



Effect of Cycling and Washing in Fouling Level from Theaflavin Filtration on Ultrafiltration Membrane

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Abstract

Ultrafiltration membranes, characterized by their molecular weight cutoff ranging from 50 to 100 kDa, are extensively employed for the filtration of macromolecules including suspended solids, carbohydrates, proteins, and various other components. However, the prolonged usage of these membranes leads to a notable decline in their performance due to fouling. Membrane fouling entails the accumulation of filtered foulant media and the formation of deposits, adversely impacting the filtration efficiency. This study focuses on evaluating the impact of repeated filtration cycles on membrane fouling and investigates the effectiveness of membrane washing in restoring membrane performance. The ultrafiltration membrane was subjected to five consecutive repetition cycles using theaflavin as the filtration media. The observed flux exhibited a substantial decline of 87%, decreasing from an initial value of 0.0574 L/m².hr to 0.0073 L/m².hr. To mitigate fouling and enhance membrane performance, a thorough washing process was carried out. Following the washing procedure, the flux was found to increase to 0.034 L/m².hr, indicating a significant improvement in the membrane's performance. These findings are further supported by the morphological analysis of the membrane using Scanning Electron Microscope (SEM) photographs, which reveal the distinct differences between the membrane's morphology before and after washing.

Keywords: Ultrafiltration; Washing; Cycling; Theaflavin

Introduction

In the pursuit of efficient and sustainable separation processes, ultrafiltration (UF) membranes have emerged as invaluable tools for the filtration and purification of diverse biocompounds (Vijayakumar et al., 2008). Over the past few decades, significant progress has been made in the development of UF membranes, accompanied by a comprehensive understanding of the underlying filtration mechanisms and associated fouling phenomena. This convergence of knowledge has enabled researchers to optimize UF membrane performance, thereby enhancing the reliability and efficiency of these filtration systems (Muarif & Mulyawan, 2022).

The development of UF membranes has witnessed a remarkable evolution, driven by the demand for improved separation capabilities, enhanced selectivity, and prolonged operational lifetimes. Extensive research efforts have focused on tailoring membrane properties such as pore size, surface charge, morphology, and surface chemistry, aiming to achieve optimal separation performance for specific applications. These endeavors have resulted in the synthesis of innovative membrane materials, including polymeric, ceramic, and composite membranes, each exhibiting distinct characteristics and performance advantages (Liquids, 2019; Voisin et al., 2017).

Furthermore, the exploration of advanced fabrication techniques has contributed significantly to the advancement of UF membranes. State-of-the-art methods, such as phase inversion, electrospinning, and molecular self-assembly, have revolutionized membrane synthesis, enabling precise control over membrane structure and morphology (Kim et al., 2016; Vriezokolk et al., 2016). Such control has facilitated the development of membranes with enhanced permeability, selectivity, and mechanical stability, crucial for the efficient separation of biocompounds (Dwi Nyamiati et al., 2023). Despite these remarkable advancements, fouling remains a persistent challenge in UF membrane filtration processes. Fouling refers to the accumulation of undesirable substances on the membrane surface or within its pores, leading to a decline in filtration performance over time. It is a complex phenomenon influenced by various



factors, including feedwater quality, operating conditions, membrane properties, and the characteristics of the biocompounds being processed (Kumar et al., 2013; Shen et al., 2019).

Understanding and mitigating fouling is essential for ensuring the long-term sustainability and economic viability of UF membrane systems. Extensive research efforts have been directed towards elucidating the fundamental mechanisms of fouling and developing effective strategies to combat its occurrence. These strategies encompass both preventive and remedial approaches, including the modification of membrane surfaces, the application of pre-treatment techniques, and the implementation of advanced cleaning and regeneration protocols (Shen et al., 2019; Yu et al., 2014).

Theaflavins (TF) are characterized by their high antioxidant content, which confers numerous health benefits to humans. These benefits include the inhibition of cancer cell growth, prevention of conditions such as diabetes, obesity, and overweight, as well as their antiviral properties (Bhardwaj et al., 2020). Extensive research on antivirals and pharmaceutical compounds has highlighted the potential of TF as a candidate for chemical structure screening and as an inhibitor of Mpro, an enzyme crucial to the activity of SARS-CoV-2. Notably, TF demonstrates the ability to bind to the catalytic pocket adjacent to the active Mpro site in SARS-CoV-2, SARS-CoV, and mer-Cov, effectively obstructing their active sites. Furthermore, theaflavin is a constituent catechin compound responsible for imparting black tea with its characteristic color, taste, and aroma (Yulianto et al., 2022).

In this study, we aim to provide an updated and comprehensive overview of the recent advancements in UF membrane development and the strategies employed for mitigating fouling. This study provides valuable insight into the impact of repeated filtration on membrane fouling and highlights the efficacy of membrane washing in rejuvenating membrane performance. The optimization of ultrafiltration membranes and the development of effective fouling mitigation strategies are very promising to increase the overall efficiency and life of these membranes in the biocompound filtration process, the research was confirmed by repeated flux calculations and confirmed by SEM photos to see the level of membrane fouling morphologically.

Material and Methods

Material

This study utilizes tea leaves obtained from PT. Rumpun Sari, Medini, Ungaran, as the primary raw material. The filtration process of Theaflavins involved the utilization of Polyethersulfone Membrane Ultrafiltration MWCO 1000 Da, sourced from Merck.

Methods

Morphology of Membranes

Scanning electron microscopy (SEM) was employed to examine the cross-sectional and surface morphologies of the membranes, both pre- and post-theaflavin treatment. The analysis of membrane surface morphology was conducted using an electron microscope (SEM). In order to facilitate SEM analysis (Hitachi, FlexSEM 1000, Japan) with 5 kVA, the membranes were frozen in liquid nitrogen, fractured, and subjected to a coating process.

Performance of Membrane

The determination of water flux and Theaflavin rejection was conducted utilizing a crossflow system, comprising of essential components such as pressure gauges, a pump, a reservoir, valves, and a flat sheet membrane module (Ramadhani et al., 2021). To calculate the membrane flux, the following equations (1) were employed :

$$J = \frac{Q}{A \cdot \Delta t} \quad (1)$$

Where Q represents the quantity of permeate in liters (L), A denotes the effective membrane area in square meters (m²), and Δt represents the sampling time in hours (h) (Nyamiati et al., 2021).

Backwashing Methods

Membrane backwashing involves the reversal of the filtration process, where the permeate flow is temporarily stopped, and a cleaning solution is introduced on the feed side. The backwash operation aims to dislodge and remove fouling deposits from the membrane surface. The choice of backwash mode with hydraulic and air scouring and parameters flow rate, duration, and frequency plays a crucial role in determining the effectiveness of the backwashing process (Syamani, 2020).

Result and Discussion

Recent advancements in backwashing techniques have led to the development of innovative approaches that offer enhanced fouling reduction and operational efficiency. Backwashing involves the reversal or periodic interruption of the filtration process to remove accumulated foulants from the membrane surface (Moriya et al., 2012). The application of a hydraulic force during backwashing induces shear stress on the membrane, dislodging and removing fouling layers. Membrane backwashing offers a practical and effective approach to reduce fouling and enhance the performance of ultrafiltration membrane filtration processes. Understanding the fouling mechanisms, optimizing

backwashing parameters, and considering the specific requirements of each application are key factors for successful implementation (Asapu et al., 2014).

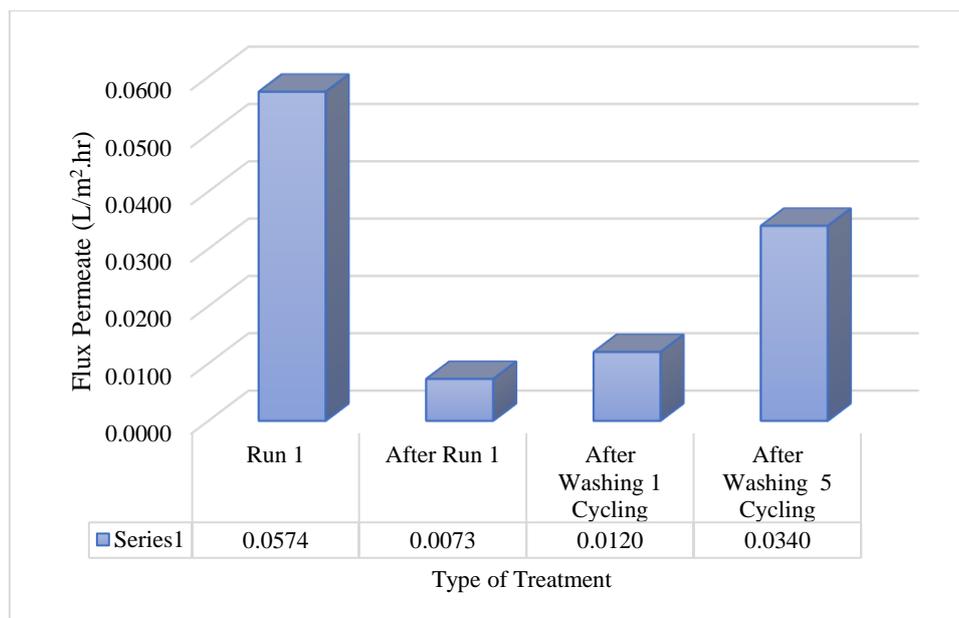


Figure 1. Performance and Flux Permeate with Variation of Treatment

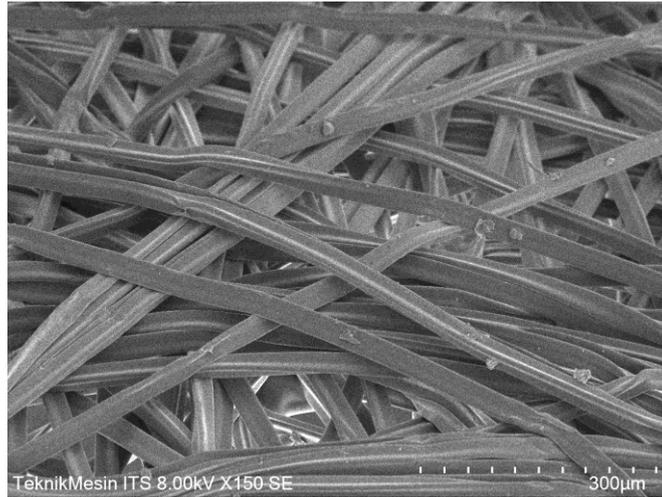
Flux permeate represents the quantity of product obtained per unit membrane area, multiplied by the filtration time. Permeate flux serves as a critical parameter to evaluate the effectiveness of the utilized membrane (Dayarathne et al., 2017). In the filtration of theaflavin biocompound, a straightforward permeate running flux value of 0.574 L/m².hr was initially achieved and can be seen in figure 1. However, over a period of 2 hours, the membrane filtration process witnessed a decline in performance, as evidenced by a reduction in the produced permeate flux, specifically dropping to 0.0073 L/m².hr. This decline signifies an 87.2822% reduction in membrane performance. Consequently, the diminished membrane performance leads to a lower filtration rate and decreased product output.

Table 1. Performance and Flux Permeate with Variation of Treatment

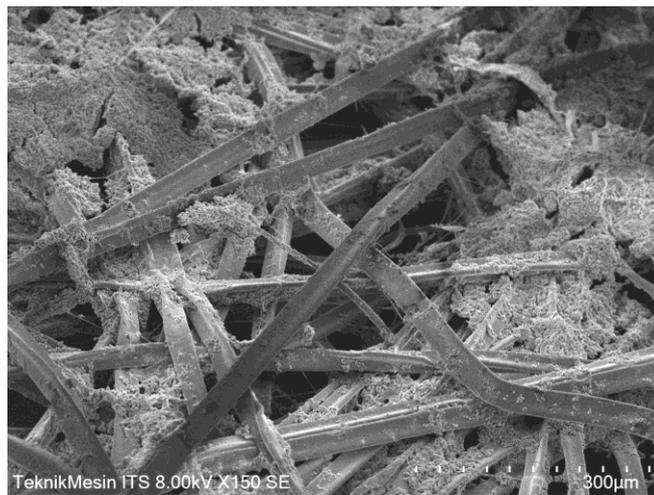
Type of Treatment	Flux Permeate	% Decrease Performance	% Increase Performance After Washing
Run 1	0.0574	-	
After Run 1	0.0073	87.2822	
After Washing 1 Cycling	0.0120	79.0941	9.3812
After Washing 5 Cycling	0.0340	40.7666	53.2934

To rectify this situation, a single backwashing cycle was conducted, whereby the product container feed was filled with washing water. Subsequently, a reverse process was implemented to eliminate any dirt or fouling adhered to the membrane. Following the first washing cycle, an improvement in membrane performance was observed, with an increase of 9.3812% can be seen in Table 1. The corresponding flux value also rose to 0.012 L/m².hr.

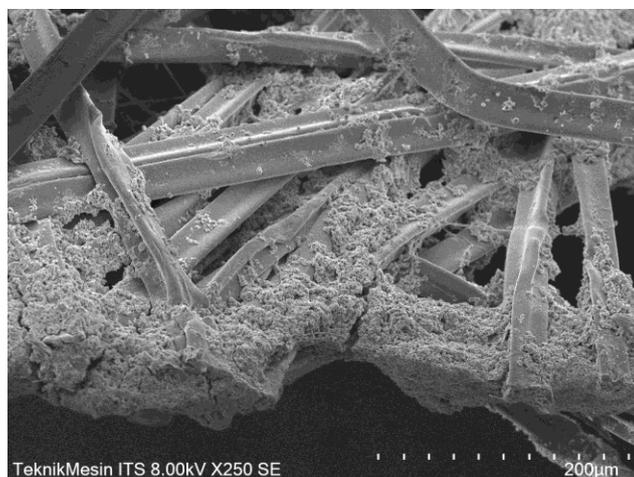
Encouraged by this outcome, the process was further repeated five times, resulting in a performance increase of 53.2934% and a flux value of 0.0340 L/m².hr. By employing this washing method, the adverse effects of fouling in the biocomponent filtration process can be mitigated (Oh et al., 2009).



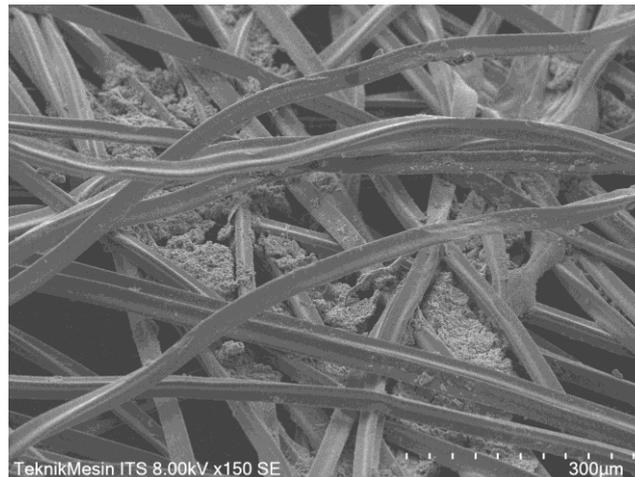
a)



b)



c)



d)

Figure 2. Morphology of Membrane a) Before Filtration; b) After Filtration; c) After Washing 1 Cycling; d) After Washing 5 Cycling

Figure 2 presents the morphological changes of the membrane throughout various stages, including before filtration, after filtration, and after the washing treatment. Figure 2b specifically illustrates the pristine state of the ultrafiltration membrane, characterized by well-organized and unblemished fibers. Upon undergoing the filtration process (as shown in Figure 2b), the membrane exhibits cake buildup resulting from the separation and retention of dissolved solutes. The corresponding cake percentage is detailed in Table 2, indicating a cake formation of 58.245%. Subsequently, after a single washing cycle, the membrane's cake content decreases significantly to 21.471%. Furthermore, when the membrane undergoes five washing cycles, a substantial reduction is observed, with the cake content reducing to a mere 4.606%. These results highlight the remarkable effectiveness of employing the five-times washing method in mitigating membrane fouling levels (Jhaveri & Murthy, 2016).

Table 3. Mass and % cake of membran with variation of treatment

Type of Treatment	Mass of Membrane (gr)	% Cake
Run 1	0.076	
After Run 1	0.182	58.245
After Washing 1 Cycling	0.097	21.471
After Washing 5 Cycling	0.079	4.606

Conclusion

Membrane backwashing represents a crucial technique in combating fouling issues in ultrafiltration processes. Through the reversal of filtration flow, backwashing dislodges and removes accumulated foulants, thereby restoring membrane performance and prolonging its lifespan. Membrane backwashing offers a practical and effective approach to reduce fouling and enhance the performance of ultrafiltration membrane filtration processes. Understanding the fouling mechanisms, optimizing backwashing parameters, and considering the specific requirements of each application are key factors for successful implementation. Further research and development efforts should focus on exploring innovative strategies and technologies to achieve sustainable and efficient membrane fouling control. After conducting a thorough analysis of five washing cycles, it was determined that the membrane's performance improved significantly by up to 53.293%. This conclusion is substantiated by the examination of membrane morphology through scanning electron microscopy (SEM), which clearly reveals the beneficial impact of washing in reducing the cake formation on the membrane from an initial level of 58.245% to a mere 4.606%.

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Notation List

J	= Flux Permeate [L/m ² .hr]
Q	= Quantity of Permeate [L]
A	= membrane area [m ²]
Δt	= Sampling time [h]

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