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Progress in treatment of oilfield produced water using membrane distillation and potentials for beneficial re-use

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ABSTRACT

Membrane distillation (MD) has proved worthwhile in treatment of hypersaline feeds demonstrating near complete rejection of dissolved solutes without any effect on the process conditions. This makes it potential treatment option for hypersaline oilfield produced water (PW) with salinity level far greater than that of seawater. Polymeric membranes have recently garnered more attention than their ceramic counterparts in oily wastewater treatment owing to ease of synthesis and relative cost advantage. However, lower mechanical durability and the propensity for fouling of these membranes due to presence of low surface energy organics in PW ultimately affects MD performance in its treatment. Studies elucidating the mechanism of fouling between PW feed and membranes in MD is scarcely reported in the literature. Various fouling mitigation approaches have shown promise towards the realization of MD as a viable option for PW treatment. Hybridization of MD, use of (super)hydrophobic MD membranes with feed pre-treatment using other technologies and/or membrane post-cleaning, integrated MD systems and recently omniphobic and Janus modifications of MD membranes have all been reported for treatment of PW showing more promise towards achieving ultrapure-distillate treatment. In this article, the performance of these polymeric MD membranes used in PW treatment in comparison to the other conventional treatment options as well as advances in MD as a cost-effective alternative for beneficial re-use of PW is reviewed, highlighting the areas requiring further study for this line of research. Because MD is still largely energy inefficient, several efforts to realize it as an all-round competitive technology focusing on long-term stability, brine handling capacity and potentials for cost savings with alternative and rather sustainable energy source are also discussed.

1. Introduction

Wastewaters generated by oil and gas industry is simply referred to as produced water (PW). Depending on its origin, PW is broadly classed as oilfield PW, natural gas PW, coal bed methane PW and shale gas PW [36,124]. Its sources are primarily from water injected into the reservoir for enhanced oil recovery (EOR) purposes, flow back water from hydraulic fracking as well as underlying aquifer or formation water where present/inherent in the reservoir formation [68]. They have contributed to mainstreams of wastewaters with hyper salinity and organic chemical [44], impacting heavily on the environment by contaminating fresh water sources, distorting aquatic habitation and even rendering arable lands infertile when discharged to the environment without proper treatment steps [197]. The US Environmental Protection Agency (EPA) classifies oily saline discharges as among the most hazardous wastewaters owing to presence of recalcitrant compounds, aromatic compounds, heavy metals and salts in high concentration that pose difficulty for treatment [128]. As such, there is need for cost effective strategies for treatment of saline oily wastewater that minimizes environmental impact [230]. It is for this reason stringent regulation regarding

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discharge are put in place.

Conventional treatment options ranging from use of phase separators, hydrocyclones, coalescers, coagulation and flocculation, media filters and polishing steps have been utilized to significantly reduce organic species concentration in PW to meet discharge regulations. However, these processes do not address the very high concentration of salt present in PW [21,33,150,222,235] . Thermal processes including the use of brine concentrators and crystallizers, multi-effect distillation (MED), multi-flash distillation (MSF) and electrodialysis (ED) proved rather unsuitable due to large footprint, effect of salt scaling on equipment and high energy requirements [88,129,193,213].

A grandeur shift to use of pressure-driven membranes in treating PW saw the use of ultrafiltration (UF) [38,288–289], nanofiltration (NF) [303], microfiltration (MF) [90,136] and reverse osmosis (RO) membranes [56,146,187],with remarkable treatment capacity although the membranes suffered from fouling by organics. RO in particular has been extensively studied and has been predominant as a desalination technology that has shown capacity for removal of monovalent ions and producing high purity permeate [103]. However, RO membranes also suffer from fouling by organics and more severely from osmotic pressure build-up resulting from concentration of the feed causing it to exceed the operating pressure of the system and thus affect the process performance [279–280].

Membrane distillation (MD) has recently generated increasing attention and acceptance, and can be a potential replacement for conventional desalination processes [190]. MD process performance is not affected by salinity of the feed solution [163]. This technology has various promising characteristics such as lower operating temperatures in comparison with other thermal desalination technologies as the feed solution does not require heating up to boiling point temperature. The constraint of space on onshore and offshore platform makes MD an interesting candidate to consider for treatment of PW because of its small footprint [239]. Lower operating pressure than reverse osmosis (RO) and 100% theoretical rejection of the non-volatile solutes in the feed solution makes MD more ideal.

MD has shown promise in treatment of feedwater with hybrid impurities such as oils, salts and surfactants typical of PW with dedicated membranes. The major concern with respect to this type of wastewater is the propensity for membrane fouling by low surface energy contaminants present in PW. Various approaches for fouling mitigation in MD have been reported in literature [60,156,282] . A recent review on fouling phenomena and mitigations strategies in MD was reported by Choudhury et al. [50]. They examined the interactions between foulant and different types of MD membranes and the influence of different operating conditions on the occurrence of fouling and wetting. In another review article, Siyal et al.[243] gave an overview of various membrane materials developed for MD applications for the treatment of wastewater with hybrid impurities typical of emulsified industrial or shale gas wastewater. They posited that further advancement in materials suited for treatment of such wastewater type is required and that integrated MD systems may be the way forward. Similarly, Nthunya et al. [200] have reviewed the use of nanoparticles to enhance the performance of MD membranes in wastewater, brackish water and seawater treatment applications and discussed the cost implications, commercial viability, environmental sustainability and future prospects of MD. In a more specific review, Lu et al. [170] comprehensively reviewed the design approach and fundamental principle of omniphobic membranes giving insights to developing robust anti-wetting membranes and their novel applications in MD. In a recent article on polymeric membranes for MD [221], we reviewed the intrinsic trade-off relationship that exists due to various membrane properties and how they can be optimized towards achieving the ideal MD membrane. It also brought to fore the need to further study these properties.

Growing concerns for environmental protection and drive for attaining sustainability makes the continuous evolution of treatment processes for complex wastewaters such as PW pertinent. MD offers potential for green treatment process in removal of various organic and inorganic compounds from wastewater and seawater alike [1,119,122 121,120]. It is important to bring to fore the advances made in this area particularly with green technologies such as MD in a bid to further bridge the gap between the laboratory and commercialization.

Increase in efforts to realize MD as a useful separation technology for PW has seen a significant rise in publications over the last decade, particularly for polymeric membranes. Fig. 1 presents the recent trend of publication in the Scopus database using the keywords 'produced water' and 'membrane distillation' limiting the search to title, abstract and keywords of journal articles. Interestingly, the trend rose sharply after 2013 which featured the first article showcasing MD for treatment of de-oiled PW for water reclamation from a steam-flooded oil exploration facility by Singh et al. [241] using a hollow fiber membrane module. Subsequent increasing trend highlight the growing interest in this field by researchers, amidst remarkable success. It can also be noted the corresponding increase in oily wastewater related publications detailing specifically the use of omniphobic membranes in MD. The very first publication of which entailed the modification of hydrophilic glass with nanoparticle and subsequent coating with polymer was reported by Lin et al. [160]. The results demonstrated that such types of modification are critical for MD application with feed such low surface energy contaminants such as oil field PW. The increasing trend thereafter also reflects the growing interest in this line of application.

Hence, in this article, we review the applications of various polymeric membrane types for treatment of PW in MD to achieve cleaner distillate, their constraints and potentials for process optimization. The trend in treatment of PW ranging from conventional methods and pretreatment options to other membrane technologies are systematically reviewed in order to clearly demonstrate to the reader the potentials of MD in this field. We also briefly highlight the class of nanoparticles frequented in MD applications for enhanced performance as well as novel carbon nanomaterials like carbon nanotubes that have good photothermal/energy storage properties and thus shown potential for improving MD energy efficiency. Fouling phenomena typical for PW in MD and the problem of scaling by divalent salt ions present in PW and their effect on MD process as well as mitigation strategies are also presented. The potential of achieving beneficial re-use of PW using MD as a cost-competitive alternative is also captured. This review article has contributed to the sustainable development goal (SDG) No.6 of the United Nations (UN) which is aimed at clean water and sanitation as shown in Fig. 2. Specifically, it encompasses two human rights; 1) universal and equitable access to safe and affordable drinking water 2) access to adequate and equitable sanitation and hygiene for all. Interestingly, clean water and sanitation have only become human rights in recent years. The UN's 2030 agenda has delineated that the SDG 6 promotes the success of all the other 16 SDGs. Considering the importance of clean water as a strategic resource especially in the wake of diminishing fresh water sources, it is important to strike the right balance between economic growth and environmental protection without sacrificing either in the battle against global water pollution.

2. Membrane distillation

MD is an emerging technology designed to only allow the passage of vapor across a hydrophobic membrane material [76,216] . This vapor transport is facilitated by a partial vapor pressure differential created by the difference in temperature from feed side to the permeate side of the membrane [77]. MD is one of the most advanced water treatment processes utilized for extremely polluted water sources. In this process, only water vapor will be transported through the micro-porous hydrophobic membrane [149].The separation process involves vaporization through heating of the feed solution. Popular MD configurations, as illustrated in Fig. 3, include direct contact membrane distillation (DCMD) where there is direct contact between the liquid on both feed and permeate sides with the membrane at the vaporization and condensation interfaces

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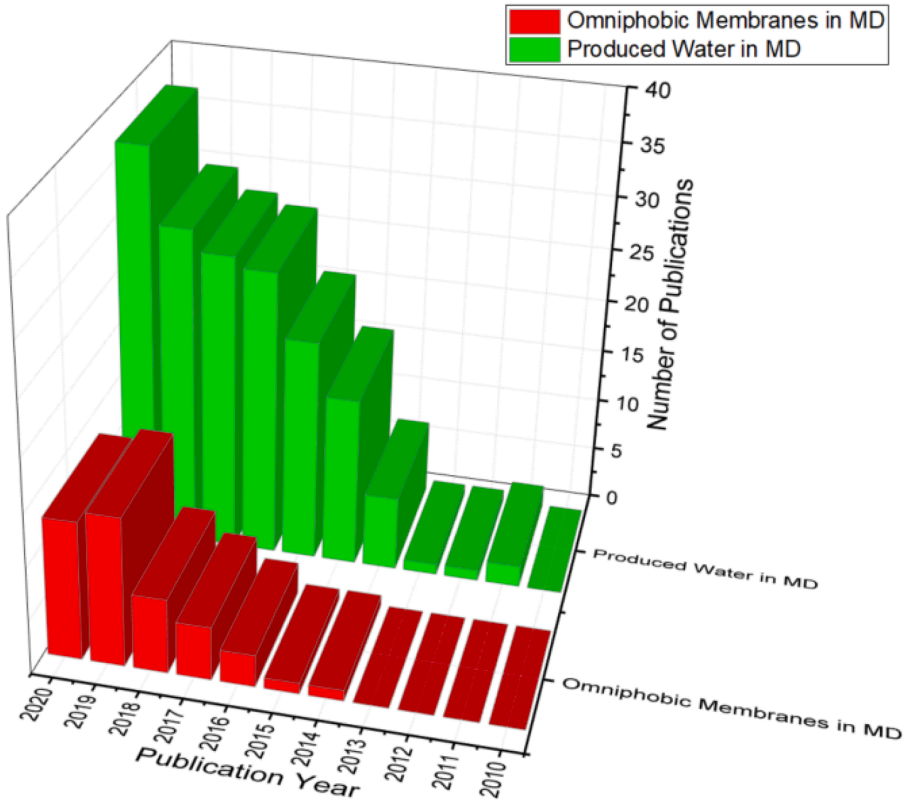

Fig. 1. Trend of publications regarding MD and PW treatment in the last decade from Scopus database (Data retrieved on June 6, 2021).

Fig. 2. Clean water and sanitation as SDG No. 6 of the United Nations (*Water and Sanitation – UN SDGs*, 2016).

respectively [273], vacuum membrane distillation (VMD) where an external vacuum pump is used to aid vapor collection and condensation takes place outside the module [144]. Air gap membrane distillation (AGMD) features a gap between a condensation and the membrane aimed at minimizing heat loss [135], while sweeping gas membrane distillation (SGMD) involves transportation of vapor generated to a condenser outside the module using inert gas [142]. The difference between all the configurations is the type of contact medium with the membrane on the permeate side, the feed solution however is in contact with the membrane at the feed side for all the configurations [307].

2.1. Ideal properties and materials for MD membranes

In comparison to ceramic membranes, polymeric membranes have received much attention due to their intrinsic hydrophobicity, cheap cost and ease of fabrication. At present, polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF) and polypropylene (PP) comprise of the most popularly used polymeric membrane material for the MD applications. PTFE is the most suitable material for MD owing to its higher hydrophobicity, remarkable thermal stability and chemical resistance than the rest of the materials. However, PTFE is difficult to be processed due to its low solubility with regular solvents [65,273]. Meanwhile, PP possesses high crystal forming ability alongside excellent solvent resistance with advantages in relation to material and manufacturing costs. However, it has a moderate thermal stability at higher operating temperatures which significantly affect the integrity of the MD process [273]. PVDF on the other hand has a high solubility in regular solvents such as dimethylformamide (DMF), N-methyl-2-pyrrolidone (NMP), triethyl phosphate (TEP) and dimethylacetamide (DMAc) and has good processability. It is generally fabricated with the phase inversion technique [273].

Membrane properties dictate the performance of the membrane and as such, an optimized membrane precisely designed for MD operations could improve its performance [78]. The ideal membrane comprises of at least one hydrophobic layer that will not be wetted by the feed solution. Membrane properties such as water permeability, pore size distribution, membrane thickness and surface energy require precise tuning for desired performance in MD process. These properties directly affect the wettability of the membranes typified by the liquid entry pressure

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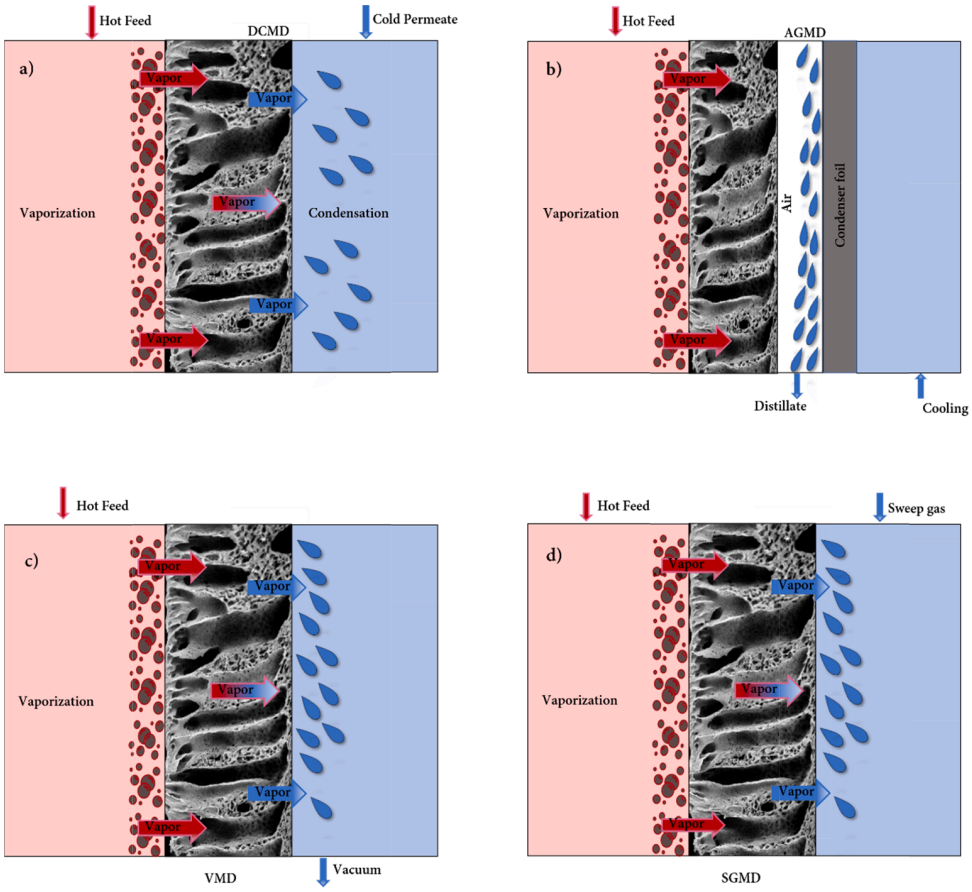

Fig. 3. Diagrammatic representation of popular MD configurations; a) DCMD b) AGMD c) VMD d) SGMD.

(LEP) and water contact angle values as well as membrane thickness optimization which ensures minimal heat loss and hence long-term performance. The intrinsic relationship of these properties and their corresponding effect on MD membranes can be found in the literature [221,243] .

2.2. Membrane wetting in MD

In desalination membranes, MD which is a thermal process offers the advantage of overcoming the drawbacks of RO [95]. Increase in osmotic pressure as the salt concentration increases through the desalination process is beyond the maximum operating pressure of current RO technology [280]. Superhydrophobic modifications to create surfaces with excellent anti-wetting properties with enhanced MD performance are essentially synthesized by imparting roughness on hydrophobic surfaces through incorporation of inorganic nanoparticles such as silica, zinc, titanium, alumina and/or iron oxide nanoparticles [5,20,42,57,96,104,140,242,295,309]. While most of the inorganic nanomaterials are relatively cheaper, they are mostly hydrophilic and several synthesis steps often involving expensive chemicals are required to make them intrinsically superhydrophobic and/or superamphiphobic to be suited for hypersaline PW feed as discussed in subsequent sections. Attention has recently focused on carbon-based nanomaterials including various forms of graphenes and carbon nanotubes that are readily hydrophobic with structures providing excellent permeability, mechanical strength and even photothermal properties for improvement of MD energy efficiency.

2.3. Carbon-based nanomaterials and designs for robust MD membranes

Carbon-based nanomaterials such as graphene oxide (GO) and carbon nanotubes (CNTs) have been reported in polymeric membrane synthesis [2,151,263]. These nanomaterials are hydrophobic in nature due to their aromatic rings composed of relatively non-polar C—C and C—H bonds which are not solvated by water molecules [9]. They also render the improvement of membrane mechanical strength which is important in MD [191]. These classes of nanomaterials are seen to be 'changing the game' in performance enhancement of membranes. In a review of application carbon composites membranes thermal driven membrane processes such as MD and pervaporation, Yuan et al. [301] highlighted three common approach for their application, as depicted in Fig. 4. While free-standing carbon membranes and surfaces modified by carbon materials offer fast water transport occasioned by nanochannels and smooth surfaces, their high thermal conductivity result in loss of latent heat for vaporization. Mixed matrix membranes (MMM) demonstrate more promise due to good thermal insulation provided by the polymer matrix. Hence, incorporating carbon materials in MMMs is an effective approach to improve their hydraulic and even mechanical properties ([217].

To synthesize a high performance membrane for MD process, Bhadra et al. [30] immobilized GO on PTFE using PVDF as a binder material. The resultant membrane exhibited a high flux of 97 kg/m^{2}·h and high salt rejection with a feed containing 34,000 ppm of salt, in a DCMD test. The high flux is expected to be attributed to the presence of GO causing reduced temperature polarization, selective sorption and nanocapillary effect. Leaper et al. [151] also reported enhanced flux by 86% with APTES-functionalized GO incorporated in PVDF in comparison to pristine PVDF. Similarly, Abdel-Karim et al. [2] recently used reduced

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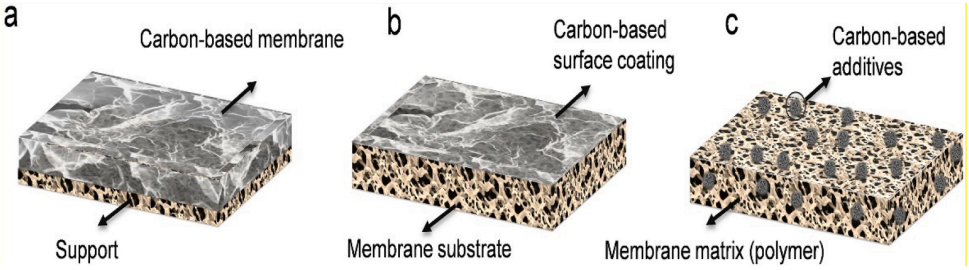

Fig. 4. Common approaches for use of carbon composites in thermal-driven membranes a) Carbon-based membranes b) surface modification by carbon materials c) Carbon composite MMMs [301]

graphene oxide (rGO) nanoplatelets as fillers in PVDF membrane matrix in a bid to study the levels of oxygen reduction (36%, 58%, 65% and 69%) on the performance of MD. The resultant MMM with 0.5 wt% rGO (reduction of 58%) exhibited flux enhancement of 169% in comparison to pristine PVDF membrane with salt rejection of 99.99% in an AGMD test of 96 h. In another study by Bhadra et al. [31], they incorporated CNT onto PTFE composite membranes with a nonwoven fabric support in a bid to enhance the permeate flux for a test period of 120 days. Multi-walled CNTs (MWCNTs) were functionalized by carboxylation in a microwave and was added to the PTFE polymer solution. The resultant carbon nanotube immobilized membrane (CNIM-*f*) displayed stable flux of 69 kg/m^{2}·h and salt rejection above 99.9% (feed concentration of 34,000 ppm) throughout the test period. As illustrated in the vapor transport mechanism in CNIM-*f* (Fig. 5), CNTs and even graphene can serve to provide a well-defined nanochannel with little mass transfer resistance coupled with enhanced water permeability [266], and a unique salt sieving property due to carbon fibrous structure preventing salt ions from having contact with water–vapor interfaces [42].

In an effort to enhance the mechanical properties of PVDF hollow fiber and flat sheet membranes for MD process, Lu et al. [173] introduced n-butylamine functionalized graphene oxide (GO-NBA). The GO-NBA proved to give better enhancement of mechanical properties of the membranes than the conventional GO. A burst pressure and LEP enhancements of 67% and 15% were recorded respectively for a GO-NBA concentration of 0.5 wt%. A flux of 61.9 kg/m^{2}·h and salt rejection of 99.9% was achieved for a DCMD test with model seawater as feed. Similarly, Grasso et al. [92] reported a PVDF flat sheet membrane functionalized with styrene in order to enhance adhesion of graphene on the membrane surface. The styrene functionalized PVDF membrane showed remarkable flux (16 kg/m^{2}·h) and rejection of 99.9% but lost mechanical properties. The graphene-PVDF composite however had lower flux with improved performance stability.

In a study comparing the performance of graphene nanoplatelets (GNPs) and MWCNTs to fabricate dual layer PVDF hollow fiber membranes by Ravi et al. [220], they showed that both nanomaterials improved the membranes' surface roughness, water contact angle and wetting resistance which were observed to increase with increasing concentration of both GNPs and MWCNTs. Both nanoparticles were oxidized then coated them fluoroalkysilanes to induce hydrophobicity before incorporating in polymer dope solution. Interestingly, due to higher fluorine content of GNP-modified membranes thereby exhibiting higher water contact angle and wetting resistance, they displayed higher permeate flux (8.27 kg/m^{2}h) than the MWCNT-modified membrane (3.82 kg/m^{2}h) for the same nanoparticle concentration. All the membranes also exhibited salt rejection of>99.9%. However, the flux values were quite low compared to similar works in the literature, attributable to thick membrane walls of dual layering and dense membrane surface. The work featured the creation of a hydrophobicity gradient across the

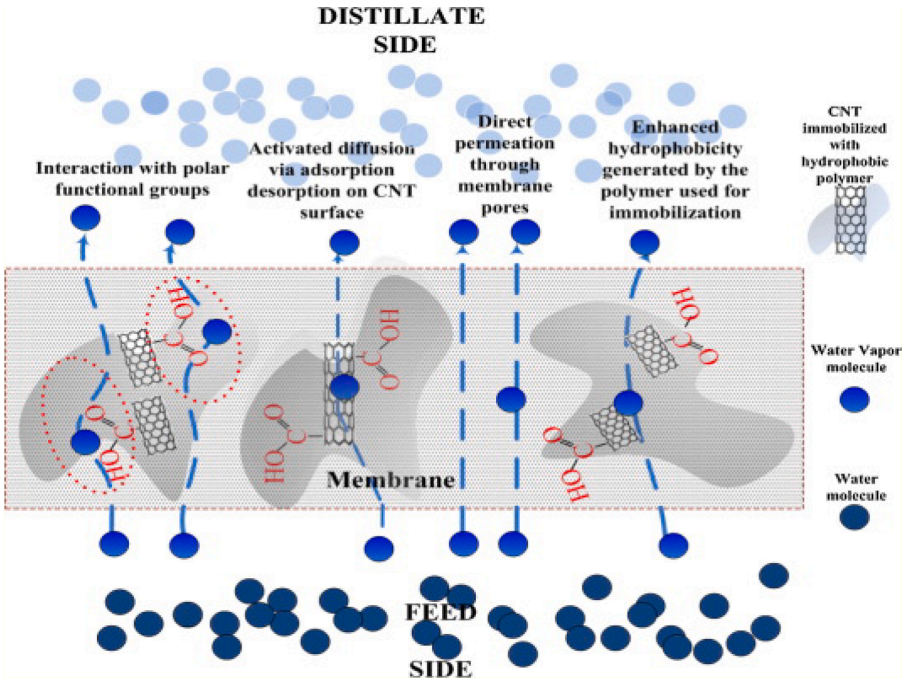

Fig. 5. Mechanism of transport through CNIM-f (Bhadra et al., 2016b).

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membrane path with the inner layer being hydrophilic which has been reported to facilitate better flux for MD membranes. Fouladivanda et al. [83] also reported the synthesis of octadecylamine-reduced graphene oxide (ODA-rGO) for modification of PVDF-HFP nanofibrous membranes for saline water desalination using AGMD. Contact angle increased by about 14.5% percent with 0.1% loading of ODA-rGO in comparison to pristine membranes. Modified membranes were hot-pressed in a bid to improve the wetting resistance through pore size reduction and thus increase in LEP, as well as increase mechanical durability of the membrane. The resultant membrane displayed average permeate flux of 21.1 kg/m^{2}h and 99.99% salt rejection in a 4-days run without loss of rejection.

Recently, carbonaceous nanomaterials have provided excellent platforms for use as energy storage agent for localized heating on the membranes surface [98–99,219,276]. Although more studies are required to understand their behaviour with complex feeds like PW. Despite promising results in enhancing hydraulic and mechanical properties of MD membrane, these carbon composite membranes can still be wetted by low surface tension organics such as oils and surfactants limiting their potentials in wastewater treatment [58]. Subsequent sections discuss the applications of various nanomaterials including inorganic and carbon nanomaterials for treating oily wastewater particularly PW in MD, bringing to fore their merits, demerits and the advances made thus far. A brief overview of PW characteristics and contaminants of environmental concerns present in PW as well as prominent technologies for handling PW are first presented.

3. Produced water properties

The composition of PW water varies greatly across various fields [68], depending largely on the type of hydrocarbon present, the production method, geographic location of the field and the type of geologic formation. The properties and volume of PW can change along the production life of the reservoir [133]. Treated PW currently constitutes the largest waste stream discharged into the sea from offshore facilities globally [29].

The general categorization of PW constituents are the following: dispersed hydrocarbons, dissolved gases, dissolved organic and inorganic compounds, suspended solids and production chemicals [68]. So basically, these include salts (i.e. salinity or TDS), oil and grease (O&G), benzene, toluene, ethylbenzene and xylenes (BTEX), polyaromatic hydrocarbons (PAHs), phenols, organic acids, calcium, magnesium, sulphates, barium, biocides and corrosion inhibitors. Table 1 present a summary of major PW constituents and their typical range of concentration in oil fields and gas fields.

Most regulatory policies and requirements in the industry are targeted at O&G content. This would include dispersed oil and water soluble organics with the exception of low molecular weight phenols and acids as well as BTEX [133]. Removal of dispersed oil in PW is during primary treatment is particularly important as the discharge regulations are based on its concentration [124]. Owing to strict regulatory guidelines for discharge to the environment in oil and gas operations, treatment of PW by operators is unavoidable. Regulations become increasingly stringent as more evidence reveal the impact of PW to the environment (Witze, 2015). PW quality requirements will depend on the after-use of the stream: disposal or re-injection. For disposal, 15 ppm oil concentration can be achieved with an optimized treatment facility; 35 – 40 ppm for a treatment facility designed to meet discharge limits in accordance with most international guidelines for seawater disposal; 100 ppm for poorly treated PW stream [133]. Table 2 presents the current discharge regulations for various offshore fields around the world.

Table 1
Typical PW constituents for oil fields [4,158]

Parameter	Oil field	Gas field
Total oil/grease (mg/l)	2 – 565	2.3–60
Total organic carbon (mg/l)	500–2000	67 – 38,000
Total suspended solids (mg/l)	1.2 – 1000	8–5484
Total dissolved solids (mg/l)	247,000	2600 – 360,000
Chemical oxygen demand (mg/l)	1220	2600 – 120,000
Sodium (mg/l)	132–97000	520 – 120,000
Chloride (mg/l)	80–200000	1400 – 190,000
pH	4.3 – 10	3.1 – 7.0

3.1. Environmental footprint of PW

It is also important to consider the footprint of MD technology particularly as it relates treatment of wastewater with hybrid impurities such as PW, to assay its sustainability with regards to the environment. This can be viewed in light of its capacity to handle toxic component of PW such as dissolved salts, BTEX and radium isotopes [311], that may cause harm to environment which could in turn cause harm to human and animal health. For instance, radioactive contaminants in PW can cause cancer and dental fracture while a possible brain, liver and kidney damage may result from a combination of bromide with disinfectant chlorine that yields disinfection by-products [312]. In a study highlighting the ecological effects of PW discharge into the sea, Bakke et al. [23] reviewed endocrine, non-endocrine as well as genotoxic effects of contaminants in PW on fish close to discharge points. The effects are confined to 1–2 km from discharge outlets and the risk of widespread impact are generally low although information is scarce on such effects on fish populations or communities. In a pilot study to determine the fate of radium contamination in marine sediment during PW discharge leading to precipitation of naturally occurring radioactive materials (NORMs) the likes of radio-barite and/or radio-strontium as a result of PW mixture with seawater, Ahmed et al. [6] fully characterized the precipitates formed from their mixture as well as the radium uptake from sediments obtained from discharge sites in a bid to understand the mechanism of formation using XRD, X-ray fluorescence (XRF), SEM imagery and radioactivity. They confirmed the co-precipitation of radiostrontobarite and inferred its deposition in shallow marine environments due to lack of dispersion. They suggested further studies to highlight the environmental impact of such solid phase in marine environment of shallow water depth. Using PW discharge from 26 platforms, Nepstad et al. [198] also modelled the PAHs exposure in the early life of Atlantic cod using different simulation years to study

Table 2
Discharge regulation for oil and grease content in PW across various fields around the world.

Field/Location	Regulation/Regulator	Oil and grease limit (mg/l)	Ref.
Norwegian Continental Shelf (NCS) – Norway	Oslo-Paris Convention (OSPAR)	30	[68,271]
U. S. A	United States Environmental Protection Agency (USEPA)	29 (monthly average)	[133]
North Sea	OSPAR	30	[308]
Santos Basin – Brazil	CONAMA Resolution 393	20 (old resolution) 29 (new resolution)	[85,228]
Australia	–	30	[72,308]
Niger Delta – Nigeria	Department of petroleum resources (DPR)	40	[249]
Spain	MAH/285/2007	50	[133]
Upper Zakum, Arabian Gulf - UAE	ADNOC	20	[316]
Alberta - Canada	C-NLOPB CNSOPB	30	[158]
Kuwait	Kuwait Convention	40	[177]

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variability in annual exposure. They found that there is marked difference between PAH discharge concentrations and predicted internal concentrations with a factor 1000 less than concentrations usually associated with severe narcotic effects. A detailed review on environmental effects of offshore produced water discharges can be found elsewhere in the literature [29].

In onshore operations, risks posed to human and animal health as well as environment may be associated with poor handling of PW stream with spills occurring during transportation to disposal sites, surface discharges or groundwater contamination from deep wells [79,102] . Other common PW handling options that pose risk to environment include use of unlined retention/evaporation pits where PW is first disposed in a receiving pit where oil and grease are skimmed off after floating and the remaining water is transferred to another pit for evaporation [48]. Evaporation ponds use natural solar energy to evaporate waste water and concentrate brine to a sludge that is later on taken to a landfill for disposal [97]. This management option has been associated with air pollution, groundwater aquifer contamination, wildlife impact and even spills that have affected soil health [211], and represents about 40% of PW disposal in California [248]. Fig. 6a shows an illustration of how poor handling of PW can contaminate groundwater sources and pose risks to health while Fig. 6b depict cattle drinking from a seep that is adjacent PW disposal pond. The water oozes out from the ground about 300 feet from the unlined disposal pit, likely a seepage therefrom. Incidences in Louisiana and Pennsylvania where cattle were exposed to these wastewaters led to serious health complications and subsequent death [113].

Efforts have been made to optimize PW handling using pits. Although this process is slow and dependent on climate, mechanical evaporators can be installed to enhance evaporation. Pre-treatment steps to remove solids and oil is also necessary as floating oil in the ponds decreases the rate of evaporation [79]. Maugans and Banda [179] presented a work on fresh water and produced water storage in oilfield pits with priority focus including evaporation, volume monitoring, leaks and spill. They presented a plan for a digital evaporation pit that has gradually sloping edges and is lined (usually with HDPE) to avoid infiltration into the soil. The risk of overfills and leaks are minimized as level sensors indicate depth and volume with alarms. However, the problem of fouling due to microbial growth still remain a challenge for evaporation ponds . Hence, handling of PW streams effectively and efficiently is still necessary.

3.2. PW treatment trend

Primary PW treatment involves the use of gravity separator, otherwise known as skimmers, on onshore platforms to remove dissolved gases, larger dispersed oils and suspended solids. This primary treatment stage can reduce the dispersed oil content to between 50 and 200 ppm [206]. The secondary treatment stage involves further reduction of dispersed oil content by gas flotation technique which disperses gas bubbles in the water phase and their attachment to oil droplets [235]. Offshore platforms often have the flotation unit in either the dissolved gas flotation (DGF, where gas bubbles are generated by nucleation) or in induced gas flotation (IGF, where gas bubbles are generated by mechanical dispersion) and the process can effectively lower the concentration to below the discharge requirement limits [68]. Tertiary treatment for PW otherwise referred to as polishing aims to reduce the concentration of dissolved oils to below 10 ppm. The processes involved at this stage are gas stripping, biological treatment, coalescing filters, walnut shell filtration, etc., although it is mostly used onshore [80].

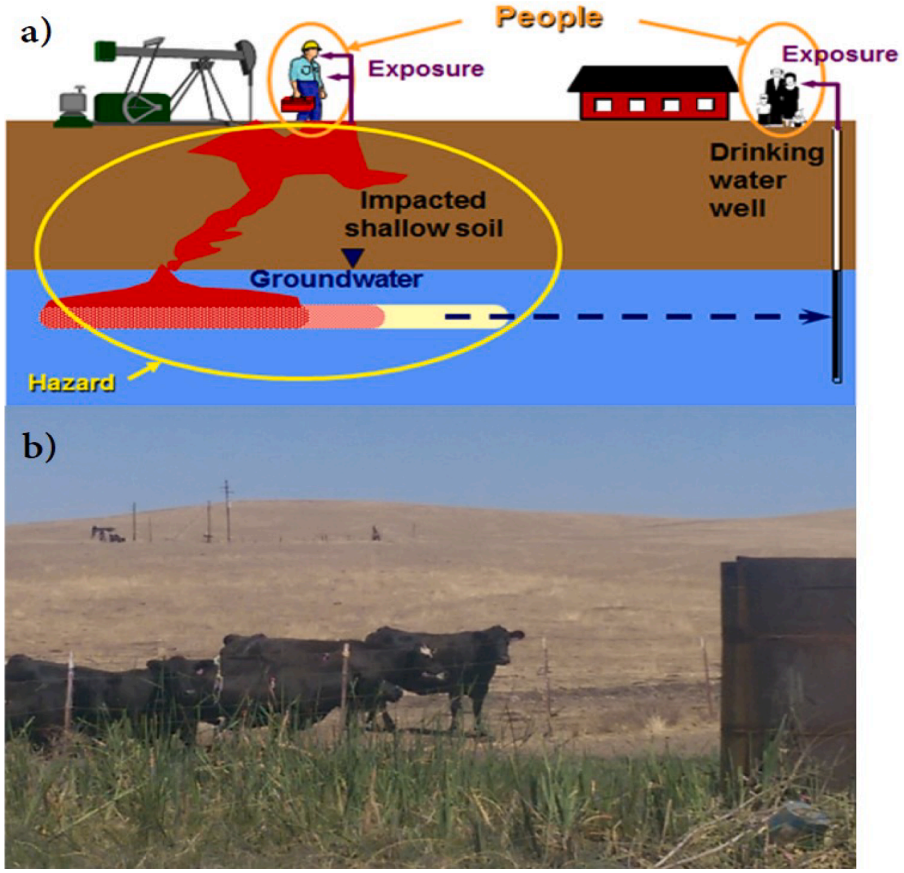

Fig. 6. a) Illustration of possible pathways to groundwater contamination and health risks b) Image of a seep 300 ft from a PW disposal pit in Kern – California [79,113]

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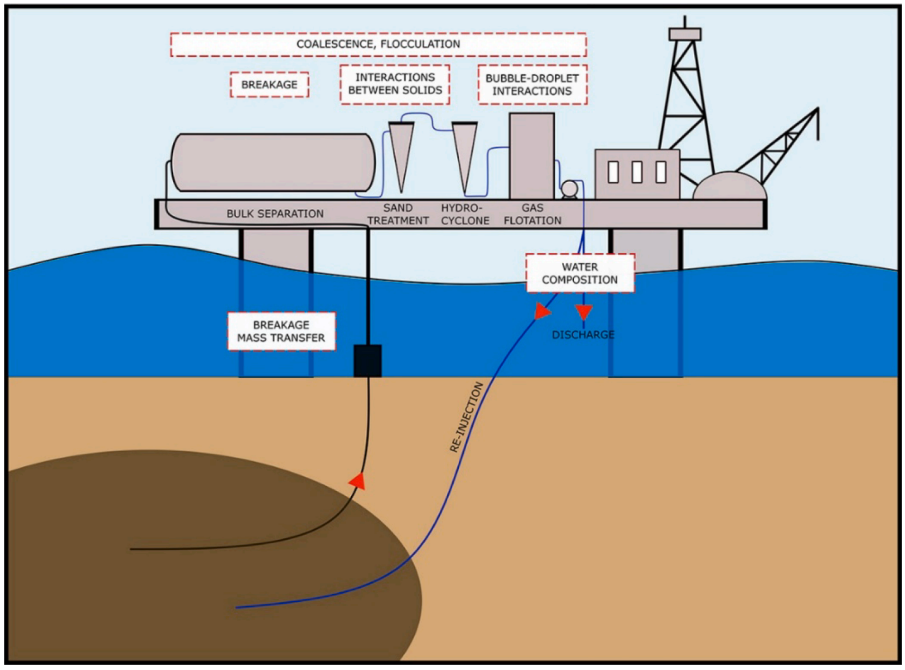

Fig. 7. Schematic illustration of a typical offshore produced water treatment facility for re-use/disposal limits [68]

Fig. 7 shows a typical offshore produced water treatment facility from the primary separation of phases to secondary and tertiary treatments aimed at reducing oil-in-water concentration and subsequent disposal to the sea or re-injection for use in EOR.

Membrane technology has been put forward for effective treatment of PW with the capacity of reducing oil concentrations further below 5 ppm, following primary treatment [60,288–289] . Several works have highlighted the use of various filtration membranes for treatment of PW with remarkable results [38,56,103,146,187,288–289] . These methods however either had fouling related issues [255]. RO can desalinate PW and remove last traces of dissolved and dispersed oil. It is also capable of removing various heavy metals in PW. The main drawback of RO especially for treatment of hypersaline feeds like PW is build-up of osmotic pressure with increasing salt concentration in the feed which eventually exceeds the operating pressure of the system. It also requires pre-treatment steps to reduce the effect of membrane fouling and it is energy-intensive [3,159,209,285] .

Primary treatment steps are important to properly isolate the PW for further secondary and/or tertiary treatment steps which further improve the PW quality by eliminating suspended solids, reducing oil concentrations and stripping dissolved gases [21,33,52,150,222,235] . However, these methods do not address the very high salt concentrations in PW. Conventional thermal desalination processes such as MED, brine concentrators MSF are costly with high energy demand and often requiring feed pre-treatment to remove organics [89,183,193,199,260] .

Recent advanced membrane processes that are popular for desalination of PW include ED and MD. ED systems can operate at relatively higher concentrations of organic foulants in feed water than RO systems, where charged compounds are targeted and removed in feedwater via an electrical potential difference-driven process across a membrane stack [129,213]. Flushing allows for less dependence on use of pre-treatment chemicals. However, the process is also highly energy dependent and hence means more costs. MD on the other hand is capable of 100% TDS removal at very low operating pressures while not being affected by osmotic pressure build-up, thus surpassing the drawbacks of RO [4,105,258,270].

4. Produced water handling using MD

The suitability of MD in treating hypersaline wastewater with hybrid impurities typical of PW using membranes with dedicated anti-wetting and anti-fouling properties has been reported [243]. This offers the potential of re-collecting high purity water for domestic use on offshore platforms as well as for irrigation agriculture [210], for fields situated onshore. Pre-treatment of PW prior to subjecting it to MD is important in ensuring integrity and longevity of the MD system. As discussed earlier, pre-treatment processes can reduce the oil-in-water concentrations to as low as 10 ppm when PW polishing is involved [80]. Similarly, the effect of salt concentration in the feed solution is negligible on MD processes compared to other pressure driven processes like RO and NF, since it is a thermal process and as such not affected by osmotic pressure build up [12,279–280,299] . However, presence of divalent ions of magnesium and calcium in PW feed often led to scaling on the MD membrane surface in addition to fouling by organic species which together compromise the process integrity of the MD system [82,117] . Thus, it is thus important to understand the mechanism of organic and inorganic fouling particularly for PW feed in MD. However, more effort is needed to design optimum anti-fouling membranes with stable rejection of salts and organics over longer operation time. Subsequent sections highlight fouling and scaling problems for MD and advances in mitigation strategies for treatment of PW.

4.1. Membrane fouling in MD for PW

A critical review of fouling mechanism on the membrane surface and within the pores by emulsified oils for pressure driven membranes was done by Tummons et al. [265]. They summarized a number of mechanisms by which dissolved species affect emulsion stability to include disturbance of partition between the oil and aqueous phase and salt-induced changes in critical micelle concentration of surfactants amongst others. The interaction between oil droplets with the membrane surface and other species such as salt and surfactants govern membrane fouling. Studies of oil fouling behaviour with MD membranes via evaluation of surface energy interaction have also been reported [162,292,315] . However, study elucidating MD membranes fouling by

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surfactant stabilized oily wastewaters is scarce, owing to membrane pore wetting issues which inhibits the oil–water separation [270]. Surfactant-like compounds (commonly used as emulsifiers and dispersants) are present widely in saline waters, such as oil and shale-gas PW and dyeing wastewater [54]. Through significant reduction of the LEP by surfactants, commercial hydrophobic membranes are easily wetted [278] . The surfactant lowers the surface tension of feed solution because of its amphiphilic nature that allows the passage of water with its dissolved salts across MD membranes, instead of just water vapor.

Chew et al. [46] studied the mechanisms involved in the interactions between PVDF membrane surface and a surfactant-stabilized emulsion. The results revealed that the surfactant concentration had an influence on membrane fouling and wetting behaviour. Hou et al. [117] also studied the effect of surfactant SDS wetting on MD process with PVDF and PTFE membranes. The mechanism of fouling was quantitatively studied by extended Derjaguin-Landau-Verwey-Overbook (XDLVO) theory. Using XDLVO theory which constituted the acid-base (AB) interactions, Lifshitz-van der Waals (LW) attractions and electrostatic (EL) repulsion, they demonstrated the interfacial free energy between the different \membrane surface chemistries (hydrophobic and omniphobic) and the SDS aggregates in the aqueous media. They showed that deposition of SDS colloids on the membrane surface is majorly attributed to attractive forces between the membrane surface and foulant. Fourier transform infrared spectroscopy (FTIR) analysis of the distillate side of the membrane confirmed the permeation of SDS during the DCMD test.

Table 3 presents a brief summary of methods that have been used to elucidate membrane fouling for MD of PW. While EDX, SEM, FESEM and FTIR have often been used to study foulant constituents typically after MD operation, the fouling behaviour itself for MD of PW is scarcely reported. XDLVO offers the advantage of analysing the interactive energies between the membrane surface and fouling species to better understand the conditions for their attachment or repulsion. Similarly, the direct observation through the membrane (DOTM) offers real-time study of the fouling during the MD operation. However, the technique even though cheap and easily accessible, has a low resolution which hinders detection of smaller sized foulant and is limited to two-dimensional imaging with limitations towards investigation of depth [227,264] .

Other improved techniques that have been used in fouling studies for pressure driven membranes include optical coherence tomography (OCT) [109,107,164,264] and electrical impedance spectroscopy (EIS) [115,262,304]. As posited by Rudolph et al. [227], it is possible to study the thickness, concentration, composition distribution and structural properties in high resolution with these improved techniques although no one method can avail all the information to enable complete monitoring of fouling progression. It is also interesting to note that there is little information in the literature regarding the use of these techniques for MD fouling studies, much less so for studies focusing on fouling by PW in MD.

4.2. MD with PW feed

One of the early works in the treatment of oilfield PW in MD was reported by Macedonio et al. [176]. Pre-treatment of the PW was done using microfiltration and activated carbon in an effort to remove oils, suspended solids and hydrogen sulphide, H_2S. The DCMD configuration employed fabricated PVDF membranes as well as commercial PP membranes fed with wastewater with the salinity of 247,900 ppm and the TOC of 18.1 ppm. The membranes exhibited TDS rejection of above 99.7% and TOC rejection of up to 92.9 % in a 7 h operating time. MD PW treatment cost analysis revealed 1.28 $/m^3 and 0.72 $/m^3 for the plant feed temperatures of 20 °C and 50 °C, respectively. However, there was no mention of membrane fouling analysis at the end of the MD process.

Chew et al. [46] prepared a synthetic surfactant-stabilized oil-in-water emulsion to mimic real oilfield PW usually having oil concentration range of 2 – 565 ppm [80], for desalination in a DCMD process using PVDF membranes in a bid to understand the relationship between the membrane surface and the emulsions during the process. The results revealed that membranes with better anti-fouling and anti-wetting properties are required for prevention against fouling and to do that, surface modification to impart such properties is necessary. Similarly, Lokare et al. [165] studied the fouling behaviour of PW from unconventional gas well of the Marcellus Shale in Pennsylvania with TDS of up to 300,000 ppm using commercial hydrophobic MF membranes in DCMD. The TOC from one of the sites was about 19 ppm. Long term experiment to assess fouling propensity carried out for 72 h revealed 99.99% dissolved solids rejection and 78.9% TOC rejection. SEM images of the used membranes revealed dark parches of scaling which was confirmed by EDX to be sodium chloride deposits.

Recently, Zhang et al. [306] carried out a study that focused on treating shale oil and gas PW from Wattenberg field – Colorado, in DCMD with two pre-treatment steps of precipitative softening (PS) and walnut shell filtration (WSF), using commercial PVDF flat sheet membranes. The PS and the WSF pre-treatment significantly reduced the

Table 3
Summary of methods used to study fouling for MD of PW.

Method	Mechanism	Interpretation	Ref.
XDLVO	Energy interaction between a colloid and membrane surface expressed as the sum of Lifshitz-van der Waals (LW), acid-base (AB) and electrostatic double layer (EL) interactions. A negative value for the summation of interactions indicates an attraction with a lower value showing a stronger attraction, while a positive value indicates a repulsive force.	For emulsified water, a high negative summation of the interactions implies severe membrane fouling by organics while a high positive value implies less susceptibility to fouling.	[44,106,117,174,254,315]
Force spectroscopy	Oil-probe interaction with membrane surface is studied at 3 stages i.e approach event; probe driven towards membrane at a constant speed, contact event; probe contacts the membrane surface, compression event; probe is further driven towards them membrane after contact with oil. Probe is then retracted until oil droplets splits. Shape of force curve gives insight on oil-membrane interaction.	A positive attractive force is observed upon contact for a membrane that is attractive to oil droplet. A negative attraction means there is repulsion.	[108,116,272,280]
Gas/Water permeation	Dry gas or pure water is passed through the pristine and used membranes and the differences in flux gives an idea of fouling propensity.	Effect of temperature polarisation during real MD test may result in discrepancies.	[46,166]
Molecular dynamic simulation	A simulation and visualization steps are used to create a forcefield model and study the interaction between membrane surface and fouling species.	Interaction energies between the membranes and the fouling species give an insight on their fouling propensity for the different feed formulations.	[55,182,252,270]
DOTM	Fouling evolution is visualized in real time using a camera coupled to a light microscope with the objective focused on the membrane surface.	The rate of change of the surface coverage by the foulant is captured at intervals and it is in turn used to elucidate degree of fouling through the MD process.	[253,256,254]

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concentration of organic foulant in the feed PW prior to the MD process. A combination of PS and WSF reached up to 98% BTEX removal with salt rejection up to 99.9% at a recovery of 82.5%. This process also reduced the total volatile petroleum hydrocarbon (TVPH) concentration to only 0.9 ppm in the distillate, way below the discharge limit requirements. On the other hand, the treatment of raw PW in MD without pre-treatment resulted in only about 40% recovery with vapor transport resistance resulting from membrane pore blockage caused by organic foulant, organic scalants and suspended solids present in the raw PW. This further buttressed the importance of pre-treatment prior to MD for shale oil and gas PWs.

In a very recent work, Zou et al. [313] reported the desalination of PW from the Permian basin using a novel hollow fiber membrane in a 200 h continuous DCMD process. The PW had a TDS concentration of 154,200 ppm and non-purgeable organic carbon (NPOC) content of 57.6 ppm. In this study, the salt rejection of 99.99% was stable for the first 24 h after which it began to decline down to about 98.4% for sodium and chloride ions at the end of the 200 h. The permeate flux also declined from 25.41 kg/m^2·h to 15.21 kg/m^2·h. This decline was attributed to a likely partial membrane pore wetting caused by foulant. The examination of the membrane surface morphology after the DCMD operation confirmed the membrane was covered by multi layers of particulate deposition. EDX analysis further revealed that the chemical compositions of the depositions as containing calcium carbonates which could have significantly affected the membrane flux. Attenuated total reflection-FTIR (ATR-FTIR) also revealed the presence of proteins, aliphatic hydrocarbons and humic substances usually present in oilfield PW. This is buttressed by the fact that only about 5 ppm of NPOC was detected in the distillate. They further demonstrated that the membrane fouling can be effectively reduced from 40% to 10% by simple physical cleaning with permeate water from the DCMD process.

The requirement for achieving omniphobicity on the surface of membranes involves a combination of a very low surface energy and surface roughness giving a re-entrant structure that provides a kinetic barrier for transitioning from the stable Cassie-Baxter state to the fully wetting Wenzel state for low surface tension liquids such as hexadecane, SDS, ethanol, oils etc [153] as demonstrated in Fig. 8. The air pocket is depicted to reduce the contact angle between the membrane surface and the feed liquid thereby increasing the omniphobicity, and also reported to increase the permeate flux making it ideal for treating oily saline wastewater in MD [298].

Boo et al. [32] fabricated the first omniphobic membranes for the treatment of shale gas PW from the Texas Permian Basin in MD by grafting PVDF surface with SiNPs followed by coating with fluoroalkyl silane (FAS) to lower the surface energy. (3-Aminopropyl) triethoxysilane (APTES) functionalization of the pristine PVDF surface introduced NH^+ ions which enable the grafting of SiNPs (having negative surface charge) through electrostatic adhesion to give a stable bond. The spherical SiNPs created a re-entrant structure on the membrane surface in addition to inducing the membrane surface roughness while the FAS (perfluorodecyltrichlorosilane, FDTS) ensured air pockets between the SiNP spheres; a requisite for omniphobicity. The modified PVDF surface showed CA>150° for water and > 130° for SDS and mineral oil. Mineral oil-in-water concentration reaching 80 ppm in the synthetic feed solution with a corresponding salt concentration of 1.0 M (58.44 g/L) NaCl were used in this study. The performance of the modified membrane in the 8 h DCMD process showed a complete salt rejection and stable water flux with increasing mineral oil concentration (of up to 80 ppm). When the system was challenged with real pre-filtered shale gas PW containing surfactants, O&G, total volatile organics, TSS and TDS of 3.2 ppm, 2.0 ppm, <0.14 ppm, 41 ppm and 101,000 ppm respectively, the modified membrane exhibited stable performance with stable flux of around 13.6 kg/m^2·h. Interestingly, contrary to the hydrophobic control PVDF membrane, the modified membrane displayed resistance to surface attachments of mineral oil droplets expected to occur through nonpolar-nonpolar interaction.

In order to improve anti-fouling properties and reduce oil adhesion onto the membrane surface, Wang and Lin[281] used low surface energy perfluoroalkyl functional groups to modify composite membrane surface as well as SiNPs to induce roughness; together imparting omniphobic properties on the membrane. Spray coating of the CTS/PFO-PVDF composite with fluorosurfactant (FS) ethanolic solution to further lower the surface energy was carried out in this study. The composite membrane was challenged with crude-oil in water emulsion with oil and salt concentrations of 1000 ppm and 35,000 ppm respectively in an AGMD process. The composite membranes showed underwater oil CA of above 130° confirming oleophobicity. Ultrapure distillate with no oil permeation and salt rejection above 99.9% was achieved with the CTS/PFO-PVDF membrane which performed the best in MD fouling mitigation. Apart from exploiting surface chemistry, membrane fouling in MD can also be managed through formation of surface patterning. The patterns promote mixing nearby membrane surface that sway away foulant [141,178]. Such approach is attractive to be implemented for developments of MD membrane customized for PW treatment. Fig. 9 is a

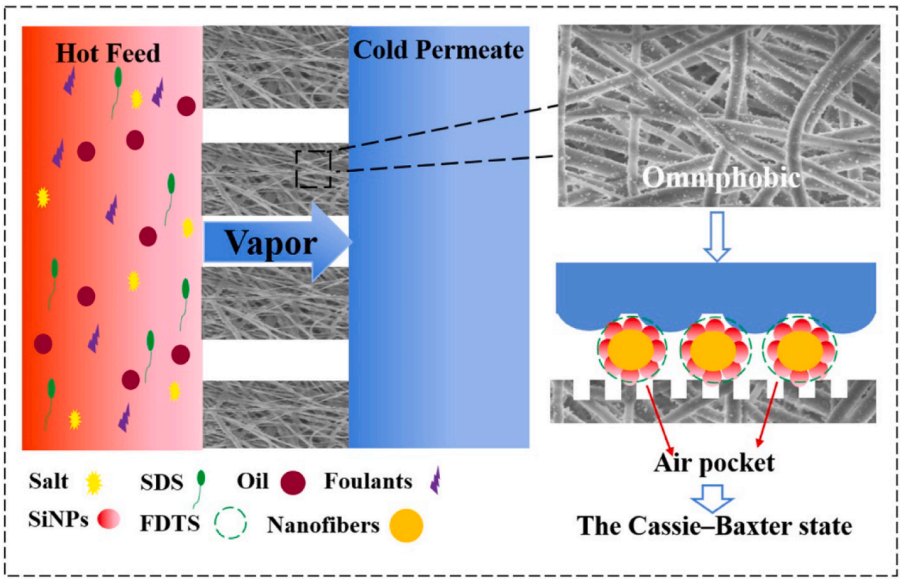

Fig. 8. Omniphobic membrane surface for robust separation of oily saline wastewater in MD [298]

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Fig. 9. Photographic image of crude oil emulsion feed solution (left) and the MD distillate (right) [281]

picture of the crude oil-in-water emulsion before and after treatment in MD.

In a similar study for shale gas PW, Lu et al. [168] modified PVDF hollow fiber membranes with SiNPs followed by Teflon AF 2400 coating to lower surface energy. The feed solution having 3.5% NaCl and SDS concentration increased at intervals until it reached a concentration of 0.6 mM were used in this experiment. The modified membrane exhibited good rejection toward liquids including water, ethylene glycol, cooking oil as well as ethanol and stable performance in a 7 h VMD test with membranes exhibiting salt rejection of higher than 99.5%. It showed a stable flux average of 11.9 kg/m^2·h throughout the experiment owing to good repulsion of low surface energy feed, with recovery of about 90%. Table 4 gives a summary of other efforts in treating PW in MD with various polymeric membranes. It is evident that the performance of polymeric membranes in PW treatment is critically affected by their surface chemistry being either hydrophobic, superhydrophobic or omniphobic. PTFE polymer has stronger hydrophobicity to repel water, but also poses higher oil affinity. PVDF has been frequented for studies of PW in MD owing to superb processability, cheap cost and availability. Polymeric membranes used without modification of surface chemistry towards omniphobicity suffer from oil fouling resulting from hydrophobic-hydrophobic attraction between membrane surface and hydrophobic tails of oil. Presence of surfactants has been shown to severely aggravate membrane surface and pore wetting that subsequently facilitates salt transport across the membrane leading to MD failure. Pre-treatment of PW has proved helpful in reducing concentration of fouling species making subsequent PW treatment in MD practicable. However, scaling by salt present in the feed remains an issue for MD membranes. Omniphobic membranes offer potential for achieving high purity permeate and simple physical cleaning like flushing suffices for any scalants that may have adhered to membrane surface. While VMD may offer more flux, cost is incurred from installation of more pumps, making it not economically viable. DCMD has been favourably more studied due to ease of assembly and remarkable performance. However, it also requires improvement to address issue of temperature polarization due to direct contact between hot feed and membrane surface. An MD set-up utilizing alternate heat sources such as solar or geothermal energies with dedicated superamphiphobic membranes capable of robust anti-fouling and surfactant wetting will be ideal for high purity permeate recovery from adequately pre-treated PW feed. It is important to also consider the effect of surface modification of membranes on eventual performance properties like porosity, thickness and durability for prolonged performance as well as the complex chemistry of real PW feed. As such, the selection of a cheap and appropriate pre-treatment option particularly targeting organic as well as scale-forming species and subsequent subjection to an MD process using high flux omniphobic membranes is necessary.

4.3. Membrane scaling and mitigation for MD of PW

Inorganic fouling largely refers to scale formation on the membrane surface that entails the precipitation of salts such as NaCl, calcium carbonate ($CaCO_3$) or calcium phosphate ($Ca_3(PO_4)_2$) resulting from nucleation and crystallization on the feed side consequent upon temperature changes and evaporation of water ([15]. Monovalent ions such as sodium are also likely to form aggregates when they interact with organic functional groups, although they were found to not likely deposit on the membrane and cause flux decline [67,196]. Pore wetting study using molecular dynamic simulation by Velioglu et al. [270] further revealed that NaCl increased the SDS-PVDF affinity leading to decreased surface tension and worsening of pore wetting occasioned by increased likelihood of LEP exceedance. This buttressed their earlier findings that it was not the oil itself that affected the process but a coupling of SDS and salt [105]. Similarly, Zou et al. [313] also reported that increased salt concentration in the feed resulting from continuous water vapor transfer had caused scale formation that clogged the membrane pores ultimately leading to flux decline.

Multivalent ions are however more prone to formation of scaling than monovalent ions [27], with calcium and magnesium ions being predominant [82]. When the solubility limits of the constituent minerals is exceeded, scales are deposited on the membrane surface [111]. Scaling by salts present in PW is one of the threats to sustainable MD operations. This has over the years received attention from researchers in a bid to overcome fouling and scaling related issues in MD. Like fouling by organic species, inorganic scaling in MD has been studied by a number of methods, including the use of SEM imagery of used MD membranes and EDX to analyse the constituent of salt scaling on MD membrane surface [166,296,313], as well as simulation studies [270]. Recently, more accurate online methods like light microscopy (LM) and OCT have been proposed for better quantification and analysis of the scaling layers using non-invasive in-situ visualization [25,145;154;185]. Despite being a more expensive method, OCT allows for detection of 3D datasets enabling the volumetric calculation of scaling layers [26].

Scaling inhibitors and pH control are a common way of controlling scale formation and subsequent deposition [53]. Inhibitors prevent the crystal nucleation and growth while controlling the pH between 5 and 7 prevents the formation of magnesium and carbonate salts by keeping their ions in solution [10,39;110;175]. However, these methods do not

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Table 4
Summary of articles on MD membranes for PW treatment showing various configurations, polymer materials, fabrication and modification methods.

Year	Polymer Material	Synthesis and modification method	Module and configuration	Pre-treatment	Feed/ permeate inlet Temp (°C)	Impact	Performance	Ref.
2020	PVDF membranes	PVDF hollow fiber membranes synthesized via dry-jet wet spinning with Janus modification showcasing an outer superhydrophobic SiNPs-covered layer and an inner hydrophilic PVDF-PEG layer for treatment of PW from a facility in Carlsbad, New Mexico.	Hollow fiber in DCMD set-up.	PW with TDS of 154,220 ppm and NPOC of 57.6 ppm was pre-treated with a spin down sediment water filter to eliminate solids in suspension having diameters>50 μm.	85.1/20	Scales of salts and dissolved organic matter were found to have accumulated on the membrane surface effectively affecting process performance. Physical cleaning of membrane with permeate water showed potential to inhibit performance decline from 40% to 9.7%.	LEP: - WCA: 137.6° Flux: 25.41 – 15.21 kg/m^2·h Rejection: >98.4% Operating time: 200 h	[313]
2020	PSF membranes	Phase inversion solution casting with PEG as pore former and varying NPs (SWCNTs, MWCNTs, Al_2O_3-NPs and CuO-NPs) for MD treatment of oilfield PW from Sert basin - Libya	Flat module in enhanced DCMD set-up	–	60/20	PSF membrane with 0.5% SWCNT showed the most flux of 20.91 kg/ m^2h when testing with synthesis brine of salinity of 123,136 ppm exhibiting 99.99% rejection. Hence it was used for saline oilfield PW treatment. However, no report of membrane fouling analysis was given.	LEP: - WCA: 73 ± 2.5° Flux: 5.97 kg/ m^2·h Rejection: >99.99% Operating time: 8 h	[240]
2019	Electrospun cellulose acetate (CA)	Electrospinning of CA nanofibers and surface modification with SiNPs using sol–gel method followed by CVD of FAS to lower surface energy imparting omniphobic properties for PW treatment.	Flat nanofibrous module in DCMD setup	Emulsion mimicking PW was used as feed with addition of SDS up to 0.4 mM	70/20	Addition of surfactant SDS in increasing concentration had no effect on the performance of omniphobic nanofibrous membrane as it exhibited excellent wetting resistance as well as resistance to mineral oil and ethanol.	LEP: 58 kPa WCA: 150.6 ± 4.0° Flux: 68 kg/m^2·h Rejection: ~100% Operating time: 10 h	[61]
2018	Commercial PVDF, PP and PE hollow fiber membranes	Membranes were used without modification, for treatment of shale gas PW form Eagle Ford, Texas – USA.	Hollow fiber module in a DCMD set-up	Pre-treatment in a vortex-based anti-fouling membrane (FMX-B), flocculation-sedimentation (FS) and flocculation-sedimentation-microfiltration (FSMF) reduced DOC, TDS and turbidity to 110.9 ppm, 87,000 ppm and 0.55 NTU, respectively.	70/20	MD test with raw and treated PW had significant differences in initial flux and flux decline ratios with the latter showing lesser declines. Similarly, PVDF membranes had the higher initial flux as well as the highest decline. Post-DCMD membrane fouling analysis with SEM revealed lesser fouling by organics for pretreated feed. Removal of DOC ranged from 86.3% to 91.7%	LEP: 270 kPa WCA: - Flux: 5 kg/m^2·h Rejection: >99.9% Operating time: 33.3 h	[49]
2018	Polyoxadiazole membrane	Phase inversion hollow fiber spinning without		Pre-treatment by aeration was carried out at 50 kPa for 24 h	70/20	PW with TDS concentration of	LEP: 550 kPa WCA: 120°	[296]

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Table 4 (*continued*)

Year	Polymer Material	Synthesis and modification method	Module and configuration	Pre-treatment	Feed/ permeate inlet Temp (°C)	Impact	Performance	Ref.
		further surface modification for treatment of real produced water supplied by Saudi Aramco	Hollow fiber module in DCMD set-up	and the top-most clear solution was taken as the feed.		84,000 ppm and TOC of 41 ppm were treated in DCMD and the result revealed high salt rejection and a TOC concentration of 1.6 ppm in the permeate. SEM and EDX of the hollow fibers after the test revealed salt scaling on membrane. However, physical cleaning was sufficient to ensure membrane reusability as no severe fouling was observed.	Flux: 12.5 – 11.8 $kg/m^2{\cdot}h$ Rejection: >99.7% Operating time: 100 h	
2018	PVDF-HFP nanofibrous membrane	Electrospinning of PVD-HFP with F-POSS synthesized by thiol-ene click reaction of vinyl-POSS and perfluorododecanethiol under UV irradiation.	Flat nanofibrous module	Emulsion with progressive addition of SDS surfactant in the MD feed.	60/20	The modified membrane showed wetting resistance towards water, 3 mM SDS solution, mineral oil and ethanol with corresponding contact angles of 154.5 ± 2.6°, 149.5 ± 4.7°, 148.8 ± 0.7° and 128.2 ± 6.2°, respectively. The pristine PVDF-HFP membrane however showed instant wicking by mineral oil and ethanol. SDS addition during MD sharply reduced flux for pristine PVDF-HFP membrane.	LEP: - WCA: 154.5 ± 2.6° Flux: ~9 $kg/m^2{\cdot}h$ Rejection: >99.9% Operating time: 8 h	[168]
2018	Hydrophobic, superhydrophobic and composite PVDF membranes	Hydrophilic layer coating was synthesized on the hydrophobic PVDF to give Janus properties and SiNPs with fluorination by FAS in another batch for treatment of PW from Wattenberg field in northeast Colorado.	Flat sheet module in a DCMD set-up	–	60/20	The Janus membrane had an under-water oil CA of 160 ± 2° while the omniphobic membrane exhibited in-air hexadecane CA of > 100°. PW samples with TDS of 40,000 ppm and TOC up to 1460 ppm were used as feed for the DCMD experiment. The Janus membrane had higher initial flux as well as the best fouling resistance when treating PW sample. SEM and EDX revealed inorganic scaling and organic fouling to be responsible for flux decline. However, omniphobic membrane demonstrated	LEP: - WCA: 159 ± 2 Flux: 26.2 ± 0.2 $kg/m^2{\cdot}h$ Rejection: >99.9% Operating time: ~ 19 h	[66]

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Table 4 (*continued*)

Year	Polymer Material	Synthesis and modification method	Module and configuration	Pre-treatment	Feed/ permeate inlet Temp (°C)	Impact	Performance	Ref.
						reusability above the others.		
2018	PVDF membrane	PVDF phase inversion flat sheet membranes were modified by layer-by-layer assembly of silica aerogel and FTCS to give omniphobicity for treatment of RO brine from coal seam gas (CSG) PW.	Flat sheet module in AGMD set-up	Pretreatment of CGS PW feed was by RO resulting in a highly concentrated brine for feeding into MD.	60/20	Addition of SDS in the feed significantly reduced the LEP of the modified membrane (17 kPa). However, the modified membrane showed stable flux and excellent salt rejection even with SDS concentration reaching 0.5 mM. Similarly, addition of 20 ppm humic acid had no effect on the modified membrane performance.	LEP: 282 ± 0.13 kPa WCA: 177 ± 0.4° Flux: 11.22 kg/ m^2·h Rejection: >99.9% Operating time: ~ 72 h	[290]
2017	Commercial PVDF membrane	Pristine commercial PVDF were challenged with oily saline wastewater mimicking PW to study the effect of SDS, oil and NaCl in the feed.	Flat sheet module in DCMD configuration	–	65/15	The effect of SDS, oil and NaCl in the feed and their interaction with the membrane surface during MD process was systematically studied. Higher oil concentration (200 ppm) resulted in more pronounced flux decline occasioned by membrane fouling and increased permeate conductivity regardless of SDS presence. SDS addition stabilized the emulsion and reduced fouling but increased wetting by reducing surface tension of the liquid.	LEP: 110 kPa WCA: 111° Flux: 19.2 – 19.5 kg/m^2·h Rejection: >99.9% Operating time: ~ 20 h	Han et al., 2017b
2017	PTFE membrane	PTFE flat sheet membrane on a PE support net for membrane distillation crystallization of Shale Gas PW from the Marcellus Shale, Pennsylvania – USA.	Flat sheet module in a modified DCMD set-up integrating crystallization.	–	60 ± 0.5/ 20 ± 0.5	SEM, EDX and X-ray diffraction (XRD) were used to analyze membrane wetting mechanism and LEP measurements to investigate the effect of O&G on membrane wetting. Results revealed O&G concentration above 150 ppm led to a sharp decrease in LEP signifying wetting. SEM-EDX results shows spherical scales on membrane synonymous with shape of calcium carbonates and barium carbonate in the second PW sample. Integration	LEP: ~300 kPa WCA: - Initial flux $_{(SGPW1)}$: ~13 kg/ m^2·h Initial permeate conductivity: 1 µS/cm Operating time: -	[147]

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Table 4 (*continued*)

Year	Polymer Material	Synthesis and modification method	Module and configuration	Pre-treatment	Feed/ permeate inlet Temp (°C)	Impact	Performance	Ref.
						with crystallizer allowed for longer process without inorganic scaling and raised recovery by 62.5%.		
2017	Commercial PVDF membrane	PDA/PEI was deposited on the PVDF substrate to produce an in-air-hydrophilic/underwater superoleophobic composite membrane for water recovery from PW	Hollow fiber module in a DCMD set-up	Surfactant-stabilized petroleum-in-water emulsion containing 450 ppm of petroleum, 50 ppm of surfactant (dodecyltrimethylammonium bromide, DTAB) and TDS (3.5 wt% NaCl), respectively mimicking oil, surfactant and salt present in real PW was prepared as feed solution.	60/20	The modified membrane displayed underwater superoleophobicity maintaining a stable flux for up to 7 days of operation. Surface hydration and electrostatic repulsion provided by the hydration layer resultant from the interaction of hydrophilic moieties on membrane and water molecules made it difficult for oil attachment to membrane surface.	LEP: - WCA: 25.7 ± 4.0° Flux: ~13 kg/ m^2·h Rejection: >99.9% Operating time: ~ 150 h	[46]
2016	PP membrane	Phase inversion casting of PP blended with MWCNTs to improve membrane performance for treatment of PW supplied by Gemsa Petroleum Company – Egypt	Flat sheet module in a vacuum enhanced DCMD (VEDCMD) set-up	Pre-treated PW samples were collected at the point of disposal with TDS of 231985 ppm and oil-in-water concentration of 5 ppm	55/15	The MWCNTs enhanced the flux performance of the membrane by 58% and preventing pore wetting, in comparison to the neat membrane. The effect of polymer concentration, membrane thickness, feed temperature and feedwater concentration were studied. No membrane fouling analysis reported.	LEP: - WCA: 118° Flux: 19.6 kg/ m^2·h Rejection:99.99% Operating time: 6 h	[201]

remove the ions but keep them in 'check'. Pre-treatment by softening technologies has proved effective towards removal or reduction of divalent cations like Ca^{2+} and Mg^{2+} that can react to form scales [82]. Electrodialysis (ED) has been suggested to be suitable for softening PW by preferential removal of multivalent ions in seawater with desired process conditions [28,86]. Sosa-Fernandez et al. [247] explored the utilization of ED to desalinate polymer flooding PW, particularly targeting multivalent ions as they increase the risk of scaling as well as reservoir souring. They demonstrated that it was possible to remove divalent cations, including sulfate (anion), when employing low current and high temperature of 24 A/m^2 and 40 °C respectively. Other works have also highlighted the potency of using ED to isolate divalent ions from saline feeds [100,157;246;294] with increasing attention from researcher, although further efforts are required to reduce the energy consumption of the system [63]. Nanofiltration is another membrane-based techniques for PW softening [229], although it has a limitation of having the ability to remove only divalent salt ions and is affected by scaling and fouling on the membrane surface [138,286].

Several works have featured modifications to polymeric membranes using organic and inorganic nanomaterials to impart special surface properties for robust anti-fouling and anti-scaling application in MD including omniphobic membranes [172], Janus membranes [313] and slippery surface designs [139]. For instance, in addition to lowering surface energy and creating re-entrant structure (a requisite for omniphobic modification), Chen et al. [45] created a slippery surface (OMNI-SLIP) by heat-pressing spun fibers at 170 °C and dip-coating in Teflon AF 2400. The resultant membrane with a sliding angle of 10.79 ± 1.63° prevented scaling by $CaSO_4$. The fact that MD is not affected by osmotic pressure build-up puts in the limelight for handling hypersaline feeds such as PW. However, efforts are continuous to improve the process integrity of the MD system for sustainable and long-term operation, amongst which is integration with other technologies that serve as pre-treatment in order to isolate MD membranes from complex species in PW feed.

4.4. MD integrated with other technologies for PW handling

It is important to highlight efforts to reduce the inherent effect of fouling and scaling on MD membranes by complex feeds and their overall effect on the system performance through combination of other

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processes with MD in an integrated or hybrid set-up. Prime amongst them with the aim to pre-treat the PW before feeding to MD for desalination is incorporating forward osmosis (FO) to MD (FO-MD) [13,114,305] , EC-MD [232] as well as coupling with crystallization unit to achieve zero liquid discharge (ZLD) [147]. While most studies have highlighted their application in other industries [194,245,300], specific studies focusing on handling raw PW streams are scarcely reported.

In an earlier study, Sardari et al. [232] combined EC and MD for treatment of high salinity PW. Turbidity, TSS and TOC were removed by 96%, 91% and 61% respectively using the EC phase. Subsequent treatment in DCMD using ethylene chlorotrifluoroethylene (ECTFE) flat sheet membranes resulted in fouling and scaling on the surface of the membrane confirmed by brownish cake layer from digital and SEM imagery, leading to significant decline in water flux. In subsequent effort to overcome the problem of fouling for MD hybrid, Sardari et al. [233] introduced FO to their erstwhile EC-MD hybrid. The EC first reduced suspended solids and organic foulants in PW prior to feeding into FO-MD system. This achieved efficient removal of TOC, TSS and water recovery of up to 78%, 96% and 76% respectively, in a prolonged operation. However, membrane modification is required to overcome pore wetting issues with increasing DS concentration and the problem of fouling occurs for the FO membrane which inhibits process performance. There is also the inherent limitation of lack of efficient draw solution in the market.

In a different study, Jebur et al. [130] combined EC and microfiltration to MD operated in batch mode for treatment of hydraulic fracking PW. The EC component served to reduce TOC from 120 ppm to 64 ppm in the feed while MF (using polyethersulfone membrane) was used to separate particulate matter further driving TOC down to 44 ppm. Final treatment in MD achieved a TOC of 1 ppm and TDS of 56 ppm from an initial 245,300 ppm. An interesting fouling and scaling study was presented using three different membranes (commercial PVDF, electrospun PVDF-HFP and MWCNTs coated PTFE) to evaluate the EC-MF-MD system. They showed that although EC was able to reduce solids and organic species that could clog the pores and foul the surface of membrane, some floc still attached to the surface during the MF stage. However, recirculation with DI water for 1 h effectively regenerated the membranes. The MWCNTs-coated PTFE exhibited highest constant flux and the best performance during the MD stage although membranes were quickly fouled and irreversibly so due to hydrophobic-hydrophobic attraction between CNTs and organic species in PW feed. The flux for the electrospun PVDF-HP dropped rapidly although the membranes were more easily regenerated while flux dropped to zero after the first run for the commercial PVDF after recovering 131 ml of the desired 250 ml. The membranes were regenerated and the total permeate volume of 250 ml was recovered in the 2nd run although some absorbed species could not be removed by simple flushing. Scaling was generally controlled by favouring precipitation in the feed tank and not on the membrane surface via lowering the feed tank temperature to 36 °C and increasing the temperature in the MD module to 60 °C, thus supressing precipitation. However, there is the adverse effect of increased energy consumption.

Nawaz et al. [196] recently reported an FO-MD hybrid for treatment of different PW streams. By choosing the PW stream from water-oil separator outlet to be the draw solution for FO, they highlighted a possible reduction in operational cost by boycotting the otherwise typical synthetic DS. The FO stage achieved flux ranging between 8.3 LMH to 26.78 LMH with different streams while an average flux of 14.41 LMH with salt rejection above 99% was achieved for the MD phase. Fouling was observed in FO membranes in the form of colloidal calcite scaling while pore blocking occasion by $CaSO_4$ was the main reason for flux decline in MD. The $CaSO_4$ scaling was found to be resistant to simple flushing by DI water. However, a complete flux recovery was possible in standalone MD using ethylenediamine tetraacetic acid (EDTA) to eclipse the effect of divalent scale-causing calcium ions as well as the dissociation of humic acid, thus reducing their negative effect on flux.

The integration of MD with crystallization (MDC) units in order to achieve ZLD has also been reported [14,147]. While the set-up has shown potential for increased total recovery as well as prolonged operation with controlled salt scaling, MD itself is energy insufficient and additional crystallization unit also means more energy consumption. Other hybrids of various configuration have been extensively researched and published in several review articles [8,87,155,194]. These have opened myriads of insights to curbing one form of shortcoming or the other for the various integrating parts for application in various fields, it is however pertinent to always consider the cost implication vis-a-viz desired output as well sustainability with regards to the environmental footprint and even long-term operation of the individual technologies.

5. Competitiveness of MD in handling PW brine

The portrayal of MD as an alternative desalination technology has seen tremendous recognition over the years mainly because of its excellent salt rejection performance as well as tolerance for hypersaline feeds with TDS up to 350,000 ppm [36]. A review of the state of desalination and brine production across the world by Jones et al. [134] revealed RO as the predominant technology accounting for about 84% of total operational plants (as depicted in Fig. 10a) while multi-effect distillation (MED) and multi-stage flash distillation (MSF) are the popular thermal technologies accounting for 18% and 7%. Nanofiltration (NF), electrodialysis (ED) and EDI total about 5% with small volumes of desalinated water. Interestingly, a subsequent review of current membrane technologies for PW desalination by Ahmad et al. [7] put MD in light of major membrane-based technologies such as RO and forward osmosis (FO) with a rising interest particularly for MD, as depicted in Fig. 10b. The potentials of MD for desalination of PW as depicted by increasing interest amongst researchers is represented by about 39.22% of publications in the last two decades, followed by FO and then RO with 35.29% and 25.49% respectively. Recent advances have seen the incorporation of various surface properties unto MD membranes for desired end use including anti-fouling and anti-scaling modifications, as previously discussed. Despite these advances however, MD is still largely in the lab stage with a few pilot tests. There is therefore need to further look at recent efforts to make MD competitive with respect to other popular PW desalination technologies like ED, MED, MSF as well use of evaporation ponds, taking into consideration the cost of desalination, brine handling and disposal as well as long-term stability.

5.1. Long-term performance

The stability of MD membranes over long term performance is critical in understanding membrane durability over said period by studying possible changes that occur at the membrane interface during the operation [180]. Bench-scale studies typically have operation in the range of minutes to hours while pilot testing record longer periods of time. However, many researchers have reported operations from just about 48 h (2 days) to 300 h as long term [139,184,250,261,275], giving the usual nature of reporting much less operating time for MD. However, a more ideal test to depict long term may be represented by duration upwards of a few days or even weeks as reported by other researchers ([143,152;180;291] .

McGaughey et al. [180] studied the long-term performance of commercial PTFE flat-sheet membranes in a 100-days DCMD operation treating a feed with moderate to high salinities in a bid to clearly identify their effect on the feed and permeate side of the membranes. Results showed that there was significant alteration to surface morphology as well as deterioration in hydrophobicity for the period of investigation. Specifically, foulants were found to have accumulated. This was attributed a possible entry into a partially opened auxiliary feed tank, trace quantity of organic species in DI used to make the feed solution as well as the combined effect of elevated temperature and hydrophobicity which aggravated attraction of organic matter to the membrane. Flux

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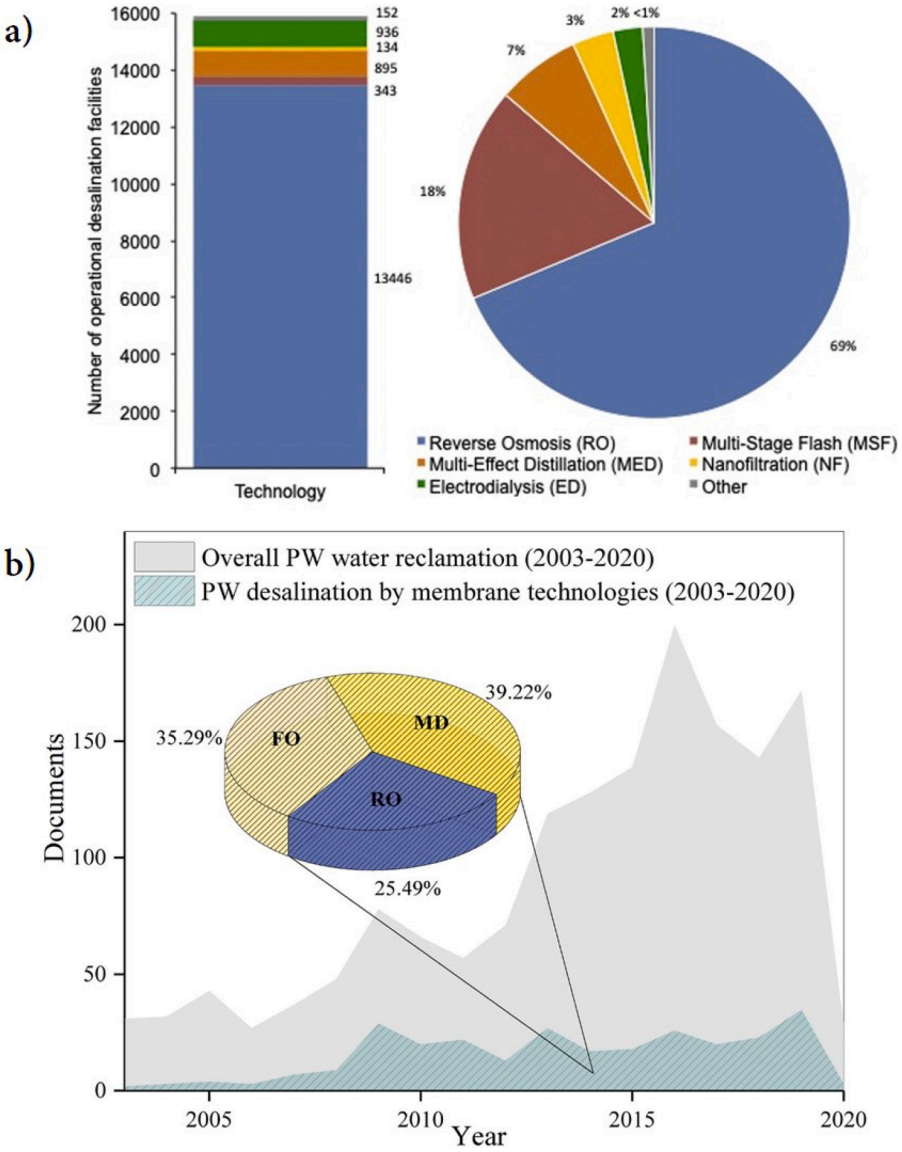

Fig. 10. Shows a) number of desalination plants for popular technologies across the globe b) publication trend portraying rise of interest in PW reclaimation and particularly for MD with respect to FO and RO [7,134] .

declined by 16% as a result and membrane hydrophobicity also gradually reduced due to fibril widening on the distillate side, although salt rejection remained above 99.98% all through. Khayet et al. [143] modified the surface of polyetherimide (PEI) hollow fiber membranes to offer robustness via single casting step by surface segregation for long-term MD operation over a period of 54 days. The addition of FPA in the polymer blend significantly increased the hydrophobicity and in turn the MD performance of the membranes, recording salt rejection above 99.99% throughout. Permeate flux was also stable throughout the experiment, attributed to high liquid entry pressure value, which in turn kept the pore dry through the MD operation. Addition of FPA was also noted to have significantly improved the mechanical strength of the membranes. Gustafson et al. [101] also examined the morphological changes and creep recovery behavior of expanded-PTFE using three 30-day MD experiments with different feed temperatures and salinities in the absence of foulants to better understand the changes occurring at the feed-side and their effect on performance. Increase in temperature at the feed side from 45 to 65 with salinity of 5 g/L resulted in a much-pronounced deviation of microstructures from the virgin membrane with fibrils widening and flattened nodes, indicating the important role played by temperature in the microstructural changes at the membrane surface. Increase in salinity to 200 g/L with feed temperature of 65 however did not contribute any significant microstructural changes on the surface of the membrane. Membrane thickness and porosity were observed to have reduced on exposure of e-PTFE to long-term MD operating conditions, which ultimately reduced water flux – suggesting that membrane morphological changes alone can also negatively affect performance. While this study is a pointer to the fact that hypersaline feed like de-oiled PW may not have much effect on membrane in long term operation, there is little reportage in the literature to that effect.

A 7-day long MD of PW was conducted by Chew et al. [46] using a modified PVDF membrane with Janus properties offering remarkable anti-fouling and anti-wetting against organic species in surfactant-stabilized PW emulsion. The hydrophilic hydration layer prevented the attachment of oil to the membrane surface which offered steady flux and remarkable salt rejection. Another long-term study of MD performance for treatment of PW with TDS up to 123 g/L was carried out by Thakur et al. [259]. Using PTFE flat-sheet module in a SGMD 125 h operation, they showed the performance of the membranes in the presence of shale oil PW containing high concentrations of organic and

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inorganic species, with salt rejection remaining above 99.9%. Modelling study was used to show that a well-optimized process could mitigate the strontium scaling observed on membrane surface. Gryta [94] also conducted a 1500 h long MD of oily saline wastewaters using polypropylene membranes. They associated the major reason for pore wetting to scaling by inorganic species in addition to oil adsorption on membranes which resulted in rapid increase in distillate conductivity and flux decline. Periodic rinsing with 5 % HCl solution was sufficient to remove scaling. Suga et al [250] studied the characteristic properties of membrane in an attempt to decrease water production cost in a VMD long term operation experiment using PVDF hollow fiber membranes. They discovered LEP to be the most important factor for stable long-term performance of the system. Contrary to theoretical possibility of operating MD without pore-wetting if the membrane LEP is just higher than the vapor pressure across the feed and permeate sides, a much higher LEP was required practically to achieve stability. Although the study was using seawater with 3.5 wt% NaCl aqueous solution, it can be expected to be applicable for hypersaline feeds like PW since MD is tolerable to increased salinity.

Generally, surface modification of membranes to offer anti-fouling/anti-wetting properties such as omniphobic/superamphiphobic designing have proved useful for treating PW in MD as previously discussed, although more studies are required to isolate the individual effect of feed temperature, hyper salinity and surface morphological and microstructural changes on the overall performance of MD during long-term treatment of PW.

5.2. Economic assessment of PW handling options

Despite the potentials for prolonged operation with little effect on process conditions as demonstrated by researchers, it is important to consider energy consumption over time and hence the economic viability of MD projects. A number of studies have reflected the cost of MD particularly for seawater and brackish water desalination in comparison to major desal tech like RO [16,71;186;238;269]. Similarly, a number of studies have highlighted and compared RO to other desal techs for PW handling, being the most popular desalination tech in the industry [137,195;205;212]. However, limited work can be found in the literature featuring holistic economic analysis of MD in relation to major PW desal technologies. Generally, because of difficulty to obtain data from plants, many assumptions are made regarding location and cost variables as well as critical datasets including critical factors affecting MD systems, leading to discrepancies in reportage and hence rendering most economic analysis unreliable [268].

Generally, an initial treatment to remove oil and suspended solids is done before subsequent management option, the cost of which is estimated at 1$/bbl [181]. One of the commonest PW brine handling practice in the USA is underground disposal by injection into wells . The costs associated with deep-well injection is estimated between 1.57 and 15.72 $/m^3 [62]. In a situation where disposal site is far from the field and no pipelines laid, the cost of trucking needs is put at 0.20$/m^3/mile [51]. While disposal in these well is one of the cost-effective options of PW management, it has been associated with inducing seismicity in the ground [236], specifically linked to injection volumes and rates in the case of Oklahoma [223]. Regardless of injection pressure, induced seismicity have been reported to influence earthquake occurrence in regions in close proximity to disposal wells [226]. Handling of TDS in PW with traditional thermal technologies such as multi-effect distillation and brine concentrators requirement extensive pre-treatment of feed and have high energy requirement making the processes costly [183,193].

Treatment of PW is considered more cost-effective than disposal in wells [257]. Recently, there is more attention and revisiting of desalination technologies that are capable of concentrating brines close to saturation conditions, termed zero-liquid discharge (ZLD) systems [202]. Single and multi-effect evaporation (SEE/MEE) study showcasing the desalination performance for shale gas flowback water with and without multi-stage compression was carried out by Onishi et al. [203]. They showed that the multi-effect evaporation with multi-stage compression (MEE-MVR) is the most cost-effective process for ZLD with an estimated cost ranging from 6.7 to 10.9$/m^3. However, the cost of desal is still significantly high and the environmental implication is such that significant amount of CO_2 is yielded and hence more sustainable approach is necessary. In a new multi-objective modelling study, Onishi et al. [204] considered minimizing thermo-economic and environmental impact of the ZLD system by integration of solar energy to power a MEE-MVR plant for treating hypersaline shale gas wastewater. The study revealed that environmental impact can be considerably reduced by enlarging the area of solar parabolic trough collectors thereby reducing utilization of natural gas which also means savings in cost of operation.

Other studies featuring ZLD have considered integrating MED with evaporative crystallization as studied by Chen et al. [41]. The cost of brine treatment was pegged at 4.17 $/m^3 attributed mainly to thermal energy cost. They demonstrated that by selling freshwater and salt, the system can be more competitive in comparison to other systems in addition to consideration of alternative sustainable powering source. Under the scenario of waste heat utilization to power MED-TVC system for ZLD, Panagopulos [207] showed that cost can be reduced to 1.69 $/m^3 for brine with TDS up to 70,000 mg/L. Increasing the number of effects from also contributed the reduction in freshwater cost with a 4-effect system having the shortest payback period of 3.49 years, for a scenario where external heat is supplied at a fixed cost. For a scenario where waste heat is available on-site in the plant at zero cost, freshwater cost increases with number of effect and as such a 2-effect system which also represented an operating cost reduction by 80%. As opposed to disposal, freshwater produced from brine treatment in MED-TVC following scenario 2 can be profitably sold at1.51 $/m^3. A general comparison is given in Table 5 between MD, ED, RO, MED and MSF for PW desalination. Advanced membrane desalination processes such as RO even though relatively cheaper have limitations in terms of TDS levels as osmotic pressure builds up in the system, with an estimated treatment cost of $0.80/m^3 while the emergence of MD as an alternative desalination technology for PW further portends a reduction of cost to $0.30/m^3 with an alternative heat source. While most of the techniques generally have limit for TDS handling, MD can handle TDS up to 350,000 mg/L [36]. Increasing salinity for RO and ED also affects the cost of desal as it results in reduced recoveries . Despite promising performance in desalination, high energy consumption for MD makes it less attractive than RO [218], although recent advances in using alternative cheap energy source to MD may mean cost-competitiveness.

5.3. Alternative heat source for MD

The quest for cheaper alternative to powering MD systems has led researchers to explore various options with a view to further drive down the cost of desalination using the technology with a potentials for reduction in costs to as low as 0.74 $/m^3 by integrating with waste heat sources [257]. Prime amongst the alternatives include the deployment of solar energy-driven process [118,302], utilization of waste heat [22,64;167;224] and recently photothermal membrane distillation (PMD) [34,89;293].

A recent optimization-based model by Tavakkoli et al. [258] featuring a techno-economic assessment of MD for shale gas PW treatment for the Southwest and Northeast Pennsylvania Shale play revealed that an on-site treatment option at natural gas compressor stations where readily available waste heat can be used presents more economic advantage translating to savings of over $16 million per year as against use of disposal wells. This is important and timely as costs saving with regards to management of PW is a win for producers and good news for its potential market; farmers for example, as PW has been recently confirmed to not threaten crop safety. Said et al. [231] explored photo-

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Table 5
Comparison between popular PW desalination technologies.

Technology	Typical plant capacity (m^3/day)	Energy Consumption (kWh/m^3)	Cost of Desalination ($/$m^3$)	TDS handling (mg/L)	Ref.
RO	Up to 395,000 (commercial app)	3 – 4 including water intake, pre-treatment and post-T. ~2 for the RO stage	0.8	80,000 150,000 (for high pressure RO)	[84,137;208]
ED	2 – 145,000	2.64 – 5.5	0.6 – 1.05	>70,000	[11,244;274]
MD	1600	49 to 350	0.3 – 5.7	350,000	[17,91;169]
VC (thermal)	10 to 30,000	23–42	0.8 – 0.95	-	[11,161]
MEE-MVR	900	-	6.55	70,000	
MED	600 to 30,000	14.45 – 21.35	0.9 – 1.5	-	[11]
MSF	10,000 to 35,000	19.58 – 27.25	0.56 – 1.75	50,000	[11,244]
Disposal (deep well)	Up to 375,000 (4 wells)	–	0.06 –6	-	[35,62]

thermally active carbon-black NPs coated on a polypropylene membrane and used for solar energy-driven MD for treatment of raw PW. Activated carbon cartridge filtration was deployed as pre-treatment targeting suspended solids and dispersed oil in raw PW with potential of causing irreversible membrane fouling prior to the nanophotonic-enabled solar MD (NESMD). TDS and DOC reduced from 132,000 ppm and 113 ppm to 83,000 ppm and 90 ppm, respectively, in one of the samples. Further treatment in NESMD reduced TDS to < 50 ppm (>99% rejection) and TOC to < 3.7 ppm. However, SEM and EDX analysis of membrane surface suggested the occurrence of fouling by organic matter and inorganic salts.

Following this feat, a pioneer study featuring a photothermal at the same time omniphobic MD membrane was reported by Chen et al. [43]. The direct solar membrane distillation system (DSMD) utilized carbon black (CB) nanoparticles-modified PVDF membranes for providing localized heating by absorbing sunlight and subsequent photothermal conversion for vaporization. The omniphobic modification on the other hand ensured robust anti-wetting properties in the presence of surfactant (0.4 mM SDS). This novel dual functional MD set-up recorded salt rejection up to 99.99% with an increased energy efficiency of 55.6% in comparison to DCMD. An increase in CB-NPs loading up to 0.2 wt% realized a corresponding increase in permeate flux by about 25% beyond which flux declined, attributed to reduction in pores. Bamasag et al. [24] also reported a solar powered submerged VMD placed in an evacuated-tube collector (ETC) coupled with either aeration or internal circulation for feed agitation in an effort to overcome temperature and concentration polarization, in addition to slowing the onset of fouling. Contrary to previous studies for submerged MD mimicking design of conventional cross-flow shell and tube modules, they proposed a closed system where the feed stays in the module without the need for circulation pumps, thus further reducing energy consumption. while aeration improved average permeate flux by 10%, internal circulation achieved 22% improvement. They reported thermal efficiency of up to 51%, highlighting the drawback of substantial solar radiation channelled to heating the feed before the onset of distillation. Despite being attractive as sustainable energy sources, temporal fluctuations of solar energy make them unattractive for commercial implementation [251].

A recent study by Robbins et al. [224] investigating the viability of waste heat from electrical loads and natural gas to operate an on-site MD facility for desalination of waste stream from Denver-Julesburg Basin in Colorado concluded that waste heat alone is not sufficient to power MD for long-term PW treatment. Similarly, waste heat from hydraulic fracking is only viable for the short-term while natural gas which is co-produced on-site can serve as alternate or supplementary energy source for long term MD of PW. However, further investigation is necessary to ascertain if the implied cost saving compensates for costs associated with heat storage and recovery systems.

Microwave irradiation as a source of heat for MD membranes has also been researched. Earlier studies found that irradiation had no effect on the mechanical properties and hydrophobicity of membranes, but induced the deposition of carbonate salts for feed solutions containing Ca^{2+} [131,132]. Microwave heat source is reported to consume at least 20% less energy than conventional heating and realizes improved vapor flux as high as 52% attributed to localized superheating as well as dissociation of salt-water clusters [225]. Using CNT-immobilised membranes, Humoud et al. [123] sought to curtail the problem of scale formation for feed solutions containing high concentration of aqueous calcium carbonate, calcium sulfate and barium sulfate by microwave induced MD. In comparison to conventional heating, they showed that reduction in scale formation was between 50 and 79% when using microwave heating, owing to smaller particles formation as confirmed by SEM and dynamic light scattering studies. Nonetheless, scaling still affected MD process [132]. Much reduced scaling $CaSO_4$ crystals as a result of high frequency collisions of ions in the feed was reported by Anvari et al. [18] in a radio frequency-induced heating MD study. They used thermally conducting membranes modified by CNTs functionalized with iron nanoparticles resulting in enhanced flux and reduced scaling as evident with detection of small crystals in the concentrate stream. Despite promising results from using microwave as heat source more MD, more detailed studies covering cost-competitiveness in comparison to other sustainable heat sources are needed, in addition to understanding the process performance when desalting complex feed such as raw PW in microwave-induced MD.

6. MD for beneficial re-use of PW

The growing potential for beneficial re-use of PW particularly for agriculture must be seen to make economic sense to both farmers and producers as the costs involved in treating and transporting PW either by pipeline network or trucking may not be encouraging for either the farmers or the producers . In addition to health hazards, water demand and environmental concerns, the risks associated with increased seismicity further motivates treatment and re-use of PW [312]. The reuse option for produced water may include irrigation, livestock watering groundwater recharge or even in industries, depending on the site-specific scenarios that will allow comprehensive decision framework [102]. However, the most obvious sector for reuse of PW outside the oil and gas industry is irrigation [237].

The use of PW for irrigation purposes is an interesting substitute for the risks and environmental concerns associated with deep-well disposal, particularly for arid region experiencing water scarcity [75]. In southeast Arabian Peninsula for instance, PW for irrigation accounts

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for 31% of the water volume consumed by the agricultural sector, annually. Echchelh et al. [73] studied the potentially sustainable irrigation schemes in arid Qatar with PW from a gas field. Using a soil–-water model for sugar beet irrigation, they tested an array of schemes including over-irrigation, blending PW with treated sewage effluent as well as desalination of PW in a bid to curb the adverse effect of salinisation and sodification on soil and aquifers. The simulation study revealed that an irrigation volume up to 300% of the crop water needs blending 2/3 of PW and 1/3 of treated sewage effluent or desalinated PW was ideal for preserving the soil in addition to crop yield and groundwater quality. While diluted PW have been used in irrigating crops in some states in the U.S, the effect on soil health and has not been studied. Miller et al. [188] investigated the efficacy of reusing minimally treated PW for irrigating wheat with a focus on studying the corresponding effect on eastern Colorado soil health and microbiome and discovered that inadequately treated PW is detrimental to soil health and led to decreased yields. An earlier study by same researchers also indicated that increased concentration of organics in water used for irrigation led to deterioration in crop immune response to pathogens [189]. Following a previous study where they found that PW salinity and sodicity can be harmful to soil structural stability as well as crop yield [73], Echchelh et al. [75] further considered the operational cost of utilizing PW for irrigation in comparison to erstwhile PW disposal taking into account environmental impacts. Although cost of handling PW for irrigation is up to 2.5 times higher than disposal, they showed through simulation that a cost-competitive option was to mix raw and desalinated PW in a 1:4 ratio and reduce irrigation amounts to meet only crop needs.

The cost of treatment is a determinant factor for the volumes of PW that will be available for re-use. The cost of treating PW for agricultural use is reported to range up to $3/bbl which is dependent on plant capacity, quality of feed as well as price of electricity [51,62,297]. While traditional disposal practice of deep well injection cost between 0.31 and 16.67 $/m^3 [80], an even more cost-competitive option involving reduction of irrigation amount just a little above crop water needs while blending PW with four equivalent volumes of desalinated PW costing around 0.46 $/m^3 was proposed by Echchelh et al. [73]. In Kern county – California, treated PW from certain fields with low salinity (TDS ~ 750 mg/L) is blended with other fresh water sources and used for irrigation purposes [93]. A recent doubling of capacity for handling PW in a membrane-based technology by Wasco plant in Bakersfield – California for reclamation and reuse, from 210,000 gallons to 420,000 gallons per day reporting a potential for lowering energy costs by 40% as well as reduction in the need for consumables such as chemicals by an estimated 19% opens a myriad of economic benefits to producers, farmers and most importantly help reduces pressure on municipal water resources as commercial users can utilize the reclaimed water [19].

Similar feats are all the more feasible with MD by capitalizing on its unique advantage of having tolerance for hypersaline feed and achieving near-absolute salt removal and high purity distillate, placing it in the limelight for PW treatment. In considering MD of PW for agricultural re-use, a dual functional membrane as reported by Chen et al. [43] would be ideal. The omniphobic MD membrane would protect crop and soil from salt and organics while the utilization of the solar power to drive the process would make it competitive, cost wise. Although there is general concern from public over potable re-use of PW particularly when it concerns recharging groundwater aquifers [102] , many researches on pre-treated PW desalination using MD has shown capacity of complete salt removal as well as residual organic pollutants as previously discussed. As shown in Table 6, [312] presented a list of PW constituents and their allowable range for re-use in irrigation, livestock watering and drinking. For agro-environmentally sustainable irrigation, the sodium adsorption ration (SAR) or sodicity and salinity of the soil have to be controlled. Most of the constituents have been demonstrated to be removal in other MD studies below the limits/standard for the re-use applications [69,70,112,125,277,310,314] . This makes MD a possible source of potable water, perhaps for use on offshore platforms where it's a valuable resource. It is however important to consider the scale of demand viz-a-vis the ability of MD to meet them in a long-term and continuous basis.

Table 6
PW constituents and their allowable range for drinking, livestock watering and irrigation.

Component	Drinking (g/m^3)	Livestock (g/m^3)	Irrigation (g/m^3)
Li^+	–	–	2500
Na^+	200	2000	Based on SAR
NH_3	1.5	–	–
Ca^{2+}	–	–	Based on SAR
Mg^{2+}	–	2000	Based on SAR
Cl^-	250	1500	–
SO_4^{2-}	500	1500	–
TDS	500	5000	2000
SAR	–	–	0.6

7. Future direction

The potentials for significant improvement of synthesis methods and modification techniques as well as MD process still allow for continuous optimization of the techniques involved for the treatment of oilfield PW. A nanoparticle-free approach of creating omniphobic surface on electrospun PVDF membrane using a solvent-thermal induced roughening to impart hierarchical nanofin structures and fluorination achieved super repellence (>150°) towards low surface tension liquids was recently carried out by Qing et al. [215]. The membranes maintained stable water flux and salt rejection in the presence of 0.4 mM SDS and 480 ppm mineral oil. Similarly, in-situ growth of nanoparticles on membrane surface resulting in robust multi-scale re-entrant structures on the membrane surface has recently garnered attention resulting in membranes with excellent wetting and scaling resistance that are not affected by intense process conditions [298]. Nucleation sites are created on the surface of the membranes for subsequent growth of nanoparticles. This has opened a new path towards simple

More comprehensive studies of fouling mechanism in MD for PW is necessary to propose better membrane materials that pose high hydraulic performance while showing resistance to fouling. A combination of two or more improved techniques that enable 3D monitoring of fouling in real-time with robust dynamic simulation to better capture all the variables in play during the MD process of PW is necessary. In a pioneer study of fouling behaviour of oil emulsion using OCT in a dead-end filtration set-up, Trinh et al. [264] reported the internal and external fouling of membranes by oil in contrast to erstwhile studies focusing on biofilms and solid particles. This approach can further be improved and adapted for MD of PW. The effect of high temperature at the feed side which is typical of MD needs to be studied particularly for PW treatment.

Pioneer work highlighting a novel technique and tool for quantifying wetting and subsequent salt transport using detection of dissolved tracer intrusion (DDTI) by Jacob et al. [127,126] , visualizes wetting at the pore scale with the aid of spectroscopic and microscopic approaches paves way for elaborate understanding of the wetting phenomena in MD of complex feeds like PW. Ion chromatography has also been reported to measure ion concentration at the permeate side in the work by Zou et al. [313]. This allows for the study of rejection efficiencies of individual anions and cations present in the feed water as against the traditional measurement of conductivity at the permeate side. Studies elucidating membrane scaling have also utilized various online and in-situ methods with remarkable insights to scaling as it occurs. However, more studies using other unique methods like EIS particularly for understanding scaling by PW constituents on MD membranes is necessary since EIS allows distinguishing electrochemical properties of membrane bulk and boundary layers [294]. This will help in further understanding the controversy around the predominant constituent of salt scaling for MD

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in PW desalination.

While the use of alternative heat source for powering MD opens a myriad of interest to further realize a more energy efficient process, more studies are necessary to overcome the challenges of temporal fluctuation for solar and waste heat sources. Microwave and radio frequency heating as a source for stable and more energy-efficient source of powering MD require further studies to overcome induced scaling problem due to direct heating of the membrane surface. In the same vein, cost-benefit and perhaps techno-environmental assessment of systems integrated with MD is necessary to justify their integration, despite promising results.

8. Conclusion

MD is a promising thermal desalination process that offers great potential for the recovery of high purity water from hyper saline wastewater sources like oilfield PW, hypersaline groundwater and even concentrated RO rejects using hydrophobic membranes, and recently superamphiphobic/omniphobic membranes. It capitalizes on the drawbacks of RO where osmotic pressure exceeds the operating pressure of the process. In MD however, the process is not affected by the build-up of osmotic pressure since it is thermally driven. It is further blessed with the potential of utilizing low grade heat from waste streams, solar energy and even geothermal hotspots as potential sources of vapor drivers. The various modification methods discussed have shown great potentials for game-changing breakthrough in oily wastewater treatment and management in MD for beneficial re-use. However, MD itself is still mostly at the laboratory scale owing to intrinsic setbacks that hinders its pilot testing amongst which are its energy demand being a thermally driven process. Despite promising results from recent advances in using alternate heat sources for powering MD systems for treating hypersaline oily wastewater, it is important to improve membrane synthesis process and materials such that highly porous and permeable membranes with higher flux are realized, without necessarily trading the cost advantages of photothermal MD.

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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