

Development of high performance pervaporation desalination membranes: A brief review



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ARTICLE INFO

Article history:

Received 11 October 2021

Received in revised form 14 January 2022

Accepted 27 January 2022

Available online 29 January 2022

Keywords:

Pervaporation

Desalination

Composite membrane

Ceramic

Crosslink

ABSTRACT

Water scarcity rises as the level of water pollution continues to increase with the progress of urbanization, industrialization and exponential growth of population. Therefore, saline water of the sea should also be made suitable rather than river water to meet the huge global demand of clean and safe drinking water. Pervaporation (PV) desalination, among many purification and separation processes, is a promising technology to reduce the crisis of global drinking water supply. From this perspective, the key success of PV desalination relies on its remarkable salt rejection from highly saline water with appropriate flux to obtain fresh water by using a suitable membrane. In this review we aim to provide a comprehensive assessment of PV desalination membrane materials, transport phenomena, the advantages of the process over comparable technologies (e.g., fractional distillation, membrane distillation, reverse osmosis) and the advantages of crosslinking during the preparation of composite membranes. This review further highlights the advantages of inorganic ceramic substrates as a support of composite membranes and the use of hydrophilic polymers as active layer for preparing stable and robust crosslinked PV desalination membranes.

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1. Introduction

The world is facing a crisis of drinking water due to the extensive population explosion in the past few decades, resulting in a critical situation that calls for effective water treatment policies and efficient separation technologies. Only 2.5% of the water on earth exists as freshwater and remaining is in saline form (Oki and Kanae, 2020). Thus, water treatment technologies have become essential to meet the demand. Saline water is largely available all over the world and improved desalting technology may solve some of the issues related to lack of fresh water (Eljaddi et al., 2021). Membrane technology is considered as the most common and efficient technology for desalination and thus has been integrated with other intermediate processes in several water treatment plants worldwide (Lively, 2016; Jones et al., 2019; Cai et al., 2020). It is highly efficient with respect to other conventional technologies due to several advantages (Mondal et al., 2013). Various membrane technologies, such as reverse osmosis (RO) (Tyler et al., 2021), electrodialysis (ED) (Tran et al., (2014), pervaporation (PV) (Prihatiningtyas et al., 2019; 2020 a,b) and membrane distillation (Alobaidani et al., 2008) have been used for water desalination. Technological differences, operational methods and limitations of these desalination processes are discussed in details in Section 3. RO is the most common technology that is currently applied in large-scale plants for desalting water to produce fresh water. In spite of its multiple advantages, RO faces some real challenges such as i) high energy requirement due to the very high operating pressure for seawater desalination (40–82 bar) (Tyler et al., 2021; Zhang et al., 2020) ii) lack of ability to handle brine that contains more than 4% NaCl (Zhang et al., 2020; Tyler et al., 2021). In this context, pervaporation (PV) is the latest promising technology for desalination (Li, 2016; Manshad et al., 2016; Wang et al., 2016a).

Theoretically, PV can completely separate salts from sea water and this suggests its potential as an emerging technology for desalination. This process is also widely applied to separate azeotropic liquid mixtures like alcohols and water (Jyothi et al., 2019), polar aprotic solvents and water (Vane, 2019). PV is very useful for bioethanol dehydration (Zheng et al., 2019), non-alcoholic wine and beer production (Castro-Muñoz, 2019). However, practical application of PV has been reported to desalinate sea water (Korngold and Korin, 1993; Xie et al., 2018). Development and use of suitable membrane materials remain a challenge for PV for specific applications. Furthermore, the techno-economic maturity of separation and purification of high salinity sea water to obtain pure drinking water using membranes may have some remaining issues like poor mechanical strength, long-term stability, efficiency, cost, industrial application, etc. To alleviate these issues, the key success of PV desalination process mainly depends on the membrane stability and flux enhancement from the perspective of dense composite membrane fabrication by using a suitable hydrophilic polymer (Cho et al., 2011; Li, 2016; Manshad et al., 2016; Yoshida and Cohen, 2003; Huth et al., 2014). Such dense hydrophilic membranes can be effectively used in PV technology. The dense and hydrophilic nature of the membrane is required to suppress fouling of the membrane by preventing salt deposition on membrane surface (Wu et al., 2018). Membranes can be classified mainly into two broad categories – dense membranes and porous membranes. For PV desalination, hydrophilic dense membranes can be effectively used where water vapor is permeated through these membranes from a mixture of

water and salt(s) under a partial pressure difference at elevated temperatures.

This review article aims to provide a compelling literature survey on reported PV desalination membranes, especially focusing on ceramic composites, which is discussed in detail. Comparisons are made between PV and other competing desalination technologies, and the advantages of PV are underlined. The review further highlights the merits of inorganic ceramic substrates as support of composite membrane and the use of selective polymers for PV desalination membrane preparation. Furthermore, the use of appropriate functional layer polymers and crosslinking (thermal and chemical) of these polymers with support material as key factors for fabrication of stable PV desalination membranes with enhanced permeability is also discussed to give a perspective for improved membrane preparation. Future directions on robust ceramic-polymer composite PV membrane preparation for desalination are also addressed.

2. Principle and historical background of pervaporation

Desalination has contributed to the development of human civilization since about thousand years ago. Seawater was thought to give hope to acquire drinking water via boiling of saline water based on evaporation technique centuries ago (Birkett, 2010). Kober, in the year 1910, had noticed that water permeation takes place in a selective manner through a membrane made of collodion and parchment, this was described by the term 'pervaporation' as the combination of two words 'permeation' and 'evaporation' (Kober, 1917). The early experiments on PV, which were performed in the years 1935–1983, were mainly on laboratory scale (Farber, 1935; Heisler et al., 1956; Binning et al., 1961; Aptel et al., 1976; Neel et al., 1983). In this process, the permeating component (liquid) is converted into a vapour phase using the enthalpy of vaporization due to the low partial vapour pressure on the permeate side. This low vapour pressure is generated during operation by placing a vacuum pump on the permeate-side of the membrane. It is a relatively simple process and parameters for process optimization are mainly temperature and vacuum pressure.

In case of PV desalination, non-volatile salt is separated from water using a hydrophilic membrane, in the form of vapour at a downstream side that is connected with liquid N₂ cold trap or a chiller (Fig. 1). The membrane may be non-porous or very finely-porous, polymeric and/or inorganic in nature that displays an affinity towards the component that needs to be removed. Additionally, PV desalination involves a phase change and does not require to overcome the osmotic pressure of saline water. Thus, it offers potential efficiency in terms of energy because of the low pressure requirements (Volkov, 1994; Feng and Huang, 1997; Shao and Huang, 2007).

The separation mechanism of PV is solution-diffusion and the separation via diffusion can be controlled by size exclusion (Wang et al., 2016a; Wu et al., 2018). The transport system through membrane in PV involves an interaction between feed, permeating substances (penetrants) and membrane (Feng and Huang, 1997; Shao and Huang, 2007). Mass transfer through PV membranes can be described by the Thomas Graham's solution-diffusion model (Volkov, 1994; Wijmans and Baker, 1995). This sorption-diffusion model is based on three important steps regarding permeation of feed components through dense membrane (Fig. 2).

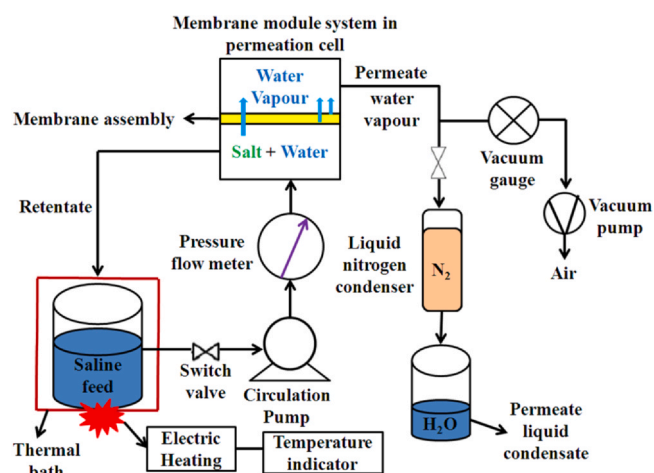


Fig. 1. Schematic of pervaporation desalination system.

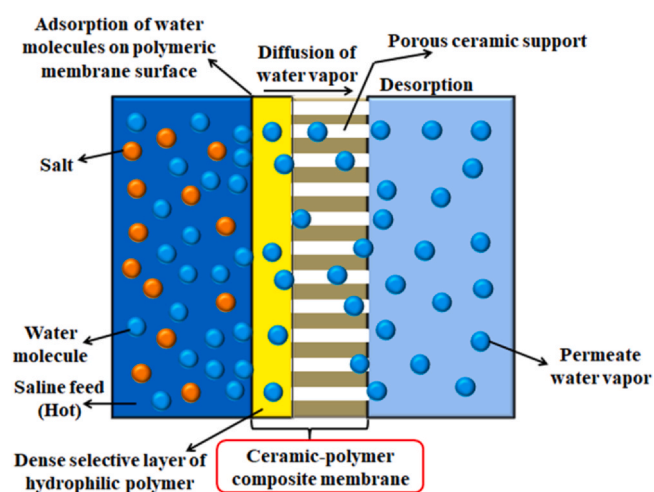


Fig. 2. Mass transport through hydrophilic dense pervaporation desalination membrane.

- (i) Selective adsorption of molecules of the components from the liquid feed solution onto the membrane surface layer.
- (ii) Diffusion of the components through the membrane matrix.
- (iii) Desorption via evaporation of the components into the vapour phase at the downstream permeate side of the membrane.

Among the mentioned three steps, the first and last step can attain equilibrium state at a faster rate but the second step where diffusion takes place is marked as the “rate-controlling step” according to Fick’s first law. The chemical potential difference (partial vapour pressure difference, concentration gradient) is thought to be the overall driving force in the PV system according to this solution-diffusion model. This model is generally used to establish the equation of mass transport for membrane processes. The solution-diffusion model can be applied successfully to all aspects of PV (Feng and Huang, 1997; Wijmans and Baker, 1995; Ong et al., 2016).

The first PV membrane was brought into the field of commercial application by Gesellschaft für Trenntechnik (GFT) in 1982. It had a composite structure with cross-linking network of polyvinyl alcohol (PVA) thin film coating supported by porous poly(acrylonitrile) (PAN) (Tusel and Bruschke, 1985). Industrialization of the first PV system took place in Brazil in 1983 for ethanol dehydration (Nunes and Peinemann, 2006). More than 100 PV plants have been successfully completed since 1982, of which the largest plant that has a

capacity to produce 5000 kg h⁻¹ of ethanol, was located in Bethenville, France (Shi and Chung, 2013). The history of PV and desalination (before World War-II – 2006) is summarized in Fig. 3 and the new trend of research has been started from 2007 onward.

3. Comparison between pervaporation desalination and comparable desalination processes

Thermally driven distillation technologies or purification using mechanical and/or osmotic strength (i.e., using a membrane) are mainly two types of desalination processes used in industrial scale. Among the membrane-based technologies RO (Li and Wang, 2010), PV (Wu et al., 2018) and MD (El-Bourawi et al., 2006; Camacho et al., 2013) are very well known for desalination. However, MD and PV are less mature compared to RO. In this section the advantages and disadvantages of PV compared to comparable desalination processes are being discussed.

3.1. Pervaporation vs. fractional distillation

PV is an alternative technique for the separation of water from salt mixtures present in sea water. It is a simpler method than other processes. The separation mechanism of PV is independent of the relative volatility of the components (Meng et al., 2020), whereas fractional distillation follows the relative volatility order of the components (Manshad et al., 2016). The separation mechanism of PV depends on the permeability and selectivity of the membrane and on the diffusivity of the feed solution. Fractional distillation is limited by the liquid–vapour equilibrium, (Manshad et al., 2016) but there is no such barrier in PV and from this point of view, it can be said that PV is more advantageous than distillation. Moreover, the energy-saving feature of PV (Said et al., 2021) also makes it more acceptable than distillation as evaporation takes place in PV through the dense membrane (Matsui and Paul, 2003; Vane, 2005).

3.2. Pervaporation vs. reverse osmosis

The driving force for RO is the pressure difference between the feed and permeate sides of the RO membrane. In seawater desalination by RO, the osmotic pressure is to be surpassed by applying a high external pressure at the feed side to separate water from a solution of very high salt concentration (Fig. 4) and as a consequence, the fresh water producing ability decreases, which in turn reduces the RO performance, leading to critical environmental hazards (Zwijnenberg et al., 2005; Naim et al., 2015; Li, 2016). Mass transfer in RO occurs by the combination of solution-diffusion and pore flow model (Fig. 5) (Merdew et al., 2010). The convective flux or the “coupled flow” of the solute associated with the solvent flow controls the real solute mass transfer on the high-pressure side in RO, resulting in a net solute transport in the same direction as permeation (Ramani, 1992). Concentration polarization has a significant impact on the mass transfer in case of RO and thus models like the Kimura-Sourirajan and the Spiegler–Kedem model takes this phenomenon into consideration (Chen et al., 2020).

RO has several advantages with simple operational equipment and applied widely worldwide to secure pure drinking water (Jiang et al., 2018). Recently, Wang et al., 2016a reported that RO accounts for 60% of the desalination capacity for sea water and brackish water. Furthermore, ground water desalination can also be effectively processed by RO membrane technology (Pangarkar et al., 2011). RO process can not only be functional for removing salt from saline water but also has additional advantage to remove water pollutants like suspended solids, organic matter, colloids, color, nitrate contaminants, bacteria and viruses (Shoukai, 1999; Garud et al., 2011). However, despite several advantages, RO faces some challenges during seawater desalination which can be addressed by PV

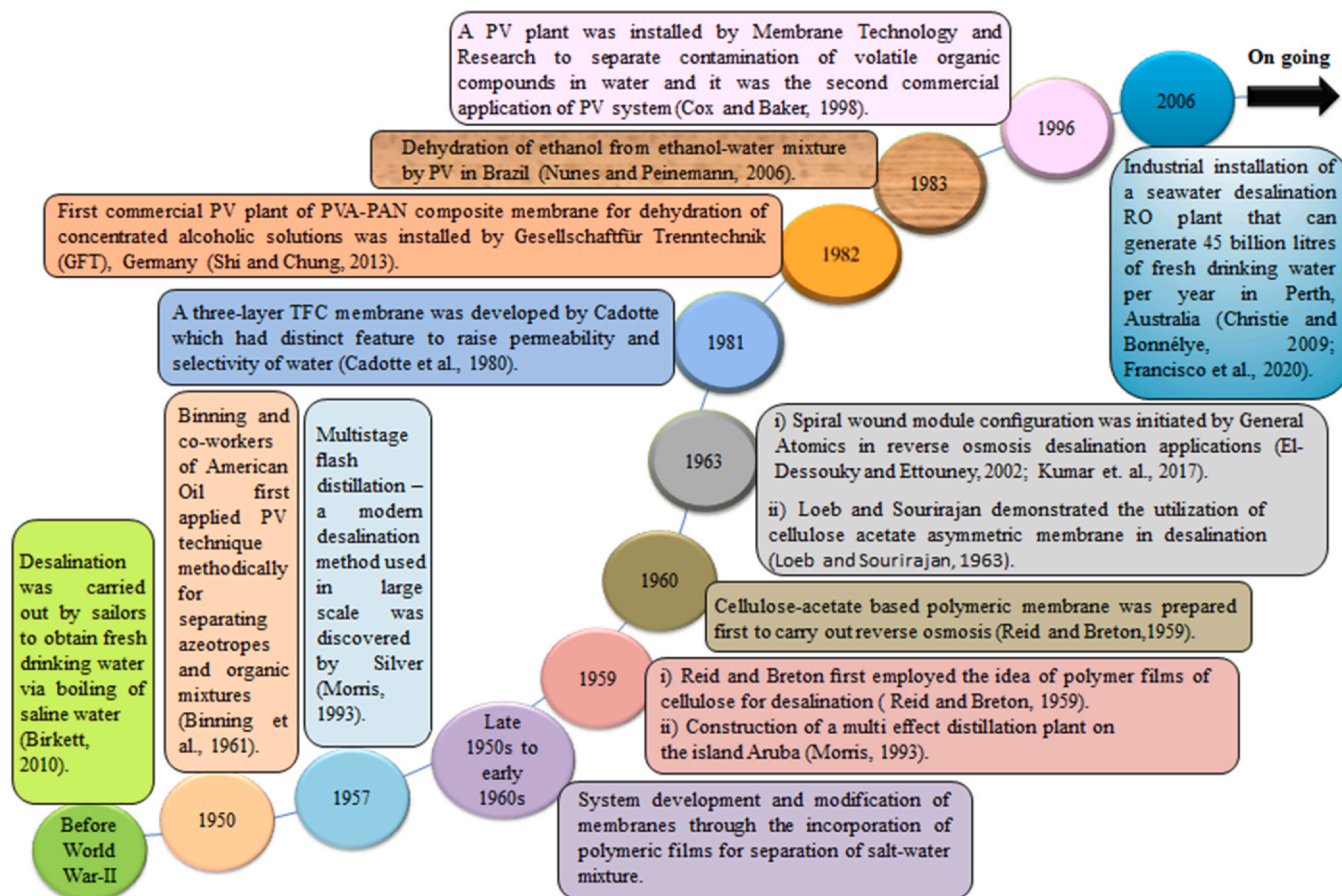


Fig. 3. History of pervaporation and desalination.

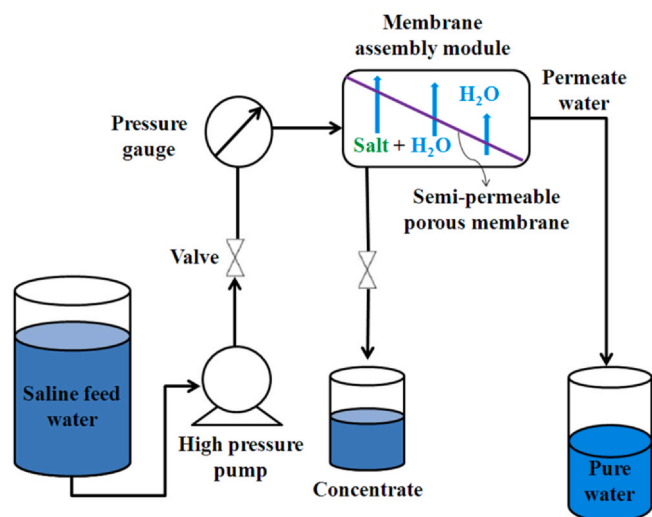


Fig. 4. Reverse osmosis filtration system.

desalination. The PV membrane material allows separating two or more components from a liquid mixture based on their difference in solubility and diffusivity in the solution. Thus, desalination of high salinity seawater may be successful with PV. An improvement of salt rejection and permeation flux may be achieved by using PV membrane for desalination. Here, the driving force is the chemical potential gradient which arises due to the difference in partial vapour pressure of feed liquid across the membrane between feed and

permeate sides (Huth et al., 2014; Ong et al., 2016). Since PV involves a phase change, there is no need to apply an excess external pressure larger than the osmotic pressure to harmonize the driving force as required in RO (Huth et al., 2014; Kuznetsov et al., 2007). More than 99% salt rejection is observed for monovalent salts (Chaudhri et al., 2015; Cho et al., 2011; Duke et al., 2009; Elma et al., 2012; Xie et al., 2011a,b) and the rejection of salt does not depend on the operating conditions (Cho et al., 2011). Thus, PV has potential to be advantageous over RO (Li, 2016).

3.3. Pervaporation vs. membrane distillation

MD and PV both are non-pressure driven membrane processes. Desalination via MD is a thermally-driven separation process (Alkhudhiri et al., 2012; Alklaibi and Lior, 2004; Camacho et al., 2013; El-Bourawi et al., 2006; Lawson and Lloyd, 1997; Drioli et al., 2015; Wang and Chung, 2015). MD involves evaporation of water vapor at liquid-vapor equilibrium, across the hydrophobic microporous membranes as distillate from the highly saline feed side liquid (Cho et al., 2011) (Fig. 6). The temperature difference between the two sides of the membrane creates a difference in vapour pressure, which is the driving force of MD (Wang et al., 2016a).

There are some notable differences between PV and MD:

i) wetting of membrane surface after a long term operation has an adverse effect on overall flux of hydrophobic MD membrane (Tijing et al., 2015; Warsinger et al., 2015). Wetting of the membrane surface takes place if the applied mechanical pressure is higher than the minimum liquid entry pressure of the membrane. It can be assumed that if salt crystals are deposited on the membrane surface as foulant during desalination, there is an increased risk of membrane

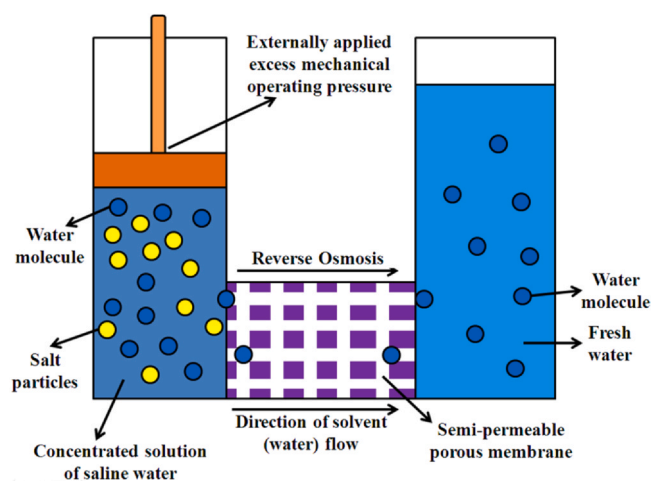


Fig. 5. Mass transport during reverse osmosis.

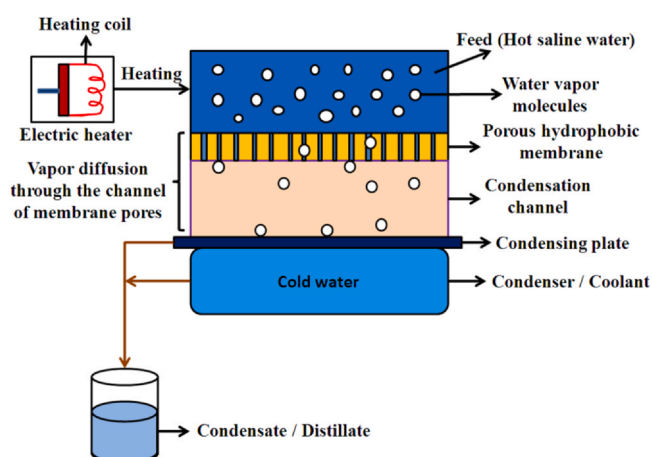


Fig. 6. Schematic diagram of membrane module assembly for membrane distillation.

wetting (García-Payo et al., 2000; Gryta, 2002, 2005, 2007; Cho et al., 2011), which is a threat for MD technique (Tijing et al., 2015; Warsinger et al., 2015).

On the other hand, in PV desalination, a hydrophilic polymer is chosen as membrane material, which is expected to be more fouling-resistant and have a better salt rejection as wetting of the dense membrane surface can be prevented (Wu et al., 2018). A hydrophilic membrane has high affinity towards water molecule due to the presence of H-bonds, which can selectively separate water vapour molecules resulting in a high permeation flux (Semenova et al., 1997; Chapman et al., 2008). Thus, the separation efficiency of PV is superior over MD.

ii) a PV membrane can actively participate in the separation, but an MD membrane does not participate in the separation process; it

rather plays a role of support medium at liquid-vapor interface (Khayet and Matsuura, 2004; Urtiaga et al., 2001).

iii) mass transport in MD occurs by three basic mechanisms—Knudsen diffusion, Poiseuille flow (viscous flow) and molecular diffusion (Alkhudhiri et al., 2012). Solution-diffusion mechanism helps to establish the mass transfer process across a PV membrane (Khayet and Matsuura, 2004; Semenova et al., 1997; Shao and Huang, 2007).

iv) permeation through a MD membrane depends on the pore structure, pore size, porosity and membrane tortuosity (Khayet and Matsuura, 2004; El-Bourawi et al., 2006; Tang et al., 2012). Permeate flux increases with membrane porosity and pore size but decreases at high membrane tortuosity value (Khayet and Matsuura, 2011; Boubakri et al., 2020). However, the permeability of a PV membrane is mainly dependent on the affinity of the membrane for water, membrane thickness and nanostructure (Korngold et al., 1996; Khajavi et al., 2010; Khayet and Matsuura, 2004).

v) commercial and industrial applications of PV are dehydration of organic solvents, removal of volatile organic compounds from aqueous phase by evaporation, separation of azeotropic mixtures (Qureshi et al., 2001; Shao and Huang, 2007; Villaluenga and Tabemohammadi, 2000; Yoshida and Cohen, 2003). The PV membrane material is highly water-selective, and does not allow organic substances to penetrate through the membrane, while such removal of organics is absent in MD because a hydrophobic membrane is generally used in MD. Thus, the separation efficiency and overall performance of PV is promising compared with MD (Li, 2016).

3.4. Pervaporation vs. vapour permeation

PV and vapour permeation (VP) are both energy efficient separation techniques as they can be operated by supplying a low temperature and pressure (Cannilla et al., 2017). These are also cost-effective routes as they involve a low operating cost (Marszałek and Kaminski, 2008; Sander and Soukup, 1988, 1991). Similar to PV, the separation process in VP is also independent of the relative volatilities of the components to be separated, but depend on relative diffusion rates of the components across the membrane material (Jyoti et al., 2015). VP also follows the solution-diffusion mechanism for separation through the membrane like PV. Despite these close similarities of PV with VP, there are also some significant differences (Ohlemann, 2012) which are discussed in Table 1.

3.5. Comparison of energy consumption between reverse osmosis, membrane distillation and pervaporation

Two types of energy are mainly required to run a PV desalination system i) thermal energy, required for heating of the feed solution and ii) electrical energy, needed to create vacuum for feed circulation. Among them thermal energy requirement is predominant which increases with rise in temperature (Mericq et al., 2011). The mechanical pressure required in RO to surpass the osmotic pressure of saline water is ranging from 5.5 MPa to 6.8 MPa (Kaminski et al., 2018) where energy is consumed approximately between 2–4 kWh/m³ (Amy et al., 2017; Gude, 2011). PV itself can generate the driving

Table 1
Differences between pervaporation and vapour permeation.

Pervaporation	Vapour permeation
Feed mixture is always in liquid phase.	Feed mixture is in gaseous or vapour phase.
Driving force in PV is the difference of chemical potential (between the feed and permeate side of the membrane).	Difference in partial pressure of the component gases or vapours is the main driving force.
PV is a phase transformation process from liquid to vapour which requires enthalpy of vaporization.	No phase transformation takes place.
Membrane and feed solution are directly in contact. So, membrane surface spoiling can occur.	Membrane and feed solution are not in direct contact with each other. So, longevity of the membrane can be expected.

force to run the system, where there is no need to overcome the osmotic pressure of saline water by applying external mechanical pressure. So, the energy consumption of PV does not include the energy requirement for mechanical pressure. The energy criteria for vacuum membrane distillation (VMD) and PV desalination are very much similar from the perspective of laboratory setup which requires thermal energy to get hot feed and electrical energy to generate vacuum at the permeate side (Criscuoli et al., 2008; Alkhdhiri et al., 2012). However, the more common experimental type of MD is direct contact membrane distillation (DCMD) in which the hot feed and permeate side both are in contact with cold water (coolant). On contrary the main problem associated with DCMD is that quite an amount of energy gets diminished while the membrane comes in contact with water (Fig. 6) (Ashoor et al., 2012; Rahimpour et al., 2019). Bolto et al. (2010) from their observation opined that PV desalination is a much more energy efficient process than MD, thus making PV a cost-effective process compared to its counterpart i.e., MD. Therefore, energy saving feature of PV makes it more suitable than other desalination processes (for example, RO, MD) for water desalination. Furthermore, energy consumption and cost of desalination depends on source (sea/ocean, mines and geothermal resources) of saline water. Overall, the energy consumption of PV desalination system can be evaluated by the amount of different energies (thermal, electrical) consumed and the quantity of treated water (Deniz and Çınar, 2016).

4. Ceramic membrane substrate for pervaporation desalination

Silica and alumina are the major components of a ceramic substrate including carbon, zeolite, various metal oxides (titania, zirconia), metals like palladium, silver and their alloys (Wijmans and Baker, 1995; Wang, 2008). Depending on their structure, ceramic membrane can be classified into two broad categories: porous (amorphous and crystalline) and dense (non-porous) (Malekpour et al., 2008, 2011; Khajavi et al., 2010). The substrates can be tuned into different architectures (e.g., tubular, flat sheet, hollow fiber etc.) with variable packing densities due to their good mechanical and thermal stability. The use of ceramic membranes (e.g., zeolites and amorphous microporous silica) in PV desalination has gained interest over polymeric membranes due to their good separation capabilities, thermal and chemical stability, rigid structure, low fouling tendencies and resistance to swelling (McLeary et al., 2006). The separation performance of ceramic membranes that depends on different sizes of molecular-sieves and size of the particle to be separated, may be controlled by varying pore sizes of the membranes. Here pore structure and surface charge are responsible for mass transfer and salt rejection (Wang et al., 2016a). The transport phenomenon through a molecular-sized microporous inorganic membrane is governed by adsorption–diffusion through surface diffusion (Bowen et al., 2004; Wee et al., 2008). Elma et al., (2012) reported that for microporous silica derived membranes in PV desalination, water transport follows sorption-diffusion mechanism due to hydrophilic nature of silica based membranes which is basically a surface adsorption process. Additionally, a selective layer (e.g., polymeric) is often synthesized over a porous ceramic support membrane to increase membrane permeability, selectivity and salt rejection. Support substrate materials are sometimes functionalized with acid or alcohol and then polymeric skin layers are crosslinked over the functionalized or non-functionalized ceramic support tube for a better adherence of the two layers (Wu et al., 2015). Clinoptilolite/phosphate composites are mechanically stabilized by chemical bond formation between zeolite and phosphate (An et al., 2014). These clinoptilolite-phosphate membranes are advantageous from the view point of ion leakage prevention and enhancement of salt rejection. The flux was measured to be 15 kg/m²h for desalination of feed with 1.4 g/L concentration and greater than 95% NaCl salt

rejection was obtained at 95°C (An et al., 2014). Singh et al. (2015) developed a carbon templated amorphous silica based membrane which could deliver comparatively low water flux than usual PV desalination membrane (2.6 kg/m²h) with high salt rejection (99.9%) at 25°C feed side temperature from 40 g/L NaCl concentration. Carbon templated silica (CTS) membranes were also fabricated by Wijaya et. al. (2009) via a sol-gel process and the effect of carbon chain length of surfactant templates was investigated. The longest carbon chain surfactant (cetyltrimethylammonium bromide, C16) derived CTS membrane exhibited a low water flux of 3 kg/m²h with average salt removal (97%) at 20°C operating condition when the feed concentration was 3 g/L NaCl. Kujawski et al. (2007) prepared fluoroalkylsilane (FAS)-modified PV membranes by grafting 1H, 2H, 2H-perfluorodecyltriethoxysilane (C8) molecules onto ceramic nanofiltration membranes. The PV desalination performance of the modified membranes showed a flux of 5 kg/m²h for 30 g/L NaCl feed solution at 40°C feed temperature with high salt separation. In another study, Malekpour et. al. (2011) synthesized LTA (linde type A) and MFI zeolite membranes on the surface of an α -alumina porous support by a hydrothermal method. However, they used the membranes to remove cationic and anionic species from aqueous solutions by the PV process. Therefore, ceramic supported composite membranes might be appropriate in PV desalination application and can definitely be recommended based on their notable separation performance, physical stability and mechanical strength.

5. Selective layer for pervaporation desalination membranes

The fabrication of a composite membrane is very important for obtaining a suitable water selective membrane through which water can permeate preferentially. Therefore, the selection of a polymer is a very important criterion to prepare a good dense membrane through which a better PV performance and salt rejection can be achieved. Some specific polymers that are used for the preparation of PV desalination membranes are discussed in next three Sections (5.1, 5.2, 5.3) and other polymers are summarized in Table 2.

5.1. Block copolymers

Block-copolymers are suitable materials for fabricating dense membranes. For example, a block-copolymer, poly(ether-block-amide) (PEBA), contains two segments - one rigid and one flexible polymeric part. The rigid polyamide segment provides mechanical strength and stability and the soft flexible polyetheroxide segment contributes in good permeability (Panek and Konieczny, 2008; Hamouda et al., 2011, Nigiz et al., 2017). Marian et al. (2017) synthesized three composite membranes, PEBA/PAN (Polyacrylonitrile)/PE (Polyester), PEBA/PSF (Polysulfone)/PE and PEBA+NaX/PSF/PE by phase inversion. A permeate flux of 4.93 kg/m²h with salt rejection of 99.9% was reported at feed temperature 45–50°C. The water contact angle of these three membranes was ~53°, which suggests hydrophilicity, and thus the water vapour sorption rate was high. However, the membranes are yet to be scaled up and commercialized. Nigiz et al. (2017) prepared zeolite-3A/PEBA membrane via solution mixing and casting. They obtained a flux of 4.33 kg/m²h at a feed temperature of 60°C and 30 g/L NaCl concentration with 99.16% salt rejection. At 40°C, 99.63% salt rejection was obtained using the same membrane and reported fluxes were 3.30, 3.10, 1.98 kg/m²h corresponding to varying NaCl concentration of 10 g/L, 30 g/L and 100 g/L, respectively. This occurs may be due to change in polarity of the solution. Additionally, with an increase in NaCl concentration, water content gets reduced and flux decreases due to change in driving force, for diffusion of water vapour molecules through the membrane, which is maintained by partial vapour pressure of the water (Xie et al., 2011b). Reduction of water flux also takes place as high concentration of salt decreases the thermodynamic activity of

Table 2

Advantages, disadvantages and research gap of existing PV desalination membranes mentioned in Fig. 7.

Symbol	Comment	Research gap	Ref.
	Operating condition is simple, no salt leakage and pore-plugging problems.	(a) Low water flux (b) Commercially not implemented.	(Zwijenberg et al., 2005)
	(a) Better chemical and thermal stability can be obtained with good salt rejection efficiency. (b) Pervaporation desalination by the inorganic membrane was applied for natural seawater desalination.	(a) Cost-effective production is difficult. (b) Structural defects in membrane.	(Cho et al., 2011)
	Desalination of highly saline sea water can be carried out at a feed pressure of 34–48 bar.	Very high degree of swelling (110.9%).	(Li et al., 2011)
	The final permeate concentration of ions was lower than 0.02 mg/L.	(a) Maximum impurities retained in the feed side. (b) Presence of structural defects in the membrane. (c) Very high operating temperature.	(Khajavi et al., 2010)
	Applied for treatment of highly saline waste water for irrigation.	Lower flux.	(Sule et al., 2013)
	Mechanical stability gets increased due to incorporation of PAN.	High extent of crosslinking reduces hydrophilicity due to absorption of hydrophilic functional groups.	(Liang et al., 2014)
	Exfoliation of GO particles takes place.	Thicker deposition of GO increased the thickness of GO-PAN composite, which affected water flux and membrane efficiency.	(Liang et al., 2015)
	(a) Low pressure difference, approximately 1 bar is required to desalinate brackish water and sea water. (b) Industrial application of the membrane over a long time period can be attributed to remarkable hydrostability of the membrane.	Degradation of silica membranes occurs due to hydrolysis of siloxane bonds.	(Ladewig et al., 2011)
	(a) Mechanical stability of the membrane. (b) Ion leakage can be inhibited.	(a) Inferior particle dispersion. (b) Structural defects in the membrane.	(An et al., 2014)
	(a) Chemically and thermally stable membrane with mechanical rigidity. (b) Silica initiated crosslinking.	Complicated and time-consuming fabrication process.	(Chaudhri et al., 2018)
	Mechanical stability was provided by PAN.	Hydrophilicity was decreased with enhancing degree of crosslinking as hydrophilic functional groups get removed (O–H of PVA) on crosslinking of PVA with excess PMDA. As a consequence, lower flux was obtained.	(Zhang et al., 2018)
	—	(a) Pore sizes of microporous silica film enlarged when interaction of water with silanol groups occurred and the membrane lost its stability during desalination.	(Wijaya et al., 2009)
	(a) Porosity increased when carbon content increases by 10%.	Silica membrane may lose hydrostability on hydrolysis of siloxane bonds.	(Ladewig et al., 2011)
	Incorporation of cobalt oxide in the silica matrix for betterment of hydrostability of silica.	Low productivity of water flux.	(Lin et al., 2012)
	Investigation of the PV performance at different temperatures with varying concentration of the NaCl–water solution.	High production cost.	(Drobek et al., 2012)
	High salt rejection was found.	Packing density was found to be lower.	(Feng et al., 2017)
	Desalinate NaCl–water solution.	—	(Xu et al., 2016)
	—	Enlargement of GOF membranes in water due to oxidized graphene.	(Feng et al., 2016)
	(a) Suitable for desalinating 10 and 20 wt% brine solutions. (b) PVA–SBQ is a photo-crosslinkable polymer. (c) Incorporation of nanocellulose as a transition layer to tune pore size.	(a) Some acryl groups may remain unreacted. (b) Costly fabrication process.	(Malekpour et al., 2011)

water molecules. (Korin et al., 1996; Wang et al., 2016a). Another reason for decline in water permeation flux may be attributed to concentration polarization when salt concentration near the membrane surface increases. (Huth et al., 2014; Wang et al., 2016a). Molecular sieves of the zeolite 3A gave higher salt rejection efficiency. The zeolite 3A incorporated PEBA composite membrane was moderately hydrophilic in nature with contact angles ranging from 63°–71°. In another study, Nigiz (2018), fabricated a GNP (Graphene nanoplates) / PEBA membrane with superior antifouling property using a solution mixing and casting procedure. A maximum flux of 5.12 kg/m²h was reported at 65°C feed temperature with salt rejection above 99.89% and when temperature was lowered to 35°C, the flux decreased to 2.58 kg/m²h but a high salt rejection of 99.94% was obtained. Membranes were moderately hydrophilic in nature with contact angle values 75° and 80° when 5 wt % and 2 wt % GNP were used, respectively. Wang et al. (2016b) synthesized sulfonated

poly(styrene-ethylene/butylene-styrene) (S-SEBS) following a grafting technique in which sulfonic groups from acetyl sulphate (sulfonating reagent) were grafted on SEBS triblock copolymer. A high water flux of 22.87 kg/m²h at 63°C feed temperature, 6 kPa permeate pressure and 1 wt% NaCl feed solution was obtained for the membrane with 54.1% sulfonation degree. They claimed that the salt rejection (> 99%) was independent of the sulfonation degree and operating conditions. The unmodified SEBS membrane was hydrophobic (CA ~100°) in nature, whereas modified S-SEBS membranes were superhydrophilic in nature. The water contact angle of the SEBS membrane was measured to be ~100°. The S-SEBS membrane with sulfonation degree of 54.1% exhibited the lowest water contact angle (23°). When the sulfonation degree was varied from 0% to 54.1%, the water uptake rose from 0% to 189% while the contact angle decreased from 100° to 23°. Authors claimed that with increasing degree of sulfonation, the hydrophilicity, water permeate flux and anti-

fouling property of the membrane also improved compared to other PV desalination membranes. In another study Geise et al. (2010) used sulfonated blocked co-polymer for the preparation of a PV desalination membrane. They studied a series of sulfonated pentablock copolymers to characterize their potential as chlorine-tolerant membrane materials for desalination applications. They concluded that the water and salt transport properties of this type of polymer depend on the degree of sulfonation, block molecular weights, and the film casting technique. Additionally, a greater degree of sulfonation led to an increase in water permeability of this membrane. Thomas et al. (2020) demonstrated the performance of free-standing self-assembled pentablock terpolymeric membranes in the field of PV desalination. They used Nexar™ (tradename) which was a partially sulfonated pentablock terpolymer. Following 52% degree of sulfonation, it exhibited a remarkable permeance with high salt removal of 99.5% and also a high flux of 3.32 kg/m² h at room temperature with 32 g/L NaCl feed solution. It was reported that the membrane permeance and salt separation performance of dense, non-porous Nexar™ in PV desalination was better than with commercially available membranes.

Most of the studies involving block polymer as selective layer for PV desalination membrane are focused on the development of fully polymeric membranes. The use of block copolymers for composite membrane preparation over a ceramic support is scarce. Wu et al. (2018) prepared a hydrophilic PEBA membrane via solution casting and applied for PV desalination of high salinity water. They reported an almost complete salt rejection (> 99.9%) with a low flux of 1.68 kg/m²h at optimum feed condition of 65°C. However, when salt concentrations were varied from 1–20 wt%, the flux was reduced to 50% with no change in salt rejection. Additionally, the type of salt (i.e., NaCl, MgCl₂, Na₂SO₄) did not affect salt rejection. However, with increasing feed temperature from 25°C to 65°C, the pure water flux increased from 1.16 to 1.68 kg/m²h. Irreversible fouling, which is rather common for ceramic membranes (Munla et al., 2012; Shang et al., 2015; Zuo et al., 2020), was not observed during their study.

5.2. Cellulose

Cellulose is another promising membrane material for PV desalination with high hydrophilicity. Polymer chains are densely packed in the cellulosic structure by an intermolecular hydrogen bonding network. A cotton cellulose membrane was prepared by Kuznetsov and co-workers, (2007). At 40°C feed temperature, a 6.7 kg/m²h water flux with 100% salt rejection of 40 g/L NaCl was obtained. This membrane showed a higher permeability due to its high hydrophilic nature. A super-hydrophilic cellulose acetate membrane for PV desalination was also synthesized by Naim et al., (2015) by a simple phase-inversion route with asymmetric membrane morphology. The highest flux of 5.97 kg/m²h was reported for 40 g/L initial NaCl concentration at 70°C with a salt rejection of 99.7% indicating a high performance of the membrane. They found that with increasing feed concentration, flux decreases; e.g., with 140 g/L feed concentration, the flux became 3.45 kg/m²h but the salt rejection remained almost constant (> 99.7%). Elewa et al. (2016) developed an innovative deacetylated cellulose acetate membrane, which showed a sufficient flux of 4.11 kg/m²h at 70°C feed temperature (with 99.9% salt rejection for 120 g/L feed concentration) in desalination of saline water by PV. Prihatiningtyas et al. (2019) synthesized a cellulose triacetate/cellulose nanocrystal (CTA/CNCs) nanocomposite PV membrane using solution casting. Cellulose nanocrystals (CNCs) are hydrophilic with good mechanical properties. The sponge-like structure transformed to a self-assembled structure through incorporation of CNCs into CTA. The water flux increased from 2.16 to 5.76 kg/m²h when 3% CNCs were incorporated into a CTA membrane. It was also reported that flux was three times higher if the temperature increases from 30° to 70° C; a higher temperature enhances the diffusion rate of

water molecules through the membrane. The highest flux of 11.68 kg/m²h with 99.9% NaCl removal was obtained in PV desalination for a CNCs 3%-CTA nanocomposite PV membrane at operating temperature of 70°C. The feed concentration was varied within the range of 30–90 g/L. In Prihatiningtyas et al. (2020b) successfully fabricated another cellulose triacetate/Ludox-silica (SiO₂) nanocomposite as PV desalination membrane following solution casting. The 4 wt% SiO₂ containing cellulose triacetate/Ludox-SiO₂ membrane exhibited a flux of 6.1 kg/m²h with NaCl separation of 99.8% at 70°C feed temperature with 30 g/L NaCl in the feed. Besides the capability of forming dense film, CTA as a polymeric material could also provide a high salt rejection with lower fouling tendency and mechanical strength. Silica nanoparticles are advantageous as nanofillers because of their hydrophilic nature and low toxicity. The measured water contact angle values of the developed nanocomposite membranes were within the range of 56.8°–68.9° indicating hydrophilicity of the membranes. A cellulose triacetate/Al₂O₃ membrane was also prepared by Prihatiningtyas et al. (2020a) to desalinate hyper saline solutions, which showed an excellent performance in PV desalination with a maximum flux of 6.85 kg/m²h and 99.8% salt removal at 70°C.

5.3. Polyvinyl alcohol and polyacrylonitrile

PVA and PAN are very common suitable polymers used to prepare PV desalination membranes. Liang et al. (2018) fabricated a PAN/PVA-based PV desalination membrane by dip-coating, where a sulfonic acid functionalized PVA membrane supported on PAN was used. Cross-linking between the porous PAN substrate and the selective layer of PVA was the main mechanism for densification of the membrane. 4-Sulfophthalic acid was used as cross-linking agent and sulfonating reagent that provided the passage for transport of water molecules. A higher flux of 46.3 kg /m²h with a salt rejection of 99.8% was observed for 35 g/L NaCl feed solution at 70°C and a permeate side pressure of 100 Pa. A reduction in water flux was observed when the feed salt concentration was very high (e.g., flux: 22.1 kg /m²h, feed concentration: 100,000 ppm NaCl solution, temperature: 70°C) although the salt rejection was very good (> 99.5%). A glutaraldehyde (GA) cross-linked PVA/polyvinylidene fluoride (PVDF) PV composite membrane was developed by Li et al. (2017) via a simple dip-coating method. The desalination process produced a flux of 13.7 kg/m²h at an operating feed temperature of 80°C for 100 g/L NaCl solution with a highly stable salt rejection of 99.99%. A slightly higher flux of 16.38 kg /m²h was obtained with Milli-Q water at the same operating temperature. This PVA/PVDF membrane can be considered a very suitable candidate for treatment of concentrated brine solution because of its storage stability, antifouling property and cleaning efficiency. Recently, Zhang et al. (2019) synthesized a sulfosuccinic acid crosslinked PVA/PAN membrane by solution mixing and casting. A maximum flux of 27.90 kg /m²h was reported at 70°C feed temperature for 35 g/L feed solution with 99.80% salt rejection. A higher feed concentration of 100 g/L showed a minimum flux value of 4.50 kg/m²h at 30°C. The measured water contact angle value was 77.1 ± 3°. Water-soluble PVA was crosslinked by sulfosuccinic acid in order to improve stability in water. On the other hand, PAN was responsible for the mechanical strength and thermal stability of the membrane. A multi-walled carbon nanotubes doped PVA membrane was prepared by Yilman et al. (2018) via solution mixing and casting; this membrane had over 92% salt rejection at 40°C for a mixture of NaCl, MgCl₂ and KCl. Some advantageous outcomes were obtained by the membrane including a higher hydrophilicity, film forming capability, superior antifouling nature and higher adsorption property. Similarly, Li and Wang, (2010) developed a PVA/MA (maleic anhydride)/silica membrane, which could produce a flux of 5.51 kg /m²h with > 95.50% salt rejection at 22°C. They found that with an increase of the silica content

in the membrane, the flux is lower (> 3.65) but the salt rejection remains the same (the probable reason is explained in Section 5.1). Chaudhari et al. (2018) fabricated an efficient PV desalination membrane by the reinforcement of a PVA-silica film on a porous polysulfone (PVA-SiO₂/PSF) hollow fiber. At 60°C, depending on the membrane thickness, the highest flux values varied from 20.60 to 11.90 kg/m²h with unchanged salt rejection efficiency (99.90%) in all the cases for feed concentrations of 2000 mg/L, 30000 mg/L and 50000 mg/L. A PVA-intercalated graphene oxide (GO) membrane supported on a commercial mixed cellulose ester (MCE) micro-filtration membrane was prepared by Sun et al. (2020) via pressure-assisted filtration for desalination of high-salinity water through PV, in which PVA chains were responsible for cross-linking. The optimized GO-PVA membranes showed an excellent flux of more than 98 kg/m²h with 99.99% salt separation at 85°C feed temperature with 10 wt% NaCl as feed. Three dimensionally cross-linked GO nanosheets network could implement a brick-mortar like structure in GO layers followed by a high adhesion strength leading to a stable PV performance. Carbon nanotube (CNT) incorporated PVA membranes were fabricated by Yang et al. (2020), which were actually PVA based hybrid membranes with dispersed nanochannels. The PVA/CNT hybrid membranes (PVA crosslinked with maleic acid (MA*) and functionalized multi-wall carbon nanotube) displayed 99.9% of salt rejection at a feed temperature of 30°C in desalinating 3.5 wt% aqueous solution of NaCl by PV.

Permeate flux and salt rejection are the two main performance parameters for all membrane-based desalination processes. RO processes produce permeate flux in the range of 30–60 kg m⁻² h⁻¹ consuming energy between 2–4 kWh m⁻³, whereas, present PV and MD processes provide lesser permeation (0.05–15 and 2–30 kg m⁻² h⁻¹, respectively) using a bit less energy than RO, typically around 2 kWh m⁻³ for PV and 0.6–1.75 kWh m⁻³ for MD Castro-Munoz, (2020). Ideally, water flux in case of PV desalination can be considered satisfactory from the point of view of commercial viability when it is comparable to that of RO for equivalent amount of energy consumed. Thus, membranologists are focused on development of PV desalination membranes with higher efficiencies and long-term stability since the overall process efficiency and cost is highly governed by the membrane performance.

A comparison in performance of different kinds of PV desalination membranes (tagged as 1–24) is shown in Fig. 7. It is observed that almost all the membranes exhibited greater than 99% salt removal with range of flux between 1–25 kg/m²h (Fig. 7). Name of the membranes with operating condition such as feed temperature and feed concentration are also listed in the Fig. 7. Additionally, the advantage(s), disadvantage(s) and research gap of some of these PV desalination membranes are critically evaluated in Table 2.

6. Potential membrane for stable and high flux PV desalination

In recent years, research on the application of composite membranes for PV has shifted towards exploration of new chemically and thermally stable materials suitable for use at elevated operating temperatures. Ceramic supported polymeric composite membranes are hybrid membranes in which a thin active layer of appropriate polymer is coated over a porous ceramic substrate (Roy et al., 2020). These composite membranes are prepared by coating of a polymeric film on a microporous or mesoporous ceramic support. Swelling is a critical issue compromising the long-life stability of hydrophilic PV desalination membranes. This may occur due to penetration of the water vapour molecules through the membrane, which leads to potential changes in membrane morphology. As a consequence, diffusion rates of water vapour molecules through the membrane may be enhanced (Binning et al., 1961; Mulder and Smolders, 1984). Thus, the polymeric materials which are used in the composite membrane as an active layer often have to be cross-linked (possibly

by heat treatment to enhance stability and to reduce swelling (Ong et al., 2016; Wu et al., 2018). More than 99% salt rejection is expected by these membranes due to the non-volatility and low diffusivity of salt. Composite membranes that are anisotropic in nature can be applied commercially because of their productivity and improved fluxes (Purkait et al., 2018). Thus, the development and application of ceramic supported polymeric composite membrane may be of use for the emerging PV desalination technology.

7. Role of crosslinking on pervaporation membrane

Several membrane preparation processes such as dip coating (Elshof, 2015; Sahoo et al., 2018), phase inversion (; Marian et al., 2017; Naim et al., 2015; Strathmann and Kock, 1996), and interfacial polymerization (Cadotte et al., 1980; Cadotte, 1985; La et al., 2010) have been adopted to prepare PV desalination membranes. However, these coating processes are not able to change the microporosity of the PV membranes and thus often temperature has been used during fabrication to tailor the intrinsic membrane properties towards densification (Ilinitch et al., 1999). The crosslinking temperature has a significant effect on the membrane structure and thus directly affects the membrane performance. It affects the characteristics of the active layer and simultaneously helps to tailor the membrane surface properties and pore size. Often a higher temperature leads to more crosslinking, resulting in a smaller pore size (Chen et al., 2020). Generally, two types of cross-linking are applied: thermal cross-linking by heat treatment at a particular temperature and chemical cross-linking, which depends on reaction pathways. Thermal cross-linking is widely applied during the preparation of dense PDMS (polydimethylsiloxane) and PEBA membranes for PV application (Gu et al., 2013; Wu et al., 2015; Xue et al., 2020). The low flux is the main hurdle that needs to be overcome by PV membranes prior to wide industrialization. Therefore, much effort has been given on the development of high flux PV membranes. It was observed that a very small variation in membrane structure can dramatically enhance the flux (Robeson et al., 1994). Water flux is directly related to hydrophilicity of the membrane that can be enhanced by increasing hydrogen bonding or ion dipole interaction between water molecules and selective layer materials (Lim et al., 2021). On the other hand, water diffusivity by the membrane can also be fine-tuned by regulating three parameters i) morphology of membrane nano channels, ii) size of hydrophilic domain and, iii) polymer free volume (Gavrilescu et al., 2015; Pazda et al., 2019). The feed side of the dense membrane usually takes part in sorption equilibrium during PV desalination process and the other part (i.e., the bottom edge) remains dry. Therefore, tailoring the material and chemical property of the selective layer polymer is essential to improve the process. Researchers have also suggested that polymer crosslinking facilitates water transport through the membrane (especially from the dry region) by creating additional water diffusion paths via generating carrier-water complexes (Mansur et al., 2008; Bolto et al., 2009). To address this limitation, several researchers focus on tailoring the material and chemical properties of the hydrophilic polymers that have been used to prepare PV desalination membranes. Sulfonation of an aromatic polymer is a very well-known crosslinking mechanism that can form hydrophilic nanochannels for water transport. 4-Sulfophthalic acid was used as a crosslinking agent by Xue et al. (2019) to optimize the trade-off between separation performance and chemical stability of the PV desalination membrane. They found that with higher crosslinking degree the dependency of water diffusivity on water concentration reduces. Molecular dynamic simulation indicates that sulfonic groups act as a fixed facilitated transport agent that reversibly reacts with water and the mobility of water molecules and their interaction increases with increasing SPTA content. In another study modified PVA (sodium iodate treated) was crosslinked by FS-3100 (fluorocarbon based

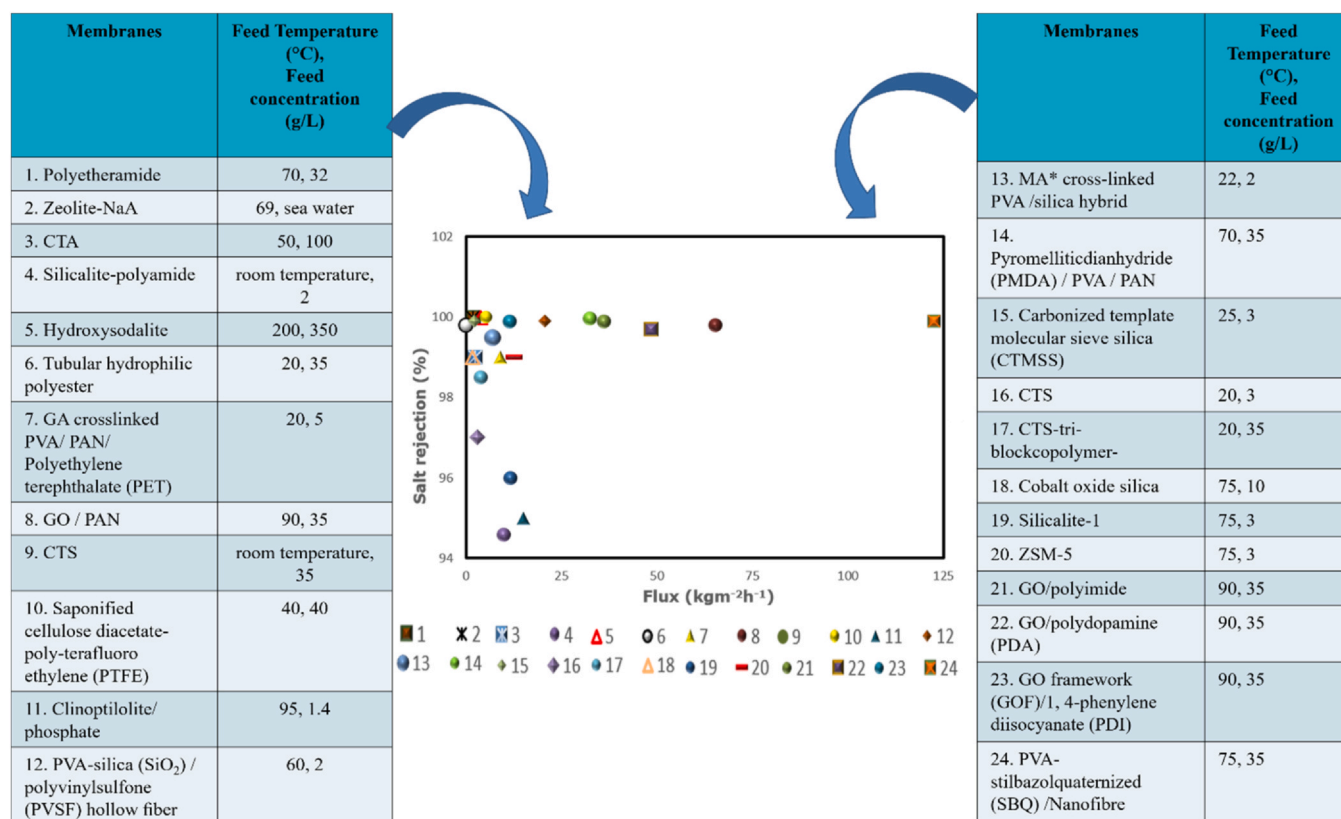


Fig. 7. Membranes for pervaporation desalination, operating conditions (feed salt concentration and temperature), flux and salt rejection. 1. Zwijnenberg et al. (2005), 2. Cho et al. (2011), 3. Huth et al., 2014, 4. Li et al. (2011), 5. Khajavi et al. (2010), 6. Sule et al. (2013), 7. Liang et al. (2014), 8. Liang et al. (2015), 9. Ladewig et al. (2011), 10. Kuznetsov et al. (2007), 11. An et al. (2014), 12. Chaudhri et al. (2018), 13. Xie et al., 2011a, 14. Zhang et al. (2018), 15. Duke et al. (2007), 16. Wijaya et al. (2009), 17. Ladewig et al. (2011), 18. Lin et al. (2012), 19. Drobek et al. (2012), 20. Drobek et al. (2012), 21. Feng et al. (2017), 22. Xu et al. (2016), 23. Feng et al. (2016), 24. Malekpour et al. (2011).

crosslinker) to improve the hydrostability and chlorine resistance of PVA. The crosslinked membrane was very stable at extreme pH condition (2–12) with 99.93% salt rejection (feed NaCl concentration: 35,000 mg/l, temp: 70°C) and considerable water flux ($34 \pm 1 \text{ kg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$). The membrane also showed excellent antifouling property with 100% flux recovery even after 1200 h operation (Zhao et al., 2020). However, since FS-3100 does not contain any polar group, the permeability was around 50% lower than in the case of sulfonation.

8. Conclusion and future prospects

PV can be used to treat highly saline water without needing to create much vapour pressure difference across the membrane. Feasible applications of pervaporative desalination can be made more promising by fabricating suitable hydrophilic membranes with high flux. Polymer crosslinked ceramic composite membranes can be promising, with the advantage of being rigid and stable (thermal, chemical and mechanical stability) as a ceramic support layer together with a tuneable polymeric active layer. In order to develop hydrophilic dense organic-inorganic hybrid (ceramic-polymer composite) membranes, the idea of polymer crosslinking is taken into consideration for upgradation physical properties and chemical tolerance, antifouling nature, separation performance, hydrophilicity. A hydrophilic polymeric active/selective layer on ceramic support can be fabricated by using a suitable polymer with hydrophilic functional groups, viz. PVA, cellulose, or chitosan. A cross-linked state of these polymeric functional layers can give greater stability and suppress swelling during PV operation. Therefore, desalination by PV can be practically implemented in coastal areas to separate salt from highly saline sea water in order to make it suitable

for drinking by rejection of more than 99% salt. The water produced by PV desalination might also be useful in agriculture. PV technique can result in major energy reductions. In the near future, this technique is expected to develop further and be implemented on a larger scale. The efficiency of PV technique should save the environment in an eco-friendly way and can be used in the area of water technology based economy.

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.

Acknowledgements

The authors are grateful to the Department of Science and Technology (DST/INSPIRE/04/2016/000597) and Science and Engineering Research Board (CRG/2018/002021), Government of India for providing financial support for accomplishment of the work.

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