

## Membrane Development and Its Hybrid Application for Oily Wastewater Treatment: A Review

Sri Martini\* & Erna Yuliwati

Chemical Engineering Department, Universitas Muhammadiyah Palembang, Jalan Jenderal Ahmad Yani 13 Ulu, Palembang, 30263, Indonesia.

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### ABSTRACT

The treatment of oily wastewater emitted from diverse industrial sources is significantly fundamental to keep environmental sustainability as well as human health. Membrane technology as one of the propitious methods has been used and developed in many ways for treating oil-containing water and wastewater. However, membrane fouling is an unavoidable occurrence regardless membrane materials including polymeric, ceramic, and metal based-membrane. In addition to this matter, hybridization system in membrane usage has attracted an increasing interest due to its benefit in decreasing fouling rate. In this paper, we begin with an overview of membrane application and its materials as well as its existence along with other existing treatment methods. Further, we specifically provide discussion into the application of hybrid membrane system in treating oily wastewater. According to the literature review which has been discussed here, conclusions then are properly drawn to highlight hybrid membrane systems that can amplify membrane resistance on fouling phenomenon.

*Keywords:* Membrane, fouling, hybrid system, oily wastewater, polymeric membrane

### 1.0 INTRODUCTION

It is undeniable that the development of industrial sector and an increasing number of global population in recent decades have been major contributors to the staggering amount of polluted-water and wastewater streams [1, 2]. There are various pollutant particles contained in industrial oily wastewater including oil, dyes, heavy metals, and other organic and inorganic compounds that have been resulted in a huge amount from various manufacturing processes [3, 4]. Several industries such as petroleum refinery companies, cooking oil manufactures, pharmaceuticals, and cosmetic industries generate and discharge massive wastewaters into the environment threatening human health

and ecosystem as they are toxic, carcinogenic, and low biodegradable in nature [5].

Carcinogenic and non-biodegradable characteristics of the wastewater refer to its difficulty to naturally biodegrade in open environment [6-13]. In order to mitigate the negative impacts of oily wastewater, the implementation of proper purification process known as wastewater treatment is a must. It will also prevent fatal environmental issues and allow water re-use [14]. In terms of oily wastewater, the existence of oil-water emulsion is the biggest challenge as it has stable nature making it difficult to be removed from wastewater [15]. It is classified as oil in water or complex emulsion. In this phase, water is the continuous phase

while oil is the disperse phase and vice versa while complex emulsion comprises tiny droplets of oil suspended in bigger droplets that are suspended in a continuous phase aforementioned [16].

To date, literature has showed several preferred methods used to treat oily wastewaters containing detrimental pollutant particles. They are advanced oxidation processes (AOPs), coagulation/flocculation, adsorption, biological techniques, and membrane technology [17]. AOPs have a particular ability to reduce the content of different organic pollutant eliminated through mineralization process [18, 19]. AOPs is characterized by hydroxyl radicals accountable for targeted organic pollutants removal. Oxidation process will transform those pollutants into water, carbon dioxide, and other harmless products [20-23]. However, costly chemical usage, sludge generation, and by-product formation have been long concerns to this option [24]. Biological method involves the existence of microorganisms to degrade pollutants concentration in the oily wastewater. Despite longer reaction time and wider treatment place needed that could be burdens of this method, it offers some advantageous points such as more environmentally friendly system and good removal efficiency [25]. Furthermore, adsorption method is also widely applied to treat oily wastewaters. In this technique, pollutant particles named sorbates are trapped on the surface or pores of adsorbents [26]. There are various types of adsorbents such as activated carbon, metal oxide, carbon-based materials, and metal organic framework that can be applied for trapping targeted pollutants contained in contaminated water and wastewaters [7, 11, 27, 28]. Adsorption is simple and reliable with less energy

consumption even though it is considered as a compliment of other main treatment process [29].

However, those techniques suffer from limited separation efficiency for wastewaters which particularly have high organic loading, complex operating operation in certain cases, and generating secondary pollutants with inefficient separation of emulsion droplet sizes less than 10 mm [17, 30]. Therefore, appropriate modification processes should also be applied on the adsorbent to increase its performance and overcome the shortfalls [4].

Recent literature then has shown that membrane technology has been preferred treatment mode to remove various pollutants as it has the ability to produce the highest purity level of treated wastewater. Compared to other treatment methods, membranes filtration has relatively higher removal efficiency related to prevalent contaminant in the oily wastewater [31, 32].

Membrane separation is not only a reliable way for oil removal, but also a dependable technique for heavy metals remediation which are mostly contained in industrial oily wastewater. There is also an increasing interest in the development of high pressure driven membranes such as nanofiltration (NF) and reverse osmosis (RO) for removing heavy metal [33].

Membrane is a semipermeable barrier managing the transport of particular substances between two adjunct phases. Therefore, membranes has an important function in many advanced separation processes, especially in industrial wastewater treatment plants. Practical operation, competitive flexible design, less chemicals and sludge production have been well known as membrane advantageous aspects [34]. However, its higher initial construction and

maintenance cost to keep membrane performance well also need particular concerns [35]. The implementation of membrane technology especially for oily wastewater treatment has had a challenging issue related to the higher chance of fouling rate phenomenon [36, 37].

Oil compounds including oil emulsion contained in wastewater take a large portion of the formation of blocking pores on membrane surface leading to flux decline over time [38]. It requires complex cleaning operation chemically or physically. As a consequence, both operational and capital expenditures would be increasing. Plain membranes including organic membranes have higher chances to experience membrane fouling as they have hydrophobic characteristics in nature related to its polymers that interact well with oil. Therefore, some further research tried to find some reliable solution to fix this problem by increasing in the hydrophilic behaviour of the polymeric membrane by adding some additive materials or applying coating [39, 40]. Regarding membrane fouling, certain acidic and alkaline based chemicals are also often used to recover membrane ability filtering fluid or wastewater stream. However, it is worthy to underscore that frequent chemical cleaning will negatively impact health and environmental from the secondary pollutants like halogenated by-products, and shorten membrane lifespan due to material degradation [39]. Therefore, the other safer and more effective option such as applying hybrid membrane system or pre-treatment stage can be an alternative. This article aim is to cover scattered discussion related to membrane fouling, membrane material development, and the application of hybridization system in membrane usage particularly for oilv wastewater treatment.

## 2.0 MEMBRANE FOULING

Membrane fouling taking place during filtration time reduces its function as advanced filter. To understand how the membrane experience fouling phenomenon, Hermia's models comprising complete blocking, standard blocking, intermediate blocking, and cake filtration models have been widely accepted and used [1]. For oily wastewater, the main concern of fouling occurrence is due to oil particles existence much beyond permissible standard level. From this point of view, Hermia's models could be applied to illustrate how oil particles, for example, could cause fouling. In cake formation model, fouling would take place when oil particles and other finite contaminants have bigger size than the average pore size of membrane forming accumulation building-up of the cake layer on the membrane surface. While intermediate blocking represents oil and other pollutant particles blocking membrane pores and creating intermediate fouling. In this model, they are assumed to have equivalent size between each other. Standard model relates to a decrease in pore diameter due to the non-uniformity of oil particles adsorption on the membrane pore. Eventually, complete blocking model confirms that oil particles with a bigger size than membrane pores would settle on the membrane surface leading to flux decline [38, 41].

As a separation process based on the presence of semi permeable layer, the principles of the membrane work are trapping the pollutant particles on membrane surface or pores within a specific size range and, at the same time, allowing particular substances to pass the membrane [42]. Its performance is mainly driven by pressure difference as an average value

of inlet and outlet pressures named trans-membrane pressure generated from applied pressure or vacuum. While operating condition called cross flow velocity contributes to the performance of the membrane when it running [43, 44], other parameters such as temperature, pH, oil and salt concentration, in most cases, also play important roles in the removal efficiency level of targeted pollutants in the wastewater [45, 46].

These operating parameters may have different effects on different membrane types and materials, as well as the characteristics of water and wastewater sample passing membrane layer. According to their pore size, membrane can be categorized into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Membrane can be fabricated and developed from various materials such as organic-based substances including certain polymer types, and inorganic-based materials including ceramic and metal [32, 34].

### 3.0 MEMBRANE MATERIAL

As explained in the previous section, membrane fouling mostly occurred due to complete blocking of pores through adsorption of continuous or disperse phases of the oil emulsion leading to the building-up of one or two phases of pollutant particles layer on the membrane surface supported by the phenomenon of concentration polarization [47, 48]. To manage this issue, several studies then have attempted some strategies including modifying feed characteristics and developing membrane materials by adding some additives substances [49, 50]. Modifying feed characteristics can be referred to the implementation of pre-treatment process to significantly decrease the concentration of oil

droplets before it entering membrane filtration system while the second approach is limited by the high energy demand [51]. Developing membrane materials can be correlated to the aim to increasing membranes resistance on oil emulsion flowing on membrane pores or surface [52]. Membrane material development aiming to reduce fouling rate have been applied through several approaches such as adding certain additives to increase membrane material resistance on oily feed and conducting chemical and physical surface coating [2].

It is worth to reemphasise that conducting pre-treatment or hybrid membrane system has been proved as an efficient way to increase membrane feed quality especially for oily wastewater leading to increasing permeate flux [1, 37, 39]. To date, combining the efficiency and effectiveness of both pre-treatment and main treatment using sole membrane filtration is still a challenge. Therefore, some other studies have tried to fabricate membrane enriched by additives substances including graphite, ZnO, certain metals or minerals for salt, heavy metals or dyes reduction from contaminated water, and they reported that those additives could increase membrane ability by lowering fouling flux to some extent [40, 53-55]. In general, there are three basic materials used to fabricate membrane including polymeric, ceramic, and metallic based membrane that are widely available for various types of oily wastewater treatment.

#### 3.1 Polymeric-based Membrane

Polymeric-based membranes have been popular due to their comparatively small footprints, varying range of operating conditions, cost-effectiveness, having superior organic solvent resistance, and ease of operating [2, 56]. There are a variety of

synthetic polymers such as polysulfone, polyvinylidene fluoride, polyamide, polycarbonate resins, and polyelectrolyte complexes that can be employed as membrane material [36]. Phase inversion is the most widely used method to produce membrane. In this method, preparing a homogeneous polymer solution by dissolving the polymer in a suitable solvent is an initial work followed by inducing stage using an external or internal factor. While external factor includes nonsolvent-induced precipitation and thermally-induced phase separation, main internal factor relates to evaporation-induced phase inversion [57].

### 3.2 Ceramic-based Membrane

Despite their favourable characteristics for many challenging applications demanding excellent thermal, chemical, and mechanical stability, ceramic-based membranes need costly basic materials such as  $\text{Al}_2\text{O}_3$ ,  $\text{ZrO}_2$ ,  $\text{SiO}_2$ , and  $\text{TiO}_2$  for their formation [58]. This could be an additional barrier for their en masse application in industries, especially for oily wastewater treatment. Therefore, researches have been started to observe the fabrication of low-cost ceramic membranes from natural mineral-based materials made from zeolite, fly-ash, kaolin, and others. It was believed that the valorisation of relevant natural mineral-based materials could produce low-cost ceramic membranes having excellent filtration performance [59].

### 3.3 Metallic-based Membrane

The increasing usage of metallic mesh-based membranes represents their potentiality to become an alternative in industrial breakthrough supported by ultra-low pressure difference involving hydrostatic pressure as well as some particular additives [2]. The

application of modified stainless-steel mesh membrane to purify engine oil contaminated water, for example, showed an excellent rejection rate for oil particles [60]. This metallic membrane observed was fabricated by depositing a rough superhydrophilic  $\text{TiO}_2$  nanofibers layer onto the surface via spray deposition technique. Then it was followed by dispersed solution of  $\text{TiO}_2$  nanofibers in ethanol while the coated mesh was sintered to achieve adequate integration of the nanofibers onto the mesh surface. The performance of membrane can be reliable regardless their materials and modification ways as stated in Table 1.

**Table 1** Membrane materials and their modification modes for treating oily wastewaters

Membrane material composition/ modification mode	Rejection (%)	Ref.
Polymer: PVDF-PDA- $\text{SiO}_2$ / Phase inversion	94	[61]
Polymer: PVDF-NiCo-LDH/ Polymerization-hydrothermal	99.7	[62]
Polymer: PVC-Bentonit/ mixed matric	97	[63]
Polymer: PVDF-PDA/ polymerization	99.82	[64]
Ceramic: Kaolin	98.52	[65]
Ceramic: Quartz	99.98	[66]
Ceramic: Fly ash	97	[67]
Ceramic: $\text{Al}_2\text{O}_3$	99.1	[68]
Metal: Stainless stell- $\text{TiO}_2$	99	[69]
Metal: Copper-Ag	99.5	[70]
Metal: Stainless steel- $\text{SiO}_2$ -CS	99	[71]

## 4.0 HYBRID MEMBRANE APPLICATION FOR OILY WASTEWATER TREATMENT

It has been proved that the application of hybrid methods for treating raw

industrial wastewaters including oily wastewaters would lead to a better output. Other than that, it could reduce the rate of certain machine breakage, therefore, hybridization system for treating wastewaters is a reliable option to get the highest quality of treated oily wastewater [72]. In this technique, two or more treatment methods are applied in sequence. This pattern enables particular disadvantageous features of individual method to be covered by the advantageous aspects of the other hybridized method and vice versa. In general, there are two main sections of a hybrid treatment system named pre-treatment process and main treatment stage. It is also known as primary and secondary treatment processes [73]. Literature shows that membrane material is a prominent example of a method combining with pre-treatment option. Specifically, it can be described that by reducing oil and grease contents, dissolved, and suspended solid contained in the oily wastewater through pre-treatment step, membrane performance would be much better [36]. In the case of treating raw wastewater that has high organic and inorganic loading, membrane can be easily clogged leading to fast permeate flux decline. Therefore, the implementation of hybrid system for membrane system on oily wastewater treatment using various compatible methods such as coagulation/flocculation, adsorption or advanced oxidation could benefit membrane itself by reducing fouling rate and chemical cost, and lengthening membrane lifespan [39, 74-79].

As one of pre-treatment options, AOPs that cover several types of oxidation reaction can be effectively hybridized with membrane. This technique could be conducted by the application of ozone, Fenton, photo-

Fenton and certain semiconductor agents such as  $\text{TiO}_2$  and  $\text{ZnO}$  [19]. This process has shown its capability as hybrid partner for membrane including for treating oily wastewater derived from varying sources. A study constructed a hybrid system comprising ozonation and membrane bioreactor to treat water process-affected of toxic oil sands, and it reported a promising output [80]. Mild ozonation followed by a modified membrane bioreactor indicated its suitability for the treatment of contaminated water process. By having a proper dose of ozone substance, toxic materials could be eliminated to a satisfactory level. In addition, they also confirmed that ozone pre-treatment improved denitrification and removal of toxicity in membrane bioreactor system. The most important effect of the use of ozonation as pre-treatment stage on membrane performance relates to delayed fouling occurrence due to the alteration of microbial community structure. Other study also proposed that a hybrid UV/ $\text{TiO}_2$  and ultrafiltration membrane was effective in treating oily wastewater derived from edible oil manufacturer [39]. It was reported that, compared to sole usage of ultrafiltration membrane to filter oily sample, hybrid  $\text{TiO}_2$ /UV-ultrafiltration membrane system significantly increased the permeation flux value by reaching oil removal efficiencies up to 86%. This hybrid system was further explored under varying values of transmembrane pressures (TMP) and cross flow velocities (CFV). They concluded that selected operating condition has affected on membrane performance to some extent. The application of hybrid Fenton/flocculation and ultrafiltration membrane for petroleum refinery wastewater treatment was also conducted [1]. This study reported there was an increasing permeate flux

up to 1.5-fold. Other than that, compared to sole UF membrane filtration, this study revealed that the permeate flux decline of hybrid membrane system experienced lower percentage from 51% to 28% due to pre-treatment process. In general, they all showed that membrane hybridization involving AOPs is a potential and reliable approach for recalcitrant organic matter removal in oily wastewater treatment.

The other work dealt with the implementation of hybrid electrocoagulation and ceramic-based microfiltration membrane in batch mode for treating oily wastewater containing oil, grease along with particular metals like Na, Cr, Cu, Pb, and Ni [81]. They conducted electrocoagulation by considering influential working parameters such as current density, electrode distance, and initial pH. This work reported decreasing oil and grease and total dissolved solid concentration from 35 to 10.2 mg/L, and from 3230 to 2780 mg/L, respectively, in just 20 min. Ceramic membrane then removed the pollutant flocks generated during the electro-coagulation process. This pre-treatment has eased membrane work by supplying flowing wastewater with better quality.

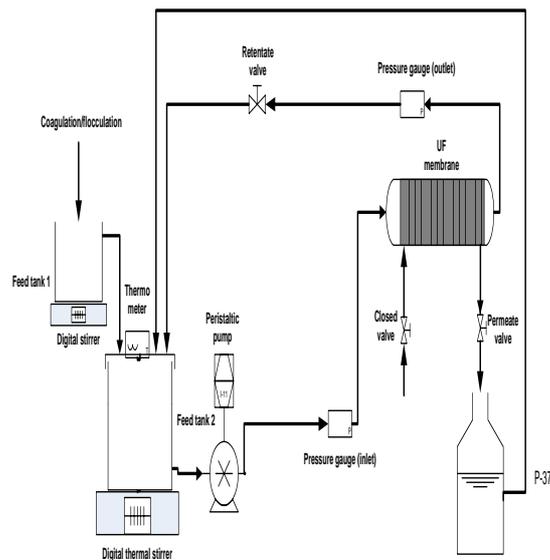
When it comes to using dual membrane system, initial construction and maintenance cost often be additional barriers. However, it is worthy to investigate its usage in certain condition needing an excellent output. The application of hybrid tubular polymeric based ultrafiltration membrane and capillary membrane distillation for the treatment of bilge water containing oil could be an example [77]. Their study substantially reduced the content of oil to remarkably less than 5 ppm. To conclude, this utilisation of dual membranes showed complete removal

achievement of oil pollutants as well as the remaining soluble substances in the feed.

An increasing membrane performance relates to a higher permeation flux. For this reason, a study investigated and found that the high amount of impurities in the raw petroleum oil refinery wastewater used as membrane feed was the major cause of low permeation flux [82]. They then emphasized the implementation of hybrid nano-porous membrane-powdered activated carbon (NPM-PAC) to overcome this issue. The experimental work indicated NPM alone was less effective in terms of steady permeation flux by reaching 78.7 L/(m<sup>2</sup>h). Reversely, hybrid NPM-PAC with optimum PAC dosage could enhance permeation flux value almost double or up to 133.8 L/(m<sup>2</sup> h), and also decreased steady fouling resistance around 46.1%. These phenomena dealt with higher porosity on membrane surface due to PAC addition [40]. Figure 1 represents the example of hybrid coagulation/flocculation and membrane design that could be used to increase membrane performance and lengthen its lifespan.

## 5.0 CHALLENGES AND FUTURE PERSPECTIVE

The main concern on this review is the usage of membrane technology especially in hybrid system for treating oily wastewater along with membrane materials and membrane fouling mechanism during purifying its oily feed. Several aspects then can be further discussed as they can be considered as challenges that need reliable solution and explanation such as filtration test method and various nature of applied oily wastewaters.



**Figure 1** An example figure of the application of hybrid membrane system

In terms of filtration test methods, two important factors including cross-flow and gravity driven should be further involved. They need to be more universal method to enable universal comparison among membrane materials and membrane type regarding operating condition and feed loading level. Another issue that is both a challenge and a prospective part of membrane is how to fabricate economical membrane technology, started from material selection to its regeneration or recovery aspects. By solving this part, the image of membrane system as an expensive option for treating wastewater can be minimized without compromising its output quality.

## 6.0 CONCLUSION

In the last decades, membrane technology has been used in numerous applications modes in accordance with wastewater treatment including oil-compounds containing wastewater.

The present review article explained the development of membrane usage along with its major problems and material sources. Other than that, this work examined varying hybrid membrane methods as an effort to treat oily wastewater and, at the same time, increase membrane performance. From the literature study, it can be observed that the presence of oil and grease and other organic inorganic particulate matter led to severe membrane fouling rate. Therefore, implementing hybrid system could help with reducing pollutant concentrate in the oily wastewater before it enters membrane system. Overall, regardless fouling occurrence which can be managed by applying hybrid technique, membrane separation is still considered as one of the most reliable oily wastewater treatment modes.

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