J. Applied Membrane Science & Technology, Vol. 9, No. 1, April 2025, 73-97

A Review on Membrane Fabrication: Structure, Properties and Performance Relationship

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Submitted: 6/1/2025. Revised edition: 25/2/2025. Accepted: 26/2/2025. Available online: 27/3/2025

ABSTRACT

Polymeric membranes have been widely explored and applied in both academic and industrial sectors. Their high separation efficiency, versatility, and scalability make them an alternative to conventional separation processes. This review presents recent developments in flat-sheet polymeric membranes for water treatment applications, focusing on fabrication methods, membrane formulation parameters, and structure-property-performance relationships. Common fabrication methods, including phase inversion, interfacial polymerization, stretching, track-etching, and electrospinning, are examined in terms of fabrication mechanisms, operating conditions and membrane structures. While these methods may utilize similar base polymers, they result in distinct membrane structures that are customized to specific applications. In addition, the influence of formulation parameters, such as polymer and solvent selection, polymer concentration, and additives used, are thoroughly discussed to obtain desired physicochemical properties for specific membrane applications. Furthermore, the membrane structural properties, including crystallinity, pore morphology, and surface characteristics, like hydrophilicity, surface charge, and roughness, are discussed to better understand their impact on membrane permeability and solute selectivity. Advanced characterization techniques for analysing these properties are also explored. Lastly, this review explores future directions for polymeric membrane technology in respect to the materials used, post-treatment of used membrane, the integration with artificial intelligence technologies, and assessment on industrial scalability of modified membrane. By integrating recent advancements, addressing existing challenges, and identifying future opportunities, this review provides a foundation for advancing polymeric membrane technologies and promoting innovation in water treatment solutions.

Keywords: Polymeric membranes, membrane fabrication, pore structure, physicochemical property, water treatment

1.0 INTRODUCTION

Membranes are synonymous with processes like separation, purification, refining, and concentration, finding extensive applications in industries [1, 2], ranging from water and wastewater treatment to biotechnology,

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DOI: https://doi.org/10.11113/jamst.v29n1.311

pharmaceuticals, and food and beverage production. Membrane technology is a well-established yet continually evolving field that aims to conserve water and valuable products through simple, effective methods. One of the key advantages of membrane separation is its versatility and scalability [3]. Membranes can be applied at various scales, from laboratory settings to industrial processes, serving diverse purposes across numerous applications. Moreover, membranes can be tailored to exhibit specific structures and properties through fabrication techniques or post-modification [4]. A variety of chemicals are available for membrane fabrication, with recent trends emphasize the use of green materials, which are safer to handle, biodegradable, and environmentally friendly.

The selection of a membrane process and its corresponding rejection mechanism is primarily determined by the driving force and the characteristics of the solute to be separated [5]. For example, pressure-driven processes such as microfiltration (MF) and ultrafiltration (UF) rely on size exclusion. MF is commonly used to remove macro-sized molecules (>0.1 µm), such as bacteria or suspended solids [6], while UF is suitable for smaller molecules (0.01-0.1 µm), such as proteins and viruses [7]. Nanofiltration (NF) combines size exclusion with Donnan exclusion, making it suitable for even smaller solutes (1-10 nm), including multivalent ions, dyes and fertilizers [8]. Reverse osmosis (RO) employs a solution-diffusion mechanism and is ideal for rejection nearly all solutes, including monovalent ions (<1 nm) [9].

In contrast, osmotically-driven process, such as forward osmosis (FO), utilizes both size exclusion and the solution-diffusion mechanism for solute rejection [10]. Electrically-driven processes like electrodialysis (ED) rely on Donnan exclusion to selectively remove charged ions. Thermally-driven processes, including membrane distillation (MD) remove non-volatile compounds via a vapor-liquid phase change [11], while pervaporation (PV) separates volatile organic compounds from aqueous solutions through selective permeability [12]. Each of these processes is tailored to specific separation needs, emphasizing the versatility of membrane technology.

Generally, membranes can be categorized into [13]: organic, inorganic, and organic-inorganic membranes. Each type features distinct structures and properties that are suitable for different industrial applications. Organic membranes are typically made up of cellulose or synthetic polymers and can be further classified into single-layer structure or thin-film composite structure. Inorganic membranes are generally ceramic and can be subdivided into metal oxide-based and carbon-based membranes. Organic-inorganic membranes combine polymeric materials with inorganic fillers such as metal oxides or carbon-based materials to achieve enhanced performance.

Among them, polymeric membranes are known for their ease of fabrication and operation, scalability, and cost-effectiveness [14]. These membranes are widely available across a wide range of pore sizes, from MF to RO. Also, they are tunable for desired physicochemical properties. However, polymeric membranes are susceptible to organic solvents, extreme pH levels, and high temperatures [13], which can reduce their lifespan. Despite this, research on polymeric membranes technology is still progressing due to their high separation efficiency, versatility and practicality.

This review article explores common fabrication methods for polymeric membranes used in water treatment, including phase inversion, interfacial polymerization, stretching, electrospinning, and track-etching. It further examines the impact of membrane formulation parameters, including concentration of polymer, as well as choice of polymer, solvent, and additives on membrane separation performance. Lastly, the article discusses the structure-property-performance relationship and associated characterization methods for each physicochemical properties.

2.0 MEMBRANE FABRICATION TECHNIQUES

Depend on the polymer types and membrane application, polymeric membrane can be fabricated using various methods. This section provides an overview of membrane fabrication methods, including phase inversion, interfacial polymerization, stretching, track etching, and electrospinning, in terms of the underlying mechanism, polymers used, advantages and limitations. Table 1 summarizes the fabrication techniques for polymeric membrane used in water treatment.

2.1 Phase Inversion

Phase inversion is a demixing process that transforms a homogeneous polymer solution from a liquid to a solid state in a controlled manner [15]. This process is influenced by the thermodynamic properties and viscoelastic behavior of the casting solution, as well as the solvent-nonsolvent exchange kinetics during membrane casting [16]. Phase inversion happens when there is an exchange between solvent and nonsolvent. Non-solvent induced phase separation (NIPS) and evaporation-induced phase separation (EIPS) are popular methods in commercial polymer membrane manufacturing due to their simplicity, scalability, and low cost [15].

The NIPS, also known as immersion precipitation, uses a ternary composition of polymer, solvent, and nonsolvent.

Table 1 Summary of various fabrication technique for polymeric membranes

Fabrication Technique	Common Membranes	Characteristics	Applications	Pros	Cons
Phase Inversion (NIPS & EIPS)	PS, PES, PVDF, PMMA, PA	High porosity, strong mechanical properties	UF, MF, NF	Simple, scalable, low cost	High water usage, toxic wastewater generation
Interfacial Polymerization	TFC, PA	Thin selective layer, high rejection rates	NF, RO	High flux, uniform polymerization	High solvent use, non-reusable waste
Stretching	PTFE, PP, Polyethylene	Large pores, precise thickness control	MD	Good mechanical properties, scalable	High energy consumption
Electrospinning	PSF, PES, Chitosan, PANi	High porosity, ultrafine fibers	Wastewater treatment, filtration	Low cost, versatile, nanoscale precision	Low mechanical strength, high voltage use
Track Etching	PSF, PTFE, PC	Precise pore size control, high stability	Diagnostics, filtration	High precision, scalable	Complex, costly, long processing time

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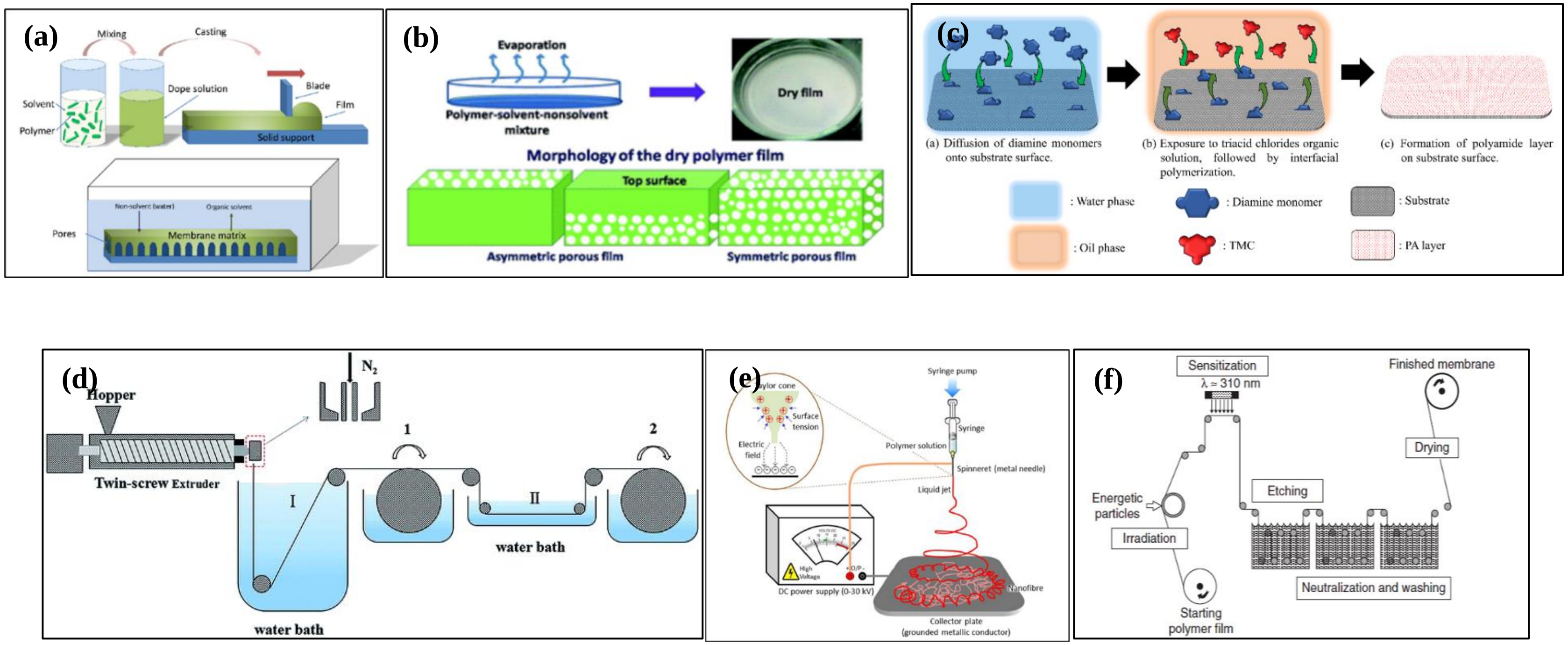


Figure 1 Illustration of various membrane fabrication techniques (a) Non-solvent induced phase separation (NIPS), illustrating the solvent exchange process leading to membrane pore formation [17]; (b) Evaporation-induced phase separation (EIPS), showing the production of asymmetric and symmetric porous membranes [18]; (c) Interfacial polymerization, demonstrating the formation of PA layers on substrates [10]; (d) Stretching process, highlighting the extrusion and pore creation in polymer membranes [19]; (e) Electrospinning setup, showcasing the production of nanofibrous membranes through an electrostatic force [20]; and (f) Track etching process, outlining the ion irradiation and etching to create precision pores in polymeric films [21]

As illustrated in Figure 1(a), membrane formation occurs as the solvent exchanges with the nonsolvent, causing polymer precipitation. This technique is commonly employed for the fabrication of polymeric membranes using polymers such as polysulfone (PSF), polyethersulfone (PES), and poly(vinylidene fluoride) (PVDF), dissolved in solvents such as N,N-Dimethylacetamide (DMAc), N,N-Dimethylformamide (DMF), and Dimethylsulfoxide (DMSO). Water is the most commonly used nonsolvent for the demixing process. NIPS membranes offer high tensile strength, porosity, and hydrophilicity, with small-pore membranes demonstrating high thermal shock resistance and improved power density [22]. However, limitations include high water consumption in coagulation baths and the generation of toxic wastewater from solvent release [23].

Similar to the NIPS method, the EIPS method also involves a ternary composition of polymer, solvent, and nonsolvent, but the solvent has higher volatility than the nonsolvent. As demonstrated in Figure 1(b), during membrane fabrication process, the solvent and nonsolvent evaporate in the air, leaving a thin polymeric film. Liquid-liquid phase separation occurs through the controlled evaporation of the solvent and nonsolvent (regulated by temperature and relative humidity) from a homogeneous single-phase polymer solution [18]. Common polymers used for this method include poly(methyl methacrylate) (PMMA) and polyamide (PA) in solvents such as tetrahydrofuran (THF) and dichloromethane (DCM), with water as the non-solvent. EIPS provides better morphology control with a simpler process [24]. Despite these advantages, EIPS faces challenges similar to NIPS, including high water usage and toxic wastewater generation [23]. EIPS also faces difficulty finding suitable solvents and non-solvents [18].

2.2 Interfacial Polymerization

The interfacial polymerization technique, shown in Figure 1(c), is widely used to fabricate the selective layer in thin-film composite (TFC) membranes. This selective layer is typically made up of polyamide and is formed on top of a porous UF or MF substrate. The fabrication of this thin selective layer involves polymerizing reactants in immiscible organic (oil) phase and aqueous phase to create a cross-linked membrane structure. The process begins with the impregnation of an amine monomer (e.g., m-phenylenediamine (MPD) or piperazine (PIP)) into the substrate, followed by exposure to an organic solution containing triacid chloride (e.g., trimesoyl chloride (TMC)) [10]. The interfacial polymerization reaction occurs at the liquid-liquid interface between the aqueous and oil phases. Rapid diffusion of diamine monomers into the oil phase reacts with the triacid chloride, forming a thin PA layer anchored on the substrate’s surface.

This technique produces membranes that are thermally and chemically stable [25] and exhibit improved characteristics, including enhanced performance, ultra-high permeance, and high rejection rates, along with uniform polymerization. However, interfacial polymerization has certain drawbacks. A significant amount of organic solvents and monomers is required for the formation of PA selective layer, yet these chemicals are non-reusable without effective recovery through post-treatment [23].

2.3 Stretching

Stretching, shown in Figure 1(d), is a solvent-free technique used for fabricating highly porous membranes from homopolymers. The process involves heating the polymer to its melting point, extruding the polymer solution into thin sheets, and stretching these sheets under controlled cold and hot conditions to form pores [26].

Initially, stretching is performed under cold conditions to form nucleate structures, followed by hot stretching to refine the final pore structure [26]. Commonly used polymers for this technique include polytetrafluoroethylene (PTFE), polypropylene (PP), and high-density polyethylene (HDPE). Each polymer is extruded into sheet at specific temperature range: PTFE is generally stretched between 100-250 °C [27], PP between of 130-145 °C [26], HDPE between 95-125 °C [28]. During the stretching process, the polymer chains align, thereby enhancing membrane properties.

The stretching process can be performed uniaxially or biaxially [29]. Figure 2 shows the schematic diagram of different stretching stages.

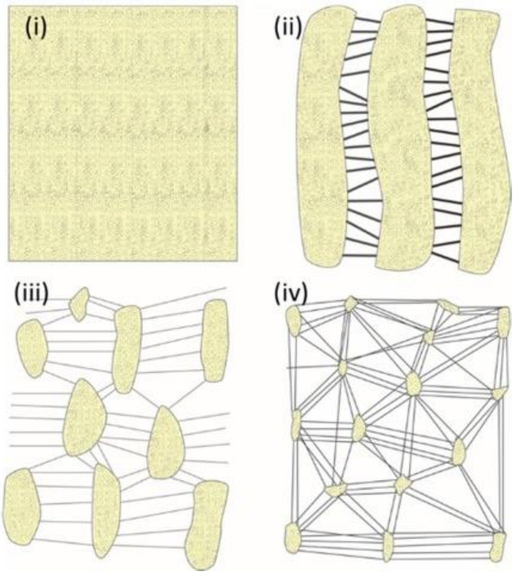


Figure 2 Schematic of PTFE stretching process: (i) raw PTFE sheet, (ii) early-stage stretching process, (iii) uniaxial tension process, and (iv) biaxial tension process [29]

Uniaxial stretching applies tension in one direction, producing elongated pores and anisotropic properties. In contrast, biaxial stretching exerts tension in two perpendicular directions, leading to a more uniform pore distribution and isotropic membrane characteristics. Stretching offers several advantages, including excellent mechanical properties, precise control over membrane thickness, and high porosity. Additionally, this method enables large-scale production with high productivity, making it suitable for applications like uranium extraction from seawater [30]. However, stretching is highly energy-intensive due to the need for both high and low operational temperatures [23].

2.4 Electrospinning

Electrospinning, shown in Figure 1(e), is a technique for fabricating nanofibrous membranes with high porosity and a large specific surface area [20]. This method commonly utilizes polymers such as PSF, PES, chitosan, and polyaniline (PANi). By applying electrostatic forces, polymeric films are generated, resulting in membranes with fine pore structures and an enhanced surface area.

Electrospinning is a highly versatile technique used in various fields, including membrane separation, Li-ion batteries, and tissue engineering, due to its ability to precisely control pore formation [31]. his cost-effective approach offers excellent thermal stability and functionality, making it suitable for applications such as filtration, distillation, and drug delivery [32]. However, electrospun membranes often suffer from low mechanical strength due to their high porosity. Additionally, this method requires high voltage supply, relies on a strong electric field, and has a relatively low production throughput [23].

2.5 Track Etching

Unlike other fabrication methods which fabricate membrane from raw chemicals, track etching method turns a nonporous film into a porous membrane. As illustrated in Figure 1(f), this process involves irradiating nonporous polymeric membranes, such as PSF, PTFE, or polycarbonate (PC), with energetic heavy ions like uranium-234 and iodine-127. This irradiation breaks polymer chains, creating weak spots that serve as the basis for pore formation. Two irradiation methods are commonly used: (1) fragment irradiation via fissioning heavy metal nuclei like californium or uranium [33]; and (2) ion beam acceleration [34]. The membrane's porosity is influenced by irradiation time and temperature, while pore size depends on etching conditions, such as time, temperature, chemical factors, and ion selectivity [35].

Track etching offers precise control over pore size distribution and density [34]. This method also provides benefits like good stability, high porosity with minimal overlapping, and stable particle flux [36]. Additionally, track etching enables easy functionalization and scalability from single to multiple pores. Despite these advantages, the technique is technologically complex and costly [36]. Certain polymers, such as PVDF, exhibit strong oxidation resistance, prolonging the pore formation process [23].

3.0 INFLUENCE OF VARIOUS PARAMETERS ON MEMBRANE SEPARATION PERFORMANCE

Previous section discusses on difference fabrication technique for membrane fabrication. For each technique, difference materials are involved and required to form membrane with desired property and performance. The performance of membrane separation systems is heavily influenced by fabrication parameters, which determine the structural and chemical properties of the membranes. This section outlines the key parameters, such as the choice of solvent, polymer type, polymer concentration, and additives, which directly affecting the critical factors like water permeability, solute rejection, and fouling resistance during membrane separation processes.

3.1 Choice of Solvent

The solvent used in polymeric membrane fabrication is critical in determining polymer dissolution, phase separation kinetics, and final membrane morphology. These properties indirectly influence key performance metrics such as hydrophilicity, fouling resistance, and thermal stability, which collectively govern separation efficiency. The selection of a solvent is influenced by the fabrication technique, membrane type, and desired physicochemical properties.

NIPS method commonly utilizes solvents such as NMP, DMAc, and DMSO for polymer dissolution. Highly miscible solvents (i.e., NMP) promote rapid phase separation, resulting in membranes with larger pores and higher porosity. This structure enhances membrane permeability but compromises solute rejection [37]. Conversely, less miscible solvents (i.e., DMAc and DMSO) delayed the solvent-nonsolvent demixing process, resulting in membranes with denser pores that offer lower permeability but improved solute rejection [38]. EIPS method requires more volatile solvents, such as acetone and THF to facilitate the demixing process. Acetone's high volatility promotes the formation of compact membranes with better selectivity but reduced permeability [39]. In contrast, slightly less volatile solvents (i.e., THF) forms porous structures, which could improve membrane permeability [40].

In interfacial polymerization, organic solvent, such as n-hexane, is commonly used for dissolution of acid chloride [10]. Bio-based solvents, such as cyclopentyl methyl ether (CPME) and 2-methyl-tetrahydrofuran (2-MeTHF), have been proposed as an alternative to n-hexane [41]. Similar to n-hexane, these solvent exhibit low solubility in water, which is ideal for the synthesis of PA selective layer [41]. CPME gives a high solute rejection for fully aromatic PA membranes, with a slight reduction in permeance [41]. In contrast, 2-MeTHF gives a high solute rejection for semi-aromatic PA membrane, together with an increase in permeance [41]. On the other hand, stretching does not use any solvent [37]. The properties and performance of the membrane depends on the material's physical properties used in the fabrication [37]. In electrospinning, polar solvents such as DMF promote finer fibers [42], producing hydrophilic membranes with better permeability and fouling resistance. Conversely, non-polar solvents like chloroform yield thicker fibers with improved solute rejection but reduced permeability and fouling resistance [43].

The selection of solvents across these techniques ultimately determines the structural and functional attributes of the membranes, emphasizing the critical role of solvent choice in achieving desired separation performance.

3.2 Choice of Polymer

Similar to solvents, polymers significantly impact the membrane's structure and, consequently, its performance. Polymers serve as the foundation of membrane material, offering unique physical, chemical, and thermal properties that must align with the application requirements. Additionally, the polymer's glass transition temperature (Tg) is a critical factor influencing processability and operational stability [44,45]. A higher Tg generally enhances structural stability, while a lower Tg improves flexibility and gas transport.

PSF and PES are widely used polymers for their ability to form homogeneous solutions, enabling phase separation (i.e., NIPS and EIPS) and producing membranes with porous sublayers that support efficient water flow and solute rejection [46]. PVDF offers superior chemical resistance and dense pore structures, enhancing selectivity but requiring hydrophilization or grafting to mitigate fouling [47]. Polymer like polyacrylonitrile (PAN) is also commonly used in phase inversion, particularly in EIPS, due to their solubility in polar solvents such as NMP, DMF, and DMAc, enabling the formation of dense membranes [38].

PA is extensively employed in TFC membranes for applications demanding high solute rejection and water permeability [10]. This polymer can withstand high pressure, temperature, and biological attacks, operate effectively over a wide range of pH, and demonstrate excellent mechanical strength [48]. However, the application of PA-TFC membranes is constrained by the hydrophobic nature of PA skin layer, which result in membrane fouling and diminished separation efficiency[49] .

PE and PP are unsuitable for TIPS due to limited solubility in common solvents but are effectively used in stretching techniques [24,26]. PP membranes provide excellent mechanical strength and durability [26], while PE membranes balance permeability and solute rejection, with hydrophilic modifications enhancing antifouling performance [24].

PC and polyethylene terephthalate (PET) are commonly processed by track-etching method due to their stability towards acids, organic solvents, and mechanical properties [37]. PC membranes offer consistent solute rejection and good thermal stability [37], while PET membranes provide hydrophilicity, fouling resistance, and high thermal stability, making them suitable for high-temperature applications [50].

PAN and polystyrene (PS) are often processed through electrospinning method. PAN membranes exhibit intrinsic hydrophilic properties due to its polar nitrile groups, good thermal stability, solvent resistance, and high mechanical strength [51]. In contrast, PS membranes features thicker fibers with better solute rejection and thermal stability but susceptible to fouling [52].

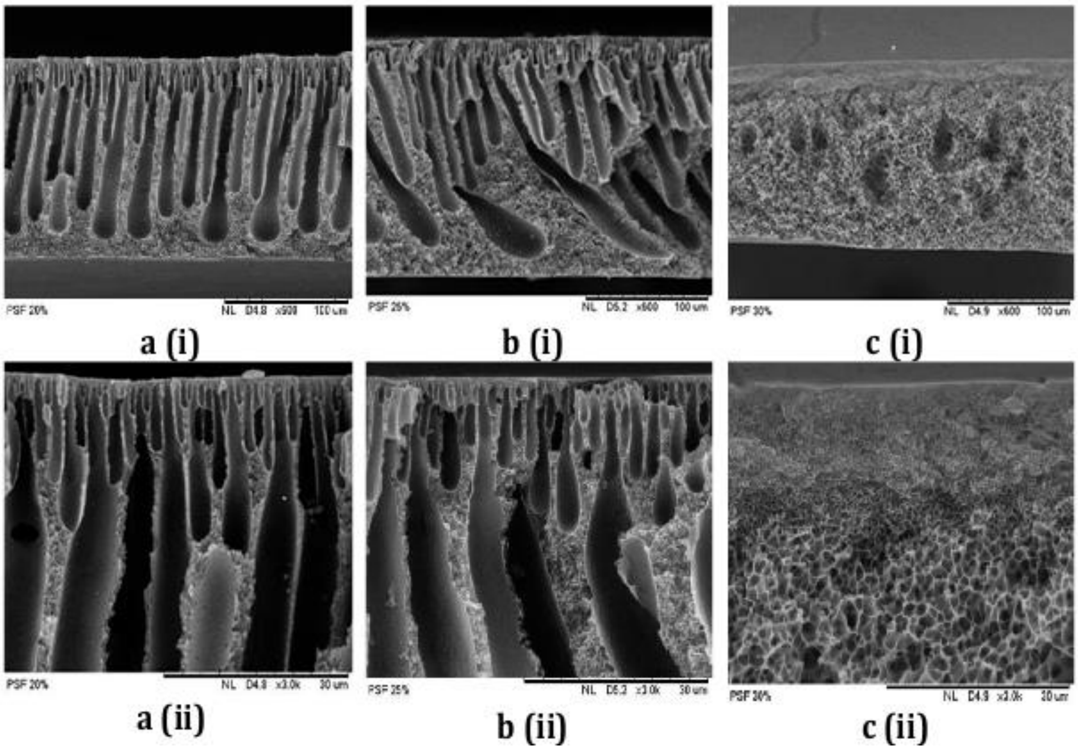
The choice of polymer is crucial to obtain a balance between permeability, selectivity, and fouling resistance used for specific applications.

3.3 Polymer Concentration

Polymer concentration is another formulation parameter that influences the membrane structure and performance. As illustrated in Figure 3, a higher concentration of polymer usually leads to a denser membrane structure, characterized by smaller pore sizes and reduced porosity [37,38]. This is attributed to the increased in number of polymer molecules in the solution, which raises its viscosity and reduces the solvent diffusion rate during the phase inversion process [53]. This increased viscosity restricts the movement of polymer chains, reducing their capacity to rearrange. As the solvent diffuses, the denser presence of polymer chains increases their likelihood of interacting and entangling, resulting in a tightly packed structure with smaller spaces between chains and, consequently, smaller membrane pores [54,55]. In contrast, low polymer concentrations exhibit a lower viscosity, allowing greater chain mobility during membrane formation. Subsequently, the faster solvent-nonsolvent demixing process results in larger pore sizes.

This dense structure enhances the membrane’s selectivity by effectively rejecting solute or contaminants [56]. However, the increased in selectivity often compromises membrane permeability, as the dense polymer structure hinders the water diffusion across the membrane [56]. Conversely, membranes fabricated with lower polymer concentrations exhibit a more porous structure with larger pore sizes. This large pore structure limits the solute rejection and reduce the membrane’s selectivity, allowing more solutes or contaminants to pass through [56]. Additionally, a higher concentration of polymers can boost the membrane's tensile strength, which in turn can boost its durability [57].

The polymer concentration significantly influences the membrane structure and separation efficiency. It is important to select optimal polymer/solvent ratio to develop membrane with high water permeability and solute rejection.



a (i) b (i) c (i)

a (ii) b (ii) c (ii)

Figure 3 Cross-sectional images of PSF membranes prepared at polymer concentrations of (a) 20 wt%, (b) 25 wt%, and (c) 30 wt%. Images were captured at magnifications of (i) 500× and (ii) 3,000× [58]

3.4 Additive to the Membrane Matrix

Additives are commonly added to the membrane to enhance the physicochemical properties and performance during separation process. The incorporation of additives results in enhanced structural, chemical and functional properties of the membrane. Subsequently, these modifications improve membrane performance, in terms of water permeability, selectivity, and fouling resistance.

Several types of additives have been explored to modify membrane properties. For example, pore-forming agents are added into the polymer dope solution to enhance the pore structure of membrane matrix. Pore-forming agents, such as LiCl and poly(ethylene glycol) (PEG), are commonly used to create more porous membranes [37], leading to improved water permeability. These agents alter the thermodynamic stability of the polymer solution, promoting rapid solvent-nonsolvent demixing. During membrane casting, the polymer solution contacts with the non-solvent and the pore-forming agents diffuse from the polymer matrix along with the solvent, leaving behind voids that develop into pores. In addition, the molecular weight and concentration of pore-forming agents can influence the membrane morphology and pore structure [37]. Higher concentrations or molecular weights can further destabilize the polymer solution, leading to accelerated demixing and the formation of larger pores and increased porosity. While these modifications improve water flux and permeability, they can also reduce solute rejection, which is the common permeability-selectivity trade-off in membrane separation [46].

Hydrophilic polymers are another type of additives that are commonly incorporated into membrane matrix to enhance the hydrophilicity and antifouling properties. Common hydrophilic polymers, such as polyvinylpyrrolidone (PVP), PEG, polyvinyl alcohol (PVA), and poly(acrylamide) (PAAm), are widely employed to enhance membrane hydrophilicity [10]. These additives introduce polar functional groups, such as hydroxyl, carboxyl, or amide groups to the membrane, enhancing the water affinity. Subsequently, these functional groups enhance hydrogen bonding interactions with water molecules, making the membrane more hydrophilic. In addition, novel copolymer, such as polysulfide-amide (PSA) [59] and carboxylic-acid containing polyamide (CPA) [60] had been explored to further boost this property when incorporated in the MF and UF membranes.

Functional additives are frequently incorporated into membrane matrices to enhance the antifouling resistance, thereby enhancing both performance and lifespan. Fouling is generally categorized into organic fouling, biofouling, and scaling, depending on the nature of feed solutes [61]. To mitigate these challenges, various antifoulant agents are employed. For example, titanium oxide and silver nanoparticles are known for their antibacterial properties, which can significantly reduce biofouling [62]. Graphene oxide and cellulose nanocrystal possess numerous oxygen-containing functional groups, which enhance surface hydrophilic and promote the formation of a protective hydration layer that helps prevent organic fouling [63,64]. In addition, to mitigate inorganic scaling caused by gypsum and silica, poly(acrylic acid) (PAA) deprotonates to form negatively charged carboxyl groups that chelate calcium ions, while polyethylenimine (PEI) protonates to create a positively charged surface that repels scale-forming species [65].

The presence of additives within the membrane matrix has a substantial impact on the membrane's performance. By carefully selecting and optimizing additives, membrane properties such as permeability, selectivity, and hydrophilicity can be adjusted to improve enhanced efficiency and durability of the membrane. However, the choice of additives should be aligned with the specific needs of the application to guarantee an effective and well-balanced separation performance.

4.0 STRUCTURE-PROPERTY-PERFORMANCE RELATIONSHIP

Previous section discussed fabrication methods and manipulation of dope solution composition, in which both play important roles in affect the structure, properties and performance of the membrane. This section further explains the influence of the membrane structure and property affecting the membrane performance. Most important parameters, such as polymer crystallinity, pore structure, hydrophilicity/hydrophobicity, surface charge and surface roughness are discussed below.

4.1 Crystallinity of the Polymer

Crystallinity refers to the degree of structural order in a polymer, significantly influencing the mechanical strength, thermal stability, water permeability, and selectivity of membranes [66]. Two phases can be found in a polymer matrix: crystalline and amorphous. Crystalline areas are made up of densely packed, organized polymer chains, which contribute to mechanical strength and chemical resistance while restricting molecular mobility [67]. These regions are essential in membranes subjected to high pressures or harsh solvents. In contrast, amorphous regions feature randomly arranged chains, providing flexibility and increased free volume, facilitating small molecule transport. However, these regions are more prone to fouling and chemical attack, compromising structural integrity.

In addition, high crystalline membranes exhibit superior thermal resistance and chemical stability due to the strong intermolecular bonds in ordered regions [67]. Membranes with high crystallinity can withstand elevated temperatures and resist oxidizing chemicals, making them ideal for industrial applications. Despite this, reduced free volume in crystalline regions limits water permeability, yet making the membranes selective to solute separation by creating size-exclusion barriers. Thus, by balancing crystalline and amorphous phases, transport properties and fouling resistance could be optimized. Crystalline regions provide mechanical strength, while amorphous regions enhance permeability [68].

Various methods have been explored to manipulate polymer crystallinity, such as thermal annealing, additive incorporation, and controlled solvent evaporation. Thermal annealing via heat treatment post fabrication, reorganizes polymer chains into more ordered structure, increasing crystallinity and mechanical strength [69]. Additive incorporation, such as nanoparticles (e.g., TiO_2, ZrO_2, graphene oxide), promotes nucleation during polymer solidification, balancing membrane permeability and selectivity [70]. Controlled solvent evaporation during membrane casting, manipulating the crystalline and amorphous region proportions with a slower evaporation rate promotes crystalline phases [68].

Various characterization techniques can be used to assess the membrane crystallinity. For example, X-ray diffraction (XRD) is used for phase distinction between crystalline and amorphous. Differential scanning calorimetry (DSC) allows estimation of crystallinity by comparing enthalpy values. Fourier transform infrared spectroscopy (FTIR) identifies specific crystalline bonds and evaluates crystallinity based on characteristic adsorption bands. Atomic Force Microscopy (AFM) provides nanoscale imaging to assess the distribution of crystalline regions.

4.2 Pore Structure

Pore structure refers to the arrangement and characteristics of the pores within a membrane, including size, shape, distribution, tortuosity and porosity within a polymeric membrane. These attributes dictate the membrane permeability, mechanical stability, and the overall selectivity in transporting ions, molecules, or particles [34]. Fine-tuning these pore structures enhances membrane performance for specific applications. For example, MF membranes, with pore sizes ranging from 0.1 to 10 μm, are designed for separating larger particles and microorganisms. UF membranes, with pore sizes between 2 and 100 nm, are suitable for retaining macromolecules like proteins and colloids. NF membranes feature smaller pore sizes, typically 1 to 2 nm, enabling the rejection of multivalent ions and small organic molecules. RO membranes are characterized by non-porous or extremely small pore structures (<1 nm), making them ideal for desalination by rejecting nearly all solutes.

Pore size corresponds to the diameter of individual pores, while pore size distribution describes the range or frequency of pore sizes in a membrane. These parameters directly influence selectivity and permeability. Porosity, also known as the ratio of void volume to total volume, also determines water flux and solute discrimination (selectivity) [71]. Tortuosity represents the complexity of pore pathways, also known as pore connectivity, and affects fluid flow resistance and overall permeability [72]. Optimized pore structures enhance selectivity, permeability, and fouling resistance. Membranes with smaller, uniformly distributed pores achieve higher solute rejection but lower flux, while membranes with larger pores and lower tortuosity promote water diffusion but may suffer reduced selectivity. This phenomenon is termed the permeability-selectivity trade-off, commonly present in membrane separation [73].

Different techniques are employed to assess pore structure. Gas adsorption and porosimetry are commonly employed for pore characterization. Gas adsorption, particularly the Brunauer-Emmett-Teller (BET) method, uses nitrogen adsorption to determine the specific surface area, pore volume and pore size distribution of a membrane. Porosimetry involves forcing mercury into the pores under pressure to analyze the pore size distribution. Porosity can be determined using gravimetric method or imbibition technique. The gravimetric method measures the weight difference between dry and solvent-saturated membranes, while the imbibition method estimates porosity based on the volume of liquid absorbed. Tortuosity is assessed using mathematical models based on porosity and diffusion coefficients. The tortuosity can also be evaluated using imaging techniques like X-ray computed tomography for 3D pore network reconstruction [74].

4.3 Surface Properties

Surface properties of a membrane play a critical role in determining overall performance. These include hydrophilic-hydrophobic properties, surface charge, and surface roughness, all of which significantly impact a membrane's interaction with solutes, solvents, and foulants.

4.3.1 Hydrophilic-hydrophobic Properties of Membrane Surface

Hydrophilic-hydrophobic properties refer to the membrane's affinity for water [75]. A hydrophilic surface attracts water molecules due to the presence of polar functional groups such as hydroxyl and carboxyl. These groups interact with water molecules via hydrogen bonding. Hydrophilic membranes tend to form a hydration layer on membrane surface, repelling the hydrophobic foulants and thus enhance the fouling resistance in water and wastewater treatment. Conversely, hydrophobic surfaces repel water but may attract organic matter, leading to fouling in aqueous applications. These membranes repel water through non-polar functional groups like $-CH_3$ and $-CF_2$. While excelling in resisting water adsorption, hydrophobic membranes are prone to fouling from hydrophobic compounds. Hydrophobic membranes are widely used in the MD due to their ability to resist wetting under high-temperature conditions, enabling vapor-only transport [68].

Hydrophilicity can be enhanced through plasma treatment or by introducing hydrophilic additives. Plasma treatment involves introducing hydrophilic functional groups onto a hydrophobic polymer membrane to improve wettability [76]. Alternatively, hydrophilic additives, such as PVA, PEG, and graphene oxides, introduced via blending or surface coating, improve water affinity and mitigate organic fouling [77].

Surface hydrophilicity is typically characterized by using static water contact angle (WCA) measurement, dynamic contact angle measurement, and surface energy analysis. WCA measures the angle between a water droplet and the membrane surface where a lower angle correlates higher hydrophilicity. The dynamic measurement is conducted if there is need of hydration behavior assessment, especially for membrane with superhydrophilic (WCA $< 10°$) and superwetting (WCA $\approx 0°$) properties [78]. Surface energy analysis estimates polar and dispersive components as insights into hydrophobic interactions, van der Waals forces and acid-base interactions.

4.3.2 Surface Charge

Surface charge is determined by the presence of ionizable functional groups, such as carboxyl or amine groups, on a membrane's surface [79]. These functional groups possess electrostatic interactions with charged solutes and foulants, significantly influencing separation and fouling behavior [80]. Positively charged membranes attract anions, which can lead to the deposition of negatively charged foulants such as sulfates and nitrates. In contrast, negatively charged membranes repel anions through electrostatic forces, thereby reducing scaling from compounds like calcium carbonate [81].

Surface charge can be manipulated using methods like chemical grafting or surface coating. The chemical grafting introduces additional functional groups (e.g., polyethyleneimine and trimethylammonium chloride for positive charge, sulfonic acid and carboxylic acid for negative charge) attached to the membrane surface to tailor electrostatic properties [82]. On the other hand, surface coating could be done via layer-by-layer assembly, alternating layers of oppositely charged polymers on to the membrane surface to enhance overall surface charge [76]. Notably, zwitterionic coatings are particularly effective as these materials combine both positive and negative charges while maintaining ionic selectivity [83]. Characterization of surface charge involves zeta potential analysis and electrokinetic analysis. Zeta potential analysis dictates electrostatic potential at solid-liquid interface under various pH conditions [80]. Electrokinetic analysis evaluates charge density of membrane surface for fouling and ion selectivity understanding [84]

4.3.3 Surface Roughness

Surface roughness refers to the micro- and nanoscale irregularities on the membrane surface, which affect membrane wettability and fouling behavior [85]. A rough surface can increase the deposition of foulants but also increases surface area, promoting better flux [86]. Smooth surfaces minimize foulant attachment and are ideal for high-purity applications, while controlled roughness can enhance permeability by reducing resistance to water flow and allowing certain components to pass.

Several post-modification methods have been explored to manipulate membrane surface roughness. This includes surface coating, etching, grafting, and plasma treatment. Surface coating smoothens or roughens the surface by adding layers of materials like polymers or nanoparticles [87].

Etching removes material to create controlled grooves [88], while grafting covalently attaches polymers to alter roughness and introduce functional groups [89]. Plasma treatment modifies roughness and surface chemistry using high-energy ions, enabling functionalization and wettability improvements [90]. Surface roughness is usually measured using techniques like AFM and SEM. AFM provides nanoscale imaging to analyze crystalline region distribution while also offering quantitative surface roughness measurements through topographical mapping. In contrast, SEM is primarily used to visualize surface morphology for qualitative assessments.

All of the stated surface properties interact synergistically in influencing membrane performance. This is true as by balancing the hydrophilicity and hydrophobicity of a membrane which will create a hybrid surface, a membrane can have the highest possible flux and still retain considerable fouling resistance [85]. Different charged surfaces have different electrostatic interactions with the solutes which changes the selectivity. The smoothness of a membrane dictates the tendency of organic fouling, which is very important in wastewater treatment.

5.0 CONCLUSION

Polymeric membranes remain a preferred choice in the field of separation due to their effectiveness in water and resource recovery, scalability, versatility, and cost-efficiency. This review provides an overview of various fabrication techniques, including phase inversion, interfacial polymerization, stretching, electrospinning, and track-etching, detailing their underlying mechanism, associated materials polymers, as well as respective advantages and limitations. Also, key formulation parameters, such as solvent types, polymer type and concentration as well as additive incorporation, were discussed in relation to their impact on the membrane characteristics and performance. Additionally, the structure-property-performance relationship is examined, focusing on factors like polymer crystallinity, pore structure, and surface properties, to highlight their influence on the membrane performance.

To advance polymeric membrane technology, several areas for improvement are suggested:

1. Develop and utilize environmentally friendly polymers and solvents while addressing their limitations in chemical, mechanical, and thermal stability.
2. Explore methods to reuse or repurpose used polymeric membranes, such as recycling them as raw materials for new membrane fabrication.
3. Employ AI technologies to optimize membrane fabrication processes and enhance performance predictability.
4. Investigate the scalability of membrane fabrication and post-surface modification techniques, ensuring their techno-economic viability for industrial-scale production.

In conclusion, this review underscores the importance of membrane designs to meet the evolving demands of industrial and environmental applications. Polymeric membranes continue to lead in membrane technology, presenting substantial opportunities for future innovations and applications.

ACKNOWLEDGEMENT

The authors gratefully acknowledge the funding for this work by Skim Geran Penyelidikan Fundamental (FRGS/1/2023/TK05/UKM/02/7) and Geran Universiti Penyelidikan (GUP-2023-032).

CONFLICTS OF INTEREST

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper.

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