8	98.5	2600	1	[36]
PDA/PTFE	Polymerization	10,000	23.4	165.6	99	2981.6	0.49	[73]
PDA @ PVDF	Polymerization	3360	53	152	98	4459.6	0.89	[81]
PVDF-PDH	Polymer blending	9900	0	156	98	6350	0.6	[96]
CNTs @ PAN_{EN}	Surface coating	83,300	Switchable		99.2 99.7	12,000 8000	0.7	[75]
SiO_2@PVDF	Delayed phase inversion	982,700	157	0	99.9	17888.9	0.9	[97]
PDA-SP@PVDF	Polymerization	10,000	0	154	100	22760.5	0.38	[82]
PAN-GO-SiO_2	Polymer blending	2000	7	155	99.8	31,510	0.1	[61]
PAN_{EN}	Electrospinning and electrospraying	10,000	0	162	99.93	515,200	0.01	[24]

Recently researches are focused on the development of low-cost ceramic membranes from natural mineral-based materials that have high percentage composition of Al_2O, SiO_2, TiO_2, and ZrO_2 such as zeolite, fly-ash, kaolin, and others. It was found that the utilization of these natural mineral-based materials effectively resulted in producing low-cost ceramic membranes while maintaining excellent filtration performance [98–100]. Therefore, is a breakthrough for wider adoption of ceramic membranes in industries as the menace of high cost has been tremendously reduced. Another advantage of utilizing these low-cost materials is their environmental sustainability. For example, fly–ash is a by-product of coal combustion in thermal power plants and its direct disposal has been a long term environmental issues. Therefore, utilizing it for ceramic membrane fabrication is a move for better environmental sustainability apart from its economic benefit [101].

Modifications of ceramic membranes for enhanced performance have been explored and showed encouraging performance [102]. Recently some reports evaluate the performance of low-cost and modified membranes in oily wastewater separation and reported an excellent performance as depicted in Table 2.

Fang et al. [103] fabricated a low-cost fly-ash based ceramic membrane and reported permeate flux of 159 $Lm^{-2}h^{-1}$ while maintaining 95% oil rejection in R100 stabilized crude oil in water emulsion filtration.

Zhu et al. [100] also used fly-ash and fabricated mullite hollow fiber support which was then coated with a thin hydrophilic TiO_2 layer via dip-coating. They reported permeability of 150 Lm^{-2} h^{-1} bar^{-1} and 97% oil rejection. They attributed the performance to the thin hydrophilic TiO_2 selective layer which was deposited onto the fly-ash based mullite hollow fiber support. The fly-ash based mullite fiber tremendously reduced the amount of the required TiO_2 in membrane fabrication and thus reduce the fabrication cost.

Emani et al. [104] fabricated a low-cost kaolin based ceramic membrane with the following compositions 40% kaolin, 15% quartz, and 25% calcium carbonate with additives as the balance. The membrane exhibited 79.704 Lm^{-2} h^{-1} water flux and 98.52% oil rejection.

The surface wetting property of a ceramic membrane can also be enhanced by using wetting modifiers. Chang et al. [105] modified the surface wetting property of Al_2O_3 tubular ceramic membrane by coating with TiO_2 nanomaterial via in-situ precipitation method. They reported the transformation of WCA from 33° to 8°. Thanks to the presence of wetting modifier (TiO_2) layer, the transformation accounted for a 52% improvement in membrane flux (of 335 Lm^{-2} h^{-1} for the modified and 220 Lm^{-2} h^{-1} for the unmodified).

Zhang et al. [102] also improved Al_2O_3 membrane surface wetting property by coating with TiO_2 nanorod array using magnetron sputtering and hydrothermal oxidation methods. The modified membrane exhibited WCA = 0° and UWOCA = 150°. Thus, maintained up to 99.1% oil rejection and 41.8 Lm^{-2} h^{-1} flux in a gravity driven filtration set-up.

Table 2
Performance summary of ceramic membranes for oily wastewater separation.

Membrane based material	Pore size (μm)/porosity (%)	Oil concentration in the feed (ppm)	Permeability (Lm^{-2} h^{-1} bar^{-1})	Rejection (%)	Pressure (bar)	CFV (m/s)	Ref.
Fly-ash	0.77/-	75	159	95	0.5	0.67	[103]
Kaolin	2.16/37.4	400	38.5	98.52	2.07	0.1344	[104]
Quartz	0.309/53	200	–	99.98	0.69	–	[106]
Zirconia oxide	0.1/-	100	120	–	2	2	[107]
Fly-ash	0.11/-	200	150	97	0.025	0.15	[100]
α-Al_2O_3	–	995,000	1926	98.6	1	Dead end filtration	[31]
Al_2O_3	0.98/29.4	32,300	–	99.1	Gravity driven filtration		[102]

5. Metallic membrane material development

Metallic membrane material development has been reported with encouraging performance [108–112] as summarized in Table 3. The application of metallic mesh-based membranes can potentially become a major industrial breakthrough thanks to ultra-low ΔP required merely through the hydrostatic pressure (driven by gravity). It can be modified via simple mechanisms such as dip-coating, spray coating, layer by layer (LBL), etc. Additives such as TiO_2, ZnO, HNTs, CNTs, SiO_2, $BiVO_4$, UiO-66 are often employed to modify the surface wetting property of the mesh and found to be effective [113–115].

Lin et al. [116] fabricated a modified metallic membrane by coating copper mesh with TiO_2 nanomaterial. The membrane exhibited superwetting property with WCA = 0°, UWOCA = 160.8°, 300 Lm^{-2} h^{-1} water flux, and 99.99% oil rejection. The performance was attributed to the hierarchical structure, superwetting property, and decreased pore size induced by TiO_2 coated layer.

Gunatilake and Bandara [108] modified a stainless–steel mesh by depositing a rough superhydrophilic TiO_2 nanofibers layer onto the surface via spray deposition technique. After spraying the dispersed solution of TiO_2 nanofibers in ethanol, the coated mesh was sintered for 45 min at 450 °C to achieve adequate integration of the nanofibers onto the mesh surface. The coated mesh exhibited superwetting property with WCA = 0° and UWOCA = 162°. While maintaining total oil rejection (100%) in gravity driven filtration of engine oil in a water mixture. A similar approach was also reported by Zhou et al. [109]. They deposited W, N co-doped TiO_2 nanobelts (WNTNBs) onto titanium mesh. They used the immersion route and subsequent calcination at 550 °C for 2 h. The membrane exhibited superwetting with WCA = 0°, UWOCA = 156°, and maintained up to 98.5% oil rejection even after the 20th filtration cycles.

Jiang et al. [57] coated a stainless–steel mesh with 3-aminopropyl triethoxysilane (SCA), PEI and trimesoyl chloride (TMC) in sequence with SiO_2 nanoparticles layer accompany via covalent layer-by-layer grafting (LBLG) method. The membrane exhibited WCA = 159° and UWOCA = 0°. And maintained up to 99.88% water rejection even after 30th filtration cycles. They attributed the performances to the amidation reaction between amino groups of PEI and palmitic acid which rendered the superhydrophobicity property of the surface while superoleophilicity property of the surface was attributed to the hierarchical structure and long carbon chain of the coated layer.

Liu et al. [117] coated stainless steel mesh surface with CS–SiO_2–GA layer using the spraying technique and reported WCA = 0° and UWOCA = 159°. Thus, maintaining up to 99% oil rejection even after the 20th filtration cycles. The performance was attributed to the presence of superhydrophilicity modifiers (amino, hydroxyl, silanol groups) in the coated layer matrices and the hierarchical structure formed due to cross-linking of amino groups of CS with the aldehyde groups of GA, and hydrolysis of TEOS that formed silanol groups of SiO_2 which reacted with the hydroxyl groups of CS.

Li et al. [118] fabricated a metallic membrane for oily wastewater treatment by spray coating of palygorskite and polyurethane mixture onto copper mesh substrate and reported up to 99% oil rejection even after the 50th filtration cycle and attributed the

Table 3
Summary of performance of modified metallic membrane for oily wastewater treatment.

Membrane formation	Method description	Oil concentration in the feed (ppm)	Flux (L m^{-2} h^{-1})	WCA (°)	UWOCA (°)	Rejection (%)	Ref.
TiO_2 nanofibers coated stainless steel mesh	Spray deposition	272,700	–	0	162	100	[108]
SCA-PEI-TMC-SiO_2 coated stainless steel mesh	LBLG	500,000	–	159	0	99.88	[57]
PDDA-HNTs coated stainless steel mesh	LBL	500,000	–	0	151.5	97	[120]
Nanosecond laser-induced oxide on stainless steel mesh		500,000	–	0	164	96	[122]
CS-SiO_2-GA coated stainless steel mesh	Spray deposition	200,000	–	0	159	99	[117]
WNTNBs coated titanium mesh	Solution route and calcination method	300,000	–	0	156	98.5	[109]
TiO_2 coated stainless steel mesh	liquid phase deposition	500,000	–		152	99	[112]
SiO_2 -PU coated stainless steel mesh	Spray coating	500,000	–	0	154	98.8	[115]
Ag coated copper mesh	Galvanic exchange reaction	500,000	–	0	153.5	99.5	[121]
TiO_2 - PU coated stainless steel mesh	Spray coating	500,000	–	<5	156.5	97.5	[119]
$BiVO_4$ coated stainless steel mesh	Immersion	375,000	–		154	98.6	[123]
TiO_2 coated stainless steel mesh	Sol-gel and dip-coating method	500,000	Water: 13,554 Oil: 7281	Switchable		99.9 99.9	[12]
UiO-66 coated stainless steel mesh	Immersion	500,000	12.7×10^4	25	150	99.99	[124]

performances to the coated layer and decreased in mesh pore size induced by the coated layer. Similarly, Li et al. [115] also used this facile method of direct spraying of surface wetting modifiers to modify the surface of a stainless steel mesh. They successfully dispersed waterborne polyurethane (PU) and SiO_2 nanoparticles into acetone, the solution was directly sprayed onto the stainless steel mesh. The modified membrane exhibited hierarchical structure and hydrophilicity induced by waterborne PU and SiO_2 nanoparticles layer, and thus maintained up to 98% oil rejection even after the 50th filtration cycles. Li et al. [119] also used the same procedure but using TiO_2 instead of SiO_2 and reported 97.5% oil rejection even after the 40th filtration cycles.

Hou et al. [120] induced hierarchical structure and superhydrophilic property onto stainless steel mesh surface by deposition of poly (diallyl dimethylammonium chloride) (PDDA) and HNTs in sequence via layer by layer assembly. The membrane exhibited 97% oil rejection even after the 20th filtration cycles.

Li et al. [121] reported up to 99.5% oil rejection even after the 45th filtration cycles using silver (Ag) coated copper mesh that was fabricated via galvanic exchange reaction and attributed the performances to the hierarchical structures induced on the mesh surface due to aggregation of Ag tress and also the hydrophilicity property of Ag coated layer. The copper mesh was initially treated in sequence with ethanol, hydrochloric acid, and deionized water to remove impurities (metallic oxide) followed by a galvanic reaction which lasted for 30 s via immersion into 0.08 M $AgNO_3$ solution, then the membrane was rinsed with deionized water and dried. The membrane exhibited superwetting property with WCA = 0° and UWOCA = 153.5°.

6. Conclusions and future perspectives

This review detailed recent membrane material development specifically towards the treatment of oily wastewater. Most studies report almost complete oil rejection and improved throughput. Permeability widely ranging from 16 Lm^{-2} h^{-1} bar^{-1} up to 515,200 Lm^{-2} h^{-1} bar^{-1} for polymeric membranes (Table 1), 38.5 to 1926 Lm^{-2} h^{-1} bar^{-1} for the ceramic membranes (Table 2), and limited hydraulic report for the metallic membranes (Table 3). The large discrepancy in the hydraulic performances can be ascribed by high variation in the membrane material development, feed composition, and characteristics as detailed in Tables 1, 2, and 3. Therefore, make it very challenging to make a direct comparison. Another important aspect is the filtration test method, in which cross-flow and gravity driven have been applied. For the gravity driven system, ΔP is very low and the membrane is not fully compacted yet, thus lead to very high permeability. The variability of the applied feed and the testing methods can be understood since each study was customized to address a certain issue. However, a more universal method is required to allow universal comparison.

Ceramic membranes offer lower permeability than the polymeric ones, due to their high membrane thickness which is directly correlated with permeability. Nonetheless, such material must be tailored to suit their advantages of treating feed at high temperatures, i.e., refinery process water.

Application of membrane surface patterning and Janus membranes for oily wastewater treatment is less matured. Tailoring this membrane material development would be one of the most promising approaches to be explored. Surface patterning maximizes in-situ cleaning efficiency, thus minimizing the membrane fouling tendency. While in Janus membranes, the accumulated foulant will be flushed by simply reversing the filtration. These two strategies of membrane materials development provide both economic and environmental benefits and need to be explored further to its full potential.

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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.

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