## Insights into Membrane Distillation Application for Textile Wastewater Treatment – A Review

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#### ABSTRACT

Textile wastewater must be effectively treated with the best available technology prior to release to receiving water bodies to prevent its impact on the environment. Apparently, membrane distillation shows great potential in treating textile wastewater a part of the complexity of the textile wastewater composition. This membrane process enables the water vapour to pass through its porous hydrophobic membrane and retains the concentrated pollutants to be transported. This paper provides data and information from previous studies using membrane distillation to treat textile wastewater. An overview of the development of membrane distillation as well as the fundamental theory is presented. Recent progress in the application of membrane distillation in textile wastewater is then discussed. The final part of the paper looked at the future orientation of this technology to be acceptable in the industrial sector, especially for the textile industry.

Keywords: Membrane distillation, textile wastewater, colour removal, microporous membrane

## **1.0 INTRODUCTION**

The textile industries play an important role in global economic development bv selling things that quickly depreciate in value because of perishable or fast fashion cycles. Although the textile industry is one of the major contributors to the local economy, it indirectly led to negative environmental impact from its input and output activities in the textile processing. In terms of inputs, the textile industry is considered a waterdemanding industry as it consumes a large volume of fresh water during the manufacturing process [1-3]. Statistics show that the textile industry uses  $0.06-0.40 \text{ m}^3$  of water for each kg of textile manufactured that is generally

used as a medium to apply dyes and finishers and remove impurities [4-6]. It should be pointed out that the water used to treat textiles often requires heating, which has led to an increase in energy consumption. Whereas on the output side, the textile manufacturing process generates an enormous amount of wastewater with a wide range of contaminant concentrations.

Textile wastewater generally consists primarily of process water, cleaning water, non-contact cooling water and storm water [6]. The effluents can be classified into four categories, according to the United States Environmental protection Agency (USEPA); hard to treat, hazardous or toxic, high volume and dispersible wastes [6]. The main sources of hard-to-treat wastes are nonbiodegradable organic and inorganic compounds. Colour, metals, phenols, certain surfactants. toxic organic compounds, pesticides, and phosphates are among the wastes. Meanwhile, metals. chlorinated solvents, nonbiodegradable, or volatile organic compounds are referred to as hazardous or toxic wastes in textile processing, which partly comes from non-process applications such as machine cleaning.

To reduce its environmental impacts, stringent environmental regulations are enforced in each country to prevent severe water pollution caused by the textile industry. Recently, membrane has distillation (MD) shown а significant contribution in removing dyes from the textile effluent or dye solution due to its flexibility to operate under low hydraulic pressure and moderate temperature [7-12]. Since the textile industry discharges effluent at relatively high temperatures (80–90°C), MD can exploit the free energy of hot effluent during the treatment process [9, 12]. Additionally, MD may use lowquality waste as well as alternative energy sources such as solar and geothermal energy to maintain feed temperature [13]. The best feature of thermally-driven this separation process is its ability to retain nonvolatile organic compounds from entering the membrane pores which cannot easilv achieved bv nanofiltration (NF) and reverse osmosis (RO) processes [14].

The main objective of this review is to provide data and information from recent research on the treatment of wastewater by membrane textile distillation. Firstly, an overview on the development of membrane distillation and the basic theory are given. Recent progress in the application of membrane distillation in textile sewage is then presented. The final part of the paper addresses the future direction of this technology to make it acceptable in the industrial sector, particularly for the textile industry.

## 2.0 BACKGROUND OF MD PROCESS

## 2.1 Chronicles of MD Development

MD has been introduced since 1963 by Bodell and the start-up phase was reported between 1960 and 1970 [15]. Extensive literature was available at the time to promote MD as a new membrane separation technology that evaporation combines and condensation processes in one system. The initial focus of the research was solely on the application of water desalination. Compared to RO, MD is hardly fouled during the separation process due to its relatively large membrane pores and unique separation mechanism that allows only vapour to pass through the membrane pores. However, the breakthrough of first high-flux asymmetric RO membranes (made via non-solvent induced phase separation (NIPS) from cellulose acetate solution) by Loeb-Sourrirajan in 1960s shifted the attention of researchers toward RO development.

As reported by Susanto [16], the MD growth study is not positive after 1970 because no literature was published during this period. Although MD is not popular during its initiation phase, it starts to gain its attention back specialized after one membrane workshop entitled "Workshop on Membrane Distillation" held at Rome, Italy in 1986 [17] The goal of the workshop is to standardize the terminology used to describe MD processes. MD development was further supported by international manufacturers such as Gore and Associates (USA), Enka AG

(Germany) and the Swedish Development Co., aiming to produce membranes for commercial application [16, 18]. During the "Growth Phase", the major problem encountered in the MD development is its process that is not commercially accepted by industrial players owing to its energy inconsistency, particularly in heat loss through conduction. The development chronology for MD is presented in Figure 1.



Figure 1 Timeline of MD development

# **2.2 Mechanism and Configuration of MD Process**

The term MD is basically reflecting to the common distillation process that operates based on the vapour-liquid equilibrium principle which requires latent heat of evaporation to produce the final distilled water [15, 19]. Due to the similarity, MD is induced by the vapour pressure gradient caused by the temperature difference through the membrane [20]. In MD, the membrane only acts as a barrier to hold the liquidvapour interface at the entrance of the membrane pores and does not involve phenomenon. а transport Unlike NF/RO membranes, MD is not selective or has "water-like" а attraction to the feed solution [18].

As illustrated in Figure 2, MD separates two aqueous solutions at different temperatures using a microporous non-wetted hydrophobic membrane with a vapour-liquid interface at the pore entrance [21]. Due surface tensions. the the to hydrophobic nature of the membrane prevents aqueous solutions from penetrating the pores [22], unless transmembrane pressure of MD process exceeds liquid entry pressure (LEP) of the membrane. The MD process involves simultaneous mass and heat transfer, and both determine the membrane's water productivity.

There are four main configurations that are usually used during the MD process that are direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD). This is further described in Table 1.

Table 1   Membrane	configuration
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Membrane configuration	Process	Advantages	Disadvantages
DCMD Feed in Liquid permeate out Liquid Feed out permeate in	<ul> <li>Throughout the process, both the feed and permeate solution are in direct contact with the membrane surface [20].</li> <li>The hydrophobic membrane functions as a physical barrier that preventing the feed solution from entering membranes pores.</li> <li>Vapour molecules pass through the membrane pores from the high-temperature side to the low-temperature side due to the temperature difference between the feed and permeate sides [30].</li> </ul>	<ul> <li>High flux [31].</li> <li>Simple design</li> <li>Low operating temperature and low hydrostatic pressure [30].</li> </ul>	• High heat loss [32,33].
AGMD Feed in Coolant out Membrane Feed out Feed out Product	<ul> <li>A permeate gap filled with air separates the membrane from the cold walling [34].</li> <li>Vapour that passed through the membrane must overcome the air gap in order for condensation process to occur [20].</li> </ul>	<ul> <li>Low operating temperature and pressure.</li> <li>High energy efficiency [31].</li> </ul>	• Low permeate flux [20].
SGMD Feed in membrane Feed out Feed out Sweep gas in	<ul> <li>Air stream is used to collect vapour [33].</li> <li>The feed solution is heated and then transfer to the membrane surface.</li> <li>The volatile substances pass through the membrane pores to the permeate side, where carrier gas collects and transports the vapour [34].</li> </ul>	<ul> <li>Less heat loss</li> <li>Lower thermal polarization</li> <li>Excellent wetting resistance [32].</li> </ul>	• Need external condenser [34].
VMD Feed in Condenser membrane Permeate Feed out Vacuum pump	<ul> <li>Vacuum is used instead of condensing medium [31].</li> <li>Porous hydrophobic membrane is placed into direct contact with the feed solution.</li> <li>Condensation process takes place outside the membrane module.</li> </ul>	• Negligible heat loss [31].	<ul> <li>Vacuum pump used required high electric consumption.</li> <li>More susceptible to fouling and pores wetting [31].</li> </ul>



Figure 2 Schematic diagram of the MD process [23]

# **2.3 Material and Characteristics of MD Membrane**

fabricated MD can be using hydrophobic polymers or by altering the surface of hydrophilic membranes. Chemical resistance polymers such as polytetrafluoroethylene (PTFE). polypropylene (PP) and polyvinylidene fluoride (PVDF) can be used to manufacture hydrophobic membranes. Some studies have shown that other polymers, such as polycarbonates (PCs), polyethylene (PE) and polyesters, are also suitable for the production of MD membrane. Depending on the properties of the membrane materials, different methods such as sintering, stretching, NIPS or thermal induced phase separation (TIPS) can be used to make porous hydrophobic membranes. However, literature reports that NIPS and TIPS are the two common methods of preparing microporous membranes for the MD process.

It should be noted that the membrane just serves as a barrier to hold the liquid/vapour interfaces at the entrance of its pores in the MD process, therefore there is no need to be selective as required in other membrane processes like NF, RO, and pervaporation. During the MD process. the membrane must not be wetted and only vapour and non-condensable gases are present within its pores. The membranes used in MD usually have pore sizes of 10 nm to 1 µm. The

membrane material be must hydrophobic with a high water contact angle and small maximum pore size to avoid pore wetting. Although a great number of membranes have been used in MD, there is no universal membrane property that can fit all the applications. Due to unavailability of commercial membranes for the MD process in the current market, some studies used microfiltration (MF) and ultrafiltration (UF) membranes for this purpose. At present, the major challenge in the MD process is the lack of readily available commercial membranes with high porosity and low membrane thickness. Other membrane characteristics also have an essential role in determining MD performance include the LEP, mechanical strength, thermal stability, and chemical resistance.

One that must know the performances of the MD with respect to permeate flux and solute rejection mostly determined by are the membrane structural properties [21]. In general, thicker membrane tends to have lower heat loss by conduction through the membrane matrix while thinner membrane may offer high flux due to relatively low mass transfer resistance. On the other side, membrane with larger pore size is preferable to enhance permeate flux compared to smaller membrane pore size. In most cases, higher LEP can be achieved with the use of membrane with superior hydrophobicity (low surface energy material) and/or by reducing the membrane surface pore size [15]. Typically, higher LEP value is required to prevent liquid penetration during the MD process even though the process is operated at a very low operating pressure. This is because of the possibility of imbalance pressure between feed and permeate sides during real operation. Compared to reducing the membrane pore size, many researchers prefer to work on

membrane hydrophobicity improvement with the aims of reducing membrane wetting [24].

Another physical characteristic that should be taken into consideration in selecting MD is membrane mechanical strength. Optimum membrane thickness and pore size are required to maintain excellent flux without deteriorating mechanical strength. Laganà et al. [25] reported that the optimum range for MD thickness is within the range of 30-60  $\mu$ m whereas Wu et al. [26] suggested the pore size should be in the range of 0.01-1  $\mu$ m. Khayet [18] on the other hand reported that the mean pore size must be controlled at below than 0.5 µm to prevent membrane pore wetting. Bonyadi and Chung [27] stated that a sharp pore size distribution of 0.3-0.5 µm should be the main target for membranes used in MD process. With respect to membrane porosity, Adnan et al. (2012) recommended that high membrane porosity (>70%) is improve the necessary to MD performance. To be recognized as thermally driven membrane process, MD should exhibit good thermal stability for the long-term stability as the feed temperature may reach as high 90-95°C. The membrane also as requires high chemical resistance, especially when dealing with harsh environments.

According to the literature, PP membranes are extensively employed in MD application for treatment of dyeing solution [8-11]. This type of polymer has greater hydrophobicity, thermal and chemical resistance, all of which are important for the treatment of hot textile wastewater through the MD process [29]. However, the production of PP membrane is difficult because it can only be fabricated using the molten extrusion technique followed by stretching or high the temperature thermal phase separation

process [15]. Due to this, PVDF received more attention as an alternative to PP in the MD process. This is due to the fact that PVDF can be dissolved in common solvents.

# 3.0 MD FOR COLOUR REMOVAL PROCESS

Table 2 summarizes MD application in textile wastewater the treatment process. The MD setup can be found in different configurations that differ in the way the permeate solution is processed on the cold side as discussed in the previous section. The most popular configurations are DCMD and VMD. Earlier studies mainly used commercial PP membranes made by Enka Microdyn, USA and Membrana GmbH, Germany [8-10,]. Different types of membrane modules were used for this application. The capillary membrane module was preferred in the earliest study as this module has a higher packing density as compared to the plate and frame module [35]. Meanwhile, the hollow fiber membrane module was chosen because it could effectively reduce the thermal polarization during the MD process. The following is an updated review on the development of MD fabrication, in particular to the fabrication of new type of membrane material for colour removal process.

In 2014, Mokhtar *et al.* [36] tested the fabricated PVDF hollow fiber membrane in reactive black 5 (RB5) solution. PVDF was chosen due to its outstanding properties such as low surface energy, low melting point, high tolerance to oxidizing agents, and good thermal stability [37-39] (Kuo *et al.*, 2008; Kim *et al.*, 2010; Edwie *et al.*, 2012). In this study, the concentration of PVDF ranged between 12% to 18% by weight. Results showed that PVDF membrane with 12 wt% of polymer concentrations exhibited the best performance as these membranes eliminated almost all dye molecules (99.78%) when tested with 0.05 g/L reactive black 5 (RB5). However, the membrane permeate flux is relatively low (max. 6 kg/m<sup>2</sup>.h) and requires more research.

Mokhtar et al. [40] further studied the effects of different types of additives (primarily as a pore former agent) for the MD treatment of dyeing wastewater. It is well known that simple blending method is the most commonly used technique by researchers to improve membrane performances. In this study, PVDF membranes were prepared with two different types of additives, ethylene glycol (EG) and polyvinylpyrrolidone (PVP). It was discovered that the membrane blended with additives (both EG and PVP) had a higher overall porosity and mean pore size than that of the PVDF membrane. However, results showed that the membrane incorporated with PVP could lead lower membrane to hydrophobicity and decrease in thermal stability and mechanical strength of the membrane.

Furthermore, when tested with 0.5 g/L RB5 solution during the MD process, the PVDF-EG membrane showed consistent separation efficiency with an average dye rejection of 99.88%. Meanwhile, the rejection of the PVDF-PVP membrane decreased slightly from 99.91% to 99.16% at the end of the test. The slight decrease in dve rejection obtained by PVDF-PVP membrane could be due to the fouling and pore wetting.

Figure 3 depicts a comparison of the inner and outer layers of PVDF-PVP and PVDF-EG membranes after the treatment process. Clearly, the inner and outer layers of the PVDF-PVP membrane were heavily stained with dye components, whereas the whitish colour of the PVDF-EG membrane changed only marginally. Direct observation of the physical colour of membranes confirmed the dye molecule penetration from the lumen side of the PVDF-PVP membrane to the outer layer. The PVDF-EG membrane's promising results are primarily due to its higher wetting pressure and degree of hydrophobicity.



**Figure 3** Stereoscopic images of (1) PVDF-EG and (2) PVDF-PVP hollow fiber membrane after MD tests, (a) inner surface and (b) outer surface [40]

A part of blending the polymer with additives to induce the porous structure of the fabricated membrane, Mokhtar *et al.* [41] was made an attempt to develop composite membrane made of PVDF and Cloisite 15A clay for further evaluation using both synthetic and real industrial wastewater. Results indicated that the membrane was able to give excellent rejection (>97%) for any type of dyes irrespective of its molecular weight.

Membrane specification	Types of feed solutions	Permeate flux	Rejection (%)	Remarks	Reference
Commercial polypropylene (PP)	Dyes: Blue LEVAFIX	Flux increased with	Dyes: 100%	This study was the first attempt to	[43]
hollow fiber membrane	E-G, Blue LEVAFIX	increasing bulk		examine the potential of MD	
Effective membrane area: 0.90 m <sup>2</sup>	E-BA and Red	temperature difference.		process for textile wastewater	
Pore size: 0.45 µm	LEVAFIX E-4BA	Greater flux decay with		treatment process. Pure water flux	
Porosity: 70%	(MW: 860-1200 g/mol)	increasing the feed		could be recovered and reused for	
Membrane thickness: 100 µm		concentration.		other purposes.	
Commercial PP capillary membrane	Dye: Methylene Blue	Flux increased with	Dyes: 100%	This work showed the membrane	[8]
from Enka Microdyn, USA	(MW: 319.85 g/mol)	increasing feed	Salts: 100%	ability to reject salt at higher	
Effective membrane area: 0.036 m <sup>2</sup>	Salt: NaCl	temperature and feed		concentrations up to 1.0 M NaCl.	
Pore size: 0.20 µm		flow rate. No obvious		The experimental results were in	
Membrane thickness: 1.5 mm		changes in the permeate		agreement with the modeling data.	
		fluxes while increasing			
		the salt concentration in			
		the dye solution.			
Commercial PP capillary membrane	Dyes: Remazol Brillant	Flux increased	Dyes: >90%	Membrane fouling was observed	[9]
from Membrana GmbH, Germany	Blue R, Reactive Black	proportionally to feed		during MD process and membrane	
Effective membrane area: 0.0028 m <sup>2</sup>	5, Indigo, Acid Red 4	flow rate and feed		wetting was found as the primary	
Pore size: 0.20 µm	and Methylene Blue	temperature but		reason for the sudden flux	
Porosity: 75%	(MW: 262.26-991.82	inversely proportional to		enhancement after treating dyeing	
Membrane thickness: 0.51 mm	g/mol)	the feed concentration.		solutions.	
		The flux depended on			
		the MW of the dye used			
		in the feed solution.	<b>a a a a a a a a a a</b>		
Commercial PTFE flat sheet	Real textile wastewater	Flux increased by 71%	Salts: 100%	The agarose layer prevented the	[12]
membrane from EF-Materials	collected from the	with the attachment of		membrane fouling and pore	
Industries Inc., Taiwan modified with	dyeing process.	agarose hydrogel layer		wetting problems and improved the	
agarose nydrogel layer		on the membrane surface		permeate flux without affecting the	
Effective membrane area: $0.04 \text{ m}^2$		compared to bare		separation efficiency.	
Pore size: $0.20 \mu\text{m}$		membrane.			
Porosity: 85%					
Membrane thickness: 130 µm					

 Table 2 Textile wastewater treatment using MD process

Membrane specification	Types of feed solutions	Permeate flux	Rejection (%)	Remarks	Reference
In-house made PVDF hollow fiber membrane Effective membrane area: 0.0109 m <sup>2</sup> Pore size: 0.09-0.14 μm Porosity:70-78% Membrane thickness: 130-170 μm	Dye: RB5 (MW:991.82 g/mol)	Flux increased when the polymer content used for membrane making was at low concentration.	Dyes: 99.78%	Due to its membrane characteristics, 12 wt% PVDF membrane attained the higher permeate flux when compared to other membranes fabricated of higher PVDF concentrations.	[36]
In-house made PVDF hollow fiber membrane (blended with different clay amount) Effective membrane area: 0.0137 m <sup>2</sup> Pore size: 0.09-0.17 µm Porosity:82-83% Membrane thickness: 127-144 µm	Dye: RB5 (MW:991.82 g/mol)	PVDF membrane incorporated with the lowest clay concentration exhibited consistent permeate flux throughout the study.	Dyes: 99.98%	Clay incorporation could improve membrane properties but excessive use of it could negatively affect membrane structure and lead to poor MD performance.	[44]
In-house made PVDF-C15A hollow fiber membrane Effective membrane area: 0.01 m <sup>2</sup> Pore size: 0.88 μm Porosity:83% Membrane thickness: 127 μm	Dyes: Crystal Violet, Acid Red 1, Reactive Orange 16, Congo Red and Reactive Black 5 (MW: 407.98 -991.82 g/mol) Salt: NaCl	Flux reduced as a result of increased dye and salt concentrations.	Dyes and salt: > 97%	The fabricated membrane demonstrated excellent results in removing nearly all dye components (except CV dye) with consistent permeate flux recorded regardless of dye MW.	[40]
In-house made PVDF-C15A hollow fiber membrane Effective membrane area: 0.01 m <sup>2</sup> Pore size: 0.88 μm Porosity:83% Membrane thickness: 127 μm	Real textile wastewater collected from the equalization tank.	Almost 50% flux declined was observed for a long- term study.	COD: 72-80%; Colour: 72- 89%	The fabricated membrane showed inconsistent flux when it was directly used for industrial wastewater. This is primarily due to its low tolerance towards certain chemical components in the feed.	[41]
Commercial PVDF and PTFE flat sheet membrane purchased from Millipore, USA and Pall Gelman, USA, respectively. Effective membrane area: 0.00098 m <sup>2</sup> Pore size: 0.20-0.45 µm Porosity: 69-84%	Methylene Blue, Crystal Violet, Acid Red 18 and Acid Yellow 36 (MW: 300 - 600 g/mol)	PTFE membrane recorded higher flux than PVDF membrane in which its initial flux was around 34 L/m <sup>2</sup> .h while PVDF flux 21-23 L/m <sup>2</sup> .h.	Colour: 100% (PTFE) and 65.6-93.2% (PVDF)	PTFE membrane exhibited promising performance due to its higher porosity and greater hydrophobicity compared to PVDF membrane.	[45]

Membrane specification	Types of feed solutions	Permeate flux	Rejection (%)	Remarks	Reference
In-house made PDMS/PVDF hybrid electrospun membrane Effective membrane area: 0.00098 m <sup>2</sup> Pore size: 0.45-0.49 µm Porosity:72-87% Membrane thickness: 98-105 µm	Methylene Blue, Crystal Violet, Acid Red 18 and Acid Yellow 36 (MW: 300 - 600 g/mol)	Flux enhancement was recorded for the PDMS/PVDF electrospun membrane compared to control PVDF electrospun membrane.	Colour: 100%	The superhydrophobic membrane can achieved high flux and dye rejection and withstand from foulin and pore wetting problem.	[42] g
Commercial PTFE and PVDF hydrophobic membrane Pore size: 0.22 µm and 0.22 µm Porosity: 85.1% and 70.4% Membrane thickness: 180 µm and 100 µm	Industrial dyeing wastewater form dyeing and printing company	PTFE membrane obtained higher flux and rejection performance than PVDF membrane.	COD: > 89% Colour: > 94%	The suspended solids accumulate o the surface of the membrane resulting in membrane wetting and fouling.	n [46]
Commercial PVDF flat-sheet hydrophobic membrane by Milipore Pore size: 0.22 µm Porosity: 75% Membrane thickness: 125 µm	Maxilon Blue 5G, Drimarena Yellow K- 2R and Sodium Fluorescein	The increased in feed temperature increased the flux. Flux gradually rose as the stirring rate increased. Flux decline with time during long term study.	Dyes: > 99%	Dye adsorption on the membrane surface and membrane pores lead to partial pore blockage which induce permeate flux decline. By simple flushing, the cake formed were partially removed.	[47] o d
Commercial PTFE (PTFE23001, Sterlitech) and PA (NF990, DOW FILMTECTM) flat sheet membrane Porosity: 76.7% and 17.1% Pore size: 0.2 µm and 0.68 µm	Reactive black and disperse black	PTFE membrane obtained higher flux than PA membrane for both RB and DB solution.	Dyes: > 97.5%	Both VMD and TPV recover water from dye solution with different separation process. VMD is based on volatility difference while TPV based on difference in membrane affinity.	[48]
Dual-layer, nanofibrous styrene- acrylonitrile membrane Porosity: 81% Commercial PTFE membrane Pore size: 0.22 µm Membrane thickness: 180 µ	Dyeing wastewater form textile and fabric factory	Fabricated membrane obtained higher flux and rejection rate compared to PTFE membrane.	COD: 98% Colour: 99.9% TDS: 99.5%	The formation of cake layer on the membrane surface resulted in decrease in membrane flux during long term study.	e [7]

Membrane specification	Types of feed solutions	Permeate flux	Rejection (%)	Remarks	Reference
Dual-layer nanofibrous SAN <sub>4</sub> -HIPS	Disperse dye Red-60,	Fabricated membrane	COD: 99.28%	As the membrane remained	[22]
membrane	Reactive dye Orange-	obtained higher flux than	BOD: 97.93%	hydrophobic after test, flux	
Commercial PTFE membrane	122 and industrial	commercial membrane.	Colour: 100%	decreased due to membrane	
Pore size 0.22 µm	textile wastewater from	Flux declines up to 42.58%		fouling rather than partial pore	
	textile dyeing factory	after 48h.		wetting.	
Commercial PTFE flat sheet	Reactive black CI	Permeate flux for RB dye	Colour:	> Surface contact angle were reduce	ed [30]
membrane	20,505 (Tiafix RBL	solution decreased while	87.84%	over time for both dye solution ar	d
Porosity: 76.788%	133%) and disperse	DB showed stable		membrane thickness increased du	e
Membrane thickness: 108.3 µm	black (mixture)(EX-SF	permeate flux.		to fouling.	
	300%)				
Commercial PTFE flat sheet	Dye: Reactive Black	Flux increases with	Dyes: > 98%	DCMD performance is highly	[3]
membrane from Sterlitech	and Disperse Black	increasing feed		dependent on the dye	
Corporation	(MW: 991.82 g/mol)	temperatures.		characteristics as well as the	
Pore size: 0.20 µm				process operational conditions	
Porosity: 84.52%					
Membrane thickness: 108 µm					
Polyetherimide (PEI) membranes and	Methylene blue	PEI and PEI-PDMS both	-	PEI-PDMS membranes shows	[49]
PEI modified using	solution (MW: 319.86	obtained high permeation		good antifouling property with	
polydimethylsiloxane	g/mol)	flux		89.4% flux racovery.	
Effective membrane area: 220 cm <sup>2</sup>					
Pore size: 79 nm and 72 nm					
Commercial PTFE membrane	Textile wastaweter	Fractionation process	-	Membrane wetting occur at highe	r [50]
(Changqi, China)	from textile company	achieved 98% water		temperature	
Pore size: 0.5 µm		recovery while only 38%			
Porosity: 90%		for ozonation process.			
Membrane area: $123.5 \text{ cm}^2$					

Using real textile effluent, it was found that the membrane could not withstand for a long period of study (40 h) and the permeate quality and quantity reduced rapidly with time (as shown in Figure 4). The separation process started with the initial flux at around  $38 \text{ kg/m}^2$ .h and tended to decline during the first 10 h of operation until it reached the critical flux (around 15 kg/m<sup>2</sup>.h). Partial pore wetting and thermal polarization were the main reasons for this sudden performance drop. Nevertheless. the fabricated membrane recorded at least 89% rejection for all the water quality parameters measured (i.e., colour, conductivity, turbidity, TDS, COD and BOD5) during short-term study and achieved minimum 72% COD and colour rejections for the long-term study.



**Figure 4** Permeate flux of the PVDF-Cloisite 15A membrane versus time [41]

Current development in membrane fabrication is not limited to blending method only, studied by An et al. [42] proven that electrospinning can be a technique reliable to construct controllable interfacial surface with inorganic nanomaterials and polymer Thev employed matrix. the electrospinning technique to produce a superhydrophobic

polydimethylsiloxane/polyvinylidene fluoride (PDMS/PVDF) membrane. The membrane fabrication process consisted of two steps, i) electrospinning for making a basic (vinylidene fluoride-copoly hexafluoropropene) **PVDF-HFP** electrospun membrane and ii) electrospraying for hybridizing PDMS microspheres onto the PVDF-HFP electrospun membrane.

This new technique had attributed to the formation of monodispersed miscrospheres on the surface of the PVDF-HFP electrospun membrane, which claimed as the main reason for the enhancement in membrane hydrophobicity and surface roughness. The presence of bumps and deep vallevs on the membrane surface enable the water and dye droplet (testing during contact angle measurement (as shown in Figure 5) to be suspended on the membrane surface without allowing water to enter the membrane pores. The mechanism basically obeys the famous theory of Wenzel and Cassie-Baxter effect. Interestingly, the newly developed membrane can reach a contact angle 155°. The value up to superhydrophobic characteristic of that membrane ensures resistance its towards membrane pore wetting.



**Figure 5** Pure water and dye droplets on the surfaces of the three different membranes (CPVDF, E-PH and E-PDMS) [42]

As expected, the modified membrane performed better compared

to the C-PVDF membrane with initial flux recorded of 34 kg/m<sup>2</sup>.h and 21-23 kg/m<sup>2</sup>.h, respectively, when treating 100 mg/L single dye solution. The dye rejection is proportional to the flux trend in which indicated that the PDMS/PVDF electrospun membrane obtained the highest colour removal efficiency throughout the MD tested as shown in Figure 6. Several explanations were provided by the authors that reflected on the promising performance of their in-house membrane. These include i) weak dyemembrane bonding, ii) repulsion strength between dye molecules and functional groups on the PDMS/PVDF electrospun membrane and iii) fluffy dye-dye structure produced before the dye molecules entered into the pores.



**Figure 6** Permeate flux versus time trend for four different dyes using three different membranes (C-PVDF, E-PH, and E-PDMS) (feed temperature =  $60^{\circ}$ C, flow rate = 0.5 L/min, and C<sub>0</sub> = 100 mg/L) [42]

#### 4.0 OPERATING CONDITIONS AND THEIR IMPACTS ON MD PRODUCTIVITY

The performance of the MD process in non-volatile solute removal is dependent on the feed properties and operating conditions. Obviously, the characteristics of the membrane itself explain the changes in quality and quantity of the permeate produced. In general, the efficiency of the separation process depends on many operational parameters such as dye characteristics (acidic/basic, molecular weight. etc), dye and/or salt concentration in the feed solution, feed/permeate temperature. and solution flow rate.

#### 4.1 Effect of Dye Characteristics

Theoretically, it is expected that the membrane is able to reject 100% of the dye content in the feed solution due to

its mechanism principle. However, studies conducted by researchers found that different properties of organic dyes would lead to different colour removal efficiency during the MD process. The explanation for this behaviour could be owing to the complicated interaction between the membrane and the dye molecules, which is caused by the electrostatic attraction between atoms with opposite charges or the sharing of electrons, as in covalent bonds [42]. Dyes can behave as electron acceptors and electron donors. The chromogenechromophore structure that normally responsible for the dyes' colour is working as an electron acceptor, while the auxochrome group that attached to the chromophore behaves as an electron donor. Since the membrane also has its own net electrical charge (either positive or negative, depending on solution pH), it can react with the dye molecules and form either strong

bonds through covalent or ionic bonds or weak bonds via dipole-dipole interactions and hydrogen bonding.

Only limited studies have been found in the literature describing the effect of dye characteristics on the permeate flux and dye removal efficiency [9, 10, 41, 45]. Most of the literature used single dye solution instead of dye mixture in their studies. Criscuoli et al. [9] studied four different types of organic dyes with MW in the range of 262.26 to 991.82 g/mol. The results showed that the permeate flux of the lowest MW dye (i.e., Indigo) did not necessarily result in the highest permeate flux. The dyes with bigger MW (i.e., reactive black 5 (RB5) and Remazol Brillant Blue R (RBBR)) have relatively high initial flux which is closed to 36 kg/m<sup>2</sup>.h. They explained that this behaviour is normal under VMD configuration. The meanwhile, dve rejection, was maintained at a very high level as only pure water was recovered at the permeate side.

Mokhtar et al. [40] on the other hand reported that when Crystal Violet (CV) dye was present in the feed solution, different interaction between dye molecules and membrane matrix was occurring and resulting in high permeate flux but low rate of removal. The in-house **PVDF** membrane generated extremely similar permeate fluxes (around 10 kg/m<sup>2</sup>.h) for all dyes studied, with the exception of CV which showed >17 kg/m<sup>2</sup>.h. The authors claimed that the strange behaviour of that CV can be possibly due to the high "affinity" of this particular dye towards the membrane matrix either from dye adsorption effect into the polymeric membrane or high diffussivity of CV in the aqueous solution. The presence of dye components on the membrane surface is strongly indicated by the detection of nitrogen (N) and chlorine (Cl) elements on the composite membrane surface (as illustrated in Figure 7). An et al. [45] also believed that dye characteristic could play a role in membrane performance. affecting They used three types of negatively charged commercial membranes in treating anionic and cationic dyes. negatively Results showed that charged dyes such as Acid Red 18 (AR18) and Acid Yellow (AY36) had strong repulsion with the membrane of high negative zeta potential. The interaction between the negatively charged dyes and the membrane surface promoted to the aggregation of flake type foulants.

Similar behaviour was observed in work studied the separate that of in-house made performance PDMS/PVDF electrospun membrane Using commercial [42]. **PVDF** membrane with pore size of 0.45 µm, the removal efficiency trend for several dyes are as follows: CV (96.54%) < MB (98.29%) < AY36 and AR18 (100%, respectively). Low rejections of CV and MB dyes were attributed to the fact that MB is a basic cationic dye that dissociates in aqueous solution into a cation (the chromophore) and an anion, Cl<sup>-</sup> whereas CV is a direct dye that is water-soluble and charged with organic compounds that can bond to ionic and polar sites on the membrane.



**Figure 7** EDX results on the membrane surface after treating CV dyeing solution [40]

#### **4.2 Effect of Dye Concentrations**

A part of dye characteristics, dye concentrations also play major role in determining the separation performance of MD process. In general, 10-20% of textile dyes are lost during the dyeing process resulting in effluent containing between 10 and 1000 ppm of dye components [51, 52]. Analyses revealed that the values of dye loss in the effluent may vary depending on the types of dyes used and the fixation degree as shown in Table 3. For example, sulphur and reactive dyes may have up to 40% and 50% loss in effluent, respectively.

**Table 3** Fixation degree of different dyeclasses on textile support [53]

Dye class	Fibre type	Fixation degree, %	Loss in effluent, %
Acid	Polyamide	80–95	5-20
Basic	Acrilic	95-100	0–5
Direct	Cellulose	70–95	5-30
Disperse	Polyester	90–100	0–10
Metal	Wool	90–98	2-10
complex			
Reactive	Cellulose	50–90	10–50
Sulphur	Cellulose	60–90	10–40
Dye-	Cellulose	80–95	5-20
stuff			

In MD, regardless of the MD configurations, increasing the nonvolatile solute concentration in the feed aqueous solution reduces the permeate flux. According to Khavet and Matsuura [15], this behaviour is related to the drop in water vapour pressure, the driving force, with the addition of non-volatile solute in water due to the decrease in water activity in the feed. The increasing in feed concentration increased feed viscosity and boundary layer thickness, resulting in improved mass movement. The effect of dye concentration on VMD permeate flux over time is shown in Figure 8. It is strongly indicated that the membrane flux performance tended to decline when treating high concentration ( $C_o =$ 500 ppm) of feed effluent. On the contrary, the membrane surface had a low fouling tendency when dealing with 25 ppm dye concentration due to the "dye suspension" created from the fluid dynamic established inside the system.

The same findings were reported by Mokhtar et al. [40] and An et al. [42] in the case where DCMD configuration was utilized. Mokhtar et al. [40] for instance noticed that the permeate flux tended to reduce by 12.4% by increasing dye concentration from 50 to 1000 ppm. Nevertheless, the dye rejection was still considered excellent with rejection recorded at >90% throughout the experiments. They reported that the permeate flux decline at a high solute concentration was due to the lower activity coefficient of water vapour pressure, which is normal to MD cases as reported by other researchers [15, 54]. Another possible reason for low flux performance was the fixation of dye particles on the surface of the membrane that resulted in partial or complete blockage of the pores. An et al. [42] has also demonstrated that the rapid flux reduction with increasing dye concentrations from 40 to 100 ppm and attributed to dye adsorption pore induced blockage and concentration polarization.



**Figure 8** Permeate flux as function of time at different feed concentrations [9]

## 4.3 Effect of NaCl

The effect of inorganic salts, in particular NaCl on the performance of the MD process is also important as large amounts of salts are used during the fixation process [55]. The concentration of NaCl in the dyebath is usually reported in the range of 0.7 – 1.4 molar (M) [56]. Prior to this condition, it was found that the high salt concentration in the dye solution did not deteriorate the permeate flux and final water quality. This may be justified by the low concentration of salt in the textile effluent versus brine water [8, 41].

Banat et al. [8] evaluated the commercial PP membrane by varying the NaCl concentration in an 18.5 ppm dye solution from 0.05 M and 1 M while keeping the feed temperature and flow rate constant at 50°C and 14 mL/s, respectively (see Figure 9). They found only a marginal change in the permeate flux as a function of salt concentration. A similar result was reported by Mokhtar et al. [41]. Nevertheless, the membrane fouling characteristics might be influenced by the salt concentration. Figure 10 shows that the membrane surface fouling was higher more severe at salt concentration. A EDX survey on the membrane surface properties further supported the findings as more Na and Cl content were detected on the membrane surface when testing using a higher concentration of salt solution. The detection of other elements such as F. C. O. Si and Al is attributed to the PVDF and Cloisite 15A that were used to fabricate the membrane. Since scaling is hydrophilic, the membrane is more likely to have pore wetting problems. The permeate flux, on the other hand, is not affected by the scaling because the evaporation area on the feed side did not decrease significantly. Besides, the solute rejections are still high, with a rate of >98%.



**Figure 9** Effect of NaC1 salt concentration on the permeate flux collected within the first hour [56]



**Figure 10** SEM images of the i) outer surface and ii) cross-section of the composite membrane after testing with 50 ppm ARI solution containing a) 0.1 M and b) 1.0 M NaCl [41]

#### 4.4 Effect of Feed Temperature

In MD process for textile wastewater treatment, the feed temperature used commonly ranges between 40°C and 90°C [22]. Figure 11 shows the common trend of permeate flux of dveing solution as a function of feed bulk temperature. As illustrated in the figure, the increase in feed temperature increased the permeate flow of the membrane. Banat et al. [8] correlated their results with temperature polarization effects. The effect of polarization temperature becomes greater as the temperature polarization coefficient approaches zero. If the

polarization factor concentration reaches 1, the temperature polarization effect is negligible. Wang et al. [57] on the other hand stated that at low feed temperatures, heat is easily lost by conduction through the membrane materials and the gas-filled membrane pores. However. as the feed temperature rises, the latent heat of water evaporation becomes the primary source of total heat transfer, resulting in a considerable increase in permeate flux [3]. Meanwhile, Susanto [16] explained that conductive heat losses can be minimized at a high feed mass temperature due to the higher partial vapour pressure on the feed side. Studied by Silva et al. [3] observed an abnormal observation, whereas with the reactive dye and the dispersal dye, the increase in feed temperature had various effects on permeate flow. Based on their finding, they claimed that the permeate flux increment is more pronounced for the reactive dye between 60-75°C, and for the dispersion dye between 75–90°C.



Figure 11 Permeate flux (J) and separation factor ( $\alpha$ ) as a function of different DCMD operating parameters for distilled water (DW) and the three studied dyes (MB, DY and SF) under different feed temperature (T<sub>p</sub> = 293.15 K) [47]

## 4.5 Effect of Feed and Permeate Flow Rates

Normally, an increase in feed or permeate flow rate will significantly water result in higher vapour production, mainly due to the enhancement of mass and heat transfer within the membrane module. As the temperature difference between the membrane surface and the bulk streams is marginally removed by raising the flow rates for both streams at the same time, the temperature polarization effect is expected to decrease [58]. However, in order to avoid wetting problem during the MD process, the hydraulic pressure must be lower than the wetting pressure. This is the reason why certain flow rate is obtained during the experiment which called as optimum value.

According to Silva *et al.* [3], the feed or permeate flow rate does affect the Reynolds number of the solution, whereas they found that increasing Reynolds number will reduce the residence time, the boundary layer, and the transfer resistances as presented in Figure 12. High turbulence flow is assumed to enhance permeate flow at any temperature, but this effect is multiplicative at high feed temperature.



**Figure 12** Permeate flux as a function of the feed and permeate flow rate and Reynolds number for the synthetic textile wastewaters [3]

### 5.0 FUTURE DIRECTION TOWARDS INDUSTRIAL IMPLEMENTATION

Other than textile industry, MD also shows good potential to treat other industrial wastewaters produced from olive oil mill, pharmaceutical and radioactive industries [11, 59-61]

These industries produce highly variable wastewater quality due to the different manufacturing steps. The characteristics of each type of wastewater are very different from each other, leading to very different and specific issues. The adoption of MD technologies for this industrial wastewater could be a great choice as this membrane process could produce less hazardous permeate and may also recover some of the valuable compounds in the feed present solutions.

The proposed DCMD process is highly effective in eliminating hazardous contaminants from textile wastewater. The permeate produced by the process is comparable to deionized water, suggesting the high efficiency level of this thermal membrane process for industrial wastewater recovery. However, more research is required to improve permeate flow, particularly long-term flow stability for industrial scale implementation. Although the fouling problem is not as serious as pressure membrane processes, it will still affect the long-term process in an actual application. Contamination induced by the fixation of organic/inorganic substances contained in wastewater on the surface of the membrane will require further research in the future.

#### 6.0 CONCLUSION

MD technology can potentially lead to a major innovation in the textile industry, particularly in the effluent treatment process. It demonstrates release of non-volatile excellent compounds from textile effluent. However, MD only takes effect if several disadvantages been have addressed. Compared to the water desalination studies, the use of MD for wastewater treatment has been gaining the attention of researchers in recent years. To date, there is still no report on the industrial scale implementation of the MD process for wastewater treatment. Since textile wastewater contains various types of chemicals, the membrane must be capable of maintaining its changing properties after interacting with these chemicals. A part of high tolerance to chemicals, the membrane must be highly hydrophobic to prevent pore wetting.

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