Textile wastewater treatment via membrane distillation

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ABSTRACT

Due to the skyrocketed population growth and to meet clean water requirements, it is mandatory to develop techniques for proper conversion of wastewater and seawater into clean water. Textile industries generate a massive amount of wastewater. Processing of wastewater obtained from textile wastewater is more significant because of the pollutants present in the wastewater. Conventional separation processes like physical treatment, chemical treatment, and biological treatment are not pertinent in the current situation because of their lower separation efficiencies. Non-conventional separation processes are playing critical roles in ensuring the highest separation possible till date. Of these, membrane distillation (MD), which works based on trans-membrane vapor pressure difference, gives nearly 100% rejection. Thus, the present review highlights the latest work related to the separation of wastewater from the textile industries. This review also studies the limitations in terms of membrane fouling in the field of MD. Lastly, future work regarding membrane modification, MD integration, and MD commercialization are discussed. It is believed that these integrated membrane-driven separation processes will be rendered into relevant innovations in this field.

Keywords: MD integration, Membrane distillation, Membrane fouling, Textile industries, Wastewater treatment

1. Introduction

Water is the essential living source for human life and plays a vital role in the sustainable development of a country by fulfilling the needs of people. With the increase in population growth and rapid industrialization, the demand for freshwater resources is continuously increasing [1]. Since freshwater availability is significantly less, the need for reusability of wastewater came into existence [2]. One of the major sources of wastewater globally is the textile industry. Complexity in separation arises because of the wide range of pollutants obtained from various processing techniques employed in textile industries, making the separation difficult [3]. The different textile industries during the manufacturing of various fabric items require the use of several processing techniques like scouring, dyeing, sizing, de-sizing, bleaching, printing, and finishing, resulting in the generation of wastewater [4]. The dyeing process utilizes high amounts of organic dyes, additives, and salts to produce better-quality textiles that eventually lead to acute water pollution. Therefore, proper treatment of wastewater is necessary before reusing it. In addition to textile wastewater, surface water is widely contaminated because of the enormous usage of pesticides which are harmful to both biotic and abiotic environments [5, 6].

Until now, many survey papers have been written by different researchers around the world on the different types of wastewater treatment using MD, although very few survey papers specifically focused on the textile wastewater treatment using MD.

Tibi et al. [7] focussed more on membrane fabrication and surface modification for different types of wastewater treatment to enhance membrane hydrophobicity and rejection efficiency. These membrane fabrication methods include improved phase inversion technique (using alcohol as non-solvent in the place of deionized water), incorporating perfluorinated polymers, and hydrophobic polymer blending. Surface modification methods include the addition of inorganic additives (incorporation of nanomaterials), electrospinning, and chemical modification (to reduce surface energy thereby increasing hydrophobicity). Nasir et al. [8] studied the preparation of polymeric nanocomposite membranes and their applications in wastewater treatment. These membranes are eco-friendly and are energy efficient. On a lab scale, these membranes possessed improved mechanical strength and antibacterial properties which reduced biofouling and enhanced membrane stability. Pavithra et al. [9] reviewed industrial wastewater treatment using various treatment techniques like removing colorants present...
in the wastewater. These treatment techniques include advanced oxidation processes, membrane processes like nanofiltration, reverse osmosis, membrane distillation, etc. These processes exhibited more than 80–90% efficiency for the removal of dyes like azo dye, methylene blue, etc. Neoh et al. [10] and Jegatheesan et al. [11] focussed majorly on wastewater treatment using membrane bioreactor integrated with other separation processes. The results promised to show high performance in organic removal which makes this process an alternative for the reusability of water.

Shirazi and Kargari [12] investigated various types of membrane distillation and its applications. The authors emphasized more on important features like 100% rejection and stable performance even at higher concentrations of feed. Also, low permeate flux and pore wetting should be taken care of. Apart from the experimental work, Madalosso et al. [13] investigated dye wastewater using DCMD experimentally and validated with MATLAB modelling. The results showed that permeate flux obtained from mathematical model are in good agreement with experimental results with error less than 10%.

With the increase in operating time, foulants get deposited on the membrane surface and significantly impacts permeate flux and rejection. To control fouling, properties of membrane like hydrophobicity should be considered. Hydrophobicity is one of the important parameters in MD, which helps in reducing fouling by increasing the contact angle of the membrane. To increase hydrophobicity, membrane modification techniques like the addition of nanomaterials and surface modification methods are widely used. Nanomaterials like SiO$_2$ [14], TiO$_2$ [15], graphene [16], zeolites etc., are mainly used for wastewater treatment. TiO$_2$ showed great promise in separating dye wastewater from these nanomaterials due to its photocatalytic activity [17]. So, TiO$_2$ stands one step ahead in water purification techniques. Incorporation of TiO$_2$ with ZSM-5 on MoS$_2$ nanosheets [18] showed 100% rejection of arsenite present in wastewater [19]. Balati et al. [20] synthesized different crystal morphologies of black TiO$_2$ nanoparticles using pulsed laser ablation in the liquid method. The synthesized modified TiO$_2$ exhibited 99% rejection for methylene blue after 60 min of operation. The authors' work followed the pathway of developing eco-friendly modified nanomaterials with photocatalytic activity for water purification and reclamation.

The membrane-based separation processes provide a better platform for effluent treatment in textile industries because of their lower operating cost [21]. The lower level of aerobic biodegradation and the presence of dissolved salts in the effluent stream makes it unsuitable for separation by conventional treatment processes like flocculation, coagulation, and adsorption [22]. After treating effluent using membrane technology, the permeate was used to dyeing polyester fabric within the same unit [23]. Membrane distillation (MD) is one of the promising non-conventional separation processes in the present world. Membrane distillation operates based on the trans-membrane vapor pressure difference across the membrane [24–26]. Apart from wastewater separation, MD is used for desalination, azeotropic mixture separation, processing of food, etc. [27, 28]. Thus, the current survey paper focused on the MD treatment for textile wastewater treatment, membrane modification techniques and concentrate on some of the hybrid MD process used for the same purpose. Table S1 summarizes the survey papers that considered wastewater treatment using MD as the central theme. Some of them also considered the textile wastewater treatment as the sub-section. Finally, the main contributions of the current review paper are encapsulating in Fig. S1.

2. Types of Textile Effluent and Their Characterization

The textile industry is one of the major export and import industries in the world. Countries like China and the European Union (EU) have a major share in textile exports [29]. Regarding textile imports, EU and USA have a significant share among other countries [30]. As far as India is concerned, major textile industries are located in Ahmedabad, Bombay, Chennai, and Coimbatore. Medium and small-scale textile industries are situated all over the country [31]. The textile industries are classified into cellulosic fibers, protein fibers, and synthetic fibers based on the fiber used. Cellulose fibers are acquired from a plant source like cotton, rayon, hemp, and lyocell [32]. Protein fibers are taken from animals and comprise wool, silk, angora, and cashmere; and synthetic fibers are composed of polyester, polypropylene, nylon, spandex, and acetate [33].

Textile wastewater is loaded with high amounts of dyes, chemical oxygen demand (COD), biochemical oxygen demand (BOD), total dissolved solids (TDS), and heavy metals. Out of them, dyes are considered the main constituents of textile wastewater because of their high concentration in wastewater [34].

The chemical structure of dyes is also an essential parameter of classification, and based on this; dyes are classified as anionic, cationic, and non-ionic dyes. Direct, reactive, and acid dyes come under anionic dyes. Major non-ionic dyes would not get ionized in the environment. Some non-ionic dyes are polymers. Cationic dyes include azo-basic and reactive dyes [35]. Based on COD, dyes are classified [36] as: (i) If COD is greater than 1,500 ppm, then the dye comes under a high concentration level; (ii) If COD is in between 800–1,500 ppm, then the dye comes under average concentration level; and (iii) If COD is less than 800 ppm, then the dye comes under lower concentration level.

Based on the fabrics, different types of dyes are used in the textile industry. For dyeing cellulose fibers, reactive dyes, direct dyes, napthol dyes, and indigo dyes are generally used. Similarly, for dyeing protein fibers, acid, and lanaset dyes, while for dyeing synthetic fibers basic and direct dyes are recommended [33].

The main properties of textile dye effluent are color, total dissolved solids, chlorine, organic materials, toxic metals [37]. The presence of dyes results in toxic effects like skin irritation, respiratory problems on human health, and disturbing aquatic life. To overcome the trouble caused by these pollutants, we go for different treatment techniques, as mentioned in the next section [38].

The major steps involved in textile wet processing are de-sizing, scouring, bleaching, dyeing, printing, and finishing. The purpose of desizing involves removing sizing agents and effluents generated with sizing agents and effluents developed with high BOD and COD [39]. Scouring operation involves pectin and lignin removal and generates high COD [40]. Bleaching helps in improving white-
ness and generates effluents like suspended solids [41]. Dyeing adds color to the dye by using chemicals like color, metals, and surfactants [42]. Printing and finishing are the final steps in textile wet processing, enhancing durability and generating no effluents [43, 44].

3. Conventional Treatment Techniques

As discussed in section 2, textile wastewater mainly consists of dyes, salt, sizing agent, and surfactants [45]. Based on the principle involved in the separation of textile discharge, the treatment methods are categorized into physical treatment, chemical treatment, and biological treatment methods.

3.1. Physical Treatment

The most commonly used physical treatment methods are adsorption, irradiation, filtration, ion-exchange, and coagulation. As there is no chemical reaction involved, these techniques require lower operating costs than chemical treatment techniques. For adsorption, adsorbents like activated carbon, silica are used [46]. In this process, a high purity product will be obtained along with secondary waste [47, 48]. Coming to ion-exchange, it is peculiar in separating desired ions. The presence of sludge and other impurities reduces the efficiency of the ion-exchange process [49, 50].

In membrane filtration, membrane clogging and high capital cost are added disadvantages [51, 52]. Electro-kinetic coagulation is highly effective in removing small particles, but excess sludge production is undesirable [53].

3.2. Chemical Treatment

Physical methods are not satisfactory in eradicating dye as the former requires further treatment in removing waste from textile effluent. Examples of chemical treatment include flocculation-coagulation, which is a physicochemical process. This technique helps in agglomeration, which results in the formation of colloids that can be separated from the water. Coagulation is effective in degrading insoluble dyes because of the rapid formation of colloids [54]. Another important chemical treatment technique is direct chemical oxidation. In this method, sodium hypochlorite is used to remove color from dyes [46]. Advances in chemical treatment are described in later sections. Major advantages include effective separation of impurities while disadvantages include excess sludge disposal [46].

3.3. Biological Treatment

Considering the ability to produce less sludge, biological treatment is the better alternative to physical and chemical treatment. If the produced wastewater contains relatively large value of COD then biological treatment is considered satisfactory for the reduction of effluents because micro-organisms employed in biological treatment have the potential to reduce COD to the lowest value [46]. Though biological treatment is not effective in decolorization, anaerobic followed by aerobic arrangement is most suitable for decolorization. Some examples include the activated sludge process, which removes biodegradable waste stabilization ponds, anaerobic reactors. Sludge disposal is a significant problem because additional cost is incurred for its removal [55].

4. Advance Treatment Techniques

Conventional treatment techniques areoutmoded because of their lower separation efficiency. Moreover, dye molecules’ biological resistance and chemical stability make it difficult to separate in traditional separation processes [56]. To increase efficiency and overcome the drawbacks of conventional treatment techniques as mentioned in the earlier section, advanced treatment techniques are used. This section of review primarily focuses on advanced oxidation processes and membrane separation processes [57–59].

4.1. Advanced Oxidation Processes (AOPs)

The principal goal of any AOPs is to produce and utilize hydroxyl free radical (HO). The generated free radical is a strong oxidant that is used to break compounds that conventional processes cannot oxidize. Based on this principle of generating hydroxyl free radicals, different methods have evolved to degrade effluents present in textile wastewater [36, 60]. The major applications of AOPs include textile wastewater treatment [36, 57, 59]. The advantages and disadvantages of different AOPs are discussed in Table S2.

4.1.1. Ozonation

Ozone is one of the most powerful oxidizing agents for the degradation of wastewater. On dissolution in water, ozone reacts with organic compounds present in wastewater by direct oxidation as O3 or by forming a hydroxyl free radical. In textile wastewater, ozone is helpful in degrading phenols and removing COD [60, 61]. The main limitation of the ozonation process is its shorter half-life period that leads to the conversion of unstable ozone to oxygen [36].

4.1.2. Electrochemical process

This process is highly effective in removing pollutants from textile wastewater by direct or indirect oxidation. For dyestuff containing electrodes like mercury, iron, and boron-doped diamond electrode, this method is used widely. This method requires small amounts of chemicals for the treatment, hence considered very economical [36, 60, 61].

4.1.3. Ozone/H2O2

The addition of H2O2 enhances the decomposition of O3 and the generation of hydroxyl free radicals. At acidic pH, the reaction of H2O2 is significantly less with O3 because of the inadequate generation of hydroxyl free radicals. More efficiency in the generation of hydroxyl free radicals is observed at basic pH, i.e., the maximum amount of H2O2 has converted to hydroxyl free radicals, which help in faster degradation of dye molecules [36, 60, 62].

4.1.4. Fenton process

The reaction between Fe2+ and H2O2 is termed Fenton’s reaction. Due to the coagulation and oxidation properties of the Fenton
reagent, degradation of dyestuff is done using the Fenton process. The advantage of the Fenton process is that no energy is required for activating H2O2 conversion into hydroxyl free radicals, which in turn helps in the degradation of dyestuff [36, 54].

4.1.5. Ozone/ H2O2/UV

UV process is helpful in initiating oxidizing agents like H2O2 and also in the degradation of dyestuff. The main advantage of this process is that no slug formation after treatment and complete decolorization of dye [60].

4.1.6. Photo-Fenton process

The Photo-Fenton process (H2O2/UV/Fe2+) is one of the advanced oxidation processes which involves H2O2 photolysis and Fenton reaction utilizing which hydroxyl free radical formation is highly increased. With the added advantage, when compared to the Fenton process, the mineralization process is highly enhanced. Efficiency in terms of decolorization is the same in both Fenton and photo-Fenton processes [54].

4.2. Membrane Separation Processes

The AOPs require high capital and high operating cost and also need further successive processes to put out residual oxidants present in it. The drawback mentioned above leads to the research for textile water treatment towards the membrane separation processes (MSP). Due to the no additional chemical requirement and no downstream processing, the MSP is generally considered a clean process. MSP produces good quality water and also helps in the removal of low molecular weight effluents in wastewater. MSP like microfiltration (MF) [63-65], ultrafiltration (UF) [66-68], membrane bioreactor (MBR) [69-71], nanofiltration (NF) [73, 74], reverse osmosis (RO) [23, 74, 75], and membrane distillation (MD) [76-78] are used to treat wastewater. Integrating these processes with each other helps in higher quality and better reusability of wastewater [69]. MF and UF are outdated technologies and have lower separation efficiencies in the field of textile wastewater treatment. These MSPs are integrated with NF/MD for good results [67, 79]. Except for MD, all the MSP are pressure-driven separation processes that require relatively high pressure. As dye molecule size decreases, more pressure is applied to get permeate flux. NF gives 60–70% separation efficiency in pressure-driven separation processes while others have lower efficiencies [80-83]. To counteract these disadvantages like low efficiency and high pressure, MD has emerged as one of the promising membrane-based separation technologies in wastewater treatment [84]. Detailed applications, advantages, and disadvantages of MSPs are mentioned in Table 1.

5. Membrane Distillation

Membrane distillation can be considered an optimal solution for wastewater treatment because of its low operating pressure and low fouling tendency [87]. The conversion to potable water from contaminated water using a hydrophobic membrane at the earliest has been ascribed to Bodell [88].

MD is a thermally-driven separation process in which the feed
section is separated from the permeate section by the hydrophobic porous membrane [75]. In MD, both heat and mass transfer co-occur across the hydrophobic membrane by the phase change process [89]. The output in the permeate section is condensed by various methods like passing cold fluid, applying a vacuum, passing air followed by cold fluid, or by flowing an inert gas through the permeate side and finally gets collected. Basic MD configurations are classified into four, viz., direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), vacuum membrane distillation (VMD) and sweep gas membrane distillation (SGMD) [90].

The major applications of the MD include desalination [91-96], wastewater treatment like textile wastewater, pharmaceutical wastewater [97-100], concentration of azeotropic mixtures [101] and the concentration of aqueous solutions [86, 102].

5.1. Direct Contact Membrane Distillation

The DCMD is shown in Fig. 1(a) [24]. The feed section and permeate section are separated from each other by the hydrophobic membrane. The feed gets converted to vapor across the membrane. Because of the vapor pressure difference, the vapor passes through the membrane. The vapor is condensed using a low-temperature solution, preferably an aqueous solution. Because of its simplicity and high flux rate, enormous research has been done on this configuration. More conductive heat loss and difficulty of detecting wetting in the membrane are the main drawbacks of DCMD. DCMD is mainly used in desalination, wastewater treatment, the concentration of aqueous solutions, etc. [89, 103-108]. DCMD is combined with other separation technologies like NF and RO for better treatment of wastewater [3].

5.2. Air Gap Membrane Distillation

To limit the excessive heat loss due to membrane conduction and also to save cost for the extra condenser in SGMD, AGMD was introduced [86]. AGMD is shown in Fig. 1(b). The hot feed will contact the membrane, and the vapor formed will pass through the air gap before condensing on the cold fluid. As a result of this, heat loss and temperature polarization will be reduced. The air gap results in extra mass transfer resistance, which in turn results in lower flux than DCMD. The resistance developed is directly proportional to the width of the air gap. Any leakage or wetting of the membrane can be detected easily because of the air gap [24]. Primary applications of AGMD include desalination integrated with a renewable energy source, separation of azeotropic mixtures [109], and removal of volatile organic compounds (VOCs) from wastewater obtained from textile industries [94, 110-113].

5.3. Vacuum Membrane Distillation

As shown in Fig. 1(c), in VMD, vacuum is applied on the permeate side, and permeate collected outside with the help of a condenser. The principal condition for vapor suction is that the saturation pressure of molecules should be more than applied vacuum pressure. Equipment cost is hiked because of a vacuum pump and extra condenser. Because of the applied vacuum, heat loss due to conduction is minimized. Hence, the thermal efficiency of the membrane is hiked. Major applications of VMD include desalination, the concentration of aqueous solutions [86, 114, 115].

5.4. Sweep gas Membrane Distillation

The SGMD, shown in Fig. 1(d), is a collaboration of DCMD and AGMD as it offers low mass transfer resistance property of DCMD and low heat loss due to conduction as in the case of AGMD. Primary applications of SGMD include the removal of VOCs from wastewater, concentration of aqueous solutions, concentration of dilute glycerol wastewater [24, 27, 108].

Fig. 1. Schematic of membrane distillation configurations patterned with a hydrophobic membrane. (a) Direct contact membrane distillation, (b) Air gap membrane distillation, (c) Vacuum membrane distillation, (d) Sweep gas membrane distillation.
6. Water Recovery and Wastewater Treatment Using the MD Process

In textile industries, the recovery of wastewater plays a vital role in plant economics. The literature shows that MD gives maximum color rejection and maximum possible permeate flux [70, 116]. Fig. 2 shows the list of papers published in textile wastewater treatment with their respective year of publication. The increasing trend of the number of publications from the year 2001 to 2021 shows the continuous research work in the field of clothing industries discharged wastewater treatment using MD. In textile industries, the wastewater coming out from dyeing operation comes out at a higher temperature sufficient to enter the MD module [3, 50, 111, 117]. The MD schematic is shown in Fig. 3.

Li et al. [97] studied the treatment of industrial dyeing wastewater and characteristic pollutants like phenol, aniline, sulfanilic acid, 3,4-dihydroxybenzoic acid, and p-chloroaniline by DCMD. This study also measured flux and rejection performance for PTFE and PVDF membranes. The PTFE membrane showed high flux and high rejection performance. The study also stated that the accumulation of suspended particles could be responsible for fouling and membrane wetting. The study states the DCMD process as one of the promising technologies for the treatment of dyeing wastewater. Ramlow et al. [118] studied the performance of VMD incorporated with PVDF membrane and thermopervaporation (TPV) combined with polyamide (PA) membrane for intensification of water reclamation from textile dyeing wastewater having reactive black (RB), and disperse black (DB) as the dye solutions. The study stated that the permeate flux increased in the case of VMD whereas TPV gave 100% dye rejection for RB and DB dye solutions. This study suggested that both processes might be integrated and placed next to the dyeing machine to overcome difficulties.

Leaper et al. [119] investigated textile wastewater treatment in AGMD for dyes like sunset yellow (SY), rose bengal (RB), and surfactants like sodium dodecyl sulfate (SDS) by using commercial polyvinylidene fluoride (PVDF) membrane. This study also has compared DCMD with AGMD. For the operating time of 8 h, the flux obtained in DCMD (12.8 L/m².h) was more than AGMD (11 L/m².h). A 20 hour-long operating time yields complete rejection of dyes and surfactants.

Silva et al. [120] studied the evaluation of steady-state conditions for different operating times of the dyeing process obtained from textile wastewater in DCMD. A flat sheet membrane made up of PTFE was characterized as per the work done by Li et al. [97]. The operating times employed in this study are 3, 12, and 24 h with different dye solutions like RB and DB. The steady-state evaluation is significant because the dyeing process is a batch process. RB dyes showed a decrease of permeate flux while DB dyes showed stable permeate flux due to differences in size and ionic character. The flux variation can be classified into sub-steady, pre-steady, and steady states. As far as steady-state is concerned, the flux drops slightly with time. In the pre-steady state, the flux begins to decline until a steady-state is reached. During all experiments with RB and DB dye solutions, a high color rejection was seen, implying the reusability of recovered water. The wastewater discharged from DCMD is at 80–90°C. The energy obtained from this can be used to heat feed the solution.

From these studies, it is evident that the performance of MD in textile wastewater treatment depends not only on membrane properties but also on the operating parameters employed. The main operating parameters include feed temperature, feed flow rate, permeate temperature, and concentration of textile effluents present in the feed. Table S3 represents the effect of operating parameters on MD performance. From the Table, it is observed that the optimum conditions are high feed temperature, high feed flow rate, low concentration of textile effluents, and low permeate temperature.

Table 2 illustrates various studies done in the field of textile wastewater treatment using MD. These studies quoted that 100% color rejection of dye is possible, and permeate flux obtained was less. Recovery of wastewater is high and can be further increased by using hybrid membrane separation processes.
| MD Type | Membrane Type | Feed and Permeate Temperature (°C) | Feed Type | Major Findings | Permeate Flux (kgm⁻²h⁻¹) | Colour Rejection (%) | Ref. |
|---------|---------------|----------------------------------|-----------|----------------|--------------------------|---------------------|------|
| DCMD    | PTFE and PP   | 80–100 and 20                    | Dye wastewater (synthetic) | a. Polypropylene membrane had higher performance because of higher porosity than PTFE. b. To enhance water recovery and improve energy efficiency, MD can be integrated with other processes like RO and FO. | 60.1               | -                   | [110] |
| DCMD    | PVDF          | 70 and 20                        | Dye wastewater | a. For S7 dye the lowest flux reduction factor was observed. b. The highest flux reduction factor for DY solution was observed. | 9.8 (model)        | 99.9                 | [121] |
| DCMD    | PTFE          | 60 and 45                        | Textile wastewater | a. Salt rejection efficiency is 99% | 20                  | 100                  | [122] |
| DCMD    | PAN-PS        | 60 and 20                        | Textile wastewater | a. The PAN-PS membrane possesses high superhydrophobic properties is because of surface hierarchical roughness, high void volume fraction, and mean flow pore size | 60.1               | -                   | [123] |
| DCMD    | PVDF with cloisite 15A nanocomposite | 90 and 25 | Industrial wastewater | a. This study successfully reduced 89% of source wastewater to quality water standards. b. There is a reduction in flux because of membrane fouling. | 18.8               | 100                  | [99] |
| DCMD    | PTFE and PVDF | 60 and 20                        | Textile wastewater | a. The flux rate is reduced by caustic cleaning to 70% of the initial flux. b. With integration with DCMD, zero liquid discharge is seemed possible. | 20                  | 100                  | [124] |
| DCMD    | PVDF combined with ethylene glycol | 80-90 and 25 | Dye wastewater | a. PVDF modified with ethylene glycol can be considered as a promising membrane for textile wastewater. b. Because of membrane design, mass transfer resistance can be reduced during vapor transport. | 9.82               | 99.75                | [125] |
| SPMDR   | Polypropylene | 65 and N/A                       | Dye wastewater (RB5) | a. TOC removal efficiency is 80.1%. b. Complete color rejection is attained. | 4.56               | 100                  | [126] |
| VMD     | Polypropylene | 60 and 20                        | Dye wastewater | a. High purity of water can be obtained. | 57                  | > 90                  | [127] |
| VMD     | Polypropylene | 70 and N/A                       | Dye wastewater (Methylene blue) | a. Feed temperature is an important parameter which in turn is used for the calculation of permeate flux. | 6.3                | 100                  | [22] |
| PMR     | Polypropylene (commercial) | 70 and 20 | Dye wastewater (Acidic dye) | a. 100% color rejection is possible | 16.7               | 100                  | [79] |
Hybrid Separation Processes Based on MD in Wastewater Treatment

Membrane distillation offers many advantages when compared with conventional separation processes. Despite having many benefits, the process has several limitations when used individually. Hence, the need for membrane-based hybrid separation processes came into existence. These membrane-based hybrid separation processes overcome the limitations of MD like performance at high concentrations of wastewater to the maximum extent and enhance the water recovery [90, 128-130]. Membrane-based hybrid separation processes are classified into two categories [129], viz., (i) Membrane process integrated with conventional separation process (MCH), which is generally applied to minimize capital costs and (ii) Membrane process integrated with other membrane separation process (MMH), which is used to overcome the limitations occurring in the membrane separation process.

Some of the MD-based hybrid separation processes are forward osmosis-membrane distillation (FO-MD), membrane distillation-crystallization (MD-C), micellar enhanced ultrafiltration-membrane distillation (MEUF-MD), and photocatalysis-membrane reactor (PMR). Ge et al. [131] studied polyelectrolyte-promoted hybrid FO-MD for dye wastewater treatment to enhance water permeate flux and increase water recovery. This hybrid combination has greater potential than the MD process alone. Table 3 shows various studies reported by different researchers in the field of hybrid separation processes. These studies show that hybrid separation processes have greater potential for wastewater treatment and stand as a promising technology in wastewater treatment.

8. MD Membrane Fouling

Fouling is a phenomenon that happens because of the accumulation of unwanted materials on the membrane surface. Fouling causes a reduction in permeate flux and irregular performance of separation. In textile wastewater treatment, fouling occurs because of dyes, effluents, and surfactants present in the wastewater. They block the pores of the membrane resulting in a decline of flux [3, 122]. Common types of fouling that happen in the membrane systems are inorganic fouling (most commonly known as scaling), organic fouling, and biological fouling (biofouling) [86, 93, 103, 121, 137-139]. Inorganic fouling is prominently caused by alkaline salts, particulate matter, and other uncharged molecules. Organic fouling is referred to as natural organic matter. Organic fouling is caused by proteins, polysaccharides, organic acids, humic substances, etc. Both organic and inorganic fouling increases with the increase in both feed temperature and pressure drop. The fidelity of micro-organisms and the formation of the biofilm layer are generally referred to as biofouling. Bio-fouling helps in the reduction of permeate flux to some extent [24, 139, 140].
| Feed Solution | Membrane Commercial/ Fabricated | Manufacturer | Membrane Properties | Fouling type | Fouling Remedial Method and Observations | Ref. |
|---------------|---------------------------------|--------------|---------------------|--------------|-----------------------------------------|------|
| Dye wastewater | High impact polystyrene         | Tabriz Petrochemical company, Iran | Mean pore size: 0.56 μm Water contact angle: 123.4° | Organic | Rejection efficiency > 99.8% Fouling can be reduced by increasing hydrophobicity of the membrane | [143] |
| Industrial Dyeing wastewater | PTFE                            | Shanghai Mingjie membrane Co., Ltd | Mean pore size: 0.22 μm Water contact angle: 133.7° | Organic | Excellent treatment efficiency Improvement in membrane properties are required to reduce fouling. | [97] |
| Seawater (Synthetic) | Modified PVDF (SiO₂-PfTS/PVDF and PVA/PVDF) | MILLIPORE® | Water contact angle of SiO₂-PfTS/PVDF: 167.3° Pore size: 0.45 μm | Organic (HA, SDBS, and kerosene) | Surface modification using superhydrophilic and superhydrophobic membranes gives the best result. | [144] |
| Dyeing wastewater | Commercial PVDF | MILLIPORE® | Mean pore size: 0.45 μm Porosity: 72.11 Thickness: 105 μm | Organic | Electrospun membranes are more promising than other fabricated membranes in wastewater treatment In long term operation, complete color removal is possible | [145] |
| | E-PH (Electrospinning) | E-PH | Mean pore size: 0.52 μm Porosity: 87.28 Thickness: 98 μm | Organic | Electrospun membranes are more promising than other fabricated membranes in wastewater treatment In long term operation, complete color removal is possible | [145] |
| | E-PDMS (PDMS on E-PH) | E-PDMS | Mean pore size: 0.49 μm Porosity: 87.84 Thickness: 102 μm | Organic | Electrospun membranes are more promising than other fabricated membranes in wastewater treatment In long term operation, complete color removal is possible | [145] |
| Dyeing wastewater (Methylene blue) | Polyetherimide-PDMS | General Electric Co, | Mean pore size: 0.72 μm Porosity: 81 Water contact angle: 103.8 ± 0.26° | Organic | 100 % rejection Surface modification with PDMS helped in reducing fouling to almost nil. | [146] |
| DI water | PTFE | Sartorius 11807-640-320PR | Nominal pore diameter 0.20 μm Water contact angle: 123° | Organic (HA) | Not only nominal pore diameter but also pore size is equally important Capillary action will draw liquid water through the fouling layer more quickly than vaporized water in MD. | [147] |
| | PVDF | Durapore GVHP | Nominal pore diameter 0.22 μm Water contact angle: 111° | Organic (HA) | Not only nominal pore diameter but also pore size is equally important Capillary action will draw liquid water through the fouling layer more quickly than vaporized water in MD. | [147] |
| Methyl orange-Aqueous solution | PTFE | - | Thickness: 140 μm Porosity: 80% | Organic | PMR showed greater resistance to membrane fouling than other membrane separation processes | [148] |
| Wastewater (Synthetic) | MDBR-PVDF (Flat sheet) | MILLIPORE® | Mean pore size: 0.22 μm | Biological | MDBR serves as a better alternative to MD. Fouling is comparatively less in MDBR than MD. | [149] |
| HA-deionized water | PTFE (Flat sheet) | MILLIPORE® | Porosity: 75% Thickness: 125 μm | Humic Acid (HA) | Since HA fouling is physical, control of fouling is easy | [140] |

PVDF: polyvinylidene fluoride; PTFE: polytetrafluoroethylene; PP: polypropylene; ?AN-PDMS: polyacrylonitrile-polymethylsiloxane; MDBR: membrane distillation bio-reactor; HA: humic acid.
8.1. Effect of Driving Force on Membrane Fouling

The formation of a fouling layer on the membrane surface results in a decrease of temperature difference across the membrane. As a result, temperature polarization increases. As a result, the driving force decreases, and permeate flux obtained will be lower than the pure membrane. Fig. S2 represents fouling areas on the membrane. Surface fouling refers to fouling present on the membrane surface, which is reversible and can be cleaned by chemical cleaning. Chemical cleaning refers to the addition of cleansing agents like EDTA to weaken the bond between foulants and membrane surfaces. This significantly helps in removing surface fouling. Continuous chemical cleaning can damage membrane morphology and membrane stability by altering physical and chemical properties of membrane thereby reducing rejection. To minimize membrane cleaning and enhance membrane stability, polymeric membranes should be doped with self-cleaning activity materials. Among these self-cleaning materials, materials having photocatalytic activity are widely used since they can neutralize fouling by using a photocatalytic property when exposed to sunlight [141]. Internal fouling refers to the presence of foulants inside the membrane pores. These block the pathway of vapor molecules across the membrane and lead to permanent damage of the membrane. To minimize this, surface modification techniques are widely employed and are discussed in the next section.

Fortunato et al. [142] investigated synthetic dye wastewater (congo red) on membrane performance and fouling using DCMD. The results exhibited 100% rejection at all experimental conditions. The author stated that the thickness of the fouling layer increased along the membrane length because of the variation of driving force over the membrane length. Some other literature in the field of membrane fouling is shown in Table 4. These observations quote that membrane fouling is a drawback of MD. The next section focuses on the minimization of fouling by using surface modification techniques.

9. Membrane Fabrication and Modification

The hottest area of research in MD is membrane fabrication and its modification. The available commercial membranes lack in terms of performance, fouling resistance, and wetting resistance. To overcome these shortcomings, the need for membrane fabrication and modification is necessary. Many studies have been reported on membrane morphology, geometry, pore size, and thickness of the membrane [102]. Hydrophobic membranes are chosen due to their easiness of membrane fabrication and modification. Fabrication methods employed to fabricate are nonsolvent induced phase separation (NIPS), thermally induced phase separation (TIPS), sintering, electro-spinning, and melt extrusion spinning [95, 100, 150-155]. Surface modification methods include grafting (chemical grafting, plasma grafting, photo grafting, and thermal grafting) and surface coating [102].

The mainly used commercial membranes in MD are PVDF, PTFE, and PP. The PTFE, being a non-polar polymer, cannot be fabricated by NIPS and TIPS methods. Hence, the methods used for fabricating PTFE are sintering and melt extrusion methods [156, 157].
PP can be fabricated from TIPS and melt extrusion methods because of its high elastic properties [158, 159]. The PVDF membrane can be fabricated from both TIPS and NIPS or a combination of both because of its solubility in dimethylformamide (DMF) and dimethylacetamide (DMAC) [160].

Camacho et al. [161] fabricated different hydrophobic membranes like PTFE, PP, and PVDF based on different fabrication methods with varying sizes of pore. The PTFE membranes are prepared using a sintering technique with porosity ranging from 10% to 40% and pore size of range 0.2 to 20 μm. The PP and PTFE membranes are also fabricated from stretching technology with porosity around 90% and pore size ranging from 0.2 to 20 μm. The PVDF membranes are manufactured by using the phase inversion technique. The membrane prepared has a porosity of about 80% and pore size ranging from 0.2 to 20 μm.

Hendren et al. [162] studied surface modification of nanomaterials for DCMD. The alumina anodic membranes are modified and turned into hydrophobic using perfluorodecytrihydroxysilane (FPS) and trichloromethylsilane (TCS) by using the grafting technique. The studies stated that FPS treated membrane showed a more steady-state flux than membrane treated with TCS. Wang et al. [163] fabricated a composite membrane for MD to resist oil fouling. The authors modified a commercial hydrophobic PVDF membrane with nanocomposite materials like silica nanoparticles, chitosan hydrogel, and fluoro-polymer as shown in Fig. S3. The figure shows that the modified membrane displayed different surface morphology compared to commercial membrane due to the presence of nanocomposite coating. Also, the modified membrane exhibited asymmetric wettability with a modified surface resistant to oil fouling, and the other unmodified surface remained hydrophobic. The modified membrane is compared with the new PVDF membrane, which stated that the composite membrane exhibited better resistance towards oil resistance. Liao et al. [164] fabricated electro-spun superhydrophobic dual-layer membranes for MD. These membranes exhibited superhydrophobicity and showed resistance towards emulsions and salting water. These membranes showed high mechanical strength and are durable in MD operations. Table 5 shows different membranes along with their mode of modification. The observations from this section show that membrane porosity is increased, and membrane wetting is decreased because of membrane modification.

10. Conclusions

Membrane distillation has been used for the purification of different types of wastewater like oily wastewater, dye mixture containing wastewater, salt included wastewater, etc. However, MD was found very successful in textile wastewater treatment, but the membrane fouling and wetting encountered during the operation are major concerns. Therefore, the development of fouling resisting membrane is a crucial point in MD’s future research direction. Thus, the present review highlights the different aspects of textile wastewater treatment using MD starting from textile effluents to the recent advancement of membrane modification. Other types of conventional treatment techniques and their merits and demerits are also discussed. Along with the types of MD configuration, the membrane fouling study has also been explored. The present review also focuses on the recent advancements in membrane modification and fabrication technology which helped in significantly reducing membrane fouling and membrane wetting, making membrane withstand for an extended period.

Nowadays, the membrane distillation process is gaining wide attention in wastewater treatment for high water recovery. The thermal energy requirement for heating feed and lower permeate flux makes it difficult for long-term operations in industries. To overcome these situations, MD is combined with other membrane separation processes to treat wastewater. As discussed in the current review, these hybrid-based membrane separation processes help improve permeate flux and enhance water recovery, making 100% separation possible.

Author Contributions

A.S.R. (Master Student) has done the scientific literature review and written the manuscript along with Figures and Tables. S.K. (Assistant Professor) has revised and corrected the manuscript. Z.V.P.M (Professor) edited and added some important parts to the final manuscript.

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