Journal of Environmental Chemical Engineering 10 (2022) 107631

Contents lists available at ScienceDirect

Journal of Environmental Chemical Engineering

journal homepage: www.elsevier.com/locate/jece

A review on recent progress in environmental applications of membrane contactor technology

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ARTICLE INFO

Editor: Dr. Zhang Xiwang

Keywords:
Membrane contactor
Environmental application
Greenhouse gas capture
Water and wastewater treatment

ABSTRACT

Membrane contactor as a high-performance and cost-effective separation technology has attracted considerable attentions for environmental protection. It offers promising advantages as it can combine with absorption, desorption, extraction and even distillation in one equipment. In this article, the membrane contactor processes are briefly introduced followed by comprehensive review on the latest progresses for environmental applications. Greenhouse gas capture by membrane gas absorption and the membrane development are critically reviewed. In addition, water and wastewater treatments through various membrane contactor processes are discussed for applications such as oil and dye removal, heavy metal ions and radioactive materials separation, ammonia recovery as well as degasification. This article also highlights the current status and future direction of the technology to provide indications for industrial implementation. Although there are not many pilot/full-scale plants using membrane contactor technology in operation, an increasing interest is expected in near future due to strong potential of the technology for environmental applications.

1. Introduction

Continuous environmental problems can be related to the rapid economic growth and industrialization in the recent decades. Although most of the modern industrial devices are energy efficient, they still rely on fossil fuels which could result in emission of greenhouse gases – the main culprit of climate change [1–5]. In addition, continuous consumption of water in most of industrial sectors leads to the production of a large volume of effluent which can adversely affect quality of water sources if it is discharged without proper treatment [6–9].

Nowadays, high-performance and cost-effective separation technologies are highly sought-after due to the stricter environmental regulations and concerns on energy usage [10]. Therefore, the research on environmental applications of the membrane separation technology is in progress owing to its unique advantages including small system footprint, easy operation, scalability and excellent separation efficiency [11, 12]. Among the membrane processes, membrane contactor is very promising as it can combine with other processes such as absorption, desorption, extraction and even distillation in one equipment. Thus, it can completely achieve the advantages of both processes. It should be noted that a dispersive phase contact for mass transfer and separation is occurred through the traditional contactors such as packed bed and tray towers. Hence, the separation of the phases is required after completion of mass transfer which results in a difficult scaling-up.

Compared to traditional contactors, membrane contactor provides a non-dispersive contact between the phases which results in a wide range of phases flow ratio without experiencing problems such as flooding, foaming, channeling, loading and weeping. In the membrane contactors, a very high interfacial contact area between the phases is occurred which significantly improves mass transfer coefficient and compaction of the separation system. These properties are obvious in the cases where thousands of hollow fiber membranes are packed in a single membrane module. Table 1 compares the characteristics between the gas-liquid membrane contactor and some conventional contactors. The distinctive advantages of the membrane contactors have been well documented in previous review articles [13–18]. Indeed, due to its advanced features, membrane contactors have been recognized as promising alternative solution for industrial applications.

Technically, the membrane contactor can be applied for both gas and water treatment. For gas separation, the porous membrane used in the contactor module could provide a non-dispersive contact between gas and liquid absorbent to achieve desire separation. This kind of

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https://doi.org/10.1016/j.jece.2022.107631
Received 15 February 2022; Received in revised form 24 March 2022; Accepted 27 March 2022
Available online 31 March 2022

Table 1
Comparison between membrane contactor and some conventional contactors [37].

Contactor type	Surface area/ volume (m^2/m^3)	Mass transfer coefficient ($K_l.a$) (s^{-1})$\times 10^{-2}$	Gas/liquid flow ratio (%)
Bubble column	50 – 600	0.5 – 12	60 – 98
Packed column	10 – 350	0.04 – 7	2 – 25
Venturi scrubber	150 – 2500	8 – 25	5 – 30
Membrane contactor	1000 – 10000	5 – 50	1 – 99

membrane process is also known as membrane gas absorption. Removing acid gases from flue gas is the common application of the gas-liquid membrane contactor [19–24]. For water and wastewater treatment, water vapor can diffuse through the porous structure of membrane followed by condensation on the other side to produce high quality permeate (as demonstrated in membrane distillation) [25–29] or impurities can be effectively transferred from the water/wastewater to the extracting liquid to achieve desire separation factor (as found in membrane extraction) [30-32]. Typical examples of these processes include seawater desalination, volatile hydrocarbons removal and recovery, heavy metals elimination and ammonia removal. On the other hand, water degassing can take place using a gas-liquid membrane contactor in which the unwanted gas is transferred from water to the sweeping gas phase [33–36]. This process which is also known as membrane gas stripping (MGS) can be employed to separate oxygen (O_2), carbon dioxide (CO_2), ammonia (NH_3) and methane (CH_4) from water/wastewater.

In this article an attempt is made to provide an updated review on the development of membrane contactor processes for environmental applications. More specifically, we will focus on the progress of membrane contactors used for both greenhouse gas control and water/wastewater treatment. Moreover, the challenges associated with membrane contactor and future direction of the technology are discussed.

2. Type of membrane contactor processes

In this section, different types of membrane contactor processes applied for environmental protection such as membrane gas absorption (MGA), membrane gas stripping (MGS), membrane liquid-liquid extraction (MLLE) and membrane distillation (MD) are briefly discussed.

2.1. Membrane gas absorption

MGA process has been investigated as promising alternative for environmental protection in different ways such as greenhouses gas control, volatile organic compound (VOCs) removal and ozonation of wastewaters [38–44]. CO_2 removal from flue gas using MGA process in particular has attained considerable attentions owing to its greenhouse effect. The first MGA process was used for oxygenation of blood using flat sheet polytetrafluoroethylene (PTFE) membrane in 1975 [45]. The indication of CO_2 capture from the gas stream by MGA process was later introduced by Qi and Cussler in 1985 [46,47]. The researchers applied a microporous hollow fiber membrane made of polypropylene (PP) and used sodium hydroxide (NaOH) aqueous solution as liquid absorbent during MGA process. Numerous works have been conducted on development of the membrane materials, liquid absorbent and module configuration/design in order to enhance the separation efficiency of the MGA process [16–24].

Fig. 1(a) shows the schematic diagram of MGA process and the concentration profile. The gas stream and the liquid absorbent usually

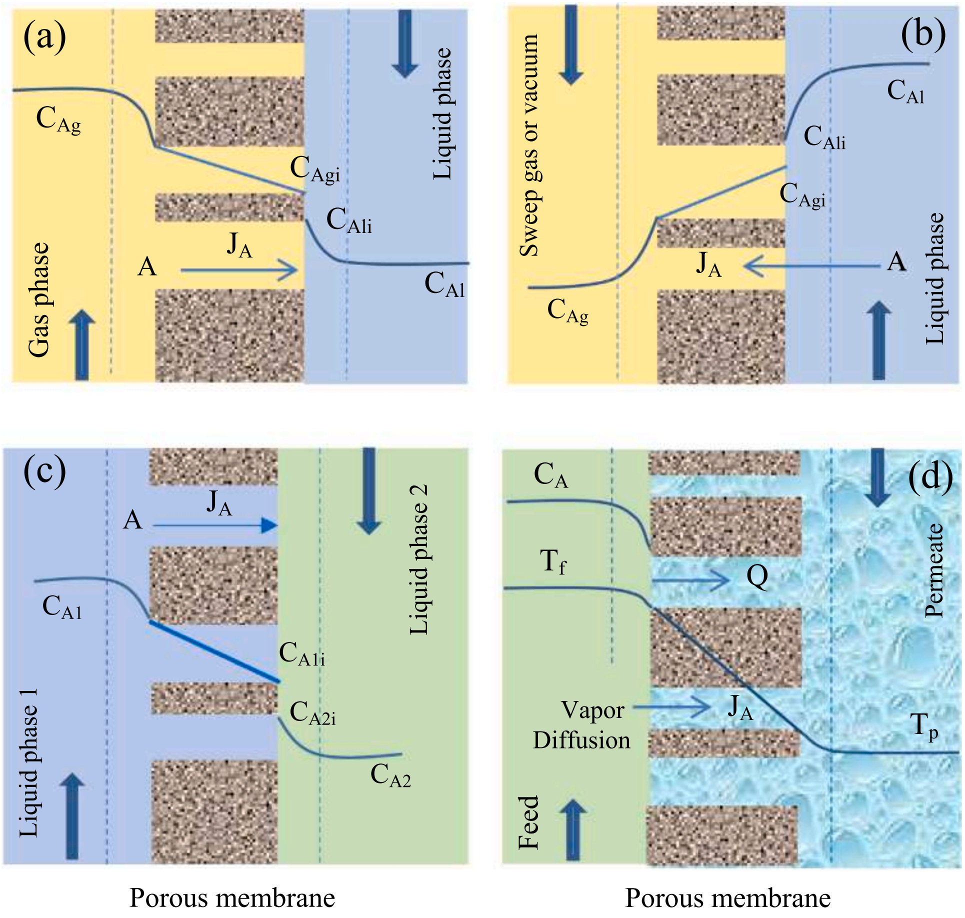

Fig. 1. Schematic diagram of (a) MGA process, (b) MGS process, (c) MLLE process and (d) MD process in treating different phases (A= solute; J_A= mass flux of solute; Q= heat flux; C_{Ag}= bulk gas concentration; C_{Al}= bulk liquid concentration; C_{Agi}= interface gas concentration; C_{Ali} = interface liquid concentration; C_{A1}= bulk liquid phase1 concentration; C_{A2}= bulk liquid phase2 concentration; C_{A1i}= interface liquid phase1 concentration; C_{A2i}= interface liquid phase2 concentration; T_f= feed temperature; and T_p = permeate temperature).

flow counter-currently in both sides of the porous hydrophobic membrane. Due to the concentration gradient (driving force), the gas component (A) is transferred from the gas phase through the porous membrane and absorbed by the liquid phase. The liquid absorbent is selected based on its high affinity to the gas component. Chemical (e.g., alkanolamine, K_2CO_3 and NaOH) and physical (e.g., water and propylene carbonate) absorbents can be used in the MGA processes. The porous membrane as the key element of the process provides a non-dispersive contact between the gas and liquid phases, eliminating the operating problems related to the conventional contactors. The porous membrane has no contribution to the selectivity that is related to the affinity of the liquid absorbent with the gas component. Instead, the membrane plays a role in separating the phases and increasing gas-liquid contact area. It should be noted that due to the driving force of concentration gradient, the membrane contactor can be operated at a low pressure, offering a relatively low operating cost compared to the pressure driven membrane processes.

2.2. Membrane gas stripping

MGS is one usage of gas-liquid membrane contactors for removing dissolved gases from liquid phases. Degassing of water/wastewater has been investigated as an alternative solution to remove different types of gases from the solution. For instances, removal of O_2 and CO_2 from seawater and boiler feed water, separation of H_2S from natural water and wastewater, removal of NH_3 from industrial wastewater and recovery of CH_4 from wastewater [48–54]. Furthermore, MGS has broadly been used in energy processes for the reduction of liquid absorbents such as CO_2 removal from alkanolamine solutions [54–59]. Indeed, MGS has potential for replacing the conventional vacuum towers and forced draft degasifiers for water and wastewater treatment.

Fig. 1(b) illustrates the MGS process in which the liquid containing dissolved gas can flow over hollow fiber membrane or through its lumen side. A vacuum or sweep gas is used to maintain the driving force of mass transfer. The concentration gradient between the liquid and gas phases can separate the dissolved gas from the liquid phase and transfer it through the membrane pores to the gas phase. The solubility of gas in the liquid phase is a key factor for gas stripping which can be promoted by increasing temperature or decreasing pressure of the stripping process.

2.3. Membrane liquid-liquid extraction

MLLE is a non-dispersive membrane contactor process which can be used for treatment of contaminated water/wastewater by an extractant phase. The selectivity of the process is related to affinity of the extractant with the contaminated component. The extraction of various components such as ammonia, phenolic, heavy metals, organic acids and pharmaceutical from wastewaters has been investigated through MLLE [60–65].

Fig. 1(c) represents the schematic of a MLLE process. The concentration boundary layer in the water phase is formed and the impure component diffuses through the liquid filled pores to the extractant liquid phase. The liquid-liquid contact is immobilized on the mouth of each pore and the component is reacted with the extractant agent. The contaminated component is concentrated in the extractant phase which can be directly reused or regenerated. The water phase meanwhile is controlled at slightly higher pressure than the extractant phase ($\geq$ 0.1 bar) to prevent the penetration of extractant phase into water phase [66,67]. Generally, the pores of the membrane are filled with the contaminated liquid phase in order to minimize the membrane mass transfer resistance. The surface of the membrane can be either hydrophobic or hydrophilic depending on the application.

In contrast with conventional liquid-liquid extraction process, non-dispersive membrane contactor presents various advantages. Firstly, it does not require an extra process for the separation of water phase and the extractant phase. Second, the flowrate of the water phase and the extractant phase can be independently manipulated to achieve an easy process optimization. Third, it possesses high contact area which could result in high mass transfer and small system footprint. Lastly, the density difference in the phase is not critical in affecting the membrane performance [68].

2.4. Membrane Distillation

MD is another example of membrane contactor process [69,70], but mainly used in vapor-liquid separation. In this process, the volatile component is evaporated from the liquid feed followed by transfer through the porous hydrophobic membrane to the permeate side. Since the driving force of the process is based on the vapor pressure difference across the membrane, increasing temperature difference between both sides of the membrane can significantly improve the vapor rate and the permeate product. In general, the MD processes can be classified as vacuum membrane distillation (VMD), sweep gas membrane distillation (SGMD), air gap membrane distillation (AGMD) and direct contact membrane distillation (DCMD). Over the past decade, the MD processes have attracted considerable attentions for water desalination and wastewater treatment [71–75].

A schematic diagram of MD process is shown in Fig. 1(d). In VMD, for increasing driving force (decreasing vapor pressure), a vacuum is used at the permeate side and the vapor condensation takes place outside the membrane module. A negligible boundary layer is formed in the vacuum side which indicates a small conductive heat loss through the membrane and improvement of the VMD performance. In SGMD, an inert gas stream is required to sweep and transfer the vapor at permeate side to an external condenser for vapor condensation and pure permeate collection. The sweep gas stream in the membrane module results in a greater mass transfer coefficient and a minimum heat loss is experienced compared to AGMD and DCMD [76]. The SGMD has a great potential for removal of organic volatile components from water.

In AGMD, an air gap is introduced between the membrane and a condensation surface inside the membrane module. The driving force for evaporation of the volatile component is generated by a temperature difference between the feed solution and the cold surface. Due to the presence of air in the permeate side, a low conductive heat loss through the membrane is occurred. However, the presence of stagnant air in the permeate side can increase mass transfer resistance, leading to low permeate flux [76]. In DCMD, the membrane is in direct contact with the liquid feed and the liquid permeate. Due to the hydrophobic characteristic of the membrane, the condensed permeate in the module may wet the membrane and reduce its separation performance. DCMD is the most applied configuration of the MD processes which has been widely used for seawater desalination, brackish water desalination and concentration of aqueous solutions in food industries [71]. However, compared to other MD processes, DCMD presents a higher conductive heat loss and this adversely limits its commercial application.

3. Environmental applications of membrane contactor technology

3.1. Greenhouse gas capture

Population growth and industrial development had caused an annual increase of fossil fuels combustion to meet the energy demand. This adversely causes severe environmental impacts such as air pollution and global warming. Greenhouse gases such as CO_2, nitrous oxide (N_2O), sulfur dioxide (SO_2) and VOCs can be released into the atmosphere via flue gas of power plants and purge gas of chemical industries. Although fossil fuels are the main culprits behind the environmental issues, they are still utilized as the main energy source globally. This is mainly because the production of renewable energy is still challenging due to high production cost and low efficiency [77]. Nowadays, conventional absorption plants (using alkanolamine solutions) are mainly used for greenhouse gas control in which they conquer more than 90% of the market [78]. In order to propose a new separation technology that can compete the proven technologies, more attentions should be paid on the properties such as product quality, environmental issues, energy efficiency, cost and safety.

In the past decades, MGA using hollow fiber membrane contactors for greenhouse gas capture is progressively developed as a promising alternative to conventional contacting devices. Despite several advantages of gas-liquid membrane contactors over conventional contactors, they present some drawbacks associated with its additional transport resistance and membrane wetting by the liquid absorbent. To address

Table 2
Summary of recent research on greenhouse gas capture, biogas upgrading and NG treatment by hollow fiber MGA process.

Gas type	Membrane	Porosity (%)	Absorbent solution	Gas mixture	Absorption flux (mol/m^2 s)	Ref.
Carbon dioxide (CO_2)	Polyvinylidene fluoride (PVDF)	46	Nanofluid of methyldiethanolamine (MDEA)+carbon nanotube (CNT)	CO_2/N_2 (20/80)	1.14×10^{-3}	[86]
	Mixed matrix PVDF+hydrophobic modified SiO_2 nanoparticles	79	Water	Pure CO_2	3.24×10^{-3}	[87]
	Ceramic (Al_2O_3)	43	Monoethanolamine (MEA)	CO_2/N_2 (20/80)	6.5×10^{-3}	[88]
	Non-porous poly dimethylsiloxane (PDMS) composite	–	MEA	Flue gas (CO_2 12.3%)	2.3×10^{-3}	[89]
	Polypropylene (PP)	40	Ionic liquids	CO_2/N_2 (15/85)	1.2×10^{-4}	[90]
	PP (3 M Liqui-Cel™)	–	Potassium glycinate amino acid salt	CO_2/N_2 (10/90)	2.27×10^{-4}	[91]
	Mixed matrix PVDF+superhydrophobic silica nanoparticles	39	Soybean-based solvent containing amino acid salts	CO_2/N_2 (12/88)	1.8×10^{-4}	[92]
	Hydrophobic coated ceramic	–	MEA	CO_2/N_2 (13/87)	4.08×10^{-3}	[93]
	Polytetrafluoroethylene (PTFE)	50	3-diethylaminopropylamine	CO_2/air (20/80)	4.0×10^{-3}	[94]
	Hybrid PVDF–HDTMS (hexadecyltrimethoxysilane)	76	Diethanolamine (DEA)	CO_2/N_2 (19/81)	2.23×10^{-3}	[95]
	Fluorinated TiO2/PVDF composite	89	MEA	Pure CO_2	12.7×10^{-3}	[96]
	Alkylated polyacrylonitrile (PAN)	89	Distilled water	Pure CO_2	1.9×10^{-3}	[97]
	PTFE	50	Dimethylethanolamine (DMEA)+ Piperazine (PZ)	CO_2/N_2 (15/85)	1.55×10^{-3}	[98]
	Hydrophobic PP-methyl grafted silica nanoparticles (CH_3SiO_2) composite	49	MEA	Pure CO_2	1.4×10^{-3}	[99]
	PVDF+ 8% phosphoric acid	–	Distilled water	CO_2/N_2 (19/81)	1.31×10^{-3}	[100]
	Superhydrophobic polyether ether ketone (PEEK)	–	Activated K_2CO_3	CO_2/N_2 (13/87)	2.5×10^{-3}	[101]
Nitrogen oxides (NOx)	Polysulfone (PSF)	–	$H_2O_2 + HNO_3$	NOx/air	8.3×10^{-7}	[102]
	Superhydrophobic PP	–	$H_2O_2 + HNO_3$	NOx/air(600 ppm)	3.1×10^{-4}	[103]
	PP	45	NaCl (5 wt%)+ 0.2 wt% H_2O_2	NO/N_2 (185 ppm)	1.59×10^{-4}	[104]
	PP	–	HNO_3 (0.5 M)+ 0.5 wt% H_2O_2	N_2O/air (1500 ppm)	2.2×10^{-6}	[105]
	PTFE	52	NaOH (0.5 M)	$N_2 + CO_2 + NO_2$	0.9×10^{-4}	[106]
	PVDF	–	Na_2SO_3 (0.2 M)	$NO_2 + N_2$ (200 ppm)	1.8×10^{-3}	[107]
Sulfur dioxide (SO_2)	Mixed matrix PVDF+MOFs	83.45	NaOH (0.625 M)	$SO_2 + N_2$(7346 ppm)	1.09×10^{-3}	[108]
	Commercial PP (Liqui-Cel™)	40	NH_4OH (0.05 M)	SO_2 (2000 ppm)+ 15% CO_2 $+N_2$ balance	–	[109]
	Multichannel ceramic	–		SO_2 (1000 ppm)+ N_2	3.3×10^{-5}	[110]
	Superhydrophobic polyethersulfone-silica (PES/PES-SiO2)	–	Ethanolamine (0.625 M)	$SO_2 + N_2$	1.24×10^{-3}	[111]
	Hydrophilic ceramic	–	NaOH (0.4 M)	SO_2 (1000 ppm)+ 10% CO_2 $+ N_2$ balance	5.5×10^{-5}	[112]
	Commercial PP	–	CaO (0.001 M)	SO_2 (100 ppm)+CO_2 15%+N_2 balance	1.9×10^{-4}	[113]
	Hydrophobic ceramic (Al_2O_3)	40	Water	$SO_2 + N_2$ (1000 ppm)	4.7×10^{-5}	[114]
	Hydrophilic ceramic (Al_2O_3)	35	NaOH (0.5 M)	$SO_2 + N_2$ (1000 ppm)	8.05×10^{-5}	[115]
Volatile organic compound (VOC)	Hydrophobic PP	45	N-formylmorpholine/H_2O (50/50)	Benzene+N_2(10.86 mg/l)	1.5×10^{-3}	[116]
	Commercial PP (Celgard X-30)	–	Silicone oil	Methyl ethyl ketone (MEK)+ air (41.2 ppmv)	4.9×10^{-6}	[117]
	Commercial PP (Celgard X-10)	83.2	Silicone oil	Acetone+N_2(993 ppmv)	1.27×10^{-4}	[118]
	Commercial PP (Celgard X-10)	62.5	Silicone oil	Methylene chloride+air (999 ppmv)	2.6×10^{-6}	[119]
Biogas	In-house made PTFE	44.22	Deionized water	CH_4 /CO_2 (60/40)	7.2×10^{-4}	[79]
	Commercial-grade PVDF	70	K_2CO_3 (0.025 M) + potassium sarcosine (0.025 M)	CO_2 40%+ H_2S (300 ppm)+ CH_4 balance	2.45×10^{-4}	[80]
	Commercial dense skin poly (phenylene oxide) (PPO)	–	Distilled water	CO_2/CH_4 (30/70)	3.0×10^{-5}	[81]
	Composite fluorinated TiO_2-SiO_2/PVDF	–	MEA solution (1 M)	CO_2/CH_4 (40/60)	8.0×10^{-3}	[82]
Natural gas	Commercial PVDF	56.3	MDEA solution (40 wt%)	CO_2/CH_4 (70/30) (P = 60 bar)	1.18×10^{-3}	[120]
	PTFE	–	Aqueous mixture of MDEA+PZ	CO_2-rich natural gas (P = 54 bar)	7.9×10^{-4}	[121]
	polytetrafluoroethylene-co-perfluoroalkylvinylether (PFA)	–	K_2CO_3 solution (0.5 M)	CO_2 (5%) +H_2S (2%) + CH_4 balance (P = 50 bar)	7.3×10^{-4}	[122]
	PVDF	39.2	MEA solution (0.5 M)	CO_2/CH_4 (10/90)	1.5×10^{-3}	[123]
	Commercial PP (Celgard X-50)	40	RO water	CO_2/CH_4 (10/90)(P = 5 bar)	1.9×10^{-4}	[124]

these issues, numerous attempts were made including new membrane material synthesis, optimization of membrane properties and development of new liquid absorbents.

It is worth mentioning that upgrading of biogas by MGA process using various membranes and liquid absorbents has been an intensive research focus in recent years [79–82]. In fact, biogas as a renewable energy resource has attracted considerable attentions in carbon reduction and controlling greenhouse gas emission [83]. Generally, biogas is produced through anaerobic digestion of municipal and industrial wastewater in which it contains CH_4 (53–70 vol%) and CO_2 (30–47 vol %) as well as trace of H_2S, NH_3, N_2, H_2, water vapor and volatile components [84]. Moreover, treatment of natural gas (NG) as the fastest growing energy resource through MGA process has attained significant interests. Indeed, the presence of significant amount of CO_2 in NG can cause serious issues such as corrosion of pipeline and equipment, reduction of calorific value and increment of transportation cost. Since the pressure of raw natural gas is in the range of 20–70 bar [85], several studies on high pressure MGA process for acid gas capture can be found in the literature. Table 2 summarizes recent studies related to the use of hollow fiber-based MGA process for greenhouse gas capture, biogas upgrading and NG treatment.

As can be seen from Table 2, most of the studied are focused on CO_2 capture as the main greenhouse gas. Although SO_2 and NO_x have also been responsible for air pollution, acid rain, urban smog and human health, only limited research can be found on their removal by MGA processes in the open literature. In addition, the release of VOCs from various petroleum and natural gas processes into air can affect public health and environment due to their carcinogen effect. However, there is no recent research on removal of VOCs through the MGA processes. Hence, more studies are required to improve MGA process for removal of VOCs, SO_2 and NO_x from gas streams, in addition to CO_2. In order to improve separation efficiency of greenhouse gases, development of ideal membrane structure as well as liquid absorbent are equally important.

Typically, the liquid absorbents used in the conventional contactors for CO_2 absorption are also found to be suitable for the MGA process provided the absorbent exhibits high CO_2 solubility and good regeneration property. Indeed, a stable long-term operation of membrane contactor can be achieved by using the liquid absorbents that have high compatibility with the membrane material. Previous studies have shown that alkanolamine solutions which is commonly used as CO_2 absorbents can react with hydrophobic polymeric membranes which affects the membrane structure, causing membrane wetting and deteriorated CO_2 absorption [125–127]. To address this issue, liquid absorbent with high surface tension and low viscosity is required to minimize membrane wetting and pressure drop during the process. Various physical and chemical liquid absorbents such as water, alkanolamine solutions, ionic liquids, potassium carbonate, amino acid salts, ammonia solution and recently nano-fluids have been used in the membrane contactors for CO_2 capture. Details of this subject topic can be found elsewhere [15,77].

As for the membrane development, it can be seen that hydrophobic polymeric and ceramic membranes are commonly used for the MGA processes. The techniques for development of the membrane structure are summarized in Table 3. In addition to surface hydrophobicity, other membrane characteristics such small pore size, high porosity, significant thermal and chemical stability are required to achieve a long-term stable MGA operation with high separation efficiency. Several attempts have been made to fabricate porous membranes with enhanced properties including phase-inversion control, thermal and stretching methods, composite and mixed matrix fabrication and surface modification.

3.2. *Water and wastewater treatment*

In recent years, much attention has also given to the potential use of hollow fiber membrane contactor process for water/wastewater treatment as an alternative solution to resolve not only the issue associated with water shortage but also to recover important resources such as

Table 3
Summary of techniques for development of the membrane structure used in MGA process.

Technique	Remarks	Ref.
Non-solvent additives in polymeric solution	Propionic acid and lithium nitrate were used as additives in PVDF solution to control phase-inversion rate. The addition of propionic acid was found to improve the PVDF membrane structure, leading to the highest CO_2 absorption flux.	[128]
	Glycerol was used as phase-inversion promoter to develop porous polyetherimide (PEI) and PVDF membranes. Using water as absorbent, the PVDF membrane showed a higher CO_2 flux compared to the PEI membrane.	[24]
	Phosphoric acid, methanol and lithium chloride were used as non-solvent additives in the PEI solutions. Phosphoric acid could improve porosity and wetting pressure of the membrane, leading to CO_2 absorption flux as high as 2.7×10^{-2} mol/m^2 s.	[129]
	Porous polysulfone (PSF) hollow fiber membranes were fabricated by phase-inversion process using low molecular weight additives.; Precipitation rate of the polymeric solution was followed the trend of glycerol > acetic acid > PEG200 > ethanol.; As a comparison, glycerol resulted in an improved structure with a higher wetting resistance and CO_2 flux (1.5×10^{-3} mol/m^2 s).	[130]
Surface modification	A wet-chemical hydrophobic modification using octadecylamine for poly amide-imide (PAI) hollow fiber membrane was performed in order to alter membrane morphology and structure.; The modification resulted in 30° increase in the membrane water contact angle, reaching 106.5°.; The resultant membrane also exhibited a stable long-term CO_2 absorption performance for up to 15 days using 2 M sodium taurinate aqueous solution.	[131]
	Heptadecafluoro-1,1,2,2-tetrahydrodecyltrimethoxysilane was used as a coating layer on the ceramic hollow fiber membrane which increased its water contact angle to about 126°.; The modified membrane was able to treat 0.175 Nm^3/h of flue gas with 90% CO_2 removal efficiency.	[132]
	Surface modification of poly ether ether ketone (PEEK) membrane was performed with a functional perfluoro oligomer. Surface modification improved membrane's CO_2 capture to about 90% with its purity higher than 95% in one stage process when both activated MDEA and activated K_2CO_3 were used as solvents.	[101]
	Silicon rubber was coated on the inner surface of the PEI hollow fiber membranes with the aim of increasing its surface contact angle and wetting resistance. An internal surface coating by 0.5% PDMS resulted in a maximum CO_2 flux of 2.5×10^{-3} mol/m^2 s.	[133]
	Hybrid PVDF-hexadecyltrimethoxysilane (HDTMS) membranes were fabricated to enhance surface hydrophobicity.; The membrane exhibited a super-hydrophobic structure with DEA contact angle of 150°.; By adding 1.5 wt% of HDTMS in the polymeric solution, a maximum CO_2 flux of 2.23×10^{-3} mol/m^2 s was achieved.; The CO_2 flux of the improved membrane decreased by 17% after 17 days of the operation and then remained stable.	[95]
	Surface modifying macromolecule (SMM) was blended with PVDF to improve surface hydrophobicity of the hollow fiber membranes.; Using 2 wt% SMM, the membranes showed large mean pore size and high hydrophobicity in which a stable long-term CO_2 absorption operation was found during 140 h.	[134]
		[135]

(*continued on next page*)

Table 3 (*continued*)

Technique	Remarks	Ref.
Dense composite fabrication	A thin skin selective layer (Teflon AF®, PTMSP) was coated on a porous PP hollow fiber support.; The results showed that such coating could produce highly permeable membrane with very stable performance over long time (using MEA solutions) for CO_2 capture.	
	A composite hollow fiber membrane with an aminosilane-modified zeolitic imidazolate framework-8 (mZIF-8) based dense skin layer was fabricated.; mZIF-8 nanocrystals was first dispersed into PDMS solution before being depositing on a porous PVDF substrate.; The results showed that the long-term stability of the modified membrane for biogas upgrading during 15 days monitoring period.	[136]
	A composite membrane was fabricated by coating a dense layer of Teflon AF2400 on the top of porous PP support.; Surface contact angles of over 125° and 105° were found for NaOH (3.9 wt%) and MEA (30 wt%) solutions, respectively.; The maximum overall mass transfer coefficient for CO_2 absorption was found to be 34×10^{-4} m/s and 59×10^{-4} m/s for using NaOH (3.9 wt%) and MEA (30 wt%) solutions as the liquid absorbent, respectively.	[137]
Mixed matrix fabrication	ZSM5 zeolite-filled PVDF mixed matrix membranes (MMMs) were fabricated by a wet spinning process.; The gas permeability, surface roughness, wettability resistance, and mechanical stability of the membranes were considerably improved upon nanomaterials incorporation.; For the membrane modified by 5 wt% ZSM5, its CO_2 flux was ~ 175% higher than the neat PVDF membrane.	[138]
	A superhydrophobic polyethersulfone-silica (PES/PES-SiO_2) mixed matrix membrane was fabricated by a spin coating method.; SiO_2 nanoparticles was found to enhance hydrophobicity of PES membrane, increasing its water contact angle up to 159°. The improved anti-wetting property enhanced the membrane's SO_2 absorption flux and SO_2 removal efficiency, reaching 1.24×10^{-3} mol/m^2 s and 63.8%, respectively.	[111]
	SiO_2 nanoparticles modified with hexamethyldisilazane (HMDS), dimethyldichlorosilane (DMDCS) and PDMS were respectively used to fabricate MMMs for CO_2 absorption.; The modified membrane demonstrated 77.6% structural porosity, high wetting resistance and superior thermal and mechanical stability compared to the pristine PVDF membrane.; Although the CO_2 absorption flux of the MMMs could not challenge the 2.84×10^{-4} mol/m^2 s flux of the pristine PVDF membrane, they demonstrated better selectivity.	[39]
	Cu@ 4 A as filler was used to fabricate PVDF MMM contactor.; The Cu@ 4A-loaded MMM was reported exhibit 10 times higher SO_2 adsorption capacity than the neat PVDF membrane.; When 40 wt% Cu@ 4 A was used to modify the membrane, the highest SO_2 removal efficiency (73.6%) and SO_2 absorption flux (9.1×10^{-4} mol/m^2 s) was able to achieve.	[139]
	Metal organic frameworks (MIL-101(Cr)) was found to be potential to improve PVDF membrane for high-efficiency SO_2 removal.; The resultant MMM showed competitive hydrophobicity with the water contact angle of 119°.; For the best performing membrane loaded with 15 wt% of MIL-101(Cr), the results showed promising SO_2 absorption performance with SO_2 flux and SO_2 removal efficiency of 1.09×10^{-3} mol/m^2 s and 89.51%, respectively.	[108]
Thermal and stretching method	Porous PTFE hollow fiber membranes were fabricated through a paste extruder followed by stretching and sintering processes.; The improved membrane showed mean pore size of 0.152 µm and overall porosity of 44.22%.; Using a woven membrane module for biogas upgrading, the CO_2 flux of 0.72×10^{-3} mol/m^2 s and removal efficiency of about 100% were found at gas and liquid velocity of 0.055 and 0.034 m/s, respectively.	[79]
	A porous PTFE hollow fiber membrane was fabricated by stretching and thermal methods.; The average pore diameter and overall porosity of 0.31 µm and 41.9% were found for the membrane, respectively.; Using MEA aqueous solution (1 mol/L), a stable CO_2 absorption flux of about 10.8×10^{-4} mol/m^2 s was achieved during 180 days of the continues operation.	[140]
	PP-CH_3SiO_2 composite hollow fiber membranes were fabricated by a thermally induced phase separation (TIPS) method.; The membrane presented water contact angle of 145°, mean pore size of 0.154 µm and porosity of 49%.; Using aqueous solution of MEA (30 wt%), the CO_2 flux reduction of about 26% was observed after 30 days of the operation where the CO_2 flux was 1.1×10^{-3} mol/m^2 s.	[99]
	Commercial PP hollow fiber membrane (Celgard® X50), which is fabricated through a melting extrusion process followed by annealing and stretching steps, was used for CO_2 absorption by aqueous MEA solution (1 M). The membrane presented mean pore size of 0.019 µm and overall porosity of 58.2%. During a long-term membrane contactor process (12 days), the CO_2 flux was decreased from 2.2×10^{-2} to 8.0×10^{-3} mol/m^2 s.	[141]

ammonia, methane, pharmaceutical substances, etc. from industrial/domestic wastewater [6]. MGA, MGS, MLLE and MD processes have been used for water/wastewater treatment which will be discussed in the following sections.

3.2.1. Ozonation of water and wastewater through MGA process

Since chemical oxidation can destroy the target pollutants present in water/wastewater, it can be practically applied for the conversion of emerging organic micropollutants into less harmful products [142]. The main types of oxidation processes for water/wastewater treatment can be summarized as [143]: (1) conventional processes without formation of reactive species (e.g., chlorine, chlorine dioxide and potassium permanganate); (2) processes at elevated temperature/pressure based on free radical reactions (wet oxidation and supercritical oxidation); and (3) advanced processes based on formation of hydroxyl radicals at ambient temperature/pressure (O_3/H_2O_2, Fe/H_2O_2, UV/H_2O_2 and UV/TiO_2).

Ozonation as an advanced oxidation process (AOP) for water/wastewater treatment has been successfully used in different applications such as disinfection, removal of organic compounds, color, odor/taste, and control of biofilm growth [142,144]. Since ozone (O_3) is produced on-site by passing dry compressed air or pure O_2 across a high voltage electrode, ozonation is a relatively high energy consuming process (around 10 kW/kg O_3) [145]. In addition, conventional ozonation devices such as bubble columns and packed beds possess drawbacks including large footprint related to low mass transfer of ozone to water/wastewater, and some operational problems (e.g., flooding, uploading, emulsion and foaming) related to the dispersion of ozone bubbles into the aqueous phase.

In order to develop industrial applications of the ozonation processes, some of the key issues that have to be addressed are high utilization of ozone in the contactor device, reduction of ozone production cost and destruction of ozone in the outlet gas [146]. Therefore, MGA process has been introduced as a suitable alternative for water/wastewater ozonation, which can solve the aforementioned issues by improving gas-liquid contact and mass transfer coefficient through a bubbleless operation.

Fig. 2 shows the schematic diagram of ozonation through a lab-scale MGA process using tubular membrane [147]. Ozone-resistant tubes and fittings were used to connect gas and liquid flow lines with membrane module. An inline ozone generator was applied to produce ozone from high purity oxygen. The ozone-oxygen mixture (less than 20% ozone) then flowed through the shell side of the module while wastewater flowed counter-currently through the tube side of the membrane by a diaphragm pump. Using non-porous PDMS membrane, mass transfer coefficient and molar flux of ozone were found to be 2.4×10^{-6} m/s and 1.1×10^{-5} mol/m^2 s, respectively.

Due to the importance of the membrane contactor technology for ozonation process, it gains a considerable attention in recent years. A few review articles have been published on different aspects of the membrane contactors for ozonation of wastewater [13,148]. The recent research on ozonation of wastewater through the MGA process is summarized in Table 4. As can be seen, hydrophobic polymeric and ceramic membrane contactors have been used for ozonation of water/wastewater. Due to high oxidative nature of the ozonation process, the durability of the commonly used polymeric membranes such as PVDF and PDMS can be a main challenge for a long-term operation. On the other hand, although the ceramic membranes have shown high thermal, chemical and mechanical resistances, a costly hydrophobic surface modification process is still required for their ozonation application. In addition, operating parameters such as temperature, pH, ozone concentration, gas flowrate and liquid flowrate play important role on the ozonation efficiency of the MGA process. Therefore, development of a durable membrane and optimization of the operating parameters are key factors for industrial ozonation of the MGA process.

3.2.2. *Oily wastewater treatment through MD process*

A large volume of oily wastewater is annually produced through various industries such as oil and gas, petrochemical, metallurgical and food processing. Therefore, development of cost-effective technologies for treatment of oily wastewater is required due to environmental impacts, water reuse, and oil recovery. Membrane separation processes such as microfiltration (MF) and ultrafiltration (UF) have been used as highly effective technologies for oily wastewater treatment [155–157]. However, membrane fouling due to surface adsorption and deposition of oil droplets is the main problem of these pressure driven processes. Severe surface fouling is irreversible and can cause rapid flux reduction, product quality deterioration, shorten membrane lifetime as well as increased energy consumption [158].

Compared to the pressure driven membrane processes, the MD processes have shown a lower fouling tendency due to their larger pore sizes and lower operating pressure. Since the rejection of contaminants in the MD process is based on chemical species volatility, it is possible to perform particular separation and desalination simultaneously [159]. In addition, low sensitivity to the feed concentration and moderate operating conditions are the most attractive characteristics of the MD process. Based on the design of the permeate side, four configurations (Fig. 3) have been proposed for MD to improve the system performance in terms of production and thermal efficiency [75]. Details of the MD configurations can be found elsewhere [25].

Despite several advantages of MD technology, it still associated with some drawbacks. High consumption of the thermal energy is a major problem for an industrial application of the MD process. However, the MD process can be coupled with low-grade energy sources such as waste heat, solar thermal energy and renewable energies in order to decrease the operational costs [160,161]. Other drawbacks of MD process are low permeation flux, sensitivity to temperature polarization, membrane wetting tendency, low separation of organic solvents with low surface tension and significant heat loss by conduction [159].

In recent years, the applications of MD have expanded to the areas beyond desalination such as wastewater treatment and resource recovery [162–164]. The distinct properties of MD technology make it an economical and sustainable challenger for oily wastewater treatment. The potential of MD technology has been acknowledged by several review papers for treatment of oil-containing feed streams [28,72,163,165,166].

For treatment of wastewaters containing oil and organic pollutants through the MD processes, fouling can occur due to strong hydrophobic-hydrophobic interaction between the foulant and the membrane [167]. Oily pollutants can attach on the hydrophobic membrane surface, which results in blocking of the membrane pores and subsequently fouling layer formation and permeation flux reduction. In order to increase oil-fouling resistance, the composite membranes with a hydrophobic substrate and an underwater super-oleophobic top surface have been developed for the MD processes. For this purpose, several methods have been used including dip-coating, chemical bath deposition (CBD), layer-by-layer (LBL) assembly, plasma treatment and electrospinning

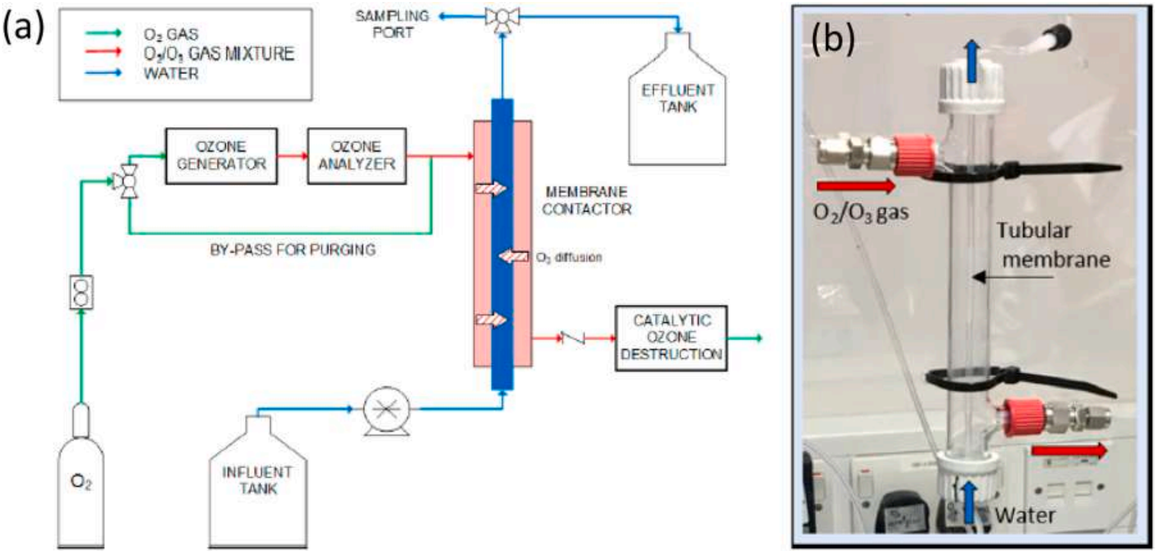

Fig. 2. (a) Schematic diagram of ozonation process using lab-scale membrane contactor and (b) photograph of a lab-scale tubular membrane with water passing through its lumen while O_2/O_3 mixed gas contacting its outer surface [147].

Table 4
Summary of recent research on ozonation of wastewater through MGA process.

Membrane	Operating condition	Ozone absorption flux (mol/m^2 s)	Ref.
Hydrophobized tubular ceramic (α-Al_2O_3)Pore size= 0.1 µmPorosity=NA	pH= 6; ozone concentration in the gas= 60 mg/L; temperature= 15 °C; gas velocity=NA; deionized water velocity= 0.05 m/s.	2.5×10^{-7}	[149]
PVDF hollow fiberPore size= 0.17 µmPorosity= 67%	pH= 2; ozone concentration in the gas= 50 mg/L; temperature= 20 °C; gas velocity= 0.024 m/s; water velocity= 0.5 m/s.	7.5×10^{-6}	[150]
PTFE hollow fiberPore size= 0.3 µmPorosity= 40%	pH= 7; ozone concentration in the gas= 40 mg/L; temperature= 28 °C; gas velocity= 0.12 m/s; water velocity= 0.9 m/s.	1.1×10^{-5}	[151]
PVDF hollow fiberPore size= 0.2 µmPorosity= 70%	pH= 7; ozone concentration in the gas= 50 mg/L; temperature= 25 °C; gas velocity= 0.01 m/s; water velocity= 0.5 m/s.	6.2×10^{-6}	[152]
Hydrophobized tubular ceramic (α-Al_2O_3) MWCO= 1000 kDa	pH= 7.8; ozone concentration in the gas= 40 mg/L; temperature= 20 °C; gas velocity=NA; contaminated water velocity= 0.14 m/s.	4.2×10^{-5}	[153]
Non-porous tubularPDMSPore size=NAPorosity=NA	pH= 7.1; ozone concentration in the gas= 110–200 g/m^3; temperature= 21 °C; gas flowrate = 100 mL/min; water velocity= 0.002–0.226 m/s.	1.1×10^{-5}	[148]
Commercial PDMSPore size=NAPorosity=NA	pH= 7; ozone concentration in the gas= 5 mg/l; temperature=NA; gas flowrate = 30 mg/h; water velocity= 0.015 m/s.	7.3×10^{-7}	[154]

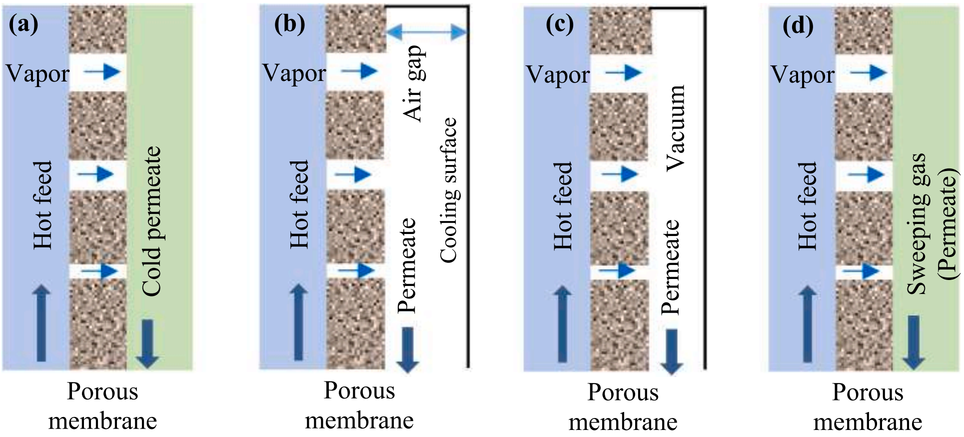

Fig. 3. Four main configuration of membrane distillation with feed (hot liquid) and permeate (cold liquid/cold vapor/sweeping gas): (a) DCMD, (b) AGMD, (c) VMD and (d) SGMD.

[163]. For example, as shown in Fig. 4, the composite membranes were prepared via electrospinning of hydrophilic polyacrylonitrile (PAN) nanofiber on a hydrophobic PTFE porous membrane [168]. The composite membrane showed the underwater oil contact angle > 150°, which was related to the re-entrant surface structure and surface hydrophilicity of the electrospun coating layer. The improved membrane indicated a stable permeate flux of around 15.2 L/m^2.h during the DCMD of a saline and oily feed (1000 mg/L crude oil), while a quick oil fouling and permeate flux reduction was observed for the hydrophobic PTFE membrane.

Tang et al. [169] used a facile breath figures templating (BFT) technique to develop novel composite membranes with hydrophobic PTFE substrate and a hydrophilic cellulose acetate (CA) coating with through-pores. Increasing CA concentration decreased membrane pore sizes but increased its porosity and underwater oleophobicity. For DCMD of a saline feed containing 1000 mg/L crude oil, a stable permeate flux of around 16.85 L/m^2.h and salt rejection of near 100% were achieved. Khan et al. [170] fabricated an anti-oil-fouling omniphobic polyethersulfone (PES) membrane using fluorinated silica nanoparticles (F-SiO_2 @PES) combined with perfluorodecyl triethoxysilane and PDMS. The prepared composite membrane presented an effective anti-oil fouling and anti-wetting performance during DCMD of oil-in-water emulsions. Flux reduction of 5–15% and oil rejection of > 99% was observed for various combinations of feed solutions.

On the other hand, Zhu et al. [171] prepared an asymmetrically Janus membrane with underwater super-oleophobicity and in-air super-hydrophobicity properties via coating of the hydrophobic silica nanoparticles on the hydrophobic PVDF substrate. The composite membrane presented an underwater super-oleophobicity with oil contact angle of 164° and an in-air super-hydrophobicity with water contact angle of 166°. The developed membrane demonstrated a stable condition during DCMD of oil-in-saline water emulsion with high permeate flux of 25 L/m^2.h and robust anti-oil fouling resistance. More studies on enhancement and performance of the composite membranes for MD of oily wastewater can be found in the review papers published elsewhere [28,163,165].

From the literature survey, MD processes have been applied for various oily water and wastewater treatment. The commercial hydrophobic membranes are widely used for wastewater treatment by the MD processes, in which the membrane fouling and permeation flux recovery are the main research focus. Integration of the pre-treatment processes with the MD processes can be a promising alternative to enhance the membrane performance. Although a stepwise washing procedure can be used for flux recovery and reuse, the implementation of continuous washing procedures for flux recovery is not practical. Therefore, modification of hydrophobic membranes and preparation of composite omniphobic and Janus membranes can be effective alternatives for treatment of oily wastewaters. Although the improved composite membranes have indicated durable anti-oil-fouling and wetting resistance, they in general showed lower permeation flux compared to the plain commercial membranes. Hence, more research on development of the membranes for MD of oily wastewater is required. Cost-effective and state-of-the-art approaches including the membrane materials and surface modification need to be considered with the possibility of the MD process scaling-up. In addition, long-term operations of MD for oily wastewater treatment need to be investigated in order to attain a

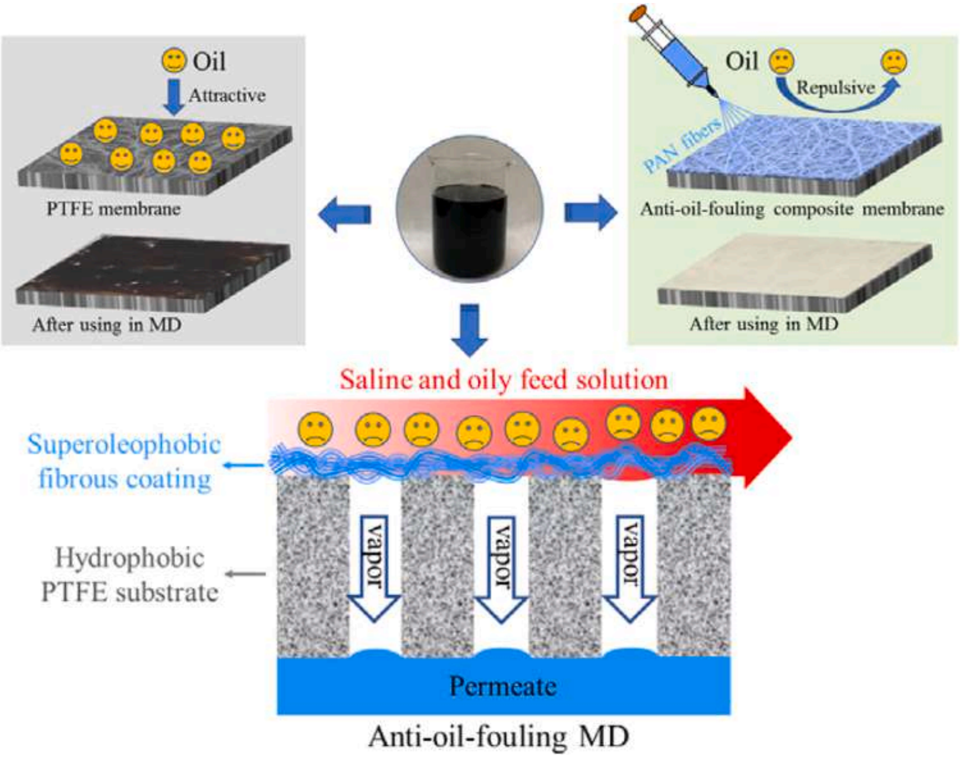

Fig. 4. Schematic illustration of anti-oil-fouling composite membrane for MD process of saline and oily wastewater treatment (The surface properties were improved by coating of superoleophobic PAN nanofibers) [168].

sustainable process.

3.2.3. Dyeing wastewater treatment through MD process

A large volume of water is annually consumed through textiles, cosmetics, food coloring, carpet and paper industries and all these activities generate a massive amount of dyeing wastewaters that can possess threat to the environment. Generally, dyeing wastewaters contain complex pollutants with high levels of chemical oxygen demand (COD) and biochemical oxygen demand (BOD) [172]. Indeed, it is really hard to treat dyeing wastewaters and the development of treatment techniques are still in progress. The traditional dyeing wastewater treatment techniques such as biological and physicochemical (coagulation, adsorption, radiation, oxidation and photocatalysis) processes have shown various weaknesses such as high cost, low separation efficiency, complexity and large amounts of sludge production [173–175]. In comparison to the traditional dyeing wastewater treatment techniques, membrane separation processes have attracted considerable attentions due to their easy operation and maintenance, modular design and high separation efficiency [176,177]. Especially, MD has gained interest of scientists for dyeing wastewater treatment due to its high rejection rate at low operating pressure. Moreover, dyeing wastewaters (especially those produced in textile industry) are usually released at

Table 5
Recent research on treatment of dyeing wastewater by the MD processes.

Membrane	MD process	Dye solution	Operating condition*	Permeate flux (kg/m^2 h)	Rejection (%)	Ref.
Commercial PVDF	DCMD	MB+NaCl+water	T_f= 60 °C; Tp= 20 °C; C_f= 100 mg/L; Q_f= 0.6 L/min; Q_p = 0.6 L/min; $t_{op.}$= 50 h.	22	100	[180]
Composite PVDF	VMD	MB+water	T_f= 50 °C; $P_{vac.}$= 31.3 kPa; C_f= 500 mg/L; Q_f= 0.85 L/min; $t_{op.}$= 100 h.	10.35	99.6	[181]
Commercial PTFE	AGMD	RB+NaCl+ SDS+water	T_f= 70 °C; T_p = 20 °C; C_f= 100 mg/L; Q_f= 0.38 L/min; $t_{op.}$= 20 h.	11	100	[182]
Commercial PP	DCMD	Reactive black+water	T_f= 60 °C; T_p = 20 °C; C_f= 30 mg/L; Q_f= 1.5 L/min; Q_p = 0.5 L/min; $t_{op.}$= 3 h.	22	100	[183]
PVDF	DCMD	RB5 +water	T_f= 90 °C; Tp= 20 °C; C_f= 100 mg/L; V_f= 0.034 m/s; V_p = 0.016 m/s; $t_{op.}$= 2.5 h.	6	99.8	[184]
TEOS crosslinkedPolystyrene	VMD	MB+NaCl+water	T_f= 30 °C; $P_{vac.}$= 250 kPa; C_f= 10 mg/L; Q_f=NA; $t_{op.}$= NA.	65	99.8	[185]
PDMS/PVDF hybrid electrospun	DCMD	MB+water	T_f= 60 °C; T_p = 20 °C; C_f= 100 mg/L; Q_f= 0.5 L/min; Q_p = 0.5 L/min; $t_{op.}$= 24 h.	25	100	[186]
PVDF-Cloisite 15 A nanocomposite	DCMD	Textile wastewater	T_f= 90 °C; Tp= 25 °C; C_f= 346–526 Pt/Co; V_f= 0.023 m/s; V_p = 0.002 m/s; $t_{op.}$= 40 h.	15	89	[178]
Commercial PTFE	DCMD	Polyester dyeing wastewater	T_f= 60 °C; T_p = 20 °C; C_f= 8520 COD; Q_f= 1.5 L/min; Q_p = 0.5 L/min; $t_{op.}$= 3 h.	16	98.5	[187]
PEI surface coated by Zonyl® BA	AGMD	MB+water	T_f= 65 °C; T_p = 5 °C; C_f= 100 mg/L; Q_f= 0.2 L/min; and $t_{op.}$= 8.3 h.	7.5	98	[188]
PEI surface coated by PDMS	SGMD	MB+water	T_f= 60 °C; T_p = 20 °C; C_f= 80 mg/L; Q_f= 0.2 L/min; and $t_{op.}$= 135 h.	18.3	100	[26]

high temperature, making the MD process a possible economic option for the treatment [178].

Ideally, non-volatile dyes can be completely rejected by the MD process when the membrane is in the non-wetting mode. Since dyeing wastewaters would contain a mixture of dyes, the MD processes are generally applied for water recovery rather than the dyes recovery [179]. It is worth mentioning that type of dye can affect the MD separation performance in which various dyes such as anionic, cationic, reactive and direct have been investigated.

Form the literature, it was found that few research have been conducted in treatment of dyeing wastewater through the MD processes, where the DCMD process has been the main focus. Table 5 shows the recent research on treatment of dyeing wastewater by the MD processes. As can be seen, most of the works have been conducted using commercial membranes such as PP, PTFE and PVDF membranes due to their reasonably good hydrophobic characteristics and availability in the market. In addition, surface modification of the membranes by coating, cross-linking and composite preparation methods have shown improved results for dyeing wastewater treatment.

As an energy saving water treatment process, MD has potential to recover thermal energy of waste streams and low cost sources. Since textile industries are using thermal energy in various operations, the MD process can be integrated into the plant to recover waste heat for wastewater treatment. Furthermore, in comparison with traditional wastewater treatment processes, the MD processes do not require chemical substances during the treatment operation.

As discussed in the previous section, membrane fouling and wetting are main problems in the development of the MD process for wastewater treatment. Hence, development of the membrane structure and integration with a pre-treatment process are necessary for dyeing wastewater treatment by the MD processes. In addition, in order to minimize the effects of thermal and concentration polarization as well as fouling, some of the strategies that can be considered are increase mass transfer area per unit volume (compactness) and design a proper membrane module configuration (especially hollow fiber).

It should be noted that the technical part of the MD processes has been widely investigated for dyeing wastewater treatment. However, there is still a lack of an economic analysis to demonstrate clear cut advantages over the conventional wastewater treatment processes. High demand of thermal energy and membrane replacement can be the main economic disadvantages of the MD process, while water reuse and heat recovery from the hot effluents are some economic advantages.

3.2.4. *Phenols removal from wastewater through MLLE process*

Phenol and phenolic compounds are usually found in wastewater of various industries in different range of concentration such as oil refineries (6–500 ppm), coking operations (28–3900 ppm), coal processing (9–6800 ppm) and petrochemicals (2.8–1220 ppm) [189]. Phenolic compounds are highly toxic and environmental regulations require their concentration at < 1 ppm upon discharge [190]. Various separation methods like solvent extraction, adsorption and biological treatment have been developed for removal of phenols from wastewaters. These traditional methods have shown some limitations such as extensive use of organic solvents, emulsion formation, organic phase carryover, back mixing, high energy consumption and production of secondary pollutants [191]. Therefore, dispersion-free extraction method through a MLLE process which is known as pertraction has been introduced in order to overtake these drawbacks.

In MLLE process, the interface between the aqueous and the organic phases is immobilized by using a hydrophobic membrane. However, the membrane creates an extra resistance to the mass transfer process. By using thousands of fine hollow fibers in a compact membrane contactor module, the mass transfer resistance can be compensated by providing a large surface area per unit volume [192]. An industrial scale pertraction process has been installed in 1998 at the chemical firm - KoSa Netherlands BV in Vlissingen, The Netherlands. The process was developed to extract an aromatic compound from the wastewater [193]. A Liqui-Cel® membrane contactor module containing PP hollow fiber membranes (see Fig. 5) was used for treating 15 m^3/h wastewater produced from a chemical reactor where a removal efficiency of about 95% was achieved.

The extractant solvents such as alcohols [194,195], alkylamines [196], trialkylphosphine oxides [197] and tributyl phosphate (TBP) [198] have been used in the liquid–liquid extraction processes for removal of phenolic compounds from water and wastewater. Ideally, extractant solvents should present properties such as great extraction performance, low water solubility, low volatility and low cost [199]. The first work on extraction of phenol from water by methyl isobutyl ketone (M1BK) solvent was reported by Prasad and Sirkar in 1988 [200]. The authors employed hydrophobic PP hollow fiber (Celgard X-20) and hydrophilic regenerated cellulose hollow fibers (CUPROPHAN) membrane contactors to perform the separation. It was found that the height of transfer units (HTU) value of the membrane contactor was comparable to the lowest reported for conventional extractors. Although membrane extraction has shown advantages over traditional solvent extraction methods, the works on removal of phenol by MLLE process are limited in the literature. A summary of recent studies on phenol removal from wastewater by MLLE process is presented in Table 6. As can be seen, most of the studies have been conducted by microporous hydrophobic PP hollow fiber membrane extractors. A high phenol removal efficiency was achieved by using organic solvent extractants such as TBP, 1-decanol and pentanol. It can be said that a cost-effective MLLE process can be achieved for phenolic substances removal from wastewater when a developed membrane structure, a proper module configuration and solvents with high capacity are applied.

3.2.5. *Ammonia removal from wastewater through MLLE process*

Ammonia (or ammonium ion) is a common pollutant which is considered toxic even at low concentrations. The presence of ammonia in industrial, domestic and farming wastewater is a major environmental concern because of its accumulation in water bodies that could cause eutrophication and oxygen depletion. Typically, municipal wastewater and landfill leachate might contain 0–200 and 1000–4000 mg/L ammonium, respectively [204]. Higher concentration of ammonium (2000–7000 mg/L) could be found in effluent discharged from food and pharmaceutical industry [205]. For the wastewater discharged from the manufacturing process of uranium dioxide (UO_2) kernel, ammonia concentration could be as high as 25,000 mg/L [205].

Xu et al. [206] reported the practicability of using MLLE process to recover ammonia from an industrial wastewater discharged from a

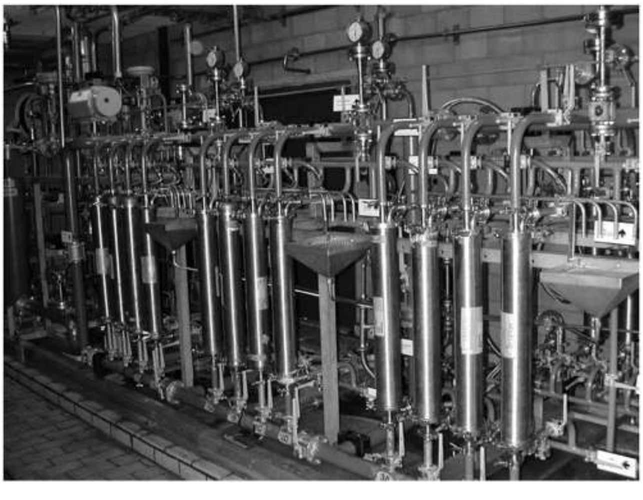

Fig. 5. Industrial pertraction installation used for aromatic compound removal using Liqui-Cel® membrane contactor module [193].

Table 6
Summary of recent research on phenol removal from wastewater by MLLE process.

Membrane	Phenol in feed water (mg/L)	Extraction phase	Operating conditions	Phenol removal (%)	Ref.
PP hollow fiber (Memtec, Australia)	1000	50% (v/v) Pentanol/ xylene	Feed pH= 5.9; $Re_{aq.}$= 2.98; $Re_{org.}$= 3.61; T = 20.5 °C; and P = 100 kPa.	88	[201]
electrospun TPU/ PDMS/ PMMA	2000 (10 g/L NaCl solution)	DI water	Feed pH= 5.6; Q_{feed}= 0.9 L/h; $Q_{Ex.}$= 1.26 L/h; and T = 25 °C.	24.9	[202]
PP hollow fiber	1000	10% (v/v) TBP/ Shellsol 2046 kerosene	Feed pH= 6.5; $Q_{aq.}$= 3.49 mL/ min; $Q_{org.}$= 5.16 mL/ min; and T = 25 °C.	74	[199]
electrospun PDMS/ PMMA	2000 (10 g/L NaCl solution)	DI water	Feed pH= 5.6; Q_{feed}= 0.9 L/h; $Q_{Ex.}$= 1.62 L/h; and T = 20 °C.	21	[203]
PP hollow fiber (Celgard X50 Liqui-Cel®)	800	Emulsion of NaOH solution (10% wt.)/ 1-decanol	Q_{feed}= 2440 mL/ min; $Q_{Ex.}$= 1800 mL/ min; T = 40 °C;	99.4	[191]

catalyst factory. Results showed that the membrane process attained an average NH_3 removal rate of 93.5% during 5-month continuous operation for the feed containing 500–1500 mg/L NH_3-N. In this study, sulfuric acid solution with concentration of > 0.5 M was used as absorbent and the wastewater before feeding into MLLE process was subject to pH adjustment and prefiltration (10 μm) to minimize membrane fouling tendency. In a separate work, Amaral et al. [207] demonstrated the efficiency of commercial PP membrane (X50 from Liqui-Cel®) in recovering ammonia from landfill leachate. After pre-treating the leachate with microfiltration membrane (pore size: 0.4 μm), the sample with total ammonia nitrogen of 881 mg/L and chemical oxygen demand (COD) of 1389 mg/L was fed into the membrane contractor. The authors were able to achieve 99.9% of ammonia removal rate with the MLLE process using 0.1-M sulfuric acid solution as extraction. The treated sample was reported to meet the effluent standards required by the local regulations. The experiment also showed that ~80% of ammonia from the wastewater could be recovered in the extraction solution, offering a solution for fertilizer production.

In 2020, a full-scale ammonia recovery process using MLLE system was established in Germany using Liqui-Cel® membrane modules with total effective surface area of 3520 m^2 and treatment capacity of up to 30 m^3/h [208]. Based on the results obtained from initial phase of operation, the membrane process was found to be applicable for an efficient nitrogen elimination from municipal process water with high nitrogen removal efficiency (>95%). However, the authors highlighted several key issues that needed particular attention in order to ensure long-term stable membrane. These included further optimization of process conditions and appropriate cleaning protocol.

In fact, based on the lab-scale findings from previous studies, one can see the impacts of process conditions on the efficiencies of MLLE during ammonia removal. For instance, Ashrafizadeh and Khorasani [209] and Ahn et al. [210] reported that increasing the pH of ammonia feed solution and increasing its velocity were beneficial in improving ammonia removal rate. Hasanoglu et al. [211] on the other hand found that higher operating temperature of ammonia feed solution could enhance kinetics of the reaction and of transportation through the membrane which led to efficient ammonia removal. They also reported that the best circulation configuration for the hollow fiber contactors was to deliver feed solution in the shell side and receiving solution in the lumen side of the membranes. Reig et al. [212] also agreed that the feed solution containing ammonia had to be passed through the membrane shell side while receiving acid solution to be delivered through fiber lumen. They found that this flow orientation worked even better when the membrane module was arranged in vertical position. With respect to the properties of receiving solution, it is normally reported that that the concentration of sulfuric acid solution did not play significant roles in impacting the efficiency of MLLE process.

In addition to process conditions, the characteristics of membranes used for MLLE process could play a critical role in influencing the process outcomes. Lauterböck et al. [213] evaluated 22 flat sheet membranes from various manufacturers (e.g., Pall, Millipore, Koch Membrane and Sartorius) and of different materials (e.g., PP, PTFE, PVDF and PES) with the aim of providing better insights towards the mass transfer behaviors of ammonia in the hydrophobic membranes. The common membrane parameters that were taken into consideration in the study were pore size, bubble point, contact angle, breakthrough pressure, airflow and thickness. The experimental findings revealed that membrane resistance played significant role in limiting mass transfer of ammonia and the observed transfer coefficient could vary by more than the power of 10 (between 0.04 × 10^{-3} and 24.59 × 10^{-3} m/h). Although the authors found that the membrane characteristics determined in this study were suitable to estimate the free ammonia mass transfer coefficient, variation in the transfer coefficient might still take place and this can be explained by the fact that some membrane structural and morphological properties are not able to be directly measured.

Compared to the treatment of dilute ammonia streams (<25 mg/L NH_4^+) [214,215], Liu and Wang [205] reported that radioactive wastewater produced in the manufacturing process of UO2 kernel could be a more practical source of recovering ammonia as this wastewater could contain as high as 25,000 mg/L ammonia. Liu and Wang [205] used self-fabricated PVDF hollow fiber membrane (Average pore size: 0.01–0.13 μm, Porosity: 76%) and assessed its performance in treating simulated radioactive wastewater containing 7750 mg/L NO_3^-, 6000 mg/L urea and 10,000 mg/L tetrahydrofurfuryl alcohol (THFA). The results indicated that ammonia removal rate could reach > 90% in 120-min operation using the simulated wastewater with pH ~11.5. It is important to note that the presence of co-existing compounds such as THFA and urea played almost no effect on the membrane separation against ammonia.

The potential of using MLLE process in treating high concentrated ammonia solution was also demonstrated in the work of Hossain and Chaalal [204]. Instead of using acid stripping solution, the authors used an environmentally friendly sunflower oil containing di(2-ethyhexyl) phosphate (D2EHPA) as carrier to recover ammonia from feed solution with highly concentration ammonia (25,600 mg/L). The experimental results showed that although the commercial hollow fiber membrane (Celgard X-30 type) was able to recover ammonia from the feed, its removal efficiency was rather low (43–73%). This could be due to the low absorption efficiency of edible oil compared to the commonly used sulfuric acid solution.

To further enrich the content of ammonia nitrogen in the wastewater, Yan et al. [216] integrated hollow fiber membrane extraction with water splitting with the aim of achieving highly efficient ammonia capture. The mechanisms and experimental setup of ammonia separation and recovery using this integrated system are shown in Fig. 6(a) and (b), respectively. In Fig. 6(c), it is found that ammonic capture ratios for membrane extraction could consistently reach 99% for 10 different batches of operation. The $C_{NH_4^+-N}$, on the other hand, in the basified wastewater could be as low as 10 mg/L compared to the initial concentration of 5000 mg/L. With respect to pH, it is reported that the wastewater exhibited reduced pH (from > 1 to ~9) after membrane treatment. The authors explained that the captured ammonia in the

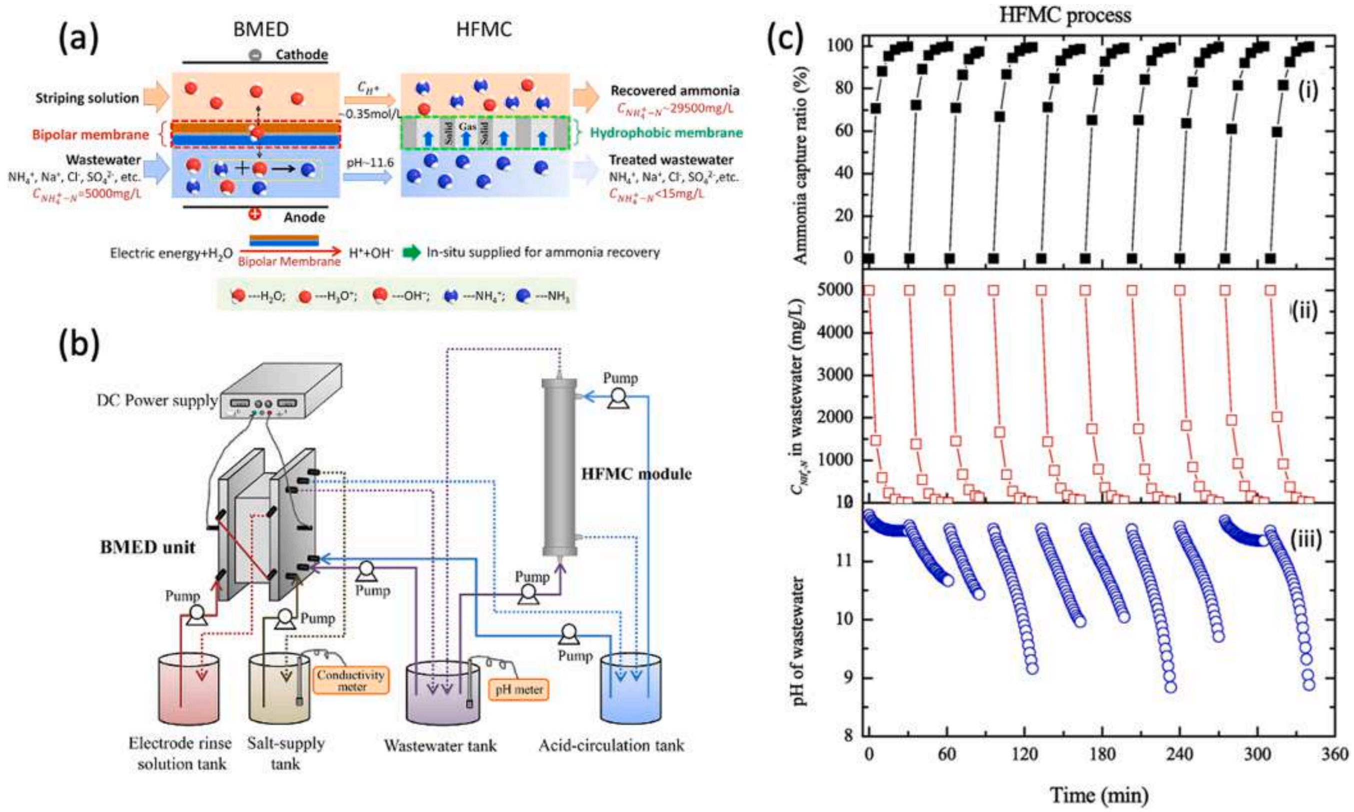

Fig. 6. (a) Mechanisms of integrated bipolar membrane electrodialysis (BMED) and hollow fiber membrane contactor (HFMC) process for ammonia separation and recovery, (b) Experimental setup of BMED and HFMC for ammonia separation and recovery and (c) results of ten batches of the ammonia capture process as a function of time, (i) ammonia capture ratio, (ii) $C_{NH_4^+-N}$ in wastewater and (iii) pH of the wastewater [216].

$(NH_4)_2SO_4$ form with a high concentration can be potentially utilized as nitrogen fertilizer or in the field of electrical power generation.

Another integrated process that has been recently demonstrated for ammonia recovery and liquid fertilizer production is ion-exchange coupled with hollow fiber liquid-liquid membrane contactors. In this work, Reig et al. [212] evaluated different operational parameters e.g., membrane position (vertical or horizontal), type of acid solution, flow rate of each stream, number of membrane contactors, etc. on how they could recover ammonia economically and efficiently for the production of liquid fertilizers. Under the optimized conditions, this integrated process was able to achieve 96% ammonia recovery rate and produce liquid fertilizer with N composition (from $NH_4{}^{+}$) as high as 4.6%. Furthermore, the ammonia-treated stream with NH_3 level of ~200 mg/L could be reused for the regeneration of granular zeolite.

3.2.6. *Removal of heavy metal ions/radioactive materials from wastewater through MLLE process*

The use of MLLE process for heavy metal separation and recovery was studied as early as 1980 s. In 1984, Kim [217] employed microporous membranes made of different types of polymeric materials (i.e., PA, PSF, PP and fluoropolymer) with MWCO ranging from 2000 to 100, 000 Da as an interface between a copper aqueous solution and organic solvent containing liquid ion exchanger (LIX 64 N). The author explained that this process was potential to selectively recover metals from ore leachates or metal-containing wastewater. Nevertheless, in this pioneering work, the author reported that it was still difficult to carry out an accurate economic analysis on the use of the membrane solvent extraction system for large-scale hydrometallurgical operation based on the stage of development at that time.

In 2009, Witek-Krowiak et al. [218] presented a new concept of membrane contactor which was enhanced by micellar solubilization. The author utilized 0.0106-m^2 PSF membrane with MWCO of 20 kDa for simultaneous removal of Cr(III) ions and p-cresol from water solution and found that the proposed method was still efficient for a low level of solute concentration in water, i.e., 70 mg/L p-cresol and 100 mg/L Cr (III) ions. When 30-mM sodium dodecyl sulfate (SDS) was introduced into p-cresol/copper solution, almost complete retention of Cr(III) ions was able to achieve. The promising result was caused by the trapping of the Cr(III) ions at the surface of the micelles due to electrostatic interactions. Meanwhile, the rejection coefficient of p-cresol was 75% under the same conditions.

Using MLLE process that was based on self-developed modified polyether ether ketone (PEEK-WC) hollow fiber membranes, Be et al. [219] could extract up to 99% Cr(VI) ions from aqueous solution containing $K_2Cr_2O_7$ using Aliquat-336 as carrier in kerosene. The developed membranes possessed an average pore size of 0.08–0.09 μm with porosity in the range of 70–80%. Although PP hollow fiber membrane could exhibit higher extraction values than that of sponge-like structure PEEK-WC membrane, it was suffered from high degree of swelling which possessed high risk of feed contamination by the organic phase and reduced performance stability. On the contrary, the hydrophilic PEEK-WC membranes was a better candidate for the Cr(VI) extraction with reduced dispersion phenomena of the organic phase into the aqueous one.

The use of toxic organic solvent during MLLE process is always the main concern for industrial applications. In order to address the issue, Hossain [220] employed sunflower oil as a non-toxic solvent to remove Cr(VI) ions from the ground water using Liqui-Cel® hollow fiber membrane. The reactive carrier used in the organic phase was Aliquat-336. Compared to Cr(III) ion, Cr(IV) ion is more toxic and could easily penetrate biological membranes. Furthermore, it is also a major part of chromium in waters. The experimental findings indicated that the distribution coefficient (DE) for Aliquat 336/sunflower oil was as good as those reported for systems using kerosene, showing its potential to replace the commercially used kerosene. When the process was applied to the groundwater samples at its natural pH (without addition of chemicals, ~94% of Cr(VI) ions were able to be rejected by a bench-scale hollow-fiber membrane contactor, leading to the production of treated water that can meet the local drinking water standard.

Using commercial PP hollow fiber membrane (Liqui-Cel®) manufactured by Celgard™, Younas et al. [221] successfully extracted Cu(II) ions from aqueous solutions employing widespread used trifluoro-acetylacetone (TFA) diluted in 1-decanol and methyl isobutyl ketone (MIBK) as organic solvents. In this work, copper-bearing aqueous phase of known Cu(II) concentration (3.15 and 15.75 mol/m^3) was contacted in counter current flow with TFA diluted in organic phase. It was found that the degree of extraction was mainly related to partition coefficient. Under the same conditions of concentrations, temperature and flowrates, MIBK was proved to be a proper diluent for TFA during Cu(II) extraction process. However, 1-decanol is preferred for Cu(II) extraction at higher temperature. By increasing temperature of the extraction system, not only the diffusion coefficient of solute improved (faster extraction) but it also enhanced the partition coefficient (efficient extraction).

Besides TFA, other studies reported that LIX64N [222] and di (2-ethylhexyl)phosphoric acid (D2EHPA) [223] which are dissolved in kerosene could also be potentially employed to extract Cu(II) ions from aqueous solution via MLLE process. Both studies indicated the experimental data agreed reasonably with the calculated time profiles of aqueous metal concentrations based on the principles of extraction reactions and transport properties of the relevant geometry (hollow fiber).

In addition to the abovementioned heavy metal ions (i.e., Cr(III), Cr (VI) and Cu(II)), the performances of MLLE process have also been assessed for many other heavy metal ions including arsenic (As), zinc (Zn), lithium (Li) and radioactive materials. For example, Bey et al. [224] fabricated microporous PVDF hollow fiber membranes with different properties by varying the bore fluid composition and its flow rate (during spinning process) and used the membrane to extract As(V) with Aliquat-336 as extractant. The authors were able to produce sponge-like membrane with pore diameters of ranging from 0.2 to 0.28 μm and porosity of 80%. The membranes however only achieved ~70% of removal rate in 6-h operation for a feed solution containing 100 ppm As(V). The authors recommended to have back-extraction to remove As(V) from the organic phase before it was recirculated to the membrane contactor module. This could further increase the extraction degree.

On the other hand, Fouad and Bart [63] employed commercial PP membrane (Liqui-Cel®) to remove Zn ions from sulfuric acid solutions via MLLE process. Di(2-ethyl hexyl phosphoric acid) (D2EHPA) dissolved in iso-dodecane was flowing in counter-current mode with the acid solution under different aqueous (0.38–0.80 L/min) and organic flow rates (0.22–0.57 L/min). The authors reported that by increasing D2EHPA concentration from 2 v% to 8 v%, k_w was increased from 5×10^{-6} to 25×10^{-6} m/s owing to the great dependence of overall mass transfer coefficient on the concentration of a complexing agent. Nevertheless, it must be noted that the authors did not provide information about the long-term performance stability of the membranes in recovering Zn from highly acidic solution (pH 0.1–2.1). Generally, PP only exhibits fair resistant against sulfuric acid with concentration of 75–100%.

Separately, Lum et al. [225] pointed out that although Zn ions could be effectively extracted using MLLE process which was based hollow fiber membrane contactor, care must be taken to pre-filter the solution prior to the membrane contactor in order to minimize crud formation and interfacial effects. Careful transmembrane pressure control is also necessary particularly when large membrane module is used in industrial process to eliminate breakthrough of one phase into another.

Owing to the importance of Li ions for different applications in particular Li-ion battery manufacturing, Xing et al. [226] developed a new type of solvent resistant membrane which was prepared from block copolymer poly(ethylene-co-vinyl alcohol) (EVAL) via immersion precipitation and used it for extracting Li ions from a real salt-lake brine. Fig. 7(a) presents the long-term stability of EVAL membrane with respect to extraction flux for ~1037 h. The small change in the flux clearly indicated the promising stability of the membrane against organic solvent (kerosene). In addition, the data shown in Fig. 7(b) revealed that the membrane exhibited relatively good mechanical stability with about 20% drop recorded in both elongation and tensile stress at breakage. The stable performance of EVAL membrane could be attributed to the unique structure of the polymer that consisted of hydrophobic ethylene units and hydrophilic vinyl alcohol units.

In order to recover U(VI) from oxalate supernatant waste, Gupta et al. [227] employed a PP hollow fiber membrane contact with effective surface area of 381 cm^2 to extract U(VI) in concentration of 35 g/dm^3 from nitric acid solution. Fig. 8(a) illustrates the process of extraction in which acid solution containing U(VI) was fed through the lumen of hollow fiber membrane while organic solvent (n-dodecane) flowing on the shell side of the membrane. Fig. 8(b) indicates that extraction efficiency of U(VI) was increased with increasing TBP concentration in n-dodecane from 5% to 50%. However, by taking into consideration the economical plant scale operation, using 30% TBP was more practical.

On the other hand, Biełuszka et al. [228] studied the efficiencies of extracting U(VI) from an acid solution using Liqui-Cel® hollow fiber membrane by comparing the performance of five different extractants, i.e., TBP, triethylamine (TEA), di(2-ethylhexyl)phosphoric acid (D2EHPA), tri-n-octylamine (TnOA) and trioctylphosphine oxide (TOPO). They reported that TnOA and D2EHPA were the most favorable extractants for U(VI), recording extraction efficiency of 97–99%. The extraction efficiency was followed by TOPO (76%), TEA (16%) and TBP (6.4%). The authors also tested the real solutions obtained from leaching uranium ores with the system and obtained extraction efficiency of 74–79% and distribution coefficient of 2.88–3.85.

3.2.7. Degasification of water and wastewater through MGS process

In 1998, a review written by Mahmud et al. [229] indicated the potential of using microporous membrane for removing VOC via MGS process by summarizing the impacts due to solution pH, membrane packing density, ozone, chlorine and fouling on the performance of hollow fiber membranes made of PP. At that time, the research studies focused only on contaminated groundwater with low concentration of VOC.

Research related to VOC removal using MGS process (sometimes it is also known as membrane air-stripping (MAS)) could be found in the literature since the 1980 s. In 1989, Semmens et al. [230] demonstrated a novel approach using microporous membrane to transfer VOCs (i.e., 1, 1,2-trichloroethane, chloroform, trichloroethylene, tetrachloroethylene and carbon tetrachloride) from water to organic-free air. They found that this membrane process offered several unique features over conventional packed-tower aeration, including no upper limit on the flow rate of gas used, increased overall mass transfer coefficient as well as elimination of tall structure and mist.

During air stripping process, the presence of surfactants in the water could possess some challenges for efficient VOC removal. Cheng et al. [231] found that it was rather difficult to attain high level of VOC removal in the cases where surfactant existed in the water as it resulted in additional mass transfer resistance in air stripping. To overcome the issue, the authors employed multipass/multistage process which could be operated at low air/liquid ratios and could still achieve almost 100% contaminant removal as a result of reduced mass transfer limitation during each stripping pass/stage. This approach offered promising strategy for removing VOCs from surfactant micellar solutions and for regenerating the solutions for reuse.

Compared to the asymmetric microporous membranes made of phase inversion method, nanofiber membrane which exhibits larger surface area and greater porosity was reported to be useful for VOC removal via MGS process. In 2012, Feng et al. [232] employed PVDF

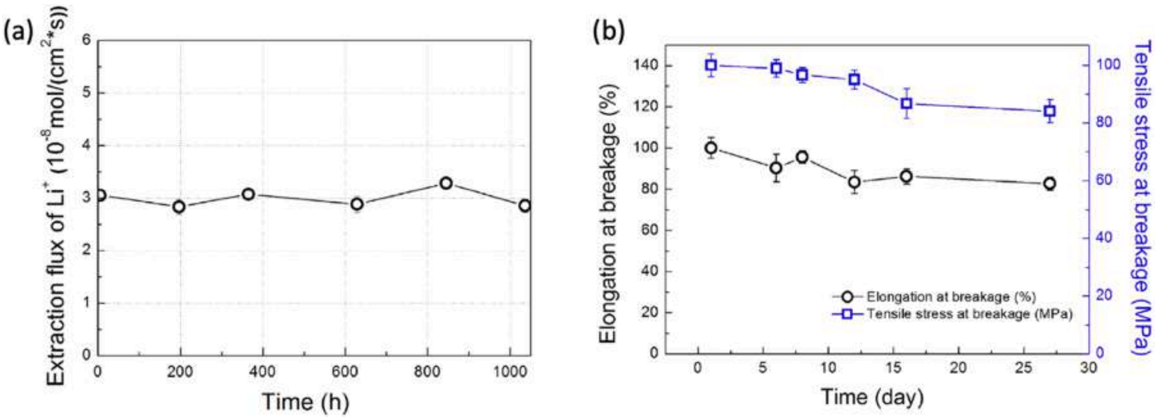

Fig. 7. Stability test of the EVAL membranes in dynamic tests. (a) Li ion extraction flux of the membranes in a continuous single-stage MLLE process (Note: each flux value was measured after refreshing the feed solution (real salt-lake) and organic extractant ($NaFeCl_4.2TBP(o)$) and (b) Changes of tensile stress and elongation of membrane in $NaFeCl_4.TBP(o)$ and brine water [226].

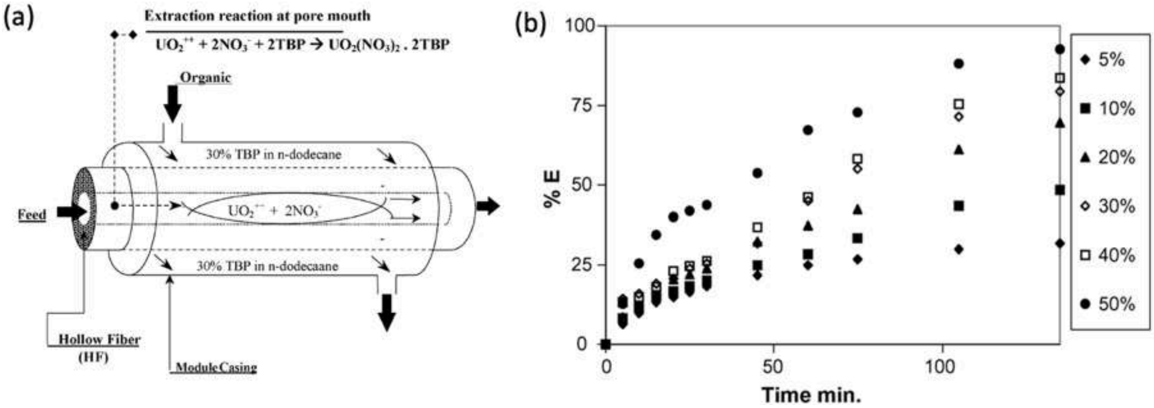

Fig. 8. (a) Extraction mechanism of U(VI) through hollow fiber contactor and (b) Influence of organic concentration on U(VI) extraction (Testing conditions: concentration of U(VI): ~35 g/dm^3, feed acidity: 3 M HNO_3, feed flow rate: 5.83 cm^3/s and organic (pseudo-emulsion) flow rate: 1.53 cm^3/s) [227].

nanofiber membrane with porosity of 76%, pore size of 0.32 μm and water contact angle of 128^o to remove chloroform from 5000-ppm chloroform feed using nitrogen stripping gas. The authors reported that the overall mass transfer coefficient of chloroform through the nanofiber membrane was even better compared to the process using hollow fiber membrane, recording a value of 2.40×10^{-5} m/s. The improved separation performance was mainly attribute due to lower resistance of boundary layer for the microporous nanofiber membranes.

In addition to VOC removal, MGS process is also widely used to address the greenhouse gases particularly CH_4. For instance, Cookney et al. [233] utilized PDMS hollow fiber membrane contactors to recover CH_4 from anaerobic process effluent using nitrogen as sweep gas. The researchers were able to achieve a maximum degassing efficiency of 72% in terms of dissolved CH_4 and further demonstrated that the recovered CH_4 could increase net electrical production and reduce net carbon footprint to only 0.01 kg CO_2/m^3.

Henares et al. [234] reported that the performance of PP membrane contactor for CH_4 recovery from anaerobic reactor effluent could be optimized by taking into consideration the operational parameters, operation mode and fouling issue. In particular, they found that higher removal efficiency could be achieved by prefiltration of the effluent (to remove particulate matters) and by conducting 30-min water cleaning daily. This could prevent severe membrane fouling in short period of time and prolong membrane lifespan.

Rongwong et al. [235] on the other hand, presented an energy analysis on the hollow fiber membrane contactors for its CH_4 recovery performance during the treatment of effluent obtained from an anaerobic membrane bioreactor wastewater treatment process. From their analysis, it was shown that the energy recovered from the dissolved CH_4 per 1-m^3 effluent could be optimized using high strip gas flow rate and lower vacuum condition (~0.9 atm). Under this condition, the energy obtained was 0.178 MJ/ m^3, which was equivalent to ~8% of the total energy recovered from the dissolved CH_4. Nevertheless, they explained that the obtained energy could be lower when membrane cleaning activities are required to perform periodically to address the membrane fouling issue induced by microorganism and organic matters in the effluent.

A recent review written by Mora and Chernicharo [236] highlighted the key challenges associated with membrane contactors for removing and recovering dissolved CH_4 from anaerobic reactors effluents in full scale process. Although membrane contactors are generally known for its potential for CH_4 recovery, there are very scarce studies that focus on the composition of the recovered waste gas, e.g., CO_2, N_2 and H_2S for both vacuum and sweeping gas operating modes. Other issues are about the long-term impacts of organic and biofouling on the membrane and the cost/benefit relationship of recovering CH_4 from anaerobic effluents from wide range of characteristics.

Other gas that has been previously studied to remove from water using MGS process is oxygen. Removing dissolved oxygen (DO) from water is necessary in many industries including power, food, semiconductor and pharmaceutical. For instance, to prevent boilers' and pipes' corrosion, the water in the power industry needs to be pre-treated to bring down the level of DO to ~5 ppm [237]. Semiconductor industry on the other hand requires extremely low level of DO (~0.1 ppb) during silicon wafers washing [237].

Shao et al. [53] carried out a pilot study to remove DO from boiler feed water using 42-m^2 hollow fiber membrane system via vacuum degassing process. The authors studied the effects of different parameters including water temperature, water flow rate and vacuum level on the mass transfer coefficient of oxygen ad its removal rate. In most of the cases, the membrane system could achieve $> 80\%$ DO removal rate and consistently produced the treated water with < 2.0 ppm DO. Nevertheless, the performance of membrane was adversely affected after long separation period using surface water as source due to the presence of aluminum silicate and organic matters in the feed water. Thus, the authors emphasized the requirement of proper cleaning process to maintain the membrane performance.

Owing to the limitations of membrane packed with bundles of hollow fibers during degassing (e.g., shell side channeling and bypass in the parallel flow and high pressure drop in lumen side), Peng et al. [54] utilized propriety transverse flow membrane contactor packed with PP woven fabric hollow fibers to address the issue. The experimental results using commercial membrane module with effective surface area between 3.7 and 81 m^2 indicated that the contactor packed with woven fabric attained higher mass transfer coefficient and greater degassing process attributed to its internal structure design that resulted in transverse flow. The pilot membrane system was able to effective remove DO from water by achieving up to 97.5% O_2 removal.

Besides removing DO from water, Criscuoli et al. [35] reported that MGS process was capable of controlling water pH for RO desalination plant using hydrophobic flat sheet membrane with gas stream of N_2 or CO_2. Results showed that although both N_2 and CO_2 were able to remove O_2 from water in a very similar extent, the final pH of water increased or remained constant after the tests with N_2 whereas decreasing after using CO_2. A maximum pH reduction of 33% and an increase of pH up to neutral was able to attain using this system. The findings also revealed the possibility of removing DO from water and controlling its pH simultaneously without the use of chemicals.

Over the years, we have seen many research studies successfully demonstrated the potential of MGS process in separating different types of gases from liquid solutions, in addition to VOC, CH_4 and DO. Literature revealed the prospects of using membrane contactor in removing not only corrosive and poisonous H_2S from sour water [238] but also pungent NH_3 from chicken manure digestate [239]. More interestingly, MGS process was even found useful in extracting aroma compounds from a highly diluted aqueous feed [240]. Aroma extracts are an important process in food industry to improve flavor of formulated foods or to compensate for the flavor losses of natural raw products during processing.

4. Current status and future direction

Membrane contactor as a device for process intensification of contacting different phases has gained various applications in the area of chemical processes. Since Esato and Eiseman [45] developed the first gas-liquid membrane contactor in 1975 for oxygenation of blood, numerous research was conducted on various membrane contactor processes for separation of gas and liquid streams. The importance of the membrane contactor technology can be shown through the published papers over the past 30 years, as given in Fig. 9. As shown, MD as the vapor-liquid membrane contactor process showed the most contribution (~54.4%) due to the high demand for the production of clean water worldwide. About 28% of the published papers is related to the MGA processes which are mostly used for greenhouse gas control, natural gas and biogas treatment. The MLLE processes for water and wastewater treatment showed a lower contribution of about 13% which can be attributed to the low separation efficiency and the requirement of an auxiliary separation process. In addition, the MGS processes demonstrated the lowest contribution (<5%) due to the low stripping efficiency.

It should be noted that most of the works on membrane contactors are still at the stage of laboratorial due to some technical hurdles. Nevertheless, there are some pilot/full scale systems are employed for environmental protection. Kvaerner developed a pilot gas-liquid membrane contactor unit for treating flue gas from a gas engine (520 kW) at the Statoil Gas Terminal in Norway [241]. Using a PTFE hollow fiber membrane contactor module with the alkanolamine solution, about 85% of the CO_2 is removed from the flue gas with the flowrate of 2610 kg/h. Separately, a pilot plant of MGA process for post combustion CO_2 capture was developed by Netherlands organization for applied scientific research (TNO) [242]. The improved liquid absorbent (CORAL) was used with a PP membrane module (0.27 m^2) at the pilot plant operating conditions of 0.5–4 m^3/h flue gas flowrate, 0.5–20 L/h liquid flowrate and 0.05–10% CO_2 concentration in the gas stream. At atmospheric pressure the capacity of the pilot plant was about 0.8 kg CO_2/h with less than 1% CO_2 content in the gas stream.

Chabanon et al. [243] applied a pilot scale membrane contactor module (10 m^2) based on PTFE hollow fibers for post-combustion CO_2 capture in the presence of 30% monoethanolamine (MEA) aqueous solution. The inlet gas contained 15% CO_2 and the liquid and the gas flowrates were in the range of 0.50–3.33 L/min and 5–30 L/min, respectively. CO_2 outlet concentration of about 2% was achieved at the liquid and gas flowrates of 150 L/h and 5 L/min, respectively.

Recently, a membrane contactor pilot plant designed by the researchers from University of Melbourne was tested at a black coal fired power station (New South Wales, Australia) for post-combustion CO_2 capture [89]. A non-porous PDMS hollow fiber MGA module (10 m^2) was used to process up to 20 kg/h of the flue gas containing 12.3% CO_2

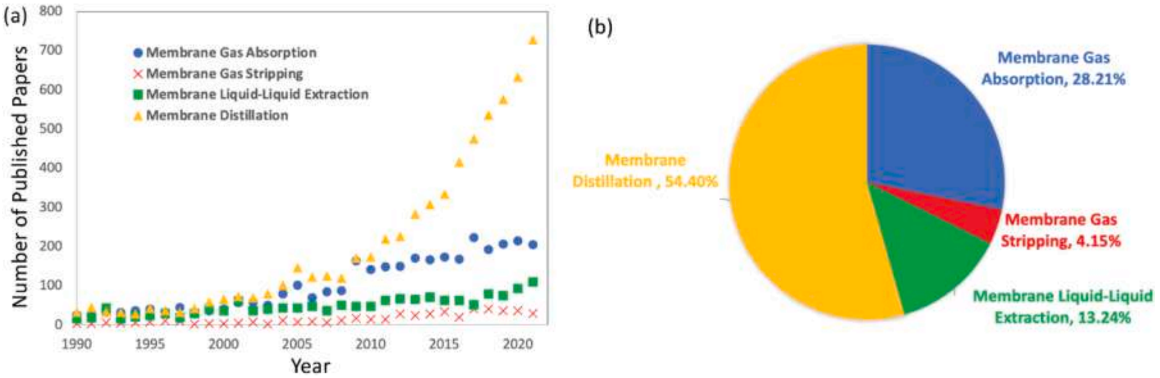

Fig. 9. Summary of literature data reported on four different membrane contactor processes, (a) Number of papers published between 1990 and 2020 and (b) Breakdown of publications for the past 30 years. The data were extracted from Scopus on January 10, 2022.

in the presence of 30 wt% MEA solution. In addition, another PDMS hollow fiber MGS pilot module (50 m^2) was used for regeneration of the MEA solution. A stable CO_2 absorption flux of about 0.0023 mol/m^2 s was achieved for over 30 days.

An industrial MGA unit for ammonia recovery from off-gas stream has been in operation since 1999 at Aliachem in Pardubice, the Czech Republic [68]. The capacity of this MGA unit was 50 kg/h of absorbed ammonia with proven very easy operation. Using a microporous PP hollow fiber membrane module (TNO design transversal flow), the ammonia was recovered as 27 wt% ammonia solution and about 99.9% ammonia emission to the environment was decreased. Dow et al. [244] installed a DCMD pilot trial for achieving zero liquid discharge (ZLD) on site at Australian Textile Mills. The photo of installed DCMD pilot plant and process flow diagram are shown in Fig. 10. The membrane module was fabricated by Victoria University (6.38 m^2) and no membrane wetting was observed during 90 days of the DCMD operation. The initial water flux was 5 L/m^2 h, but decreased to 2 L/m^2 h after more than 65-day operation. By caustic cleaning, the flux was recovered to 79% of the original flux. It was found that by applying waste heat integration and reverse osmosis/nanofiltration for reducing brine volume, the MD process can be feasible for treating wastewater of the textile plants.

An AGMD pilot scale was developed by Woldemariam et al. [245] for purification of effluent from a municipal wastewater treatment plant. A high pharmaceutical removal efficiency was achieved even for highly concentrated feeds. From an economic evaluation, the authors found that majority of the cost came from the heat cost during operation. On the other hand, a MLLE pilot scale system was used for selective separation of zinc from industrial spent pickling acids (SPAs) [246]. The pilot plant was equipped with four microporous PP hollow fiber membrane contactor modules (3 M™ Liqui-Cel™) with 80-m^2 total surface area. The extractant solution of 50 v% of TBP in ShellsolD70 was used at flowrate of 150 L/h in the module shell side. During 30-h operation, zinc concentration in the feed was reduced from 75 to 24 g/L.

Another MLLE pilot plant installed for separation of phenol from the hydrocarbon fraction by an aqueous NaOH solution (16 wt%) [247]. This pilot-plant was installed at the phenol and acetone plant of the PKN ORLEN Company. The MLLE process was operated in two stages each applying eight parallel Liqui-Cel extra flow contractor modules (4” x 28”). The plant capacity was 550–650 kg/h of hydrocarbon fraction containing 2–4 wt% of phenol. The very high separation efficiency of 98.5–99.5% was achieved where the dephenolized product was below 0.02 wt%.

Although the membrane contactor technology has demonstrated advantages over the conventional contacting devices (such as small footprint and high removal efficiency), a comparative economic evaluation in terms of membrane long-term stability, operating/maintenance cost and energy consumption is still lacking. The main challenges such as development of the membrane structure to minimize wettability, upgrading the liquid absorbent and extracting agents have been the experimental research focus. For further application of the technology at industrial level, some critical issues need to be overcome and much research efforts are required, as summarized below.

a) the membrane creates an extra resistance to the mass transfer process compared to the conventional contactors, a membrane with ultrathin skin layer is an advantage. High surface porosity is also necessary to provide a great contact area between the phases. The membrane surface hydrophobicity is the key factor which can minimize wetting and mass transfer resistance. Small pore size and surface hydrophobicity can increase breakthrough pressure of the membrane which is necessary to extend the application for high pressure operations.
b) Channeling and bypassing in the shell side of the membrane contactor module can significantly affect the separation efficiency. Therefore, the membrane module design needs to be upgraded especially for large scale.
c) Heat loss in MD process is a serious problem which can be minimized by selecting a low thermal conductivity membrane material and/or improving the module configuration/design.
d) The evaluation of energy requirement and techno-economic analysis for different membrane contactor processes have seldom studied in the literature. In fact, they are the main critical factors to achieve the level of commercialization.
e) Integration of membrane contactor processes with other chemical separation units or various membrane processes, and waste heat and/or renewable energy sources needs more investigation in order to accomplish new sustainable technical solutions.
f) Most of the studies on membrane contactor processes are focused on simulated feed streams by ignoring the presence of impurities in the feed which may act as competitors for separation or even affect membrane structural properties. It is required to study the membrane contactor performance in a real feed condition to evaluate the membrane stability for a long-term operation.

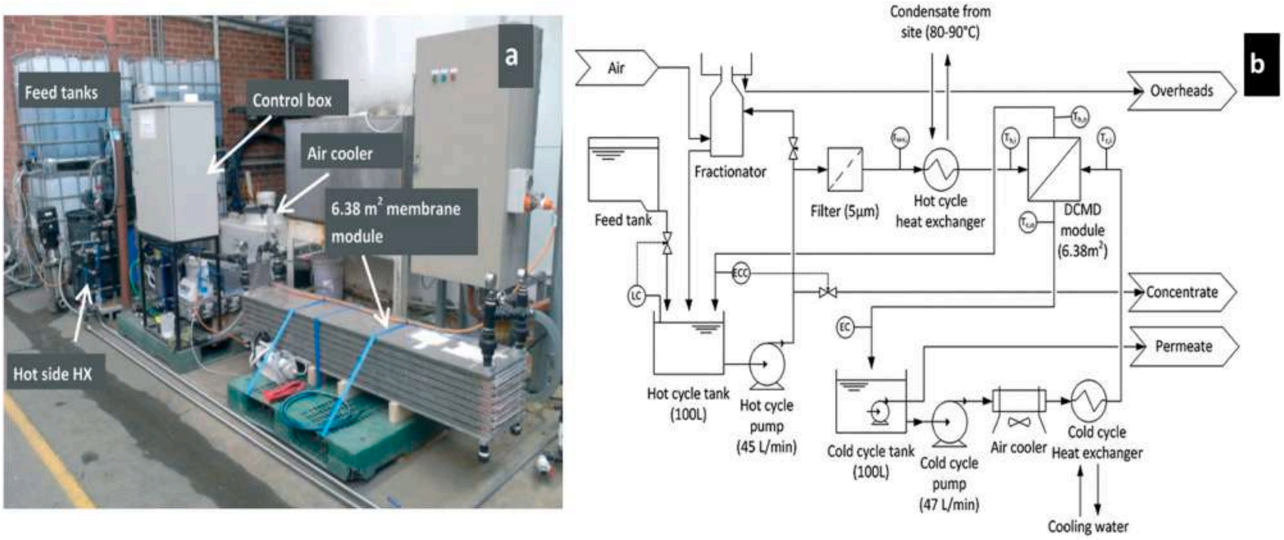

Fig. 10. (a) Photo of DCMD pilot plant equipped with 6.38 m^2 module for treating wastewater of Australian Textile Mills and (b) process flow diagram of the pilot plant [244].

g) Fouling in membrane contactor processes has been rarely studied compared to membrane wetting. Indeed, contaminants such as fine particles, aerosols and water vapor in the flue gas streams can cause fouling which might reduce the gas absorption performance. In addition, organic compounds in wastewaters can cause strong interaction with hydrophobic surface of membranes and affect the membrane contactor performance. Therefore, fouling mechanism in membrane contactor processes is necessary to be clarified in order to maintain the membrane integrity for a long-term operation.

5. Conclusions

Membrane contactor as an innovative separation process can offer interesting environmental treatment motives to many industrial challenges. In membrane contactor processes, a non-dispersive mass transfer is occurred between two different phases which can praise a new generation technology with high efficiency, compact and flexible separation. In this article, the membrane contactor processes were concisely introduced and the latest research on environmental applications of the technology were reviewed. In addition, the current status and future direction were discussed to provide indications for the application of membrane contactor at industrial level. Environmental applications of membrane contactor processes include but not limited to greenhouse gases control, recovery of VOCs from off-gas streams, recovery of hydrocarbons, heavy metals and radioactive materials from water and wastewater streams, degasification of water and wastewater, oily and dyeing wastewater treatment, as well as ammonia and phenols removal from wastewater. Several large membrane contactor pilot plants which are in operation were also reviewed. It can be said that there will be increasing interest in the coming years due to strong potential of the technology in environmental applications. Nevertheless, it must be pointed out that membrane wetting is still the main problem to achieve a practical application. Therefore, the future direction of membrane contactor development is related to the enhancement of membrane material, membrane structure, absorbent solution and extraction agent. Moreover, integration of membrane contactor processes, evaluation of energy requirement and economic benefits play important roles in commercialization of the technology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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