# **GREEN MEMBRANES FOR SUSTAINABILITY**

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#### Abstract

Membranes are inevitable and effectively eliminate a wide range of hazardous substances, both organic and inorganic compounds, including metals, heavy metals, or other pollutants, and the most efficient tools, particularly for water treatment. In the last decade, membrane technology has played a pivotal role in addressing global sustainability challenges by offering efficient solutions for various applications, i.e. water and wastewater treatment, desalination, ion exchange, dairy industry, gas separation, chemical separations and energy applications, and so on. However, this advanced technology faces challenges, including toxicity and fouling of contaminants, posing safety risks in producing environmentally friendly and sustainable from the points of membrane structure and processes. Factors like sustainability, non-toxicity, performance enhancement, and commercial viability are pivotal concerns in green membrane fabrications. Therefore, it is essential to systematically and comprehensively review critical aspects of toxicity, biosafety, and the mechanistic intricacies of green membrane fabrications for sustainability. This review covers various facets of membrane fabrication to perform sustainability, encompassing their fabrication by using green nanoparticles, bio-sourced materials, green solvents, recycled materials, additive manufacturing, and their future perspectives. The evolution of green membrane fabrication presents an exciting avenue for advancing sustainability efforts, reducing environmental impact, and enhancing performance. Elucidating these aspects aims to underscore the importance and potential of green membranes in achieving sustainable development goals.

#### Keywords

Green membrane fabrication, Water treatment, Sustainability Toxicity and biosafety, Environmental impact

## Introduction

Polymer membranes are ubiquitous, and the polymer membrane market is growing quickly and is estimated to reach US\$ 7,251.4 million in 2023. Over the assessment period from 2023 to 2033, worldwide product demand is projected to exhibit a 4.4% compound annual growth rate (CAGR), resulting in a market size of US\$ 11,153.9 million by the end of 2033 (Polymeric Membrane Market Outlook, 2023 to 2033). Their role in environmental pollution mitigation, water purification, and desalination is expected to grow to meet environmental and climate goals. Polymeric membranes are used in many large-scale and advanced separation processes across water treatment, wastewater treatment, medical (includes separation processes for blood disease treatments and bio-assay), and industrial (gas separation/purification and effluent treatment). There are various types of membranes used in processes such as microfiltration, ultrafiltration, pervaporation and osmosis (used materials such as fluoropolymers, polyarylsulfones, and polyolefins); nanofiltration (NF), reverse osmosis (RO) (materials include cellulose-based and polyamide composites), and iondialysis and filtration membranes (materials exchange: include fluoropolymers, polyaryl sulfones, and nonwovens). They are used in a number of domestic products, from electrical appliances to oil and water filters, as well as HVAC and other air treatments. They may be important in CO<sub>2</sub> capture, charge storage (membranes in electrochemical cells), and new molecular sensors.

Nowadays, most of the polymers used for membrane manufacturing are poly(vinylidene fluoride) (PVDF), polysulfone (PSU), poly-(ether sulfone) (PESU). poly(ethylene terephthalate) (PET), polyamide (PA) and poly(ethylene glycol) (PEG), etc. (Razali et al, 2015). All these polymers used for membrane development are fossil-based, non-biodegradable, and harmful to both humans and the environment. In addition, solvents such as N,N-dimethylacetamide (DMAc), dimethylformamide (DMF), or 1-methyl-2pyrrolidinone (NMP), which are mainly used for membrane fabrications, also highly harmful and toxic to both health and environment concern and they are classified as substances of very high concern (SVHC) (Faggian et al, (2014) by European REACH Regulation (EC, 2006) and all these need to be recycled or reused in circular economy. In other words, many of the polymers used are based on fossil-fuel-derived systems, and many are fluorinated, necessitating the production of 'forever chemicals' and by-products of high global warming potential through manufacture or degradation. In order to eliminate these disadvantages and contribute to the European Green Deal (EC. 2020a) and The 2030 Climate target plan (EC. 2020b), the best alternative solution is to use and develop biodegradable membranes that offer a variety of potential uses, however they have a number of challenges and limitations in terms of biocompatibility, mechanical properties and degradability. It is crucial to thoroughly evaluate the biocompatibility, degradability, and mechanical properties of the membranes, including their degradation behavior and rate, to determine their suitability for specific applications (Ehsani et al., 2022). Degradation rate/time depends on the same factors: type of material, enzyme and microorganism concentration, pH, humidity, oxygen, and the conditions of the surrounding environment (Haider et al, 2019). Long-term biodegradation of the polymer membranes also leads to a high risk for environmental problems due to increased waste disposal. Therefore, a thorough understanding of the mechanisms of membrane degradation needs to be evaluated in terms of the membrane's performance in a particular application.

In recent years, there has been a trend towards using more environmentally friendly materials that reduce the use of single-use polymeric membranes. Therefore, the reusability of these biodegradable membranes is also important to minimize waste disposal. Moreover, examining biodegradable membranes' thermal stability, mechanical stability, and swelling rate is essential. To enhance their performance, nanoparticles and cross-linking agents can be added to the membranes, improving the biodegradation rate and making them non-toxic and biodegradable. These environmentally responsible materials can aid in preserving the sustainability of the membranes and enhance their biodegradation rate in the natural environment.

In our global and future perspective, sustainability is a major challenge for us as well as our future generations, demanding a delicate equilibrium between economic priorities and environmental consciousness. In this context, the new, innovative, and more sustainable industrial technologies, characterized by reduced energy consumption and minimized waste and wastewater generation within a circular economy framework, assume paramount importance (Geissdoerfer M. Et al. 2017). Consequently, membrane technology has gained prominence since its inception in previous decades due to its inherently lower energy demands, superior efficiency, and costeffective points. Membranes have emerged as transformative elements in redefining separation processes, assuming a pivotal role across diverse applications in advanced separation methodologies (Kim and Nunes, 2021; Nunes et al, 2020). Membrane technology applications have also been used in new fields encompassing energy conversion, resource recovery, the production of value-added products, and innovative smart drug delivery systems (Diroli et al., 2021). In all these fields, membrane technology applications have been approved due to their extraordinary properties, including significantly reduced energy consumption, markedly heightened selectivity, diminished carbon footprint, and simplified operational procedures. Despite advancements in membrane technologies over recent decades, there remains a pressing need for further enhancements. This includes requirements for reduced energy consumption, decreased fouling susceptibility, heightened physicochemical durability, enhanced cleanability, superior selectivity, increased permeability, zero waste in manufacturing, cost reduction, and improved membrane module reusability/recyclability (Teodora et al., 2022). Presently, commercial membranes, whether polymeric or inorganic, rely on a limited range of materials that were not originally designed for membrane production for sustainability.

In order to address these challenges, new innovative green membranes are expected to be an alternative solution for conventional membranes for sustainability, produced from renewable materials, including wood-based polymers, or re-use and recycling of biopolymers in terms of flux and antifouling properties. In many cases, biodegradable membranes have shown comparable performance to commercial ones like PES, PVDF, and PTFE. There's a wide scope for creating new eco-friendly membranes and modifying natural or full biomaterials to perform the membrane properties with conventional membranes, expanding their potential uses. However, there are still a number of challenges and limitations with green or biodegradable membranes for application purposes in the market, including their degradation rate and composition with non-biodegradable materials, which can lead to waste disposal and environmental pollution. Further research is needed to understand the degradation mechanisms and how microorganisms and enzymes behave in various environments. Additionally, as green membranes are typically produced on pilot scales, more efficient scale-up models are needed to expand production.

The use of polymer membranes in applications such as  $CO_2$  capture and batteries, as well as healthcare and mitigation, are wholly necessary to meet climate and environmental goals. They will be continually developed in applications such as those detailed above in order to reach net zero 2050 targets. However, their role as largely single-use plastics, their lack of recyclability, their non-biodegradability, and their fossil fuel sources negate their potential benefits, and there is a clear need to develop polymer membranes from materials that themselves have low environmental or climate impact and are based on renewable resources rather than fossil fuels. Therefore, a fully innovative green process desperately needs to be developed for the membrane preparation process, replacing the conventional process, and using green solvents from renewable sources. In addition, used solvents should be efficiently treated or recycled before discharge. In conclusion, all the conventional membrane fabrication processes need to be replaced with the innovative green process, which has to be fully biobased materials for the circular economy.

## **Green Nanoparticles**

Using green chemistry principles in the fabrication of nano-enhanced membranes presents a promising avenue to reduce reliance on petroleumbased, potentially hazardous membranes and their associated costs (Rethinam et al., 2020). Additionally, concerns surrounding the disposal and recyclability of synthetic nano-enhanced membranes, there are still a number of significant challenges for manufacturers (Razmjou et al., 2019a, 2019b; Landaburu-Aguirre, 2016; Lawler, 2012). Recently, increased attention and investment have been directed toward developing green membranes and their components by using different nanomaterials for green membrane fabrication (Wang et al., 2023). Although preparing these green membranes typically involves chemical reactions that may not be entirely sustainable, they demonstrate a superior environmental profile compared to some commercially used chemicals. Despite the rapid advancement in manufacturing green-synthesized nanomaterials and their application in membrane development, a comprehensive review and discussion regarding the mechanisms underlying the properties and performance of green nanomembranes in water treatment are currently lacking.

Nowadays, most green membranes have been prepared using green nanoparticles as mixed matrix membranes (MMM) and thin-film nanocomposite (TFN) membranes. Green nanoparticles have a wide variety of properties, such as size, shape, porosity, and reactivity. In this regard, carefully selecting the source and the green reaction medium is primely important. In recent years, there's been an increasing focus on exploring the synthesis and application of green nanoparticles across various fields, notably in preparing nanocomposite membranes (Hamid et al., 2020 & Nthunya et al., 2019), reported the potential of natural extracts which are incorporation into membrane materials as green nanoparticles.

Green membrane technology encompasses using environmentally friendly nanoparticles and innovative novel fabrication methods in the nanofabrication approach. These nanofabricated membranes possess a high surface-area-to-volume ratio, enhancing their efficiency while reducing the requisite material quantity for specific applications. Notably, nanofiber membranes are designed by using different techniques such as electrospinning, electroblowing, or blown-spinning. These methods significantly curtail the use of polymers and solvents compared to conventional fabrication approaches (Sanaeepur et al., 2022). In the near future, using natural extracts, such as apples or other natural extracts, to synthesize nanoparticles and modify them for various applications might be possible. Researchers have the opportunity to explore how these altered nanoparticles could potentially elevate membrane performance, like boosting salt rejection and bolstering mechanical resilience.

Membrane technology using nanofibers has found extensive applications across multiple sectors encompassing energy, medicine, biology, and environmental domains. Nanofiber-incorporated membranes, known for their high efficacy at low pressure, hold potential in various purification processes such as blood, air, and wastewater purification. Their superior adhesion to biological surfaces makes the membranes particularly suitable for applications in transdermal drug delivery systems (Yoo et al., 2009). Typically exhibiting cross-sectional diameters ranging from 10 to 100 nm, nanofibers boast a significant specific surface area and a high surface area-tovolume ratio. Additionally, their structure allows for creating highly porous fibers with extensive network connectivity.

Furthermore, nanofiber membranes stand out due to their exceptional porosity, high vapour permeability, and interconnected open pore structures, setting them apart from standard membranes. The outstanding qualities of nanofiber membrane-based technology, including flexibility, ease of control and scaling, energy efficiency, and eco-friendliness, have spurred swift advancements in membrane science and technology (Yadav et al., 2021).

Nanofibers fall under two classification criteria: the first pertains to their raw materials—organic, inorganic, carbon, and composite fibres; the second focuses on their structures—nonporous, mesoporous, hollow, and core-shell fibres [Gugulothu et al., 2019). This multifaceted structure grants greater adaptability in tailoring nanofiber properties to meet specific requirements through chemical ingenuity. Traditional methods often necessitate substantial solvent usage, and mismanagement of residual wastes and non-compliance with safety regulations exacerbate environmental concerns. Hence, ensuring the integration of safety measures and green chemistry practices is crucial in mitigating these challenges.

Moreover, in pursuing green strategies for membrane production, several pivotal factors are required to ensure the resulting membranes possess desirable characteristics. These include achieving high membrane packing density and selectivity, effectively managing concentration polarization and membrane fouling, and ensuring efficiency in processing, maintenance, and scalability, all while minimizing expenditure and energy consumption. Accomplishing these objectives without relying on non-green materials and methodologies or generating substantial amounts of toxic and hazardous residual waste represents a significant achievement.

While recognizing the importance of a comprehensive understanding of greener approaches in nanofiber membranes, the current knowledge remains somewhat limited. Hence, this review aims to present a cutting-edge approach encompassing a complete strategy for green nanofiber membranes. It covers aspects ranging from raw materials, solvents, methods, and techniques employed in membrane fabrication to post-treatment processes. Ultimately, this review summarizes key findings and highlights future challenges and prospects in this domain.

Nanofiber membranes have garnered recognition in filtration and separation arenas due to their distinct attributes: adjustable nanoscale diameters and morphology, high porosity, substantial surface area-to-volume ratio, robust internal connectivity, and exceptional mechanical properties. However, prioritizing human health, environmental impact, and expanding applications necessitates developing and integrating a green strategy for nano-enhanced membranes—ones that are safer, non-toxic, sustainable, and eco-friendly. Employing green polymer-based materials, including natural polymers and bio-synthesized and synthesized chemicals, aligns with the ethos of sustainable progress, ensuring eco-friendliness.

## **Bio sourced materials**

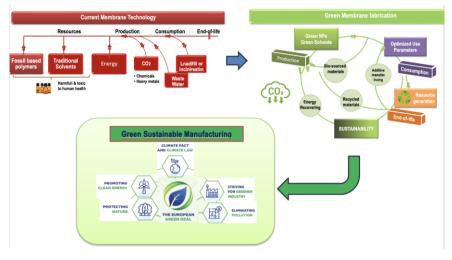
In conventional techniques for membrane production, the primary reliance lies on non-biodegradable polymers and inorganic materials sourced from petroleum-based resources. This association of conventional polymers with membrane fabrication has implications for the global energy crisis. Conversely, green membranes utilize nontoxic, biodegradable polymers, alongside bioinspired or biomimetic materials, offering a more sustainable and environmentally benign alternative to traditional synthetic polymers. The emergence of bioinspired or biomimetic materials, mirroring the structure and functionality of biological membranes, can be crafted from natural or renewable sources and applied across various domains, including water treatment, desalination, and gas separation processes etc. (Lustenberger & Castro-Munoz, 2022; Bandehali et al., 2021).

Various membrane operations like microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), pervaporation, membrane distillation, and electrodialysis are commonly employed for many applications. However, pore sizes decrease as membrane separation efficiency increases, necessitating higher pressure application. Consequently, mechanically stable, flexible, and thermally resistant synthetic polymers such as PVDF, PAN, PTFE, PS, PES, PEI, and polyethylene are predominantly used. Yet, the adverse environmental and health impacts of these fossil-based polymers have spurred a shift towards biodegradable alternatives like polylactic acid (PLA), polyhydroxyalkanoates (PHA), polybutylene succinate (PBS), cellulose, chitin, chitosan, alginate, collagen, lignin, sporopollenin etc. (Xie et al., 2020; Jiang, 2020).

Over recent decades, membrane technology has emerged as a widely adopted and swiftly expanding separation method, offering numerous advantages over conventional techniques, such as easy setup and operation, minimal energy and chemical usage, cost efficiency, compact space requirements, and decreased environmental impact (Xie et al., 2021). However, certain aspects of membrane production, mainly fossil-based polymers and solvents, lack sustainability. The reliance on fossil-based polymers and extensive use of hazardous organic solvents in membrane fabrication pose severe risks to both health and the environment (Nanda et al., 2022). The membrane market has long been dominated by petroleum-derived nonbiodegradable polymers, extensively utilized across various sectors like water & waste-water treatment, desalination, gas separation, food& beverage, pharmaceutical, and

medical industries, despite their notable adverse environmental effects (Bandehali et al., 2022). These polymers, being nonrenewable and resistant to degradation, raise concerns regarding their production and disposal practices, often generating microplastics that negatively impact ecosystems and living organisms (Nguyen Thi et al., 2022). To address these challenges, researchers focus on developing bio-based or biodegradable polymers for membrane manufacturing, aiming to drive a greener transformation in membrane technology. Several strategies, inspired by the 12 principles of green chemistry and green chemical engineering and embracing a cradle-tograve approach, have been proposed to enhance the environmental sustainability of membrane technology, as illustrated in Figure 1. In the forthcoming decades, the chemical industry is anticipated to transition towards wholly sustainable and environmentally friendly processes. Within the membrane manufacturing phase, five distinct strategies have been suggested to enhance sustainability (Xie et al., 2021). Firstly, replacing entirely or partly the nonbiodegradable fossil-based polymers with renewable bio-based counterparts holds significant promise due to their biodegradability, biocompatibility, versatility, low carbon footprint, reduced toxicity, and societal acceptance (Xie et al., 2021; Nanda et al., 2022). Secondly, substituting toxic traditional solvents, such as 1-methyl-2pyrrolidinone dimethylformamide (NMP), (DMF), and N.Ndimethylacetamide (DMAc), with greener alternatives (Haque Mizan, et al. 2023). Thirdly, wastewater-primarily composed of organic solvents and polymers generated during membrane manufacturing, as a substantial volume remains untreated, estimated at over 50 billion litres globally annually (Razali et al., 2015). The fourth strategy involves streamlining membrane fabrication steps to reduce the use of toxic solvents, thereby lowering energy consumption and costs. Lastly, considering room temperature for casting solution preparation could minimize energy consumption in the process.

#### Figure 1.



Green membrane fabrication concept for sustainability

## **Recyclable Materials**

Over millions of membrane modules, mainly reverse osmosis (RO), have been manufactured, and their use time in the process is about 5 years; many of them are discarded and could result in the disposal of hundreds of thousands of tons of intricate composite items, with only approximately 5% by weight comprising direct membrane materials, while the rest comprises spacers and casings. While disassembling these membrane modules is labour-intensive, there might be potential to recover the polymeric materials used in the spacers and main casing components. However, recycling the membrane materials themselves poses significant challenges as they require harsh chemical treatments for valorization. New strategies are urgently required to tackle these challenges and enhance production efficiency while reducing waste. An additional facet of green membranes involves incorporating recycled materials into membrane production. For instance, employing recycled plastic waste as a constituent for polymeric membranes can significantly mitigate the environmental impact associated with conventional manufacturing methods. Recent investigations have also explored the use of waste materials such as animal bones and keratin for membrane fabrication. Consequently, reevaluating waste materials as valuable resources for membrane production presents a viable avenue towards establishing the membrane industry as a sustainable industrial model (Goh et al 2021).

The circular economy, centered on efficient resource management to curb waste generation and foster more sustainable production, revolves around three key principles: reduce, reuse, and recycle (Abou Taleb & Al Farooque, 2021; Lieder & Rashid, 2016), Membrane technology, developed for various applications spanning water treatment, desalination, gas purification, dairy and beverage processing, and energy generation, owes its widespread adoption to its potential for selective separation, ease of configurability, moderate energy consumption, reduced carbon footprint, recovery of valuable substances, and substitution of processes involving harsh and hazardous chemicals (Nunes et al., 2020, Xie et al., 2021). With mounting concerns regarding the adverse effects of conventional fabrication materials and techniques on human health and environmental pollution, a paradigm shift in membrane technology that emphasizes new material sources and reduces raw material usage has emerged (Yadav et al., 2021; Ding et al., 2021). Notably, the membrane industry heavily relies on non-degradable materials and highly toxic chemicals and solvents, many of which were developed during periods of lax environmental regulations compared to today's standards (Shi et al., 2014; Zou et al, 2021). Thus, aligning membrane fabrication with sustainability and environmental protection demands a shift towards greener practices.

Recent suggestions aimed at transforming the membrane industry toward sustainability include strategies proposed by Xie et al. (2020), advocating for the adoption of biodegradable polymers, greener solvents, sustainable fabrication processes, the reuse of wastewater-generated ingredients, sustainable module design, and module recycling. These strategies aim to enhance the membrane industry's sustainability and promote sustainable membrane and membrane process development. When assessing materials for membrane fabrication, numerous potentially reusable and recyclable resources emerge within and beyond the membrane industry (Al-Mutwalli et al., 2023). Discarded plastics, waste paper, newspapers, and reusable elements sourced from waste and wastewater generated during membrane production stand as viable candidates for creating new membranes. Notably, the yearly output of wastewater from the membrane industry amounts to 50 million cubic meters, containing polymers, solvents, and additional additives. The focus should be on reclaiming these constituents to enable cost-effective and environmentally sound membrane production. Waste originating from agricultural produce and animal byproducts, alongside postconsumer plastics, represent primary sources for membrane fabrication materials. For instance, animal-derived waste materials such as wool, hair, and poultry feathers (Goh et al., 2021), as well as crop residues like corn husks, maize, rice straws, sunflower, and wheat stalks, exhibit numerous advantages; abundant supply, renewability, and low energy requirements for conversion into new raw materials suited for membrane fabrication (Debnath et al., 2021). Assessments on polymers and postconsumer plastics indicate a staggering production of 6.3 billion tons of plastic waste over the last 70 years, with a mere 9% being recycled (Li et al., 2021). This substantial volume of discarded plastic positions it as a viable and recyclable raw material within the membrane industry. Leveraging these newly identified resources for reusing and recycling discarded materials, both within and beyond the membrane industry, could significantly propel the shift toward more sustainable and environmentally friendly membrane manufacturing processes (Li et al., 2021). This review delves into organic and inorganic recyclable materials pivotal for membrane fabrication within the context of recycled materials. The aim is to establish a comprehensive framework outlining recyclable materials, thus fostering a crucial and leading role in steering the membrane fabrication industry toward a greener and more sustainable transformation.

While the potential of biopolymers is extensive, the bioplastics market encounters several limitations and hurdles it must overcome. Factors like material costs, cost-effective manufacturing, compatibility with existing infrastructure, and waste management significantly influence this market. Conversely, elements such as material availability, end-of-life product management, and waste remediation favor bioplastics, as many raw materials are sourced from nature and are readily biodegradable. However, there's limited understanding of the synthesis of bioplastics for some of these biomaterials, driving the search for more cost-effective extraction methods and exploring their potential for large-scale production. Bioplastics, known for their biodegradability and eco-friendly extraction, offer potential in the current membrane market. Yet, the biodegradability of certain bioplastics remains debatable, requiring proper remediation technologies and sorting facilities for distinct waste types instead of direct landfill deposition. Different membrane fabrication techniques can tailor functional groups of various bioplastic materials, yet more research is needed regarding their stability, simple chemical synthesis, or biosynthesis. These biomaterials can substitute more environmentally hazardous nanomaterials in surface modification of membranes.

## **Green Solvents**

Membranes are widely used and approved as a clean and environmentally friendly separation process with lower energy consumption compared to conventional techniques. However, the utilization of highly toxic chemicals. non-degradable hydrocarbon-based polymers, and harmful solvents, as used for the current membrane materials and fabrication techniques, have been identified as harmful materials for the sustainable development goals of the United Nations. Consequently, the materials used for membrane development are seriously questioned. Furthermore, concerns persist regarding the impact of membrane production, polymers, and chemicals on human health, water source contamination, global warming, and marine ecotoxicity (Yadav et al., 2021; Ding et al., 2021). Therefore, there is a critical need to envision the next iteration of membranes and their fabrication techniques, employing markedly cleaner and more sustainable approaches. Various alternative strategies have been explored and developed thus far, such as methods utilizing greener polymers, fewer chemical solvents (e.g., electrospinning, electroblowing), using eco-friendly solvents (e.g., DMSO) in the fabrication process, solvent-free techniques like 3D printing, and the utilization of bio-sourced, biodegradable, and/or natural polymers. Figure 1 illustrates the primary transformative alternatives in green membrane technology, categorizing these advancements into two main groups: green membranes and green membrane processes (Yusuf et al., 2020). Notably, 'green' membrane technology encompasses various approaches, including the utilization of biodegradable polymers and non-toxic solvents for membrane coating/casting, substituting non-biodegradable petroleumbased products and hazardous solvents. Other strategies involve minimizing preparatory steps to reduce energy consumption and waste generation, recycling useful membrane components, managing waste brine or sludge, reducing fouling tendencies, and harnessing energy from waste (Xie et al., 2020; Jiang and Ladewig, 2020; Aburabie et al., 2020; Carner et al., 2020).

The driving force behind innovation and new discoveries in sustainability and environmental impact mitigation stems from the adoption of the green chemistry approach. This approach is motivated by health concerns and a growing environmental consciousness. Consequently, extensive research and development efforts are channeled toward identifying eco-friendly alternatives in raw materials, methodologies, solvents, and energy usage. The emergence of groundbreaking advancements in green material production, waste reduction, recycling, and energy/cost efficiency signals promising prospects for a more sustainable and environmentally conscious future. Moreover, integrating economic, social, and environmental components is the cornerstone for establishing sustainability (Mohamed & Yousef, S., 2021, Clarke et al., 2018).

Green methodologies, like solvent-free electrospinning, have been focused on their ability to generate ultrafine fibers efficiently, with zero solvent evaporation, minimal toxic waste, and the production of less soluble fibers, vet challenges persist. These challenges encompass high setup requirements. relatively larger fiber diameters, lack of systematic theoretical guidance, and incomplete large-scale production methodologies. Despite recent strides in developing the green strategy for nanofiber membrane technology, achieving widespread utilization in industries demands continued effort. Future research should prioritize the development and adoption of water-soluble polymers, the elimination of harmful organic solvents, and the optimization of solvent-free electrospinning devices. Shifting from expensive, limited material sources to more affordable, non-toxic, and environmentally friendly alternatives is imperative. Furthermore, enhancing mechanical performance and structural design will broaden potential industrial applications. Introducing green materials, using water-based or less toxic solvents, and embracing solvent-free or reduced-solvent methods for fabricating greenbased nanofiber membranes are crucial steps in aligning with industrial production practices.

Green membrane fabrication is an essential aspect of sustainable development, aiming to minimize the environmental impact of membrane production. Several studies have highlighted the importance of employing green chemistry and sustainable materials in membrane fabrication and emphasized the significance of green chemistry in minimizing the generation of hazardous compounds during membrane preparation. Furthermore, successfully replaced toxic solvents with a green solvent, PolarClean, during ultrafiltration membrane fabrication, marking a significant step towards sustainable membrane production (Xie et al., 2019).

However, despite the progress that membrane fabrication is yet to be fully green, indicating the need for further advancements in this area (Park et al., 2021), also highlighted that while membrane-based operations are generally considered green and sustainable, the membrane fabrication process itself

still lags behind in terms of sustainability (Meringolo et al., 2018). Moreover, emphasized that sustainable membrane fabrication remains an unsolved challenge, essential for overall membrane filtration to be considered truly green (Le Phuong et al., 2019). Several studies have proposed the use of green solvents and eco-friendly materials in membrane fabrication. For instance, emphasized the importance of employing green solvents in membrane fabrication, as highlighted in recent review articles (Park et al., 2021). While some biomaterials display antimicrobial properties against common microorganisms, their effectiveness against the various microorganisms found in feed wastewater remains uncertain. Bio-based cross-linkers like phenalkamine (FA) which is produced through the Mannich reaction of cardanol with certain amines, which results in a partly bio-based polymer (Pirada, 2022) and can replace synthetic ones but might require additional nanomaterials to enhance stability. Despite the functional benefits bio-based polymers and additives offer in membrane fabrication, the process has not yet entirely transitioned to green technology. Green solvents like triethyl phosphate (TEP) and methyl-5-(dimethylamino)-2-methyl-5-oxopentanoate (Rhodiasolv PolarClean) show promise in membrane fabrication, potentially transforming synthetic membranes into greener alternatives by replacing conventional organic solvents (Russo et al., 2023).

## Additive Manufacturing

Additive manufacturing (AM) shows potential to reduce environmental impacts in manufacturing by cutting material use and enabling local production (Jung et al., 2023). However, high production costs and increased energy use in mass production limit its viability compared to traditional methods. The economic and environmental preference for AM depends on factors like production volume and part size. Addressing challenges such as material costs and production speed requires advances in material science and technology beyond just scaling up AM operations. Comparative studies show that AM has lower environmental impacts at very low production volumes unless the part geometry is specific. Higher production costs and environmental impacts of AM at large volumes can be balanced by factors like small part size or performance advantages. Future advancements in material production and AM processes could enhance its environmental and economic benefits. Additionally, studies should incorporate lifecycle energy consumption, CO<sub>2</sub> emissions, human toxicity, and lifecycle cost metrics.

Advancements in solvent-free fabrication techniques, such as 3D printing, hold promise in diversifying membrane technology. Employing 3D printing to create different membrane module components presents an avenue for unparalleled customization and process optimization. However, this technology necessitates further exploration and refinement to enable the production of defect-free membranes and modules at larger scales, especially in fabricating membranes featuring nanoscale pores (Tijing et al., 2020). Additive manufacturing, also known as three-dimensional (3D) printing, involves creating numerous nanofibers that are subsequently compressed to shape objects according to specific dimensions, shapes, and porosities. Comparing 3D printing to traditional manufacturing methods reveals several advantages which offer flexibility in integrating diverse designs and geometries with precise control over object porosity, thickness, and structure, layer-wise control of pore density with varied particle sizes, enhanced resolutions, the ability to produce mechanically robust and chemically resistant objects, a swift and scalable process, and reduced reliance on toxic solvents compared to conventional methods (Sreedhar et al., 2023).

The potential of 3D printing is foreseen to address numerous challenges in membrane production. Enhanced designs in membranes could significantly improve performance and durability. Presently, two widely used membrane types are ceramic and polymeric membranes. Additive manufacturing stands to bridge this gap by enabling the fabrication of cost-effective membranes, offering better control over fabrication parameters (Sreedhar et al., 2023). However, limitations exist in the current state of AM-based membranes, particularly in their restricted resolutions and comparatively lower printing speeds. Furthermore, 3D printing presents opportunities for manufacturing other essential components of membrane modules, overcoming various design constraints. However, despite its potential, challenges persist; for instance. commercializing microfluidic-based products encounters limitations in manufacturing, leading to renewed interest in using AM techniques to craft tailor-made microfluidic devices (Sreedhar et al., 2023). Researchers aim to integrate porous materials into 3D-printed nonporous fluidic devices for applications in membrane separations. Yet, this endeavor faces limitations in material choices, printing control within microfluidic environments, and mitigating fouling and undesirable solute-material interactions. Despite its advantages, 3D printing in membrane technology encounters drawbacks compared to conventional manufacturing, such as limitations in printable materials, desired porosities, and object dimensions. Optimization based on the polymer's chemical nature and membrane structure is crucial, considering 3D printing's relative costliness, limited build volume, resolution, and material choices.

## **Challenges and Outlook**

This review focuses on various innovative approaches to green membrane manufacturing, including the utilization of green solvents, bio-sourced polymers, recycled materials, and additive manufacturing strategies. However, these approaches are presently confined to laboratory-scale experiments and necessitate further demonstration to achieve commercial readiness. Nevertheless, the proliferation of industry-linked research projects in this domain underscores the escalating demand for such innovations on a larger scale.

The emergence of eco-friendly approaches and sustainable practices in separation science enables the more environmentally conscious processing and reutilization of membrane materials. Creating a circular economy requires establishing interconnected closed-loop systems that eliminate waste streams and prioritize recycling or repurposing materials to sustain or improve their value. This transformation, crucial in membrane science, engineering, and various industries, calls for innovative engineering solutions that support fresh design perspectives. Developing eco-friendly solutions can streamline material recycling in a circular manner, aiding in the formulation of more efficient strategies from the outset. This approach ultimately reduces environmental impact and energy requirements, marking a significant stride toward sustainability.

Despite advancements made thus far, hurdles persist in the widespread adoption of environmentally friendly membrane technology. The substantial manufacturing costs associated with biodegradable membranes are a formidable challenge, necessitating focused research and development endeavors. Advancements in materials science and engineering offer promise in introducing novel membrane materials with improved selectivity and permeability, driving sustainability efforts forward. Moreover, fine-tuning membrane fabrication techniques and processes themselves could augment the efficiency and longevity of green membrane technologies. The future course of green membrane technology entails synergizing with other ecofriendly technologies, such as leveraging renewable energy sources like hydrogen, solar, wind, or geothermal energy. Additionally, tapping into industrial waste heat to power membrane-based processes represents a pivotal integration (Shirazi et al., 2023). These strategic amalgamations harbor significant potential for substantially reducing the carbon footprint while fortifying overall sustainability initiatives. With continued commitment to advancement and collaboration, green membranes can play a pivotal role in tackling environmental challenges and fostering a more sustainable and resilient future.

A critical area that warrants further attention is the methodical assessment of the life cycle of membrane materials and systems. It's imperative to systematically integrate such tools into the design of membrane materials to ensure viability, scalability, and environmental relevance. Frequently, intriguing material solutions offered at the laboratory level are not commercially feasible due to evident cost and sustainability concerns. Additionally, the exploration of biodegradable membranes needs further evaluation to mitigate waste disposal issues and support the breakdown of materials into non-toxic compounds through natural enzymatic or environmental processes. Biodegradable membranes hold promise as potential alternatives to conventional ones. Moreover, there's a significant call for comprehensive lifecycle assessments that encompass the entire span of existing and newly developed green membranes. These evaluations, covering everything from production and operation to eventual disposal or recycling, would yield a deeper understanding of the environmental impact of these membranes and their processes.

### **Future perspectives**

Membrane technology stands as a cornerstone in the pursuit of sustainable practices across industries. Its versatile applications, spanning from water purification to biomedical fields, highlight its pivotal role in addressing pressing global challenges. While effective, traditional membrane fabrication methods often pose significant environmental concerns due to the use of nonrenewable resources and energy-intensive processes. The advent of green membrane fabrication offers a transformative shift towards sustainable practices, minimizing environmental footprints while maintaining or enhancing performance metrics.

The concept of green membranes revolves around the integration of ecofriendly materials and environmentally conscious fabrication techniques. These membranes are designed to harness the principles of sustainability throughout their life cycle, from material selection and fabrication processes to application and disposal/recycling. By embracing biodegradable, renewable, or recycled materials and employing low-energy, solvent-free, or bioinspired fabrication methods, green membranes aim to reduce resource consumption, minimize waste generation, and mitigate environmental impact compared to conventional counterparts.

The rationale for delving into green membrane fabrication stems from the urgent need to address environmental degradation, resource depletion, and the growing demand for sustainable technologies. This exploration is driven by the inherent potential of green membranes to offer solutions that mitigate environmental harm and optimize performance, durability, and cost-effectiveness. Understanding the significance and possibilities of green membranes is paramount in fostering a sustainable future across diverse sectors, ranging from water treatment and energy production to healthcare and beyond. The future of green membrane fabrication rests on its ability to scale sustainably. Innovations will focus on seamlessly transitioning eco-friendly processes from laboratory-scale experimentation to large-scale industrial production. Circular economy principles will drive the adoption of recycling and reuse strategies, minimizing waste and resource consumption throughout the membrane life cycle.

Advancements in fabrication techniques will revolutionize the scalability and precision of green membrane production. Additive manufacturing and 3D printing methodologies will enable intricate membrane designs using ecofriendly materials, fostering unparalleled control over membrane architecture. Integrating artificial intelligence and machine learning algorithms will optimize fabrication processes, ensuring efficiency and reproducibility at industrial scales. Although additive manufacturing (AM) has progressed significantly, its full potential in membrane technology remains untapped. The application of 3D printing in membranes and membrane modules is still at an early stage, necessitating further exploration. To enhance its deployment and applicability, several future perspectives were outlined (Koo et al., 2021). The direct printing of membranes with submicrometer-sized pores faces challenges due to the limited resolution of current 3D printers. Implementing hybrid AM in membrane technology could redirect future studies, enhancing spacer properties, such as chemical and mechanical stability, for broader commercial applications. However, limitations in hybrid AM regarding materials compatibility, process interconnectivity, and interfacial bonding need addressing (Koo et al., 2021). Most current research on 3D printing in membrane technology is confined to lab-scale testing, often under unrealistic conditions (Atkinson, 2018; Nunes et al., 2019). Scaling up and resolving material issues are key to realistically testing these structures/units in larger industrial applications. Addressing upscaling challenges involving build size, printing speed, and manufacturing power is essential to commercialize 3D printing in membrane technology. Moreover, current 3D printing speeds lag significantly behind conventional manufacturing methods, impacting commercial feasibility. Additionally, material issues, such as material selection based on implementation and environmental conditions, are critical for industrial-scale 3D printing realization in membrane technology.

The emergence of four-dimensional (4D) printing holds promise for membrane technology's future. This technology adds a fourth dimension to 3D printing, utilizing intelligent materials that respond to specific stimuli. Intelligent materials offer self-repair, multifunctionality, and self-assembly properties. Yet, their exploration of membrane technology, including spacers, membranes, and modules, remains unexplored. Future studies in 4D printing could transform 3D-printed membranes into commercial reality, fabricating optimized fouling-resistant and self-cleaning spacers, smart membranes with varying properties under different conditions, and components responsive to stimuli like concentration, pH, and charge (Koo et al., 2021).

Surface modification and coating of polymer membranes serve to either create new or enhance existing properties by altering the surface chemistry, a process known as functionalization. This modification aims to elevate separation performance while preserving the inherent properties of the membrane. These approaches can occur during the membrane fabrication phase or as post-treatment methods after membrane production. In water purification applications, treating the membrane surface plays a pivotal role in combating fouling, which typically falls into five primary categories: biofouling, protein fouling, antifoam fouling, colloidal fouling, and scaling fouling.

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