

Direct contact membrane distillation for textile wastewater treatment: a state of the art review

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ABSTRACT

To meet surging water demands, water reuse is being sought as an alternative to traditional water resources. Direct contact membrane distillation (DCMD) has been increasingly studied in the past decade for its potential as an emerging cost effective wastewater treatment process and subsequent water reuse. This review presents a comprehensive overview of the current progress in the application of DCMD for textile wastewater treatment based on the available state of the art. There are already published review papers about the membrane distillation process, but the difference in the present work is that it focuses on the textile area, which consumes a lot of water and generates large amounts of wastewater, and still needs innovations in the sector. A review focused on the textile sector draws the attention of professionals to the problem and, consequently, to a solution. Current issues such as the influences of feed solution, membrane characteristics and membrane fouling and new insights are discussed. The main performance operating conditions and their effects on the separation process are given. Likewise, challenges associated with the influence of different dyes on the DCMD results are explained. This review also highlights the future research directions for DCMD to achieve successful implementation in the textile industry.

Key words | color removal, DCMD, dyeing effluent, dye recovery, textile dyes, water reuse

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INTRODUCTION

Water scarcity due to rapid and continuous industrialization, urbanization and population and agricultural growth has raised considerable concerns on the sustainability of water resources, leading people to find alternatives to alleviate the lack of freshwater (Chekli *et al.* 2016; Elminshawy *et al.* 2016; Goh *et al.* 2016).

According to the *World Trade Statistical Review 2016* by the World Trade Organization, the current value of world textile exports totaled US\$291 billion in 2015 (World Trade Organization 2016). Despite being an important contributor to the economy, the textile industry represents one of the bigger industrial freshwater-consuming sectors (Angelis-Dimakis *et al.* 2016), generating a large quantity of wastewater. The World Bank estimates that about 17–20% of industrial water pollution is due to the textile industry (Kant 2012). Under this condition, the textile industry is not supporting a sustainable development and has to face the pressure to recover and reuse its wastewater, so as to

fulfill the demand of increasingly strict regulations. Soon many textile companies will have to reuse dyeing secondary effluent to achieve environmental and economic benefits.

Membrane technology is an attractive method to treat and reuse textile wastewater. Particularly, direct contact membrane distillation (DCMD) has been demonstrated to be potentially applicable. However, the use of DCMD for textile wastewater treatment has been limited to only a few academic publications. Most of the current DCMD researches are focused on desalination for water treatment. According to searches in the literature, in the last 5 years more than 95% of the total academic publications about DCMD applied to desalination, rather than to textile wastewater treatment. Therefore, new research is fundamental to enable textile wastewater treatment by the DCMD process.

The aim of this work is to present and discuss the current available state of the art of DCMD applied specifically to textile wastewater treatment.

MD principle

Membrane distillation (MD) is a thermally driven separation process in which only vapour molecules transfer through a porous hydrophobic membrane from the feed/retentate side and are collected, after condensation, at the permeate/distillate side (Alkhubiri *et al.* 2012). In other words, a temperature difference across membrane surfaces induces the vapour pressure difference on both sides of the membrane.

The nature of the driving force and the hydro-repellent character of the membrane reject theoretically 100% of non-volatile solutes such as macromolecules, colloidal species, inorganic ions and other non-volatile compounds (Drioli *et al.* 2015). The performance of the MD system is measured mainly by the quantity and quality of the permeate flow. MD is not fully implemented yet in industry (Khayet 2011), due to membrane and module design, membrane pore wetting, low permeate flow rate and flux decay as well as uncertain energy and economic costs.

MD processes are commonly classified into four configurations. In the DCMD process, the membrane is in direct contact only with the liquid phases, solution on one side and freshwater on the other (only the gas phase exists inside the membrane pores). Out of the four configurations, DCMD is the most favourable configuration due to its inherent simplicity without the need for external condensers (Alklaibi & Lior 2005) and more stable distillate flux (Aneesh *et al.* 2016). Figure 1 shows a schematic diagram of a typical DCMD process unit (in this case, an example of a flat sheet membrane module with countercurrent flow). There are other MD configurations, but this study refers only to the DCMD process. In this configuration, the feed solution is heated before entering the module. When in direct contact with the membrane surface, some of the feed volatile molecules evaporate. The remaining feed solution returns to the feed tank to be heated again. The vapour of the feed solution penetrates through the membrane pores and condenses in the permeate side, which is cooled with a chiller.

A large variety of membrane modules, including flat sheet (plate-and-frame and spiral-wound modules) and tubular (tubular, capillary and hollow fiber modules), have been tested in MD applications. An appropriate module design can reduce the thermal/concentration polarization, fouling and energy consumption of the process and provide compactness (Drioli *et al.* 2015).

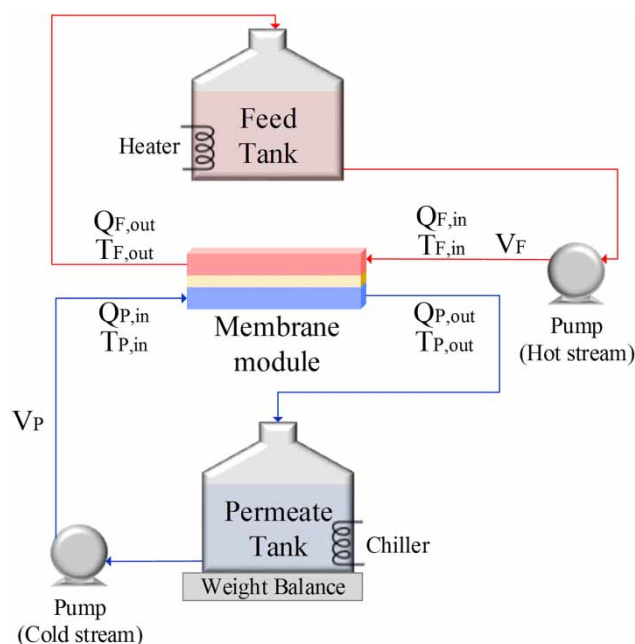


Figure 1 | Schematic diagram of a typical DCMD unit, where Q_F and Q_P are the mass flow rates, T_F and T_P are the temperatures and V_F and V_P are the velocities of feed and permeate sides, respectively.

Textile wastewater treatment

Along the entire value chain of the textile industry, large amounts of freshwater are consumed (Yang *et al.* 2015). The dyeing process leads to the most severe water pollution in textile wastewater. The main chemicals usually used during the textile dyeing processes are a mixture of different types of dyes and different auxiliary products (Arslan-Alaton & Alaton 2007), resulting in a wastewater with high pH, intensive color and salinity (Bilinska *et al.* 2016).

It is estimated that approximately 21–377 L of water is consumed for 1 kg of textile product (Asghar *et al.* 2015), confirming the potential for water reuse in the textile industry. The fabric retains only a certain quantity of dye during the dyeing process, resulting for example in a considerable amount (10–60%) of unfixed reactive dye in textile wastewater, causing highly colored effluent release (Rosa *et al.* 2015).

Dyes are resistant to chemical degradation on exposure to water, light and many chemical treatments, due to their chemical structures (Chen *et al.* 2015). Highly colored dye solutions are aesthetically undesirable and can potentially damage ecosystems because the dyes reduce light transmission, which will further disturb biological processes. Some dyes are toxic, non-biodegradable and can be harmful to human health (Métivier-Pignon *et al.* 2003; Tan *et al.* 2008).

The usual wastewater treatment methods consist of physical–chemical and biological processes (Blanco *et al.* 2014). However, these techniques have certain disadvantages such as high capital cost and operational costs, or a secondary sludge disposal problem (Ravikumar *et al.* 2005). These methods are able to meet legislative requirements but they do not enable water reuse in textile processes (Barredo-Damas *et al.* 2010).

As an alternative to conventional wastewater treatment methods, membrane-based processes have attracted attention due to their inherent simplicity, scalable modular design, easy maintenance and excellent rejection rate (Tang & Chen 2002; Dasgupta *et al.* 2015). Pressure-driven membrane processes, such as nanofiltration and ultrafiltration, have some limitations such as strong tendency to fouling and scaling, limited product water recovery and huge electrical energy consumption required to work at extremely high operating pressures (Liu *et al.* 2011; Ong *et al.* 2012; Baghbanzadeh *et al.* 2015).

DCMD is promising to overcome the limitations of pressure-driven membrane processes, due to a low operating pressure under mild temperatures (An *et al.* 2016; Mokhtar *et al.* 2016). The membrane acts as a barrier between the two faces and selectivity is determined by the vapour–liquid equilibrium involved. When comparing to conventional wastewater treatment, based on the coagulation–precipitation method and biological processes, the size of the DCMD unit requires less area to operate, due to the decreased vapour space necessity that arises from the high mass transfer area per unit volume of the process (Dow *et al.* 2017). Moreover, the DCMD process does not demand chemical products during operation, whereas for conventional treatment, there is consumption of chemical products, such as aluminium and iron salts. Although the technical part of the DCMD process applied to textile wastewater treatment is already in progress, there is still a lack of an economic assessment in order to evaluate the economic feasibility and to compare possible advantages to conventional treatment. The economic disadvantages of the DCMD process include thermal energy and electricity consumptions and membrane replacement costs. However, some economic advantages are saving in water reuse in the industrial process and potential for recovery of heat by hot textile effluent for thermal energy during DCMD operation. A complete study is necessary to assess the economic planning of the DCMD process applied to textile wastewater treatment, since the return on the investment's cost is possibly fast due to its economic advantages.

The dye house is of particular interest since water at a temperature that can be higher than 100 °C is utilized (Calabro *et al.* 1990). Thus, DCMD is interesting for textile wastewater treatment application, since a recovery of heat is possible directly near to the fabric dyeing machine. DCMD can exploit the free energy given by hot effluent discharged by the textile industry and requires significantly lower energy consumption during operation (Mokhtar *et al.* 2016).

Dyes are non-volatile substances, therefore are theoretically completely separated by the DCMD process (Mozia *et al.* 2010). A mixture of dyes composes textile wastewater; thus, application of the DCMD process for this wastewater is focused mainly on water recovery rather than recovery of dyes. Therefore, a concentrated dyebath resulting from the remaining feed solution needs to be further treated. Some sustainable alternatives arise for the treatment of the remaining feed solution, for example recovery of dye by adsorption (Mu & Wang 2016; Ngulube *et al.* 2017) and studies of saturation by applying evaporation and concentration of the dye (and other auxiliaries) in order to separate it from the aqueous solution. If the DCMD process is applied directly near to the dyeing machine, another sustainable alternative would be the reuse of the concentrated retentate in the dyeing step by adjusting its composition.

Table 1 shows an overview of the most common textile dye classification (Akbari *et al.* 2002; Lau & Ismail 2009). It is important to point out that different classes (and even different dye colors of the same class) show different results in the DCMD process.

CURRENT ISSUES AND RECENT ADVANCES

Most of the MD academic publications applied to textile wastewater treatment are focused on DCMD configuration, likely because of its relatively simple operation mode and low maintenance cost (Mokhtar *et al.* 2015a). To the best of our knowledge, searches in the literature show that for textile wastewater treatment application, there are 22 academic publications focused on DCMD and only three academic publications focused on the submerged photocatalytic MD reactor (Qu *et al.* 2014) and vacuum MD (Banat *et al.* 2005; Criscuoli *et al.* 2008).

The DCMD academic publications for treating textile industrial/synthetic wastewater are presented as follows. Mokhtar *et al.* (2015a, 2016) evaluated the performance of a polyvinylidene fluoride (PVDF) 3% Cloisite 15A membrane. An *et al.* (2017) developed a hybrid PDMF/PH membrane

Table 1 | Overview of textile dye classes

Class	Characteristics	Chemical types	Substrates	Method of application	Examples	Degree of dye fixation (%)
Acid	Water-soluble, anionic functional groups	Nitroso, nitro, azo, triphenylmethane, triarylmethane, xanthene, quinoline, azine, anthraquinone	Wool, nylon, silk, leather	Usually from neutral to acidic dyebaths (pH range 3.0–7.0)	Acid red 1 (AR1), acid red 18 (AR18), acid yellow 36 (AY36), acid orange 8 (AO8), methyl orange (MO)	89–95
Basic	Water-soluble, cationic functional groups	Azo, triarylmethane, xanthene, acridine, quinoline, methine, thiazole, azine, oxazine, thiazine, anthraquinone, phthalocyanine	Acrylic, modified nylon, polyester	Applied from acidic dyebaths	Crystal violet (CV), methylene blue (MB)	95–100
Direct	Water-soluble, anionic functional groups (larger structure than acid dyes)	Azo, stilbene, oxazine, anthraquinone, phthalocyanine	Cellulose fibers, rayon ^a , leather, nylon	Applied from neutral or slightly alkaline baths containing inorganic salts	Congo red (CR), direct green 99 (DG99)	70–95
Disperse	Very low water solubility	Azo, methine, polymethine, anthraquinone, phthalocyanine	Polyester, acetate, polyamide, acrylic	Fine aqueous dispersions often applied by high temperature pressure or lower temperature carrier methods	Disperse violet 1, disperse black 9	90–100
Reactive	Water-soluble, anionic functional groups	Azo, anthraquinone, phthalocyanine,	Cellulose fibers, nylon, wool, silk	Reactive site on dye reacts with functional group on fiber to bind dye covalently under influence of heat and pH (alkaline)	Reactive orange 16 (RO16), reactive black 5 (RB5)	50–90
Sulfur	Water-insoluble, colloidal	Sulfur	Cellulose fibers, rayon ^a	Aromatic substrate vatted with sodium sulfide and re-oxidized to insoluble sulfur-containing products on fiber	Sulfur black 1, sulfur green 6	60–90
Vat	Water-insoluble, colloidal, carbonyl group	Sulfur, aminoketone, hydroxyketone, indigoid	Cellulose fibers, rayon ^a	Dyes solubilized by reducing with sodium hydrosulfite, then exhausted on fiber and re-oxidized	Indigoid vat dye, anthraquinone vat dye	80–95

^aAlso known as viscose.

(E-PDMS) with superhydrophobicity and compared it with a poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) electrospun membrane (E-PH) and a

commercial PVDF membrane (C-PVDF). *An et al.* (2016) investigated a polytetrafluoroethylene (PTFE) membrane and two PVDF membranes. *Lin et al.* (2015) studied a

PVDF membrane attached with a thin layer of agarose hydrogel. Mokhtar *et al.* (2015b) investigated an 18 wt.% PVDF membrane blended with ethylene glycol (PVDF-EG). Mokhtar *et al.* (2015c) and Mokhtar *et al.* (2013) studied PVDF membranes blended with ethylene glycol and polyvinylpyrrolidone. Mokhtar *et al.* (2014a) evaluated PVDF membranes with different polymer concentrations (12, 15 and 18 wt.%). Mokhtar *et al.* (2014b) studied a PVDF-Cloisite 15A membrane with different Cloisite 15A concentration (3, 5 and 10 wt.%). Huo *et al.* (2013) researched a PVDF membrane in a photocatalytic membrane reactor combining a visible-light BiOBr photocatalyst and DCMD process. Ge *et al.* (2012) investigated a PTFE membrane in a forward osmosis-DCMD hybrid system. Mozia *et al.* (2005, 2006, 2007, 2009a, 2009b, 2010), Mozia & Morawski (2006) and Grzechulska-Damszel *et al.* (2010) studied a polypropylene (PP) membrane in a hybrid system coupling photocatalysis with DCMD. Calabro *et al.* (1991, 1990) evaluated a PP membrane and observed that it was possible to concentrate all dye solutions with the DCMD process. Hereafter current issues and recent advances of DCMD applied to textile wastewater will be presented and discussed.

Feed composition

Figure 2 shows the number of academic publications for the DCMD treatment of textile wastewater according to the source of the feed solution. Most of the DCMD academic publications applied to textile wastewater treatment utilized synthetic solutions as wastewater model. Currently there are only three studies which employed industrial textile wastewater: combined textile effluent either untreated or treated with flocculation and biological processes (Dow *et al.* 2017), textile factory wastewater from different sewing threads and industrial yarn processes (Mokhtar *et al.* 2016)

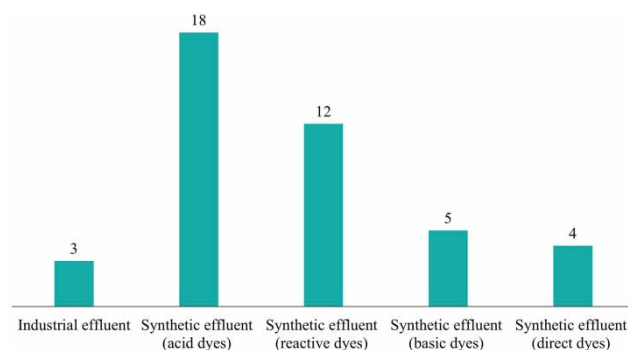


Figure 2 | Number of academic publications about DCMD applied to textile wastewater treatment according to the source of the feed solution.

and polyester dyeing wastewater (Lin *et al.* 2015). For synthetic textile wastewater, the most studied textile dyes are in the following order: acid dyes (including azo dyes) > reactive dyes > basic dyes > direct dyes. This order is justified, because acid dyes are the largest group of dyes used in the textile industry (Lucas & Peres 2006; Lau & Ismail 2009) and are the most problematic compared to other forms of dyes and must be completely removed from wastewater (Lee *et al.* 2006).

The dye class and its concentration, due to the complex composition of dyeing wastewater, are important parameters for the efficiency of DCMD treatment. Table 2 shows a summary of the studied dyes and their concentrations in DCMD researches applied to textile wastewater

Table 2 | Summary of studied dyes and their concentrations in DCMD researches applied to textile wastewater treatment (see Table 1 for dye nomenclature)

Feed solution	Feed concentration (mg·L ⁻¹)	Reference
Industrial effluent	N/A	Dow <i>et al.</i> (2017)
MB; CV; AR18; AY36	100	An <i>et al.</i> (2017)
Industrial effluent	N/A	Mokhtar <i>et al.</i> (2016)
MB; CV; AR18; AY36	100	An <i>et al.</i> (2016)
Industrial effluent	N/A	Lin <i>et al.</i> (2015)
AR1; CR; CV; RO16; RB5	50	Mokhtar <i>et al.</i> (2015a)
RB5	500	Mokhtar <i>et al.</i> (2015b)
RB5	500	Mokhtar <i>et al.</i> (2015c)
RB5	50	Mokhtar <i>et al.</i> (2014a)
RB5	50	Mokhtar <i>et al.</i> (2014b)
RB5	500	Mokhtar <i>et al.</i> (2013)
MO	15	Huo <i>et al.</i> (2013)
AO8	50	Ge <i>et al.</i> (2012)
AR18; AY36; DG99	30	Mozia <i>et al.</i> (2010)
AR18	30	Mozia <i>et al.</i> (2009a)
AY36	30	Mozia <i>et al.</i> (2009b)
AR18; AY36; DG99	10	Grzechulska-Damszel <i>et al.</i> (2010)
AR18; AY36; DG99	30	Mozia <i>et al.</i> (2007)
AR18	30	Mozia <i>et al.</i> (2006)
AR18	30	Mozia & Morawski (2006)
AR18	30	Mozia <i>et al.</i> (2005)
Red E-4BA; blue E-G	5,000	Calabro <i>et al.</i> (1991)
Blue E-G; blue E-BA; red E-RA	5,000	Calabro <i>et al.</i> (1990)

treatment. Dye concentration in the academic publications varies from 10 to 5,000 mg·L⁻¹. AR18 is the major chosen dye in the academic publications because of its wide use in the textile industry (Khosravi *et al.* 2016), followed by RB5, probably because it is highly toxic, mutagenic and carcinogenic in nature and cannot be easily treated by conventional methods (Umpuch & Sakaew 2015).

An *et al.* (2016, 2017), Mokhtar *et al.* (2015a), Mozia *et al.* (2010), Grzechulska-Damszel *et al.* (2010) and Mozia *et al.* (2007) obtained different permeate fluxes for different dye classes and similar permeate fluxes for a same class. Likewise, Calabro *et al.* (1990, 1991) obtained similar permeate fluxes for the same dye class. Most of the academic publications demonstrated experimental data of DCMD process for a synthetic solution composed of one dye class (Mozia *et al.* 2005, 2006, 2009a, 2009b; Mozia & Morawski 2006; Ge *et al.* 2012; Huo *et al.* 2013; Mokhtar *et al.* 2013, 2014a, 2014b, 2015b, 2015c).

Mokhtar *et al.* (2015a) documented that both permeate flux and dye rejection decreased when increasing dye concentration from 50 to 1,000 ppm. Likewise, Calabro *et al.* (1990) observed that an increase in the dye concentration reduces the permeate flux. The permeate flux decline at high solute concentration can be caused by the phenomenon of lower activity coefficient of water vapour pressure, by the increasing temperature polarization (Martínez-Díez & Vázquez-González 1999) and by the increase of feed viscosity and boundary layer thickness, leading to higher mass transfer resistances (Liu & Wang 2013). The reduction of permeate flux and dye rejection at high concentration of dye solution is also possibly because of dye particles attached on the membrane surface, which leads to either partial or full pore blockage, causing an additional fouling layer (Mokhtar *et al.* 2013, 2015a).

Chemical bonds between the dye molecules and the membrane are based on an attraction between the atoms within the chemical compounds (An *et al.* 2017). Dye molecules can be absorbed onto the membrane surface due to physicochemical interactions, i.e., hydrophobic interactions (dispersion forces), polar interactions (dipole forces), and charge transfer (hydrogen bonding), and their absorption onto the membrane may affect the permeate flux (An *et al.* 2016). As an example, basic dyes are cationic (positively charged), reacting well with negatively charged material, while the acid dyes (negatively charged) react well with positively charged material and are typically a salt of sulfuric, carboxylic or phenolic organic acid (An *et al.* 2016, 2017). The authors corroborated that when the membrane possesses negative charges, the treatment of acid azo dyes, which have the same membrane charge, show a better DCMD performance than treatment of basic dyes (MB

and CV). Likewise, Mokhtar *et al.* (2015a) obtained very similar permeate fluxes for all dyes studied, except for CV, possibly due to its high affinity towards the aromatic rings of the dye molecule and the membrane via van der Waals bonds. Moreover, the authors explained that the high diffusivity of CV in aqueous solution could be another reason for less promising dye removal.

Cotton is the most important and widely used textile fiber in the world and consists of around 88–96% of pure cellulose (Gamal *et al.* 2010), which commonly carries negative charge, which creates repulsion with anionic dyes. Therefore, a high amount of salt is needed to promote dye–fiber fixation, leading the effluent to have high salt concentration (Mokhtar *et al.* 2015a). In addition to dyes, salts were also evaluated in the academic publications. Mokhtar *et al.* (2015a) and Banat *et al.* (2005) concluded that an increase in feed salt concentration had a negligible effect on the permeate flux performance. However, membrane scaling, i.e. when the feed salt concentration reaches supersaturation, due to high product water recovery and concentration and temperature polarizations, has an extreme impact on the process performance, requiring more research. Calabro *et al.* (1990) tested a Na₂SO₄ 5,000 ppm solution and obtained a steady state value of the permeate flux after 3–4 h of experiment; nevertheless, the authors also tested a NaCl solution, which resulted in an 85% permeate flux reduction after increasing salt concentration from 1 M to 6 M. It must be considered that the presence of non-volatile components such as salts in diluted aqueous solutions containing organic compounds may alter the vapor pressure of the solution (El-Bourawi *et al.* 2006).

Membrane and module characteristics

Figure 3(a) shows the percentage of academic publications on DCMD treatment of textile wastewater according to

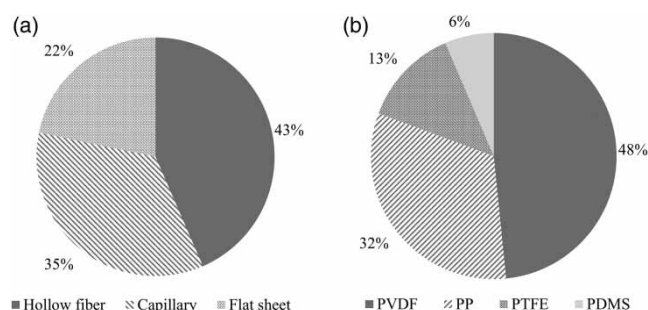


Figure 3 | Percentage of academic publications about DCMD applied to textile wastewater treatment according to (a) membrane module and (b) membrane material.

membrane module. Most of the DCMD academic publications employ hollow fiber and capillary membrane modules, possibly due to their high rate of mass transfer, which is a result of their high surface density area.

The membrane material is a very significant determining factor for the efficiency of DCMD. Figure 3(b) shows the percentage of academic publications on DCMD treatment of textile wastewater according to membrane material. Most of the DCMD academic publications used PVDF and PP membrane materials. Actually, most of the membranes employed in MD are indicated for the microfiltration process. Accordingly, the problem of developing microporous membranes with high hydrophobic character and stability, with a narrow distribution of pore size and improved structural and morphological characteristics, is a crucial aspect to the preparation of new specific membranes for MD (Curcio & Drioli 2005).

Currently, PVDF has been the major polymer material used for MD process, due to its advantage of dissolving in a variety of solvents at room temperature (Mokhtar *et al.* 2014a). PVDF membranes have moderate thermal stability and good chemical resistance, while PP membranes have a lower membrane performance (moderate thermal stability at elevated temperatures) (Curcio & Drioli 2005). A PP membrane has complicated processability, and can only be fabricated either by molten extrusion technique followed by stretching or by thermal phase separation process at high temperature (Khayet 2011). This is a possible reason for PP membranes not being used in recent academic publications.

Despite not being so employed in the academic publications, PTFE membranes have good thermal stability and good resistance to chemical attack and result in high water vapour permeability and high wetting resistance (high porosity and high hydrophobicity) (Zhang *et al.* 2012). Zhang *et al.* (2010) and An *et al.* (2016) verified that PTFE produced higher flux than PVDF as a DCMD membrane material.

In order to improve DCMD process performance, new membrane materials, such as ceramics and metals, are being studied. Mokhtar *et al.* (2014b, 2015a, 2016) obtained promising results with a PVDF-Cloisite 15A polymer membrane composed of montmorillonite clays. An *et al.* (2017) fabricated a PVDF-HFP electrospun membrane by hybridizing polydimethylsiloxane (PDMS) polymeric microspheres with superhydrophobicity onto the E-PH membrane via electrospinning.

Table 3 shows a summary of some membrane and module characteristics researched in the academic publications for the DCMD process applied to textile wastewater treatment.

Mokhtar *et al.* (2013) concluded that the degree of hydrophobicity is an important parameter in the MD process as the hydrophobic character of the PVDF-EG membrane could have greater pore wetting resistance and reduced fouling compared to the polyvinylidene fluoride–polyvinylpyrrolidone (PVDF-PVP) membrane. The authors confirmed that the membrane with higher contact angle has greater stability in terms of flux and dye rejection compared to the membrane with low hydrophobic property. Ge *et al.* (2012) reported a contact angle of the PVDF membrane of 114° and an excellent membrane wetting resistance, thermal efficiency maximization and permeation flux enhancement, due to its excellent hydrophobicity, suitable pore structure and high porosity. An *et al.* (2016) obtained a wettability reduction when using a PTFE membrane, due to its higher surface hydrophobicity (dye particles were attached on the membrane surface rather than in the membrane pores). Compared to the PVDF membrane, PTFE and E-PDMS membranes showed not only higher water contact angle but also higher dye contact angle (An *et al.* 2016; 2017).

Porosity in the academic studies varies from 69.24 to 87.84%. An *et al.* (2016) indicated that the PTFE membrane achieved greater permeate flux than the PVDF membranes due to its higher porosity and hydrophobicity. This can be explained because higher porosity membranes have a larger evaporation surface area (Alkhubdhiri *et al.* 2012).

In the academic publications, the mean pore size varies from 0.088 to 0.52 µm, 0.2 µm being the most studied mean pore size. A large pore size is required for high permeate flux and low mass transport resistance, while the pore size should be small to avoid liquid penetration. As a result, the optimum pore size should be determined for each feed solution and operating condition.

Permeate flux increases with an increase in the membrane pore size and porosity, and with a decrease of the membrane thickness and pore tortuosity (Drioli *et al.* 2015). The following works corroborate this fact. Mokhtar *et al.* (2014a) obtained the best result with the 12 wt.% PVDF membrane owing to its highest membrane porosity and largest mean pore size. Mokhtar *et al.* (2014b) found that among the three PVDF–Cloisite 15A membranes, the one with the incorporation of 3 wt.% Cloisite 15A was the ideal, because of its good properties and complete dye rejection with consistent permeate flux due to its highest liquid entry pressure, lowest thickness, highest overall porosity and highest contact angle value.

According to Ashoor *et al.* (2016), DCMD process performance is largely affected by flow path of the feed and permeate (countercurrent or concurrent), material of

Table 3 | Summary of membrane and module characteristics studied in DCMD researches applied to textile wastewater treatment

Module	Material	Mean pore size (μm)	Porosity (%)	Flow configuration	Contact angle (°)	Thickness (μm)	Reference
Flat sheet	PTFE	0.5	N/A	Countercurrent	N/A	N/A	<i>Dow et al. (2017)</i>
Flat sheet	PVDF-C; E-PH; E-PDMS	0.45; 0.52; 0.49	72.11; 87.28; 87.84	Countercurrent	118.3; 137.2; 155.4	105; 98; 102	<i>An et al. (2017)</i>
Hollow fiber	PVDF + nanocomposite	0.088	83.70	Countercurrent	97.7	N/A	<i>Mokhtar et al. (2016)</i>
Flat sheet	PVDF 0.22; PVDF 0.45; PTFE	0.22; 0.45; 0.20	69.24; 72.11; 84.32	Countercurrent	120.2; 117.9; 137.9	N/A	<i>An et al. (2016)</i>
Flat sheet	PTFE + agarose hydrogel	0.2	85.00	Countercurrent	127	130	<i>Lin et al. (2015)</i>
Hollow fiber	PVDF + nanocomposite	0.088	83.70	Countercurrent	97.72	N/A	<i>Mokhtar et al. (2015a)</i>
Hollow fiber	PVDF	0.15	73.18	Countercurrent	83.7	155	<i>Mokhtar et al. (2015b)</i>
Hollow fiber	PVDF-EG; PVDF-PVP	0.15; 0.17	N/A	Countercurrent	87; 68	N/A	<i>Mokhtar et al. (2015c)</i>
Hollow fiber	PVDF12; PVDF15; PVDF18	0.14; 0.10; 0.09	78.08; 75.86; 70.77	Countercurrent	82.2; 86.47; 91.96	130–170	<i>Mokhtar et al. (2014a)</i>
Hollow fiber	PVDF + nanocomposite	0.088	83.70	Countercurrent	97.72	127	<i>Mokhtar et al. (2014b)</i>
Hollow fiber	PVDF-EG; PVDF-PVP	0.15; 0.17	N/A	Countercurrent	87; 68	N/A	<i>Mokhtar et al. (2013)</i>
Flat sheet	PTFE	0.45	80.00	Countercurrent	145	140	<i>Huo et al. (2013)</i>
Hollow fiber	PVDF	0.28	78.80	Concurrent	114	N/A	<i>Ge et al. (2012)</i>
Capillary	PP	0.20	N/A	Concurrent	N/A	N/A	<i>Mozia et al. (2010)</i>
Capillary	PP	0.20	N/A	Concurrent	N/A	N/A	<i>Mozia et al. (2009a)</i>
Capillary	PP	0.20	N/A	N/A	N/A	N/A	<i>Mozia et al. (2009b)</i>
Capillary	PP	N/A	N/A	Concurrent	N/A	N/A	<i>Grzechulska-Damszel et al. (2010)</i>
Capillary	PP	N/A	N/A	Concurrent	N/A	N/A	<i>Mozia et al. (2007)</i>
Capillary	PP	0.20	N/A	N/A	78	54	<i>Mozia et al. (2006)</i>
Capillary	PP	0.20	N/A	Concurrent	N/A	N/A	<i>Mozia & Morawski (2006)</i>
Capillary	PP	0.20	N/A	Concurrent	N/A	N/A	<i>Mozia et al. (2005)</i>
Hollow fiber	PP	0.45	70.00	Countercurrent	N/A	N/A	<i>Calabro et al. (1991)</i>
Hollow fiber	PP	N/A	N/A	Countercurrent	N/A	N/A	<i>Calabro et al. (1990)</i>

N/A: not available.

system construction, means of heat exchange, mechanical and thermal properties of feed and permeate, and system configuration. Most of the academic publications are based on countercurrent flow configuration. However, it was noticed that flow path and system configuration/design were hardly explained in the academic publications. *Drioli et al. (2015)* indicated that the structure of the thickness

film and the module geometry affect DCMD performance. Nevertheless, this information was not often provided.

Process operating conditions

Major determinants of the performance of the DCMD are its operating conditions. The fundamental operating parameters

of the MD process are the feed and permeate temperature and mass flow rate.

Table 4 shows a summary of the operating conditions researched in academic publications for the DCMD process

applied to textile wastewater treatment. The academic publications demonstrated a feed velocity/flux of 0.016–0.5 m·s⁻¹/0.5–4 L·m⁻¹ and permeate velocity/flux of 0.002–0.18 m·s⁻¹/0.2–3 L·m⁻¹. Mokhtar *et al.* (2015a)

Table 4 | Summary of the operating conditions researched by academic publications for the DCMD process applied to textile wastewater treatment

Feed velocity/ flux (m·s ⁻¹) (V _F)	Permeate velocity/flux (m·s ⁻¹) (V _P)	Feed temperature (°C) (T _F)	Permeate temperature (°C) (T _P)	Mean permeate flux (kg·m ⁻² ·h ⁻¹) (short-term study) (Q _P)	Mean permeate flux (kg·m ⁻² ·h ⁻¹) (long-term study) (time) (Q _P)	Reference
0.041	0.041	60	20	5 L·m ⁻² ·h ⁻¹	2 L·m ⁻² ·h ⁻¹ (1,560 h)	Dow <i>et al.</i> (2017)
0.5 L·min ⁻¹	0.5 L·min ⁻¹	60	20	22–23 L·m ⁻² ·h ⁻¹ (C-PVDF); 32–33 L·m ⁻² ·h ⁻¹ (E-PH); 32–33 L·m ⁻² ·h ⁻¹ (E-PDMS) ^a	14–20 L·m ⁻² ·h ⁻¹ (C-PVDF); 23– 29 L·m ⁻² ·h ⁻¹ (E-PH); 23–29 L·m ⁻² ·h ⁻¹ (E-PDMS) (24 h) ^a	An <i>et al.</i> (2017)
0.023	0.002	90	25	38	15 (40 h) ^a	Mokhtar <i>et al.</i> (2016)
0.5 L·min ⁻¹	0.5 L·min ⁻¹	60	20	20.5–21.5 L·m ⁻² ·h ⁻¹ (PVDF 0.22); 21.5– 23 L·m ⁻² ·h ⁻¹ (PVDF 0.45); 31– 34 L·m ⁻² ·h ⁻¹ (PTFE) ^a	14.5–18.5 L·m ⁻² ·h ⁻¹ (PVDF 0.22); 15.5– 18.5 L·m ⁻² ·h ⁻¹ (PVDF 0.45); 24–30 L·m ⁻² ·h ⁻¹ (PTFE) (24 h) ^a	An <i>et al.</i> (2016)
2 L·min ⁻¹	2 L·min ⁻¹	60	21	20 L·m ⁻² ·h ⁻¹ ^a	20 L·m ⁻² ·h ⁻¹ (24 h) ^a	Lin <i>et al.</i> (2015)
0.023	0.01	70	20	N/A	12.42 (N/A) ^b	Mokhtar <i>et al.</i> (2015a)
0.016	0.01	60	20	8.3 ^a	9.82 (6 h)	Mokhtar <i>et al.</i> (2015b)
4 L·min ⁻¹	3 L·min ⁻¹	60	20	N/A	9.82 (PVDF-EG) (5 h)	Mokhtar <i>et al.</i> (2015c)
0.034	0.016	80	20	5.5 (12PVDF) ^a	5.8 (12PVDF) (2.5 h) ^a	Mokhtar <i>et al.</i> (2014a)
0.023	0.01	70	20	10.5 ^a	10 (2.5 h)	Mokhtar <i>et al.</i> (2014b)
4 L·min ⁻¹	3 L·min ⁻¹	60	20	8.39 (PVDF-EG); 10.5 (PVDF-PVP) ^a	9.81 (PVDF-EG); 8.4 (PVDF-PVP) (2.5 h) ^a	Mokhtar <i>et al.</i> (2013)
N/A	N/A	45	15	11	11 (35 h) ^c	Huo <i>et al.</i> (2013)
0.5 L·min ⁻¹	0.2 L·min ⁻¹	66	20	N/A	N/A	Ge <i>et al.</i> (2012)
0.5	0.18	62	20	N/A	12.6 L·m ⁻² ·h ⁻¹ (5 h) ^c	Mozia <i>et al.</i> (2010)
0.5	0.18	62	20	N/A	12.6 L·m ⁻² ·h ⁻¹ (5 h) ^c	Mozia <i>et al.</i> (2009a)
0.33	0.095	70	20	N/A	14 L·m ⁻² ·h ⁻¹ (5 h) ^c	Mozia <i>et al.</i> (2009b)
0.5	0.18	62	20	N/A	N/A	Grzechulska-Damszel <i>et al.</i> (2010)
0.31	0.09	60	20	N/A	14.17 L·m ⁻² ·h ⁻¹ (5 h) ^c	Mozia <i>et al.</i> (2007)
0.33	0.095	70	20	N/A	14 L·m ⁻² ·h ⁻¹ (5 h) ^{a,b,c}	Mozia <i>et al.</i> (2006)
0.31	0.09	67	20	N/A	14 L·m ⁻² ·h ⁻¹ (20 h) ^c	Mozia & Morawski (2006)
0.33	0.095	70	20	N/A	16.7 L·m ⁻² ·h ⁻¹ (140 h) ^{a,b,c}	Mozia <i>et al.</i> (2005)
0.026	N/A	50	35	1.512; 1.62	0.396; 0.432 (N/A)	Calabro <i>et al.</i> (1991)
1.5 L·min ⁻¹	1.5 L·min ⁻¹	50	35	N/A	16.25; 15.83; 13.75 L·m ⁻² ·h ⁻¹ (N/A) ^a	Calabro <i>et al.</i> (1990)

^aObtained from the graphics.

^bObtained for AR1 (50 ppm) and NaCl (1.0 M).

^cResults of a combined photocatalysis–DCMD process.

N/A: not available.

obtained only a 25% increase of the feed temperature when the feed velocity was changed from 0.010 to 0.023 m·s⁻¹. Calabro *et al.* (1991) also reported a concentration factor increase when increasing feed velocity. El-Bourawi *et al.* (2006) indicated that feed velocity and feed stirring rate increase the heat transfer coefficient in the feed side and reduce the temperature and concentration polarization effects, thus resulting in higher MD permeate fluxes. Nevertheless, the effect of both feed and permeate velocity on the permeate flux was hardly studied in the academic publications.

In order to determine DCMD performance, it is important to take into account the permeate flux. In the academic publications, the membrane permeate flux (J_v) (kg·m⁻²·h⁻¹) is calculated according to Equation (1).

$$J_v = \frac{\Delta W}{(A \times \Delta t)} \quad (1)$$

where W (kg) is the weight of permeate collected over a pre-determined time t (h) of process and A (m²) is the effective membrane area.

The permeate flux varied in the range 1.512–38 kg·m⁻²·h⁻¹/5–34 L·m⁻²·h⁻¹ for a short-term operation. Of course, these results were obtained for different conditions and, therefore, cannot be comparable.

According to the academic publications, feed and permeate temperatures varied respectively from 45–90 °C and 15–35 °C. Mokhtar *et al.* (2015a) obtained a permeate flux increased by 200% on increasing the feed temperature from 50 to 90 °C. The works of Ge *et al.* (2012), Mozia *et al.* (2010, 2009b) and Calabro *et al.* (1990) confirmed likewise that the higher the feed temperature, the higher the permeate flux. Calabro *et al.* (1991) reported a concentration factor increase when increasing temperature gradient. This propensity is explained by the Antoine equation, which predicts an exponential relationship between the vapor pressure difference and temperature. The effect of permeate temperature was not studied in the academic publications. It is also necessary to evaluate the feed temperature from an economic point of view, taking into account also the energy consumption.

Long-term performance

Process stability during long-term exposure to different operational conditions is one of the important aspects that can influence the industrial implementation of any

new technology. According to Table 4, the permeate flux for a long-term operation varied in the range 0.396–15 kg·m⁻²·h⁻¹/2–30 L·m⁻²·h⁻¹.

The phenomenon of flux decay in MD has been often observed in long-term operation. The most accredited explanation considers that transmembrane flux falls down as a consequence of fouling (Curcio & Drioli 2005). It is important to consider fouling in MD, since it increases the costs of energy consumption, downtime, cleaning and membrane replacement, and deteriorates product water quality due to pore wetting (Kim *et al.* 2015; Warsinger *et al.* 2015). Dye molecules tend to adhere easily onto the membrane pores, due to chemical interaction/bonding between the aromatic rings of the dye molecule and the material membrane. Therefore, the chemical structures of the different dye classes play an important role during DCMD performance. Organic fouling is the result of the interaction between the chemical characteristics of the organic fouling materials (dye molecules) and the membranes (An *et al.* 2016). There is an influence of dye concentration on the membrane fouling. An *et al.* (2017) obtained a higher permeate flux at higher dye concentrations.

In order to reduce foulants accumulated on the membrane external surface and, consequently, membrane fouling and pore wetting, an improvement of the membrane surface properties is necessary for a long-term operation of the DCMD process (Mokhtar *et al.* 2016). During the DCMD process, the membrane must remain hydrophobic to avoid wetting, otherwise the feed solution will penetrate through the pores (Lin *et al.* 2015).

In order to improve some process parameters during DCMD operation, it is important to consider the surface tension of the liquid and of the membrane material. They are related to intermolecular forces such as dispersion forces, polar forces and hydrogen bonding and are measured through contact angles between the solution and the material.

Currently, pre-treatment and membrane cleaning are the main techniques to control fouling. Other possible solutions to reduce membrane fouling and wetting phenomena are membrane surface modification using advanced materials (Lin *et al.* 2015; Lu *et al.* 2016). Ashoor *et al.* (2016) indicated that the stability of PVDF membranes for long periods of DCMD operations, ranging from several weeks to months, could still not be ascertained from DCMD studies. Existing researches have been limited only to short-term test operating times, most likely due to fouling and wetting issues. Therefore, it is necessary for more research to address fouling and pore wetting problems, especially related to surfactants (Mokhtar *et al.* 2016).

Rejection rates

Membrane rejection, R (%), of the parameters, is calculated as shown in Equation (2).

$$R(\%) = \left[1 - \frac{C_P}{C_F} \right] \times 100 \quad (2)$$

where C_P and C_F stand respectively for permeate and feed concentration ($\text{mg}\cdot\text{L}^{-1}$).

Table 5 shows a summary of the rejection rates for color, chemical oxygen demand (COD) and total organic carbon (TOC) obtained in academic publications for the DCMD process applied to textile wastewater treatment. According to the academic publications, color rejection varied from 40 to 100%, COD rejection from 72 to 80% and TOC rejection from 80 to 100%.

Some studies demonstrated that the DCMD process results have shown better performance if compared to the other commonly used pressure-driven membrane processes (Mozia et al. 2005, 2006, 2007; Grzechulska-Damszel et al. 2010; Mokhtar et al. 2016).

CONCLUSION

The presented study showed a critical review of the state of the art of the DCMD process applied to textile wastewater treatment. DCMD issues such as membrane configuration, characteristics, modules, fouling, operating parameters and rejection rates were covered. A specialized review of this research area is paramount for further studies.

It was identified that academic publications demonstrated that DCMD performance was better than membrane process results based on pressure difference, due to better rejection rates. New insights about solution characteristics, such as different ions, salt, surfactants and pH, and module geometry and system configuration need to be more investigated, since they affect DCMD results.

This review reveals that some potential research areas pertinent to DCMD for textile wastewater treatment deserve further exploration. In particular:

- An energy and sustainability analysis. Although DCMD seemed to be technically suitable, the capital and operating costs of this technology must be reduced to facilitate its economical implementation. Capital costs may be reduced considering the optimization of permeate and feed velocities and temperatures, as well as the

Table 5 | Summary of rejection rates for color, COD and TOC obtained in academic publications for the DCMD process applied to textile wastewater treatment

Color rejection (%)	COD rejection (%)	TOC rejection (%)	Reference
N/A	N/A	N/A	Dow et al. (2017)
96.54–100.00	N/A	N/A	An et al. (2017)
72.00–89.00	72.00–80.00	N/A	Mokhtar et al. (2016)
96.54–100.00	N/A	N/A	An et al. (2016)
N/A	N/A	N/A	Lin et al. (2015)
99.92 ^a	N/A	N/A	Mokhtar et al. (2015a)
99.86	N/A	N/A	Mokhtar et al. (2015b)
99.86	N/A	N/A	Mokhtar et al. (2015c)
99.78	N/A	N/A	Mokhtar et al. (2014a)
99.98	N/A	N/A	Mokhtar et al. (2014b)
99.6–99.9 ^b	N/A	N/A	Mokhtar et al. (2013)
100.00 ^c	N/A	N/A	Huo et al. (2013)
N/A	N/A	N/A	Ge et al. (2012)
100.00 ^c	N/A	N/A	Mozia et al. (2010)
N/A	N/A	N/A	Mozia et al. (2009a)
100.00 ^c	N/A	91.90–100.00 ^c	Mozia et al. (2009b)
100.00 ^c	N/A	N/A	Grzechulska-Damszel et al. (2010)
40.00–100.00 ^c	N/A	N/A	Mozia et al. (2007)
100.00 ^c	N/A	80.00 ^c	Mozia et al. (2006)
100.00 ^c	N/A	N/A	Mozia & Morawski (2006)
100.00 ^c	N/A	80.00 ^c	Mozia et al. (2005)
100.00	N/A	N/A	Calabro et al. (1991)
100.00	N/A	N/A	Calabro et al. (1990)

^aObtained for AR1 (50 ppm) and NaCl (1.0 M).

^bObtained from the graphics.

^cResults of a combined photocatalysis–DCMD process.

N/A: not available.

membrane supply, the possible water reuse and the efficiency of pre-treatment and cleaning procedures. In this way, textile industries will obtain economic benefits, besides the environmental advantages.

- Flux decay due to fouling was reported in all academic publications; however, membrane cleaning or pre-treatment to reduce fouling impacts was not so well covered. The presence of diverse foulants, not only dyes, in the textile wastewater implies the need for other studies for a good DCMD performance. Chemical

or physical membrane cleaning associated with a pre-treatment of the textile wastewater could be a crucial item for DCMD industrial implementation. Reduced propensity for fouling translates to reduced operating costs.

- Influence of other dye classes, such as sulfur, disperse and vat dyes, on the DCMD performance. Dye classes have different compartments during the DCMD process, mainly because their chemical types have distinct interaction with the membrane material through chemical bonds.
- Most of the conducted DCMD studies are based on laboratory-scale experiments employing a synthetic solution as wastewater model. Large-scale DCMD studies covering industrial application represent an interesting area of research. A DCMD module could be installed directly near the fabric dyeing machine in order to maximize the heat recovery. A long-term industrial performance of DCMD, i.e. several weeks to months, would evaluate DCMD stability.
- Development of new membrane materials specific for MD with high hydrophobicity deserves more emphasis, since the results with superhydrophobic membrane were promising. Other commercial membrane materials, for example polyamide, deserve also to be researched, since recent studies are based only on PTFE and PVDF materials.

Although DCMD is already an established process for desalination, its application for textile effluent treatment is based on few studies. Even though the process is the same for both applications, the features of the solution change the whole study and focus in the work, being necessary to carry out specific studies for the reuse of water. Water reuse is becoming more and more important in the textile industry and, therefore, membrane separation processes, such as DCMD, have a promising applicability with several advantages.

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