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A review on recent advances of cellulose acetate membranes for gas separation

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This review thoroughly investigates the wide-ranging applications of cellulose-based materials, with a particular focus on their utility in gas separation processes. By focusing on cellulose acetate (CA), the review underscores its cost-effectiveness, robust mechanical attributes, and noteworthy CO_2 solubility, positioning it as a frontrunner among polymeric gas separation membranes. The synthesis techniques for CA membranes are meticulously examined, and the discourse extends to polymeric blend membranes, underscoring their distinct advantages in gas separation applications. The exploration of advancements in CA-based mixed matrix membranes, particularly the incorporation of nanomaterials, sheds light on the significant versatility and potential improvements offered by composite materials. Fabrication techniques demonstrate exceptional gas separation performance, with selectivity values reaching up to 70.9 for CO_2/CH_4 and 84.1 for CO_2/N_2. CA/PEG (polyethylene glycol) and CA/MOF (metal–organic frameworks) demonstrated exceptional selectivity in composite membranes with favorable permeability, surpassing other composite CA membranes. Their selectivity with good permeability lies well above all the synthesised cellulose. As challenges in experimental scale separation emerge, the review seamlessly transitions to molecular simulations, emphasizing their crucial role in understanding molecular interactions and overcoming scalability issues. The significance of the review lies in addressing environmental concerns, optimizing membrane compositions, understanding molecular interactions, and bridging knowledge gaps, offering guidance for the sustainable evolution of CA-based materials in gas separation technologies.

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1. Introduction

The escalating carbon dioxide (CO_2) emissions in recent years pose a grave environmental threat, primarily contributing to global warming. The concentration of CO_2 in the atmosphere is projected to surpass a critical threshold of 417 parts per million (ppm) by 2022, marking a staggering 50% increase from the 278 ppm recorded at the onset of the industrial era.[1,2] This surge is predominantly attributed to the widespread utilization of fossil fuels for energy production, underscoring the urgent need for effective strategies to mitigate CO_2 emissions.[3,4] The primary motivation for CO_2 separation from CH_4 in natural gas purification is not directly related to environmental remediation, but rather to improve the calorific value and quality of the natural gas product. The presence of CO_2 in natural gas can significantly reduce the calorific value and energy content of the fuel, making it less desirable for end-use applications. By selectively removing the CO_2 from the natural gas stream, the purity and energy density of the methane-rich natural gas can be enhanced, ensuring it meets the necessary specifications for various applications, such as residential, commercial, and industrial use.[5,6]

In contrast, the separation of CO_2 from flue gas streams is more directly tied to environmental concerns and the need to mitigate greenhouse gas emissions. Flue gas, which is the exhaust gas from the combustion of fossil fuels in power plants and industrial facilities, is a major source of anthropogenic CO_2 emissions. Capturing and separating the CO_2 from these flue gas streams can enable its sequestration or utilization, thereby reducing the overall carbon footprint and contributing to environmental sustainability.[7]

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Efficient separation techniques are essential in this context, driving the exploration of membrane-based CO_2/CH_4 separation technology. This approach has garnered significant attention due to its notable advantages, including high energy efficiency, minimal capital investment, operational reliability, and simplicity.[8] Cellulose acetate (CA) is a compelling material for gas separation membranes, offering a combination of cost-effectiveness, robust mechanical properties, superior fouling resistance, ease of processing, and high CO_2 solubility.[9] The intrinsic characteristics of CA, such as sustainability and mechanical stability, position it as a prominent candidate for fabricating polymeric gas separation membranes, presenting an eco-friendly alternative to traditional petrochemical-based materials.[10] The increasing need for gas separation membranes with enhanced performance is motivated by the prospect of substantially reducing energy consumption in chemical processing.[8]

Due to their notable chemical resistance and mechanical stability, CA membranes are extensively used in various gas separation applications, such as CO_2 capture, hydrogen recovery, and nitrogen generation.[11–13] CA possesses notable mechanical properties, including tensile strength and Young's modulus, which contribute to its reputation for mechanical strength compared to other polymers. Studies have reported that CA typically exhibits a tensile strength ranging from approximately 45 to 70 MPa, depending on factors such as the degree of acetylation and processing conditions. Moreover, Young's modulus of CA is usually in the range of 2 to 4 GPa. This range is comparable to polyimides (2–5 GPa) and polysulfones (2–3 GPa), which are known for their mechanical strength and rigidity. These mechanical characteristics position CA as a robust polymer, particularly in comparison to other polymers commonly used in similar applications.[14–16]

CA membranes for gas separation use various synthesis methodologies to enhance their performance and selectivity. Notably, the grafting of imidazole ionic liquid onto CA films demonstrated excellent CO_2 permeation properties, showcasing a significant increase in selectivity compared to pure CA. Gamma irradiation of CA–polyethylene glycol composite membranes also proved effective, enhancing selectivity for CO_2/CH_4 gas separation. Additionally, the modification of CA films with fluorine treatment showcased a reduction in permeability, indicating potential applications in controlling gas transport. The use of room-temperature ionic liquid membranes, whether homogeneous or biphasic, revealed variations in permeability and selectivity, emphasizing the influence of casting methods on membrane performance.[17–20]

Different blends, such as CA/polysulfone(CA/PSF), CA/thiazole-based polyimine (PM-4), CA/polyimide (PI), and CA/Pebax, have notably enhanced gas separation performances. The incorporation of polymer has shown remarkable potential in enhancing the gas separation performance of membranes. CA/PI blends exhibited CO_2/CH_4 selectivity up to 50 compared to pure CA ($\sim$30). CA/PSF blends doubled the CO_2/N_2 selectivity from 25 to 50. CA/Pebax blend membranes exhibit enhanced efficiency in CO_2/N_2 separation, with significant improvements in CO_2 permeability and selectivity upon the addition of Pebax. CA/Pebax, there is an approximate 25% and 59% increase in CO_2/N_2 selectivity and CO_2 permeability, respectively.[21–23] Each blend presents a unique gas permeability sequence, emphasizing their application-specific advantages.

On the exploration of CA-based mixed matrix membranes (MMMs). Noteworthy examples include enhancing mechanical strength and permeability in cellulose acetate/nanoclay (CA/NC) composite membranes.[24] The stability and CO_2 adsorption performance improvement achieved by incorporating Cu-MOF-GO in CA.[25] The studies extend to gas separation characteristics of MMMs based on metal–organic frameworks (MOF) and cellulose acetate/gamma-cyclodextrin MOF, showcasing advancements in CO_2/CH_4 separation with enhanced selectivity.[26] Furthermore, the introduction of nanomaterials like multi-walled carbon nanotubes (MWCNTs) and ZIF-62 glass in cellulose acetate membranes demonstrates promising characteristics for liquid and gas separations.[27,28]

MMMs composed of CA/MOFs demonstrate enhanced selectivity and permeability for CO_2/CH_4 separation compared to other composite membranes illustrated in Fig. 7. The position of data for CA on the Robeson plot serves as a useful indicator of its potential as gas separation membranes. For instance, CA/MWCNT and CA/PI are positioned lower on the graph, indicating lower permeability, whereas CA/PEG exhibits greater permeability among the synthesized cellulose acetates. CA/IL and CA/silane show promising permeability potential, positioned between CA/MWCNT and CA/PEG on the Robeson plot. CA/MO and CA/MOF demonstrate exceptional selectivity in composite membranes with favorable permeability, surpassing other composite CA membranes.

As the research progresses, a distinct focus is placed on the challenges associated with experimental scale separation, emphasizing the need for cost-effective and time-efficient alternatives. This prompts a shift towards MD simulations, a powerful tool capable of elucidating physical properties and morphological alterations at the molecular level. The last section includes specific MD simulation studies exploring the performance of cellulose-based materials. For instance, the research a encompasses son CA dissolution behavior[29,30] and the compatibility of cellulose acetobutyrate with plasticizers.[31] The collective findings highlight the progress in enhancing gas separation efficiency and underscore the persistent challenges that necessitate a holistic approach, combining experimental insights and computational simulations.

This review navigates through cellulose-based materials; this research sets the stage for a comprehensive exploration of their potential applications, innovative compositions, and the critical role of MD simulations in advancing this field. This review comprehensively explores the properties and application of CA. Moreover, various synthesis methodologies employed for CA membranes are discussed, providing an in-depth examination of their application and advancements. The synthesis methods discussed encompass grafting techniques, showcasing their specific application advantages. The review further delves into the advancements of CA-based MMMs, emphasizing mechanical strength, permeability enhancement, stability, and CO_2 adsorption performance. Additionally, the challenges linked to