Asian Journal of Environmental Research
E-ISSN: 3047-4930
DOI: https://doi.org/10.69930/ajer.v3i2.783
Review Article

ScienceTech. Group

Vol. 3 (2), 2026
Page: 147-167

Biopolymer-Based Functional Membranes for CO_2 Capture: Fundamentals, Sustainable Material Design, and Emerging Composite Strategies

Rini Perdana

Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Negeri Makassar, Indonesia
*Email (corresponding author): rini.perdana@unm.ac.d

Abstract. *The increasing concentration of atmospheric carbon dioxide (CO_2) has intensified the need for sustainable, energy-efficient, and scalable capture technologies. Membrane-based CO_2 capture has attracted considerable attention due to its compact configuration, continuous operation, and relatively low energy demand compared with conventional absorption processes. However, many polymeric membranes still face limitations related to the permeability–selectivity trade-off, poor humidity tolerance, weak mechanical stability, and limited CO_2 affinity. In this context, biopolymer-based functional membranes have emerged as promising platforms because of their renewable origin, processability, biodegradability, and tunable surface chemistry. This review discusses the fundamentals and recent progress of biopolymer-based membranes for CO_2 capture, focusing on the relationship between membrane composition, molecular interaction, and capture performance. Key mechanisms, including physisorption, chemisorption, carbamate and bicarbonate formation, and facilitated transport, are highlighted to explain CO_2 uptake and selective transport in functional membrane systems. Special attention is given to cellulose acetate, chitosan, poly(vinyl alcohol)-based matrices, nanocellulose-reinforced membranes, and amine-functionalized composite strategies. The roles of porous fillers, amine immobilization, interfacial compatibility, water stability, and regeneration behavior are critically discussed. Although biopolymer-based membranes offer strong potential for sustainable CO_2 capture, challenges remain in long-term cyclic stability, humid mixed-gas operation, filler dispersion, and scale-up fabrication. The development of sustainable, low-cost, and regenerable CO_2 capture membranes can contribute directly to mitigating climate change, reducing atmospheric greenhouse gas concentrations, supporting net-zero emission targets, and advancing SDG 13 (Climate Action) and SDG 7 (Affordable and Clean Energy).*

Keywords: *CO_2 Capture; Biopolymer Membrane; Nanocellulose; Composite Membrane; Sustainable Materials*

1. Introduction

Carbon dioxide (CO_2) capture has become one of the most urgent research priorities in environmental chemistry, materials science, and sustainable engineering due to the continuous increase of anthropogenic greenhouse gas emissions from fossil fuel combustion, industrial processes, power generation, and hydrogen production (1). The accumulation of CO_2 in the atmosphere has intensified climate change, global warming, and environmental degradation, thereby demanding efficient, scalable, and environmentally benign capture technologies. Conventional CO_2 capture processes, particularly amine-based absorption, have demonstrated high capture efficiency and industrial maturity (2). However, their large-scale implementation remains constrained by high thermal regeneration energy, solvent

Received: 25 May 2026 / Accepted: 5 June 2026 / Available online: 8 June 2026

degradation, corrosion, operational complexity, and secondary environmental impacts. These limitations have stimulated extensive research into solid adsorbents and membrane-based systems as alternative technologies for low-energy and sustainable CO_2 capture (3).

Membrane technology has emerged as a promising platform for CO_2 separation because of its compact design, continuous operation, modular configuration, relatively low energy requirement, and ease of integration with existing industrial processes. Compared with conventional absorption, pressure swing adsorption, or cryogenic separation, membrane systems can operate without phase change and can potentially reduce operational complexity. (4). Nevertheless, the performance of polymeric membranes is often limited by the permeability–selectivity trade-off, physical ageing, plasticization, swelling, poor mechanical durability, and moisture sensitivity. In practical flue gas environments, membranes are exposed not only to CO_2 and N_2 but also to water vapor, oxygen, acidic gases, and particulate matter, which can significantly alter gas transport behavior. Therefore, the development of advanced functional membranes that combine high CO_2 affinity, structural stability, humidity tolerance, and sustainable material design is essential for moving membrane-based CO_2 capture from laboratory-scale studies toward practical deployment. (5).

Recent advances in CO_2 capture membranes have increasingly shifted from single-component polymer films toward composite and mixed-matrix membrane architectures. These systems incorporate porous fillers, functional groups, ionic species, or nanostructured reinforcements into polymer matrices to improve CO_2 adsorption, diffusion selectivity, and mechanical integrity. Metal-organic frameworks (MOFs), covalent organic frameworks (COFs), porous carbon, zeolite, ionic liquids, and amine-functionalized polymers have all been investigated as functional components. (6). For example, defect-engineered Zr-MOF nanocrystals incorporated into polyetherimide membranes have been shown to improve CO_2/CH_4 separation by enhancing CO_2 adsorption and interfacial compatibility, with reported CO_2 permeability of approximately 400–440 Barrer and CO_2/CH_4 selectivity of about 33–47 for optimised UiO-66-based membranes (7). However, although many advanced fillers provide excellent separation performance, some of them still face challenges associated with complex synthesis, high production cost, limited scalability, interfacial defects, and stability under humid or mixed-gas conditions. (8).

In this context, sustainable functional membranes based on biopolymers, nanocellulose, zeolite, and amine-functionalized composite systems have attracted increasing attention. Biopolymers such as cellulose acetate, chitosan, alginate, and poly(vinyl alcohol)-based systems offer attractive advantages, including renewability, biodegradability, abundance, processability, and the presence of reactive functional groups such as hydroxyl, amino, and carboxyl groups (9). These properties enable chemical modification, crosslinking, and hybridization with inorganic or organic fillers. However, pristine biopolymer membranes often exhibit limited CO_2 affinity, insufficient mechanical strength, and poor resistance to water-induced swelling or leaching. Therefore, rational composite design is required to transform sustainable biopolymers into high-performance CO_2 capture membranes (10).

Nanocellulose has emerged as a particularly important reinforcing and functional component in sustainable membrane systems (11). Its high aspect ratio, abundant hydroxyl groups, tunable surface chemistry, mechanical strength, and renewable origin make it suitable for constructing robust membrane networks. More importantly, chemically modified nanocellulose can act not only as a mechanical reinforcement but also as an interfacial bridge for retaining CO_2-philic species such as polyethyleneimine (PEI) (12). A recent study on

cellulose nanofiber–polyethyleneimine functionalized membranes demonstrated that oxalic acid-treated cellulose nanofibers could improve PEI retention, membrane strength, and CO_2 adsorption performance through ammonium–carboxylate interactions. The best membrane, containing 4 wt% PEI and cellulose nanofibers pretreated with 75 wt% oxalic acid, achieved CO_2 adsorption capacities above 4 mmol g^{-1} and maintained reusability over five adsorption–desorption cycles (13). This finding highlights the potential of nanocellulose as a key material for designing sustainable amine-functionalized membranes with improved durability.

Zeolite and natural zeolite represent another important class of sustainable or semi-sustainable porous fillers for CO_2 capture membranes. Zeolites possess ordered microporous structures, high thermal stability, ion-exchange capacity, and relatively low cost compared with many MOF and COF materials (14). Their aluminosilicate frameworks and exchangeable cations can promote electrostatic interactions with CO_2, while their rigid porous structure may provide additional diffusion pathways in mixed-matrix membranes (15). Nevertheless, zeolite-based systems also present limitations, including reduced performance under humid conditions, limited flexibility for functional modification, and potential filler–polymer incompatibility. Review literature on COF-based CO_2 adsorbents notes that zeolites are thermally stable and low-cost but often suffer from performance loss in humid environments and have more limited functional tunability than advanced porous frameworks (16). Thus, the integration of zeolite with biopolymers, nanocellulose, and amine groups provides a promising route to balance cost, stability, functionality, and CO_2 affinity.

Amine functionalization remains one of the most effective strategies for enhancing CO_2 capture because CO_2, as an acidic gas, can interact reversibly with basic amine sites. Primary, secondary, and tertiary amines can promote CO_2 adsorption through chemisorption, carbamate formation, bicarbonate formation under humid conditions, and facilitated transport (17). Polyethyleneimine is frequently used due to its high amine density and strong CO_2 affinity, but its practical use in membranes is limited by leaching, swelling, and instability in the presence of water. This limitation makes amine immobilization a central design challenge. Strategies such as ionic interaction with carboxylated nanocellulose, covalent grafting, crosslinking, amine-functionalized zeolite, and polymer–filler interfacial engineering are therefore essential for improving the durability of amine-based composite membranes. In addition, amine-functionalized systems must be evaluated not only under dry CO_2 adsorption conditions but also under humid and mixed-gas environments to represent realistic operating conditions (18).

Humidity is one of the most critical yet complex factors affecting CO_2 capture membranes. Water vapor may negatively affect membrane performance by occupying adsorption sites, forming water clusters, blocking micropores, inducing swelling, or causing leaching of active species. On the other hand, controlled humidity can enhance CO_2 capture in amine-containing membranes by facilitating bicarbonate formation and improving amine utilization (19). Recent work on humidity-tolerant CO_2/N_2 membranes demonstrated that water can play a dual role: excessive water causes pore blockage and competitive adsorption, whereas properly engineered water transport pathways can assist CO_2 migration through H_2O–CO_2 cluster formation. In that study, magneto-responsive channels in a PIM-1-based membrane achieved CO_2 permeability of 6355 Barrer and a CO_2/N_2 separation factor of 82.53 under saturated humidity, with 300 h continuous stability(20). Although this system is more complex than most sustainable membranes, it provides an important conceptual basis for

designing humidity-tolerant membranes that do not simply resist water but strategically regulate water–CO_2 interactions.

Despite rapid progress, several critical gaps remain in the development of sustainable functional membranes for CO_2 capture. First, many studies still focus on single-gas adsorption or ideal gas mixtures, whereas practical applications require validation under humid, mixed-gas, and impurity-containing conditions. Second, the long-term stability of amine-functionalized membranes is often insufficiently evaluated, with many studies reporting only a limited number of adsorption–desorption cycles. Third, the interface between polymer matrices, nanocellulose networks, porous fillers, and amine sites remains a major determinant of membrane performance, yet it is not always systematically optimized. Fourth, while advanced MOF, COF, ionic liquid, and engineered polymer systems have achieved impressive performance, their cost and scalability may limit broad implementation. These gaps create an opportunity for sustainable composite systems based on locally available biopolymers, nanocellulose, natural zeolite, and immobilized amine groups (21,22).

Therefore, this review aims to critically discuss recent progress in sustainable functional membranes for CO_2 capture, with a particular focus on biopolymer-based matrices, nanocellulose-reinforced structures, zeolite and natural zeolite fillers, and amine-functionalized composite systems. The review first summarizes the fundamental mechanisms of CO_2 capture in functional membranes, including physisorption, chemisorption, carbamate/bicarbonate formation, and facilitated transport. It then examines the roles of biopolymers, nanocellulose, zeolite, and amine groups in improving CO_2 affinity, mechanical stability, interfacial compatibility, humidity tolerance, and regeneration performance. Finally, this review identifies current challenges and future research directions toward low-cost, regenerable, humidity-tolerant, and scalable CO_2 capture membranes. By integrating sustainability, interfacial engineering, and realistic operating considerations, this review provides a framework for the rational design of next-generation functional membranes for carbon capture applications. In a broader environmental context, advancing sustainable and low-cost CO_2 capture membranes is expected to contribute to climate change mitigation, reduction of atmospheric greenhouse gas concentrations, transition toward net-zero emissions, and the achievement of SDG 13 (Climate Action) and SDG 7 (Affordable and Clean Energy).

2. Fundamentals of CO_2 Capture Using Functional Membranes

Functional membranes for CO_2 capture operate through the integration of molecular transport, adsorption, and selective interaction between CO_2 and active sites distributed within the membrane matrix. Unlike conventional dense polymer membranes that rely primarily on the solution–diffusion mechanism, functional composite membranes are designed to combine several capture pathways, including physisorption in porous domains, chemisorption at reactive functional groups, facilitated transport through reversible carrier interactions, and diffusion through engineered free-volume or filler-derived channels (23). This combination is particularly important for sustainable composite membranes based on biopolymers, nanocellulose, zeolite, and amine-functionalized components, where the membrane is expected not only to separate CO_2 from other gases but also to adsorb, retain, transport, and release CO_2 under realistic operating conditions (24).

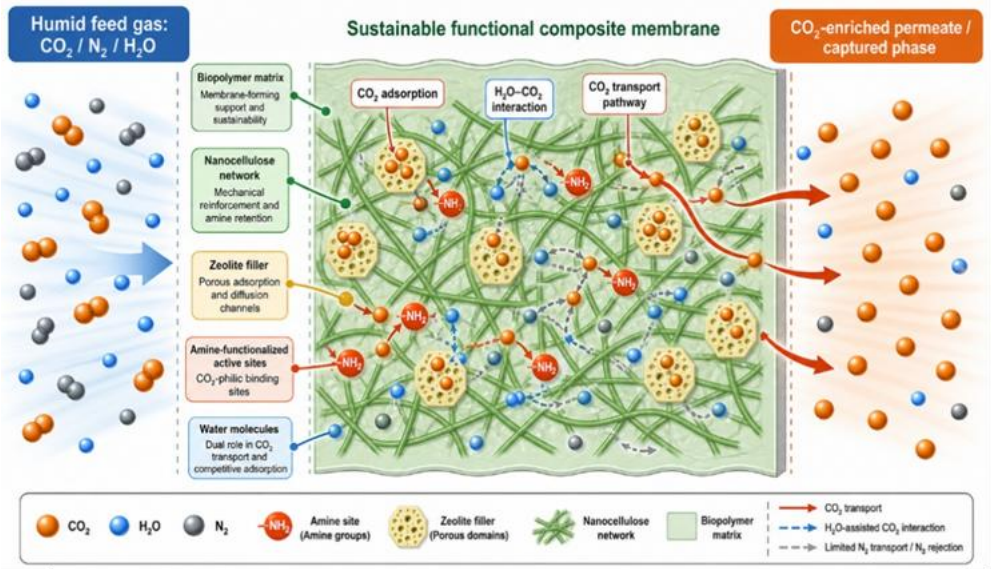

Figure 1 Schematic Overview of Sustainable Functional Membranes for CO_2 Capture

Figure 1. Schematic overview of sustainable functional membranes for CO_2 capture. The membrane architecture integrates a biopolymer matrix, nanocellulose network, zeolite or porous filler domains, and amine-functionalized active sites. The biopolymer matrix provides membrane-forming ability and sustainability, nanocellulose contributes mechanical reinforcement and amine retention, zeolite fillers provide porous adsorption and diffusion pathways, and amine groups serve as CO_2-philic binding sites. Water vapor may either assist CO_2 transport through bicarbonate formation and H_2O–CO_2 interactions or reduce performance through competitive adsorption and pore blocking (9).

2.1 CO_2 transport in membrane systems

Gas transport through polymeric and composite membranes is commonly explained by the solution–diffusion model. In this mechanism, gas molecules first dissolve into the membrane surface, diffuse through the membrane phase, and desorb at the permeate side. The overall permeability is therefore governed by both gas solubility and diffusivity. CO_2 generally exhibits higher solubility than N_2 or H_2 because of its larger condensability and quadrupole moment, while its kinetic diameter allows transport through many polymeric and microporous domains (25,26). However, selectivity does not depend only on molecular size; it is also influenced by gas–membrane affinity, polymer chain mobility, filler distribution, free-volume structure, and the presence of specific interaction sites. Studies on MOF–polymer mixed-matrix membranes show that solubility selectivity and diffusion selectivity jointly determine final separation performance, and that competitive adsorption in mixed-gas systems can weaken the apparent selectivity compared with pure-gas conditions .

In composite membranes, the transport pathway becomes more complex because the gas molecules may pass through the polymer phase, filler pores, polymer–filler interfaces, or non-selective voids. Properly designed porous fillers can introduce additional transport channels and increase CO_2 adsorption, while poorly dispersed fillers can create defects that decrease selectivity (27). For example, MOF-based mixed-matrix membranes may exhibit improved CO_2 permeability when porous fillers provide additional gas pathways, but excessive filler loading can lead to aggregation and nonselective void formation, thereby

reducing selective transport. Therefore, the fundamental design of CO_2 capture membranes must balance sorption enhancement, diffusion control, filler dispersion, and interfacial compatibility (28).

2.2 Physisorption in porous and polar domains

Physisorption is one of the basic mechanisms of CO_2 uptake in functional membranes. It involves weak but reversible interactions such as van der Waals forces, electrostatic interactions, quadrupole interactions, and confinement effects within porous domains. In sustainable composite membranes, physisorption can occur on zeolite surfaces, activated carbon domains, nanocellulose interfaces, porous fillers, or defect sites within inorganic particles (29). Zeolite and other aluminosilicate fillers are particularly relevant because their microporous structure and exchangeable cations can enhance electrostatic interactions with CO_2. Similarly, MOF- or COF-type fillers provide tunable pore structures that can increase CO_2 affinity and create selective pathways (30).

The strength of physisorption is generally lower than chemisorption, but this can be advantageous for regeneration because CO_2 can be released with lower energy input. Physisorption-dominated membranes often show good reversibility and faster kinetics, although their performance may decrease under humid conditions when water competes with CO_2 for adsorption sites. This issue is important for zeolite- or MOF-containing membranes because polar surfaces and metal nodes may preferentially interact with water. Therefore, in sustainable membrane design, physisorption sites must be optimized not only for high CO_2 uptake but also for selectivity and moisture resistance (31).

2.3 Chemisorption on amine-functionalized sites

Chemisorption is a key mechanism in amine-functionalized membranes because CO_2 is an acidic molecule that can react reversibly with basic amine groups. Primary and secondary amines are commonly associated with carbamate formation, whereas tertiary amines and hydrated amine environments can promote bicarbonate formation. Polyethyleneimine (PEI), aminosilanes, amine-functionalized zeolite, chitosan, and amino-functionalized MOFs are widely used because they provide CO_2-philic binding sites. In nanocellulose–PEI membranes, PEI contributes a high density of amine groups, while modified nanocellulose can help retain PEI within the membrane matrix through ionic interactions, improving both CO_2 capture performance and structural stability (32).

The general carbamate pathway under relatively dry conditions can be represented as:

$$2RNH_2 + CO_2 \rightleftharpoons RNHOO^- + RNH_3^+$$

This pathway requires two amine sites per CO_2 molecule, which may limit amine utilization efficiency. In the presence of water, however, CO_2 can be converted into bicarbonate species through a hydrated pathway:

$$RNNH_2 + CO_2 + H_2O \rightleftharpoons RNH_3^+ + HCO_3^-$$

This bicarbonate route can improve the theoretical efficiency of amine utilization because fewer amine sites are needed per CO_2 molecule. For this reason, controlled humidity can be beneficial in amine-functionalized membranes. However, excess water may dilute or block active sites, induce swelling, and promote leaching of physically immobilised amines. Therefore, amine-functionalized membranes require careful control of amine loading, immobilisation strategy, hydrophilicity, and water stability (33).

2.4 Facilitated transport of CO_2

Facilitated transport refers to the selective movement of CO_2 through reversible interactions with carrier sites in the membrane. In amine-containing membranes, CO_2 can bind to amine groups, migrate through a sequence of adjacent carrier sites, and be released at the permeate side. This mechanism differs from simple diffusion because specific chemical interactions assist transport. The process can be described as a sequence of binding, migration, and release. When properly designed, facilitated transport can increase CO_2 selectivity over weakly interacting gases such as N_2, CH_4, or H_2 (34).

In composite membranes, facilitated transport may be enhanced by distributing amine groups along nanocellulose networks, zeolite surfaces, polymer chains, or porous filler interfaces. The presence of a continuous or semi-continuous network of CO_2-philic sites can reduce transport resistance and increase CO_2 mobility. However, if amine sites are isolated, overloaded, or blocked by water clusters, facilitated transport becomes less effective. Therefore, the spatial distribution of amine groups is as important as their total concentration. This is one reason why interfacial engineering is central to high-performance composite membranes (34).

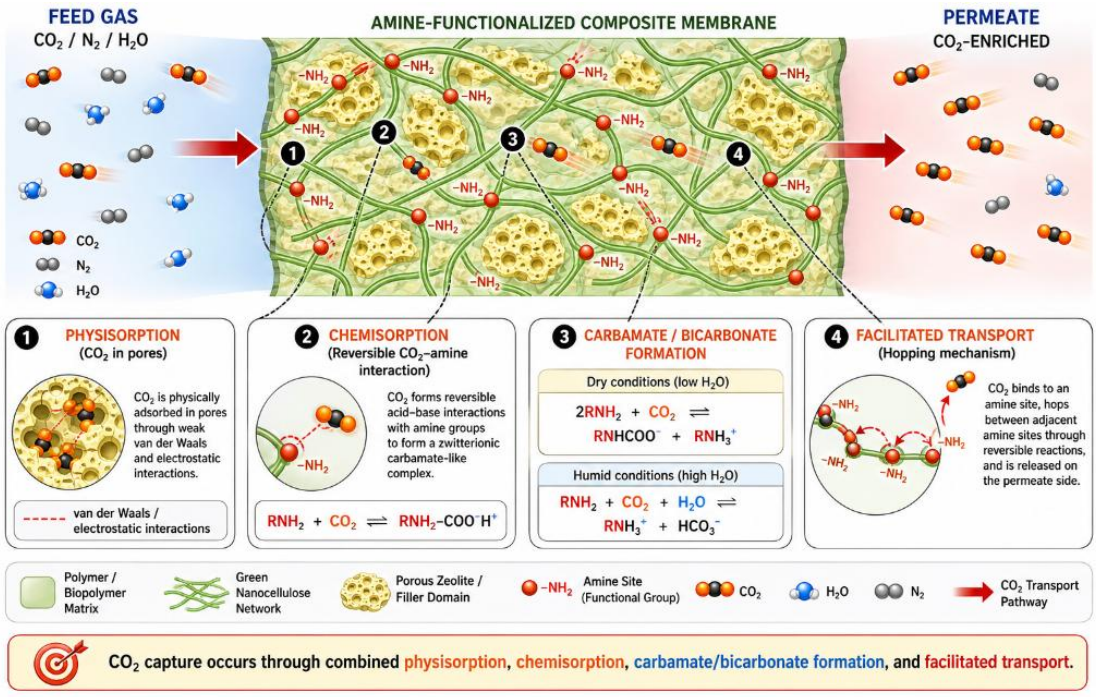

Figure 1. Mechanism of CO_2 Capture in Amine-Functionalized Composite Membranes

Figure 2. Mechanism of CO_2 capture in amine-functionalized composite membranes. CO_2 capture occurs through multiple pathways, including physisorption within porous filler domains, chemisorption through reversible interaction with amine sites, carbamate formation under dry conditions, bicarbonate formation under humid conditions, and facilitated transport through carrier-mediated binding, migration, and release. The schematic highlights the need for stable amine immobilization, accessible transport pathways, and controlled water–CO_2 interactions to improve CO_2 capture and separation performance(35).

2.5 Role of porous fillers and mixed-matrix architecture

The incorporation of porous fillers into polymer or biopolymer matrices is intended to overcome the intrinsic limitations of pure polymer membranes. Porous fillers may increase

free volume, provide selective adsorption sites, create fast diffusion channels, and improve mechanical or thermal stability. However, the actual performance of mixed-matrix membranes depends strongly on the quality of the polymer–filler interface. If the filler is well dispersed and compatible with the matrix, the membrane can show improved permeability and selectivity. If the filler aggregates or adheres poorly to the polymer, interfacial voids may form, allowing non-selective gas flow (36).

Several studies emphasise that interfacial defects, filler aggregation, and incomplete utilisation of porous transport pathways remain major limitations in mixed-matrix membranes. MOF-based membrane reviews describe that although MMMs are scalable and mechanically robust, their separation performance is often constrained by interfacial voids and poor filler–polymer affinity (37). DES-based membrane reviews also note that poor compatibility between inorganic fillers and polymer matrices can produce interfacial defects, while ternary designs using an additional compatibilizing component may improve filler dispersion and reduce void formation(38). These principles are directly relevant to biopolymer–nanocellulose–zeolite–amine membranes, in which nanocellulose can serve as a reinforcing network and interfacial bridge, while amine functionalization can provide CO_2-specific interaction sites.

Because the choice of filler strongly influences membrane cost, CO_2 affinity, humidity response, and polymer–filler compatibility, a comparative assessment of commonly used porous fillers is necessary. Table 1 summarises the relative advantages and limitations of zeolite, MOF, COF, and activated carbon as functional fillers in biopolymer-based CO_2 capture membranes. This comparison is intended to guide the selection of appropriate fillers according to application requirements, material availability, and desired membrane performance.

Table 1. Comparative characteristics of porous fillers for biopolymer-based CO_2 capture membranes

Filler type	Relative cost	Typical CO_2 capture role	Humidity stability	Compatibility with biopolymer matrices	Main advantages	Main limitations
Zeolite / natural zeolite	Low to moderate	Provides microporous adsorption sites, electrostatic interaction with CO_2, and diffusion channels	Moderate; performance may decline under high humidity due to competitive water adsorption	Good after particle size reduction, activation, or surface modification	Low cost, thermal stability, availability, ion-exchange capacity	Limited functional tunability, possible filler aggregation, moisture sensitivity
MOF	Moderate to high	Offers tunable pore size, high surface area, open metal sites, and functionaliz	Variable; some MOFs are moisture-sensitive, while Zr-based MOFs	Good if nanosized or surface-modified; poor compatibility may cause interfacial voids	Highly tunable structure, high CO_2 affinity, strong structure–	Higher cost, complex synthesis, scalability challenges

Filler type	Relative cost	Typical CO_2 capture role	Humidity stability	Compatibility with biopolymer matrices	Main advantages	Main limitations
		ed CO_2-philic sites	show better stability		performanc e control	
COF	Modera te to high	Provides ordered porous organic networks, tunable functional groups, and lightweight porous structures	Generally better chemical stability than many MOFs, depending on linkage chemistry	Potentially good with organic/biopol ymer matrices but often requires surface engineering	Covalent stability, low density, functional design flexibility	Complex synthesis, limited large-scale membrane studies
Activate d carbon / biochar	Low	Provides high surface area and physisorptio n sites; improves adsorption capacity and kinetics	Generally good, but hydrophilici ty depends on surface oxygen groups	Good in chitosan, PVA, cellulose-based systems when well dispersed	Low cost, scalable, chemically stable, biomass-derived options	Broad pore size distribution, lower selectivity unless functionaliz ed

Natural zeolite is frequently considered a promising low-cost filler for sustainable composite membranes because of its thermal stability, ion-exchange capacity, and microporous aluminosilicate framework. However, its contribution to CO_2 capture membranes depends strongly on particle size, surface activation, filler loading, and dispersion within the polymer or biopolymer matrix. Excessive filler loading may promote aggregation and non-selective void formation, while overly large particles can reduce membrane homogeneity and mechanical stability. Therefore, future studies on natural zeolite-containing biopolymer membranes should report zeolite particle size, loading percentage, activation method, surface functionalization, and their effects on adsorption capacity, permeability, selectivity, humidity tolerance, and mechanical performance

2.6 Humidity effects on CO_2 capture and transport

Humidity is one of the most important factors controlling the practical performance of CO_2 capture membranes. In real flue gas environments, membranes are exposed to $CO_2/N_2/H_2O$ mixtures, and water vapor may strongly alter membrane transport behavior. Under high-humidity conditions, water can occupy adsorption sites, form hydrogen-bonded

clusters, block micropores, induce polymer swelling, and reduce CO_2 solubility. In PIM-1-based membranes, humidification has been shown to decrease CO_2 and N_2 permeabilities as well as CO_2/N_2 separation factor, mainly because water molecules competitively adsorb on polar groups and form clusters that block transport channels (39).

However, water does not always play a negative role. In amine-containing membranes, moderate water content can facilitate bicarbonate formation and improve amine utilization. Water may also participate in H_2O–CO_2 cluster transport when the membrane structure is engineered to support rapid water migration. A recent humidity-tolerant CO_2/N_2 membrane study demonstrated that magneto-responsive channels can accelerate the transport of water and H_2O–CO_2 clusters, achieving a CO_2 permeability of 6355 Barrer and a CO_2/N_2 separation factor of 82.53 under saturated humidity (37). This finding highlights an important design principle: future sustainable membranes should not merely resist water but regulate water–CO_2 interactions to maintain or enhance CO_2 capture performance.

2.7 Key performance parameters

The evaluation of functional CO_2 capture membranes requires multiple performance indicators. For adsorptive membranes, the most common parameter is CO_2 adsorption capacity, usually expressed in mmol g^{-1}, cm^3 g^{-1}, or mg g^{-1}. For separation membranes, the key parameters are permeability, permeance, selectivity, separation factor, and flux. Permeability describes the intrinsic ability of a membrane material to transport a gas, whereas permeance is normalized by membrane thickness and is often used for thin-film composite membranes. Selectivity reflects the preference of one gas over another, such as CO_2/N_2, CO_2/CH_4, H_2/CO_2, or N_2/CO_2. (40).

Beyond these conventional parameters, practical evaluation must also include adsorption–desorption kinetics, cyclic stability, regeneration energy, mechanical strength, humidity tolerance, and mixed-gas performance. Reviews on integrated membrane–adsorption systems emphasize that comprehensive assessment should include capacity, selectivity, and cyclic stability because these parameters determine whether a material can operate in modular capture systems (41). In addition, membrane–adsorption integration studies show that membrane systems are often evaluated using CO_2/N_2 and CO_2/CH_4 selectivity as well as permeation flux, and that composite membranes may exceed the performance limits of conventional polymer membranes when multiphase interfaces are properly engineered (41).

2.8 Trade-off between permeability, selectivity, and stability

A central challenge in membrane-based CO_2 capture is the trade-off between permeability and selectivity. Highly permeable membranes often suffer from reduced selectivity because fast gas transport may also allow non-target gases to pass through. Conversely, highly selective membranes may have lower permeability due to restricted diffusion pathways. Functional composite membranes attempt to overcome this trade-off by combining CO_2-philic sites, porous fillers, optimized free volume, and selective diffusion channels. However, introducing fillers or functional groups may also produce new limitations, such as pore blockage, rigidified polymer layers, excessive swelling, or active-site saturation (42) .

Operational conditions can further complicate this trade-off. For example, temperature may increase gas diffusion but reduce CO_2 adsorption if the adsorption process

is exothermic. Pressure may enhance gas solubility but can also saturate adsorption sites or alter transport mechanisms. Mixed-gas testing is also critical because pure-gas selectivity may overestimate practical performance. A study on N_2-selective thin-film composite membranes showed that an optimized membrane achieved high N_2 permeance and N_2/CO_2 selectivity under pure-gas conditions, but mixed-gas performance declined over time due to competitive occupation of adsorption sites and reduced CO_2 partial pressure (43). This reinforces the need for evaluating sustainable CO_2 capture membranes under realistic feed compositions.

2.9 Implications for sustainable membrane design

The fundamental mechanisms discussed above indicate that sustainable functional membranes must be designed as integrated systems rather than simple polymer films. A biopolymer matrix provides processability and sustainability, nanocellulose offers reinforcement and interfacial anchoring, zeolite contributes porous adsorption and diffusion pathways, and amine groups provide CO_2-specific binding and facilitated transport. The effectiveness of this design depends on how well these components are integrated at the molecular and interfacial levels. Strong polymer–filler compatibility, stable amine immobilization, controlled hydrophilicity, and balanced pore structure are essential to achieve high CO_2 capacity, selectivity, humidity tolerance, and regeneration performance (44).

Therefore, the fundamentals of CO_2 capture in sustainable functional membranes can be summarized in four interconnected principles. First, CO_2 must be preferentially adsorbed through polar, porous, or amine-functionalized sites. Second, CO_2 must be transported selectively through continuous pathways without excessive resistance or non-selective defects. Third, water must be controlled so that it supports bicarbonate formation or water-assisted transport without causing pore blocking, swelling, or amine loss. Fourth, the membrane must maintain performance over repeated adsorption–desorption cycles and under mixed-gas conditions. These principles provide the basis for evaluating biopolymer-, nanocellulose-, zeolite-, and amine-based composite membranes in the following sections of this review (45).

3. Biopolymer-Based Membranes for CO_2 Capture

Biopolymer-based membranes have gained increasing attention as sustainable platforms for CO_2 capture because they combine renewable origin, processability, chemical functionality, and environmental compatibility. In contrast to many synthetic high-performance polymers or advanced porous frameworks, biopolymers are generally derived from abundant natural resources and contain reactive groups such as hydroxyl, amino, carboxyl, ether, and acetamide functionalities. These groups provide opportunities for chemical modification, crosslinking, filler incorporation, and amine immobilization. For CO_2 capture applications, the role of biopolymers is not limited to serving as passive membrane matrices; they can also participate in adsorption, facilitate interfacial compatibility, stabilize CO_2-philic species, and improve mechanical integrity. Therefore, biopolymer-based membranes represent an important direction in the development of sustainable functional membranes (46,47).

3.1 Rationale for using biopolymer membranes

The growing interest in biopolymer-based membranes is closely associated with the need to reduce the environmental burden of CO_2 capture materials. Conventional amine

absorption remains effective but is limited by high regeneration energy, corrosion, solvent degradation, and operational complexity. Membrane separation offers a lower-energy and compact alternative, yet many membrane materials still rely on petroleum-derived polymers or expensive porous fillers. Biopolymers provide a more sustainable basis for membrane fabrication because they are renewable, biodegradable or partially biodegradable, and chemically tunable. In addition, many biopolymers can be processed through relatively simple techniques such as solution casting, phase inversion, electrospinning, and crosslinking, making them attractive for scalable membrane development (48,49).

The main advantage of biopolymers lies in their intrinsic functionality. Cellulose derivatives contain abundant hydroxyl and ester groups, chitosan contains amino and hydroxyl groups, alginate contains carboxylate groups, and starch- or lignin-derived materials contain oxygen-rich moieties. These functionalities can interact with CO_2 directly through weak polar interactions or indirectly by anchoring amine carriers, porous fillers, ionic groups, or crosslinking agents. However, most pristine biopolymer membranes do not provide sufficient CO_2 affinity or gas separation performance by themselves. Their use in CO_2 capture therefore requires functionalization strategies, especially amine incorporation, nanocellulose reinforcement, zeolite filling, or hybridization with porous adsorbents (4,50).

3.2 Cellulose acetate and cellulose-derived membranes

Cellulose acetate is one of the most widely studied cellulose-derived polymers for membrane fabrication because it can form dense, asymmetric, and fibrous membranes through phase inversion or electrospinning. Its film-forming ability, availability, and relatively good mechanical properties make it suitable as a membrane matrix. However, cellulose acetate lacks a strong intrinsic affinity for CO_2, which limits its adsorption capacity and selectivity when used without functional additives. This limitation has encouraged the incorporation of CO_2-philic compounds such as polyethyleneimine, amine-functionalized fillers, ionic liquids, or nanocellulose-based components (10,51).

A representative study on cellulose acetate–polyethyleneimine membranes highlights both the potential and the limitations of cellulose-derived membranes. Polyethyleneimine provides a high density of amine groups and can interact with acidic CO_2 molecules, but it does not form mechanically robust membranes on its own and can be leached from the cellulose acetate matrix in the presence of water. The incorporation of oxalic-acid-treated cellulose nanofibers into the cellulose acetate/PEI system was proposed to overcome these limitations by improving PEI retention and mechanical strength through ammonium–carboxylate interactions. The optimized membrane containing 4 wt% PEI and cellulose nanofibers treated with 75 wt% oxalic acid achieved CO_2 adsorption capacities above 4 mmol g^{-1} and maintained performance over five adsorption–desorption cycles (13).

This finding is important for biopolymer membrane design because it shows that cellulose-derived membranes require a multifunctional architecture. Cellulose acetate provides processability and membrane-forming ability, PEI provides CO_2-reactive amine sites, and modified nanocellulose provides reinforcement and active-site retention (Hou et al., 2020). Such an approach shifts the function of cellulose-based membranes from passive supports to integrated CO_2 capture systems. It also demonstrates that sustainable membrane performance depends not only on the presence of amine groups but also on their stability within the membrane matrix (44).

3.3. Chitosan-based membranes

Chitosan is another important biopolymer for environmental membrane applications. It is derived from chitin and contains abundant amino and hydroxyl groups, making it attractive for pollutant adsorption, metal ion removal, and CO_2 capture. The amino groups in chitosan provide basic sites that may interact with acidic species, while the hydroxyl groups enable hydrogen bonding, chemical modification, and crosslinking. Compared with cellulose acetate, chitosan has a stronger intrinsic affinity for various pollutants because of its amino functionality (52). However, pristine chitosan membranes often suffer from poor mechanical stability, swelling in aqueous environments, and limited porosity. These drawbacks require crosslinking, blending, or composite formation (53).

A recent chitosan-based membrane study developed an activated carbon–aspartic acid crosslinked chitosan membrane for dual environmental remediation, targeting both Cr(VI) removal and CO_2 sequestration. The study emphasizes that chitosan is biodegradable and rich in amino and hydroxyl groups, while activated carbon contributes high surface area and porosity. Aspartic acid crosslinking stabilizes the composite structure and introduces additional active sites, thereby improving adsorption behavior (54). This example illustrates the broader potential of chitosan membranes as multifunctional environmental materials rather than single-target adsorbents.

For CO_2 capture, chitosan-based membranes can be designed through several strategies. First, chitosan can serve as an amine-containing matrix that interacts directly with CO_2. Second, its functional groups can anchor porous adsorbents such as zeolite, activated carbon, silica, or biochar. Third, crosslinking can improve water stability and reduce swelling. Fourth, chitosan can be blended with more stable polymers such as PVA, cellulose acetate, or polyethersulfone to improve mechanical and transport properties. These strategies make chitosan relevant to sustainable CO_2 capture membranes, especially when dual-function or multifunctional environmental remediation is desired (55,56).

3.4 PVA and other hydroxyl-rich biopolymer matrices

Poly(vinyl alcohol) is often used in sustainable membrane systems because it is hydrophilic, film-forming, chemically modifiable, and rich in hydroxyl groups. Although PVA is synthetic, it is frequently discussed together with biopolymer-based systems because of its water solubility, biocompatibility, and ability to form hydrogen-bonded networks with natural polymers such as cellulose, chitosan, starch, and nanocellulose. In CO_2 capture membranes, PVA can act as a flexible matrix for incorporating amine carriers, zeolite particles, nanocellulose, or other porous fillers. Its hydroxyl groups can participate in crosslinking reactions and improve compatibility with oxygen-rich fillers (57,58).

However, the hydrophilicity of PVA also creates challenges. Excessive water uptake can lead to swelling, decreased mechanical stability, and changes in gas transport pathways. Therefore, PVA-based CO_2 capture membranes usually require crosslinking or blending with more rigid components. Crosslinkers such as glutaraldehyde, citric acid, sulfosuccinic acid, or other multifunctional agents may be used to regulate swelling and enhance structural stability. When combined with nanocellulose and zeolite, PVA can form a reinforced composite matrix in which nanocellulose improves mechanical strength and zeolite provides adsorption sites or diffusion pathways. This makes PVA-based hybrid membranes highly relevant for the development of low-cost functional membranes using locally available materials (59,60).

Other biopolymer or bio-derived matrices, including alginate, starch, lignin, and cellulose derivatives, can also support CO_2 capture membrane development. Alginate provides carboxylate groups that may interact with amines or metal ions. Starch offers abundant hydroxyl groups and low cost, but requires modification to improve water stability. Lignin contains aromatic and oxygenated groups that can contribute to sorption and filler compatibility, although its structural heterogeneity can complicate membrane fabrication. Overall, these materials are promising but generally require composite design to achieve competitive CO_2 capture performance (61).

3.5 Functionalization strategies for biopolymer membranes

Because pristine biopolymer membranes often show limited CO_2 capture performance, functionalization is essential. The most common strategy is amine incorporation, either through physical blending, impregnation, grafting, or ionic interaction. PEI is widely used because of its high amine density, but it must be immobilized to prevent leaching. The cellulose nanofiber–PEI membrane study demonstrates that carboxylated nanocellulose can retain PEI through ammonium–carboxylate interactions, resulting in improved stability and CO_2 uptake (13). This strategy is particularly relevant for sustainable membranes because it avoids relying solely on physical entrapment of amines.

Another strategy is crosslinking. Crosslinking can reduce swelling, improve mechanical strength, stabilize the membrane network, and prevent loss of active components. In chitosan-based membranes, aspartic acid crosslinking was used to stabilize the membrane while contributing additional active sites (54). In cellulose- or PVA-based systems, crosslinking may also help regulate hydrophilicity and maintain membrane integrity under humid conditions. However, excessive crosslinking can reduce free volume and hinder gas diffusion, so optimization is required.

A third strategy is filler incorporation. Biopolymers can be combined with zeolite, activated carbon, silica, biochar, MOFs, or other porous materials to improve adsorption capacity and transport pathways. Activated carbon incorporation in chitosan membranes demonstrates how porous carbon can enhance adsorption capacity and kinetics (54). For sustainable CO_2 capture membranes, natural zeolite is especially attractive because it is low-cost, thermally stable, and porous. However, filler–polymer compatibility must be controlled to avoid aggregation and non-selective voids. Nanocellulose can play a useful role as a bridging or dispersing component because it can interact with both polymer matrices and fillers through hydrogen bonding or electrostatic interactions.

3.6 Water stability and humidity-related challenges

Water stability is one of the most critical challenges for biopolymer-based CO_2 capture membranes. Most biopolymers are hydrophilic, which can be beneficial for bicarbonate formation in amine-containing systems but problematic when water uptake becomes excessive. Water may induce swelling, reduce mechanical strength, block pores, compete with CO_2 for adsorption sites, or promote leaching of amine carriers. In cellulose acetate/PEI systems, PEI loss in the presence of water has been identified as a major limitation, particularly at higher PEI contents where electrostatic repulsion may promote outward diffusion of PEI chains (13).

The challenge is not simply to make biopolymer membranes hydrophobic, because controlled water content may improve CO_2 capture through bicarbonate pathways. Instead,

the goal is to regulate water–membrane interactions. A good biopolymer membrane should allow beneficial hydration near amine sites while preventing excessive swelling, pore blockage, and active-site leaching. This requires a balanced design involving crosslinking, amine immobilization, filler selection, and pore structure control. For example, carboxylated nanocellulose can help retain PEI, while zeolite or activated carbon can contribute structural and adsorptive domains. The optimal membrane architecture should therefore be hydrophilic enough to support CO_2–amine chemistry but stable enough to withstand humid gas streams (62).

3.7 Adsorption performance and regeneration behavior

Performance evaluation of biopolymer-based CO_2 capture membranes should include adsorption capacity, kinetics, selectivity, mechanical stability, and reusability. The cellulose nanofiber–PEI system provides an important benchmark, where the optimized membrane achieved CO_2 adsorption exceeding 4 mmol g^{-1} and retained its adsorption performance over five cycles without significant loss (13). This indicates that properly immobilized amine sites in a cellulose-based membrane can provide both capacity and cyclic stability.

For chitosan-based membranes, regeneration studies in pollutant adsorption systems provide useful insight into the durability of bio-based composites. The activated carbon–aspartic acid crosslinked chitosan membrane retained substantial adsorption capacity after repeated regeneration cycles, although a gradual decline was observed after five cycles (54). While this example focused primarily on Cr(VI) removal, it demonstrates that bio-based composite membranes can be regenerated and reused, which is essential for practical CO_2 capture applications.

Nevertheless, many biopolymer membrane studies still report limited cycling data. Five cycles are useful for initial proof of reusability, but they are insufficient for claiming industrial relevance. Future studies should evaluate 20–50 adsorption–desorption cycles, long-term humid exposure, mixed-gas testing, and mechanical durability after regeneration. This is especially important for amine-functionalized systems because repeated hydration, heating, or vacuum treatment may alter amine distribution, membrane morphology, and filler–polymer interfaces (63).

3.8 Advantages and limitations of biopolymer-based membranes

Biopolymer-based membranes offer several advantages for sustainable CO_2 capture. They are derived from renewable or bio-based resources, contain reactive functional groups, can be processed through relatively mild methods, and can be integrated with nanocellulose, zeolite, amines, and porous fillers. They also align with green chemistry principles and circular economy concepts, especially when derived from agricultural residues, marine biomass, or industrial biowaste. In addition, biopolymer membranes can be designed for multifunctional environmental remediation, such as simultaneous water pollutant removal and air pollutant capture (64).

However, their limitations are equally important. Pristine biopolymer membranes generally show modest CO_2 affinity, poor gas selectivity, sensitivity to water, and limited long-term stability. They may also suffer from batch-to-batch variability due to differences in biomass source, degree of deacetylation, molecular weight, crystallinity, or functional group density. From a membrane separation perspective, many biopolymer systems are still evaluated mainly as adsorptive membranes rather than true gas separation membranes.

Therefore, more data are needed on permeability, permeance, CO_2/N_2 selectivity, CO_2/CH_4 selectivity, and mixed-gas operation (65).

3.9 Outlook for biopolymer-based sustainable CO_2 capture membranes

The future of biopolymer-based CO_2 capture membranes lies in rational composite design. Biopolymers should be combined with nanocellulose for reinforcement and interfacial control, zeolite for porous adsorption and diffusion pathways, and immobilized amines for CO_2-specific binding. Such systems can potentially overcome the individual limitations of each component. For example, a PVA or cellulose acetate matrix can provide processability, nanocellulose can improve strength and amine retention, zeolite can enhance porosity and adsorption, and PEI or other amines can provide selective CO_2 interaction (66).

To advance this field, future studies should focus on four priorities. First, biopolymer membranes must be tested under humid and mixed-gas conditions rather than only under dry pure CO_2. Second, amine immobilization must be improved through ionic, covalent, or crosslinked networks to prevent leaching. Third, filler dispersion and interfacial compatibility must be optimized to avoid non-selective voids. Fourth, performance evaluation should include not only adsorption capacity but also permeability, selectivity, kinetics, regeneration stability, and mechanical durability. Addressing these priorities will allow biopolymer-based membranes to move from sustainable material concepts toward practical CO_2 capture technologies (67).

Conclusions

Biopolymer-based functional membranes offer a promising sustainable platform for CO_2 capture because they combine renewability, processability, chemical tunability, and environmental compatibility. Materials such as cellulose acetate, chitosan, PVA-based matrices, and nanocellulose can serve not only as membrane supports but also as active components when combined with amine groups, porous fillers, and crosslinking strategies. CO_2 capture in these membranes occurs through multiple mechanisms, including physisorption, chemisorption, carbamate/bicarbonate formation, and facilitated transport. Amine-functionalized components enhance CO_2 affinity, while nanocellulose and other biopolymer networks can improve mechanical stability, active-site retention, and interfacial compatibility. However, membrane performance is strongly influenced by humidity, filler dispersion, amine stability, and membrane morphology.

Despite their potential, several challenges remain, particularly limited long-term stability, amine leaching, swelling under humid conditions, and insufficient testing under realistic mixed-gas environments. Future research should focus on designing robust biopolymer-based composite membranes with controlled hydrophilicity, stable amine immobilization, strong polymer–filler interfaces, and scalable fabrication methods. Overall, biopolymer-based functional membranes represent a viable route toward low-cost, sustainable, and regenerable CO_2 capture systems. Their future development should integrate fundamental transport mechanisms, sustainable material design, and practical performance validation to support real-world carbon capture applications

Funding

This research received no external funding

Conflicts of Interest

The authors declare no conflict of interest.

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