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Membrane fouling and fouling mitigation in oil–water separation: A review

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ABSTRACT

Oil–in–water emulsion separation is a challenging separation process in industries due to the large volumes of emulsified oil produced. Membrane technology has become more important in emulsified oil treatment due to its high separation efficiency and straightforward operational procedure. Up to now, microfiltration and ultrafiltration are the most utilized membrane processes for oil-in-water emulsion separation. However, the membrane for emulsified oil deals with several problems. The presence of flexible oil droplets creates a fouling tendency as well as a reduction in the overall rejection of the membrane. These phenomena have attracted researchers to develop antifouling strategies for membranes, including membrane materials engineering and membrane operational management. This review highlights the current progression of membrane used for oil-water separation started from membrane materials perspectives, followed by the discussion on fouling and fouling mitigation during oil-water separation. The review is concluded by future perspectives of membrane technology for oil-water separation.

1. Introduction

Oily wastewater is considered as one type of wastewater that is challenging to separate and can harm the environment. Many types of industries produce oily wastewater, such as refinery, metallurgical, textile, leather, pharmaceutical, and food and beverage industries. In addition, residential activities also produce oily wastewater. Up to now, oil refineries are one of the largest oily wastewater producers, and the production tends to increase every year [1,2]. Oily wastewater from refinery industries is often referred to as oil-produced water. Produced water represents the biggest wastewater in most oil and gas production. Basically, produced water contains water from the sea or freshwater trapped with oil and gas inside the reservoir and residual water used for production. Vast amounts of produced water are discharged into the sea; hence regulation regulates a maximum of 30 mg l^{-1} of the oil-water emulsion that can be found in the ocean [3,4].

The more stringent regulation has required industries to take action by applying different technologies to treat oily wastewater. Techniques for oily wastewater treatment include gravity or centrifugal force, electrostatic precipitation, cyclones, floatation, physical or chemical demulsification, heat treatment, and adsorption. The techniques mentioned above have been utilized in many industries; however, these technologies suffer from relatively low separation efficiency, especially for emulsions with small drop sizes, high cost, and complex operational steps [1,5,6]. In recent years, membrane separation techniques have been developed and considered an effective technique for separating oil from water, especially small droplets-containing wastewater [2,4,6,7]. In addition, the membrane technology offers low cost and easy operation [8–10]. In general, membranes for oily wastewater treatment are fabricated from polymeric as well as inorganic materials. Polymeric membranes are cheap and easy to scale up, however, their fouling susceptibility has motivated researchers to develop different kinds of modifications of polymeric materials. On the other hand, inorganic membranes from ceramic and metals offer good separation performances with some challenges such as high cost and relative challenging to scale up.

In addition, research on the developments of antifouling strategies, such as polymer modification, polymer blending, and surface modification, is still actively conducted by researchers. Antifouling strategies are required to maintain membrane performance during the oil-water separation process. Consequently, deep knowledge of membrane

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Nomenclature

APTES	3-Amino-propyltriethoxysilane
BOD	Biological oxygen demands
CA	Cellulose acetate
CNTs	Carbon nanotubes
COD	Chemical oxygen demand
CP	Cellulose propionate
DAF	Dissolved air floatation
DEA	Diethanolamine
DOPA	3,4-Dihydroxy-L-phenylalanine
EDTA	Ethylenediamine tetraacetate
FAS	Fluoroalkylsilane
FO	Forward osmosis
FPEOAA	Perfluoroalkyl polyethoxy acetic acid
GO	Graphene oxide
HEA	Hydroxyethyl acrylate
HMO	Hydrous manganese oxide
LMH	Liters per square meter per hour
MD	Membrane distillation
MEA	Monoethanolamine
MF	Microfiltration
MMMs	Mixed matrix membranes
MOFs	Metal organic frameworks
MWCNTs	Multiwalled carbon nanotubes
NF	Nanofiltration
O/W	Oil–in–water
OTMS	Octadecyltrimethoxysilane
PA	Polyamide
PAM	Polyacrylamide
PAN	Polyacrylonitrile
PANEN	Polyacrylonitrile electrospun nanofiber
PANI	Polyaniline
PBI	Polybenzimidazole
PCP	Porous coordination polymer
PDA	Polydopamine
PDMSPEG	Poly(dimethylsiloxane) and poly(ethylene glycol)
PEG	Polyethylene glycol
PEGDA	Poly(ethylene glycol) diacrylate
PEI	Polyethylenimine
PES	Polyethersulfone
PFTS	Perfluorode cyltriethoxysilane
PK	Polyketone
P(MMA-co-GMA)	Poly(methyl methacrylate-co-glycidyl methacrylate)
POME	Palm oil mill effluent
PSf	Polysulfone
PSMA	Poly(styrene-co-maleic anhydride)
PTFE	Polytetrafluoroethylene
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
PVP	Poly(vinyl pyrrolidone)
rGO	Reduced graphene oxide
RO	Reverse osmosis
SDS	Sodium dodecyl sulfate
SPEEK	Sulfonated poly(ether ether ketone)
SWCNTs	Single-walled carbon nanotubes
TA	Tannic acid
TDS	Total dissolved solid
TMP	Trans membrane pressure
UF	Ultrafiltration
VSEP	Vibratory shear enhancing process
W/O	Water–in–oil
ZIF	Zeolitic imidazolate framework

fouling by oil drops and the choice of suitable antifouling strategy is very important. Table 1 summarizes the review papers published by different researchers on membrane processes for oily wastewater treatment. Most of the reviews emphasis on membrane fouling as a sub-section of their work or if it is focused on membrane fouling then the discussion on fouling mitigation focuses on membrane materials development. Hence, this review focuses on fouling and fouling mitigation by various techniques during oil-water separation by membranes processes. In terms of membrane processes, microfiltration and ultrafiltration are two types of membrane processes that have been actively investigated by researchers for the oil-water separation process, as depicted in Fig. 1. The Scopus database shows that microfiltration and ultrafiltration are investigated more frequently for oil-water separation than other membrane processes. This data indicates the potentiality of these two processes in separating oil from water. Therefore, this review aims to discuss the current techniques for oil-water separation, including membrane, followed by focused perspectives on materials development for membranes applied for oily wastewater treatment. Fundamental of fouling of membranes by oil-water emulsions and mitigations approach to minimize fouling are discussed followed by the industrial application perspectives and future outlook.

2. Characteristics of oily wastewater and recent techniques for its treatment

In general, oily wastewater can be classified into three categories, such as free-floating oils, unstable oil-in-water emulsions, and stable emulsified oils. Free-floating oils have oil droplets larger than 150 μm and can be found as spilled oils on the ocean. Unstable or dispersed oil-in-water emulsions have oil drops around 20–150 μm and are easy to coalesce. On the other hand, emulsified oils have oil drops with a diameter under 20 μm and are usually stable because of surfactants. Oily wastewater consists of various chemicals compounds, and the compositions depend on the process production in each industry. The composition of emulsified oily wastewater is complicated because it can contain mineral, vegetable, or synthetic oil, fatty acids, emulsifiers (anionic and non-ionic surfactants), corrosion inhibitors (amines), bactericides, and other chemicals. Oily wastewater can also contain petroleum hydrocarbons, phenolic compounds, and naphthenic acids.

However, the composition of oily wastewater can be classified as organic and inorganic components. The inorganic component mainly consists of inorganic oils, gasoline, heavy metals, oils sludge, solvents, and particulate matter. The organic components in oily wastewater mainly consist of petroleum hydrocarbons with nickel, cadmium, lead, vanadium, and organometallic complexes as minor elements. Organic and inorganic components exist as dispersed and free-floating oils, respectively [17]. Each oil refinery has specific processes, and therefore wastewater produced from its production will have a different composition. For instance, petroleum refinery wastewater can include oil and grease, phenols, sulfides, ammonia, suspended solids, cyanides, nitrogen compounds, and heavy metals, e.g., chromium, copper, and zinc.

There are many methods to treat oily wastewaters, as summarized in Fig. 2. The appropriate treatment method depends on the oil droplet size in wastewater. The treatment methods can be classified into physical, chemical, and biological techniques. Dissolved Air Floatation (DAF) is a physical method to remove the oil by adding μm-size bubbles and floatation. DAF is used for removing free and dispersed or unstable oils [12,18,19]. Other physical techniques include gravity separation and skimming, suitable treatment methods for removing free oil from wastewater. In addition to DAF, froth floatation using additional

Table 1
Summary of important review papers on membrane processes and membrane fouling during oily wastewater treatment.

Focus of the paper	Key contribution	Ref.
Oily wastewater treatment	A wide survey on various membrane processes for oily wastewater treatment and surface modification of membrane to mitigate fouling	[6]
Microfiltration for oily wastewater treatment	A comprehensive review on the application of microfiltration for oily wastewater treatment. A brief overview of membrane fouling and mitigation were provided with the focus on membrane material modification.	[11]
Oily wastewater treatment	Review on various membrane processes applied for oily wastewater treatment and techniques to quantify membrane fouling	[12]
Oily wastewater treatment	Review on fouling mechanism and membrane material modification to alleviate fouling by oily wastewater	[13]
Oily wastewater treatment	Primarily focusing on a deep study on membrane materials applied for oily wastewater treatment and modification to alleviate membrane fouling	[14]
Oily wastewater treatment	A comprehensive review on hydrophilic membrane materials development and modifications to mitigate fouling during oily wastewater treatment	[15]
Emulsified oil treatment	Review on membrane fouling by oily wastewater with emphasis on how the properties of emulsion affect the separation performance. In addition, fouling mitigation based on membrane materials development is limited to surface modification	[16]
Oily wastewater treatment	A comprehensive review on membrane fouling mechanism and mitigation from the perspectives of membrane materials and operational management	This review

surfactants and aeration is also a physical method applied for floating oil-saturated bubbles [20]. While DAF can achieve 95% oil removal efficiency, the froth floatation can only attain 55% removal efficiency with a much higher oil concentration. Another physical technique for oil-water separation is adsorption using porous materials or adsorbents [12,18]. One challenge for adsorption is to reduce the chemicals used for the regeneration process and find suitable adsorbents for a particular separation process.

Chemical demulsification techniques for oil-water separation are solvent extraction [21], a combination of extraction-fractional distillation [21], coagulation-flocculation [22,23], and the Fenton process [24]. Solvent extraction can remove free oil or dissolved oil-containing wastewater until 93% removal efficiency, while if combined with fractional distillation could achieve more than 95% removal efficiency. The conventional coagulation-flocculation process is applied by attaching a coagulant to create flocs that sink and can be removed. However, the separation efficiency of this process alone is only around 90%. On the other hand, a novel Fenton process can only remove around 65% of oil drops with the addition of Fenton reagent or poly-ferric sulfate.

The separation efficiencies from those methods are high enough; however, all these traditional techniques have disadvantages. These methods cannot effectively remove oil droplets with a diameter less than 10 μm [25,26]. They are useful only for free oil solutions and dispersed or unstable oil-in-water emulsions [27], and low oil concentration [28]. Other disadvantages are low efficiency [25,28], high operation costs [7, 25,28], corrosion and recontamination problems, the use of toxic chemicals, space for installation and secondary pollution, and significant energy requirement.

3. Membrane based oil–water separation

Membranes for oily wastewater are usually applied for the secondary treatment of oil emulsion. In general, the treatment of oily wastewater can be categorized as primary and secondary treatments. In the primary step, mechanical processes are applied to isolate free-floating oils from water and emulsified oils. Membranes are then used as a potential technique in the secondary treatment to separate stable or emulsified oils from water. In addition to the ability of the membrane to separate small oil droplets from water, membranes also have several advantages compared to other techniques for oily wastewater treatment. The apparent advantages of the membrane process would be lower capital cost, the absence of chemical additives, and the subsequent generation of oily sludge. The retentate that contain oil in very high concentrations could be reused or disposed of into existing incinerators [29].

Pressure-driven membranes are commonly used for oily wastewater

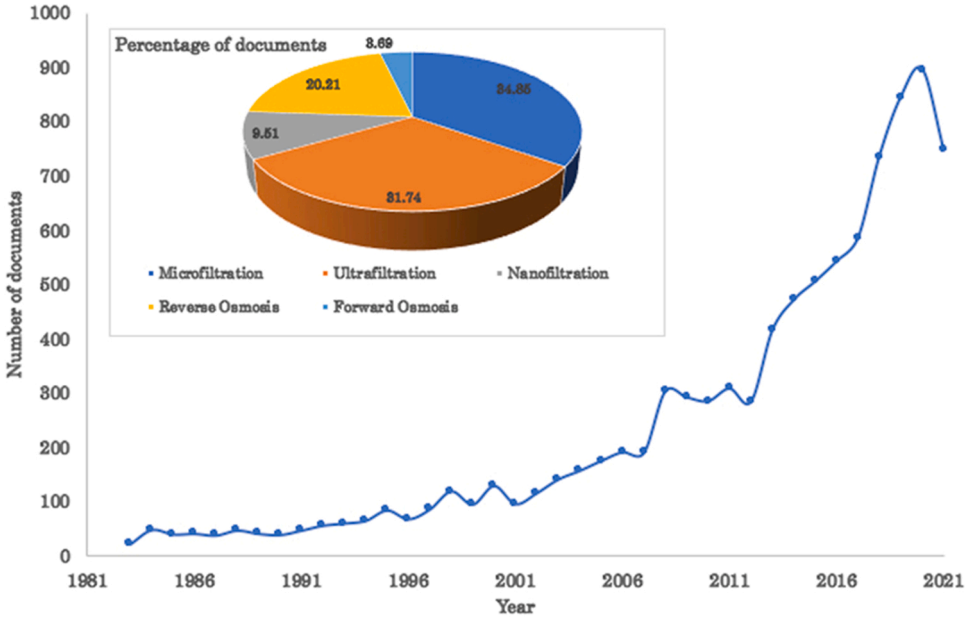

Fig. 1. The number of journal publications on the membrane for oil-water separation processes. The data were retrieved from the Scopus database with keywords Oil AND water AND membrane on 24 August 2021.

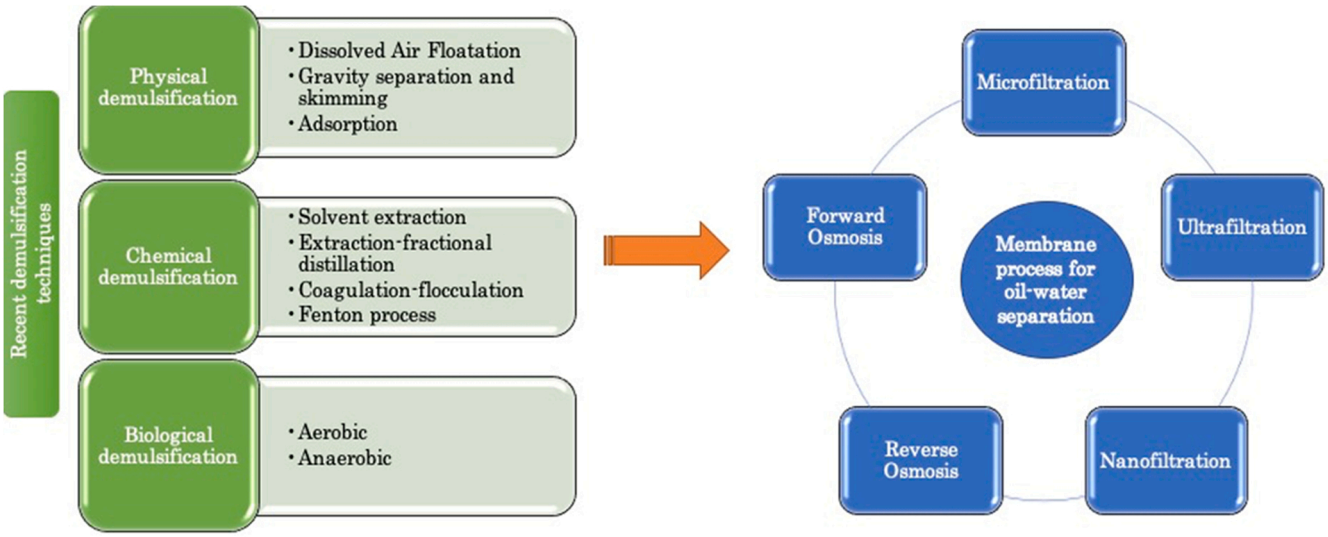

Fig. 2. Recent and membrane-based demulsification techniques.

treatment [30]. Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF), Reverse Osmosis (RO), and Forward Osmosis (FO) are pressure-driven membranes that are commonly used to replace and to be combined with conventional techniques for oil-water separation, as shown in Fig. 2. Microfiltration has the most considerable pore size in the family, while feed pressure used to produce permeate and retentate in microfiltration is the lowest among other membranes. For oil-water separation, up to now, microfiltration and ultrafiltration membranes have been used more frequently than nanofiltration and reverse osmosis.

MF is one type of membrane filtration that has been widely used in oil-water separation. MF membrane can be defined as a porous membrane that discriminates the flow of a suspension containing colloidal or fine particles with dimensions within the size range of 0.1–10 μm through the membrane [18]. MF membranes can produce high permeate flux or product throughput; however, the deformation of oil drops usually occurs due to the large pore size of the membranes [31]. In a cross-flow MF system, the permeate or pure water passes through the membranes, while the oil drops accumulate on the surface of the membrane and form a polarization layer and/or a thin cake on the membrane surface. The build-up of the aggregates continues until a steady-state condition is reached. However, since the feed flows tangentially to the membrane surface in crossflow MF, the accumulation of aggregates is limited, and the pressure drop is lower than the conventional filtration process. This phenomenon is called membrane fouling.

UF is a pressure-driven membrane process that concentrates and fractionates macromolecular solutes and separates suspended species from water. UF provides a non-destructive separation, which can be performed without any phase transition [18]. The concentrate may contain up to 50% of oil. The oily concentrate can be further separated by centrifugation [32]. UF membranes, which have tight pores, have been selected in most applications to ensure steady permeate quality. The flux frequently declines with time; this has been attributed to surfactant adsorption on the pore walls, the buildup of a polarized layer of concentrated emulsion at the membrane surface, and pore plugging by oil droplets [16]. However, this technique has two drawbacks; concentration polarization and membrane fouling. As a filtration process, membranes used are truly porous, and separation is a physical process requiring elevated pressure to achieve passage of fluid through the filter. The water and other low molecular weight solutes pass through the membrane pores. The larger molecules or aggregates are rejected. The successful UF separation performance is usually obtained when oil's discrete and stable emulsion particles, more significant than the membrane pore size, are maintained. However, the separation mechanism of oil and water in UF involves the size differences among various solute and solvent molecules under consideration and adsorption and surface charge characteristics of membranes [33].

Several studies have proved that membrane technology, especially UF and MF, is suitable for treating oil-in-water emulsion [34–41]. Hlavacek investigated membrane coalescence to separate the two phases of an industrial emulsion from the aluminum industry. The demulsification process occurred when the emulsion permeated through an MF membrane. It was found that the oil concentration in the lower layer of the permeate was as low as 30 ppm compared to 30,000 ppm in the original emulsion. Then, using a pilot plant fitted with a 0.2 μm MF membrane of 1 m^2 filtration area, it was shown that the process was reliable during four weeks of operation [34].

In another study, Ohya et al. [35] investigated the effects of average pore size in porous glass tube membranes on the performance of the cross-flow MF using oil-in-water emulsions. Then the flux decline with filtration time was divided into 2 stages. The filtration mechanism in the first stage can be explained by a type of blocking filtration model and the second stage by the cake filtration model. According to the pore size, blocking filtration mechanisms are different; for 0.27 μm pore diameter, complete blocking filtration occurred; for 0.75 μm pore diameter, intermediate blocking filtration occurred, while for 1.47 μm pores diameter, standard blocking filtration occurred. The transition time from blocking to cake formation occurred around 10 min for all membranes with different pore sizes. Oil rejection was found to decrease with time initially but as soon stabilized at a constant level. Surfactant transmission was reported as 49% for 0.27 μm membrane, 80% for 0.75 μm, and 99% for 1.47 μm membrane.

Another study has been conducted using cross-flow MF with rapid back pulsing to remove suspended solids and dispersed oil from an aqueous stream by Ramirez and Davis [42]. Experiments on clay suspensions demonstrated that rapid back pulsing could maintain the permeate flux at a level that is more than 10-fold over the long-term flux in the absence of back pulsing, without any reduction in the permeate quality. Experiments using dilute oil-in-water dispersions showed that rapid back pulsing can increase flux by up to 25 times but cannot be maintained over the life of the membrane.

Nanofiltration (NF) is used most commonly to treat water or wastewater that contains components with a molecular weight larger than 1000 Dalton, and the membrane can retain divalent ions. NF has also been studied for the oily wastewater treatment process [40]. On the other hand, Reverse Osmosis (RO) is one type of membrane filtration

applied for treating a wide variety of industrial effluents to separate oil from water. Sridhar et al. [43] used the pilot-scale RO to determine the feasibility of treating the wastewater from a vegetable oil industry. The system was very suitable for treating oil effluents with total dissolved solid (TDS) concentrations up to 52,000 mg/L. RO has been a good separation process for treating oil industry effluent and water recovery due to the high fluxes obtained alongside the significant rejection of TDS, chemical oxygen demand (COD), biological oxygen demand (BOD), and color. Application of RO units in conditions when the feed may be contaminated with crude oil and fuel oil spillages with feedwater comprising NaCl/water solutions of 2000–35,000 mg/L concentration is also reported [44]. In this work, hydrocarbons retained in solution in water were not found to exhibit damaging effects on the performance of the RO membranes. However, it was reported that diesel contamination possibly posed harmful effects with a capacity to reduce membrane fluxes to zero if present in significant concentrations for even short periods of time. Hexane, which is one of the lighter crude oil fractions, can also cause severe deterioration of the performance of RO membranes when in contact, in pure or emulsified form. The damage is worse in more concentrated hydrocarbon mixtures and at longer exposure times. The damaging effects of the hydrocarbon contaminants are also dependent on types of membrane.

One attractive alternative of conventionally pressure-driven processes is Forward Osmosis (FO). FO is a relatively novel membrane technology used for water treatment. The separation processes in FO involve two crucial steps, i.e., (i) filtration step, where water flows from the feed side with lower osmotic pressure into a draw solution with higher osmotic pressure due to simple diffusion through the membrane. In this case, other components will be rejected by the membrane, and the draw solution is mixed with water that permeates from the membrane, and (ii) water recovery step, where the water is recovered from the draw solution, for example, by evaporation process back to its initial condition and can be reused [45]. Compared to commonly used UF, this osmotically driven process is less energy-intensive while demonstrating better oil rejection and lower fouling tendency. Several studies have attempted to apply FO in the oily wastewater treatment process [46–49]. In order to be competitively used in both bench and industry scales, the challenges dealing with the membrane performance and

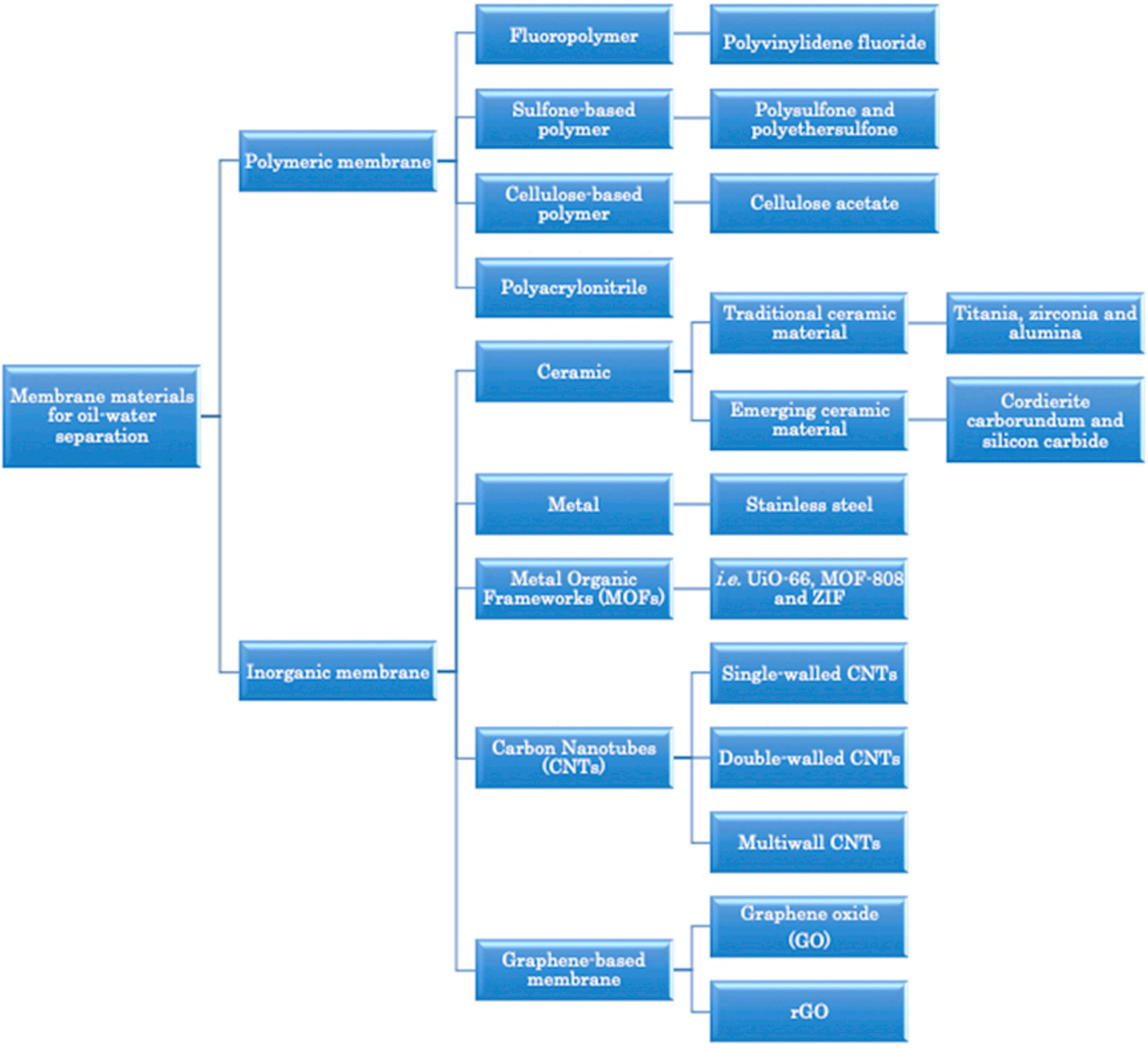

Fig. 3. Summary of membrane materials that are commonly used for oil-water separation.

economic viability of draw solution regeneration must be resolved [27,46,47,50].

4. Membrane materials for oil–water separation

The discussion on membrane technology for oil-water separation should be started from the membrane's materials perspective. This is mainly because the membrane fouling propensity of the membranes used for oil-water separation depends on the materials of the membranes. In general, similar to other types of separation using membranes, the membranes materials for oil-water separation can be categorized as organic/polymeric and inorganic membranes, as shown in Fig. 3. Each material has been applied in industrial as well as laboratory scales to treat oil-water emulsion. Polymeric membranes for oil-water separation offer several advantages over their inorganic counterparts due to low cost for fabrications and ease of scaling up. However, polymeric membranes are relatively easy to be fouled by contaminants or oil drops.

4.1. Polymeric membranes for oil–water separation

Polymeric membranes are used more widely than inorganic membranes for oily wastewater treatment and wastewater treatment in general due to their low cost and ease of fabrication. Polymeric membranes for oily wastewater treatment can be fabricated from various polymers, such as fluoropolymer (Polyvinylidene fluoride or PVDF), sulfone-based polymer (polysulfone or PSf and polyethersulfone or PES), cellulose-based membrane (cellulose acetate or CA), and polyacrylonitrile (PAN) [15,17,51–60]. PVDF membranes have several advantages, such as outstanding antioxidation, superior thermal and hydrolytic stabilities, and good mechanical and membrane-forming properties. However, the hydrophobic nature of PVDF hinders its ability to separate oil–in–water emulsion. Hence several studies have been conducted to modify the surface of the PVDF membranes [17,61]. PSf and PES are polar in nature, and they have amorphous structures. PSf also has a very flexible chain. Hence PSf can be synthesized to form a robust membrane matrix [33]. Cellulosic polymers are polysaccharides with molecular weights up to 1,500,000 g/mole. They can be formed of esters, such as cellulose acetate and cellulose nitrate, or ethers, such as ethylcellulose [33]. Compared to PSf-based membranes, cellulose-based membranes show the distinct advantage, such as easy cleaning [32]. In addition, membranes fabricated based on CA often show high hydrophilicity, high water permeability, and reduced membrane fouling tendency. Membrane from PAN has been synthesized as an Ultrafiltration membrane and showed 100% oil rejection in a previous study [62]. However, the long-term operation resulted in a decrease in flux because of membrane blocking.

4.2. Inorganic membranes for oil–water separation

Inorganic membranes have good mechanical properties and long-term chemical and thermal stability. Hence regeneration can be achieved by heating. They also have a relatively narrow pore size distribution and higher porosity, resulting in better separation characteristics and higher flux. This leads to a lower membrane area needed and a smaller footprint. However, the high fabricating cost is one of the biggest challenges for the large-scale application of inorganic membranes. One inorganic membrane that is commonly used for the treatment of oily wastewater is a ceramic membrane. Ceramic membranes are made of inorganic materials. The ceramic membranes are mainly synthesized from common ceramic materials, such as Al_2O_3 or alumina, zirconia or ZrO_2, titania or TiO_2, and emerging inorganic materials, as cordierite carborundum, and silicon carbide [1,63–65]. Different combinations of titanium, aluminum, and zirconium oxides have been tested for produced water treatment. These materials enhance the hydrophilic property of the membrane surface via the presence of hydroxyl groups on the surface. The hydrophilic nature, in turn, prevents oil attachment and thus effectively prohibits permeation flux decline. However, the addition of aluminum, titanium, and zirconia nanoparticles is expensive. Therefore, other low-cost ceramic membrane modification methods have been gaining popularity in the literature, such as clay, kaolin, fly ash, and the combination of these materials [26,66–69]. Several studies mentioned that the role of the surface of the membranes in the treatment of oily wastewater is significant [26,70,71]. The hydrophilic membranes reduce the fouling by minimizing the formation of the cake layer on the surface. Prior to the sintering process, ceramic membranes are usually hydrophilic in nature due to surface hydroxyl groups (-OH), however, during the sintering process at high temperature, the surface of the membrane changes to hydrophobic. Composite membranes were also prepared to obtain the hydrophilic modification of the membrane surface.

A ceramic and cellulose acetate composite was synthesized as a hydrophilic membrane and tested its separation ability with oil-water emulsions. The oil rejection of 92.54% was achieved with the feed concentration of 200 mg/L in a dead-end flow filtration [26]. Another study prepared Al_2O_3 supported TiO_2 membrane using $Ti(SO_4)_2$ as a precursor by in-situ hydrolysis method and intended to treat oil-water emulsions. The membrane showed around 99.75% rejection for the oil concentration of 4000 mg/L in a cross-flow filtration [71]. Another ceramic membrane based on zirconia was also prepared in another study [70]. Zhou et al. The membranes employed alumina as support using $ZrCl_4$ as a raw material via in-situ precipitation method to treat stable oil-in-water emulsions. The prepared membrane demonstrated 97.8–99.2% rejection for 9–13 mg/L oil concentration in a crossflow filtration.

In addition to ceramic membranes, metal membranes have been used for the treatment of oily wastewater. Because of their low cost, high plasticity, high thermal stability, and good mechanical properties, metal materials have been well studied for use as filter membranes with special wettability for oil-water separation. This special wettability can be achieved by coating the membranes with metal nets and porous metal through physical and chemical methods. A study conducted by Feng et al. [72] was the first study employing metal membranes for oily wastewater. In their study, a stainless-steel mesh membrane was sprayed by hydrophobic polytetrafluoroethylene (PTFE) to create a superhydrophobic–superoleophobic filter membrane. Following that study, many porous metal filter membranes with special wettability, fabricated through coating [73], surface oxidation [74], and chemical surface modification [75], were successfully used for oil-water separation. Other metal membranes were also developed by Holdich group that possessed a slotted structure and were used for several studies on oily wastewater treatment [36,76–78].

Other inorganic membranes that have been successfully synthesized are carbon nanotubes (CNTs) membranes, molybdenum disulfide (MoS_2) membranes, titanium carbide ($Ti_3C_2T_x$) or MXenes membranes, and graphene-based membranes. As one of the members of the fullerene family, CNTs are structurally composed of cylindrical graphite sheets that are rolled up into a seamless tube-like structure with a diameter of the nanometer order and appearance of lattice fencing. Depending on the layers of graphene shells, CNTs can be further classified as single-walled carbon nanotubes (SWCNTs), double-walled carbon nanotubes (DWCNTs), and multiwalled carbon nanotubes (MWCNTs) [64]. Direct vacuum-assisted of CNTs nanosheets can fabricate CNTs membranes onto the surface of a porous substrate. Several CNTs membranes have been fabricated and tested for the oil-water filtration process [79–81]. Two emerging materials for membranes, such as MoS_2 and MXenes, have been utilized to treat oily wastewater. MoS_2 is two-dimensional (2D) material that has high chemical, mechanical and structural strengths and can be tailored to create pores on its surface as has been applied on graphene [82]. MoS_2 materials have been fabricated as nanoporous MoS_2 membranes, layer-stacked MoS_2 membranes, and MoS_2-based composite membranes [82,83]. In a recent study [84], functionalized MoS_2 materials were deposited on alumina substrate to

form membrane to treat oily wastewater. MoS_2 were functionalized by reduced graphene oxide and zeolitic imidazolate framework (ZIF-8). The functionalization process increased the anti-fouling property of MoS_2. In addition to MoS2, MXene is another 2D material that has been applied as membrane for oily wastewater treatment [85,86]. MXenes have high surface area, hydrophilic, and high mechanical stability that create possibility to be used either as pure membrane on a substrate or composite membrane [87]. An ultrathin MXene membrane with the thickness of 30 nm has been successfully coated on a polyethersulfone substrate and was applied to treat emulsion [85]. Produced membranes showed excellent performances in terms of anti-fouling and permeate flux due to hydrophilicity property of MXene as well as the inter layer spacing between two MXene sheets. Other inorganic materials that have been applied as membranes for oily wastewater are graphene and graphene oxide [88]. Graphene oxide or GO possess hydrophilic property that can reduce fouling possibility inside the membrane. Several studies utilized vacuum filtration to create GO layer on the surface of substrate [89,90].

In recent years, Metal Organic Frameworks or MOFs, also known as Porous Coordination Polymers (PCP), or porous coordination networks, have lately arisen as a broad class of crystalline materials with ultrahigh porosity (up to 90% free volume) and massive internal surface areas, extending beyond 6000 m^2/g [91,92]. MOFs have received a growing interest from many researchers as membranes and potential fillers in composite membranes [93]. MOFs are considered hybrid porous solids as they contain metal cations connected by an organic linker. The types of metals and linkers will significantly determine the chemical structures and properties of the resulting MOFs. The concept of secondary building units (SBUs) in MOFs permits a theoretically unlimited number of conceivable MOFs structures. MOFs have a coordination network and bonding, which makes them less stiff and brittle than zeolite. Hence, MOFs are able to show flexibility in terms of the gate opening and linker dynamics [92,94–96]. MOFs are named in a way analogous to that adopted by zeolite particles. Various MOFs can be used as membranes with anti-fouling properties, such as UiO-66, because of their hydrophilic property [92]. Another MOFs is ZIF-8, which is potentially useful to synthesize a superhydrophobic membrane used in the water–in–oil emulsion separation process [97]. Several studies have conducted the fabrication of pure MOFs membranes for the oil-water separation process, and several excellent review papers are available for MOFs membranes [92,94,98].

5. Membrane fouling

5.1. Fouling phenomenon

The long-term application of membranes in oil-water separation depends on the extents of two parameters, such as the product throughput through the membranes that is usually denoted as permeate flux and the oil rejection or oil rejection efficiency. The permeate flux and oil rejection during filtration can be altered by several factors, i.e., changing the driving force, applying membranes with different pore sizes, and the propensity of fouling the membranes by the components on feed solution. The fouling phenomenon in oil-water separation using membranes depends on some aspects, such as surface chemistry, membrane's pore size, membrane's surface structure, surface charge, surfactants, operating conditions, and the presence of other components in the wastewater.

5.1.1. Feed–membrane surface interaction

The surface chemistry between the membrane and oil drops is one of the most critical factors in membrane fouling by oily wastewater. Surface chemistry involves the interaction between the oil drops and the membrane under water. In general, a hydrophilic membrane will have an oleophobic nature when it interacts with oil. It is to mention that a hydrophilic membrane will retain oil droplets on its surface and allow water to permeate. On the other hand, a hydrophobic membrane tends to show its tendency to attract oil droplets to pass through (shows its underwater oleophilic nature). This will open the opportunity for the oil droplets to foul the membrane. Hence, creating a membrane with a hydrophilic surface is one way to reduce the possibility of membrane fouling in oily wastewater filtration.

5.1.2. Membrane's pore size

Ideal separation through the membranes requires that contaminants should be larger than the pore size of the membranes [99]. However, the pores of the membranes should not be too small to avoid the squeezing and blocking of the membrane's pores by contaminants or oil drops. Hence, the control of pore size and the choice of membranes with appropriate pore size will prevent membrane fouling.

5.1.3. Membrane's surface charge and structure

The surface charges of the membrane surface and oil droplets underwater determine the extent of membrane fouling. The fouling phenomenon occurs because of the difference in charges between the membrane's surface and oil droplets. In terms of surface structure, the difference in surface roughness of the membranes determines the interaction between the oil droplets with the membrane's surface. It is also reported that surface charges can influence membrane fouling by modifying its wettability towards oil droplets.

It is reported that a membrane with hydrophilic nature will attract water on its surface, then the contact of the oils with the membrane's surface will appear as the interaction not only oil and membrane but also between the oil and the composite of water and membrane's surface [26]. However, the surface roughness of the membranes also determines the occurrence of fouling [100]. A higher degree of roughness leads to higher interaction with oil droplets and increases the tendency of the membrane to be fouled by the oil droplets.

5.1.4. The presence of surfactants

The existence of surfactants in oily wastewater can alter the interaction between the feed water and the surface of membranes. The surfactants can exist as the foulant for the membrane. In this case, the surfactants can be adsorbed and accumulated on or inside the membranes and increase the flow resistance through the membranes. In addition, the surfactant micelles may also block the pores leading to flux decline. On the other case, the surfactants can alter the wetting phenomenon of the membrane's surface by the oil droplets. The study showed that because of the occurrence of surfactants, the hydrophilicity of the membrane could be changed by the surfactants. Hydrophilic membranes might reduce their hydrophilic nature or become oleophilic, or on the other hand, the surfactants can increase the hydrophilicity of the membrane. This might happen because the polar end groups of surfactants would be adsorbed onto the hydrophilic membrane surface, whereas the hydrophobic hydrocarbon chains are likely to be exposed outwards. Hence, the membranes are more susceptible to fouling. Meanwhile, the membrane would also lose its selective wettability towards oil and water upon surfactant adsorption, adversely influencing its efficiency in oil-water separation.

5.1.5. The presence of contaminants

A contaminant in oily wastewater that can alter the fouling phenomenon in membranes is salts. Salts can change the oil-water interfacial tension, influencing droplet deformation and penetration through the membrane. In addition, salts may promote droplet coalescence due to electrostatic screening, hence increasing the propensity of fouling. Moreover, antifouling membranes may gradually lose their antifouling property under saline water because of the decomposition and corrosion of hydrophilic components of the membrane. At last, during the treatment of saline wastewater, the membranes can also be contaminated by salt crystals.

5.1.6. Filtration's operating condition

The fouling phenomenon in membrane filtration is also determined by the operating conditions employed during the filtration process. Feed flow velocity and feed pressure are considered the main operating conditions affecting the polarization concentration and fouling phenomena. In cross flow filtration mode, higher feed velocity can reverse and sweep the polarization of contaminants on the membrane surface. In dead-end filtration mode, the control of feed pressure can affect the polarization and fouling phenomena. Oily wastewater contains oil drops that are not rigid and can squeeze through the pores of the membranes. Hence, the control of operating conditions during filtration is significant for long-term operation.

Despite the successful utilization of microfiltration and ultrafiltration on the treatment of emulsion on a laboratory scale, there are still limited successes in realizing these techniques on an industrial scale. It is considered very important to find a suitable model from a laboratory scale that can be employed to predict the behavior of membranes when the membranes are applied to treat real emulsions on an industrial scale. Research on the investigation of flux behavior during oil-in-water (O/W) and water-in-oil (W/O) emulsions has been conducted recently using synthetic emulsion. The flux decline during microfiltration of O/W emulsion using porous glass tube membranes was investigated by Ohya et al. [35] The flux decline can be explained by two stages or models, i.e., blocking filtration and cake filtration. On another line of research, Tien and Ramarao evaluated four filtration models, i.e., cake filtration, deep bed filtration complete blocking, and intermediate blocking [101]. Hermans and Bredèe first developed these models in 1936. In the proposed models, four cases might appear during the filtration process. The cases include cake filtration, deep bed filtration, complete blocking, and intermediate blocking as described in Fig. 4.

In A's case, the separation process is conducted with 40 wt% slurries that can form a cake layer on the filter's surface. Case B explains the separation process using a filter with relatively big pore sizes; thus, many particles will pass through to the permeate side. In the case of C, the filtration is done using the filter with small pore sizes, while Case D uses a filter with pore sizes in between the pore sizes used in case 3 and case 1 or 2. All cases can also be explained by mathematical expressions as follows:

Case A. cake filtration:Filtration rate:

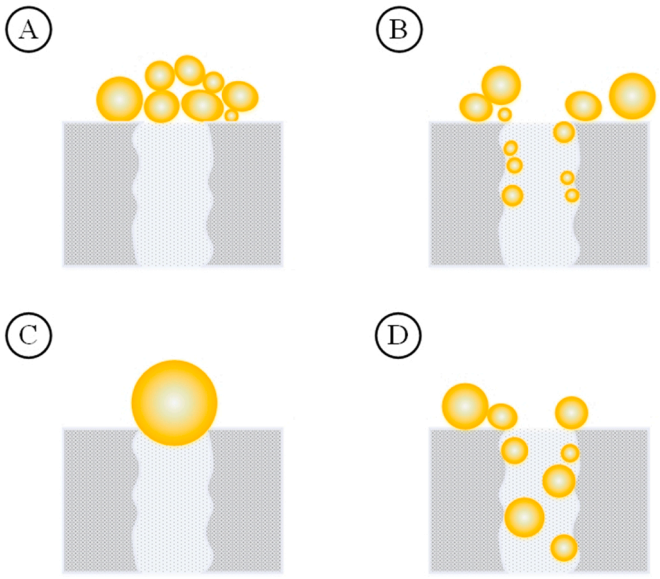

Fig. 4. Schematic representation of filtration model: (A) cake filtration, (B) deep bed filtration, (C) complete blocking, and (D) intermediate blocking.

$$\frac{\mathrm{dV}}{\mathrm{dt}} = \frac{\mathrm{P}}{\mu(\mathrm{AV}+\mathrm{B})} \tag{1}$$

The correlation between V and t:

$$\mathrm{t} = \frac{\mu}{\mathrm{P}}\left(\frac{\mathrm{AV}^2}{2}+\mathrm{BV}\right) \tag{2}$$

where V is permeate volume, t is time, μ is viscosity, and P is trans membrane pressure. A and B are constants.

Case B. deep bed filtration:Filtration rate:

$$\frac{dV}{dt} = \left(\frac{dV}{dt}\right)_o (1-aV)^2 = \frac{(dV/dt)_o t}{\left(1+a(dV/dt)_o t\right)^2} \tag{3}$$

The correlation between V and t:

$$\mathrm{V} = \frac{\left(\frac{dV}{dt}\right)_o t}{\left(1+a(dV/dt)_o t\right)} \tag{4}$$

Case C. complete blocking:Filtration rate:

$$\frac{dV}{dt} = \left(\frac{dV}{dt}\right)_o (1-aV) \tag{5}$$

The correlation between V and t:

$$V = \frac{1}{a}\left[1-exp\left\{-a\left(\frac{dV}{dt}\right)_o t\right\}\right] \tag{6}$$

Case D. intermediate blocking:Filtration rate:

$$\frac{dV}{dt} = \frac{(dV/dt)_o}{(1+At)} \tag{7}$$

The correlation between V and t

$$V = \frac{\left(\frac{dV}{dt}\right)_o}{A} ln(1+At) \tag{8}$$

Several studies have attributed membrane fouling by emulsified oil to one or more of the above mechanisms [101,102]. However, the validity of the blocking laws applied to emulsion separations is still under investigation for further study [16]. Several reasons are raised for the applicability of blocking law for oil-water separation. First, the blocking laws were derived for monodisperse suspensions of spherical and non-deformable particles. At the same time, oil droplets have shape and size that depend on several factors (e.g., the composition of the two liquid phases, interfacial tension, shear rate) and can change as a result of transient processes such as droplet deformation, coalescence and break-up. Second, the blocking laws assume no back-transport of the foulants away from the membrane surface. This assumption precludes application of blocking laws to permeate flux data obtained in stirred dead-end and crossflow filtration tests, where a number of particles back-transport mechanisms operate simultaneously. Even in unstirred conditions, back-transport can still be significant for sufficiently small droplet sizes and large concentration gradients near the membrane surface. Another mechanism that was proposed to explain membrane fouling by oily wastewater is the formation of a gel layer on the membrane surface [16]. However, the investigation on this mechanism has been conducted only by indirect observation [16].

5.2. Oil drops deformation phenomenon

Another phenomenon that should be considered seriously in separating oil from water using a membrane is the deformation of oil drops in

the membrane's pore. The rejection of oil drops determines the successful separation of oil-in-water emulsion using membranes, significantly drops bigger than the membrane's pores. Oil drops are deformable and big drops can squeeze and appear in the permeate of produced water. Fig. 5 below is a typical description of the deformation of a big drop on the surface of membrane:

The oil drop initially has a radius of R. When it enters the pore under appropriate conditions, the body of the drop, which contacts the pore entrance, or the leading edge of the drop, will tend to squeeze and form a more petite shape with a diameter r. The rear side of the drop and outside surface of the filter form a contact angle, while the leading edge of the drop and internal pore surface form a contact angle. Many factors and conditions influence the squeeze of a significant drop. The conditions include the emulsion properties, membrane pore size and morphology, and interactions between the oil droplet and the membrane surface. Another factor is the velocity of the feed solution, which can be correlated to the shear rate on the surface of the membrane [13,80]. If the oil droplet is much smaller than the nominal pore size, the oil enters membrane pores and contributes to intrapore fouling. The permeate flux and oil rejection depend on the operational parameters. The trans membrane pressure relative to the membrane pore size and size distribution of oil drops in the feed has also been reported to be critical operating variables: membrane composition and solution chemistry. To avoid intrapore fouling by emulsified oil, membrane pore size should be smaller than oil droplet size. Under such conditions, the oil may still enter membranes pores but only if the transmembrane pressure is higher than the critical pressure (*P crit*). The critical pressure required to squeeze the oil drops can be calculated using Eq. (9) [31,104].

$$P_{crit} = 2\gamma_{ow}\frac{cos\theta}{r_{pore}}\left(1 - \left[\frac{2 + 3cos\theta - cos^3\theta}{4\left(\frac{r_{drop}}{r_{pore}}\right)^3 cos^3\theta - \left(2 - 3sin\theta + sin^3\theta\right)}\right]^{\frac{1}{3}}\right) \quad (9)$$

where y_{ow} is the interfacial tension between oil and water, and θ is the contact angle measured from the water side, i.e., $\theta = 180^\circ - \theta_{ow}$. r_{pore} and r_{drop} are radii of the pores and oil droplets, respectively.

Several conclusions can be drawn from Eq. 9. First, the underwater oil contact angle θo*w* determines the sign of P_{crit} For $\theta_{ow} < 90^\circ$, P_{crit} is negative, implying that the oil can wet and fill the pores of the membrane spontaneously even under zero pressure. The filtration may fail because the oil can easily pass through the membrane. Thus, θ_{ow} should be more significant than 90° to obtain successful filtration. It is preferred to be considerably higher since it allows a high transmembrane operation pressure, essential to increasing the permeate flux. Second, when θ_{ow} is $> 90^\circ$, the critical pressure increases with decreasing pore radius. This means that membranes with smaller pores have higher rejection efficiency to oil droplets. Lastly, Eq. (2) also indicates that larger droplets have a higher critical pressure, and thus smaller droplets are easier to pass through the pores under pressure. If the droplets are smaller than the pore size, the droplets would freely pass through the membrane pores, leading to filtration failure. For droplets of infinitely large size (e.g., an oil film covering the membrane), the critical pressure becomes $P_{crit} = 2\gamma_{ow}\cos\theta / r_{pore}$ [13].

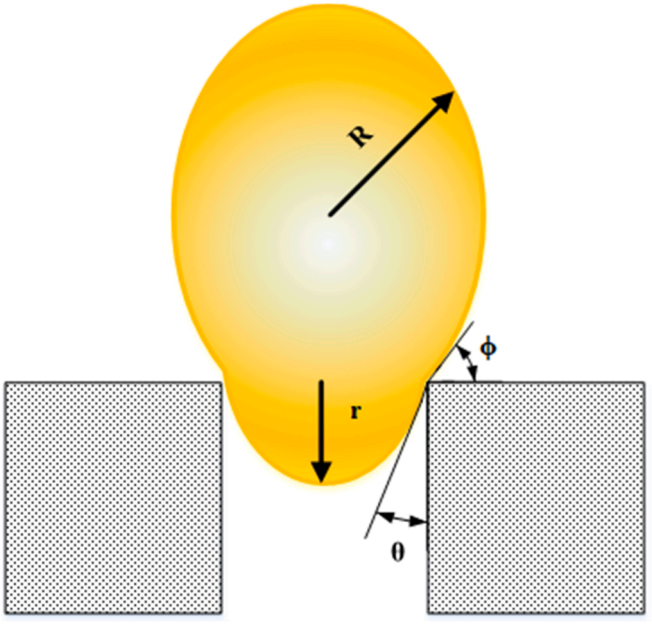

Fig. 5. Schematic of an oil drop at pore entrance of membrane.

Hence, another fouling model has been proposed by Huang et al. [13]. During the filtration of oily wastewater, emulsified oil droplets move onto the surface of the membrane because of the permeate flow. The deposited droplets would partially block the membrane pores at the early stage of filtration. During filtration, the droplets accumulate on the surface of the membrane and form a cake layer. In crossflow filtration, as the crossflow could also carry oil away from the cake layer, a continuous cake layer is expected to form once a balance between oil deposition and oil removal is reached. Since oil droplets are deformable, the resultant cake layer can be densely packed and shows high resistance to water permeation [41]. Oil droplets in the cake layer are thermodynamically unstable and tend to coalesce. In some experiments, it was found that coalescence led to the formation of larger oil droplets which were easier to remove by crossflow [41,103]. This can be understood by considering the critical penetration pressure of oil droplets. Larger droplets have higher critical pressure based on Eq. (9); thus they are less likely to permeate through the membrane and more probable to be carried away by the crossflow (the critical droplet size beyond which the oil droplet can be carried away by crossflow has been predicted based on the force balance on the droplet) [103]. In this respect, the coalescence of oil droplets helps to mitigate membrane fouling. However, suppose severe pressure is exerted on the oil droplets. In that case, they may experience a wetting transition on the membrane (especially for less oleophobic membranes), accompanying a significant decrease of their oil contact angles under water [40]. These collapsed oil droplets might coalesce laterally on/within the membrane, and consequently, a continuous oil film will form. It is expected that the contiguous oil film forms more easily on membranes, which are underwater oleophilic. [8] Such contiguous oil film obviously brings serious membrane fouling. Droplets smaller than or comparable to membrane pore size could enter or be forced into the pores by the permeate flux. This leads to internal oil fouling within the pores, also a type of membrane fouling that is difficult to clean [41,103].

In another study, other mechanisms were also developed by Salama et al. [105]. In their study, two mechanisms were proposed, where in the first mechanism, the accumulation of pinned oil droplets that are neither permeated nor detached are assumed to block the pores of the membranes and reduce the pores available for filtration. Then, this model was extended into the second model. The second model accommodates the more general scenario contributing to the problem of fouling in oily water filtrations. This mechanism is generally attributed to the coalescence and clustering of oil droplets with pinned droplets to make larger oil chunks or laminates that reduce the membrane area available for filtration. In this model, the details of the fates of individual oil droplets are irrelevant due to the dynamic process that changes the shape, size, and orientation of oil droplets. Instead, a coarser framework is constructed. The oil, the water, and the membrane are dealt with as three separate continua rather than the multitude of continua of both the oil and membrane required in the previous model.

6. Fouling mitigation during oil–water separation

The fouling mitigation of membrane during oil-water separation has been the focus of research in oily wastewater treatment using membranes [13,15,17,106]. In general, the discussion on the minimization of fouling can be categorized and analyzed from the membrane and operating conditions perspectives. Hence, the discussion in the following sections will be started from the review of current trends and research on the efforts to modify, choose, and combine various materials and techniques related to the membranes. Then the review will be focused on the several efforts conducted to manipulate operational conditions during oil-water separation and filtration.

6.1. Perspective from membrane materials selection

In general, membranes with hydrophilic nature and oleophobic under water are desired to minimize the polarization concentration and fouling phenomena during oil-water filtration. The ability of the membranes to allow water to pass through while repelling oil can provide efficient separation without the use of energy and other treatments, which is preferable as the treatment would be cost-efficient and easily operated. Hence, the wetting properties of membrane materials are very useful in oily wastewater treatment because they can determine how specific membranes can be applied in the filtration process. The hydrophilicity of particular materials often depends on interfacial interaction in which it differs from hydrophobicity in terms of competition between the interfacial free energy of cohesion of the solid immersed in liquid (in this case, water). Usually, the hydrophilic surface is assumed to be fully wetted by water denoted with a contact angle of 0 degrees.

Various approaches have been proposed and conducted to create more hydrophilic membranes, such as polymer-polymer blending [107], polymer – inorganic mixing inside mixed matrix membranes (MMMs) [108–111], surface modification of membranes [6,112,113], employing membranes with different pore geometry [36,76–78,114,115] as well as using emerging hydrophilic materials like graphene oxide (GO), MOFs and CNTs as free-standing membrane [88,106,116,117].

6.1.1. Polymer–polymer blending. Many researchers have tried to blend polymers from different sources or mix certain polymers with additives from the material selection perspective. The blending process is intended to change or increase the hydrophilicity of the base polymer, hence the fouling phenomenon can be minimized. Two main additives are used to improve hydrophilicity, namely hydrophilic polymers and amphiphilic copolymers [118]. These two materials exhibit the best affinity to water but exhibit weak compatibility with polymer matrices, such as PVDF, PSf, and PAN. Therefore, the content of hydrophilic polymers in the membrane is limited and gives rise to insufficient hydrophilicity. In addition, the blended hydrophilic polymers are easy to release from the membrane during long-term filtration. It will cause the attenuation of membrane hydrophilicity gradually. Various types of polymers and additives have been employed, including polyvinylpyrrolidone (PVP) [38,119,120], polyethylene glycol (PEG) [38,121], polyvinyl alcohol (PVA) [37,122], Pluronic [25,123], polyaniline [124], sulfonated poly (ether ether ketone) (SPEEK) [125], polyethylenimine (PEI) [126–129], poly(acrylic acid) [130], and poly(2-hydroxyethyl methacrylate) [131], poly(vinyl pyrrolidone) (PVP) and segmented amphiphilic copolymer of poly(dimethylsiloxane) and poly(ethylene glycol) (PDMSPEG) [132], bifunctional copolymer based on poly(methyl methacrylate-co-glycidyl methacrylate) (P(MMA-co-GMA))[133], Polyacrylamide (PAM) [134], and polymer additive that has both hydrophilic and oleophobic groups synthesized from CA via one-step esterification between the active hydroxyl groups and a functional fluorine-containing compound, perfluoroalkyl polyethoxy acetic acid (FPEOAA) [135].

A simple blending technique was commonly used to increase the hydrophilicity of the base polymer. Zhang et al. have successfully fabricated an antifouling membrane by employing a simple blending technique. They blended aliphatic polyketone (PK) and PVA. PK is a polymer with intrinsic intermediate hydrophilicity and good membrane formation ability that is usually synthesized using a non-solvent induced phase separation process. To improve the hydrophilicity of PK, the polymer was mixed with PVA that was introduced as a pore-former and more importantly, a hydrophilicity enhancer to perfect the underwater oil repellency. Fouling-resistant membrane filtration of oil-in-water emulsions was readily achieved in the cross-flow mode with a high flux up to 420 L m^2 h^1 at 0.1 bar and a high flux recovery of 93–96% being maintained, even for the highly adhesive and highly fouling soybean oil emulsions (up to 100,000 ppm). A small amount of the PVA additive trapped within the PK membrane matrix can help reach the desirable underwater superoleophobicity. The composite membrane can enable outstanding fouling-resistant cross-flow filtration of highly fouling oil-in-water emulsions with very low or no fouling [122]. In another study, the combination of more than two polymers were also conducted [121]. For instance, PEG-400 was introduced into the mixture of PES and CA polymers. The PEG-400 introduction was intended to improve the hydrophilicity of composite membranes, hence enhancing the permeate flux. The membranes synthesis was conducted by phase inversion process, and produced membranes showed increased water flux compared to membranes without PEG-400. The higher water flux of the PES/CA membrane was related to the membrane's higher hydrophilicity and larger pore sizes.

In terms of membrane operation, both MF and UF have been synthesized from different polymers, such as cellulosic esters, polyamides (PA), PSf, PAN, and PVDF, to obtain the specific characteristics. The composite UF membranes from blended polymers are also developed for oil and water separation purposes [107,136]. The ultrafiltration performances in the oil-water separation of each type of polymer are also investigated. The investigation on the effect of PVP and PAN blending on membrane morphology and oil separation performances has been conducted [137]. The addition of PVP did not change the overall morphology of membrane. The enhancement on flux and oil rejection were recorded after the blending of polymers. From continuing investigation from both research and real applications, ultrafiltration becomes the most appropriate process for oil-water separation from industries.

On another line of research, the blending process is conducted to improve both polymers' performances in terms of antifouling property and increasing permeate flux. In this case, PAN membrane that can produce high water flux, but the poor antifouling property was blended with CA membrane that produces low flux but shows superior antifouling property [138]. A novel membrane material was synthesized by grafting PAN onto CA powder via free radical polymerization to achieve high permeation flux and fouling resistance. The synthesized CA-graft-PAN (CA-g-PAN) copolymer was employed to fabricate asymmetric ultrafiltration membranes by the phase inversion method. It was found that CA-g-PAN membranes exhibited remarkably high-water permeability (about 100 times) than CA membranes. Ultrafiltration experiments also demonstrated that CA-g-PAN membranes have the excellent oil-fouling-resistance ability even under higher operation pressure and higher oil concentration in oil/water emulsion. Most of the deposited oil droplets on CA-g-PAN membrane surfaces can be easily washed away by simple hydraulic washing, and the fluxes of CA-g-PAN membranes were nearly completely recovered. After three times oil/-water emulsion ultrafiltration, the feed solution flux was kept at a relatively high level of about 110 L/(m2 h), indicating good oil/water separation performance of CA-g-PAN membranes.

Another material to blend with the polymer is zwitterionic polyelectrolyte [118,139]. It is well known that sulfobetaine zwitterionic polyelectrolyte contains both anionic and cationic species in the polymer side chain. Due to the strong interaction between ions and water, a stable, hydrated layer can be formed around this polymer chain, effectively preventing foulants, such as protein, marine organisms, and oil droplets, from attaching. Therefore, zwitterionic polyelectrolytes have

been widely used for designing materials with improved anti-fouling performance [118]. The mixture of PVDF/ copolymer poly (3-(N-2-methacryloxyethyl-N,N-dimethyl)ammonatopropanesulto-ne)-co-2 hydroxyethyl methacrylate) (PSH) membranes showed water flux recovery after filtration experiment as high as 98%, demonstrating superior antifouling performance when separating oil-in-water emulsions.

6.1.2. Polymer–inorganic particle blending inside mixed matrix membranes (MMMs) and composite membranes. In another line of research, the application of mixed matrix membranes (MMMs) in oily wastewater treatment has just been started recently [140]. In MMMs, the inorganic fillers are incorporated inside the polymer matrix. Hence, the separation mechanism inside the membranes will combine the separation mechanism of fillers and pure polymeric materials. The successful separation process using MMMs is determined mainly by the absence of voids and defects on the interface between the inorganic fillers and the polymer matrix. Voids and defects in polymer-particle combination in some cases can increase the flux through the membranes but will decrease the selectivity of the membranes. The formation of void and defect-free MMMs is growing as an exciting field of research in the pervaporation process. In addition to MMMs, MMMs-based composite membranes have also been utilized in oily wastewater treatment. The different morphologies of polymer particles that will determine the separation mechanism through the composite membranes and MMMs are depicted in Fig. 6. The morphologies include ideal morphology, sieve-in-a-cage, the rigidification of polymer, and the blockage of particle pores. These defects can be caused by several factors, such as the incompatibility between polymer and particle, the evaporation of solvent during membrane formation that stresses the interface of polymer-particle, and the weak adhesion between the particle and polymer.

Four morphologies in MMMs can be described as (1) idealized or 'hard to obtain' morphology, (2) voids at the interface morphology, (3) rigidified polymer layer morphology, and (4) reduced permeability region with in sieve morphology.

Case 1. is usually explained by the Maxwell model that is formulated by Eq. (10):

$$P_{mm} = P_c\left(\frac{P_d + 2P_c - 2\varnothing_d(P_c - P_d)}{P_d + 2P_c + \varnothing_d(P_c - P_d)}\right) \tag{10}$$

where P_{mm} is the effective permeability of an MMMs, ϕ is volume fraction, while c and d denote the continuous and dispersed phases, respectively.

Case 2. or sieve-in-a-cage morphology is the easiest to diagnose. It is also easy to make this undesirable morphology, especially with glassy polymers. Case 2 results in increased permeability with essentially no change in selectivity. Although this may seem to be a favorable trade-off, we believe it will be exceedingly difficult to prepare such morphologies without a dramatic loss of selectivity.

Case 3. demonstrates the formation of the rigidified region on the interface between the polymer matrix and inorganic particles. The rigidified region arises because of stress experienced by two materials during the preparation of MMMs. When the rigidification of polymer

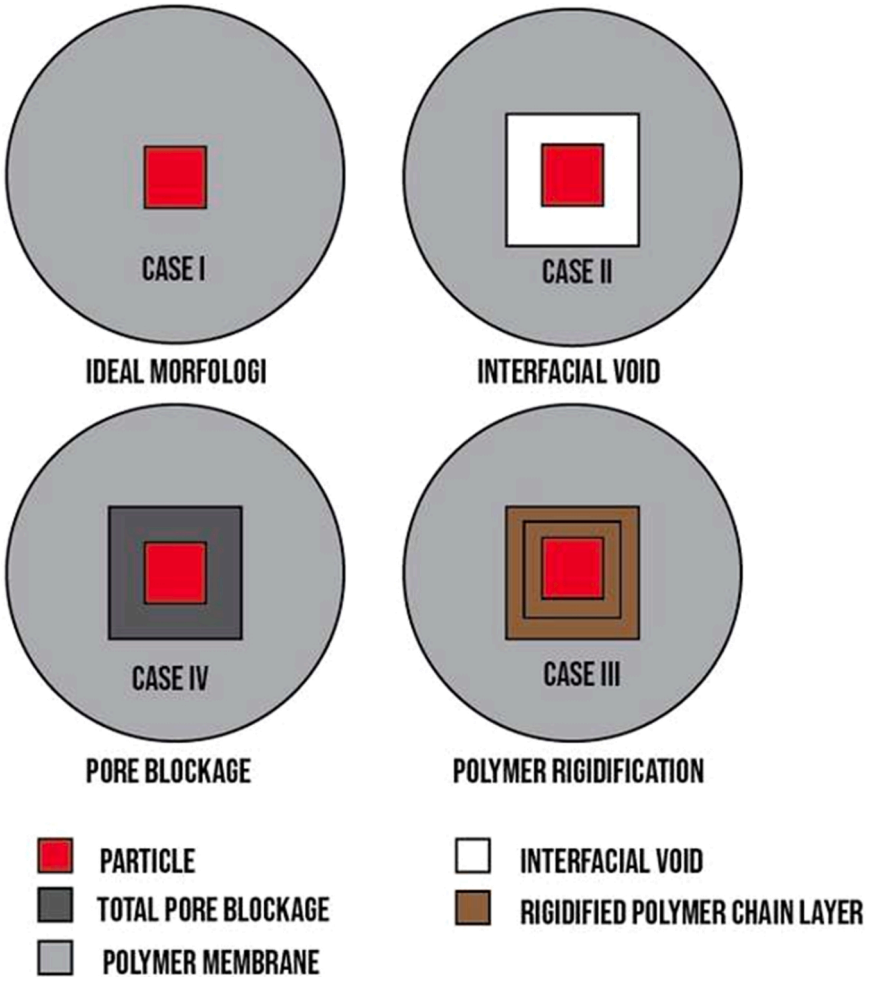

Fig. 6. The morphologies and typical defects in MMMs.

occurs, it can be expected that the polymer layer near the particle surface has low chain mobility that will improve the resistance to penetrants, decrease the permeability, and increase the membrane selectivity. This phenomenon can be observed by the increasing value of the polymer's glass transition temperature [24]. While in case 4, the interphase comprises a reduced permeability region in the outer layer of the particles or the whole particles.

The non-ideal morphology of MMMs could be avoided and minimized by several techniques, such as proper choice of combination between polymer and particle, modification of polymer or inorganic filler before mixing, priming protocol during mixing, optimizing the threshold concentration of particle inside MMMs, and employing dual fillers. Up to now, various polymers and inorganic particles have been utilized inside MMMs as well as composite membranes to treat oily wastewater. For polymers, pure or single polymer and polymer blends have been used as the matrix for MMMs. Polysulfone [141–143], polysulfone – polyethyleneimine blend [126], polysulfone - polyvinylpyrrolidone blend [144], polyethersulfone [136,145–147], polyacrylonitrile [123,148,149], PVDF [150–152], cellulose acetate [108,111,153], and polybenzimidazole (PBI) [154] are typical polymers applied that have been combined with inorganic fillers.

In terms of inorganic fillers, multiwall carbon nanotubes (MWCNTs) [117,141,150,155], halloysite nanotubes [110,146,147], montmorillonite [111,144], $CaCO_3$ nanoparticles [123,126], graphene oxide (GO) and reduced graphene oxide (rGO) [142,145,152], nickel-cobalt layered double hydroxide (NiCo-LDH) nanomaterials [151], titanium dioxide (TiO_2) [152,153], silica (SiO_2) [112], iron acetate [143], zwitterionic nanoparticles – polydopamine -sulfobetaine methacrylate or P (DA-SBMA) [108], hydrose manganese oxide [109], metal-organic frameworks (MOFs) [148,149], and the combination of fillers mentioned above are inorganic particles that have been used as discontinuous phase inside MMMs. Among all fillers, several fillers, including CNTs, halloysite nanotubes, GO or rGO, and MOFs have been used more frequently than other fillers.

CNTs possess high specific surface area, easy functionalization, chemical stability, and proper compatibility that make them one of the potential fillers to be explored in the future for the synthesis of MMMs and nanocomposite membranes. In addition, CNTs can help to create a rougher surface due to their rigid cylindrical nanostructures with a diameter ranging from about 1 nm to dozens of nanometers and length ranging from hundreds of nanometers to micrometers, which could lead to the increase of efficient filtration area and permeability of the composite membranes [156]. However, the high tendency of CNTs to form aggregates due to strong van der Waal's forces has driven several studies to improve the dispersion of CNTs into the polymer matrix. One way to improve CNTs dispersion is using GO nanosheets as a dispersing agent because of their high contents of hydrophilic functional groups [117]. GO/MWCNTs-based MMMs have been synthesized and used for Palm Oil Mill Effluent (POME) treatment. The incorporation of particles can improve the antifouling property of the membranes and enhance water permeability due to improvement in hydrophilicity, the thinner skin layer, and forming a repulsive boundary barrier [117].

Graphene is another material that is considered prospective to be utilized as a filler in MMMs and nanocomposite membranes. Graphene is quoted as the mother of graphite material as graphene is the 2-dimension form of graphite and is usually produced by mechanical exfoliation of graphite. Graphene gains much interest in material research as graphene is one of the strongest materials in terms of mechanical stability because of its sp2 hybridized bond [157,158]. Its 2-dimensional structure opens a possibility to be utilized as an inorganic filler in MMMs or nanocomposite membrane because a small and thin particle can be produced from graphene material.

In addition to graphene, GO and rGO have also been utilized as fillers in MMMs and nanocomposite membranes. Chemical conversion of graphite to graphene oxide is considered the best way to synthesize graphene in large amounts. In this typical method, graphite is first oxidized using sulfuric acid, nitric acid, and potassium permanganate to produce graphite oxide or GO (Hummers method). GO is hydrophilic material and can be exfoliated in water or N,N-dimethylformamide, tetrahydrofuran, or N-methyl-2-pyrrolidone or ethylene glycol to produce graphene oxide. The basal plane of graphene oxide contains hydroxyl and epoxy groups and a small amount of lactol, ester, acid, and ketone carbonyl groups at the edge, making it possible to be functionalized. However, these functional groups can produce defects on graphene oxide that unfortunately can be partially recovered by chemical reduction of graphene oxide, so its conductivity is a magnitude lower than pristine graphene. The reduction of graphene oxide into rGO or graphene can be conducted using thermal reduction or by adding hydrazine to reduce the hydroxyl functional group. The carboxyl group can be deprotonated by pH adjustment using ammonia solution. Other chemical agents that are usually used to reduce graphene oxide are sodium borohydride or hydrogen gas [159].

To treat oily wastewater, GO and rGO have been incorporated into the polymer matrix to improve the performances of pure polymeric membranes [88,145,152]. GO and rGO were used as single filler and mixed with other components before being incorporated into a polymer matrix or deposited on a support. The use of GO as a single filler has been demonstrated by a recent study [145]. The GO nanosheets were incorporated inside the matrix of PES in both flat sheet and hollow fiber forms. A significant enhancement of hydrophilicity was observed for both configurations (flat sheet and hollow fiber), where the water contact angle value decreased from 74.8° to 42.2° and 71.4–49.8°, respectively. The flux of the composite membranes increased up to 150% compared to pristine polymeric PES membranes, especially when 1.0 wt % of GO was added. A composite of rGO and SiO_2 that has been modified by Polydopamine (PDA) was successfully coated on a PVDF porous support via a surface deposition method. Produced membranes showed an improved performance on the treatment of oils and cationic dye from wastewater compared to both pure PVDF membrane and non-PDA modified membranes. The membranes also showed superior anti-fouling property and sustainable stability simultaneously. In addition, the recovery flux after ten cycles can reach 87.2% when filtering oil-in-water emulsions, and 78.9% when processing dye. This robust antifouling performance indicated our membrane was promising for practical applications [152]. The combination of PVDF polymer and GO particles was also realized in a composite membrane. To improve the underwater superoleophobicity of the PVDF/GO-based membranes, the membranes were crosslinked with acrylic acid and ethylene glycol dimethacrylate [160]. Produced membranes depicted good separation performances for different types of surfactant-stabilized emulsions, such as n-hexane – water, dichloroethane – water, petroleum ether – water, and toluene – water emulsions. The crosslinking process proved to be an effective method to increase the superoleophobicity of the membranes including in harsh environment. In addition, the stability performance of the membrane was also relatively high after several cycles of filtration process.

In addition to graphene-based nanosheets and CNTs, halloysite nanotubes have also been attempted to be incorporated in the polymer matrix. Halloysite is hydrophilic in nature due to the presence of hydroxyl functional groups (- OH) and exhibits a high specific surface area [110]. The presence of many -OH groups on the membrane surface is the critical factor determining both membrane hydrophilicity and water permeability and thus anti-fouling properties during the oily wastewater treatment process. The incorporation of halloysite nanotubes or the composite of halloysite and ferrihydrate into PES matrix decreased the water contact angle of the produced membranes and finally improved the flux and oil rejection capacities of the membranes [110,146,147]. Current studies on MMMs and nanocomposite membranes for oily wastewater treatment processes have attempted to utilize emerging materials, such as MOFs. The zeolitic imidazolate framework (ZIF) and MOF-808 have been blended with a polymer matrix to form MMMs for

oil-water separation [148,149]. MOF-808 was selected because it possesses excellent stability, high adsorption capacity, and it is relatively easy to modify [149]. MOF-808 can be modified with EDTA, and the modification and incorporation of MOF-808 into PAN matrix could improve membrane's separation efficiency, especially to adsorb selected ions presented in oily wastewater. In addition, the membrane demonstrated excellent recyclability and corrosion resistance. Overall, the membrane is highly efficient in treating wastewater.

The performance of single filler-based MMMs and nanocomposite membranes can be potentially improved by utilizing dual fillers inside the polymer matrix. The efforts to combine more than one filler have just been started recently [109,117,152]. Combining fillers is considered a good way to either improve the dispersion of particles inside the polymer matrix or maximize each material's separation capacities, including polymer. For instance, the combination of TiO_2 and hydrous manganese oxide (HMO) was successfully incorporated into PES matrix to form MMMs. The presence of HMO could not only minimize the costly TiO_2 consumption during the membrane fabrication process but also helped TiO_2 to increase the overall hydrophilicity of the PES membranes. The presence of HMO-TiO_2 dual fillers could produce higher water flux compared to pristine PES membranes. In addition, the MMMs exhibited a significantly lower degree of flux decline due to improved surface resistance against oil fouling and are of potential for long-term operation [109].

6.1.3. Membranes with different pore geometry. Membrane pores geometry has been considered one crucial aspect of the efforts to minimize fouling of membranes during oil-water separation processes [36,78,114,115]. Membranes with circular geometry have been used more frequently than membranes with other geometries due to their versatile and easy fabrication. However, the fouling tendency of membranes with circular geometry is relatively high. Hence, several studies developed and analyzed membranes with slotted pore geometry from metal [36,76]. In principle, different mechanisms govern the droplet passing through the circular and slotted pore membranes when used for oil-water separation processes. For circular pores membrane, the transport of oil droplets is governed by the trans membrane pressure (TMP). On the other hand, the drag force around the droplets induced by the motion of the fluid determines the transport of oil drops through the slotted pore membranes. In a circular pore membrane, the fouling tendency of membranes by the deformation of big oil drops is relatively high. However, in a slotted pore membrane such as shown in Fig. 7, the big oil drops are not possible to completely plug the slot and there will always be space available around the drops for the permeate flow to pass through. Hence, the pressure differential forcing the retained drops through the pores of a filter will be different: in the case of the plugged circular pore filter, it is the pressure drop across the filter, whereas, in the case of the slot, it is the liquid drag that acts to force the drop to deform and pass into the permeate [36,76].

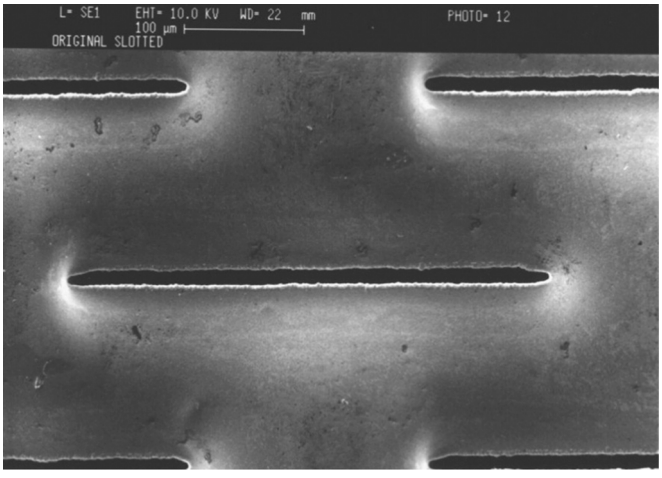

Fig. 7. SEM image of the surface of slotted pore filter.
(Reproduced with permission from [115]).

In a typical study, Holdich et al. [161] employed slotted true surface filters with narrow pore size distributions and maximum pore diameters of 6.8 and 8 μm to separate oil emulsions of drop diameters similar, or below, those used in the development work undertaken for the testing of hydro cyclones in the North Sea oil industry. The filters operated successfully and provided stable flux rates and rejections up to 96% under various operating conditions. The operating pressure required to produce permeate flow was a small fraction of that needed when filtering with conventional microfiltration membranes. For some tests, the filter tubes contained tightly fitting helices of pitches: 14, 42 and 70 mm to induce rotational flow and minimize oil drop deposition suspended in water. For these experiments, filtrations were run under constant flux and pressure conditions, and this filtration process needed low values, ranging from 3 to 9 kPa. Another experiment using true surface filters conducted by Ullah et al. [115]. They tested a slotted pore membrane with 4-μm slot width and 400 μm slot length with oil emulsion. The shear rate on the surface of membranes helped to reduce the increase of TMP, and the membranes could produce permeate flux as high as 1200 LMH. Cumming et al. have also investigated oil using a stirred cell fitted with Nuclepore filters with pore sizes of 2, 5, 8, and 10 μm [31]. The filters are produced by nuclear bombardment followed by chemical etching. They can be classified as true filters because they have pores that pass directly through the filter from one side to the other side. A simple theory based on interfacial tension, contact angle, and pore size is described. The results are compared with experimental results, in which treated oil emulsion drop size in the range 1–40 μm stabilized by polyvinyl alcohol. There is a good similarity between model and experiment for the 2 μm filter, but the model reliability decreased with increasing pore size. A mathematical model to explain the interaction of oil drop and slot of the membrane was also developed by Kosvintsev et al. [76]. In another study, Darvishzadeh et al. [114] examine the influence of fluid properties on the critical pressure of permeation of an oil micro-droplet into a slotted pore by employing the Navier-Stokes equation. A model was developed to estimate the critical permeation pressure from a force balance model that involves the drag force from the flow around the droplet and surface tension forces as well as the pressure variation inside the pore. Several factors have been found that determine the critical pressure, including oil-to-water viscosity ratio, surface tension coefficient, contact angle, and droplet radius. The results from this study are beneficial to optimize the design and performance of microfiltration systems with slotted pores during the oil-water filtration process.

6.1.4. Surface engineering of inorganic membrane. The successful treatment of oily wastewater using membrane requires enhancement in permeate throughput and oil rejection efficiency of the membranes. To achieve these goals, surface treatment of membrane is considered key to improving membrane's characteristics during oil-water separation. In terms of surface modification of membranes for oily wastewater, researchers have tried to modify the surface to make it more hydrophilic. Hence, in recent years, surface modification research has focused on finding materials and techniques that can either change the functional groups of the membrane's surface or coat the membrane's surface with more hydrophilic materials. Both organic and inorganic membranes for the oily wastewater treatment process have been successfully modified.

In terms of inorganic membranes, different inorganic membrane materials including steel mesh, ceramic, glass, and metal have been tested and modified. Various chemicals and techniques have been used, including poly(dimethylsiloxane) (PDMS) [162], ionic polymer [163], fluorosilane and GO [164], silane compounds [165], ZIF-8 [166], homogeneous emulsion containing low surface-energy material (teflon,

adhesive polyvinyl acetate, dispersant polyvinyl alcohol, surfactant sodium dodecyl benzene sulfonate, and thinner [72], hydrogel [73], nano-sized ZrO_2 [70], nano TiO_2 [71,167], oxygen plasma and polymer coating [168], fluoroalkylsilane (FAS) [169], poly(acrylic acid) grafted poly(vinylidene fluoride) (PAA-g-PVDF) nanofibers [170], cupric phosphate ($Cu_3(PO_4)_2$) [171], and polydopamine [172,173].

In recent years, inorganic membranes such as mesh membranes have also transformed into what is currently known as Janus membranes. In most cases, the Janus membrane is referred to as a membrane with opposing properties opposing wettability on each surface [174]. In their study, Liu et al. [175] fabricated a Janus wire mesh by a combined laser structuring and fluorosilane/GO modification of the two sides of the mesh, respectively. Process production of the Janus membrane is presented in Fig. 8a. The Janus membranes were then utilized for oil–water separation process. The mesh membranes for oil-water separation can be simply classified into two categories, the superhydrophobic/oleophilic mesh membranes, and the hydrophilic but under-water superoleophobic mesh membranes. The former realize oil-water separation by unidirectional oil transportation since water cannot pass through the mesh due to the superhydrophobicity; whereas the latter separate oil from water by discharging water, because oils that are lighter than water cannot permeate through the superoleophobic water/mesh interface. For this circumstances, Janus membrane is a good option for the mesh membrane used in their study. The basic principle is creating a superhydrophobic and superoleophilic surface on the wire mesh. The separation performances of the Janus membranes were very good where the oil contents in water were measured to be 0.02%, 0.01%, 0.02%, 0.03% and 0.05% by weight for the separation for bean oil/water, n-heptane/water, methylbenzene/water, water/perchlormethane and water/trichloromethane mixtures, respectively. Water residuals in bean oil, n-heptane, methylbenzene, perchlormethane and trichloromethane were measured to be 0.04%, 0.02%, 0.05%, 0.07% and 0.08%, respectively as can be seen in Fig. 8b. These results suggested that the Janus mesh shows very high separation efficiency for various oil/water mixtures. The Janus mesh is also very stable, indicating its potential to be used in a practical situation.

6.1.5. Surface modification of organic/polymeric membranes. For polymeric membranes, surface modification of membranes is directed to increase the membranes' hydrophilicity to reduce the membranes' fouling tendency during oil–water filtration. Various materials and methods have been attempted. Such materials and techniques include: PDA [142,151,152,176–181], co-deposition of PDA and PEI [129,174], hydrophobic 1H, 1H, 2H, 2H-perfluorode cyltriethoxysilane (PFTS) modification (F–SiO_2/PVDF) followed by a unilateral plasma-etching treatment (P–SiO_2/PVDF) [182], hydrophilic polyacrylonitrile electrospun nanofiber (PANEN) membrane with a single-side hydrophobic CNTs network coating [183], poly(styrene-co-maleic anhydride) (PSMA) and polyethyleneimine (PEI) [127], conductive polymer PANI (polyaniline) [124], monoethanolamine (MEA)-diethanolamine (DEA) [184], octadecyltrimethoxysilane (OTMS) [185], levodopa (L-DOPA)/3-amino-propyltriethoxysilane (APTES) reaction [186], deposition of a polyamide (PA) layer using interfacial polymerization [143], plat polyphenol [187], sodium bis(2- ethylhexyl) sulfosuccinate [188], SPAN 80 [189], poly (ethylene glycol) diacrylate (PEGDA) via low-pressure plasma-induced graft polymerization [190], ethylenediamine tetraacetic acid disodium salt [191], hydrophilic hydroxyethyl acrylate (HEA) monomer [192], Tannic acid (TA) and ferric ions (FeIII) [193], tannic acid and diethylenetriamine [194].

One material that has been used more frequently than any other material to modify the membrane's surface is polydopamine. Polydopamine (PDA) has been considered one material that can alter the hydrophilicity of the membrane's surface. Since the breakthrough discovery by Messersmith and co-workers [195], the facile method to coat materials with PDA has been widely implemented for different applications, including nanotechnology and membrane separation,

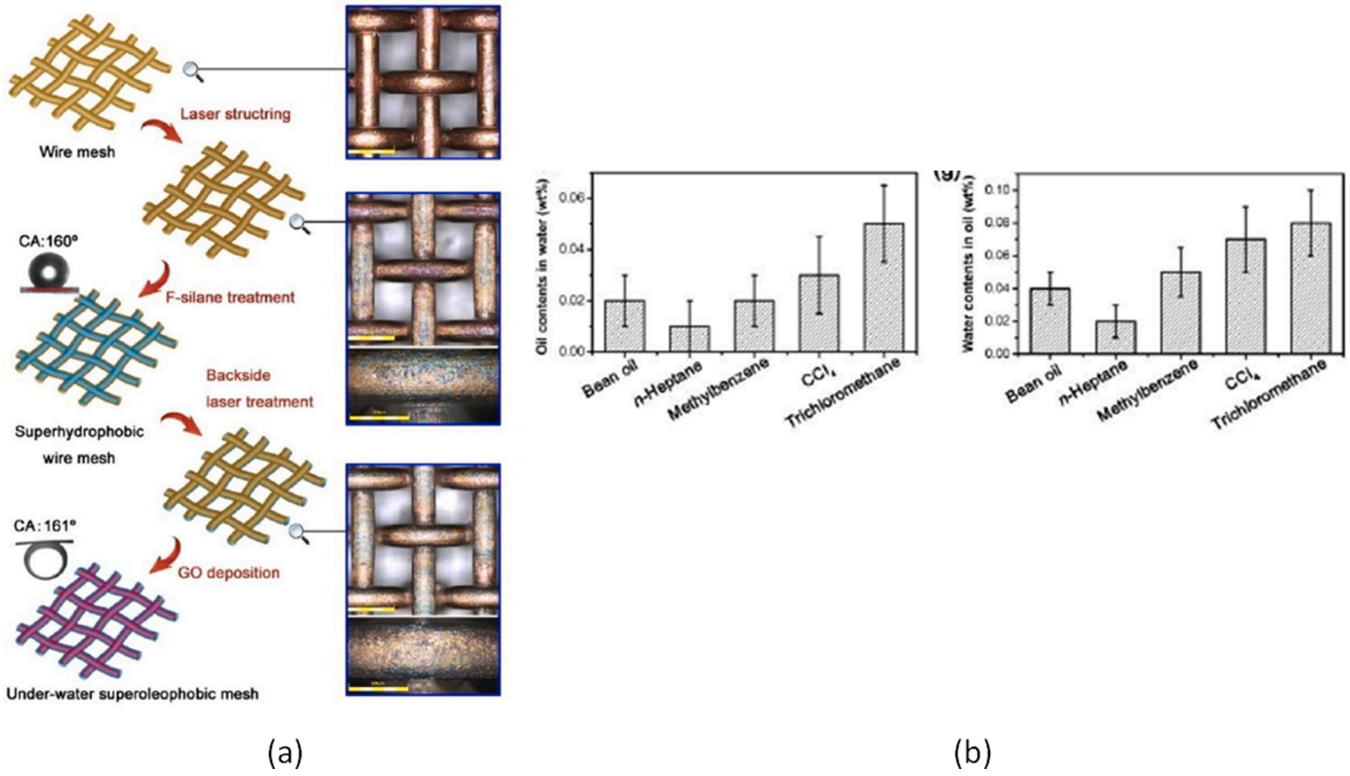

Fig. 8. (a) The surface modification of mesh to produce Janus membrane and (b) Separation efficiency of Janus mesh membrane: (left) residual oil content in water and (right) residual water content in various oils.
(Reproduced with permission from [175]).

especially the fabrication of MOFs membranes [196,197]. In nature, mussels have shown their capability to attach to virtually all types of surfaces. This ability may arrive from the amino acid composition of proteins discovered near the substrate interface that contains plentiful 3, 4-dihydroxy-L-phenylalanine (DOPA) and lysine amino acids [195].

Based on this postulation, PDA has been applied to coat various substrates, including metals, metal oxides, ceramics, synthetic polymers, and an extensive range of other hydrophilic and hydrophobic materials. The polymerization of dopamine was carried out inside tris(hydroxymethyl)aminomethane hydrochloride (Tris HCl) buffered dopamine solution [195,198]. The PDA coating technique applied resulted in a thin PDA layer and robustness. One-step coating of PDA is also efficient compared to layer-by-layer coating and Langmuir-Blodgett deposition. Despite its success as a coating layer, the chemical structure of PDA has not yet been fully investigated [198,199]. Some proposed structures of PDA are depicted in Fig. 9. In scheme I, Messersmith and co-workers proposed a typical structure where the oxidized and cyclized monomers were covalently linked through aryl-aryl connections. Analogous covalent-based structures were also proposed, as shown in schemes II and III in Fig. 9. In contrast, scheme IV proposed a stack of oligomers covalently attached via π-π and noncovalent connections to develop supramolecular complexes [198].

Considering the abundant hydrophilic groups, PDA has been used as hydrophilic modifiers for hydrophobic polymer membranes. One example is a study conducted by Xiang et al. [200], who prepared a PDA modified PVDF membrane and investigated immersed time's effect on membrane performance. The hydrophilicity and superoleophobicity under seawater were achieved for the membrane immersed in dopamine solution for 24 h. The modified membrane obtained stable superoleophobicity under seawater with an oil contact angle of 152 ± 0.3° and extremely low oil-adhesion. The permeability and selectivity of the modified membrane were significantly higher than that of traditional filtration membranes. Moreover, the introduction of PDA coating endowed the membrane with good fouling resistance to proteins and quick flux recovery after membrane cleaning. In another study, membranes with SiO_2 nanoparticles anchored PVDF@PDA membrane via simple hydrolysis and physical blending process. Underwater oil contact angels revealed that the superoleophobicity property of PVDF@PDA@SiO_2 was obtained after nano-SiO_2 modification, leading to a better performance in dichloroethane/water emulsion separation [112].

Even though PDA coatings have successfully improved the properties of membranes used for the oil-water separation process, PDA-based coatings still have their limitations. It is argued that during PDA coating, chemicals for post-functionalization are restricted to those containing amine or thiol groups. In addition, the coating process is still relatively time-consuming [201], produces inhomogeneous roughness [202], inadequate surface wettability [203], and poor stability in extreme pH conditions [204]. To improve the polymerization process of dopamine to PDA, the addition of an aggregation-prevention agent could be beneficial. In recent studies, the aggregation of PDA can be prevented by the presence of PEI based on Michael-addition or a Schiff-based reaction between amine and catechol groups present in both chemicals [205,206]. This co-deposition process was very effective in shortening reaction time and preventing PDA aggregation. A study conducted by Fang et al. [113] successfully co-deposited PDA/PEI/TA on PP membrane support to produce a hydrophilic and underwater superoleophobic microfiltration membrane based on Michael addition and Schiffs base reactions. PEI provides lots of amino groups, which can form strong covalent bonds with catechol/pyrogallol groups (derived from TA or DA molecules) via Schiff base/Michael addition reactions, increasing the stability of the coating. The as-prepared TA/DA/PEI coating shows great wettability and outstanding stability, due to that, PEI can crosslink with TA and DA. After the deposition process, the produced membrane showed excellent hydrophilicity, outstanding acid, alkali, and organic solvent resistance, as illustrated by Fig. 10. Using the membranes, oil-water separation filtration could produce permeate flux of more than 475 L m^{-2} $h^{-1,}$ and separation efficiency could reach

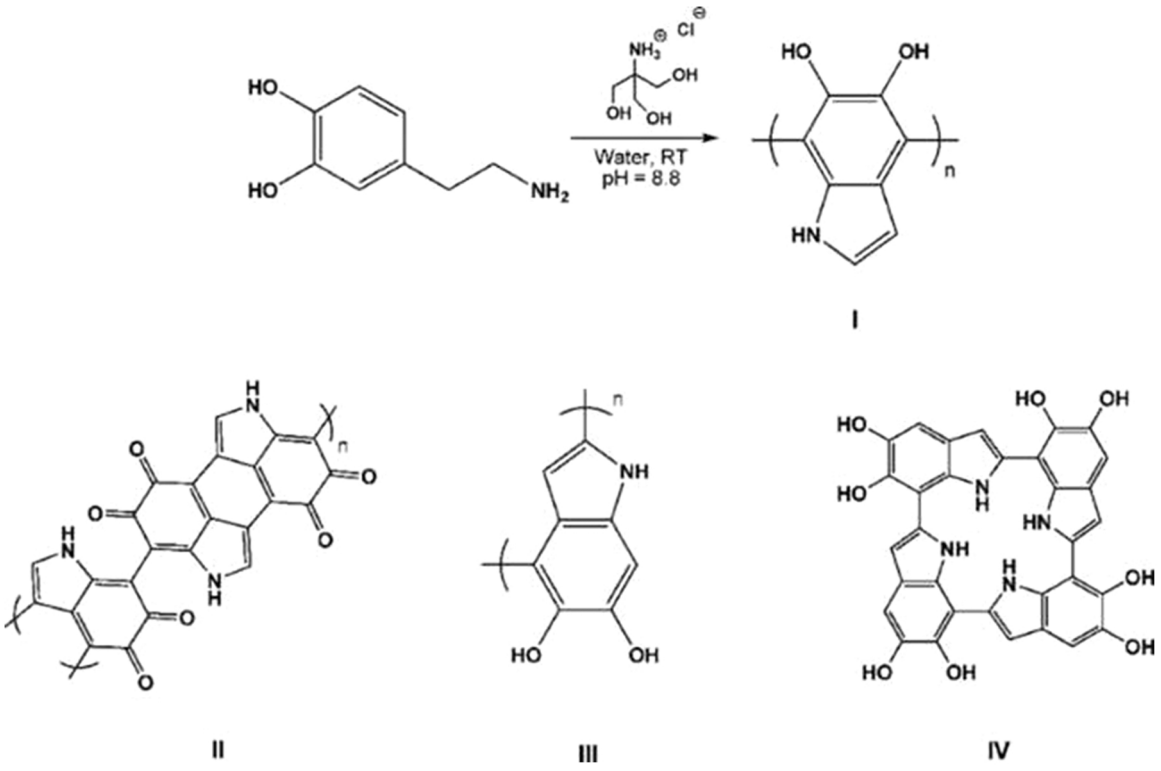

Fig. 9. (Top) Synthesis and structure of poly(dopamine) as proposed by Messersmith and others and (bottom) additional proposed structures of poly(dopamine). (Reproduced from [198] with permission).

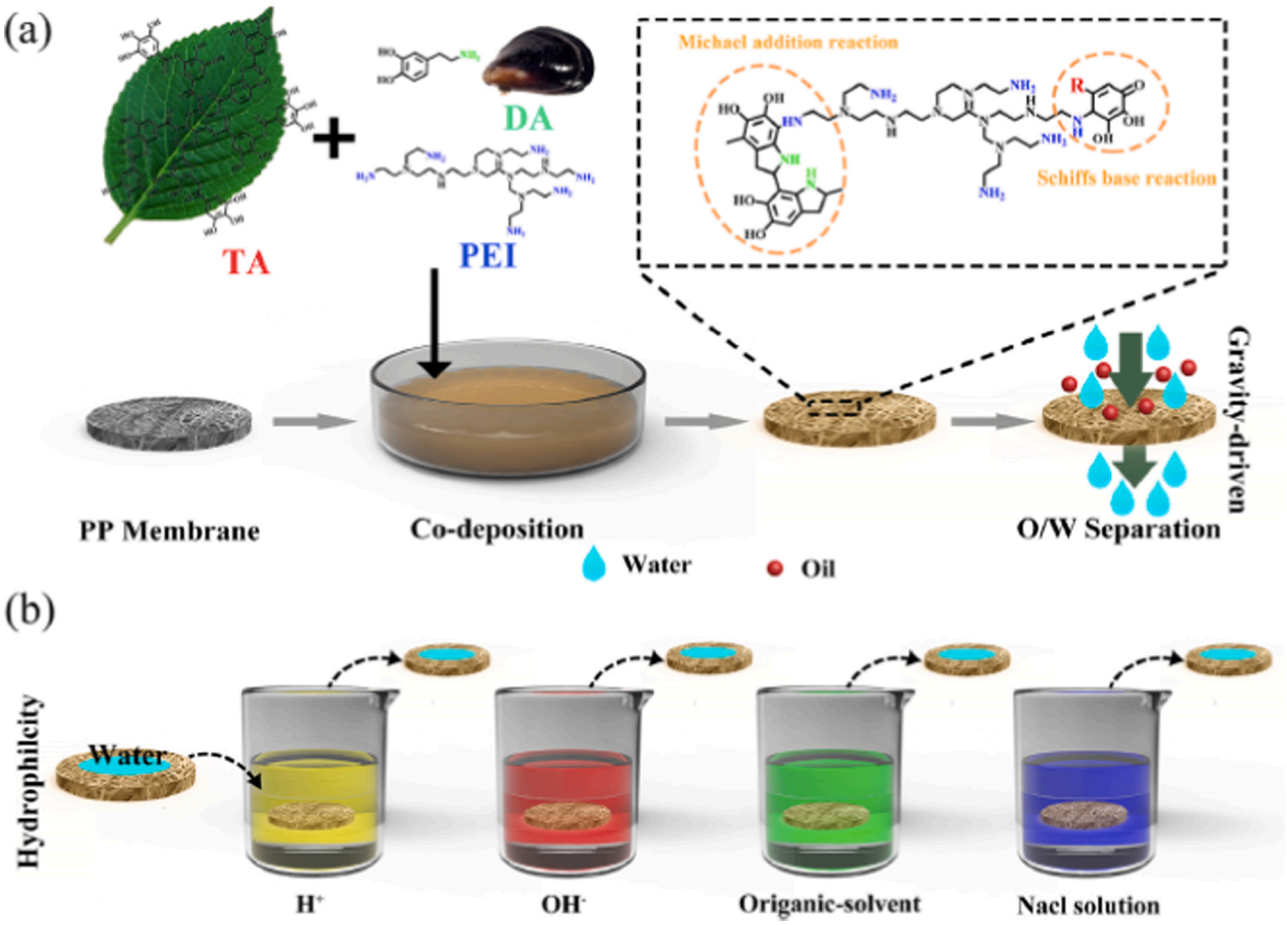

Fig. 10. (a) The illustration of the co-deposition process of TA/PEI/DA on PP membranes and (b) the stability of produced membrane after the co-deposition process. (Reproduced with permission from [113]).

99.8%. These results depict the potential of mussel-inspired chemicals for the surface modification process of the membrane in oil-water separation process. The utilization of PDA combined with MOFs particles, such as UiO-66-NH_2 nanoparticles, to produce super wettable property of composite membrane was also conducted in a recent study [207]. Hydrophilic MOFs nanoparticles and PDA functionalization improved the hydrophilicity of PVDF substrate. The membranes from the study were not only able to separate tetracycline from surfactant stabilized emulsion, but were also able to demulsify various emulsion with very high membrane rejection efficiency.

Similar to inorganic membranes, the efforts to produce Janus membrane with opposing properties for oily wastewater treatment process using polymeric membranes have been started recently [128, 129,155,182,183]. The main motivation behind the development of Janus membranes is that the surface wettability mainly determines the oil/water mixture separation at the interface between liquid and solid. The superhydrophobic/superoleophilic materials can selectively filtrate the oils and vice versa. However, most current membranes used for oil/water separation only have a single-sided wetting selectivity. Only one specific type of oil or water can be separated from oily mixtures, severely impeding the practical membrane applications in comprehensive emulsions separation. In a recent study by Lin et al. [182], PVDF porous supports were transformed into Janus membrane by using surface silicification followed by a hydrophobic 1H, 1H, 2H, 2H-perfluorodecyltriethoxysilane (PFTS) modification (F–SiO2 surface) incorporated with unilateral O_2/N_2 plasma-etching treatment (P–SiO_2/PVDF), as depicted in Fig. 11. Janus membranes from this study exhibited opposing surface wettability where their contact angle difference between the two sides reached up to 150°. The filtration performances of Janus membranes showed permeate flux of 7200 L m^{-2} h^{-1} bar^{-1} for oil-in-water emulsion. The separation efficiencies up to 99.8% could be achieved through the Janus membranes. In addition, the Janus membranes showed excellent reusability, where a nearly 100% recovery ratio of permeating flux was obtained after several cycles of oil-water separation tests. Another technique to develop Janus membrane has been conducted by biomimetic PDA interface regulation and superhydrophobic attapulgite (SOATP) spraying of regenerated cellulose membrane [208]. Janus membranes from this study produced very good rejection of more than 99% for oil – in – water and water – in – oil emulsions. The flux during oil – in – water emulsion filtration was higher than the flux during water – in – oil emulsion filtration. This might be due to the difference in particle sizes of the dispersed phase in each emulsion and the difference in pore size of surfaces available in Janus membranes. In addition, the membranes showed stable performance in terms of cycle time and chemical stability.

Another interesting progress on the development of membrane material for fouling mitigation is membrane with self - cleaning ability. Self - cleaning ability of the membrane ensures the minimization of fouling propensity of the membrane. In a recent study, self - cleaning membranes have been successfully fabricated by coating NH_2-MIL-88B on quartz fibrous membrane through solvothermal technique. The incorporation of NH_2-MIL-88 increased the hydrophilicity of the membranes, however the porosity of the membranes decreased. This created lower flux and higher oil rejection efficiency from modified membranes compared to unmodified membranes. Compared to unmodified membranes, the modified membranes from this research showed self - cleaning ability that was created by a possible photo – Fenton self - cleaning mechanism. The self - cleaning ability of the membranes could

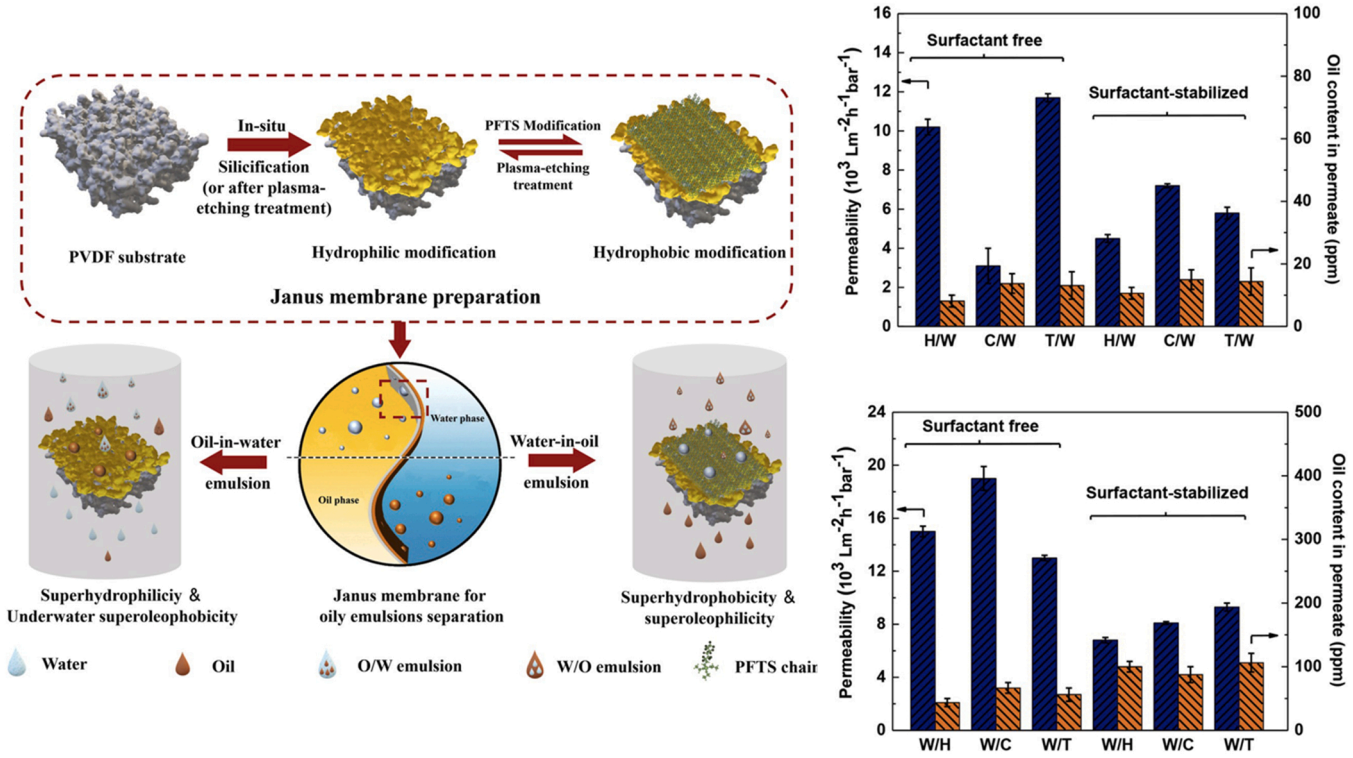

Fig. 11. (a) Schematic representation of the fabrication of PVDF-based Janus membranes and (b) Filtration performances of Janus membranes: (upper diagram) oil–in–water and (below diagram) water–in–oil filtration tests.
(Reproduced with permission from [182]).

potentially retain a relatively lower flux decrease compared to membranes with no NH_2-MIL-88 [209]. Another study was also conducted to produce membrane with self - cleaning ability [210]. Layered double hydroxide or LDH was utilized and combined with carbon cloth to produce membranes that were intended to separate oil from water. To improve the separation performance, the carbon cloth/LDH membranes were immersed in $AgNO_3$ solution. The deposition of Ag provided self – cleaning ability to the membranes that was supported by stable long - term performances of the composite membranes.

6.2. *Perspective from operational management*

In addition to the membrane's material and surface modification, the operating conditions during filtration are also essential to control the extent of fouling. Fouling is affected by the feed and membrane properties and the operational parameters such as crossflow velocity, shear rate, and transmembrane pressure, temperature, and pH [12,211]. For a membrane study in rejecting oil droplets, it was found that oil droplets should be retained entirely, and the continuous phase is permeated. However, because the oil droplets are deformable, they can be squeezed through the pores and contaminate the permeate depending on the applied pressure. Nazzal and Wiesner investigated the effects of transmembrane pressure and membrane pore size on the deformation of oil droplets through the membrane's pores [104]. If the transmembrane pressures were set below a critical pressure, emulsion rejection efficiency could be maximized. Another study using slotted pore membranes suggested that the critical permeation pressure weakly depends on the droplet radius [114].

Koltuniewicz et al. investigated the microfiltration fouling for separating oil-in-water emulsions by using three types of microfiltration membranes [212]. They performed simultaneous experiments in both dead-end and cross-flow modes at various pressures, and the latter case, at different cross-flow velocities. After an initial phase of pore blocking, the buildup of a layer of droplets and the resultant switching to cake filtration dominated the membrane's performance [114,212]. Because of membrane fouling, the filtration process has to be stopped frequently for membrane cleaning to restore membrane permeability. Membrane cleaning, as well as other measures for fouling control, significantly increased the cost and complexity of the processes. Another strategy to reduce fouling tendency in terms of operational parameters is by controlling the cleaning solution's pH or temperature. A study by Dunderdale et al. [213] exhibited an effective cleaning of membranes after oil filtration after the pH was reduced. Oil drops tend to strongly adhere at high pH because of the more oleophilic surface, resulting in increased contact angle hysteresis.

Creating a high shear rate on the surface of the membrane is another way to minimize the fouling tendency. The shear rate can be generated in various ways, such as employing a cross-flow mode of operation, mixing the feed solution in the dead-end mode of operation, rotating the membrane [214], employing oscillation on the membrane surface [215–217], utilizing vibratory shear enhancing process (VSEP) [218]. Oscillations at the membrane surface can be developed by oscillating the membrane surface itself or inside the flowing liquid. The oscillation can enhance the heat and mass transfer on the top of permeate flux. The generated shear by oscillation reduces the possibility of the oil drops moving toward the membrane surface. In addition, the oscillation also reduces the transmembrane pressure, increasing the oil rejection and reducing the propensity of fouling [216]. The deformation of oil drops through slotted pore membranes under oscillation conditions was studied by Ullah et al. [215]. Oil emulsions with different interfacial tensions, i.e. 4 and 30 mN m^{-1} were filtered at four vibrational shear rates, i.e., 0, 1200, 3200, and 8000 s^{-1} and two permeate fluxes, i.e., 200 and 1000 L m^{-2} h^{-1}. The experimental results exhibited that oil with lower interfacial tension was more deformable because the increase in TMP was less due to the passage of even larger drops to the slot opening. In addition, the oil rejection efficiency was also lower.

In addition to controlling operational parameters during the filtration process, membrane cleaning plays an important role in minimizing the propensity of fouling. A common technique to clean the membrane is

by using the backwash or backflush method, where the direction of the permeate flow is reversed to dislodge foulants accumulated on the membrane surface or inside the pores. Backflush was found to be more effective at higher applied pressures, higher crossflow velocities, and lower oil concentrations [219]. In another, a short duration of back-flushing was found to be effective when a zirconia tubular membrane was utilized to treat oil emulsions [220]. Backwash using hot water and the alkaline solution was also attempted for ceramic membranes [221] and the process could recover over 95% of the initial flux with almost no change in oil rejection efficiency. Oil droplets that adhere for a long time on the membrane's surface could be removed by using a hot alkali solution as the backwash solution. In other studies [222,223], chemical cleaning was also conducted to minimize fouling phenomenon. The sodium hydroxide (NaOH), disodium ethylenediamine tetraacetate (EDTA), sodium dodecyl sulfate (SDS), sodium pyrophosphate (SPP) and hydrochloric acid (HCl) were studied and used in combination to clean polyamide-based nanofiltration membranes [222]. The formulation of cleaning chemicals and procedures was developed. The optimized formulation consisted of 0.05% EDTA, 0.2% sodium pyrophosphate, 0.2% SDS with optimized pH of 11.0. The cleaning steps should follow two consecutive procedures, i.e., cleaning with the alkaline formulated cleaning agent for 0.5 h followed by cleaning with HCl solution (pH=2) for 0.5 h.

Another way to avoid membrane fouling is to combine membranes with different driving forces and modes of operation and also to combine membranes with non-membrane techniques as pretreatment before the oily wastewater is treated inside the membranes [40,218,224,225]. Zhang et al. [218] studied the combination of UF membrane operated in a VSEP machine with a chemical demulsification process. The demulsification or emulsion breaking process was applied as a pretreatment step to reduce the organic loading and increase the floc size distribution of oil droplets, and then effluent was further treated by VSEP to remove the residual oil. Through this combination, the UF membrane's flux can be maintained up to 245 L m^{-2} h^{-1} with lower oil content in the permeate compared to using VSEP alone. The combination of Forward Osmosis (FO) and Membrane Distillation (MD) was also attempted in another study [225]. The study to combine these two processes was stemmed from the idea that by using FO to treat oil-water emulsion, a stable water flux with high recovery and high rejection of the contaminants may be achievable. It would effectively recover a large portion of water for reuse and significantly reduce the amount of concentrated waste stream for disposal. MD after FO is necessary to continuously re-generate the draw solution for FO and reused in areas where high salinity water is not available in a large amount. Their experiments exhibited several promising results, such as large water flux and high oil removal ratio were observed through FO process, while ultrahigh NaCl rejection was shown by MD process. In addition, high water recovery (90%) was achieved by the combined FO and MD at high feed salinity, and ultimately rejected oil and NaCl were also achieved by the combination of FO and MD.

7. Future outlook of membrane for oil–water separation

The future application of membrane in the oil-water separation process depends on developing techniques to minimize and reduce the fouling phenomenon by oil droplets and other contaminants. As discussed in previous sections, fouling caused by oil droplets deformation has generated problems on membrane flux and oil rejection efficiency. In addition, the presence of other contaminants has been considered a severe problem for the real application of membrane in the oil-water separation process. Up to now, several membrane companies have commercialized their membranes to be used in oil filtration. Such companies include Osmonics, Filtration Solution Inc., Clean Water Tech PTE, Ltd., Koch, Veolia Water Technologies, and Hydranautics [12,226,227]. Most commercial membranes available are UF membranes in different module configurations, and most of them are fabricated from polymeric materials. The low cost and versatility of polymeric membranes compared to inorganic membranes are the major driving forces for the commercialization of polymeric membranes. However, as polymeric membranes are susceptible to the fouling phenomenon, future membranes for the oil-water separation process still need to be improved in their anti-fouling capacities.

Anti-fouling polymeric membranes urgently need to be synthesized to improve the commercialization of membranes for the oily wastewater treatment process. Modification techniques that have been discussed in this review should be deeply studied and upgraded to a large scale. Such promising modification techniques include polymer blending, mixed matrix membranes and nanocomposites membranes, and membrane surface modification. Since most modification techniques are still on a laboratory scale, this is an opportunity for further research to improve the techniques and scale up the techniques. Another way to enhance the broad application of membranes in oil filtration is to combine a particular membrane process with other membrane processes or combine membranes with other technology [228]. The combination of forward osmosis and membrane distillation has shown a promising alternative to improve membrane capacity in treating oily wastewater.

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CRediT authorship contribution statement

Putu Doddy Sutrisna: Conceptualization, Data curation, Writing – original draft. **Kiki Adi Kurnia**: Data curation, Writing – review & editing. **Utjok W.R. Siagian**: Writing – review & editing. **Suryadi Ismadji**: Writing – review & editing. **I Gede Wenten:** Conceptualization, Writing – review & editing.

Declaration of Competing Interest

The authors report no declarations of interest.

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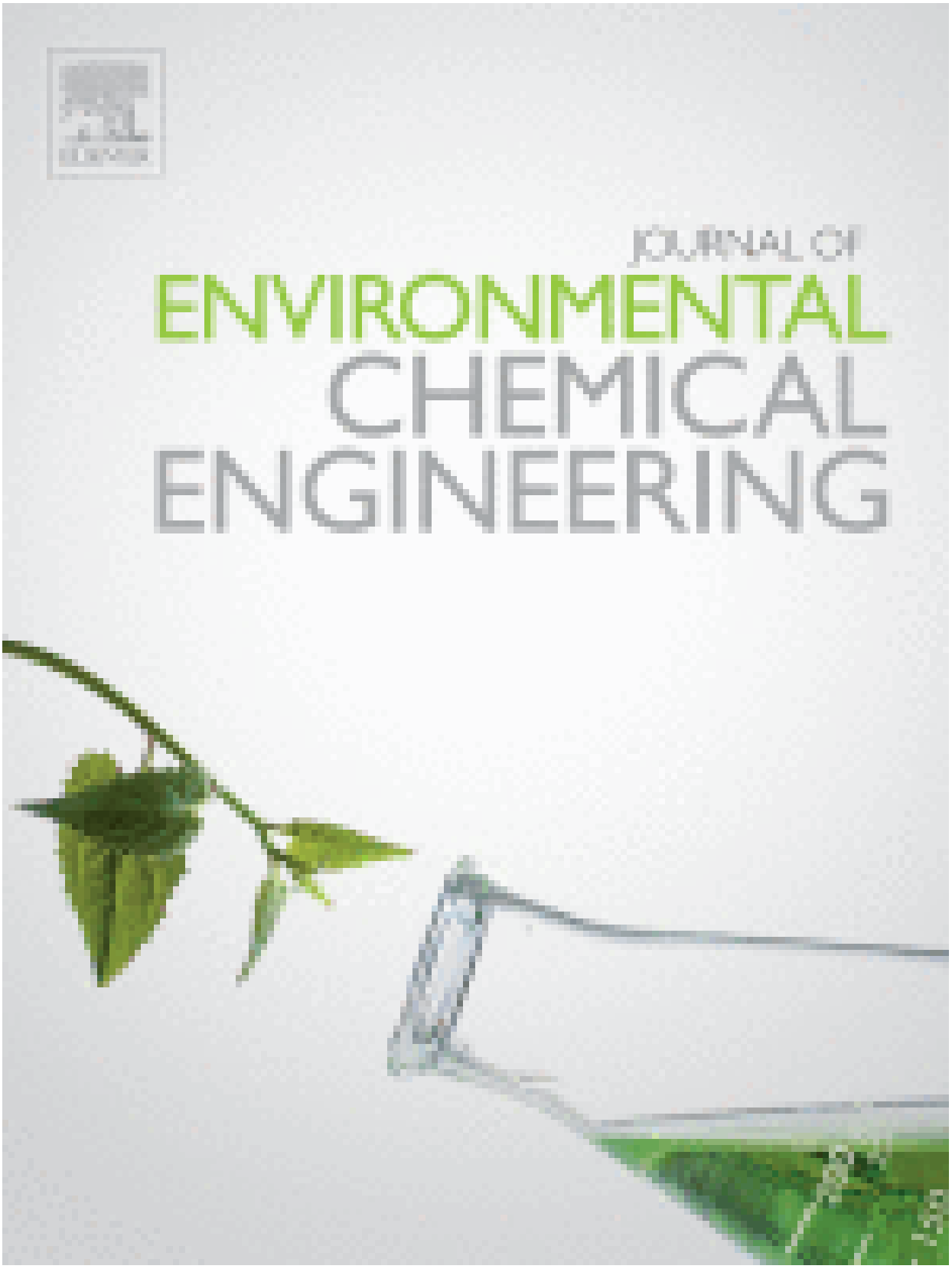
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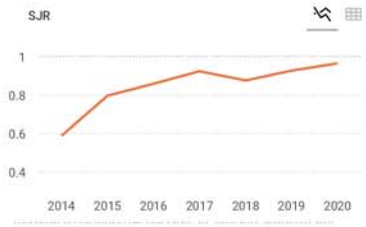

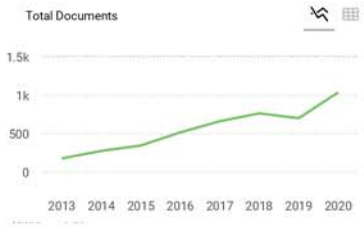

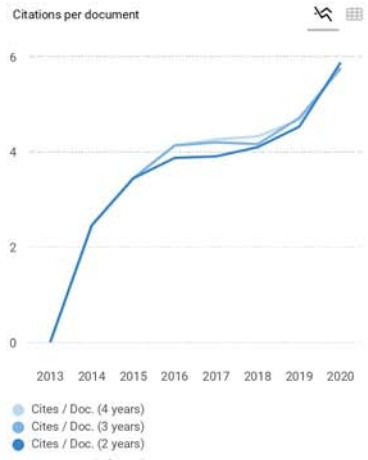

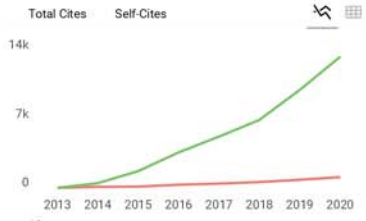

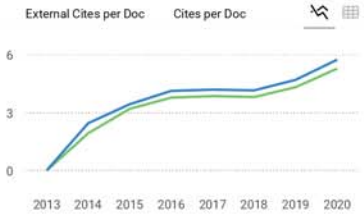

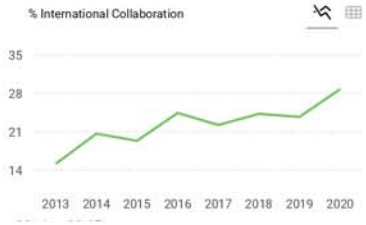

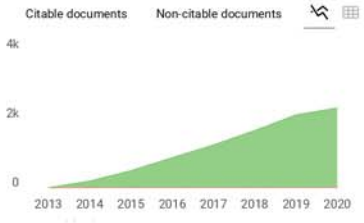

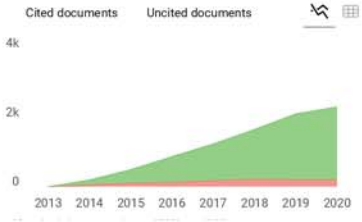

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A **Ali** 7 months ago

Hi
How can publish a letter to the editor in this journal?
my letter is about a previously published article in this journal that I found there are some misleading data in that paper.

Best regards.

reply

SCImago Team

Melanie Ortiz 7 months ago

Dear Ali, thank you very much for your comment, we suggest you look for the author's instructions/submission guidelines in the journal's website. Best Regards, SCImago Team

C **Chi Lye** 2 years ago

In June 2020, I submitted my manuscript to Journal of Environmental Chemical Engineering. After more than 2 weeks with the editor, the paper was rejected for "receiving more submissions than the journal would approve".
I just wonder, Did the editor have to keep me waiting for more than two weeks to tell me that they could not proceed to review the paper because they got a lot of articles? This is ridiculous!

reply

Abhishek 2 years ago

Is this an SCIE indexed journal?. Is this journal recently been added to the SCIE list?

SCImago Team

Melanie Ortiz 2 years ago

Dear Abhishek,
Thank you for contacting us.
SJR is a portal with scientometric indicators of journals indexed in Elsevier/Scopus. Unfortunately, we cannot help you with your request referring to the index status. We suggest you consult Scopus database (see the current status of the journal) or the mentioned database for further information. You can also check that information in the journal's website or contact directly with the editorial staff.
Best Regards, SCImago Team

K **Khalid Z. Elwakeel** 2 years ago

The journal received 2019 Impact Factor of 4.3

reply

M **Mohammad Ali Nasiri** 2 years ago

https://www.sciencedirect.com/journal/journal-of-environmental-chemical-engineering

SCImago Team

Melanie Ortiz 2 years ago

Dear Khalid,

Thank you for that information. Could you please reflect your data source?

Best Regards, SCImago Team

N **Neghi** 2 years ago

Hi..what is the source record id of 'journal of environmental chemical engineering' in scopus indexes journals

reply

SCImago Team

Melanie Ortiz 2 years ago

Dear Neghi, thank you very much for your comment, unfortunately we cannot help you with your request. We suggest you to consult the Scopus database directly. Remember that the SJR is a static image of a database (Scopus) which is changing every day. Best regards, SCImago Team

A **A. R.** 3 years ago

Hi
This journal is apparently not on the JCR of 2019, when will the journal be granted by impact factor? Isn't it the time after six years of publication?

reply

J **Jafar Ali** 3 years ago

Is SCI mago means the same term used for sci journals? or it is a different category to rank journals ??? kindly guide about the status of Journal of Environmental Chemical Engineering.

it's not in the web of science and Journal Citation Report (JCR) ?

and it's in 5th year still no impact factor, because I have published one paper in this journal and to graduate, I need 2 sci papers will it fulfill my requirement or not?

Thanks looking for your kind response

reply

SCImago Team

Scopus data, our impact indicator is the SJR. Check our page to locate the journal. We suggest you consult the Journal Citation Report for other indicators (like Impact Factor) with a Web of Science data source.

Best Regards,
SCImago Team

K **Khieu** 3 years ago

Hi.
Does this journal belong to ISI?
Thanks.

reply

E **E.S. Agudosi** 3 years ago

Yes

SCImago Team

Elena Corera 3 years ago

Dear Khieu, SCImago Journal and Country Rank uses Scopus data, our impact indicator is the SJR. Check our page to locate the journal. We suggest you consult the Journal Citation Report for other indicators (like Impact Factor) with a Web of Science data source. Best Regards, SCImago Team

A **atieh** 4 years ago

Hi
has this journal requested for an impact factor?

reply

SCImago Team

Elena Corera 4 years ago

Dear Atieh,

thank you very much for your comment. Unfortunately, we cannot help you with your request, we suggest you contact journal's editorial staff so they could inform you more deeply.

Best Regards,
SCImago Team

O **Olukanni** 4 years ago

In this case, is the IF equivalent to 0.92 or 3.74

reply

SCImago Team

Elena Corera 4 years ago

Dear Olukanni, the impact indicators are not exactly equivalent to each other. These are two different calculations that provide different values. In the case of the IF is the value is an average of the number of citations, in the case of the SJR the value is a position in the thematic category. Best Regards

F **Farhad** 4 years ago

When does this journal have an impact factor?

reply

SCImago Team

Elena Corera 4 years ago

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The users of Scimago Journal & Country Rank have the possibility to dialogue through comments linked to a specific journal. The purpose is to have a forum in which general doubts about the processes of publication in the journal, experiences and other issues derived from the publication of papers are resolved. For topics on particular articles, maintain the dialogue through the usual channels with your editor.

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Source details

Journal of Environmental Chemical Engineering

Scopus coverage years: from 2013 to Present

Publisher: Elsevier

ISSN: 2213-3437

Subject area: Environmental Science: Pollution | Environmental Science: Waste Management and Disposal | Chemical Engineering: Process Chemistry and Technology | Chemical Engineering: Chemical Engineering (miscellaneous)

Source type: Journal

CiteScore 2020
7.5

SJR 2020
0.965

SNIP 2020
1.354

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CiteScore | CiteScore rank & trend | Scopus content coverage

Improved CiteScore methodology

CiteScore 2020 counts the citations received in 2017-2020 to articles, reviews, conference papers, book chapters and data papers published in 2017-2020, and divides this by the number of publications published in 2017-2020. Learn more >

CiteScore 2020

$$7.5 = \frac{23{,}550 \text{ Citations } 2017 - 2020}{3{,}123 \text{ Documents } 2017 - 2020}$$

Calculated on 05 May, 2021

CiteScoreTracker 2021

$$7.6 = \frac{35{,}753 \text{ Citations to date}}{4{,}694 \text{ Documents to date}}$$

Last updated on 06 March, 2022 • Updated monthly

CiteScore rank 2020

Category	Rank	Percentile
Environmental Science — Pollution	#17/132	87th
Environmental Science — Waste Management and Disposal	#17/108	84th
Chemical Engineering — Process Chemistry	#12/59	80th

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