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Review

Water and Wastewater Treatment Systems by Novel Integrated Membrane Distillation (MD)

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Abstract: The scarcity of freshwater has been recognized as one of the main challenges people must overcome in the 21st century. The adoption of an environmentally friendly, cost-effective, and energy-efficient membrane distillation (MD) process can mitigate the pollution caused by industrial and domestic wastes. MD is a thermally driven process based on vapor–liquid equilibrium, in which the separation process takes place throughout a microporous hydrophobic membrane. The present paper offers a comprehensive review of the state-of-the-art MD technology covering the MD applications in wastewater treatment. In addition, the important and sophisticated recent advances in MD technology from the perspectives of membrane characteristics and preparation, membrane configurations, membrane wetting, fouling, and renewable heat sources have been presented and discussed.

Keywords: membrane distillation; wastewater treatment; membrane configuration; fouling renewable heat sources

1. Introduction

Freshwater scarcity and the excessive consumption of water have been regarded as serious challenges over past decades. Several contributing factors such as an increasing population, improving living standards, agricultural sector growth, and industrialization have threatened a further reduction in the water level and given rise to this crisis [1]. Based on the type of industry, a vast amount of wastewater containing salinity and organic compounds such as arsenic, fluoride, cadmium, chromium, mercury, manganese, lead, etc., have been produced. Discharging these contaminant elements above their effluent standard has exerted catastrophic effects on aquatic and terrestrial habitats and human health [2]. To address this issue, several treatment technologies have been investigated by scientists such as reverse osmosis, disinfection, granular filtration, gravity separation, coagulation-flocculation, air stripping and aeration ion exchange, adsorption, and membrane filtration [3]. Among all the conventional techniques under study, the membrane process has become highly popular due to the potential benefits associated with the technology.

Generally, in a membrane, some particular substances are selectively allowed to pass through while others are retained (retentate phase) [4]. The permeating compounds pass through the membrane based on a driving force such as a pressure gradient, concentration gradient, temperature gradient, or electrical gradient [5,6]. This phenomenon emerges from membrane module characterization (pore size, pore shape), membrane surface characteristics (porosity, charge/hydrophobicity) and membrane configuration (geometry, dimensions) [7]. To put it another way, membrane separation processes applied in wastewater treatment are categorized as the isothermal and non-isothermal process. The former includes concentration-driven membrane processes (pervaporation and membrane extraction), pressure-driven membrane processes (microfiltration, ultrafiltration, nanofiltration, and reverse osmosis) and electrically driven membrane processes (electrodialysis, electrophoresis) while the latter is a thermally driven membrane process named membrane distillation (MD) [8–10]. According to the literature, among all the membrane processes, MD has been perfectly able to treat water with an extremely high level of salinity [11,12] and hazardous contaminants [13]. In this context, extensive research has been conducted by scientists and researchers all over the world over recent years. Drioli et al. [14] investigated the current and prospective role of membrane engineering in attaining the objectives of a process intensification strategy to improve the efficiency and sustainability of novel membrane processes including MD.

The present study assesses the evolution of membrane distillation in wastewater treatments. The work investigates the characteristics, material, module, and different configurations of MD applied in water treatment as well as covering the fouling and wetting phenomena. Furthermore, the benefits and limitations of MD processes, economic analysis, and future research directions of interest have been pointed out. While various review papers focusing on MD technology have been conducted by researchers, most of them provide a full membrane perspective, without being highly focused on the novel MD membrane designs and process configurations. In this critical review, the authors aim to review the recent advances in MD technology in terms of low-grade or renewable heat sources, such as waste heat from industrial processes which reduce transmembrane heat loss and increase the proportion of heat recovered from the permeate stream. Moreover, nontraditional anti-fouling processes and recently developed membranes prepared from surface modifications of polymers and nanomaterials such as plasma surface modification and electrospinning are investigated thoroughly.

2. Membrane Distillation

2.1. History of MD Process

Membrane distillation was patented in 1963 [15] and Findley published the first MD paper in 1967 in the "Industrial & Engineering Chemistry Process Design Development" journal [16]. Nonetheless, the MD process did not attract considerable interest until the early 1980s when more efficient module membranes such as Gore-Tex became accessible [17]. The term MD originates from the significant similarity between the MD method and typical distillation process since both of them operate based on the liquid/vapor equilibrium. Moreover, in both processes feed stream is heated by the energy source to obtain the necessary potential heat of vaporization [18]. Being a practical and effective wastewater treatment, MD has been the topic of worldwide investigation by many researchers and scientists. Besides, the interest in MD processes has rapidly grown over the recent years. Table 1 gives up-to-date and interesting information concerning the application of MD in wastewater treatment based on several patents published from 2016 to 2018.

Table 1. List of published patents in the application of membrane distillation (MD) in wastewater treatment (2016 to 2018).

Patent	Highlights
Desulfurization waste water zero-discharging treatment technology for coal-fired power plants Publication number: CN105712557A Publication date: 2016-06-29	The present invention relates to a zero-discharging treatment device for desulfurization wastewater that consists of a nanofiltration system, calcium removal sedimentation pool set, a heavy metal and magnesium removal pool set, an evaporating crystallizer, and a membrane distillation system. Crystal salt gained from evaporating crystallization can be entirely recycled, and treatment and operating cost are extremely reduced.
A membrane distillation system that is used for concentration of desulfurization waste water Publication number: CN204981458U Publication date: 2016-01-20	The utility model reveals the MD system that is applied for the handling of the concentration of desulfurized wastewater, with this considered to have some benefits such as a simple structure, safe operation, and a smaller area.
Salt-containing wastewater treatment system Publication number: CN205133326U Publication date: 2016-04-06	The present utility model includes a salt-containing wastewater treatment system that contains an electrodialysis device for receiving and handling salted wastewater, which is connected to the MD system.
Process and system for produced water treatment Publication number: US20170096356A1 Publication date: 2017-04-06	This invention relates to a system and process for produced water treatment. A heat exchanger, a vacuum tank, an adsorption-desorption, and MD crystallization process are in this process combination. The efficiency related to costs of maintenance and energy consumption is considered.
Modularly installed energy-saving membrane distillation wastewater treatment device and method Publication number: CN106865663A Publication date: 2017-06-20	The present invention represents a modularly installed energy-saving MD wastewater treatment device which consists of a post-treatment and primary treatment module. The proposed device assembles easily and has suitable energy-saving effects for the treatment of different types of sewage.
Energy -saving membrane distillation effluent treatment plant of modularization installation Publication number: CN206635064U Publication date: 2017-11-14	The utility model relates to an energy-saving MD sewage treatment plant of modularization installation and consists of post-processing and primary treatment modules. The applied used equipment in the device is energy conserving, convenient, and effective for all types of effluent treatment.
High -concentration organic wastewater treatment system Publication number: CN206318843U Publication date: 2017-07-11	The present utility model reveals a highly concentrated organic wastewater treatment system that consists of a liquid bath of raw material, anaerobic biological treatment device, and a positive osmotic membrane. Highly concentrated organic wastewater from a bioreactor is handled in the combined MD system. This system proposes greater recovery of the pure water rate and reduces the concentration of organic matters.
Multi-stage submerged membrane distillation water treatment apparatus and a resource recovery method Publication number: US20170313610A1 Publication date: 2017-11-02	This investigation related to a submerging multi-stage membrane distillation water treatment device.

Table 1. *Cont.*

Wastewater treatment system Publication number: CN207243660U Publication date: 2018-04-17	This utility model shows a wastewater treatment system that consists of an MD unit connected to magnetism loaded flocculation unit. This proposed wastewater treatment system has some benefits like simple equipment, efficient sewage treatment, and a lower energy consumption.
Concentrated decrement device of vacuum membrane distillation wastewater that frequently flows backwards Publication number: CN207734625U Publication date: 2018-08-17	This invention concerns a zero-discharging wastewater treatment device, which is particularly related to a vacuum membrane distillation (VDM). MD has some effective benefits, for instance, it enhances the service life, and shows great abilities to treat wastewater with high salinity.
Slot-type solar sea water desalination device based on membrane distillation Publication number: CN107720863A Publication date: 2018-02-23	The invention reveals a slot-type solar seawater desalination device integrated with membrane distillation. The required heat energy of the process is supplied from solar energy emitted by a slot-type condenser mirror, in which solar energy is reflected and condensed onto an arc heat collection tube.
Porous membrane for membrane distillation, and method for operating membrane distillation module Publication number: WO2018174279A1 Publication date: 2018-09-27	A membrane distillation device, with a hydrophobic porous hollow fiber membrane, and a condenser for condensing water vapor is invented for water treatment. The membrane has an average pore diameter of 0.01–1 μm.
Hollow fiber membrane module for direct contact membrane distillation-based desalinization Publication number: WO2018195534A1 Publication date: 2018-10-25	The invention is a desalination system by direct contact membrane distillation integrated with a cylindrical cross-flow module comprising high-flux composite hydrophobic hollow fiber membranes. A model is developed and directed to the system and shows the observed water vapor production rates for various feed brine temperatures at different feed brine flow rates.
A membrane distillation technique and method for treating radioactive waste water systems Publication number: CN108597636A Publication date: 2018-09-28	The invention shows a seed film distillation procedure and technology for processing radioactive waste including (pretreatment, preheating, membrane separation, condensation process) by accumulating the wastewater
Multistage immersion type membrane distillation water treatment apparatus and a resource recovery method using the same number of oil resources Publication number: KR101870350B1 Publication date: 2018-06-22	This invention provides a multistage immersion-type membrane distillation water treatment system and a viable resource recovery technique applying the same number of oil resources which can substantially reduce the heat energy.

2.2. Definition of MD

MD is a thermally driven treatment process in which the thermal gradient is generated across a microporous hydrophobic membrane [19]. Simultaneously, the process can be operated by low-grade heat and/or waste including solar energy [20], geothermal energy [21], wind, tidal, and nuclear energy, or low-temperature industrial streams [22]. It should be noted that the process is driven by the vapor pressure difference between the permeable hydrophobic membrane pores. In other words, volatile vapor molecules are allowed to pass through the MD while non-volatile compounds are retained on the retentate stream. The permeated volatile vapors are then collected or condensed by various techniques. Finally, completely pure products that are theoretically 100% free from solid, harmful substances and non-volatile contaminants are produced [23]. Figure 1 illustrates a schematic diagram of the MD process.

As the graph reveals, volatile vapor molecules in the hot feed which are vaporized at the liquid/vapor interface are able to pass through the pores of the membrane. The liquid feed, on the other hand, is prevented from transporting through the membrane pores. This phenomenon is mainly because of the hydrophobic nature of the MD membrane and its surface tension. Therefore, it is important to note that the dry pores must not be wetted by the liquid feed which is directly in contact with the hydrophobic membrane [24–26].

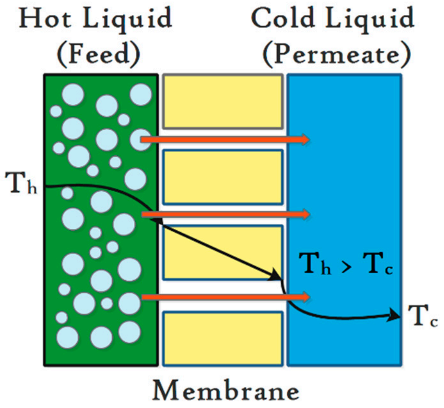

Figure 1. Schematic diagram of the MD process [10].

2.3. Limitation and Benefits of MD Process

In comparison with other conventional membrane separation systems, MD separation process brings tremendous benefits such as having low operating temperatures, being cost-effective by applying waste heat and renewable energy sources, being able to treat wastewater with a high level of purity, and being less likely to suffer from membrane fouling [18,27,28]. Therefore, these remarkable features make MD an attractive method for wastewater treatment, seawater desalination, and so many other industrial applications including environmental purification, in the food industry [29], in medicine [30,31], and in the production of acids, etc. However, employing MD in industry is limited by some significant challenges like the risk of total or partial membrane wetting [32] or not being commercially available on a large scale [33]. Regarding previous studies, the choosing of suitable membranes and the energy efficiency are the two main factors that must be taken into account when applying the MD process [18,23]. Table 2 presents a detailed overview of the positive and negative aspects of the MD process.

Table 2. A comprehensive overview of the positive and negative points of the MD process.

Advantages	Disadvantages	Reference
Low operating temperature (the process liquid is not essentially heated up to the boiling temperatures)	Lower permeate flux compared to other commercialized separation processes, such as RO.	[9,19,33–35]
Lower hydrostatic pressure required compare to pressure-driven membrane separation processes such as reverse osmosis (RO).		[9,19,33]
High rejection (99–100%) for macromolecules, non-volatile compounds (colloids, salts), and inorganic ions. In fact, 100% separation happens, theoretically.	High susceptibility of permeate flux to temperature and concentration polarization effects, partial or total pore wetting, and membrane scaling and fouling.	[19,36,37]
Lower requirements on the mechanical properties of the membrane.		[18,33]
Larger pore size and less chemical interaction between process solution and membrane lead to less fouling.	High heat loss (by conduction) and energy consumption	[33,36]
Alternative low-grade energy sources like waste heat, solar energy, and geometrical heat can be utilized.	Pore wetting risk	[8,38]
The possibility to combine with some other separation processes in order to build an integrated separating system, like an RO unit or ultrafiltration	Unclear economic and energy costs for different MD applications and configurations, just when waste heat is available MD becomes cost competitive.	[8,33]
An efficient method to eliminate heavy metals and organic from wastewater.		[33]
It is an effective and safe process to remove radioactive waste.	The lack of commercially available MD modulus manufactured for large-scale applications and high-performance membrane.	[33,39]
MD is able to work with a saturated solution or high solute concentration in a liquid stream		[33]
Fewer vapor spaces needed in comparison with common distillation process so MD can be used at a smaller scale.	Having less producers of MD technology	[8,33]
Reduced sensibility to concentration polarization.		[8,33,38]
High concentration polarization or osmotic pressure does not limit performance.		
Having low cost and less sophisticated installation and construction (because of lower operating temperature and pressure), leads to a full level of automation.	Limitations of MD permeate flux, due to a further mass transfer resistance caused by trapped air through the membrane	[18,33]
Being less sensitive to membrane pollution or concentration polarization and without a pretreatment stage.		[8,33,36]

2.4. Membrane Characteristics

Hydrophobicity is the fundamental necessity for an MD membranes process. Therefore, the membranes must be fabricated with original or modified hydrophobic polymers with low surface energies. Moreover, the membrane applied in the MD should have a low resistance to mass transfer and low thermal conductivity to prevent heat loss across the membrane. In addition, the membrane should have good thermal stability in high temperatures, and good chemical resistance to acids and bases. High permeability is another significant feature that a membrane should possess in order to be applied in the MD process. To satisfy this feature, the membrane surface layer must be as thin as possible so that the vapors are allowed to pass through the membrane in a short period of time. Another notable characteristic of a membrane is a high liquid entry pressure (LEP). LEP is regarded as the minimum hydrostatic pressure in an MD system that prevents the liquid solutions from penetrating into the membrane pores. A sufficiently high LEP can be achieved by applying a membrane material with high hydrophobicity and a small maximum pore. In addition, the surface porosity and pore size of the membrane must be as large as possible [18,40].

2.5. Membrane Materials and Modules

The most popular micro-porous hydrophobic membranes are commercially fabricated with polypropylene (PP), polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), polyethylene (PE), inorganic materials, and carbon nanotubes (CNTs) available in plate and frame, hollow fiber, tubular, spiral wound, and flat sheet modules. Currently, PTFE with unique features such as high hydrophobicity and strong resistance against severe operating conditions has dominated the commercial and laboratory applications of MD [23,41,42]. Figure 2 makes a comparison between different membrane modules with their positive and negative points [9]. Table 3 provides information about the characteristics of commercial membranes commonly applied in the MD process.

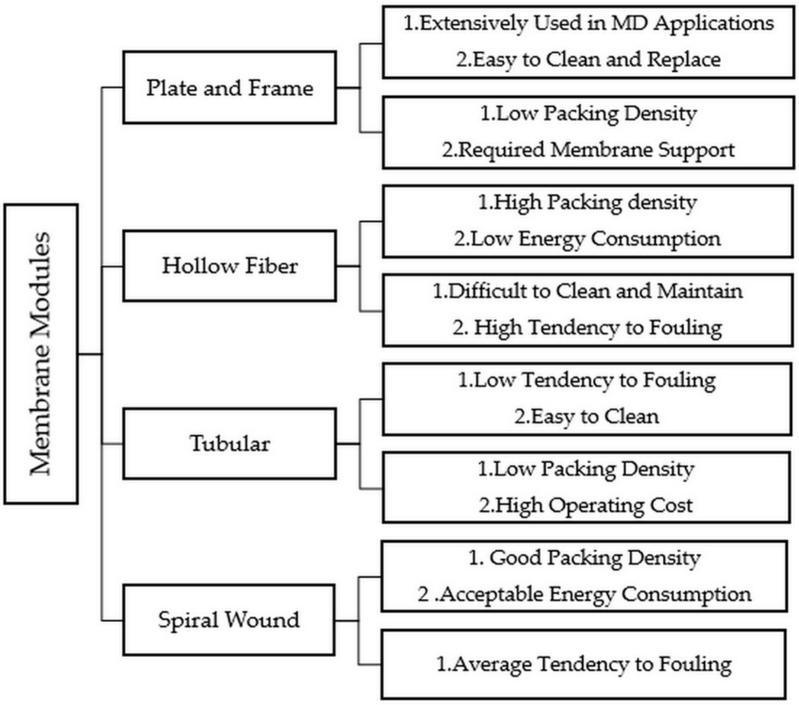

Figure 2. Advantage and disadvantages of membrane modules.

Table 3. Commercial membranes applied in MD (membrane thickness, δ; porosity, ε; liquid entry pressure of water, LEPw).

Membrane Trade Name	Material	Manufacturer	δ (μm)	ε (%)	LEP_W (kPa)	Reference
TF200	PTFE */PP **	Gelman	178	80	282	
TF450	PTFE/PP	Gelman	178	80	138	[8,18,43]
TF1000	PTFE/PP	Gelman	178	80	48	
PT20	PTFE/PP	Gore	64 ± 5	90 ± 1	3.68 ± 0.01	[8]
PT45	PTFE/PP	Gore	77 ± 8	89 ± 4	2.88 ± 0.01	
TS1.0	PTFE/PP	Osmonics Corp.	175	70	-	
TS22	PTFE/PP	Osmonics Corp.	175	70	-	[18]
TS45	PTFE/PP	Osmonics Corp.	175	70	-	
Taflen	PTFE/PP	Gelman	60	50	-	
FGLP	PTFE/PE	Millipore	130	70	280	
FHLP	PTFE/PE ***	Millipore	175	85	124	
GVHP	PVDF ****	Millipore	110	75	204	
PV22	PVDF	Millipore	126 ± 7	62 ± 2	2.29 ± 0.03	[8,44]
PV45	PVDF	Millipore	116 ± 9	66 ± 2	1.10 ± 0.04	
HVHP (Durapore)	PVDF	Millipore	140	75	105	
GVSP	PVDF	Millipore	108	80	-	[18]
Gore	PTFE	Gore	64	90	368	
Gore	PTFE	Gore	77	89	288	
Teknokrama	PTFE	Teknokrama	-	80	-	
Teknokrama	PTFE	Teknokrama	-	80	-	
Teknokrama	PTFE	Teknokrama	-	80	-	
G-4.0-6-7	PTFE	GoreTex Sep GmbH	100	80	463	
Sartorious	PTFE	Sartorious	70	70	-	
MD080CO2N	PP	Enka Microdyn	650	70	-	
MD020TP2N	PP	Enka Microdyn	1550	70	-	[8,18]
Accurel®	PP	Enka A.G.	400	74	-	
Celgard X-20	PP	Hoechst Celanese Co	25	35	-	
Accurel® S6/2	PP	AkzoNobel	450	70	1.4	
Enka	PP	Sartorious	100	75	-	
Enka	PP	Sartorious	140	75	-	[18]
3MA	PP	3M Corporation	91	66	-	
3MB	PP	3M Corporation	81	76	-	
3MC	PP	3M Corporation	76	79	-	
3MD	PP	3M Corporation	86	80	-	
3ME	PP	3M Corporation	79	85	-	
Membrana	PP	Membrana, Germany	91	-	-	
PP22	PP	Osmonics Corp.	150	70	-	
Metricel	PP	Gelman	90	55	-	
Celgard 2400	PP	Hoechst Celanese Co.	25	38	-	
Celgard 2500	PP	Hoechst Celanese Co.	28	45	-	
EHF270FA-16	PE	Mitsubishi	55	70	-	

* Polytetrafluoroethylene; ** Polypropylene; *** polyethylene; **** Polyvinylidene fluoride.

3. Conventional MD Configurations

A MD process can be categorized into four basic configurations, which plays a fundamental role in separation efficiency and processing cost. Figure 3 shows a schematic diagram of various conventional configurations including direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD), and vacuum membrane distillation (VMD) [40,45–47].

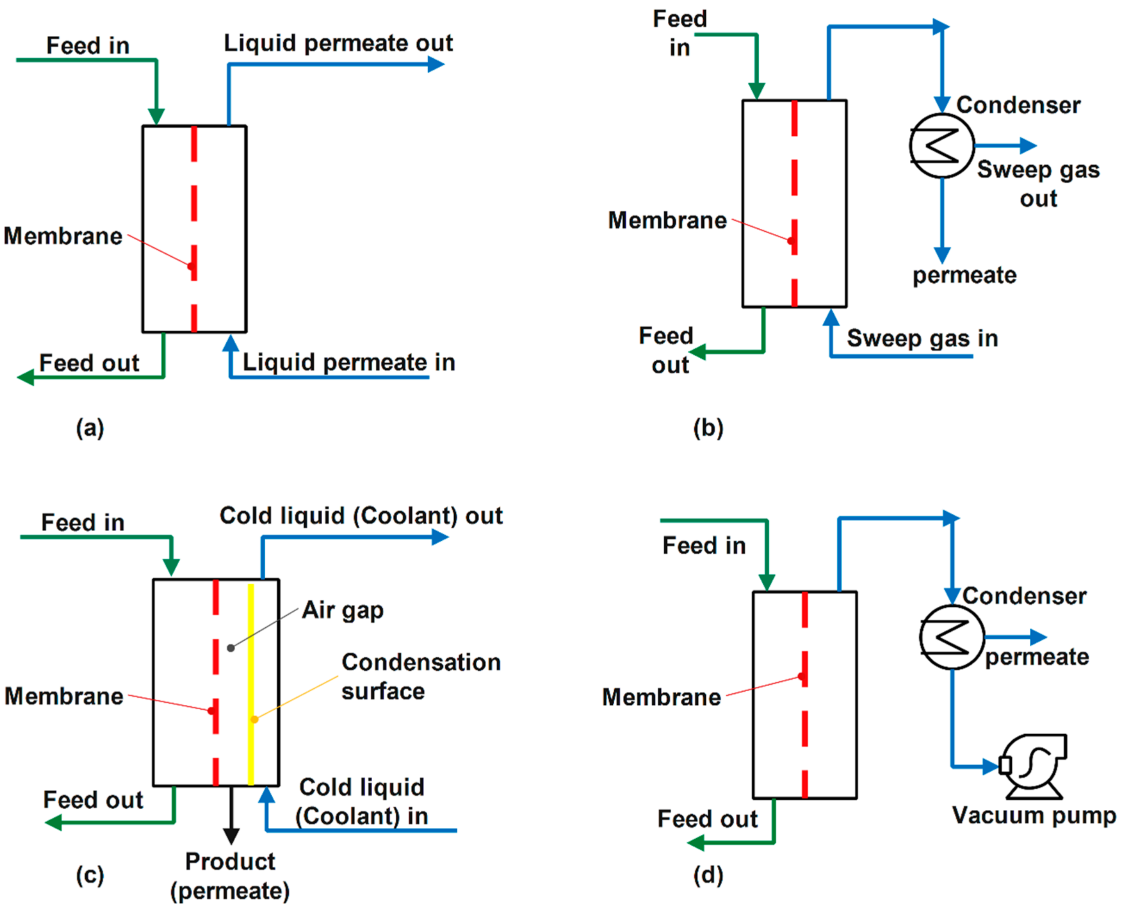

Figure 3. Schematic diagram of conventional membrane configurations (**a**) direct contact membrane distillation (DCMD); (**b**) sweeping gas membrane distillation (SGMD); (**c**) air gap membrane distillation (AGMD); (**d**) vacuum membrane distillation (VMD).

3.1. Direct Contact Membrane Distillation (DCMD)

DCMD is the simplest MD configuration, in which a liquid phase (feed) with high temperature is in direct contact with the hot side of the membrane surface, and a cold aqueous phase is in direct contact with the permeate side. Therefore, volatile compounds evaporate at the hot liquid/vapor interface at the feed side. Having been passed through the membrane pores, the vapor phase will be condensed in the cold liquid/vapor interface at the permeate side. It is notable that the vapor pressure difference is induced by the temperature difference across the membrane and the hydrophobic nature of the membrane prevents the feed from penetrating through the membrane. Despite its simplicity, the conduction heat loss associated with this process is higher than in other configurations. Membrane modules in DCMD could be shell-and-tube or plate-and-frame employed under cross-flow or longitudinal flow [38,48–52].

3.2. Air Gap Membrane Distillation (AGMD)

In this configuration, the evaporator channel is similar to that in DCMD. However, an air gap, which is the controlling factor for the heat and mass transfers, is interposed among the membrane and the cooled surface. The evaporated volatile compounds pass through the membranes and the air gap and then condense onto the cold surface. A significant point about this configuration is that the condensation surface separates the permeate (distillate) from the cold liquid (coolant). Therefore, the cold liquid can be other liquid like cold feed water. The AGMD configuration has the highest energy efficiency among the other configurations and the applied membrane could be both flat sheet and hollow fiber. Furthermore, the used condensation surface is usually a thin dense polymer or metal film [53–59]. Nonetheless, when it comes to popularity, the AGMD process lags behind DCMD and VMD processes because of its complicated module design [60]. Kalla et al. [60] comprehensively reviewed the most recent developments in the AGMD process. Based on their investigation, material gap membrane distillation, double stage AGMD unit, conductive gap membrane distillation, superhydrophobic condenser surface, multi-stage and multi-effect membrane distillation, modified air gap membrane distillation, tangent and rotational turbulent inlet flow, and vacuum assisted air gap membrane distillation process are recent advancements in AGMD process. Woldemariam et al. [61] presented an exergetic analyzing (exergy evaluations are necessary tools for analyzing the performance of separation systems, including those featuring MD.) of AGMD systems at a laboratory and pilot scale. The energy efficiency results indicated that the materials of the condensation plate play a crucial role in optimizing the performance of MD systems such as heat transfer across modules. Stainless steel and polypropylene were considered as the appropriate materials in this regard.

3.3. Sweeping Gas Membrane Distillation (SGMD)

In SGMD, which is also known as air stripping membrane distillation, an inert gas (stripping gas) is applied at the permeate side of the membrane as a carrier to sweep the vapor or collect vapor molecules from the membrane surface. Similar to AGMD, a gas barrier decreases the heat loss and significantly increases mass transfer making SGMD a process with promising future perspectives [62,63]. Nonetheless, SGMD generates a small volume of permeate vapors while needing large volumes of sweep gas and external condensers, consequently incurring extra expenses. Therefore, the process has received little attention in comparison with other MD configurations such as DCMD [19,64]. Applying metallic hollow fibers or a coating membrane with polydimethylsiloxane could significantly enhance the water vapor permeate flux up to 40% in sweep gas membrane distillation [65]. Moore et al. [66] developed a non-steady process model to simulate an SGMD system integrated with solar thermal and photovoltaic power for the desalination of drinking water. The economic analysis indicated that the optimized proposed technology for cost recovery over a 20-year service life is 84.7 $\$/\mathrm{m}^3$, which is more than alternative sources water costs. Therefore, future work on SGMD is necessary to make this system economically competitive.

3.4. Vacuum Membrane Distillation (VMD)

In the VMD configuration, a vacuum is created by a pump at the permeate side of the membrane module. Then, an external condenser is used as for AGMD if the permeate stream is the product. In addition, the vapor pressure difference is formed by continuous removal of the vapor permeate from the vacuum chamber. To form the driving force, the created vacuum must be less than the saturation pressure of volatile compounds in the aqueous feed. For the VMD configuration, the conduction heat loss is negligible and membrane wetting is avoidable [9,18,67–69]. Table 4 makes a comparison between different conventional MD configurations and represents the merits and demerits of each conventional process.

Table 4. The merits and demerits of conventional MD configurations.

Method of Treatment	Advantages	Disadvantages	Reference
DCMD	Simplest operation Least required equipment Simplest MD configuration	Not suitable for removing non-volatile organics and dissolved gasses (water must be permeating flux) Highest heat loss by conduction among other configurations	[33,38,48]
AGMD	High flexibility in MD configuration Less conductive heat loss Less tendency to fouling High flux Without wetting on the permeate side	Creation of additional resistance to mass transfer Hard module designing Minimum obtained output ratio	[8,9,55]

Table 4. *Cont.*

SGMD	A suitable configuration for removing contaminant (volatile component and dissolved gasses) Without wetting from the permeate side Lower thermal polarization	Large condenser needed due to the small volume of permeate diffuses in a large sweep gas volume Low flux	[8,9,33,70,71]
VMD	Negligible conductive heat loss High flux Suitable for aroma compounds recovery	Pore wetting risk Higher fouling Vacuum pump and external condenser	[8,9,33]

4. New MD Configurations

Several novel configurations with low energy consumption and improved permeation flux have been developed by scientists and researchers. A brief review of newly proposed MD configurations is now presented.

4.1. Thermostatic Sweeping Gas Membrane Distillation (TSGMD)

The AGMD and SGMD processes can be combined in a process named thermostatic sweeping gas membrane distillation (TSGMD). As Figure 4 clearly illustrates, the inert gas is passed through the gap between the condensation surface and the membrane. Part of the vapor is condensed on the condensation surface (AGMD) and the remainder is condensed over the external condenser (SGMD) [18,72]. This phenomenon basically takes place to minimize the temperature of the sweeping gas, which increases significantly along the membrane module length. In other words, the presence of the condensation surface in the permeate side decreases the temperature of the sweeping gas, which leads to an enhancement in the driving force and the wastewater treatment performance [33,47]. Condensate production in the TSGMD can be increased by enhancing the membrane area, recycling cool air back to the membrane module, and decreasing the airflow across the cooling fins.

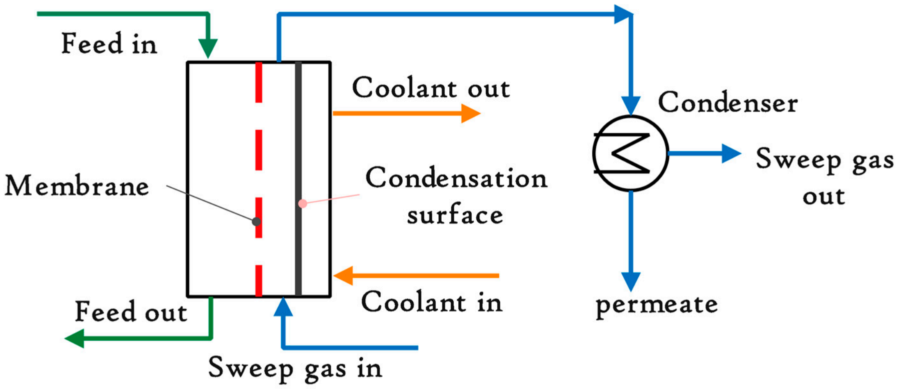

Figure 4. Schematic diagram of thermostatic sweeping gas membrane distillation (TSGMD) [73].

Tan et al. [74] developed a novel SGMD system coupled with a thermoelectric heat pump (TSGMD) to improve the energy efficiency of the water treatment system. The results indicated that applying a T-SGMD system is capable to double the condensate production per unit energy consumed. Furthermore, condensate production in the proposed system can be increased by enhancing the membrane area, recycling cool air back to the membrane module, and decreasing the airflow across the cooling fins. Cool air recycle could affect the condensate flux without a serious loss of cooling in comparison with other tested factors during the operation of the T-SGMD. More importantly, the TSGMD system was able to increase the condensate produced per unit energy without a significant loss in the cooling capacity per unit energy input. This process can be simulated by using a multicomponent Stefan–Maxwell mathematical model. Based on the model analysis, Rivier et al. [62] concluded that since the selectivity of TSGMD is far from unity and can be manipulated by changing operational conditions, this process is suitable for separating azeotropes. Furthermore, the thermal conductivity of the sweeping gas in TSGMD is four-fold lower than that of the membrane, and a small mass transfer resistance exists in the cold chamber due to the gaseous bulk. In one study, the formic acid-water azeotropic mixture was separated by this module [75]. Both experimental and modeling results suggested that TSGMD can considerably shift selectivity with respect to vapor-liquid equilibrium (VLE) data, and the system can be successfully used for the separation processes.

4.2. Multi-Effect Membrane Distillation (MEMD)

This configuration is an AGMD module with an internal heat recovery system based on the concepts of multi-effect, which is suitable for seawater desalination. The cold feed is placed under the condensation surface as a coolant to condense the permeated vapor compounds as well as to absorb heat. The pre-heated feed solution is heated once again before entering the feed channel [19,76]. Figure 5 shows a schematic diagram of the MEMD process. The source of heating for this system should be between 50 to 100 °C and the utilized membrane is usually micro-porous PTFE. It is reported that the module has very low specific energy consumption (between 56 to 100 kWh/m^3) [77].

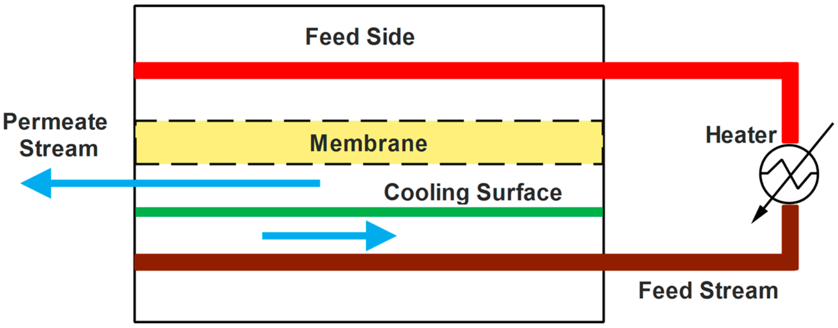

Figure 5. Schematic diagram of the multi-effect membrane distillation (MEMD) [19].

4.3. Vacuum Multi-Effect Membrane Distillation (V-MEMD)

VMEMD shares a similar concept with the multi-effect membrane distillation (MEMD) except for the vacuum enhancement. The system includes a heater, multiple evaporation–condensation stages, and an external condenser. Consequently, the distillate is created in both condensation surfaces and inside the external condenser. In addition, the vacuum condition is developed at the air gap between membrane and condensation surfaces to eliminate the excess air/vapor from the process. In this context, memsys is a state of the art technology and relatively new configuration based on vacuum multi-effect membrane distillation. This highly efficient technology consists of a novel internal heat recycling concept that results in a significant reduction in energy consumption [19,69,78–80]. Figure 6 shows a schematic diagram of the V-MEMD system. The performance of this type of membrane is affected by changing heating conditions, cooling, and feed. The main factors for its optimization and scale-up are the number of stages and the size of each stage. In an experimental study, Zhao et al. [80] found that heating and cooling temperatures are the most important factors affecting the module flux and efficiency. By comparing this module with common technologies, one can conclude that it can provide better heat and mass transfer rates [81]. Furthermore, Mohamed et al. [82] reported that flow rates are also important factors affecting the module performance. They suggested that the system performance can be improved more by performing the following tips: separating distillate and brine tanks, utilizing magnetic or float flow meters, using a separate pump for heating recirculation, and improving the heat isolation or recovering the heat loss in the brine stream.

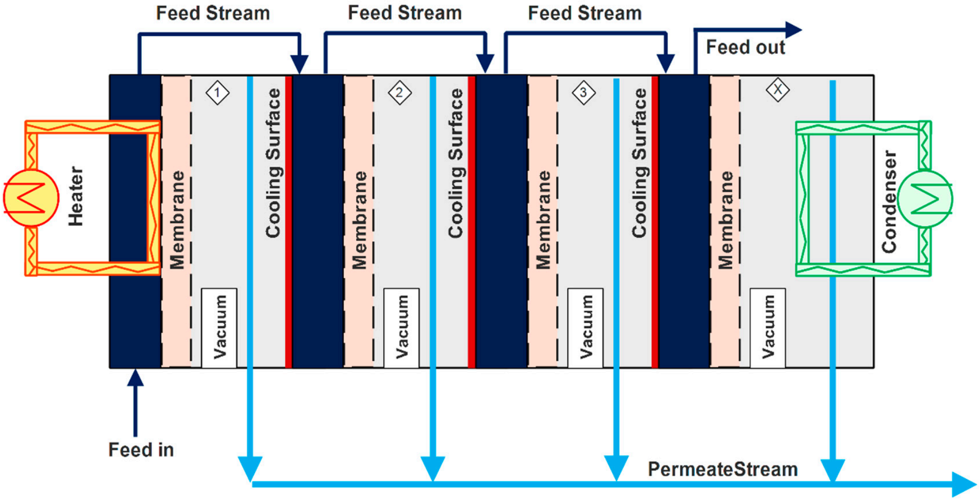

Figure 6. Schematic diagram of vacuum multi-effect membrane distillation (V-MEMD) [19].

4.4. Material-Gap Membrane Distillation (MGMD)

Since AGMD has shown the lowest permeation flux among all configurations, a new and efficient configuration named material gap membrane distillation (MGMD) was developed and designed by researchers to compensate for the weakness of AGMD. In this membrane module, the air gap is filled with either nonconductive materials such as porous support, sand, and sponge (polyurethane) or conductive materials such as the metal mesh. Therefore, the vapor compound flux increases up to 200–800% [83–86]. Figure 7 shows a schematic diagram of the MGMD system.

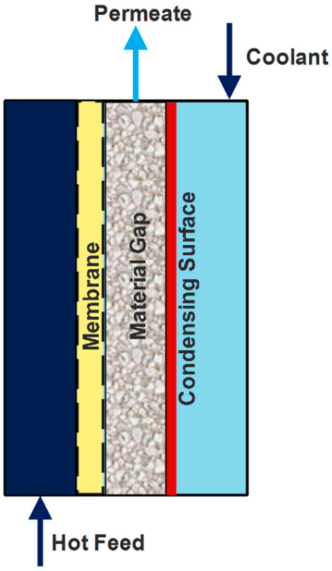

Figure 7. Schematic diagram of material gap membrane distillation (MGMD) [83].

To simulate large-scale module conditions, it is essential to perform the experiments at low-temperature gradients across the membrane. This is because at this condition, the heat recovery is maximized and the water vapor flux would be at a minimum [81].

4.5. Permeate-Gap Membrane Distillation (PGMD)

The combination of DCMD configuration and AGMD module is regarded as permeate-gap membrane distillation (PGMD) or liquid-gap membrane distillation (LGMD). In this configuration, the additional compartment between the membrane and the condensation surface is filled with a static cold liquid solution or with permeate. It is also notable that applying PGMD leads to a higher-surface-related permeate output in comparison with AGMD. This is mainly because, in AGMD, the diffusion resistance of the air layer acts as an obstacle in the process. However, PGMD has greater heat loss than AGMD [83]. Figure 8 shows a schematic diagram of the PGMD system. Winter et al. [85] proposed PGMD with internal heat recovery that was achieved by separating the distillate and coolant. As a result, any liquids such as the feed water can be utilized as the coolant. Therefore, one can place PGMD between AGMD and DCMD to lower the sensible heat transfer to the permeate, but at the cost of greater heat loss. By accepting the superior performance of PGMD over AGMD, Swaminathan et al. [84] reported that the countercurrent flow of the pure water in the gap to the cold stream results in the highest energy efficiency, and increasing the gap conductivity enhances the permeate production.

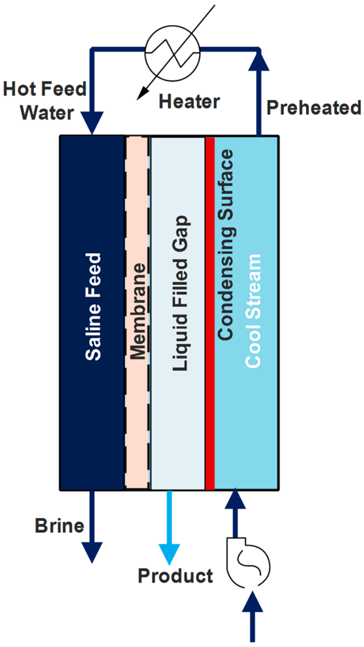

Figure 8. Schematic diagram of permeate-gap membrane distillation (PGMD) [86].

5. Application of MD

Generally, MD has been applied in various areas including desalination, the chemical industry, the food industry, the textile industry, pharmaceutical, and biomedical industries and the nuclear industry. Table 5 provides more information about each application.

Table 5. The application of the MD process in different industries.

Area	Application	MD Configuration	Reference
Chemical industry	Removing volatile organic compounds from water Acid concentrating Crystallization Azeotropic mixtures separation	VMD DCMD SGMD AGMD	[33,87–89]
Desalination	Producing pure water from brackish water	VMD DCMD SGMD AGMD	[33,34,90,91]
Food industry (Juice and Dairy)	Juice concentrating Processing of milk Temperature sensitive materials	VMD DCMD AGMD	[33,92–95]

Table 5. *Cont.*

Textile industry	Dye removal Wastewater treatment	VMD DCMD	[33,96–98]
Pulp and paper industry	Removing sodium sulfate, organic and inorganic compounds, adsorbable organic halogens (AOX), color, phenolic compounds, and chemical oxygen demand (COD) from wastewater	DCMD	[99–101]
Pharmaceutical and biomedical industries	Wastewater treatment Water removing from protein and blood solutions	DCMD	[30,33,102,103]
Nuclear industry	Producing pure water Wastewater treatment Radioactive solutions concentrating	DCMD VMD	[31,33,104,105]
Gold mining	Reusing mining effluents Removing hazardous metals and ions such as sulfate from mining effluents	DCMD	[106]
Bioethanol production plants	Recovery of ethanol from scrubber-water	AGMD	[107]

The Application of MD in Water and Wastewater Treatment

MD technology has been extensively used in the purification of wastewater produced from various industries, in order to recover valuable compounds or make wastewater less dangerous to the environment. However, in comparison with other membrane processes, MD is more difficult to apply on an industrial scale because of some serious economic and engineering problems.

When it comes to desalination, many various types of technologies are available including thermal- and membrane-based desalination processes. The thermal-based group comprises processes such as multistage flash distillation (MSF); multi-effect distillation (MED); and, single- or multiple-effect evaporation (SEE/MEE) systems, which can be coupled to mechanical or thermal vapor compression (MVC/TVC). Membrane technologies include reverse osmosis (RO), forward osmosis (FO), electrodialysis (ED), and membrane distillation (MD). Onishi et al. comprehensively reviewed the main advantages and disadvantages of each process [108].

RO has been considered as the most economical and the least energy intensive technology for large-scale seawater desalination, followed by MED and MSF [23,109,110]. However, the unique characteristics of MD have made this process an excellent option with high efficiency. For instance, MD, in particular, DCMD, has enormous potential in the desalination of highly saline wastewaters where MD fluxes can remain comparatively high, much higher than those for RO. Moreover, in small-scale applications where the quality of water is not suitable for currently established technologies such as RO-based processes, MD is an effective alternative. This process could also be co-located with industrial facilities and power generation systems to take advantage of the waste heat and low-cost thermal energy to produce high-quality water. In addition, MD is a potential treatment candidate for combining with other separation techniques such as RO, ED, crystallization, and bioreactors to enhance water recovery and decrease the amount of concentrate requiring disposal. Therefore, MD has a practical application in water treatment with zero (or near zero) liquid discharge and can be more economical than other established thermal processes in zero-liquid discharge applications. The permeate with extremely high quality in the MD process compared to RO permeate can also offer considerable benefits particularly when purified water is required as boiler feed [19,23,111]. In mining industries, the process combining ultrafiltration (UF) and reverse osmosis (RO) is widely applied for wastewater treatment, in which 80% of COD, more than 95% chroma and almost all the ferrous irons and bacteria can reject significantly. Nonetheless, the brine discharge and the water recovery ratio (limited to around 30% to 60%) of the RO process remain serious issues in this regard. Therefore, MD could be proposed to address these problems by enhancing the water recovery ratio and recovering minerals [8,112,113].

Lokare et al. [114] investigated the synergies and potential of DCMD for wastewater treatment produced during gas extraction from unconventional (shale) reservoirs in Pennsylvania (PA).

An exhaust stream from Natural Gas Compressor Station (NG CS) was used as the waste heat source for DCMD operation providing a feasible option to treat high salinity generated water. They developed an ASPEN Plus simulation of DCMD using fundamental heat and mass transfer equations and the literature correlations to optimize the design and operation of large-scale saline water desalination and estimated the energy requirements of the process. The minimum temperature gradient of 10 °C between the permeate-side and feed stream was used to achieve the optimum membrane area when several membrane modules are provided in series. According to obtained results, the amount of available waste heat of NG CS regardless of the produced water salinity is much higher than the amount of waste heat needed to concentrate produced water in PA to 30% salinity. Moreover, the results indicated that DCMD is able to concentrate all the produced water in PA utilizing NG CS waste heat. Nonetheless, the economic probability of the process must be assessed to determine major cost drivers and barriers. Ali et al. [115] evaluated the integration of a microfiltration and membrane distillation process for water treatment and minerals recovery from produced water. The results indicated that the integrated process offers the opportunity of converting generated water into salt and freshwater highly efficiently and also minimizes the issue of waste disposal. Nonetheless, to make the system commercially available, better arrangements for separating crystals of various salts should be made. Boukhriss et al. [116] simulated and experimentally studied an AGMD membrane distillation pilot for the desalination of brackish water and seawater with zero liquid discharged. The theoretical model was generated using Matlab and verified utilizing pilot-scale experimental data. Their investigation showed that the AGMD configuration is capable of producing desalinated water with zero liquid discharged even at a low hot-fluid supply temperature of 25 °C, which makes the system feasible to be coupled with low-temperature heat sources such as a solar collector. Baghbanzadeh et al. [117] investigated a zero thermal energy input membrane distillation (ZTIMD) process which was also a zero-waste system. The required thermal driving force for the process was provided by using the warm seawater of the surface as the feed and the cold water at the bottom of the sea as the coolant. The innovative concept of their invention revolutionized the seawater desalination industry. This is mainly because ZTIMD was revealed to be economically more efficient than current seawater desalination processes by simulations based on a single-pass DCMD system. In other words, under the optimum conditions, the proposed ZTIMD process could provide pure water with a cost of \$0.28/m^3 at the particular energy consumption of 0.45 kW h/m^3, which is notably lower than that of the main current seawater desalination methods such as RO (\$0.45–2.00/m^3). A novel water desalination method which couples thermal membrane distillation (TMD) with reverse osmosis (RO) was developed by Huang et al. [118]. They proposed a water–energy integration process with the strong nexus of water and energy. Furthermore, a dual-objective model was formulated to analyze the system thermodynamically and optimize the process under the objective function of minimizing fuel and freshwater consumption. The sensitivity analysis of the heat-to-power demand ratio revealed that the RO-TMD coupling water desalination process is superior to traditional single RO at a high heat-to-power demand ratio in terms of minimizing freshwater and fuel consumption. In the proposed novel water–energy integration system, the fuel and freshwater consumptions were reduced by 1.7% and 21.0%, respectively, compared with those of the conventional system.

A comprehensive summary of more MD applications in wastewater treatment on a laboratory-scale is presented in Table 6. Although the number of research papers published in the MD application in wastewater treatment has increased significantly over the past few years, tremendous effort should be taken to design and fabricate novel membrane modules to permit a successful industrial application of this separation technique. It is worth mentioning that, various excellent membrane modules have become available in the market recently. Therefore, in the near future, a MD process with industrial applications may become increasingly available.

Table 6. Summary of MD applications in wastewater treatment.

Feed	Membrane Configuration	Membrane Material	Contaminant	Removal Efficiency (%)	Scale	Reference
Radioactive wastewater (SrCl2)	VMD	PP	Sr^{2+}	Over 99.60	laboratory-scale	[104]
Metal solution (salts of Co (II), Zn (II), Cu (II), Ni (II), Cd (II) and Pb (II))	VMD	Poly(vinylidenefluoride)-titanium tetraisopropoxide PVDF-TTIP	Heavy metals	Total removal	laboratory-scale	[119]
Distilled water and crude oil	VMD	PVDF	Total Organic Carbon (TOC)	93.4–97	laboratory-scale	[120]
Olive Mill WasteWater (OMWW)	DCMD & VMD	PP	Polyphenols TOC	99.6 89 and 99.6	laboratory-scale	[121]
Industrial textile wastewater	DCMD	PVDF-Cloisite 15A nanocomposite	Colour Total Dissolved Solids(TDS) Chemical Oxygen Demand(COD)	95.3 93.7 90.8	laboratory-scale	[122]
Synthetic dye solution	DCMD	PVDF modified by ethylene glycol (EG)	RB5	99.86	laboratory-scale	[98]
Highly saline radioactive wastewater	DCMD	PP	Nuclides (Co(II), Sr(II), Cs(I)) and boron (B)	>99.97%	laboratory-scale	[123]
Synthetic wastewater and Seawater	Osmotic membrane bioreactor (OMBR)—(DCMD) hybrid system	PTFE active layer and a PP supporting layer	30 trace organic contaminants	>90%	laboratory-scale	[37]
Geothermal water	AGMD	PP, PTFE and PVDF	Boron	99.5%	laboratory-scale	[21]
High salinity hydraulic fracturing produced water (HFPW)	Combined Electrocoagulation (EC) and DCMD	Ethylene chlorotrifluoroethylene (ECTFE)	Turbidity, Total suspended solids (TSS) and TOC	96%, 91% and 61%, respectively	laboratory-scale	[124]
Industrial dyeing wastewater	DCMD combined with physicochemical and biological treatment	PTFE and PVDF	COD and color removal	96% and 100% respectively	laboratory-scale	[49]
Saline oily wastewater	DCMD	PVDF modified with silica nanoparticles and Polystyrene (PS) microspheres	Oil and gas emulsified wastewater	Highly desirable	laboratory-scale	[125]
Mining wastewater	VMD	PVDF membrane was coated by Hyflon AD materials (Hyflon AD40L, Hyflon AD40H)	Mining waste	Highly efficient	laboratory-scale	[126]
Bentazon herbicide solutions	VMD	PTFE	Bentazon	Very effective	laboratory-scale	[127]
Fermentation wastewater	DCMD	PP	COD, TOC	95%	laboratory-scale	[128]

6. Fouling Phenomenon in MD Process

The term "fouling" has been regarded as a complex phenomenon which is an inevitable part of each membrane process and adversely affects membrane performance. Generally, fouling is the precipitation and accumulation of various foulants such as particles and dissolved components on the external surface or inside the membrane as pore blocking. Figure 9 clearly illustrates external surface fouling and internal fouling pore blocking.

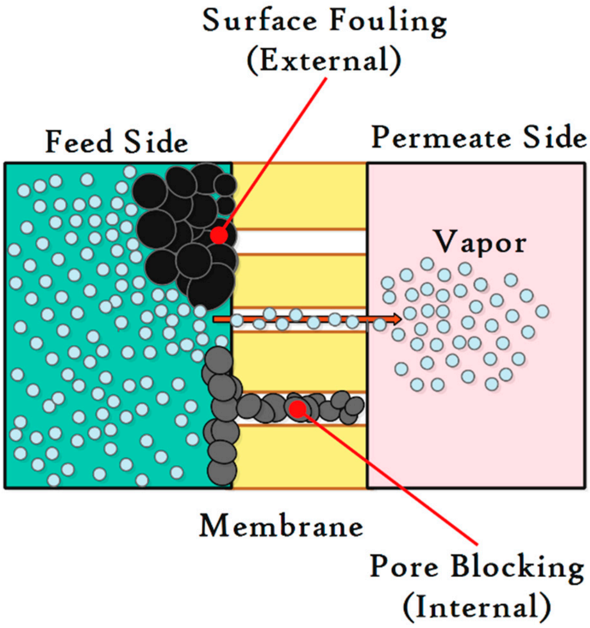

Figure 9. Schematic diagram of surface fouling (external) and pore blocking (internal) [35].

A fouling layer puts on extra thermal and hydraulic resistance to the process and decreases the temperature difference across the membrane, which means a sharp reduction in the driving force. Consequently, the permeate flux decreases drastically. If fouling does not address this properly, it will contribute to membrane damage, early membrane replacement or even shutdown of the operation [35]. The main fouling phenomena commonly occur during water and wastewater treatment and are categorized based on the foulant type as organic, inorganic (scaling), biological, and particle fouling. When suspended solids and metal hydroxide in source water accumulate on the membrane surface and inside the membrane pores, forming a cake layer, the phenomenon is known as particle fouling. Scaling is regarded as the precipitation of inorganic salts presented in source water such as calcium carbonate, calcium sulfate, silicate, NaCl, calcium phosphate, $BaSO_4$, $SrSO_4$, ferric oxide, iron oxide, aluminum oxide, inside the membrane pores, which leads to bulk, pore plugging/clogging, and membrane crystallization. Furthermore, the adsorption of natural organic matter (NOM) such as HA, fulvic acid, protein, polysaccharides, and polyacrylic polymers on the membrane has been considered as organic fouling which contributes to gel formation of the macromolecular compounds and membrane wetting. As well as that, when various aquatic organisms such as fungi, sludge, algae, yeast, and micro-organisms in source water form a biofilm on the membrane, the fouling is named biofouling [35,129]. Nevertheless, mostly, a combination of several types of fouling mechanisms occurs in actual MD processing, as opposed to a single fouling mechanism which makes the problem more complicated to address. It is worth mentioning that membrane fouling is expected to be less devastating in MD, due to the absence of hydraulic pressure in such processes compared to pressure-driven membrane processes. Nonetheless, the continuing presence of the membrane in the highly concentrated feed solution to meet the pure liquid discharge makes the MD process vulnerable to membrane fouling.

Bush et al. [130] compared the performance and fouling behavior of MD and nanofiltration (NF) processes applying silica-saturated water from 225 mg/L to 600 mg/L SiO_2 to illustrate the potential differences in the silica scaling behavior and its impacts on MD performance compared to a pressure-driven membrane process. The results showed that salt rejection during MD was >99.8% for all solutions tested and was unaffected by scaling, while rejection during NF was between 78–90% and tended to decrease after scaling. NaOH solution at $pH > 11$ was used to clean the fouled membranes for both processes, which was extremely effective at the restoring water flux but unable to remove the silica scale layer completely. Tow et al. [131] analyzed the fouling and scaling behavior of RO, forward osmosis (FO), and DCMD using a single membrane module under the same hydrodynamic conditions (flux and cross-flow velocity). During fouling experiments, calcium sulfate was used as a model inorganic foulant and alginate was utilized as a model organic foulant. Based on their results, FO showed the greatest scaling resistance while MD tolerated organic fouling much better than FO and RO. Although FO and MD each indicated a higher resistance to one type of foulant, neither process outperformed RO in the resistance to complex fouling including organic and inorganic fouling.

Typically, the characteristics of foulants (concentration, molecular size, solubility, diffusivity, hydrophobicity, charge), water (solution chemistry, pH, ionic strength, presence of organic/inorganic matters), and membrane (hydrophobicity, surface roughness, pore size, surface charge, surface functional groups), as well as operational conditions (flux, solution temperature, flow velocity), can significantly affect the fouling formation phenomenon [61,132]. In this regard, various approaches are employed by researchers in order to detect and prevent membrane fouling summarized in Table 7. Shan et al. developed a versatile approach for designing an amphiphobic membrane surface [133]. During this method, a biomimetic system was investigated to design an amphiphobic surface with a unique structure and controllable wettability. A commercial PVDF was modified via superhydrophobic nanocoating using polydopamine (PDA) followed by the fluorination of 1H,1H,2H,2H-perfluorodecanethiol. The proposed amphiphobic membrane indicated excellent superhydrophobicity with a water contact angle of $167.6° + 0.27°$ as well as remarkable chemical and thermal stability under severe conditions. Another striking feature about this membrane was its outstanding anti-fouling capability with higher flux and great salt rejection in the long-term DCMD process, which exhibits promising potentials for industrial applications.

Table 7. Summary of detection and prevention methods for membrane fouling.

Detection Method(s)	Prevention Process(es)	Reference
• Permeate flux decline • Scanning electron microscopy (SEM) • Energy dispersive X-ray spectroscopy (EDS) to evaluate the elemental composition of a fouled layer • X-ray powder diffraction (XRD) to analyze and evaluate the crystalline nature of inorganic, organic, polymers, metals, or composite materials • Atomic force microscopy (AFM) is applied to characterize the surface of the membrane • SEM–EDS/TEM (transmission electron microscopy)–EDS • Atomic absorption spectroscopy (AAS) analysis to determine the fouling composition using the absorption of optical radiation (i.e., light) by free atoms in the gaseous state • Contact angle (CA) (the angle between the liquid drop and the horizontal surface, an angle below 90° shows hydrophilic behaviour, while an angle above 90° shows hydrophobicity) • Membrane autopsy • Fourier transform infrared (FTIR) to obtain the infrared (IR) spectra of the sample for evaluating and identifying the chemical bonds and molecular structure of organic molecules and analyzing of organic and inorganic functional groups on the membrane surface • Attenuated total reflectance-fourier transform infrared spectroscopy (ATR-FTIR) to analyze organic and inorganic fouling • Gel permeation chromatography or gel filtration chromatography (HPSEC) or using flow field-flow fractionation FlFFF to determine the size or molecular weight distribution of organic matters • Liquid-chromatogram organic carbon detection (LC-OCD) • Zeta potential (to determine the electrokinetic phenomena of membranes and evaluate the possible interaction between the foulants and membrane surface)	• Membrane flushing (membrane cleaning by regular deionized (DI) water) • Ultrasonic irradiation technique • Chemical cleaning of membrane by acids, alkalis, metal chelating agents, surfactants, enzymes, and oxidizing agents (2 h water, with 0.1 M NaOH or 2–5 wt.% HCl solutions or 0.029 M Na_2EDTA and 0.058 M NaOH or citric acid followed by NaOH or by HCl) • Pretreatment (coagulation, multimedia filtration, sonication, deep-bed filtration, pH changes boiling/thermal water softening, chlorination, degasification,) • Gas bubbling • Increasing the feed flow rate • Temperature and flow reversal • Using anti-fouling membranes (such as membrane surface modification by applying different superhydrophobic coatings such as sodium alginate hydrogel, TiO_2 nanoparticles, a mixture of polydimethylsiloxane (PDMS) and hydrophobic SiO_2 nanoparticles) • Using antiscalants (chemical additives) such as condensed polyphosphates, organophosphonates, and polyelectrolytes	[35,61,128,129,132,134–150]

Table 7. *Cont.*

- Tensile strength
- Direct visualization
- EXSOD (ex-situ scale observation detector) for real-time crystal monitoring of membranes
- Autopsy
- Optical laser sensor method for investigation of the deposit thickness on a membrane
- Ultrasonic time-domain reflectometry (UTDR) to observe and evaluate the deposition of combined organic and colloidal fouling on the membrane surface as well as providing physical characteristics of the media where the waves travel and also providing the real-time measurement of the location of an interface
- Inductively coupled plasma mass spectrometry (ICP-MS) to determine the concentration (ppm, ppb) of metal and non-metal elements in a fouling layer
- Streak-plate method to determine the number of microorganisms
- A confocal laser scanning microscopy (CLSM) technique is applied to obtain high-resolution optical images with depth selectivity and also to visualize and quantify biofilms in-situ in combination with a fluorescent probe.
- Infrared thermography (IRT) technique is applied to measure the surface temperature and its distribution figure out whether a foulant is metallic or non-metallic in nature
- Excitation Emission-Matrix Fluorescence Spectroscopy (EEM) to detect proteins, acid and other organic materials present in fouled membranes

7. Wetting Phenomenon in MD Process

In the wetting phenomenon, water enters the pores of the membrane and fills them by breaking the surface tension presenting between liquid and vapor on the surface of the membrane [151]. As previously described, mass transfer through the membrane pores only takes place in the vapor phase, from a hot feed solution. Therefore, the liquid feed must not be allowed to penetrate partially or entirely through the dry membrane pores. As Figure 10 clearly illustrates, the degree of membrane wettability may vary according to the area wetted by the liquid. The water might be present only in the external surface layers of membrane pores, or a fraction of pores inside the membrane with the largest diameters might be wetted, or all the pores inside the membrane might be filled by water. To address this issue, various approaches have been developed [152–156]. Extra liquid entry pressure and membrane fouling are two principal causes of membrane wetting. Kim et al. [157] fundamentally investigated the MD process integrated with crystallization in order to prevent inorganic scaling induced by multivalent ions such as barium and calcium in shale-gas-produced water treatment. By utilizing the proposed system, scalant loading was decreased significantly and membrane wetting was mitigated dramatically. Therefore, the total recovery of the process was increased up to 62.5%. Furthermore, experimental results indicated that the pretreatment process for oil and grease before MD application is mandatory for enhancing the stability of water production and sustaining the integrity of permeate water quality.

A comprehensive review of wetting mechanisms, wetting causes, and wetting detection methods was carried out by the authors of [156]. Jacob et al. [26] developed a novel method to visualize and evaluate wetting in the MD system based on the detection of dissolved tracer intrusion, which is capable of detecting all pore wetting mechanisms at different locations on a membrane. The function of the developed method was based on the ex-situ detection of a tracer (salt) intrusion by SEM-EDX, after the performance of the VMD applying a saline solution. Guillen-Burrieza et al. [158] investigated the effects of the MD operation variables on membrane wetting. They concluded that the wetting time and rate are significantly reduced by the parameters enhancing flux. Damtie et al. [159] suggested a new methodology to treat highly polluted industrial wastewater and analyzed the wetting tendency of different membranes. They studied the performance of the most popular commercially available hydrophobic membranes including polytetrafluoroethylene (PTFE), polypropylene (PP), and polyvinylidene fluoride (PVDF). The investigation confirmed that the type of membrane material and membrane pore size greatly influence the process efficiency. Moreover, the relationship between the membrane surface wettability and MD performance is affected by the composition of feed water during the water treatment [160].

Eykens et al. [161] explored an alternative for the traditional hydrophobic membrane materials through the deposition of a hydrophobic coating using vacuum plasma on a commercial hydrophilic membrane with a microporous structure to prevent membrane wetting at high salinity. The required hydrophobicity (>100°) was obtained and membrane wetting was prevented effectively. Chen et al. [162] developed a ZnO nanostructure on alumina hollow fiber membranes with a uniform pore size of 197 nm and a thin wall of 200 μm to enhance the wetting resistance during the DCMD process with a low surface tension feed. The contact angle of the omniphobic hollow fiber (HF) membrane for a 90% v/v ethanol/water mixture was as high as 138.1°. The SEM, EDX, and AFM analyses showed that the omniphobic alumina hollow fiber membranes not only showed extraordinary wetting resistance for desalinating low surface tension wastewaters but also showed a great potential for industrial applications because of the simplicity of scaling-up.

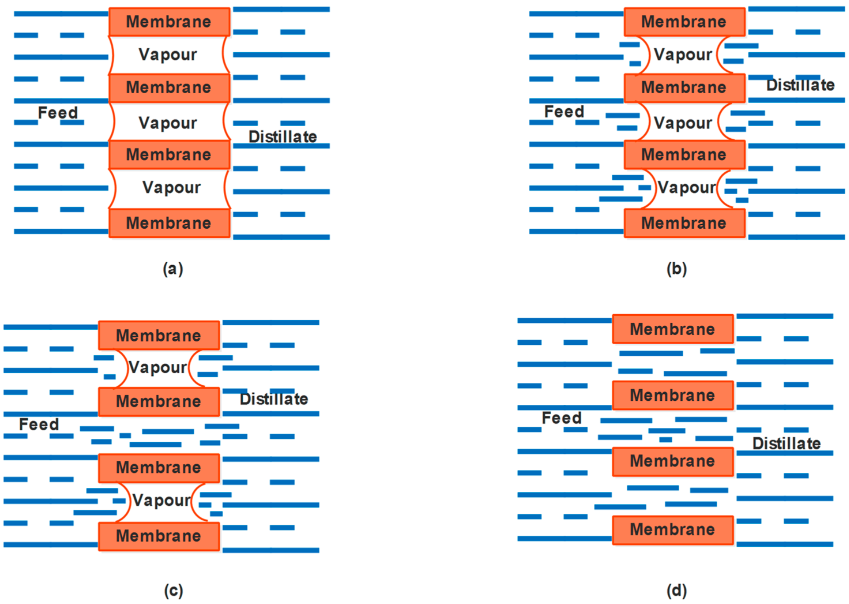

Figure 10. Schematic diagram of membrane wettability stages: (**a**) non-wetted, (**b**) surface wetted, (**c**) partial wetted, and (**d**) fully wetted [35].

8. Novel Approaches to MD Technology

Polymeric porous membranes are traditionally fabricated by conventional methods, most of which contribute to relatively low porosity. Recently, various novel membrane production techniques that have a high porosity above 80% and interconnected open pore structures with a high surface roughness are applied to enhance the membrane performance and provide high flux in MD [163]. Figure 11 illustrates a detailed classification of traditional and novel membrane production methods. Eykens et al. [163] offered a comprehensive definition of each technique and provided advantages and disadvantages of each method in their review paper.

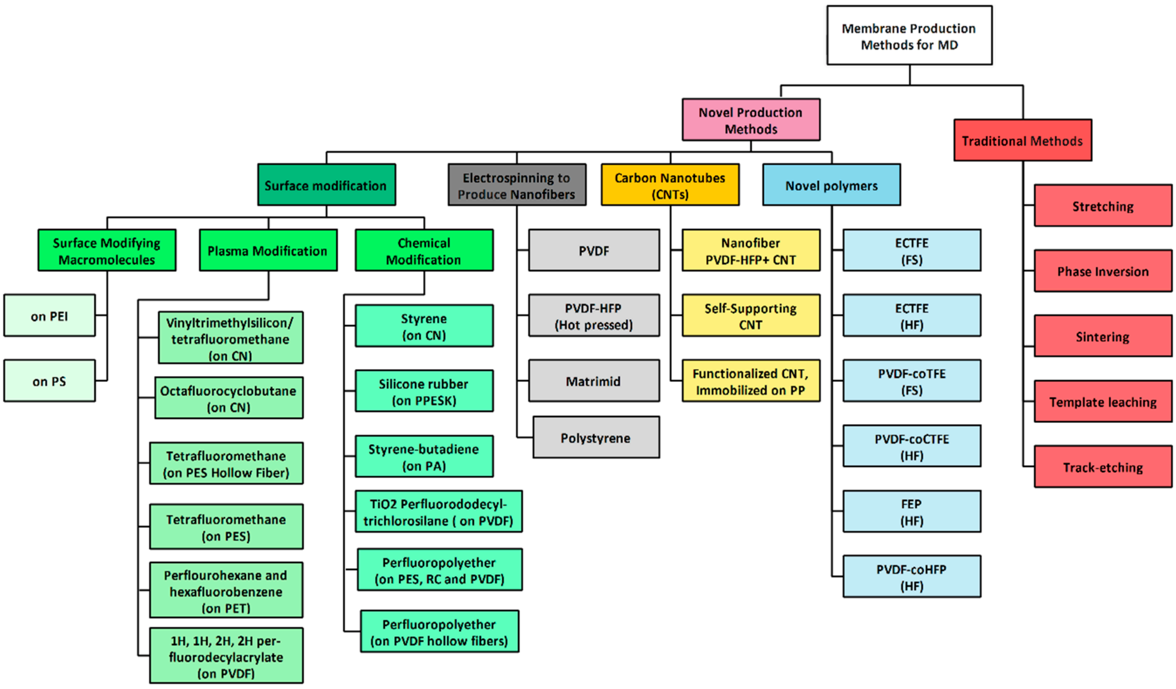

Figure 11. Schematic diagram of various methods for membrane production applied in MD technology [163].

Woo et al. [164] investigated the development and performance of an omniphobic PVDF membrane which was successfully fabricated by electrospinning and modified by tetrafluoromethane (CF_4) plasma treatment for water brine treatment with an AGMD system. They studied the effects of various durations of plasma treatment on the characteristics of the nanofiber membrane. The optimum obtained results (treatment duration: 15 min; liquid entry pressure: 187 kPa; flux: 15.28 $L/(m^2 \cdot h)$; salt rejection ~100%) demonstrated that the formation of new CF_2-CF_2 and CF_3 bonds after plasma treatment without considerably altering the morphology and physical properties could resist the wetting phenomenon by reducing membrane surface energy and providing omniphobic property for low surface tension liquids such as methanol, mineral oil, and ethylene glycol. Therefore, the proposed omniphobic membrane has great potential to treat water containing high salinity and organic contaminants. An et al. [165] fabricate an amphiphobic PVDF-co-HFP electrospun nanofibrous membrane with excellent anti-wetting properties for the MD process. They applied 1H,1H,2H,2H-perfluorodecyltriethoxysilane (FAS) to fluorinate PVDF-co-HFP fibers followed by a crosslinking process to form a network upon dealcoholization under thermal treatment. Based on their results, the FAS-coated PVDF-co-HFP nanofibrous membranes show excellent stable amphiphobicity with high contact angles of 127° against water and oil even on challenging and critical conditions such as long-term operation, presenting sodium dodecyl sulfate surfactant in the saline feed, or boiling water and strong base and acid etchings. Boo and Elimelech [166] provided a self-heating membrane for MD via CNT Joule heating which enhances the desalination efficiency of high-salinity brines. This novel technology increases the thermal driving force by increasing the temperature of the saline feed stream without the need for external heat. Joule heating that is also known as ohmic heating or resistive heating is the process in which thermal energy is produced by the resistance of a conductor to electron flow. In general, in self-heating membranes, a thin conductive composite layer is formed via a sequential spray coating of CNTs and Polyvinyl alcohol (PVA) on a hydrophobic porous substrate (polytetrafluoroethylene).

More importantly, applying novel renewable energy-driven systems in water treatment has dramatically increased with the aim of energy conservation. Figure 12 illustrates using different renewable energy sources including solar energy, waste heat, and geothermal energy as well as applying a membrane distillation crystallization (MDC) method via the precipitation of crystal salts under supersaturation conditions in a crystallizer. A detailed explanation of each method could be found in the following references [64,167]. Furthermore, MD systems with water recycling and heat regeneration could significantly enhance water recovery and thermal efficiency and, consequently, are capable to meet the actual demand [168]. Long et al. [169] investigated a DCMD system integrated with low-temperature waste heat for water treatment. They developed a modified model characterizing the heat and mass transfer in the DCMD, which was validated by excellent agreement with the experimental data. Based on their study, gain output ration (GOR) and mass recovery rate are two major factors to evaluate the performance of a DCMD system with heat recovery. Lee et al. studied the effects of two different types of seawater-coolant feed (backward feed (BF) and parallel feed (PF)) arrangements in a waste-heat-driven multistage vacuum membrane distillation with regard to the improvement of system performance [170]. Based on their investigations, the proposed system with the BF arrangement is more efficient and economical for freshwater production than the PF arrangement at a smaller number of module stages in terms of the specific thermal energy consumption. Furthermore, they comprehensively analyzed different BF arrangement scenarios and found the optimal number to be a 24-stage VMD desalination system in terms of energy efficiency and cost.

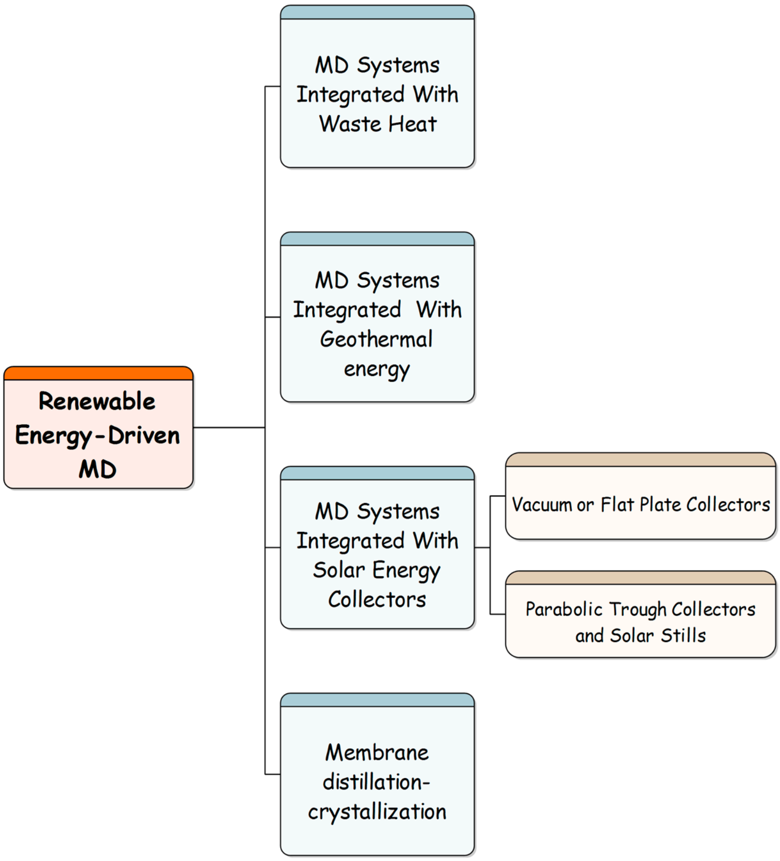

Figure 12. Schematic diagram of various renewable energy-driven MD systems.

9. Economic Analysis of MD Process

Energy-efficient water treatment and desalination processes play a crucial role in enhancing freshwater supplies without imposing considerable strain on scarce resources. By applying low-grade or waste heat, membrane distillation (MD) has shown great potential to augment sustainable water. Nonetheless, economic analyses are essential for the viability of the MD process since a huge amount of energy is used for the water evaporation in order to separate water from non-volatile contaminants and pumping [33]. Furthermore, it should be noted that an energy-efficient MD system effectively applies the thermal gradient for vapor transfer in comparison with conductive heat loss, which is measured by the membrane thermal efficiency of the membrane. In addition, the latent heat of condensation is productively reused in an economical MD system [90]. Recently, the authors of [171] critically examined the crucial factors affecting the energy efficiency of MD processes and explained how future membrane design and process development could considerably boost MD efficiency. They demonstrated that the size of the system can significantly influence the performance of the process. Moreover, they found that enhancing the porosity of the membrane and optimizing its thickness can dramatically increase the MD efficiency. In addition, the configuration of the process plays a leading role in maximizing the latent heat recovery. More importantly, the novelty of membrane materials and surface modification are essential for increasing membrane robustness. Swaminathan et al. [172] comprehensively analyzed a single-stage MD system in terms of the energy efficiency (indicate as a gained output ratio or GOR) and vapor flux for the desalination of feed streams up to a high feed salinity. The system was designed to determine the thickness of the membrane with an optimal cost as well as the size of the system as a function of the ratio of specific costs of heat energy and module area. Based on the obtained results, in the small systems with low salinity, GOR increases and flux decreases with a rise in the membrane area. Furthermore, for the solutions with high salinity, it is essential to determine a critical system size beyond which GOR begins to decrease.

Based on the performance of the MD, the total capital cost of the system, optimum flow conditions, and MD configurations, and the cost of the MD system production may vary from 0.26 to 130 $\$/m^3$. Furthermore, the total energy consumption of the process could change from 1 to 9000 kWh/m^3 based on the type and size of the system, operating conditions, sources of the provided energy, recovery approaches, and the estimated cost of the procedures. Additionally, by applying waste heat, the production cost of a 30,000 m^3/d (capacity plant) MD desalination plant could be reduced from 2.2 to 0.66 $\$/m^3$ [173,174].

A comprehensive cost evaluation of a 111 MWe solar power tower (SPT) plant integrated with the DCMD system was investigated by Soomro and [114,175]. The average freshwater production by the proposed MD system was evaluated up to 40,759 L/day with a cost of $\$0.392/m^3$. The authors of [176] evaluated the economic feasibility of MD for wastewater treatment by performing a techno-economic assessment (TEA) for a hypothetical 0.5 million gallons per day (MGD) direct contact MD (DCMD) which concentrates produced water from 10% (100,000 mg/L) TDS to 30% salinity. Sensitivity analysis showed that the TDS level of the feed and the price of thermal energy significantly affect the total cost of treating produced water. Furthermore, they revealed that utilizing a source of waste heat could considerably decrease the total cost from $\$5.70/m^3_{feed}$ to $\$0.74/m^3_{feed}$. Hitsov et al. [177] demonstrated a graphical user interface tool which could design a comprehensive MD system, comprising all of the supporting equipment and capable to estimate the price of the obtained distillate for different distillation configurations at various production scales and concentration factors. They also investigated several case studies that varied from 2 to 1000 m^3 of distillate per day, with a final brine salinity up to 20 wt.% and feed temperature up to 80 °C, and demonstrated the optimal system design for each case. The cost of distillation varied from 25 €/m^3 (the smallest scale) to 2.1 €/m^3 for the largest scale. Soomro and Kim [178] published an economic evaluation of integrating a 50 MWe parabolic-trough (PT) plant with the DCMD system for freshwater production. The economic analysis illustrated that the proposed system could be a sustainable and economical process producing up to 14.33 m^3 of freshwater per day at a price of $\$0.64/m^3$.

According to a technical economic study conducted by the authors of [106], the capital expenditure (or capital expenses) (CAPEX) and operational expenditure (or operational expenses) (OPEX) of DCMD applied in gold mining effluent treatment (for a membrane lifespan of 1–5 years) are estimated to be US$ 305,483.85 and 0.13 to 0.27 US$/$m^3$, respectively, while the amounts are US$ 575,490.30 and 2.00 to 2.10 US$/$m^3$ for the NF process. This is mainly because the required energy for NF is almost 40 times greater than that for DCMD, due to the need for cooling the feed in the NF process. Moreover, 98% of the thermal energy consumption in DCMD is reduced by applying the residual heat of the effluent. The CAPEX was measured as explained by Hitsov et al., considering a membrane area of 24 m^2 per module and the capital cost ($Ccap$) was calculated per cubic meter of effluent ($Ccap/m^3$), as explained in great detail by Reis et al. [106]. Woldemariam et al. [107] evaluated the CAPEX and OPEX of an industrial-scale district heat-driven MD process for the recovery of ethanol from scrubber water. The economy of the distillation system was obtained from the case studied plant including production rate, the amount of steam used, and other costs such as capital investment. Results of the techno-economic investigation indicated that MD could be a competitive technology for ethanol recovery when the system is supplied by low-grade heat such as waste heat or district heating network.

10. Future Trends and Conclusions

MD is a thermally driven treatment process, which has been perfectly able to treat water containing an extremely high level of salinity and hazardous contaminants. More importantly, MD has the possibility to integrate with other separation processes and renewable energy sources. Nonetheless, few studies were performed on a large scale and with long-term MD application since several challenges such as high energy consumption, fouling, scaling, and pore wetting have limited its commercial application. Therefore, it is essential to fabricate novel membranes with specific characteristics such as low resistance to mass transfer, low thermal conductivity, high thermal stability, high chemical resistance, or a membrane with surface modifications to improve MD performance and characteristics in order to minimize fouling and wetting phenomena and energy consumption, as well as enhancing the permeate flux quality and quantity. Moreover, the development of MD application in wastewater treatment needs to handle more organic and biological fouling, in combination with inorganic scaling. This makes the fouling study more complicated; therefore, more insight into the mechanisms of mixed fouling should be given careful and special attention in the future.

Generally, MD process can be categorized into four basic configurations including direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD), and vacuum membrane distillation (VMD). Additionally, various novel configurations with low energy consumption and improved permeation flux such as TSGMD, MEMD, V-MEMD, MGMD, and PGMD have been proposed recently. Polypropylene (PP), polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), polyethylene (PE), inorganic materials, and carbon nanotubes (CNTs) are the most popular micro-porous membranes commercially fabricated in the form of plate and frame, hollow fiber, tubular, spiral wound, and flat sheet. However, different novel techniques such as electrospinning and surface modification have been employed recently to produce a membrane with a high porosity of above 80% and enhance the membrane performance by providing high flux.

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Abbreviations and Symbols

AGMD	Air Gap Membrane Distillation
COD	Chemical Oxygen Demand
CN	Cellulose Nitrate
CNT	Carbon Nanotube
DCMD	Direct Contact Membrane Distillation
ECTFE	Poly (ethene-co-chlorotrifluoroethene)
ED	Electrodialysis
FEP	poly (vinylidene fluoride-co-chlorotrifluoroethylene)
FO	Forward Osmosis
FS	Flat sheet
HF	Hollow fiber
LEPW	Liquid Entry Pressure of Water
LGMD	Liquid-Gap membrane distillation
MD	Membrane Distillation
MEE	Multiple-Effect Evaporation
MED	Multiple-Effect Distillation
MGMD	Material-Gap Membrane Distillation
MSF	Multi-Stage Flash
NF	Nanofiltration
NOM	Natural Organic Matter
PES	Polyethersulfone
PET	Poly(ethylene terephthalate)
PGMD	Permeate-Gap Membrane Distillation
PP	Polypropylene
PS	Polysulfone
PPESK	Poly(phthalazinone ether sulfone ketone)
PTFE	Polytetrafluoroethylene

PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
PVDF-co-CTFE	Pol (vinylidene fluoride-co-chlorotrifluoroethylene(
PVDF-co-HFP	Poly(vinylidene fluoride-co-hexafluoropropylene)
PVDF-co-TFE	Poly(vinylidene fluoride-cotetrafluoroethylene)
RC	Regenerated cellulose
RO	Reverse Osmosis
SGMD	Sweeping Gas Membrane Distillation
SEE	Single-Effect Evaporation
TDS	Total Dissolved Solids
TOC	Total Organic Carbon
TSGMD	Thermostatic Sweeping Gas Membrane Distillation
TSS	Total suspended solids
VMD	Vacuum Membrane Distillation
V-MEMD	Vacuum Multi-Effect Membrane Distillation
ZTIMD	Zero Thermal Input Membrane Distillation
δ	Thickness
ε	Porosity

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