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Cigarette Filter-Based Membranes with Tannin and $FeCl_3$ Additives for Enhancing the Antifouling Properties of Oil Emulsion Filtration

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Abstract

Industries in Indonesia significantly contribute to the economy by increasing foreign exchange earnings and creating job opportunities. However, industrial activities also negatively impact the environment, particularly water pollution caused by liquid waste containing oil emulsions. This research aims to develop a membrane based on cigarette filters as an alternative to cellulose acetate for separating oil emulsions in water. Cigarette filters were processed into membranes with tannic acid (TA) and ferric chloride ($FeCl_3$) as additives using a vacuum-filtration coating technique. The resulting membranes were tested for their characteristics and performance, including morphological analysis, functional group analysis, hydrophilicity, water uptake, porosity, pure water flux, selectivity toward oil emulsions (artificial emulsions from CPO and sodium dodecyl sulfate (SDS)) and antifouling properties. The results showed that membranes with additives exhibited more uniform pores, asymmetric structures, and improved surface characteristics. The addition of tannic acid 2.4 mmol and $FeCl_3$ 3.7 mmol also enhanced the antifouling properties of the membranes, although it decreased the pure water flux. Membranes without additives (M1) and with additives (M2) were compared for pure water flux, separation efficiency, and antifouling properties. M2 exhibited a lower flux (15.03 $L/m^2 \cdot h$) than M1 (19.69 $L/m^2 \cdot h$) due to reduced porosity. Although the oil emulsion selectivity of M2 (97.80%) showed no significant improvement compared to M1 (97.79%), the addition of additives notably enhanced the antifouling performance. M2 demonstrated a flux recovery ratio (FRR) of 71.22%, significantly higher than M1's FRR of 46.01%, indicating the effectiveness of the additives in reducing fouling and improving membrane reusability. The membranes achieved high selectivity for oil emulsions with a separation efficiency of up to 97%. These findings suggest that using cigarette filters as the base material for membranes and adding additives can provide an efficient and environmentally friendly solution to reducing oil emulsion pollution.

1. Introduction

The growth and development of various industries in Indonesia have positively impacted the country's economy by increasing foreign exchange earnings and creating new job opportunities. However, in addition to these positive effects, industrial growth has also brought negative environmental impacts. Liquid waste generated by industrial processes leads to environmental pollution, particularly due to the presence of oil emulsions in the waste [1]. Environmental pollution caused by oil emulsion waste is a serious issue [2], as it can significantly harm ecosystems, aquatic organisms, and human health. Prolonged exposure to oil-emulsion-contaminated water may lead to severe health problems, including skin irritation, respiratory issues, and potential long-term toxic effects on internal organs [3].

Oil emulsion waste is produced daily globally, particularly by industries such as petrochemicals, oil and gas refining, food processing, metalworking, and cosmetics manufacturing. These industries contribute substantially to the volume of oil emulsion waste released into the environment [4]. The permissible oil content in water is typically between 5 and 40 mg/L [5]. To address the pollution caused by oil emulsions, efficient and renewable methods for removing oil emulsions from the water must be developed. One renewable method of separation is membrane technology filtration [6].

Various treatments have been developed to mitigate the pollution caused by oil emulsions, including chemical coagulation, electrocoagulation, adsorption, and biological treatments [7]. However, these methods have limitations compared to membrane filtration technology. For instance, chemical coagulation requires large amounts of chemicals, which can lead to secondary pollution and increased operational costs. Electrocoagulation is energy-intensive and requires constant maintenance of electrodes. Although effective, adsorption methods often require frequent adsorbent regeneration, leading to higher costs and operational inefficiencies. Biological treatments are slow and may not be effective for all types of oil emulsions.

In contrast, membrane technology offers several advantages, including selective separation, efficient energy usage, reduced chemical requirements, reusability, and environmental friendliness [4]. Membrane separation technology has emerged as an effective and sustainable alternative for removing contaminants from water, including oil emulsions. Unlike conventional methods, membrane technology is capable of achieving high separation efficiency without introducing additional pollutants [8].

Several recent studies have shown that membrane technology is highly effective in removing oil emulsions from water. For example, Jiang *et al.* [9] modified polylactic acid membranes by incorporating cellulose acetate in situ into the membrane matrix, resulting in membranes with superhydrophilic and superoleophobic properties, achieving an oil emulsion separation efficiency of 99.8%. Al-Rajabi *et al.* [10] modified polyacrylonitrile (PAN) membranes by embedding cellulose acetate nanofibers through core-shell electrospinning, producing hydrophilic membranes with a separation efficiency of 99.9%. Fahmy *et al.* [11] developed hybrid membranes using polysulfone (PSF), cellulose nanocrystals, and polyaniline via phase inversion techniques, achieving a separation efficiency of 88.2%. These studies employed cellulose acetate-based membranes due to their high hydrophilicity, facilitating the effective separation of water and oil emulsions.

Cellulose acetate has many advantages, such as biodegradability, and is thus more environmentally friendly. However, membranes made from cellulose acetate are relatively more expensive than those made from other polymers [12]. To address this issue, cellulose acetate can be replaced with environmental residues such as cigarette filters. Converting cigarette filters into cellulose acetate can be performed directly due to minimal contamination from other residues. Cigarette filters are also easily found in the environment as solid waste [13]. Given the abundant availability of cigarette butts, we innovated by replacing cellulose acetate polymer with these filters as the primary raw material for membrane production.

A previous study Liu *et al.* [14] demonstrates that cigarette filter membranes generally exhibit poor mechanical properties, which can limit their effectiveness in various applications. To address this issue, the incorporation of additional materials or additives, such as tannic acid and $FeCl_3$, has been proposed. The addition of these additives not only improves the mechanical strength of the membranes but also enhances other important characteristics, including their antifouling properties, which help prevent fouling by microorganisms, and their selectivity, which contributes to improved separation efficiency and overall performance [15].

Tannic acid and $FeCl_3$ additives can form complex bonds. The electron pairs of tannic acid compounds bind with metal ions in $FeCl_3$, forming a complex molecule. This complex formation alters the solution's color and enhances the material's physical and chemical properties. The mixed $FeCl_3$ and tannic acid solution were applied to the membrane using the coating vacuum filtration technique. In the coating process, the volume of the solution affects the thickness of the membrane surface layer. The pressure difference between the top and bottom surfaces of the membrane drove the coating solution into the membrane pores, forming a layer on the membrane surface [16].

Several studies have replaced the polymer cellulose acetate with cigarette filters. For example, a previous study [17] replaced cellulose acetate with cigarette filter waste to enhance the characterization and performance of the membranes. Morphological analysis of the membranes revealed a porous cross-sectional upper layer with high selectivity. On the other hand, Abu-Danso *et al.* [18] explored the extraction of cellulose nanocrystals and cellulose nanofibers from cigarette filters to remove diclofenac from water, and their results indicated good selectivity in the removal process. However, based on the literature review, there is a lack of research that adds additives to cigarette filter membranes to improve their characterization and performance.

This study utilizes cigarette filters as a substitute for cellulose acetate, incorporating tannic acid and $FeCl_3$ through a vacuum coating filtration process. The aim is to enhance the membranes' characterization and performance, particularly their antifouling properties, and improve selectivity and overall functionality. Unlike previous studies on structural modification, this study chemically functionalized the membranes with tannic acid and $FeCl_3$, forming complexes that improved the antifouling properties. In addition, this study offers a cost-effective and sustainable alternative to conventional cellulose acetate membranes by converting cigarette filters into membrane raw materials.

2. Experimental

2.1. Materials

Cigarette butt filters (CBF) purchased online under the Umild brand were used as the main raw polymer material. Dimethylacetamide (DMAc), tannic acid, $FeCl_3$, Tris-chloride, and sodium hydrogen phosphate ($Na_2HPO_4.2H_2O$), all sourced from Sigma Aldrich and purchased from a chemical supply store, were used as the solvent, additives, and buffers to neutralize the membrane surface, respectively. Distilled water was used as the non-solvent. Crude palm oil (CPO) was purchased directly from a palm oil industry, while sodium dodecyl sulfate (SDS), used as a surfactant to create an artificial oil-in-water emulsion solution, was also sourced from Sigma Aldrich.

2.2. Preparation of Membranes

The membrane was fabricated using a non-solvent-induced phase separation process. First, a dope solution was prepared by mixing 17% w/t of CBF into DMAc solvent, followed by stirring for approximately 24 hours. The solution was then left to stand for about 1 hour to remove any air bubbles. Next, the solution was cast onto a glass plate using a casting knife. The evenly spread solution was immersed in a coagulation bath containing a non-solvent, initiating the phase inversion process through a solvent-non-solvent exchange.

The additives were added via a vacuum-filtration coating technique for membranes containing additives. These included 2.4 mmol of tannic acid, 3.7 mmol of $FeCl_3$, and a buffer solution (Na_2HPO_4 200 mmol/L; tannic acid 0.3 mmol/L; Tris-chloride 16 mmol/L) [19]. The membrane sheet was placed in a flash filtration flask, and 25 ml of the additive solution was poured onto the membrane surface. A vacuum pump drew the liquid through the membrane, allowing the additives to coat the surface and form a layer. A clearer schematic is provided in Figure 1.

The prepared membranes were tested for their characterization and performance. CBF membrane with code M1 was fabricated without additives, while M2 was fabricated with adding additives, as shown in Table 1.

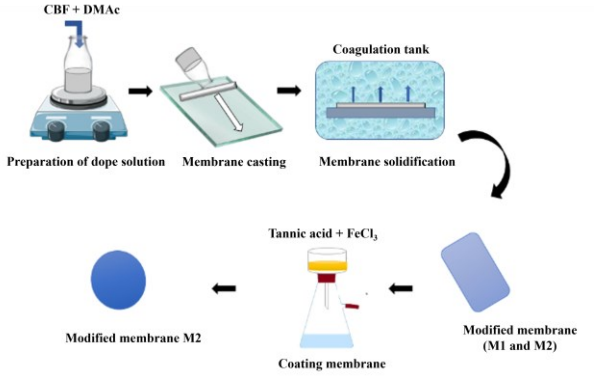

Figure 1. Membrane preparation

Table 1. Composition of CBF membranes

Membrane code	CBF (% w/t)	DMAc (% w/t)	Tannic acid (mmol)	$FeCl_3$ (mmol)
M1	17	83	-	-
M2	17	83	2.4	3.7

2.3. Characterization Test

2.3.1. Morphological Structure of Membranes

The membranes were analyzed using Scanning Electron Microscopy (SEM, JSF-7500, JEOL, Japan) to assess their morphological structure on the surface and cross-section, yielding qualitative results. Membranes M1 and M2 were initially cut and dried. For cross-sectional analysis, the membranes were immersed in liquid nitrogen to freeze and fracture them. They were then mounted on a specimen holder and placed in a vacuum coating apparatus, where gold and palladium (Au-Pd) coatings were applied. This coating enhances the sample's electrical conductivity, facilitating the observation of its morphology.

2.3.2. Functional Group Analysis

Functional group analysis was conducted using Fourier Transform Infrared (FTIR-ATR, PerkinElmer Inc.) to identify the functional groups in the membranes. The working principle involves measuring the absorption of infrared radiation at various wavelengths. The observed wavelength range is approximately 650-4000 cm^{-1}. The membrane was dried at room temperature and placed on an active cell for observation. The resulting spectra displayed peaks at specific wavelengths, which were used to analyze the functional groups.

2.3.3. Membrane Hydrophilicity

The hydrophilicity of the membrane was measured using a water contact angle (WCA) instrument, specifically a goniometer (Drop Master 300; Kiowa Interface Science Co., Ltd., Tokyo, Japan). Each membrane was dried at room temperature, and a water droplet was placed perpendicularly on the membrane surface. The angle formed on the membrane surface was measured by the instrument.

2.3.4. Water Uptake and Membrane Porosity

The gravimetric method was employed to evaluate the porosity and water uptake of the membrane. Initially, the membrane was weighed to determine its wet weight, and its thickness was measured using a micrometer screw. It was then dried in an oven at 60°C for 3 hours, a condition sufficient to ensure complete drying without causing degradation. The dried membrane was weighed again to obtain its dry weight. The resulting data were analyzed using Equation (1) to calculate porosity. Pore size was determined based on porosity and membrane flux data using Equation (2), while water uptake was assessed using Equation (3) [20].

$$\mathcal{E} = \frac{(W_1 - W_2)}{(p \times A \times l)} \times 100\% \quad (1)$$

$$rm = \sqrt{\frac{((2.9 - 1.75\mathcal{E}) \times 8 \times \eta \times l \times Q)}{\mathcal{E} \times A \times \Delta P}} \quad (2)$$

$$wu = \frac{W_1 - W_2}{W_1} \times 100\% \quad (3)$$

2.4. Membranes Performance Testing

2.4.1. Pure Water Flux (PWF)

PWF test measures the flow of pure water across the membrane surface using a cross-flow module. This test aims to determine the volume of water permeating through the membrane over a specified time. Before measurement, the membrane was compacted for 1 hour. Subsequently, the permeate weight (the amount of water passing through the membrane) was measured every 10 minutes. The experimental setup is illustrated in Figure 2. The PWF is calculated using Equation (4) [20].

$$J = \frac{Q}{(A \times t)} \quad (4)$$

Where, J is the pure water flux that successfully passes through the membrane (L/m².h), Q is the permeate volume (L), t is the filtration time (h), and A is the membrane surface area (m²).

2.4.2. Selectivity of Oil Emulsion

Selectivity testing of the emulsions was conducted using an artificial solution. The artificial oil emulsion solution was prepared by mixing 1 ml of CPO with 1000 ml of distilled water, followed by the addition of SDS as an emulsifier (surfactant) at a concentration of 0.15% of the total oil in the solution [21]. The mixture was stirred for 2 hours. The stages of the preparation process are illustrated in Figure 3.

The test was conducted to determine the membrane's ability to remove oil emulsion contaminants from water. The permeate concentration was analyzed using a UV-Vis spectrometer (Shimadzu UV-1700). The membrane's effectiveness in removing these contaminants can be calculated using Equation (5) [20].

$$R = \frac{(C_f - C_p)}{C_f} \times 100\% \quad (5)$$

Where, R is the rejection coefficient of the membrane (%), C_f is the oil emulsion concentration in the feed 1000 (ppm), and C_p is the oil emulsion concentration in the permeate (ppm).

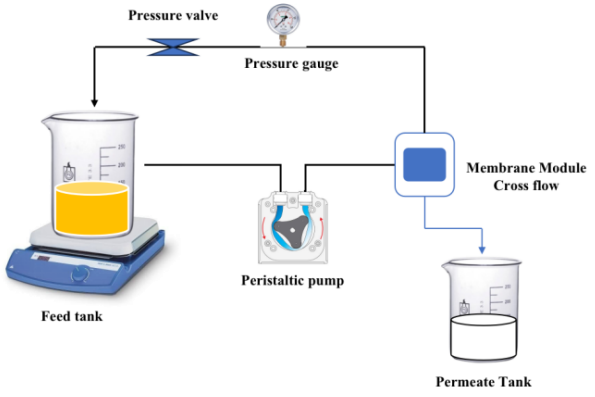

Figure 2. Filtration equipment setup

2.4.3. Antifouling Test

The antifouling test for the membrane was conducted in three stages. In the first stage, pure water was flowed through the membrane for 60 minutes, and the permeate weight was measured every 10 minutes (J_{wi}) [22]. In the second stage, an artificial oil emulsion solution was flowed through the membrane for 60 minutes, with permeate weight measured every 10 minutes (J_{ha}), followed by membrane cleaning using the backwash technique for 10 minutes. In the final stage, pure water was passed through the membrane again for 60 minutes, and the permeate weight was measured every 10 minutes (J_{wi+1}).

The data obtained from all three stages can be used to analyze the antifouling properties of the membrane by calculating the flux recovery ratio (FRR). A comparison of data from J_{wi} before and J_{wi+1} after backwashing was used to calculate the fouling on the membrane. Equation (6) is used to calculate the total fouling (R_t), which is a combination of reversible fouling ratio (R_r) and irreversible fouling ratio (R_{ir}), as described in Equations (7) to (9) [22].

$$FRR = \frac{(J_{wi})}{(J_{wi+1})} \times 100\% \quad (6)$$

$$R_t = \frac{(J_{wi} - J_{ha})}{J_{wi}} \times 100\% \quad (7)$$

$$R_r = \frac{((J_{wi+1}) - J_{ha})}{J_{wi}} \times 100\% \quad (8)$$

$$R_{ir} = \frac{(J_{wi} - (J_{wi+1}))}{J_{wi}} \times 100\% \quad (9)$$

3. Results and Discussion

3.1. Membrane Characterization

3.1.1. Membrane Morphological Structure

As shown in Figure 4, the M1 membrane exhibits highly open surface pores with uneven distribution. In contrast, the M2 membrane, modified with tannic acid and $FeCl_3$ coating, demonstrates a more uniform pore distribution [23]. This enhanced pore uniformity in M2 contributes to improved antifouling properties, resulting in superior separation performance and reduced fouling compared to the M1 membrane, whose uneven pore distribution may lead to less efficient separation and higher fouling rates [24].

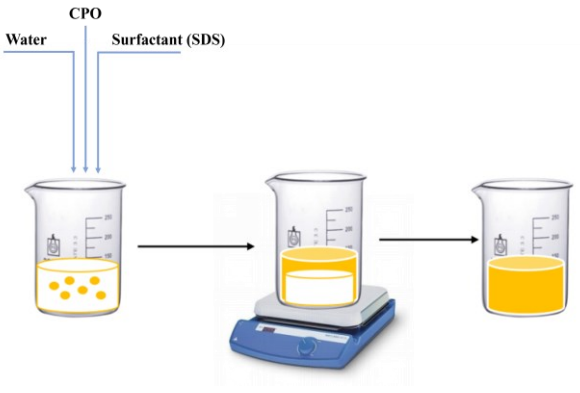

Figure 3. Preparation of artificial oil emulsion solution

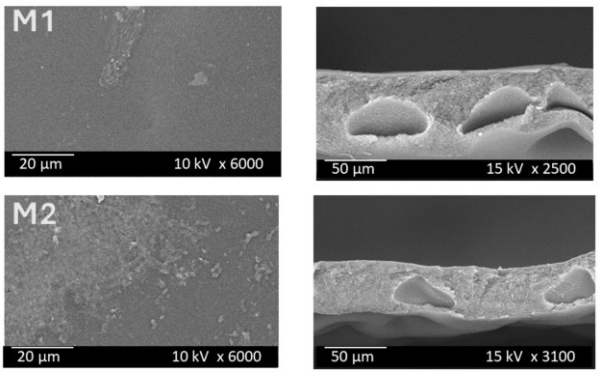

Figure 4. Surface and cross-sectional morphological structure of membranes

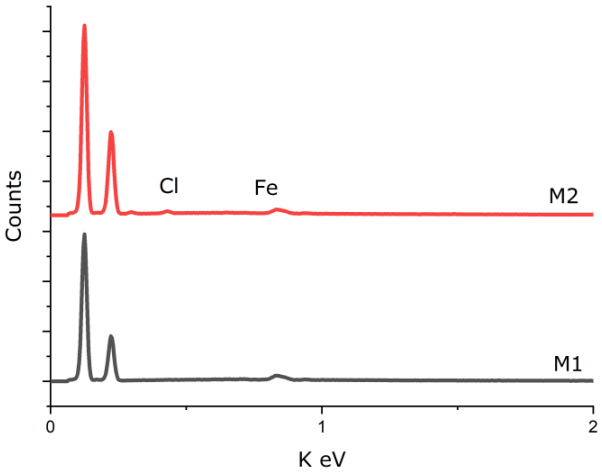

Figure 5. EDS spectrum of the membrane surface

Table 2. Chemical elemental composition of the membrane surface

	Atomic weight (%)			
Membrane code	C	O	Cl	Fe
M1	66.74	33.26	0.00	0.00
M2	63.57	35.25	0.35	0.81

The cross-sectional images of both the M1 and M2 membranes show minimal differences. This is because the addition of tannic acid and $FeCl_3$ through the vacuum filtration coating technique primarily forms a layer on the membrane surface. The cross-sectional images reveal an asymmetric structure, with a dense top layer responsible for pollutant separation, while the bottom layer serves as a support structure, influencing the membrane flux [25]. Additional observations were carried out to analyze the elemental composition of the membrane, as presented in Figure 5 and Table 2.

The M1 membrane contains only carbon (C) and oxygen (O) atoms, which are the primary elements of cellulose acetate. In contrast, the M2 membrane exhibits a greater diversity of elements, including carbon (C), oxygen (O), chlorine (Cl), and iron (Fe) [26, 27]. This finding aligns with the functional group analysis via FTIR, which shows that adding tannic acid introduces hydroxyl groups, while $FeCl_3$ contributes phenolic groups. Fe^{3+} atoms in the modified membrane further confirm the successful formation of complex bonds between tannic acid and $FeCl_3$.

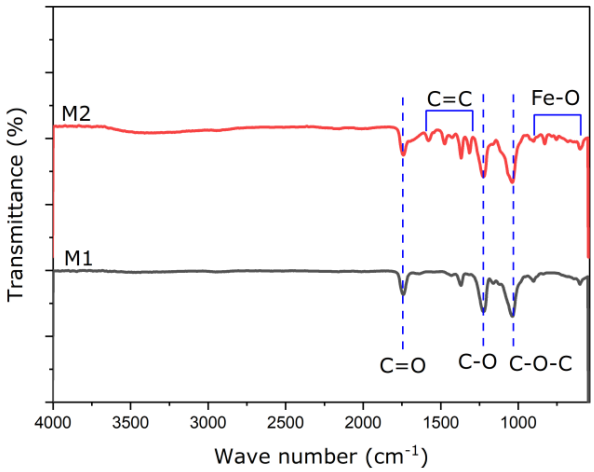

Figure 6. FTIR spectra of the membranes

3.1.2. Functional Group Analysis of the Membranes

Figure 6 shows that the M1 membrane exhibits peaks at wavenumbers 1750 cm^{-1} and 1220 cm^{-1}, indicating the presence of carboxyl groups, specifically C=O and C-O, as well as a peak at 1050 cm^{-1}, which corresponds to ether groups (C-O-C). These functional groups suggest that the polymer derived from cigarette butts contains the same functional groups as the cellulose acetate polymer [28].

New peaks appeared in the M2 membrane, indicating the presence of aromatic rings from tannic acid at wavenumbers 1350-1600 cm^{-1}, confirming the incorporation of tannic acid into the samples. Additionally, a new peak at wavenumbers 600-800 cm^{-1} indicates the presence of Fe-O groups, suggesting bonding between iron ions and hydroxyl groups in tannic acid [29]. The identification of these new functional groups demonstrates the successful modification of the membranes using the vacuum-filtration coating technique.

3.1.3. Hydrophilicity of the Membranes

The water contact angle (WCA) defines the hydrophilicity properties of the membrane. The hydrophilicity is influenced by the chemical composition of the membrane structure and is closely related to its water uptake. M1 and M2 membranes showed no significant differences in water contact angle, indicating that all membranes exhibit good hydrophilicity [30]. This finding aligns with the FTIR spectra, which identified hydroxyl (OH) groups in all membrane types.

The addition of additives using the vacuum filtration coating technique improved the hydrophilicity of the membrane [23]. As shown in Figure 7, the hydrophilicity of the M2 membrane was slightly better than that of M1, as indicated by the WCA values. M1 had a WCA of 76.00°, while M2 exhibited a slightly lower WCA of 74.63°, reflecting enhanced hydrophilicity in M2. However, the difference in hydrophilicity between M1 and M2 was minimal. This could be due to the coating layer formed by the additives not fully penetrating the membrane pores, limiting the overall effect on surface hydrophilicity [31]. Consequently, the hydrophilicity of M2 was only marginally improved compared to M1.

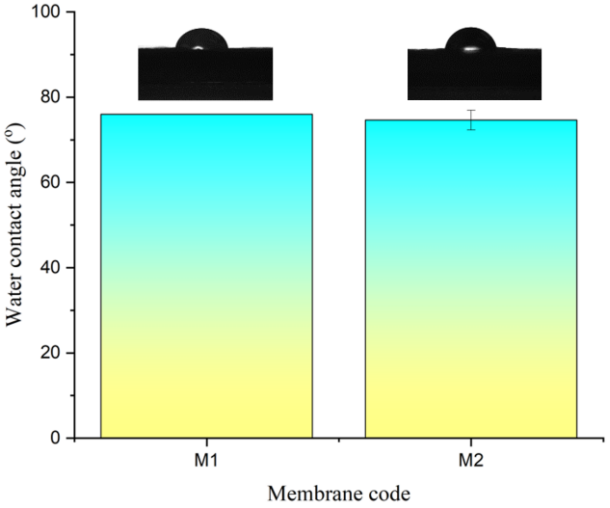

Figure 7. Water contact angle (WCA) value

3.1.4. Water Uptake and Porosity

Water uptake analysis evaluates the membrane's ability to absorb water, a value closely linked to its hydrophilicity, as measured by the WCA [32]. The water uptake value was determined using the gravimetric method, which involved measuring the difference in the membrane weight before and after water absorption, as shown in Figure 8. This method also determined membrane porosity by evaluating the volume of void spaces to quantify its empty space [32].

Upon observation, the water uptake value for the M2 membrane was 72.87%, showing no significant difference compared to the M1 membrane, which had a water uptake value of 79.94%. The M2 membrane exhibited lower water uptake than M1, likely due to the interaction between tannic acid, $FeCl_3$, and the membrane surface. The complex bonds formed between tannic acid and $FeCl_3$ likely cover some of the pores on the membrane surface, reducing water absorption [33].

The addition of additives to the membrane also affects the porosity values. The M2 membrane exhibited a lower porosity of 6.95% compared to the M1 membrane, which had a porosity of 11.93%. The coating technique used to add the additives forms a layer on the membrane surface, reducing the number of pores and, consequently, decreasing the void space within the membrane [34].

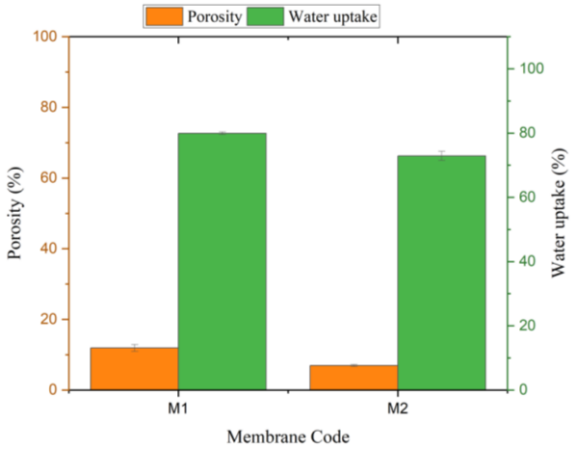

Figure 8. Water uptake and porosity of the membrane

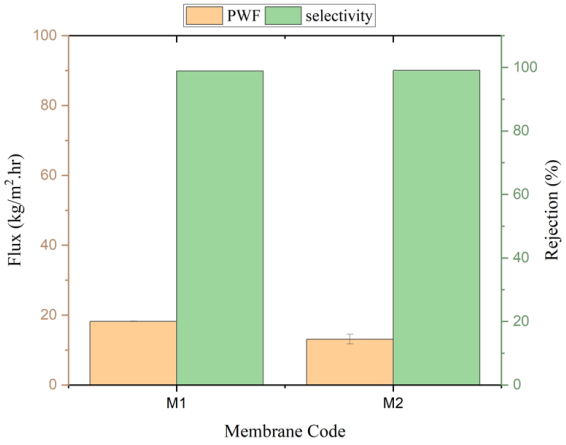

Figure 9. Pure water flux (PWF) and selectivity of the membrane

3.2. Membranes Performance

3.2.1. Pure Water Flux (PWF)

The pure water flux value for the membrane was obtained from evaluations using a cross-flow module at a pressure of 1 bar. The pure water flux is related to the morphological structure, porosity, and hydrophilicity of the membrane. Figure 9 shows the PWF values and membranes M1 and M2 selectivity.

The M2 membrane exhibited a lower flux value of 13.127 kg/m².hour compared to the M1 membrane, which had a flux value of 22.85 kg/m².hour. This reduction is attributed to the forming of a layer of tannic acid and $FeCl_3$ on the membrane surface, which covers the membrane pores. This phenomenon is evident in the morphological structure of the membrane and in the porosity values of the membrane, which are inversely related to the PWF values [35].

During the process of removing oil emulsion from water, the presence of SDS in the solution can block the membrane pores, leading to increased fouling on the membrane surface. The addition of tannic acid and $FeCl_3$ did not significantly affect the selectivity of the M2 membrane compared to the M1. The selectivity of the M1 membrane was 98.94%, while the M2 had a slightly higher selectivity of 99.09%. This result is attributed to the large particle size of the CPO oil emulsion, meaning that even membranes without additives exhibit high selectivity toward the oil emulsion [36]. The selectivity results for the oil emulsion passing through the membrane are presented in Figure 10. Visually, the artificial solution before filtration appears more turbid than the filtered oil emulsion solution.

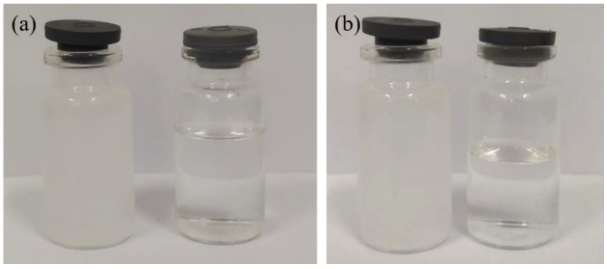

Figure 10. Oil emulsion filtration results (a) without additives and (b) with additives

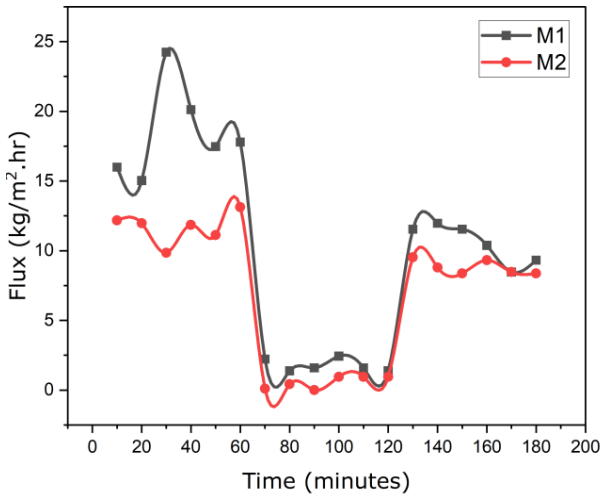

Figure 11. Flux decline and recovery profiles in the membranes

3.2.2. Antifouling Properties

Antifouling properties refer to a membrane's ability to prevent the accumulation of pollutants that could cause clogging, which is essential for the effective performance of membrane technology. Fouling occurs when pollutants build up, blocking membrane pores, but these properties can be enhanced through membrane modification, such as the incorporation of additives [37]. In this study, the membranes were treated with a surface-coating process using a vacuum-filtration coating technique aimed at reducing pollutant accumulation and preventing pore blockages.

The antifouling performance of the membranes was evaluated by monitoring flux changes, which reflect a decrease in flux during the filtration process, as shown in Figure 11. Figure 11 presents the flux decline profile, where pure water is initially filtered for 60 min (J_{w1}), followed by filtration of oil emulsion for the subsequent 60 min (J_{ha}), and a 10-minute backwash cleaning process. Finally, pure water is filtered again for 60 min (J_{w2}). This approach allows for measuring flux loss before and after the oil emulsion filtration period. The flux recovery profiles for membranes M1 and M2 are displayed in Figure 12.

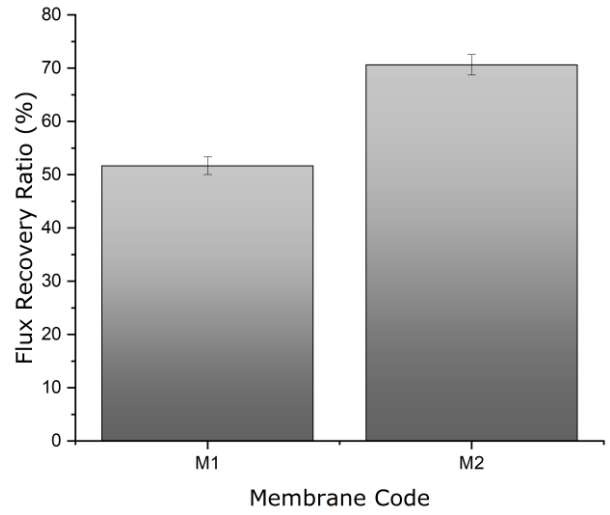

Figure 12. Flux recovery ratio (FRR) value

The oscillations observed in the flux profile reflect the influence of operational conditions, membrane stabilization, and the structural characteristics of each membrane. For the M1 membrane, the larger and unevenly distributed pores cause more noticeable flux fluctuations due to local variations in flow resistance. On the other hand, M2 shows a more stable flux profile [38]. This improvement is attributed to the uniform surface structure and enhanced hydrophilic properties brought about by the additives, which help reduce fluctuations in water transport across the membrane.

Despite the oscillations, M2 exhibits a comparatively more stable flux profile than M1, suggesting that the addition of tannic acid and $FeCl_3$ contributes to the membrane's enhanced performance. These findings indicate that the additives improve the membrane's selectivity and antifouling properties and help stabilize water flux during operation. Both M1 and M2 experienced a decline in flux following oil emulsion filtration, which can be attributed to membrane fouling. The extent of flux recovery after the oil emulsion filtration is shown in Figure 12.

The M2 membrane exhibited a higher FRR value of 70.61% compared to M1, which had an FRR of 51.66%. This improvement can be attributed to the addition of additives using the coating method, in which tannic acid and $FeCl_3$ form a layer. This layer reduces the interaction between oil emulsion molecules and the membrane surface, enhancing the antifouling properties of the membrane [39]. Thus, modifying the membrane with the addition of tannic acid successfully improved its antifouling properties.

Fouling has two characteristics: reversible and irreversible. Reversible fouling can be addressed by washing because it is typically caused by concentration polarization. Concentration polarization occurs when contaminants accumulate on the membrane surface because of the significant concentration difference between solutions above and below the membrane. Conversely, irreversible fouling is more challenging to address and often requires chemical cleaning processes. This type of fouling can also be minimized through membrane modification [40]. The R_t, which includes R_r and R_{ir}, for M1 and M2 is shown in Figure 13.

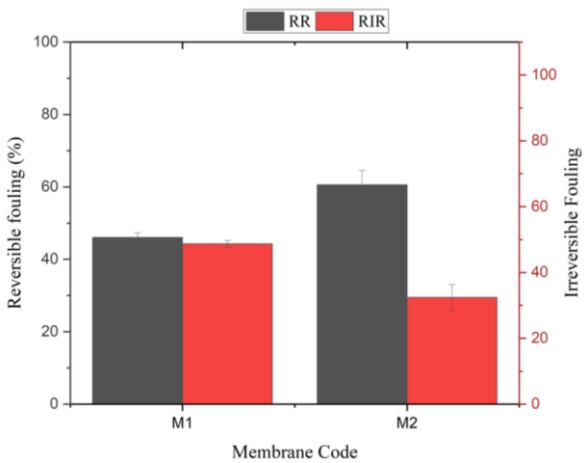

Figure 13. R_{ir} and R_r fouling values of the membrane

The M1 membrane exhibited a higher R_{ir} value compared to M2. The higher percentage of R_{ir} in M1 indicates that it is more difficult to clean membranes using only the backwash method (washing with plain water). Conversely, M2 exhibited a higher R_r than its R_{ir} value. This phenomenon indicates that fouling on the M2 membrane is more easily removed using the backwash method, thereby improving the FRR value after oil-emulsion filtration. This improvement can be attributed to the ability of the additives to coat the membrane surface, preventing the accumulation of oil emulsion particles in the membrane pores and reducing the occurrence of irreversible fouling [41].

4. Conclusion

This study successfully developed membranes based on cigarette filters, modified with tannic acid and $FeCl_3$, to enhance their characteristics and performance in separating oil emulsions from water. Morphological testing indicated that the addition of these additives resulted in a more uniform membrane surface structure and introduced new chemical components, as confirmed by EDS analysis. Functional group analysis revealed the presence of new Fe-O functional groups on the modified membranes. The porosity and water uptake tests showed lower values for the membranes with additives. Performance testing demonstrated high selectivity for oil emulsions, with the M1 membrane exhibiting a selectivity of 98.94%, and M2, with additives, showing an increased selectivity of 99.09%. Additionally, the addition of tannic acid and $FeCl_3$ improved the antifouling properties of the membranes. Membrane M1 achieved an FRR value of 51.66%, whereas M2 showed an improved FRR of 70.61%. These findings confirm the effectiveness of tannic acid and $FeCl_3$ as additives for enhancing the antifouling properties of membranes. Based on these results, future studies should focus on optimizing the concentration of tannic acid and $FeCl_3$ to further improve membrane performance. Additionally, exploring alternative methods of incorporating additives, such as blending or surface grafting, may enhance their distribution and interaction with the membrane matrix. Testing these membranes in real industrial wastewater conditions is also recommended to evaluate their performance in practical applications and to broaden their potential use across various industries.

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