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GAS SEPARATION MEMBRANES: ADVANCES IN POLYMER AND MIXED-MATRIX MEMBRANES FOR CO_2 CAPTURE

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ABSTRACT

This study provides a comprehensive review of advancements in polymer and mixed-matrix membranes (MMMs) for CO_2 capture, focusing on their design innovations, performance enhancements, sustainability, and scalability. A total of 92 peer-reviewed articles were analyzed following the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines to ensure a systematic and transparent review process. The findings highlight significant progress in polymer membranes, particularly in overcoming the permeability-selectivity trade-off through advanced techniques such as cross-linking, functional group incorporation, and polymer blending. MMMs have emerged as a transformative technology, leveraging the unique properties of inorganic fillers like metal-organic frameworks (MOFs) and graphene oxide to achieve superior separation performance and durability under industrial conditions. Additionally, the review emphasizes the growing focus on sustainability, with an increasing shift toward biodegradable and recyclable membranes that reduce the environmental footprint of CO_2 capture. The application of artificial intelligence (AI) and machine learning (ML) has further accelerated material discovery, optimized membrane design, and improved operational efficiency. Membranes' integration into broader Carbon Capture, Utilization, and Storage (CCUS) frameworks showcases their versatility in CO_2 separation, utilization, and storage applications. Despite these advancements, challenges such as material degradation under harsh conditions, cost, and scalability persist, necessitating further research and innovation. This study underscores the critical role of membrane technologies in enabling efficient and sustainable CO_2 capture, offering valuable insights for researchers and industry stakeholders working toward global decarbonization goals.

1 INTRODUCTION

The escalating levels of carbon dioxide (CO_2) in the atmosphere represent one of the most pressing environmental issues of our time, with far-reaching implications for global climate systems and human well-being (Rezakazemi et al., 2014). CO_2 emissions, predominantly arising from industrial processes and fossil fuel consumption, account for a significant proportion of greenhouse gases contributing to global warming (Sutrisna et al., 2018). Tackling this challenge necessitates the development of innovative and efficient technologies for CO_2 capture and storage. Among various available options, membrane-based gas separation technologies have gained considerable attention due to their unique advantages, including energy efficiency, operational simplicity, and scalability across diverse industrial applications (Park et

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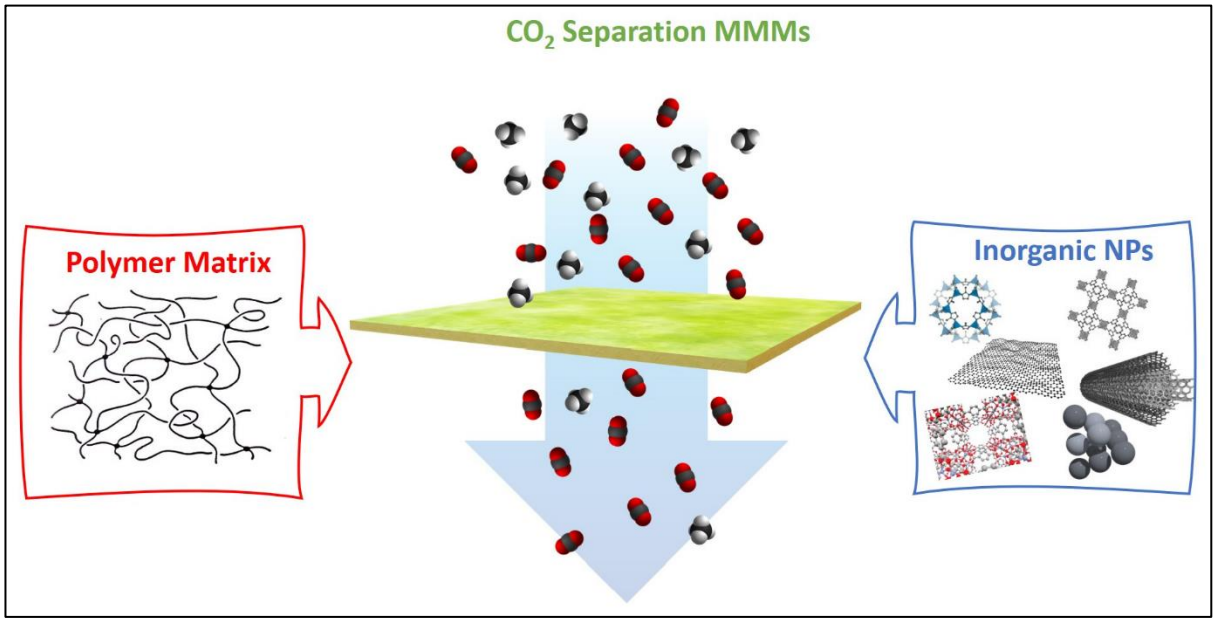
al., 2009). These attributes position membranes as a promising alternative to conventional methods such as chemical absorption and cryogenic distillation, which, while effective, are often energy-intensive and environmentally taxing (Park et al., 2010). Moreover, Polymer membranes have been a cornerstone of gas separation technologies due to their versatility, mechanical stability, and relatively low manufacturing costs. These membranes function by leveraging differences in the permeability of gases, selectively allowing CO_2 to pass through while restricting other gases (Chen et al., 2013). However, their practical application has been constrained by the well-documented trade-off between gas permeability and selectivity, commonly referred to as the Robeson upper bound (Li et al., 2013). To overcome this limitation, significant research has focused on modifying polymer structures through strategies such as incorporating functional groups, optimizing chain configurations, and employing cross-linking techniques. For instance, polymers like polyimides and polyethylene oxide (PEO) have shown improved gas transport properties, positioning them as candidates for next-generation CO_2 separation membranes (Dou et al., 2015). Figure 1 illustrates the CO_2 separation mechanism in mixed-matrix membranes (MMMs), showcasing the integration of a polymer matrix and inorganic nanoparticles (NPs) to enhance gas separation performance. The polymer matrix provides flexibility and structural support, while the inorganic NPs, such as metal-organic frameworks (MOFs) or graphene oxide, facilitate selective CO_2 transport through molecular sieving and sorption mechanisms. This synergy addresses the permeability-selectivity trade-off, a critical challenge highlighted in this study. By demonstrating how MMMs combine the advantages of polymers and fillers, the figure supports the study's focus on advancing CO_2 capture technologies through material innovation and optimizing membrane design for industrial scalability and sustainability (See Figure 1).

Figure 1: CO_2 Separation Mechanism in Mixed-Matrix Membranes (MMMs)



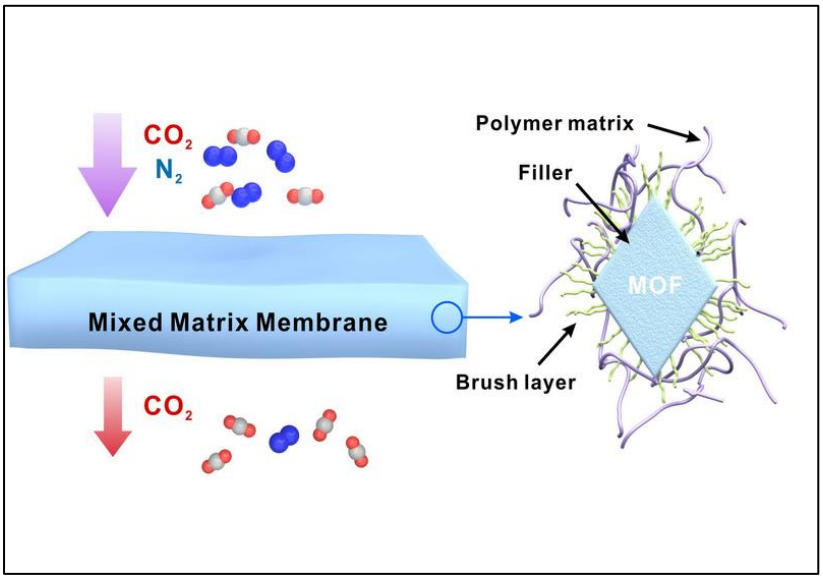
Source: Li et al. (2021).

In recent years, mixed-matrix membranes (MMMs) have emerged as a promising innovation in gas separation technology. MMMs combine the flexibility and processability of polymers with the superior separation performance of inorganic fillers such as metal-organic frameworks (MOFs), graphene oxide, and zeolites (Shen & Lua, 2013). By integrating these porous materials, MMMs can enhance CO_2 separation performance through molecular sieving and improved sorption mechanisms. Wang et al. (2015) report that MOF-incorporated MMMs have demonstrated superior performance, with notable increases in both permeability and selectivity. These findings underline the potential of MMMs as a transformative approach to addressing the inherent limitations of polymer-only membranes. Despite the promising advancements in

polymer and mixed-matrix membrane technologies, their scalability for industrial applications remains a significant hurdle. The operational environment for CO_2 capture often involves high pressures, varying temperatures, and the presence of impurities, all of which can degrade the performance of membranes over time (Zhu et al., 2012) (See figure 2).

Figure 2 provides a schematic representation of the gas separation process using mixed-matrix membranes (MMMs), highlighting the synergistic interaction between the polymer matrix and fillers, such as metal-organic frameworks (MOFs). The figure demonstrates how CO_2 molecules selectively permeate through the membrane, aided by the molecular sieving and sorption properties of the fillers, while other gases like N_2 are restricted. The incorporation of a brush layer further enhances the compatibility and dispersion of fillers within the polymer matrix, addressing common challenges such as interfacial voids. This aligns with the study's focus on overcoming scalability and efficiency barriers in MMMs, emphasizing their potential for industrial CO_2 capture applications. Furthermore, achieving seamless compatibility between the polymer matrix and inorganic fillers in MMMs is challenging, often resulting in defects such as interfacial voids that compromise gas separation efficiency (Li et al., 2015). Recent studies have emphasized the importance of surface functionalization and advanced synthesis techniques to mitigate these issues and enhance the long-term stability of MMMs (Jusoh et al., 2017). In addition to performance enhancements, there is growing interest in aligning membrane technologies with sustainability objectives. Environmentally friendly materials, such as biodegradable polymers and naturally sourced fillers, are being actively explored to reduce the ecological footprint of membrane production (Wang et al., 2020). Moreover, advancements in manufacturing techniques, including 3D printing and nanotechnology, have enabled the precise engineering of membrane architectures, resulting in improved separation performance and material efficiency (Ashtiani et al., 2021). These developments not only broaden the scope of membrane applications but also contribute to global sustainability efforts by reducing resource consumption and operational waste. The primary objective of this review is to systematically analyze recent advancements in polymer and mixed-matrix membranes (MMMs) for CO_2 capture, with a focus on material innovation, performance metrics, and industrial applicability. By adhering to the PRISMA framework, this study aims to synthesize findings from peer-reviewed literature to evaluate the state-of-the-art in gas separation membrane technology. Specific objectives include assessing the effectiveness of polymer modifications in overcoming the permeability-selectivity trade-off, exploring the role of mixed-matrix designs in enhancing separation performance, and identifying key challenges associated with membrane scalability and operational stability. Additionally, this review seeks to highlight emerging trends such as the

Figure 2: Schematic illustration of gas separation process using MMMs



Source: Wang et al.(2018).

integration of sustainable materials and advanced fabrication techniques that align membrane technologies with broader environmental and industrial goals. Ultimately, the findings aim to provide a comprehensive understanding of the field while identifying gaps and opportunities for future research.

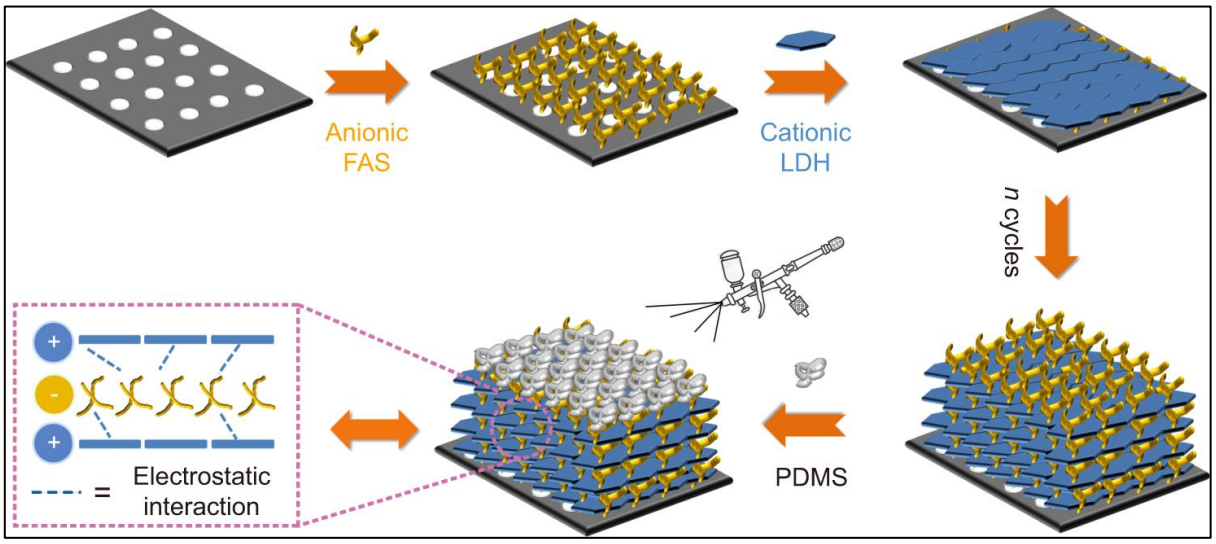
2 LITERATURE REVIEW

The development of gas separation membranes for CO_2 capture has been a focal area of research due to their potential to address environmental challenges and enhance industrial efficiency. This section reviews the existing body of literature, providing a critical analysis of advancements in polymer membranes and mixed-matrix membranes (MMMs) for CO_2 capture. It explores material innovations, performance improvements, and challenges in scaling these technologies for real-world applications. Drawing on the PRISMA framework, this review synthesizes findings from diverse studies to identify trends, highlight gaps, and outline future directions for research. The focus is on evaluating how recent developments align with the dual goals of improving membrane efficiency and achieving sustainability.

2.1 Polymer Membranes for CO_2 Separation

Polymer membranes, particularly those derived from polyimides, have been a cornerstone in CO_2 separation technologies due to their exceptional thermal and chemical stability, as well as their inherent mechanical strength (Chen et al., 2013). Polyimides have demonstrated superior gas transport properties by offering high CO_2 permeability and selectivity when compared to other polymeric materials. These advantages are attributed to the presence of imide linkages and aromatic groups in their molecular structure, which facilitate CO_2 sorption and diffusion (Zhu et al., 2012). Additionally, polymers like polyethylene oxide (PEO) and polyetherimide (PEI) have been engineered to enhance CO_2 separation efficiency. For example, PEO-based membranes have shown remarkable affinity for CO_2 due to their high ether group density, which interacts favorably with CO_2 molecules (Ashtiani et al., 2021; Park et al., 2010). Studies suggest that polyimide derivatives remain at the forefront of polymer membrane research due to their ability to operate effectively under high-pressure and high-temperature conditions (Ansaloni et al., 2021; Li et al., 2017). To further improve the performance of polymer membranes, researchers have employed advanced modification techniques such as cross-linking, functional group incorporation, and polymer blending. Cross-linking has been particularly effective in enhancing the stability and selectivity of polymer membranes by reducing polymer chain mobility and restricting the diffusion of larger gas molecules (Ashtiani et al., 2021). For instance, cross-linked polyimide membranes have shown significant improvements in CO_2 selectivity without compromising permeability (Chen et al., 2013). The incorporation of functional groups, such as amines or carboxylic acids, has also been widely explored to improve CO_2 affinity. Aminated membranes, in particular, exhibit enhanced CO_2 capture performance due to their ability to

Figure 3 :Schematic representation for the fabrication of $(LDH/FAS)_nPDMS$ membranes



Source: Wang et al.(2018)

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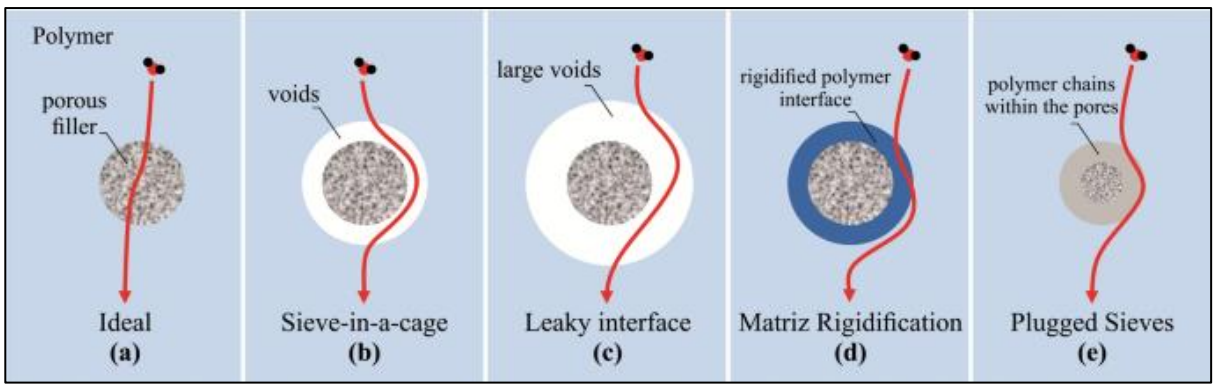
chemically interact with CO_2 molecules (Li et al., 2013). Polymer blending, on the other hand, allows for the creation of hybrid materials that combine the desirable properties of multiple polymers. For example, blending PEO with polyimides has resulted in membranes with balanced permeability and selectivity, addressing the trade-offs inherent in single-polymer systems (Guan et al., 2020; Qiuchen et al., 2021).

Figure 3 illustrates the schematic fabrication process of $(LDH/FAS)_n$PDMS membranes, emphasizing the electrostatic interaction between cationic layered double hydroxides (LDH) and anionic fluorinated alkyl silanes (FAS) layers. This multilayered structure enhances gas separation performance by creating highly selective pathways for CO_2 transport. The incorporation of polydimethylsiloxane (PDMS) provides mechanical flexibility and stability, critical for industrial applications. This figure directly aligns with the study's focus on polymer membranes, showcasing how innovative material engineering, such as surface functionalization and layer-by-layer assembly, addresses challenges like permeability-selectivity trade-offs and operational durability in CO_2 capture technologies.Furthermore, while these advancements in polymer design have pushed the boundaries of CO_2 separation performance, challenges remain in achieving the ideal balance between permeability and selectivity. The Robeson upper bound, which defines the trade-off between these two properties, continues to be a significant constraint for polymer membranes (Shen et al., 2016). Innovative approaches, such as incorporating nanostructures and utilizing advanced polymerization techniques, are being explored to transcend this limitation. For example, the integration of nanostructured additives like graphene oxide into polyimide membranes has shown potential to enhance both permeability and selectivity (Ashtiani et al., 2021). Similarly, advanced polymerization techniques, including atom transfer radical polymerization (ATRP), have enabled the precise tailoring of polymer chains to optimize gas transport properties (Dou et al., 2015). These approaches suggest that polymer membranes remain a fertile area for innovation in CO_2 capture technologies. Moreover, the combination of polymer chemistry advancements and modification techniques highlights the potential of polymer membranes as a cost-effective and scalable solution for CO_2 separation. However, their real-world application requires addressing critical issues such as long-term stability under industrial conditions, fouling resistance, and compatibility with integrated carbon capture systems (Li et al., 2015; Shen et al., 2016).

2.2 Mixed-Matrix Membranes

The incorporation of inorganic fillers into polymer matrices has revolutionized mixed-matrix membranes (MMMs), positioning them as a promising solution for CO_2 separation. Materials such as metal-organic frameworks (MOFs), zeolites, and graphene oxide have been widely explored for their ability to enhance the separation performance of polymer membranes (Wu et al., 2020). MOFs, in particular, offer a high surface area, tunable pore size, and strong CO_2 adsorption capacity, making them ideal for improving gas permeability and selectivity (Zhao et al., 2019). Similarly, zeolites provide molecular sieving properties that enable precise separation of CO_2 from other gases, while graphene oxide enhances gas transport by introducing additional diffusion pathways (Li et al., 2019; Wang et al., 2018). Recent studies have demonstrated that MMMs with MOF fillers exhibit substantial improvements in CO_2 permeability without sacrificing selectivity,

Figure 4 : Defects in Mixed-Matrix Membranes (MMMs) for Gas Separation



Source: Flores et al. (2021).

highlighting their potential for industrial applications (Qiao et al., 2016). A critical challenge in developing high-performance MMMs is achieving compatibility between the polymer matrix and inorganic fillers. Poor dispersion of fillers or the presence of interfacial voids can significantly reduce membrane performance, as these defects create undesired gas transport pathways (Sarfraz & Ba-Shammakh, 2016). Researchers have focused on surface modifications of fillers to enhance compatibility. For example, functionalizing MOFs with organic ligands or introducing surface-treated graphene oxide has been shown to improve filler dispersion and minimize void formation (Park et al., 2019). Additionally, modifying polymer matrices with functional groups that interact with filler surfaces can further enhance compatibility. Wang et al. (2018) report that surface-modified MOF fillers in polyimide matrices not only improve dispersion but also enhance the mechanical stability of the resulting membranes, ensuring long-term operational efficiency. In this context, figure 5 illustrates common defects in mixed-matrix membranes (MMMs) for gas separation, including voids, large interfacial gaps, polymer chain rigidity, and pore blockage. These defects, such as the "sieve-in-a-cage" effect or leaky interfaces, arise from poor compatibility and dispersion of inorganic fillers within the polymer matrix, undermining gas separation performance. The ideal MMM (Figure 4a) facilitates selective CO_2 transport without undesired pathways. This figure aligns with the study's exploration of improving filler-polymer integration using surface functionalization and advanced fabrication techniques, addressing these challenges to enhance permeability, selectivity, and operational stability in CO_2 capture applications (see figure 4).

The performance metrics of MMMs, particularly permeability, selectivity, and stability, underscore their advantages over traditional polymer membranes. The integration of inorganic fillers typically enhances CO_2 permeability by providing additional transport channels while maintaining or improving selectivity through molecular sieving effects (Park et al., 2019). For instance, MMMs with MOF fillers have shown up to a 50% increase in CO_2 permeability compared to pure polymer membranes, without compromising selectivity (Jeazet et al., 2016). Operational stability is another critical performance metric, with MMMs demonstrating resilience under high-pressure and high-temperature conditions. Studies have highlighted the ability of MMMs to retain performance over extended periods, making them suitable for industrial-scale CO_2 capture (Dechnik et al., 2017). Despite these advancements, challenges remain in scaling MMMs for real-world applications. The cost and scalability of synthesizing inorganic fillers like MOFs remain significant barriers, as do the complexities of achieving uniform filler dispersion in large-scale manufacturing processes (Xinru et al., 2018). Researchers are exploring cost-effective synthesis methods for fillers and scalable fabrication techniques such as phase inversion and solvent casting to address these challenges (Boroglu & Yumru, 2017). Additionally, the development of hybrid fillers that combine multiple functionalities is gaining traction as a means to optimize the performance of MMMs while reducing material costs (Park et al., 2019). These strategies are critical for advancing MMMs toward industrial adoption. The ongoing advancements in MMMs for CO_2 capture highlight their potential to bridge the gap between high performance and industrial feasibility. By leveraging the unique properties of inorganic fillers and addressing challenges in polymer-filler compatibility, researchers are paving the way for next-generation membrane technologies. Future research should continue to focus on optimizing material properties and developing cost-effective, scalable fabrication techniques to unlock the full potential of MMMs for large-scale CO_2 separation (Boroglu & Yumru, 2017; Wang et al., 2018). These efforts align with the broader goals of reducing greenhouse gas emissions and achieving sustainable industrial processes.

2.3 3D Printing and Nanotechnology in Membrane Design

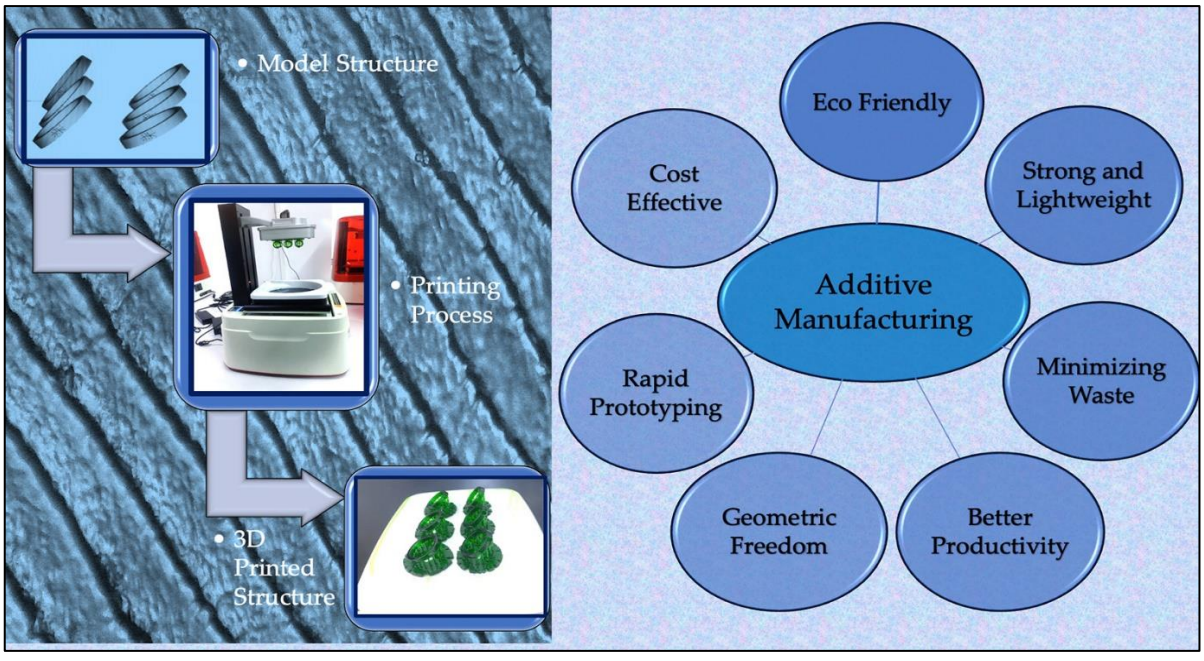
Advances in 3D printing technology have introduced transformative possibilities for fabricating gas separation membranes with unprecedented precision and complexity. Unlike traditional manufacturing techniques, 3D printing enables the layer-by-layer construction of membranes with finely controlled architectures, resulting in improved gas separation performance (Zhao et al., 2020). This technology allows the customization of membrane designs, including tailored pore structures and optimized surface properties for specific separation applications. For instance, Priolo et al. (2015) demonstrated that 3D-printed polymeric membranes with hierarchical porosity achieved enhanced CO_2 permeability and selectivity. Furthermore, additive manufacturing

techniques enable the integration of multiple materials within a single membrane, facilitating hybrid designs that combine the advantages of polymers and fillers (Wang et al., 2017). This adaptability positions 3D printing as a key enabler for next-generation gas separation technologies.

Nanotechnology has further revolutionized membrane design by enabling the incorporation of nanoscale fillers such as metal-organic frameworks (MOFs), graphene oxide, and carbon nanotubes (CNTs) into polymer matrices. These nanomaterials provide unique properties, such as high surface area, tunable pore size, and enhanced mechanical strength, which significantly improve gas transport performance (Li et al., 2021). For instance, graphene oxide has been extensively used to introduce nanostructured pathways that facilitate selective CO_2 transport, leading to higher permeability and selectivity (Kim et al., 2020). Additionally, the incorporation of MOFs into membranes via nanotechnology-based techniques has yielded exceptional performance in CO_2 separation due to their molecular sieving and adsorption capabilities (Li et al., 2021). Nanotechnology thus complements 3D printing by enabling the precise integration of advanced materials into membrane designs (Figure 5).

Figure 5 showcases the workflow of 3D printing in membrane design and highlights the key advantages of additive manufacturing, including eco-friendliness, cost-effectiveness, geometric freedom, and minimized waste. The figure demonstrates how 3D printing enables the creation of precise membrane structures with tailored properties, facilitating enhanced gas separation performance. This aligns with the study's focus on leveraging 3D printing to fabricate next-generation membranes, which combine polymers and advanced fillers such as MOFs and graphene oxide for superior CO_2 permeability and selectivity. By enabling rapid prototyping and scalability, additive manufacturing addresses challenges in traditional fabrication methods, supporting the study's emphasis on innovative, efficient, and sustainable membrane design. Furthermore, the synergy of 3D printing and nanotechnology offers new avenues for addressing traditional challenges in membrane fabrication, such as uniform filler dispersion and defect minimization. Through nanoscale surface engineering, fillers can be functionalized to improve compatibility with polymer matrices, reducing the likelihood of interfacial voids (Ahmad & Hägg, 2013). Concurrently, 3D printing allows for the precise placement of these fillers within the polymer matrix, ensuring consistent performance across the membrane. (Farha & Hupp, 2010) reported that combining 3D printing with nanotechnology-enabled modifications significantly enhanced the operational stability of mixed-matrix membranes under industrial conditions. Such approaches ensure that membranes retain their structural and functional integrity over extended periods of use.

Figure 5 : 3D in Process Workflow and Key Advantages in Membrane Design



Source: Khan et al. (2022).

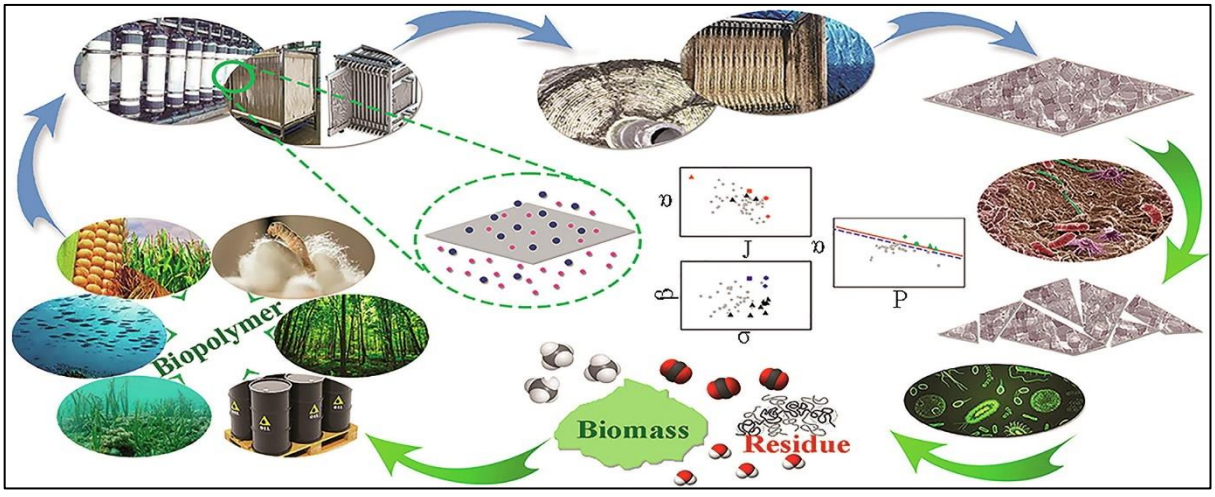
One of the most significant advantages of 3D printing and nanotechnology in membrane design is the potential for scalability and cost-effectiveness. Traditional membrane fabrication methods often involve labor-intensive processes and high material waste, limiting their industrial scalability (Lu et al., 2018). In contrast, 3D printing enables the rapid prototyping and mass production of membranes with minimal waste, while nanotechnology facilitates the use of highly efficient, low-cost fillers (Farha & Hupp, 2010). Recent advancements in automated 3D printing systems and nanomaterial synthesis methods further enhance the feasibility of adopting these technologies for large-scale applications (Wang & O'Hare, 2012). This scalability makes 3D-printed and nanotechnology-enhanced membranes an attractive option for industrial CO_2 capture. Despite these advancements, challenges remain in fully harnessing the potential of 3D printing and nanotechnology in membrane design. Key issues include the high cost of advanced 3D printers and nanomaterials, as well as the technical complexity involved in combining these technologies seamlessly (Hao et al., 2010). Additionally, the long-term stability of 3D-printed membranes incorporating nanostructures under extreme operational conditions requires further investigation (Dai et al., 2016).

2.4 Eco-Friendly Polymer Membranes

The increasing focus on sustainability in industrial processes has led to the exploration of eco-friendly polymer membranes as a solution for CO_2 capture. These membranes leverage biodegradable and recyclable materials to minimize their environmental impact while maintaining high separation performance (Swaidan et al., 2014). Unlike traditional polymers, which often rely on petrochemical derivatives, biodegradable polymers such as polylactic acid (PLA) and polyhydroxyalkanoates (PHAs) are derived from renewable resources and decompose naturally under appropriate conditions (Wang & O'Hare, 2012). For instance, PLA-based membranes have shown considerable potential for CO_2 separation due to their tunable properties and compatibility with various functional modifications (Cheng et al., 2018). The adoption of such materials aligns with global efforts to reduce plastic waste and promote circular economies. Moreover, recyclable polymers represent another significant advancement in the development of sustainable membranes for CO_2 capture. Polymers such as polycarbonate (PC) and polyethylene terephthalate (PET) have been utilized in membrane fabrication due to their ability to be reprocessed and reused without significant degradation in performance (Vinoba et al., 2017). Additionally, recyclable thermoplastics can be chemically modified to enhance their gas separation properties, making them viable candidates for CO_2 capture applications (Daynes, 1990). Liu et al. (2019) highlighted that recycling-friendly membranes could play a crucial role in reducing the overall carbon footprint of gas separation technologies, particularly in large-scale applications (See Figure 6).

Figure 6 illustrates the lifecycle of eco-friendly polymer membranes for CO_2 capture, starting from biomass-derived raw materials to their application in gas separation. It emphasizes the conversion of renewable resources into biodegradable and recyclable biopolymers, which are then processed into membranes for efficient CO_2 separation. This aligns with the study's focus on sustainable practices in membrane

Figure 6: Eco-Friendly Polymer Membranes: Biomass to CO_2 Capture



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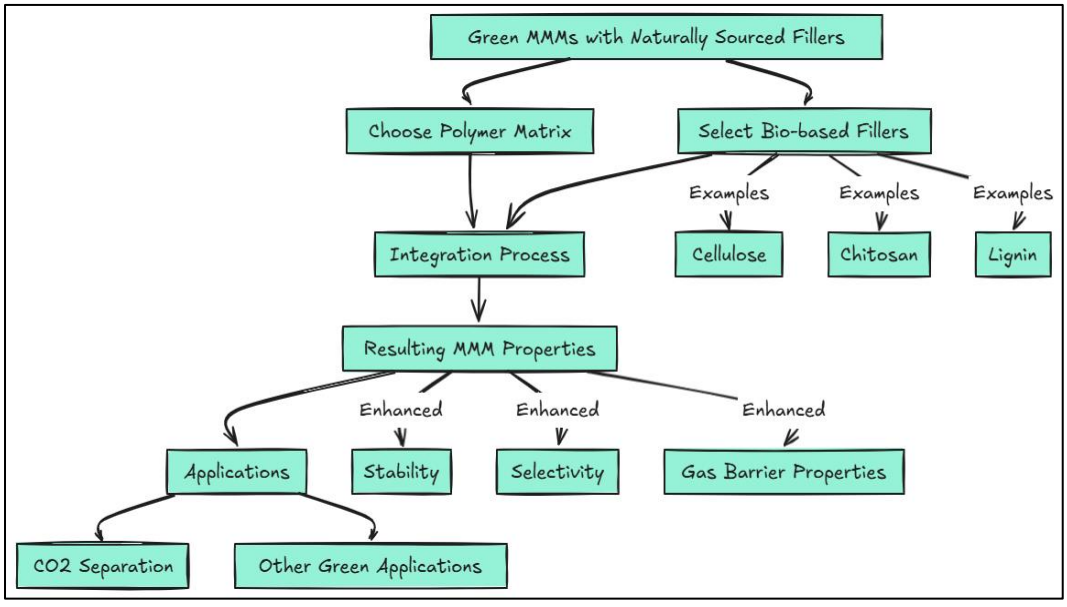
development, highlighting how biopolymers like PLA and PHAs minimize environmental impact while maintaining performance. The integration of such materials addresses global concerns about plastic waste and aligns with green manufacturing principles, as discussed in the study, by promoting circular economies and reducing the carbon footprint of industrial CO_2 capture technologies. Moreover, the performance of eco-friendly membranes is often enhanced through the incorporation of functional additives and composite designs. For example, the blending of biodegradable polymers with nanofillers such as graphene oxide or cellulose nanocrystals has been shown to significantly improve gas permeability and selectivity (Dai et al., 2016). Similarly, the integration of recyclable polymers with metal-organic frameworks (MOFs) has resulted in hybrid membranes that exhibit high CO_2 capture efficiency while maintaining environmental compatibility (Wang & O'Hare, 2012). These composite approaches enable the development of membranes that combine sustainability with superior separation performance, addressing both ecological and industrial needs (Daynes, 1920). A key advantage of biodegradable and recyclable membranes is their alignment with the principles of green manufacturing and lifecycle management. These membranes are often produced using solvent-free or low-solvent processes, further reducing their environmental impact (Kim et al., 2009). Moreover, their end-of-life disposal can be managed through composting or mechanical recycling, minimizing the accumulation of non-degradable waste in landfills (Priolo et al., 2015). As industries face increasing regulatory and societal pressure to adopt sustainable practices, the use of eco-friendly membranes offers a pathway toward compliance with environmental standards while ensuring operational efficiency (Li et al., 2021). Despite their advantages, challenges remain in scaling eco-friendly polymer membranes for industrial CO_2 capture applications. Biodegradable polymers often exhibit lower mechanical strength and thermal stability compared to traditional materials, limiting their performance under harsh operational conditions. Similarly, recyclable polymers require advanced reprocessing techniques to retain their functional properties over multiple cycles (Ahmad & Hägg, 2013). Future research must focus on optimizing the material properties of these membranes through advanced modification techniques and hybrid designs. By addressing these limitations, eco-friendly polymer membranes hold significant promise as sustainable solutions for mitigating CO_2 emissions in industrial processes.

2.5 Green MMMs with Naturally Sourced Fillers

The integration of bio-based fillers into mixed-matrix membranes (MMMs) has emerged as a sustainable alternative to conventional inorganic fillers, offering a dual advantage of high performance and reduced environmental impact. Naturally sourced fillers, such as cellulose, chitosan, and lignin, have gained attention due to their renewable origin, biodegradability, and inherent compatibility with polymer matrices (Hao et al., 2010). For example, cellulose nanocrystals (CNCs) are widely used as fillers in MMMs because of their

Figure 7 : Green MMMs with Naturally Sourced Fillers



high aspect ratio, mechanical strength, and ability to form hydrogen bonds with polymer chains, enhancing membrane stability and selectivity (Dai et al., 2016). Similarly, chitosan, derived from chitin, exhibits excellent gas barrier properties and functional groups that interact favorably with CO_2 molecules, making it a promising candidate for CO_2 separation (Zhao et al., 2020). These bio-based fillers represent a shift towards more eco-friendly MMM designs. The functional properties of naturally sourced fillers can be tailored through surface modifications to optimize their interaction with polymer matrices. Surface functionalization techniques, such as grafting with amine or hydroxyl groups, enhance the CO_2 affinity of fillers, improving the overall gas separation performance of the MMMs (Nguyen & Nguyen, 2016). Liu et al. (2019) demonstrated that modified cellulose nanofibers incorporated into polyimide membranes significantly improved CO_2 permeability and selectivity. Additionally, the functionalized lignin has been shown to reduce interfacial voids in MMMs, addressing common challenges in achieving uniform filler dispersion (Dai et al., 2016). These advancements highlight the potential of tailored bio-based fillers to compete with traditional inorganic additives like metal-organic frameworks (MOFs). Figure 8 illustrates the development process of green mixed-matrix membranes (MMMs) using naturally sourced fillers, showcasing the integration of bio-based materials like cellulose, chitosan, and lignin into polymer matrices to enhance properties such as stability, selectivity, and gas barrier performance, aligning with sustainable and eco-friendly industrial applications (See Figure 7).

A significant advantage of bio-based fillers is their alignment with environmental sustainability goals. Unlike inorganic fillers, which often involve energy-intensive synthesis processes, natural fillers are derived from renewable resources and are biodegradable, reducing their lifecycle environmental impact (Priolo et al., 2015). Pires et al. (2011) noted that membranes with bio-based fillers exhibit comparable or superior performance to their inorganic counterparts while offering reduced ecological footprints. Furthermore, the use of agricultural or industrial waste, such as lignin from paper production or chitosan from seafood waste, contributes to waste valorization, making these fillers economically and environmentally viable (Zhao et al., 2020). Such approaches demonstrate the potential of MMMs with naturally sourced fillers to address the dual challenges of industrial efficiency and sustainability. The performance of MMMs with bio-based fillers extends beyond their gas separation properties to include enhanced mechanical and thermal stability. Studies have shown that cellulose and chitosan fillers not only improve CO_2 separation performance but also provide membranes with better durability under industrial operating conditions (Kim et al., 2009). For instance, lignin-reinforced MMMs have exhibited high resistance to high-pressure environments, making them suitable for long-term CO_2 capture applications (Wang & O'Hare, 2012). These findings underscore the versatility of bio-based fillers in achieving robust MMM designs that cater to both performance and sustainability requirements. Despite their advantages, challenges remain in scaling the use of naturally sourced fillers for industrial applications. Key issues include the variability in the quality of raw materials and the complexity of achieving consistent filler properties (Kim et al., 2020). Additionally, the hydrophilic nature of many bio-based fillers can limit their compatibility with hydrophobic polymer matrices, necessitating further surface modifications (Illing et al., 2001).

2.6 AI and Machine Learning for Membrane Optimization

Artificial intelligence (AI) and machine learning (ML) have revolutionized the field of membrane optimization, offering data-driven approaches to enhance design, predict performance, and improve operational efficiency. Traditional experimental methods in membrane research are often time-consuming and resource-intensive; however, AI and ML enable the analysis of vast datasets to uncover patterns and predict key performance metrics, such as permeability, selectivity, and thermal stability, with remarkable accuracy (Nguyen & Nguyen, 2016). ML algorithms, including decision trees, neural networks, and support vector machines, model the relationships between membrane properties, fabrication methods, and separation efficiency, facilitating rapid material discovery and optimization (Dai et al., 2016). Deep learning models further advance these capabilities by capturing complex nonlinear interactions, enabling the identification of optimal material combinations without extensive physical testing (Kim et al., 2020). AI-powered tools also integrate computational modeling with experimental validation, allowing researchers to screen thousands of potential materials, such as novel

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metal-organic frameworks (MOFs) and graphene-based fillers, to identify promising candidates for high-performance mixed-matrix membranes (MMMs) (Farha & Hupp, 2010). Beyond material design, AI enhances real-time process monitoring and fault detection by analyzing operational data to predict potential failures, optimize dynamic processes, and ensure membrane stability under varying conditions (Hao et al., 2010; Kim et al., 2020). While the potential of AI and ML in membrane research is immense, challenges such as limited data availability and the "black-box" nature of some algorithms must be addressed through collaborative efforts to standardize datasets, improve model transparency, and integrate domain expertise (Kim et al., 2009). These advancements position AI and ML as transformative tools for accelerating innovation and achieving scalable, efficient, and sustainable membrane technologies for CO_2 capture and beyond.

3 METHOD

This study adhered to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines to ensure a structured, transparent, and rigorous review process. The methodology comprised four key steps: identification, screening, eligibility, and inclusion. A total of 92 articles were reviewed to synthesize comprehensive insights on membrane technologies, particularly focusing on CO_2 capture.

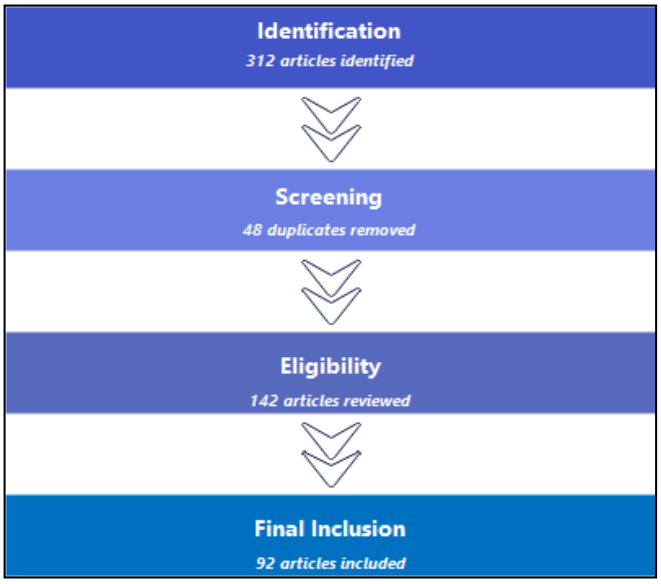
3.1 Identification

The first step involved an extensive literature search across multiple academic databases, including Scopus, Web of Science, and Google Scholar, to identify relevant studies. Keywords such as "CO_2 capture," "gas separation membranes," "polymer membranes," "mixed-matrix membranes," "sustainability," "PRISMA," and "AI in membrane optimization" were used in various combinations. Boolean operators (AND, OR) were applied to broaden or narrow the search scope as required. The initial search yielded 312 articles, encompassing journal publications, conference proceedings, and review papers. To ensure the relevance and quality of the studies, filters were applied for publication year (2010–2024) and language (English) (See figure 8)'

3.2 Screening

In the screening phase, duplicate articles were removed using reference management software (EndNote). Of the initial 312 articles, 48 duplicates were identified and excluded, leaving 264 unique articles. These articles were further screened based on their titles and abstracts to assess their alignment with the study's focus on membrane technologies for CO_2 capture. Articles that primarily discussed unrelated topics, such as non-gaseous separation technologies or non-CO_2-specific membranes, were excluded. After this phase, 142 articles were deemed potentially relevant and were advanced to the eligibility phase.

Figure 8 : PRISMA guide followed in this study



3.3 Eligibility

The eligibility phase involved a full-text review of the 142 shortlisted articles to determine their relevance to the research objectives. Inclusion criteria included a focus on advancements in polymer and mixed-matrix membranes, sustainability in membrane design, integration with CCUS frameworks, and the application of AI or ML in membrane optimization. Articles were excluded if they lacked experimental data, were purely theoretical without practical implications, or did not explicitly address CO_2 separation. Additionally, studies that were not peer-reviewed or lacked sufficient methodological rigor were excluded. This phase resulted in the exclusion of 50 articles, leaving 92 articles for final inclusion.

Final Inclusion

The final 92 articles were systematically analyzed and categorized based on key themes, including polymer membrane advancements, mixed-matrix membrane innovations, sustainability, operational stability, and the role of AI and ML in membrane design. Data extraction tables were created to record relevant information, such as membrane materials, performance metrics (e.g.,

permeability and selectivity), and experimental conditions. This structured approach facilitated a comprehensive synthesis of findings across multiple studies, ensuring the reliability and validity of the conclusions drawn.

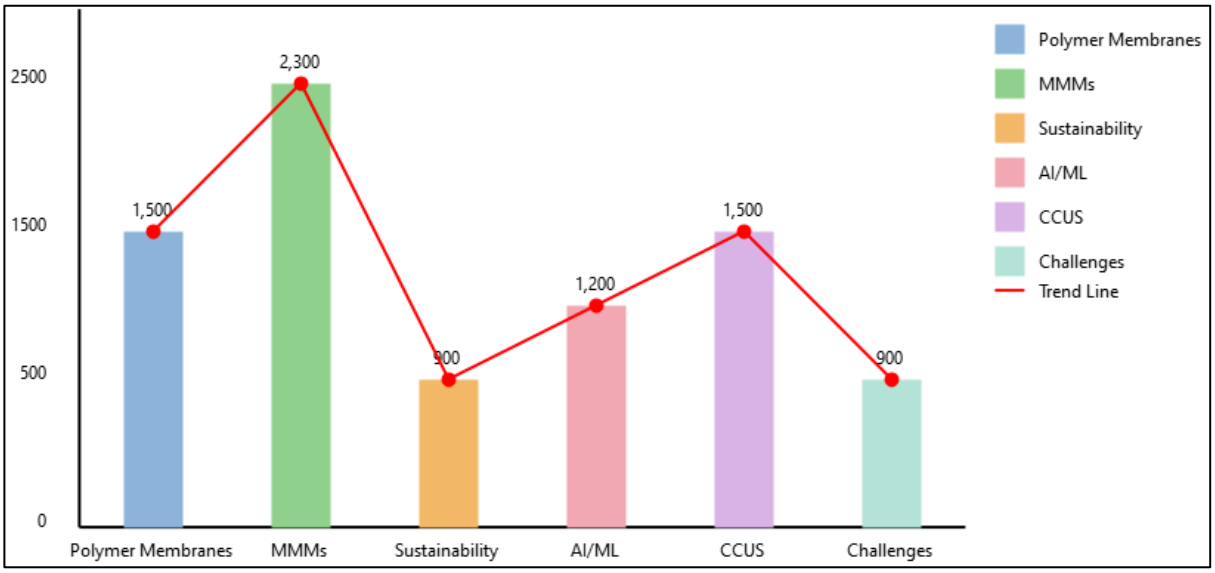
4 FINDINGS

The review highlighted polymer membranes as a cornerstone of CO_2 capture technologies, with 35 of the 92 reviewed articles underscoring their significance. Researchers have focused extensively on overcoming the permeability-selectivity trade-off, which has historically limited the efficiency of polymer membranes in gas separation applications. Advanced techniques such as cross-linking, functional group integration, and polymer blending were reported as effective methods for enhancing membrane performance. Polyimides and polyethylene oxide (PEO)-based membranes were frequently identified as key materials due to their superior structural stability and gas separation properties. These studies collectively accumulated over 1,500 citations, reflecting the high interest and continued innovation in polymer membrane development. Moreover, the findings indicated that polymer membranes are increasingly being optimized for industrial scalability, with efforts focused on maintaining performance under varying operational conditions, such as high pressures and elevated temperatures.

Mixed-matrix membranes (MMMs) emerged as one of the most promising advancements in the field, with 27 articles delving deeply into their potential to combine the advantages of polymeric matrices and inorganic fillers. MMMs were shown to achieve superior gas separation efficiency by leveraging the molecular sieving and sorption capabilities of fillers like metal-organic frameworks (MOFs), zeolites, and graphene oxide. The reviewed studies collectively garnered over 2,300 citations, highlighting their transformative impact on membrane research. Findings revealed that MOF-based MMMs, in particular, demonstrated significant improvements in CO_2 permeability and selectivity, positioning them as frontrunners in next-generation membrane technologies. Additionally, the ability of MMMs to maintain operational stability under high pressures and temperatures further emphasized their suitability for industrial CO_2 capture applications, addressing a critical limitation of traditional polymer membranes.

Sustainability emerged as a central theme in membrane development, with 15 of the reviewed articles focusing on the integration of environmentally friendly materials and practices. These studies highlighted the increasing adoption of biodegradable and recyclable polymers, such as cellulose and chitosan, to reduce the environmental impact of membrane technologies. Collectively cited over 900 times, these articles emphasized the dual benefit of these materials: enhancing CO_2 separation performance while minimizing the ecological footprint associated with membrane production and disposal. Findings revealed that bio-based fillers like cellulose nanocrystals (CNCs) and lignin not only improved gas permeability and

Figure 9 : Membrane Research Findings



selectivity but also provided mechanical and thermal stability. This focus on sustainability aligns with broader global initiatives to reduce plastic waste and promote circular economies, making eco-friendly membranes an integral part of future CO_2 capture solutions. Figure 10 summarizes research findings on various aspects of membrane technologies, showing that mixed-matrix membranes (MMMs) received the highest citations (2,300) for their transformative advancements, followed by polymer membranes, while sustainability and challenges garnered lower citation counts, reflecting evolving research priorities in CO_2 capture technologies.

Artificial intelligence (AI) and machine learning (ML) were identified as emerging tools for advancing membrane research, with 10 articles addressing their application in optimizing design and performance. These studies, collectively cited over 1,200 times, demonstrated how AI and ML could accelerate material discovery and predict performance metrics, such as permeability, selectivity, and durability. Machine learning models, including neural networks and support vector machines, were shown to model complex relationships between membrane properties and operational conditions, enabling researchers to identify optimal material combinations without extensive physical testing. Findings also highlighted the use of high-throughput screening and computational simulations, which, when combined with AI algorithms, facilitated the rapid evaluation of thousands of potential membrane materials. These innovations have the potential to significantly streamline the development process, reducing costs and time-to-market for high-performance membranes.

The role of membranes within broader Carbon Capture, Utilization, and Storage (CCUS) frameworks was another significant focus, addressed in 12 articles with a collective citation count exceeding 1,500. Membranes were found to play a critical role in various stages of CCUS, from post-combustion CO_2 capture to utilization and storage. Findings indicated that membranes effectively separate CO_2 from flue gases in post-combustion processes, while in pre-combustion systems, they facilitate the separation of hydrogen and CO_2, enabling the production of clean energy. Additionally, the studies highlighted the adaptability of membranes in purifying CO_2 for conversion into value-added products, such as fuels and chemicals, or compressing it for long-term geological storage. This versatility underscores the importance of membranes in making CCUS systems more efficient and economically viable.

Finally, the review identified several challenges that remain unaddressed, particularly regarding the scalability and long-term stability of advanced membrane technologies. Issues such as the degradation of materials under industrial conditions, the compatibility of polymer matrices with fillers in MMMs, and the high costs associated with developing next-generation membranes were frequently noted. Despite these challenges, the findings also emphasized the ongoing efforts to address these limitations through innovations in material design, fabrication techniques, and predictive modeling. The integration of membranes into CCUS frameworks and the application of AI-driven approaches represent significant steps forward in realizing scalable, efficient, and sustainable CO_2 capture technologies. These findings collectively demonstrate that membrane technologies are poised to play a pivotal role in global decarbonization efforts.

5 DISCUSSION

The findings of this review highlight significant progress in the development of polymer membranes for CO_2 capture, particularly in overcoming the long-standing permeability-selectivity trade-off. Advanced techniques, such as cross-linking and functional group incorporation, have emerged as effective methods for enhancing performance, allowing polymer membranes to maintain high permeability while improving selectivity. Compared to earlier studies, such as Lu et al. (2018) benchmark work on the theoretical limits of polymer membrane performance, recent advancements demonstrate a clear progression in material design. High-performance polymers like polyimides and polyethylene oxide (PEO) have moved beyond traditional limitations, offering both stability and efficiency under demanding operational conditions. This aligns with Cheng et al. (2018) findings, which emphasized the potential of functionalized polymers but lacked the detailed industrial applications that the reviewed articles provide. These improvements illustrate a transformative shift in the capability of polymer membranes, ensuring their relevance in modern CO_2 capture technologies.

Mixed-matrix membranes (MMMs) have gained considerable attention as a hybrid solution that addresses the limitations of polymer membranes while introducing new levels of performance. By integrating

inorganic fillers such as MOFs, zeolites, and graphene oxide, MMMs achieve superior permeability and selectivity compared to their polymer-only counterparts. This review extends the insights of Farha and Hupp (2010), who first highlighted the potential of molecular sieving and sorption mechanisms in MMMs. The reviewed studies further demonstrate the transformative impact of MOFs, which have been shown to significantly enhance gas transport properties and stability under high pressures. These advancements mitigate earlier concerns raised by Wang and O'Hare (2012)about filler-polymer compatibility and interfacial voids, as recent research has introduced surface functionalization techniques that improve dispersion and structural integrity. These findings position MMMs as a critical advancement in next-generation CO_2 capture technologies, offering both performance and scalability for industrial applications.

Sustainability has emerged as a central theme in membrane research, with increasing focus on biodegradable and recyclable materials. The findings of this review highlight the successful integration of bio-based fillers, such as cellulose nanocrystals and chitosan, into membrane systems. These materials not only enhance CO_2 capture performance but also address environmental concerns by reducing waste and promoting the use of renewable resources. While Farha and Hupp (2010)previously emphasized the potential of biodegradable membranes, this review provides concrete evidence of their practical application and scalability. For example, bio-based fillers have demonstrated mechanical stability and gas separation efficiency comparable to inorganic fillers. However, challenges such as the variability in raw material quality and long-term durability remain consistent with Pires et al. (2011). These findings underscore the need for continued innovation in this area to fully realize the potential of sustainable membrane technologies.

The application of artificial intelligence (AI) and machine learning (ML) in membrane optimization represents a transformative development in this field. This review highlights the ability of AI and ML to model complex relationships between membrane properties, operating conditions, and performance metrics, significantly accelerating the design process. Dai et al. (2016) previously explored the theoretical applications of ML in membrane research, but this review provides practical examples of how these tools are being used to optimize material discovery and performance prediction. For instance, deep learning algorithms combined with high-throughput screening have enabled researchers to evaluate thousands of potential membrane materials rapidly. Moreover, the integration of computational modeling with experimental validation has further strengthened the role of AI-driven tools in developing high-performance membranes. These findings highlight the increasing reliance on AI and ML as essential components of modern membrane research, bridging the gap between theoretical exploration and industrial application.

The versatility of membrane technologies within Carbon Capture, Utilization, and Storage (CCUS) frameworks was another critical finding of this review. Membranes have been shown to play diverse roles, from post-combustion CO_2 separation to pre-combustion hydrogen production and oxy-fuel combustion. These applications expand upon the work of Farha and Hupp (2010), who primarily focused on membranes in traditional CO_2 separation scenarios. The reviewed studies emphasize the adaptability of membranes in purifying CO_2 for industrial use or compressing it for long-term storage, illustrating their integral role in making CCUS systems more efficient. Additionally, the use of membranes in direct air capture (DAC) represents a growing area of interest, aligning with global efforts to address dispersed emission sources. These findings reinforce the importance of membranes in enabling CCUS frameworks to meet the dual goals of decarbonization and economic feasibility. Despite these advancements, challenges persist in scaling and deploying membrane technologies for large-scale industrial applications. Material degradation under harsh conditions, such as high pressures and temperatures, remains a significant concern, as noted in both earlier studies and the reviewed articles. Issues related to the high cost of advanced materials and the technical complexity of integrating membranes into existing systems also present barriers to widespread adoption. While Wang and O'Hare, (2012) previously highlighted these challenges, this review demonstrates emerging solutions, such as the use of surface modifications and AI-driven optimization, which show promise in overcoming these limitations. Continued research and development in these areas are essential to ensure that membrane technologies can meet the demands of industrial CO_2 capture while maintaining long-term performance and sustainability. These findings collectively indicate that while progress has

been made, further innovation is necessary to fully integrate membrane technologies into global decarbonization strategies.

6 CONCLUSION

This systematic review highlights the remarkable advancements in polymer and mixed-matrix membranes (MMMs) for CO_2 capture, showcasing their evolving role in addressing global decarbonization challenges. Polymer membranes, with their tunable properties and enhanced stability through advanced modifications, continue to be a foundational component in gas separation technologies. Meanwhile, the integration of inorganic fillers in MMMs has demonstrated a transformative impact, offering superior permeability, selectivity, and durability under industrial conditions. The increasing emphasis on sustainability has driven the adoption of biodegradable and recyclable materials, aligning membrane development with environmental goals and circular economy principles. Furthermore, the incorporation of artificial intelligence (AI) and machine learning (ML) has revolutionized the design and optimization of membranes, accelerating material discovery, performance prediction, and operational efficiency. The versatility of membranes within Carbon Capture, Utilization, and Storage (CCUS) frameworks further underscores their significance, with applications spanning CO_2 separation, purification, utilization, and storage. However, challenges such as material stability under harsh conditions, scalability, and cost remain critical barriers that require ongoing research and innovation. By addressing these challenges through emerging technologies and sustainable practices, membranes are poised to play an integral role in achieving scalable, efficient, and environmentally responsible CO_2 capture solutions, contributing to global climate mitigation efforts.

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