

PREPARATION OF HIGH-PERFORMANCE FO MEMBRANES BY USING RECYCLED CIGARETTE BUTTS AS SUPPORTING LAYER

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ABSTRACT

Cigarette butts (CBs), mainly composed of cellulose acetate, are one of the most common and persistent forms of waste, posing significant environmental risks due to their non-biodegradable nature and toxic leachates. At the same time, CBs represent a potential low-cost and sustainable resource for advanced material development. In this work, we aimed to enhance membrane performance by fabricating forward osmosis (FO) membranes through the recycling of CBs into nanofiber supports via electrospinning, followed by modification with ZIF-8 and a polyamide (PA) active layer. Structural and chemical characterizations revealed that CB-based membranes possessed superior fibrous morphology and functional groups suitable for ZIF-8 incorporation compared to pure cellulose acetate membranes. The optimized composition (CB:CA 1:4) achieved a high water flux of 24.93 LMH and a reverse salt flux of 3.23 gMH under FO operation using distilled water as the feed solution and 1 M NaCl as the draw solution. The addition of ZIF-8 enhanced hydrophilic-hydrophobic balance, mitigated internal concentration polarization, and improved overall selectivity, while the PA layer provided additional fouling resistance and stability. These results indicate that combining recycled CBs with advanced nanomaterials can significantly enhance FO membrane performance. Beyond technical improvements, this study offers an environmentally beneficial approach by transforming hazardous waste into functional materials for clean water production, supporting both sustainable waste management and low-energy water treatment technologies.

Keywords: *cigarette butts, cellulose acetate, electrospinning, forward osmosis, ZIF-8*

1. INTRODUCTION

Cigarette butts (CBs) are recognized as one of the most pervasive waste products worldwide, with more than 4.5 trillion discarded annually [1]. These butts, primarily composed of cellulose acetate, are highly resistant to biodegradation and can persist in the environment for decades [2, 3]. Their presence is especially problematic in coastal and urban environments, where they act as a long-term ecological threat [4-6]. Furthermore, public attitudes and improper disposal habits in various regions contribute to the increasing density of this hazardous litter [7, 8]. When improperly disposed of, they release toxic substances such as nicotine, polycyclic aromatic hydrocarbons, and heavy metals including cadmium, lead, and arsenic [9-11]. These pollutants can leach into soil and water, threatening aquatic ecosystems, food safety, and ultimately human health [12-14]. In addition to their chemical hazards, cigarette butts represent a

major economic and social burden, as their cleanup costs cities millions of dollars annually and they remain a dominant component of litter in beaches, parks, and urban streets [4, 15, 16].

From a materials science perspective, CBs are a nearly untapped resource because their main constituent, cellulose acetate, is a semi-synthetic polymer with desirable properties such as hydrophilicity, fiber-forming ability, and chemical stability [17]. Previous studies have primarily focused on recycling CBs into low-end applications, such as acoustic insulators [18], structural bricks [19, 20], or asphalt modifier [21]. Beyond basic recycling, CD-derived materials have also been explored for specialized applications such as superhydrophobic filters for oil spill cleanup [22]. However, these methods largely underutilize the inherent chemical potential and molecular functionality of cellulose acetate, treating the waste merely as a physical filler. If properly recovered and processed, CBs can serve as a low-cost raw material for the fabrication of functional materials, turning an environmental liability into a value-added product [23, 24]. Therefore, the concept of recycling CBs into membrane supports therefore represents an innovative approach that not only addresses environmental remediation but also maximizes the material's value for advanced separation technologies [25, 26].

Forward osmosis (FO) has emerged as a promising alternative to conventional pressure-driven processes like reverse osmosis (RO) due to its lower energy requirements, strong contaminant rejection, and lower fouling tendency [27-29]. These membrane technologies are crucial for the efficient removal of pathogens, heavy metals, and organic micro-pollutants from contaminated wastewater [30, 31]. FO relies on osmotic pressure differences to drive water transport across a semi-permeable membrane, making it inherently more energy-efficient and adaptable for applications such as desalination and wastewater treatment [32-34]. Despite these advantages, the widespread adoption of FO has been limited by challenges associated with membrane performance, specifically internal concentration polarization (ICP) occurring within the support layer [29, 35]. While conventional phase-inversion supports often exacerbate ICP due to their tortuous pore structures, the development of highly porous nanofiber supports has been proposed as a solution [36]. However, finding a balance between high porosity, mechanical stability, and sustainable material sourcing remains a significant challenge in current FO membranes research [37, 38].

To address these performance barriers, this study utilizes electrospinning to fabricate nanofiber supports from recycled CBs, subsequently modified with ZIF-8 nanoparticles and a polyamide (PA) active layer. While electrospinning provides the necessary porosity, the synergy between waste-derived nanofibers and ZIF-8 for enhanced molecular sieving remains largely unexplored. The integration of metal-organic frameworks (MOFs) like ZIF-8 is essential for ensuring the structural integrity and high selectivity of the resulting composite membrane. This multi-layered composite approach leverages the strengths of each component, offering a path toward high-performance FO membranes that are both technically efficient and environmentally sustainable, effectively transforming hazardous waste into a valuable resource for water treatment.

Therefore, this study aims to explore the fabrication and performance of high-flux FO membranes using recycled cigarette butts as a sustainable support layer modified with ZIF-8 and PA layers. The primary objective is to evaluate how the synergy between waste-derived nanofibers and ZIF-8 crystals enhances water permeability and fouling resistance while

mitigating internal concentration polarization (ICP). By systematically evaluating structural properties, and separation performance, this work seeks to demonstrate both the technical feasibility and environmental value of converting CB waste into efficient membranes for clean water production.

2. RESEARCH METHODOLOGY

This experimental study evaluates the fabrication of high-performance forward osmosis membranes using recycled cigarette butt nanofibers modified with ZIF-8 and a polyamide (PA) selective layer. Data collection involved structural characterization via Scanning Electron Microscopy (SEM), Fourier Transform Infrared (FTIR) spectroscopy, and Water Contact Angle (WCA) analysis to assess morphology, chemical groups, and wettability. Membrane performance, specifically water flux (J_w) and reverse salt flux (J_s), was measured using a customized FO testing system with 1 M NaCl as the draw solution. Data analysis followed a descriptive-quantitative approach, comparing the effects of varying CB:CA ratios (Samples A1–A4) to identify the optimal composition. The results were further validated against existing literature to confirm improvements in permeability, selectivity, and the mitigation of internal concentration polarization (ICP).

2.1. Material and Equipment

The main materials in this experiment included cigarettes butts, cellulose acetate (CA), Polyamide (PA), N,N-dimethylacetamide (DMAc), ZIF-8, ethanol, and NaCl solution as the draw solution. Key instruments were an electrospinning machine, SEM, FTIR, contact angle analyzer, and FO testing system.

2.2. Experimental Methods

The experimental procedure began with the extraction of cellulose acetate (CA) from discarded cigarette butts, which were stripped, purified using anhydrous ethanol to remove contaminants [24] and dried at 65 °C for 8 hours. To prepare the spinning solution, the extracted CA was dissolved in N,N-dimethylacetamide (DMAc) at a concentration of 14 wt% under continuous stirring at 90°C to ensure the formation of a homogeneous gel [26]. The electrospinning process was conducted under controlled environmental conditions (25 °C and 55% relative humidity), with operational parameters set at an applied voltage of 12 kV, a tip-to-collector distance of 15 cm, and a solution feed rate of 0.008 mL/min. The resulting membranes were subsequently characterized to evaluate their morphological and structural properties. Surface morphology and fiber diameter were analyzed using Scanning Electron Microscopy (SEM), while chemical functional groups were identified through Fourier Transform Infrared (FTIR) spectroscopy. Additionally, surface wettability and membrane thickness were determined via Water Contact Angle (WCA) analysis and digital micrometer measurements, respectively, to ensure high-performance standards for forward osmosis applications.

3. RESULT AND DISCUSSION

3.1. Morphology and Structure Characterization of Fiber Membranes by Using CBs to Replace Pure CA

As shown in the SEM **Figure 1**, the fiber morphology evolves as the composition shifts from CB-derived cellulose to pure CA. Sample A1 shows relatively uniform fibers with moderate diameters and minimal beads, indicating that CBs can produce well-defined fibers. In A2 and A3, however, the fibers become less uniform, especially A3, where bead formation and fusion are more visible, likely due to changes in polymer viscosity [36]. In contrast, A4 with the highest CA content exhibits a clean, highly regular structure with consistent diameters and very few defects, demonstrating CA's superior spinning stability.

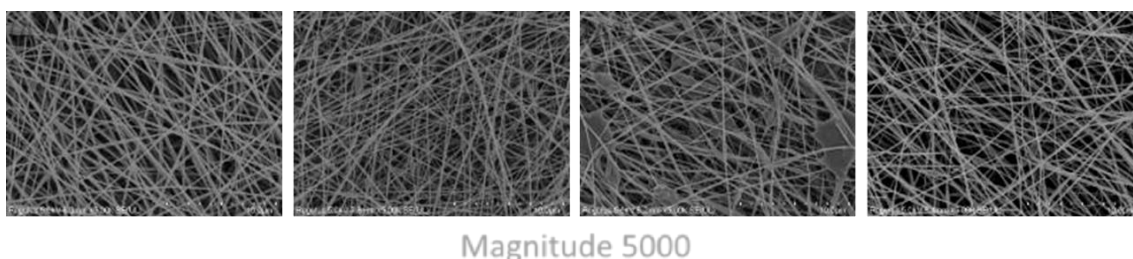


Figure 1. The morphology of cellulose acetate fiber membranes from CB

Surface wettability was also evaluated through water contact angle (WCA) measurements, where lower angles indicated higher hydrophilicity, important for water transport in FO membranes [39, 40]. In addition, FTIR confirmed the chemical structure of CA, while fiber diameter distributions were analyzed using Origin. Overall, the results confirm that increasing CA content improves fiber uniformity and reduces imperfections, while CB-based membranes still allow successful fiber formation with some irregularities, offering a sustainable alternative to pure CA.

3.2. Fourier Transform Infrared of The Nanofiber Membrane

As shown in **Figure 2**, the FTIR spectra display distinct peaks below 2000 cm^{-1} , corresponding to key functional groups such as C–H, C=O, and C–O–C, which are characteristic of cellulose acetate. As the CB content increases from A1 (1:1) to A4 (1:4), the intensity of these characteristic peaks in the FTIR spectra increases, which corresponds to a decrease in transmittance. Among all samples, A4 (CB ratio 1:4) shows the strongest peaks and highest transmittance, suggesting more effective incorporation of CB into the matrix and a more defined chemical structure.

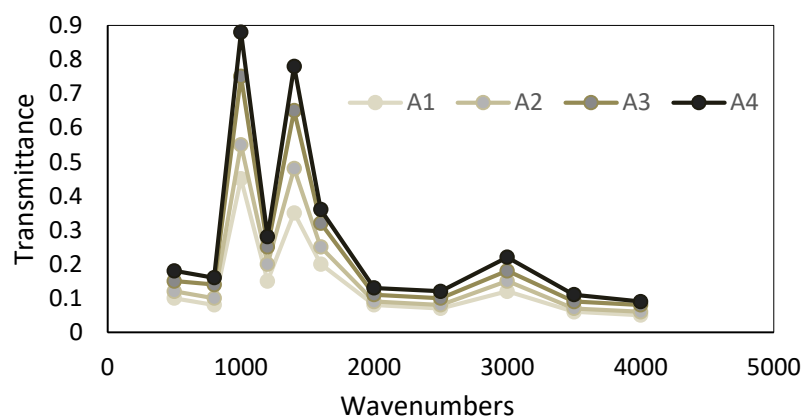


Figure 2. FT-IR spectra of CBs membranes with varying CB ratios

This enhanced FTIR response implies improved chemical properties, including higher

solubility and better thermal stability. Furthermore, the subsequent modification of A4 with ZIF-8 and polyamide layer introduces N-H stretching vibrations, confirming the successful formation of the selectivity active layer. Overall, the spectra confirm that adjusting the CB ratio directly affects molecular interactions and membrane quality.

3.3. Water Contact Angle of The Nanofiber Membrane

As shown in **Figure 3**, the measurements of the Water Contact Angle (WCA) for samples A1, A2, A3, and A4 values ranging from 87.928° to 117.546°. The highest WCA values specifically 116.132° for A1 and 117.546° for A4, indicate a hydrophobic surface character. This property is likely attributed to the lower surface energy of the recovered cellulose acetate fibers [39]. Conversely, the lower WCA 87.928° demonstrates a more hydrophilic behavior. These variations indicate that adjusting CB/CA ratio significantly influences the membranes wettability. Based on these characteristics, sample A4 was selected as the optimal substrate for integration ZIF-8 nanoparticles. Its combination of regular morphology and specific surface properties provides a more effective chemically active surface for the subsequent integration of ZIF-8 and the PA selective layer.

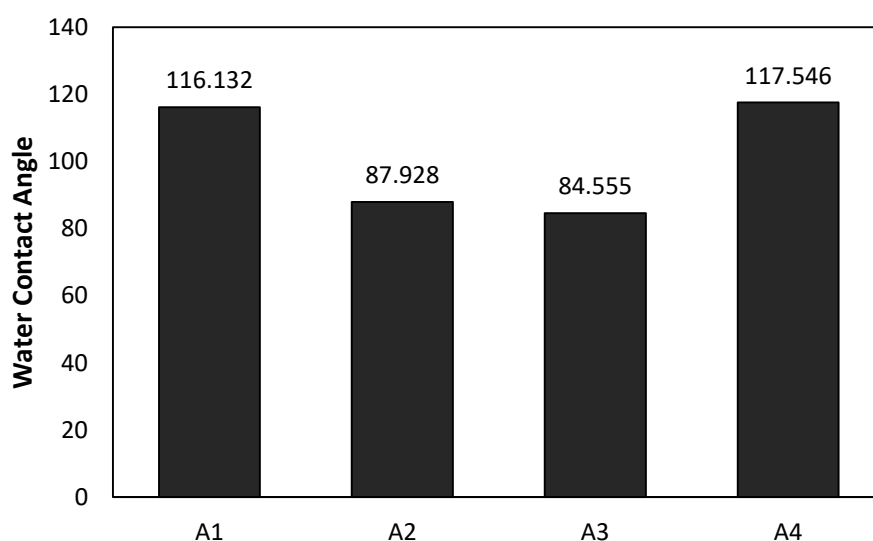


Figure 3. Water contact angle with different ratio of CBs

3.4. Membrane Thickness

As shown in **Figure 4**, the membrane thickness varies across the four samples (A1, A2, A3, and A4). The thickness measurement ranges significantly from 0.1196 to 0.1922. Sample A1 exhibits the highest membrane thickness at 0.1922 mm, followed by sample A3 at 0.189 mm and sample A2 at 0.1766 mm. In contrast, sample A4 possesses the thinnest structure in the group, with a value of only 0.1196 mm. These differences suggest that the thickness is influenced by several factors, including material composition and the parameters of the manufacturing process. For instance, the higher values in A1 and A3 may indicate a more densely packed fiber structure, while A4 represents a lighter or less dense composition. This minimized thickness in sample A4 is highly critical, as a thinner support layer effectively reduces internal concentration polarization (ICP), which is a primary limiting factor for water flux in forward osmosis (FO) systems [29, 35].

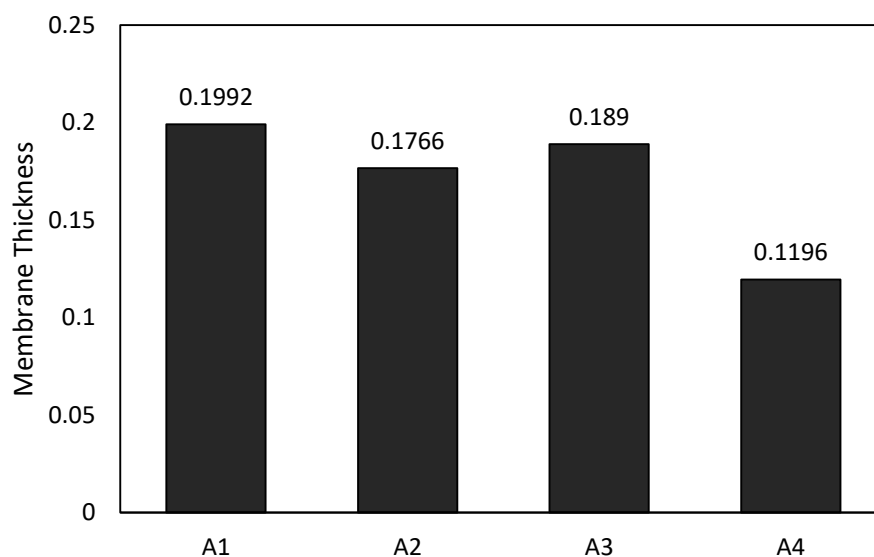


Figure 4. Membrane thickness with different ratio of CBs

3.5. The Result of Water Flux (J_w) and RSF (J_s) of FO System

Sample A4 was selected as the optimal substrate for integrating ZIF-8 nanoparticles due to its superior combination of structural, chemical, and functional properties, all critical for high-performance forward osmosis (FO) membranes. Sample A4 exhibits highly uniform fibers and an excellent morphology, as demonstrated by SEM analysis. Due to its higher cellulose acetate (CA) content, which enhances electrospinning stability, the nanofibers are regular, defect-free, and maintain a consistent diameter throughout [36]. This even, smooth surface is essential because it allows ZIF-8 particles coat or embed evenly, ensuring optimal water flux and contaminant rejection in the final membrane. Furthermore, A4 possesses enhanced chemical properties through FTIR spectroscopy. The prominent peaks correspond to key functional groups, such as C–H, C=O, and C–O–C, providing more chemically active sites. These active groups are very important because they help the ZIF-8 particles form strong chemical bonds with each other, ensuring the nanoparticles remain embedded within the membrane matrix, which increases the composite's overall strength and durability.

The surface properties of A4 also provided a strategic advantage. While FO membranes often favor hydrophilicity for water flux, A4 is hydrophobic with water contact angle of 116° – 117° . This inherent hydrophobicity helps reduce organic fouling [39]. Importantly, the hydrophilic nature of ZIF-8 nanoparticles can effectively balance this hydrophobicity when combined, creating a synergistic effect that facilitates water transport while simultaneously blocking foulants [35]. In addition, the physical structure of A4 is highly advantageous as it is significantly thinner than the other samples at 0.1196 mm. This minimized thickness is a crucial as it lowers internal concentration polarization (ICP), reducing resistance to water flow through the support layer and directly increasing efficiency [29, 35].

Overall, A4 aligns with the goal of sustainability by effectively utilizing recycled cellulose from cigarette butts (CBs) while maintaining high functional CA content. This balance results in membrane properties that are comparable to or better than pure CA, offering a cost-effective and environmentally friendly solution. Adding ZIF-8 to A4 creates

a synergistic composite membrane well-suited for high water flux, selectivity, and durability, directly supporting the development of effective water filtration systems using recycled materials.

Table 1. The result of water flux (J_w) and reverse salt flux (J_s) of forward osmosis system

No	Sample	FS	DS	Concentration (M)	J_w (LMH)	J_s (Gmh)
1	A4	Distilled water	Sodium chloride	1	24.93	3.23

Based on **Table 1**, the researcher demonstrated that the CB-CA membrane (sample A4), when modified through electrospinning optimization, ZIF-8 incorporation, and a polyamide selective layer, achieved a significantly higher water flux of 24.93 LMH indicating efficient water permeability, while maintaining a relatively low reverse salt flux of 3.23 gMH meaning only a small amount of NaCl leaks back into the feed solution. This combination of high J_w and low J_s results in a J_w/J_s ratio of about 7.7, reflecting good selectivity and efficiency [41].

These results are significant because they indicate the membrane can produce a large volume of water while maintaining quality, making it highly promising for applications such as water treatment, desalination, and water recycling with stable performance. In comparison, [26] reported that the membrane prepared from cigarette butts exhibited a water flux of 14.7 LMH and a reverse salt flux of 3.5 gMH. These results indicate that the current findings outperform previous studies by showing both higher permeability and better selectivity. The enhancement can be attributed to the optimized CA:CB ratio, the defect-free nanofiber morphology produced by electrospinning, and the synergistic effect of ZIF-8 and the polyamide layer. Together, these modifications reduce internal concentration polarization, improve antifouling properties, and enhance contaminant rejection [33]. Consequently, the present study demonstrates a more efficient and sustainable approach for transforming cigarette butt waste into high-performance forward osmosis membranes.

4. CONCLUSION AND SUGGESTIONS

This study successfully developed a high-performance Thin-Film Composite (TFC) Forward Osmosis (FO) membrane by utilizing recycled cigarette butts (CBs) as a sustainable nanofiber support. The optimized support (Sample A4, with a 1:4 CB:CA ratio) exhibited a defect-free morphology and minimal thickness (0.1196 mm), which significantly reduced internal concentration polarization (ICP) and enhanced water flux to 24.93 LMH. The integration of ZIF-8 and a polyamide (PA) active layer has created a synergistic effect, balancing hydrophilicity and mechanical strength while maintaining high selectivity, as evidenced by the effective removal of diclofenac. By transforming hazardous CB waste into a functional filtration material, this research demonstrates a viable path for the circular economy, offering a low-energy and eco-friendly alternative to conventional desalination and wastewater treatment technologies.

Future research should focus on evaluating the long-term operational stability and

durability of these CB-derived membranes under real-world wastewater conditions, which may contain complex multi-pollutant mixtures. Additionally, further investigation into the scalability of the electrospinning process and the potential recovery of ZIF-8 nanoparticles at the end of the membrane's life cycle would be beneficial. Exploring the life-cycle assessment (LCA) of this fabrication route could also provide deeper insights into its overall environmental footprint compared to traditional petroleum-based membrane production.

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