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The Future Membrane Technology: Perspectives of Advanced Membrane Applications

Anurag Prakash Sunda*

Department of Chemistry, J. C. Bose University of Science and Technology, YMCA, Faridabad 121006, Haryana, India

Abstract: Polymer materials are well recognised in the synthesis and fabrication of model membranes for water purification, liquid/gas separation processes, and for potential use in energy devices as polymer electrolytes. The recent trends in the technological development of these materials for various applications are highlighted through a broad classification into crystalline membranes, hollow-fiber membranes, hybrid/mixed membranes and molecularly imprinted membranes. The perspective aims to present some of the key developments related to crystalline and porous hybrid membrane materials. The future focus of membrane technology is to achieve cost-effective model membrane materials with high durability, selective adaptability, modularity and biodegradability. The critical part of membranes in fabrication and large-scale processing is their biocompatibility/toxicity. Thus, the insertion of biodegradable features along with the use of green solvents have become the prime necessity for industrial applications. Recent progress in screening of polymer materials through artificial intelligence may certainly provide deeper insights towards the design and development of novel membranes.
Keywords: *Reverse osmosis, Nanofiltration, Desalination, Water remediation, Machine learning, Mixed membrane.*

1. Introduction

Liquid separation research has made tremendous development in the past few decades to treat organic & non-organic substances and aqueous media. Interfacial interactions in liquid separation spurred immense interest to develop several classes of materials including polymer-based membranes, covalent organic frameworks (Yuan et al., 2019; Zhang et al., 2021), metal-organic frameworks based mixed membranes, polymer composites and nano-cellulose (Das, Lindström, Sharma, Chi, & Hsiao, 2022) materials etc. Apart from desalination and water purification, the liquid separation technology through membranes got significant attention towards food processing including dairy products, the paper & pulp industry, petrochemical refinements, and the beverage industry etc. (See Figure 1) The desirable chemical and physical separation of liquids through the membrane leads to the development of classified membranes in microfiltration, ultra-filtration and nano-filtration (Galizia & Bye, 2018; M. Li, Zhang, Zhang, Guo, & Liang, 2023) based technologies. The durability and sustainability of membranes for liquid separation are further governed by selectivity (Sadeghi, Kaner, & Asatekin, 2018; Zhao et al., 2021; Epsztein, 2022), flux-rate, fouling, fabrication strategies and morphological traits (W. Ma et al., 2023) etc. The substitution of existing membrane materials to address the unwanted accumulation of these materials and their hazardous impact on nature, the design and development of biomimetic or biodegradable membranes are of great demand (X.-C. Chen, Zhang, Liu, Zhou, & Jiang, 2022).

*Corresponding author
Email address: anurag.sunda@gmail.com

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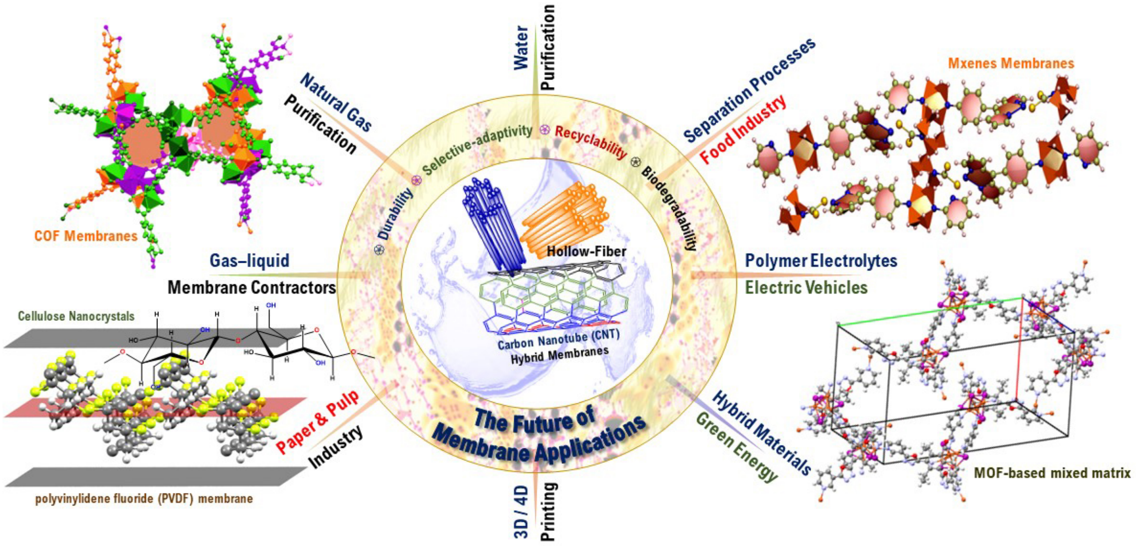

Figure 1: Schematic representation of the various membrane applications and hybrid membrane materials.

In the past few decades, the growth of advanced membrane materials could lead to a well-defined class of membranes. Some examples of specific advancement in membrane materials and their respective application based performances are highlighted in Table 1. In brief, the advancement in various membrane materials is discussed in next sub-sections:

1.1. Crystalline membranes

Well-defined and uniform porous membrane structures are achieved in crystalline membranes based on Metal-Organic Frameworks (MOF), Covalent Organic Frameworks (COFs), Aluminosilicate Frameworks (Zeolites) and Hydrogen-bonded Organic Frameworks (HOFs) (Kang et al., 2021) etc. MOFs are a combination of metal clusters with an organic linker and there are tremendous opportunities to fine-tune these material's properties based on desired applications. Furthermore, the role of ligands with groups such as aldehyde, amine, pyridine, pyrene etc. and reaction conditions are critical in COFs to network covalent bonds with desired functionalities. (Kirlikovali, Hanna, Son, & Farha, 2023; Mohan et al., 2023) Traditional approaches like solution casting, phase inversion etc. are replaced by interfacial polymerization and solvent evaporation based solution casting to produces homogeneous membranes with controlled crystalline pores. In recent years, the weak hydrogen bond interactions in organic moieties led to self-assembled porous materials with a unique 3D framework of HOFs. These attracted the membrane science community with regeneration capability, reversibility and self-healing characteristics. (C. Chen et al., 2024) For e.g. the 1,3,5-Tris (4-carboxyphenyl)benzene (BTB) ligand give rise to a 3D framework of hydrogen bonds via carboxyl groups (Yin et al., 2024). The BTB-based HOF membrane offered high crystalline features and membrane porosity with cost-effective purification. The scalability and durability of thin crystalline membranes with controlled inter-crystal defects are among the key challenges to be addressed. The separation industry is mainly targeting new innovative economical strategies for membrane processing with minimal use of solvent. Kujawa et al. presented various combination of COF materials and highlighted use of crystalline frameworks to enhance performance of membranes towards liquid separation (Kujawa et al., 2021).

Table 1: Recent advancement in membrane materials and their specific performance for various applications.

Membrane	**Material**	**Application**	**Specific Performance/ Characteristics**	**References**
COF-LZU-1	TFB with PDA	Water purification and Dye Separation	Water permeance (ca. 760 $L\,m^{-2}\,h^{-1}\,Mpa^{-1}$)	(H. Fan, Gu, Meng, Knebel, & Caro, 2018)
COF-LZU-1	TFB with TPE	Water purification and Dye Separation	Highly-open structure consisting of a void volume of 80%	(Liang et al., 2020)
COF-OEt	DFPBA with DETH	Molecular-sieving membrane	High water permeability (up to 41.7 $L\,m^{-2}$ h^{-1} bar^{-1})	(Han et al., 2022)
Imine-linked crystalline COF	TAPT and DHTA	Proton transport	Conductivity up to 0.53 $S\,cm^{-1}$ at 80 °C	(C. Fan et al., 2021)
p-toluene sulfonic acid loaded COF membranes	DMBBPyDAA	Proton transport	Conductivity up to 7.8×10^{-2} $S\,cm^{-1}$	(Sasmal et al., 2018)
Smectic Liquid Crystalline Polymer	Monoacrylate benzoic acid, smectic diacrylate (II) cross-linker and microporous polypropylene	Water purification and Dye Separation	High rejection of anionic solutes bigger than 322 g/mol	(Houben et al., 2021)
Hollow fiber-based NF	PAH/ PSS	Water Treatment	Removal of organic compounds/particles/ions in a single-step process	(Virga, de Grooth, Žvab, & de Vos, 2019)
ZAC-X	AMA and SBMA	Sulfate removal and water treatment	>99.2% SO_4^{2-} rejection	(Lounder & Asatekin, 2021)
GO based membranes	GO nanosheet with PET	Actinides/ lanthanides separation	Lanthanides permeation >90% concur with below 10% actinides	(Z. Wang et al., 2023)

The zeolite-based membranes specifically Linde Type A (LTA) have shown high selectivity in pervaporation. Still, the focus needs to further reduce the cost of the membranes (Charik et al., 2022) and to produce defect-free membranes (Wenten, Dharmawijaya, Aryanti, Mukti, & Khoiruddin, 2017). LTA-zeolite consist of a general formula $[Na_{12}(Al_{12}Si_{12}O_{48}) \cdot 27\,H_2O]$ with an alternate AlO_4 and SiO4 tetrahedra structure. (Reed & Breck, 1956) The uncontrolled flux through defects is one of the major challenges in LTA-zeolite crystalline membranes to address the shape selectivity disruption at higher temperatures and long-run application.

1.2. Hollow-Fiber membranes

The state-of-the-art separation technique immersed as a sustainable alternative is through hollow fibre (HF) membranes (K. Y. Wang, Weber, & Chung, 2022). The popularity of HF membrane is due to very high separation performance and use of cost-effective polymeric materials such as polysulfone(PSF), polyethersulfone(PES), polyimide, and polybenzimidazole(PBI) etc. (Lau & Yong, 2021; Lau et al., 2022; Wojciechowski, Wasyłeczko, Lewińska, & Chwojnowski, 2024) The permeability of HF membrane is largely governed by the aqueous phase inversion. The role of pH is equally critical and Baig et al. (Baig, Pejman, Willott, Tiraferri, & de Vos, 2022) showed variation in concentration of acetate buffer results in variation in permeability based on poly(sodium 4-styrene sulfonate) (PSS) and polyethyleneimine (PEI) HF membrane. Cellulose derivatives such as cellulose triacetate, cellulose diacetate and cellulose acetate propionate-based HF membranes demonstrated excellent rejection rate with high water permeation. (Takao et al., 2022)

The field of HF membrane have grown significantly due to their high chemical stability and less pretreatment management in commercial nano-filtration applications. Various commercial HF modules have been developed and found successful for liquid separations due to their low fouling sensitivity and wide range of pressure applications (Jonkers, Cornelissen, & de Vos, 2023). HF applications for drinking water are now becoming favorite for the treatment of industrial and textile waste water. The major concern of researcher towards the use of HF nano-filtration is its limitation associate with membrane charge at various pH and charges on ions in the medium. The rejection rate and permeate through HF is greatly influence by membrane charge and charges of ions in the feed. Recently, the polyelectrolyte multilayers based method showed a great success for the HF membranes for the treatment of charged surfactant in the waste water. For example, the cross-linking by glutaraldehyde in PAH/poly(sodium 4-styrenesulfonate) (PSS) offers high selectivity and stability in separation performance (Virga et al., 2019).

1.3. Hybrid Polymer Composites and Polymer Mixed Membranes

The membrane separation technology has grown significantly over the years being cost-effective and energy-efficient for wide applicability in water purification and desalination process and other food/chemical process industries (Divakar, Padaki, & Balakrishna, 2022). Advances in hybrid polymer composite materials research using Carbon Nanotubes (CNTs), Graphene, and MOFs, COFs (Han et al., 2022) lead to access to smart materials for nano-filtration techniques and treatment of toxic or rare-earth metals. Various synthesis strategies are adopted for the successful application of polymer mixed hybrids such as physical blending of polymer, hot-pressing coating over polymer, In-situ interweaving using polyvinylidene fluoride (PVDF) as a binder and post-synthetic photoinduced polymerization (For example UIO-66-NH_2/poly(butyl methacrylate) (Sunda & Yadav, 2021). Recently, a successful attempt has been made to mimic the bionic temperature control of nanofluidic separation. COF-based ionic membrane for separation nanofluidic system could show potential based high thermo-sensation towards the wide range of salt concentration (Zhang et al., 2021). Composite membranes based on 3D-printing are found quite helpful for significant reduction of the fouling and more insights are required for industrial applications (Al-Shimmery, Mazinani, Ji, Chew, & Mattia, 2019). Recent attempts of zwitterionic copolymers through cross linking approach provide improved fouling resistance behaviour of the membranes (Lounder & Asatekin, 2021) and more comprehensive research is required in this field.

To ensure more precise permeance, the two-dimensional nano-sheet or lamellar membranes based on graphene oxides (GO) offers structure tunability through functional groups. The class of these membranes is well recognized as MXene-based membrane (J. Li, Li, & Van der Bruggen, 2020; Huang, Ding, & Wang, 2021). The flux and selectively of MXene are greatly influenced by the electrostatic interactions with solvent and are found to be highly specific based on hydrophilic (-NH_2, -OH, -COOR) and hydrophobic (-Ph) attached to GO. The fascinating structure and superior performance of MXenes are limited by the matrix compatibility and stability in organic solvents for longer cycles. The recent study on the nuclear waste treatment using MXene is important to note for understanding the complexity of interlayer nanochannel spacing in MXenes and their electrostatic effects (Z. Wang et al., 2023). Atomistic studies (Lin & Grossman, 2015) at the molecular level using molecular dynamics are required to fully unveil the selective recognition of Mxene based on the choice of functional groups and compatibility.

1.4. Molecular Imprinted Membranes

A molecular imprinting tool in membranes was introduced in the early 1960s for target-specific separations. The affinity of the targeted molecule allows the membrane to restrict its movement and allows the remaining to pass through the membrane. The membrane surface binding affinity could be tuned with the help of chitosan, amine-grafted nanofibers, electrospun polyacrylonitrile (PAN) nanofiber and chelating ligands (Yoshikawa, Tharpa, & Dima, 2016). Apart from large-scale realization, the pre-knowledge of effective molecular imprinting materials - target molecule pair is yet to be established. Machine learning could be used to provide a pre-defined library for the best pair to set imprinting targets (Ratnaningsih et al., 2022). Moreover, the molecular imprinting ratio and site recognition localization/distribution need to be rationalised for well defined industrial processes of separation specifically for proteins and micro-organisms.

2. Artificial Intelligence (AI) in Screening of Polymer Membrane

Several decades of research in synthesizing novel polymer membranes liquid separation lead to a deeper understating of various factors associated with selectivity and sustainability. The electrostatic and pore characteristics along with nano-channels brought new dimensions in membrane separation through hybrid materials. Thus, the traditional Edisonian approach is not good enough to solve the toxicity and accumulation of membrane materials in large-scale processing (J. Yang, Tao, He, McCutcheon, & Li, 2023). Recent advances through machine learning (ML) enable researchers to search for an optimum combina-

Table 2: Recent progress in ML approach towards design and development of efficient materials for membrane applications.

Sr. No.	ML Approach	Applications	References
1	An artificial neural network with skip connections and selectivity	Mono/divalent ion-selectivity and permeability	(Deng et al., 2023)
2.	A bottom-up strategy for rational membrane design	Organic solvent nano-filtration	(M. Wang, Shi, Zhao, Liu, & Jiang, 2023)
3.	Bayesian optimization	Salt salt rejection and water permeability	(Gao et al., 2022)
4.	CatBoost/ XGBoost/ fingerprint ML algorithms	Waste-water treatment through UF membranes	(Gao, Zhong, Dangayach, & Chen, 2023)
5.	ML-based gaussian process regression (GPR) models	Separation of oily Waste-water	(Usman et al., 2024)

tion of polymer and functional groups to achieve precise selectivity and operations (Xu & Jiang, 2022). Some examples of ML approach towards design and development of efficient materials for membrane applications are provided in Table 2. For prediction of high flux with perm-selectivity, machine learning algorithms are developed to screen existing polymer materials based on solute-solvent interactions, total flux, separation factor and parity parameters (M. Wang, Xu, Tang, & Jiang, 2022). The accuracy of high separation prediction through ML largely relies on the available data set, choice of parity parameters and AI algorithms. The over-prediction of polymer screening needs to be examined through cross-validation (Maleki et al., 2022). Moreover, the availability of atomistic studies limits for hybrid/mixed membranes in separation compared to the field of polymer electrolyte limits the ML utility despite of the availability of high-performance computing.

3. Toxicity and bio-biodegradability of Polymer Membrane

Polymer membrane fabrication largely depends on organic solvents for dissolution processes as well as for membrane material purification. Solvents such as chloroform, dimethylacetamide, dimethylformamide, N-methyl-2-pyrrolidone, dimethyl sulfoxide, and tetrahydrofuran are used extensively in membrane casting and membrane material synthesis. Handling and recycling of these solvents are critical for both economical aspects of membranes as well as environmental toxicity. The use of green solvents (such as ionic liquids, and deep eutectic solvents, (Silvianti et al., 2023) Supercritical CO_2 (Nalawade, Picchioni, & Janssen, 2006) etc.) could make significant progress in the past few years. The use of microwave-assisted and Ultra-Violate assisted fabrication also reduces the polymer-solvent ratio for fabrication (Zou, Nunes, Vankelecom, Figoli, & Lee, 2021).

The major environmental issue is an accumulation of polymer membrane waste due to extensive use in daily life and the short span of membrane life. To overcome this non-biodegradability of polymer membranes, researchers are emphasising on natural bio-degradable polymers or synthetic bio-degradable polymers (Bandehali, Sanaeepur, Ebadi Amooghin, Shirazian, & Ramakrishna, 2021; Samir, Ashour, Hakim, & Bassyouni, 2022). For the natural bio-degradable polymers, bio-polymers such as polysaccharides (cellulose, lignocellulose, chitin), polypeptides (collagen, casein, gluten), poly-hydroxy-butyrate, and cross-linked lipids are under investigation. Synthetic biodegradable polymers such as polylactide (Vatanpour et al., 2022), polyglycolide, poly-butylene succinate, and poly-caprolactone are among the possible alternates to derive hybrid membranes.

4. Conclusion

The idea of future membrane science preferentially begins with the innovative manufacturing process of hybrid polymer membranes with the help of green solvents (Nunes et al., 2020). The future demand and commercial utilization of polymer membranes widen the scope and opportunities to develop novel membrane materials, fabrication methods and degradation versatility towards energy-efficient liquid separation technologies.

In summary, membrane separation technologies have made significant achievements with the help of NF membranes and hollow fibers to fulfill the pure water demands. The membrane fabrication strategies are well established for mixed membranes, MXenes, and porous crystalline membranes consisting of COFs /MOFs /Zeolites with specific selectivity and permeability. Apart from biodegradability and resolving toxicity of polymer membrane materials, it is going to become crucial to provide a realistic solution for the upcoming challenge of separation of nuclear waste and separation of radioactive nuclides (Kim, Kim, Hyeon, Kang, & Lee, 2020). For the next generation of membranes, it is important to get molecular insights to unveil interfacial characteristics of the membranes in various separation processes and to develop new innovative methods using the green approach for polymer membranes with the help of AI/ML.

Abbreviation

1,3,5-triformylbenzene (TFB); phenylenediamine (PDA); 1,3,5-triformylbenzene (TFB); phenylenediamine (PDA); tetraphenylethylene (TPE); COF-bearing ethyoxyl group (COF-OEt); 3,5-diformylphenylboronic acid (DFPBA); 2,5-diethoxyterephthalohydrazide (DETH); 1,3,5-tris-(4-aminophenyl)triazine (TAPT); 2,5-dihydroxyterephthaldeyde (DHTA); Poly(allylamine hydrochloride) (PAH); poly(sodium 4-styrene sulfonate) (PSS); Zwitterionic copolymers (ZAC-X); Allyl methacrylate (AMA); zwitterionic sulfobetaine methacrylate (SBMA), Graphene Oxide (GO); polyethylene terephthalate (PET); 3,3'-dimethylbenzidine, 2,2'-bipyridine-5,5'-diamine, 4,4'-azodianiline (DMBBPyDAA)

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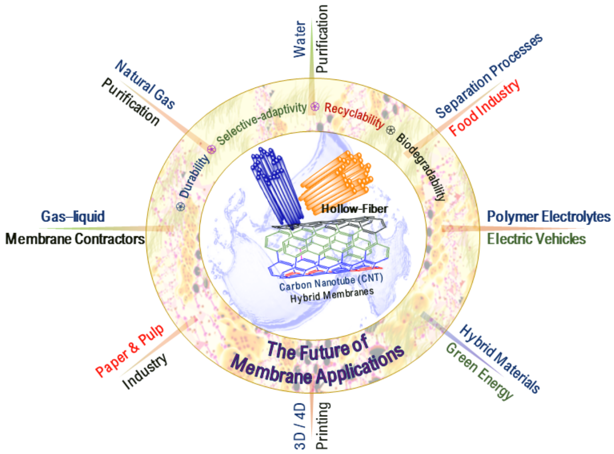
Graphical Abstract



Dr. Anurag Prakash Sunda obtained M. Sc. Chemistry (2008), and M. Phil. Energy (2009) degree from University of Rajasthan, Jaipur and later moved to National Chemical Laboratory (NCL), Pune & worked on PEM Fuel Cell Membrane Casting, Materials Synthesis and Molecular Modeling of PEM Fuel Cells. He did Ph. D. (2010 - 2014) in Chemistry from Indian Institute of Science Education and Research (IISER), Pune and further pursued post-doctoral research under the supervision of Prof. Balasubramanian Sundaram at JNCASR, Bangalore (2014-2015). He received prestigious DST INSPIRE Faculty award in Materials Sciences (2015-2020) and served as Assistant Professor (INSPIRE Faculty) in Central University of Haryana (2015-2017) and Central University of Rajasthan (2017-2019). Prior to joining YMCA, he served as Associate Professor in SBSR at Sharda University. He has been working as Assistant Professor in J. C. Bose University of Science and Technology, YMCA Faridabad since February 2020. His research interest is to investigate proton conducting materials, battery electrolytes, polymer membranes, and their composites for energy and environment applications.

