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Current Advances in Membranes for Osmotic Power Generation: A Review

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ABSTRACT

With the target of zero waste policy and renewable energy harvesting, the osmotic power generation by pressure retarded osmosis (PRO) from salinity gradient is an exciting and yet challenging problem for water management technologies to achieve water and energy sustainability. In recent years, the production of high-performance PRO membranes has earned increased concern, although many controversies are surrounding its environmental impact and practicality. Therefore, a detailed and up-to-date evaluation of key advances in PRO membrane engineering applications made in recent years is discussed in this review. Moreover, it is aimed to provide updated insight on the significant developments in advanced fabrication and modifying techniques of latest PRO membranes. The increased performance using various configurations and materials, which are also analysed in depth based on the point of view of design rationales. Furthermore, the problem faced in membrane development is addressed with suggested solutions are explored. Lastly, the potential outlook of PRO membrane application in practical scenario are also discussed.

Keywords: Osmotic power, pressure retarded osmosis, thin film composite membrane, recent trends in membrane fabrication and modification

1.0 INTRODUCTION

Rapid population growth and global climate change, demand for sustainable energy options that are clean and green. One of the proposed renewable energies is by contacting two different salinities to generate energy, also known as osmotic power [1]. Consequently, the mixing of fresh and salt water, which commonly occurs in estuaries, can be seen as a large energy treasury. The global electricity output capacity using this method was calculated at 1600 TWh per annum [2]. Confronted with stringent environmental regulation and high energy cost, the potential of osmotic power is an effective renewable energy source worth exploiting [3].

To explain the fundamental principal of osmotic power, two

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important concepts to address are forwards osmosis (FO) and pressure retarded osmosis (PRO). FO rely solely on the osmotic pressure as the only driving force for the transport of water from a solution with low salinity to high salinity. Moreover, since FO is fully driven by osmotic pressure, so the solution are not pressurised [4]. Similar to FO, PRO is also driven by the osmotic pressure. The additional feature of PRO is the hydrostatic pressure applied on the draw side, in opposite direction of the osmotic pressure. In this case, the hydrostatic pressure will slow down due to the increasing ΔP when water level rises. Therefore, it is important to maintain stable pressure and concentration at the feed side to provide constant flow. When the water volume accumulated to a certain degree, the draw side will be depressurised, and this additional volume flow will be used for power generation [5].

In general, power generation techniques currently available from osmotic pressure gradient energy use membrane-based technologies such as pressure retarded osmosis (PRO) and reverse electrodialysis (RED) [6-8]. In comparison, PRO exhibited greater efficiency and higher power density, and is better suited to extract power from high salinity gradient [8]. In a standard PRO process, water spontaneously permeates from the feed side to the pressurised high-salinity draw solution side across the semi-permeable membrane. Consequently, the diluted salt water's volume and hydraulic pressure are increased which enables the generation of power by depressurising the solution through a hydro-turbine [9]. When a source of low salinity such as river water is available, sea water can serve as the high salinity source, and energy production can take place by mixing both streams through PRO process. In fact, the concept has been implemented by a Norwegian company Statkraft, and they built a demonstration PRO power plant with a power output between 2-4kW [10]. Based on Statkraft research, the minimum power density of flat sheet and hollow fiber membranes should be at minimum 5 W/m^2 and 3 W/m^2, respectively, for commercially viable PRO processes [11, 12].

Performance of PROs can be severely affected by the membrane properties, and most problem are related to concentrations polarisation (CP). Concentration polarisation happens when the salt concentration difference across the active layer is different from the concentration difference of the bulk solutions itself [13]. Within CP, it can be further categorised into external concentration polarisation (ECP) and internal concentration polarisation (ICP). ECP occurs when a concentration layer forms at the surface of the membrane, whereas ICP forms concentration layer in the porous support layer of the membrane [14].

In PRO process, FO membranes are generally used. Commercially available FO membranes are usually asymmetric, so the membrane can be focused either in the FO mode where the active layer (AL) faces feed side (FS) (AL-FS) whereas in the pressure retarded osmosis (PRO) mode in which the AL faces draw side (DS) (AL-DS) [15]. The characteristics between the draw and feed solution are the essential to maximise PRO performance.

Today, the most widely studied salinity gradient combination is the mixture of seawater with river water that can be applied in the estuaries in coastal regions [8, 12]. For example, brines from desalination plants, which have high salinity value is more suitable for PRO applications. Furthermore, systematic studies in the operating conditions such as cross-flow rate and low salt rejection on the effect on PRO performance are still limited [16]. These factors are not only affecting the water flux but also directly related to the energy density.

This review aims to delineate the recent development in membrane used in a PRO system. Different fabrication and modification techniques are highlighted to illustrate how to fabricate a high performance thin film composite (TFC) membranes and suggest modification to obtain high performance. Finally, future development needs and perspectives are also addressed.

2.0 MEMBRANE TECHNOLOGY ADVANCES IN PRO

In recent years, with the significant progress in membrane technology and increased interest towards renewable energy alternatives. PRO specific membranes have been developed extensively, and among all, thin film composite have been most widely adapted in different studies [17-20]. TFC membranes typically consist two layers, a porous layer that provides mechanical support and a high permeability active layer that demonstrate its selectivity. Interfacial polymerization is the common practice in the forming of active layers on the substrate layer. The polyamide selective layer is commonly formed on the surface of the support substrate layer through interfacial polymerization between MPD and TMC [21, 22]. The following section addresses the latest advances in two main configurations of PRO membranes, namely, TFC flat sheet and TFC hollow fiber membranes.

2.1 TFC Flat Sheet Membranes

Wei *et al.* [23] investigated TFC membrane with reinforced support layer. High molecular weight polyvinylpyrrolidone (PVP) was added into the polysulfone substrate (PSf) layer. It was reported that PVP additive could alter the membrane morphology and pore structure by transforming the dense substrate layer into a sponge like porous structure, as PSf and PVP in the dope exceeded their overlap concentrations leading to the formation of intertwined coils which could improve the water permeate flux.

As a result, a longer phase demixing stage is achieved with PVP additives and this will suppress the formation of macro-void and forms a more porous membrane [24]. This novel TFC membrane can generate 12.9 W/m^2 using 22.0 bar of hydraulic pressure and 1.0 M NaCl brine as draw solution. Although increasing porosity could promote water permeate flux, thus increasing power density produced. However, if the free volume increment was too big, the selectivity of the membrane will be decrease due to the ICP and reverse solute flux effect [25, 26].

Therefore, while improving the structural stability and porosity of the support layer, the selectivity of the active layer should not be impeded. In what follows, a research by Tian *et al.* [27] demonstrated a reinforced selective layer can be achieved by fabricating a TFC membrane that was supported by a tiered structure polyetherimide nanofiber that was strengthened by a multiwalled carbon nanotube. It improves the membrane's ability to withstand higher applied pressure due to increased mechanical stability. Furthermore, its selective layer is an ultrathin (221 – 447 nm) polyamide layer that could minimise ICP effect and enhance permeate flux.

Conjointly, it could produce a peak power density of 17.3 W/m^2 under 16.9 bar pressure while using 1.0 M NaCl synthetic seawater brine as draw solution and deionized (DI) water as feed solution. Noteworthy, a feed spacer was included to facilitate the feed water flow. This is beneficial for water permeate performance but at the expense of additional pressure loss, thus reducing power density produced. Moreover, it could cause adverse impact on the membrane's geometry under higher applied pressure [28].

In 2020, a high performance TFC membrane exhibited both the advantages of a reinforced support layer and high selectivity active layer was presented by Kwon *et al.* [29] The polyethylene (PE) support layer was coated using polyvinyl alcohol (PVA) to form an ultrathin (~8 μm) support while maintaining a highly porous structure. With the reported low structural parameter of approximately 235 μm, it marked an increase in the porosity and at the same time, it do not increase the ICP and reverse solute flux. Besides, toluene was added in the interfacial polymerisation process when forming the selective layer. The role of toluene is to act as an organic solvent to assist in forming high permeable polyamide (PA) layer [30].

The resulting PA selective layer developed through the toluene-assisted interfacial polymerization (TIP) process has a relatively higher water permeability of 8.78 Lm^{-2}h^{-1}bar^{-1}, when compared to a commercially available membrane from HTI that produces 0.56 -1.40 Lm^{-2}h^{-1}bar^{-1}. Furthermore, the PAPE-TFC membrane attained a power density of 35.7 W/m^2 at 20 bar of applied hydraulic pressure while deionized water and 1.0 M NaCl solution were used as feed and draw solutions respectively. This result is marginally higher than other reported results reviewed in this paper and it also shows long term stability under high applied pressure when tested over a period of 12 hours. Therefore, the reported PAPE-TFC membrane has high commercialization potential for PRO usage.

Table 1 displays the features of several notable development of PRO flat sheet membranes. It can be seen in Table 1 that, majority of the studies adopted polyamide as the selective layer due to its flexible operating conditions and good binding ability with different substrate layer [31]. Recent development uses toluene, plasticiser and hydrogel to assist the interfacial polymerisation process [29, 31]. The main differences among studies lies in the choice of substrate layer, different materials have been used in fabricating the support layer and the main objectives is to allow high water permeability while maintaining a rather low structural parameter, to limit ICP and reverse solute flux effect in decreasing power density [29]. On the other hand, certain studies placed a woven mesh under the TFC membrane to accelerate permeates flow. However, it will lead to higher structural parameter and increase ICP. It explains why some studies got rid of the aid from mesh support and focuses on increasing the permeability of the TFC membrane solely. Lastly, although higher salinity gradient across feed and draw solution will increase power density, still most studies focuses to using the combination of 1.0 M NaCl (synthetic seawater) and DI water, as to justify the usage of potential implementation of PRO system in estuaries.

Table 1 Comparison of flat sheet membranes development for PRO.

No	Material	Power density (W/m^2)	Operating pressure (bar)	Draw solution	Feed solution	J_w (Lm^{-2} h^{-1})	Salt rejection, %	S (µm)	Ref
1	Polyamide-polysulfone	12.9	22	1.0 M NaCl	DI water	34.9	>90	-	[23]
2	Polyamide-polyetherimide nanofiber	17.3	16.9	1.0 M NaCl	DI water	43.9	96.1 ±1.3	474	[27]
3	Modified Polyamide-sodium dodecyl sulfate added polyimide	18.1	22.0	1.0 M NaCl	DI water	60.9	90.6 ± 0.012	-	[32]
4	Polyamide-polyacrylonitrile nanofiber	21.3	15.2	1.06 M NaCl	0.9 mM NaCl (Synthetic river water)	62.3	-	150	[33]
5	Polyamide – polyarylene ether sulfone with chlorine modification	26.6	21	1.0 M NaCl	DI water	56.7	93.1 ± 1.1	188	[34]
6	Toluene assisted polyamide – Polyvinyl alcohol coated polyethylene	35.7	20	1.0 M NaCl	DI water	175.6	85.7 ± 0.4	235	[29]

2.2 TFC Hollow Fiber Membranes

Since early 2012, the research direction for PRO membranes has been slowly diverted in considering hollow fiber configuration due to their high packing density, low footprint, and ease of scale up [35]. The development of hollow fiber membrane is aimed to control deformation problem caused by feed spacers [28]. In a general setup, hollow fiber membrane is self-supported. Therefore, unlike flat sheet membrane, it requires a higher mechanical stability that could withstand the applied pressure.

In the earlier development, Chou *et al.* [36] fabricated a notable high strength TFC membrane with high water permeability, excellent salt rejection and low structural parameter to maximise performance. It is reported that it could generate 20.9 W/m^2 of power density at 15 bar pressure, using 1.0 M NaCl draw solution (synthetic seawater) and 1.0 mM NaCl feed solution (synthetic river water). The significant difference between the flat sheet and hollow fiber membrane reported by the same researcher is its much lower reverse salt flux (0.03 mol/L) in the latter form. This is contributed by eliminating deformation problem commonly occur in the spacer of flat sheet membrane, as the membrane is self-supported. On the other hand, the water permeability and structural parameter of TFC hollow fiber is seen to be more sensitive towards pressure change reported by other literatures [35, 37]. The water trend fluctuation in this research however is relatively stable as compared to other flat sheet membranes reported [21, 22, 27, 29, 34]. If it is controlled under a desire range, the variations in pressure could actually bring positive feedback to water permeability and structural parameter.

Moving forward, one approach conducted by Li *et al.* [38] to mitigate membrane fouling was by modifying the outer surface of the inner selective layer. Polyallylamine hydrochloride (PAH) and polyacrylic acid (PAA) were deposited on the TFC polyetherimide (PEI) hollow fiber membrane. It is reported that the polyelectrolyte modification reduced pores exposed and could prevent entrance and adsorption of the negatively charged foulants onto the membrane. The study proved that the modified membrane is effective against organic foulants such as alginate and bovine serum albumin (BSA). Alginate is a naturally occurring anionic polymer commonly obtained from seaweed, while BSA is a type of animal protein. The constant power density produced is at 16.2 W/m^2 under 15 bar pressure, using 1.0 M NaCl as draw solution and DI water as feed solution. Having good resistivity against these foulants could show higher potential implementing the membrane in marine environment.

Lately, next-generation technology such as thermal rearrangement (TR) have been deployed in fabricating TFC hollow fiber member known as TR-TFC membrane. Kim *et al.* [19] presented a highly porous substrate that is thermally rearranged and subsequently, applied a polydopamine (PDA) coating to enhance hydrophilicity, mechanical stability and increase thermal, dimensional and chemical stabilities [39]. Lastly, it combined with a polyamide active layer. The end result is a support that is both thermally and chemically stable. On top of that, the membrane obtained 39.5 W/m^2 of power density at 18 bar. Although a 3.0 M NaCl concentrated brine draw solution and 1.0 M NaCl feed solution were used, it promotes the membrane performance under harsh environment. The characteristics of high mechanical and chemical stability and workability with ultra-saline brine suggested that the membrane could be utilised in a closed loop PRO system, where ultra-saline water from reverse osmosis (RO) desalination plant was used as draw solution and a heated cycle of draw solution was recirculated in the system for minimal waste discharge and water usage [40].

The operating parameters effect on the power density generated using TFC hollow fiber has been studied and summarised in Table 2. Firstly, the option of selective layer still predominantly chosen polyamide similar to TFC flat sheet membrane. However, more complicated fabricating technique can be witnessed in TFC hollow fiber, such as electrospinning, non-solvent phase separation, and dry-jet web spinning [20, 30, 34]. As a result, the choice of substrate layer across all studies reviewed appeared to be more limited, focused mainly on Polyethersulfone (PES) or polyetherimide (PEI). Yet, different processes can be done to improve the performance of the substrate. For example, inorganic additives for better mechanical stability, thermal rearrangement to alter structural morphology and PAH and PAA deposition for polyelectrolyte modification [19, 20, 38]. On a side note, since most study untended on the variation of active layer, the reaction time and concentration of monomers including MPD and TMC used in forming the polyamide layer can be looked into for a better performing active layer [41].

It should be highlighted that ICP, reverse salt diffusion and fouling are still the main obstacles for both flat sheet and hollow fibre TFC membranes, which hinder their osmotic energy harvesting efficiency. In flat sheet membrane, woven support can be removed to reduce ICP, while hollow fiber could improve the morphology of substrate by producing a thinner support. Moreover, reverse salt diffusion can be overcome, if a balance between the pore size and selectivity of the active layer is achieved. That means, the pore size should be small enough to prevent solute back flowing and large enough to not compromise water permeate flux. In addition, an approach to minimise the effect of fouling on water flux through the membranes are fabrication of membrane that has better tolerance towards inorganic scaling and organic fouling similar to the approach by Han *et al.* [18].

Table 2 Comparison of hollow fiber membranes development for PRO

No	**Material (active layer – substrate layer)**	**Power density (W/m^2)**	**Operating pressure (bar)**	**Draw solution**	**Feed solution**	**J_w (10^{-12} m/s Pa)**	**Salt rejection (%)**	**S (mm)**	**Ref**
1	Polyamide - polyethersulfone (PES)	10.6	22	1.0 M NaCl	40 mM NaCl (wastewater brine)	9.22	-	0.46	[35]
2	Polyamide - polyetherimide (PEI)	20.9	16.9	1.0 M NaCl	1 mM NaCl (synthetic river water)	4.22 ± 0.8	>95	0.61 ± 0.03	[36]
3	Polyamide - modified polyethersulfone (PES)	24.3	20.0	1.0 M NaCl	DI water	3.3	-	0.45	[26]
4	Polyamide - PAH and PAA deposited polyetherimide (PEI)	16.2	15.0	1.0 M NaCl	DI water	2.00 ± 0.28	97.5 ± 1.2	0.62	[38]
5	Polyamide – thermally rearranged nanofibrous polymer coated with PDA	39.5	18.0	3.0 M NaCl	1.0 M NaCl	3.7 ± 0.15	94.0 ± 1.63	0.15	[19]
6	Polyamide – polyethersulfone (PES) with inorganic salt additives	38.0	30	1.2 M NaCl	DI water	3.8 ± 0.20	97.3 ± 1.4	0.43 ± 0.011	[20]

3.0 CONCLUSION & RECOMMENDATION

3.1 Conclusion

Albeit osmotic power generation by pressure retarded osmosis (PRO) from salinity gradient is a promising idea, it has to first address some key issues such as concentration polarisation, reverse solute flux, fouling and mechanical stability. The practicality of the PRO membrane relies on its power density produced and long term stability under heavy usage. The ideal membrane should comprise of high water permeability, high selectivity and low structural parameter. Therefore, most studies optimise the substrate properties such as hydrophilicity, structural stability and porosity. Hence, it could effectively reduce ICP and membrane fouling [17-20, 38]. Method ranging from inorganic additives for better mechanical stability, thermal rearrangement to alter structural morphology and PAH and PAA deposition for polyelectrolyte modification [19, 20, 38] were deployed. Besides, to improve the membrane permeability, selectivity, and morphology, modification in the interfacial polymerization process of the active layer were investigated using toluene, plasticiser, and hydrogel to assist the process [29, 31]. Furthermore, the insertion of nanoparticles into the selective layer may induce nano-channels within the nanocomposite membrane, leading to improved permeability and rejection separation efficiency.

3.2 Recommendation

PRO system shows critical reliance on the selection of feed and draw solutions, and the feasibility of the practical operation of PRO can be strengthened by a larger salinity gradient. Therefore, for actual applications, hypersaline draw solutions from desalination plant and feed waters from waste stream may increase the chances of PRO to be implemented.

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