

HUMIC ACID FILTRATION EFFICIENCY OF POLYPROPYLENE-BASED ULTRAFILTRATION MEMBRANES

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Abstract

Polypropylene (PP) membranes exhibit advantageous characteristics for filtration processes, including chemical resistance, thermal stability, hydrophobicity, low cost, and mechanical durability, making them well-suited for long-term applications in water treatment, particularly for removing organic compounds. This study aims to evaluate the performance of hollow fiber PP membranes in the ultrafiltration of humic acid solutions, serving as a model organic contaminant. The experimental setup involved filtering a 15 mg/L humic acid solution in demineralized water at 1 bar for 120 minutes. Performance evaluation focused on three parameters: pure water flux, humic acid permeate flux, and rejection efficiency. The initial pure water flux reached 72 L/m²·h, decreasing to approximately 57 L/m²·h within the first 20 minutes, indicating membrane compaction. The humic acid permeate flux declined from 42 to 33 L/m²·h, attributed to fouling caused by physical and chemical interactions between humic acid and the PP membrane surface. Rejection efficiency dropped from an initial 90% to 75% by the end of the process. The observed performance degradation was linked to adsorption, pore blocking, and concentration polarization. Despite their structural advantages, PP membranes are prone to organic fouling; therefore, surface modification and feedwater pretreatment are recommended to optimize long-term performance.

Kata kunci: Flux, Fouling, Humic Acid, Polypropylene, Rejection

Abstrak

Membran polipropilena (PP) memiliki potensi besar dalam aplikasi filtrasi karena sifat ketahanannya terhadap bahan kimia, stabilitas termal, dan karakteristik hidrofobik. Kombinasi sifat ini menjadikan membran PP sangat cocok untuk memisahkan senyawa organik, seperti asam humat, dalam proses pengolahan air. Selain itu, biaya produksi yang rendah serta ketahanan mekanis yang tinggi membuatnya ideal untuk penggunaan jangka panjang. Penelitian ini bertujuan untuk mengevaluasi kinerja membran PP tipe *hollow fiber* dalam proses ultrafiltrasi larutan asam humat sebagai representasi kontaminan organik. Proses filtrasi dilakukan menggunakan larutan asam humat 15 mg/L dalam air demineralisasi pada tekanan operasi 1 bar selama 120 menit. Evaluasi dilakukan terhadap tiga parameter utama, yaitu fluks air murni, fluks permeat asam humat, dan efisiensi rejeksi. Hasil menunjukkan fluks awal air murni sebesar 72 L/m²·h menurun menjadi sekitar 57 L/m²·h dalam 20 menit pertama akibat kompaksi membran. Fluks asam humat menurun dari 42 menjadi 33 L/m²·h, menunjukkan terjadinya *fouling*. Rejeksi awal sebesar 90% menurun menjadi 75%. Penurunan ini disebabkan oleh adsorpsi, penyumbatan pori, dan pembentukan lapisan polarisasi konsentrasi. Oleh karena itu, diperlukan strategi modifikasi permukaan dan pretreatment air umpan untuk meningkatkan performa jangka panjang.

Kata kunci: Asam Humat, Fluks, Fouling, Polipropilena, Rejeksi

1. INTRODUCTION

Water is a vital natural resource for human life and the environment. However, increased industrial, agricultural, and urbanization activities have led to water pollution becoming a serious environmental problem worldwide. One of the primary contaminants in surface water and groundwater is natural organic matter (NOM), especially

humic acid (HA). This compound is a significant component of dissolved organic matter from microbial and plant decomposition. Due to its chemical stability and complex molecular structure, it is challenging to remove through conventional water treatment processes.

Humic acids are aromatic, containing functional groups such as carboxylate (-COOH)

and hydroxyl (-OH), and play an important role in forming complex compounds with heavy metals, increasing water color, and even becoming precursors for the formation of harmful organic compounds such as trihalomethanes during chlorine disinfection processes (Shao, Hou and Song, 2011a). Therefore, efficiency and effectiveness in removing humic acid from water are crucial to ensure clean water quality and public health.

Membrane technology, particularly ultrafiltration (UF), has rapidly evolved as a promising method for water treatment due to its ability to separate medium-sized organic molecules without additional chemicals. This process works through size-exclusion and surface-interaction mechanisms between the membrane and contaminants. Ultrafiltration offers low energy consumption, high efficiency, and relatively simple operation. (Li, Jiang and Li, 2018; Mundarti, 2019; Wan Osman et al., 2021).

UF membranes made from various polymer materials, such as polyethersulfone (Ahmad et al., 2019; Ambarita et al., 2021), cellulose acetate, polyacrylonitrile, and polysulfone (Mozia, Tomaszewska and Morawski, 2005) have been used for filtering organic compounds, especially humic acid. Filtration results show that the permeate flux and rejection are quite high. However, research on polypropylene (PP) membranes for filtering organic compounds remains limited. However, this membrane has many advantages due to its strong mechanical properties, thermal stability, chemical resistance, low cost, and easy availability (Ariono and Wardani, 2017; Yang et al., 2024). Despite its potential, limited research has been conducted on the use of PP membranes for HA removal via ultrafiltration, which this study seeks to address.

Therefore, research was conducted to evaluate the performance of PP hollow fiber membranes in the ultrafiltration process of humic acid solutions. The evaluated performance included measurements of pure water flux, humic acid permeate flux, and humic acid rejection during filtration. This data is important for understanding the dynamics of fouling and the effectiveness of humic acid removal by PP membranes. It also provides input for developing fouling mitigation strategies, such as modifying the membrane surface to be more hydrophilic or negatively charged.

2. METHODS

2.1 Materials

The PP hollow fiber membrane used was obtained from GDP Filter, Indonesia, with the following technical specifications: outer diameter of 0.4 mm, inner diameter of 0.3 mm, and an average pore size of 50 nm. The model solution was prepared by dissolving 15 mg/L of humic acid (HA, Sigma-Aldrich, purity $\geq 98\%$) in demineralized water.

2.2 Filtration Process

The pure water flux was determined by supplying demineralized water to the hollow fiber membrane operated in a dead-end configuration. Measurements were performed at 1 bar pressure at room temperature (Figure 1). Subsequently, membrane performance was evaluated by filtering a 15 mg/L humic acid solution for 120 minutes.

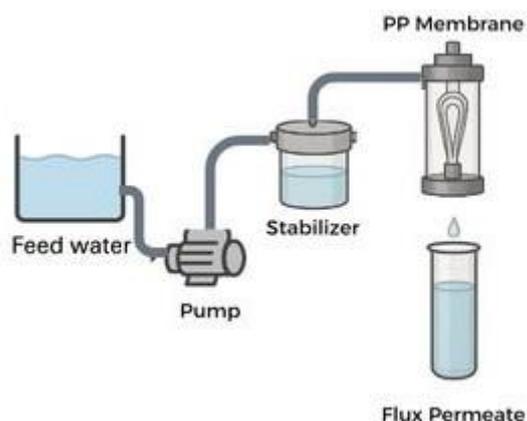


Figure 1. Experimental setup of flux measurement

The performance of the membrane system was evaluated based on two key parameters: permeate flux and rejection rate.

$$J_p = \frac{v}{A \cdot \Delta t} \quad (1)$$

$$\% \text{ Rejection} = \left(1 - \frac{c_p}{c_f}\right) \times 100 \quad (2)$$

Here, permeate flux (J_p) is defined as the permeate volume (v) per membrane area (A) over time (t) (Equation 1). Meanwhile, the rejection percentage was determined using Equation (2), where C_p represents the solute concentration in the permeate, and C_f represents the solute concentration in the feed.

2.3 Characterisation

Humic acid concentrations before and after filtration were analyzed using a UV-Vis spectrophotometer (Spectronic 20D, Thermo Fisher Scientific, USA). Spectrophotometric analysis was performed over the wavelength range of 200–800 nm, covering both UV and visible regions

3. RESULTS AND DISCUSSION

3.1 Performance of Polypropylene Membrane on Pure Water Flux

The pure water flux was analyzed to evaluate the membrane's basic permeability properties before use under actual operating conditions, namely, the filtration of humic acid compounds. This parameter reflects the efficiency of water transport through the membrane, free of contaminant obstruction, and provides an initial overview of the membrane's performance. Figure 2 shows a pure water flux of approximately $72 \text{ L/m}^2 \cdot \text{h}$ at minute 0. At minute 20, the pure water flux decreased to $\pm 57 \text{ L/m}^2 \cdot \text{h}$. After that, the flux stabilized at $53\text{--}58 \text{ L/m}^2 \cdot \text{h}$, with small fluctuations, until minute 120.

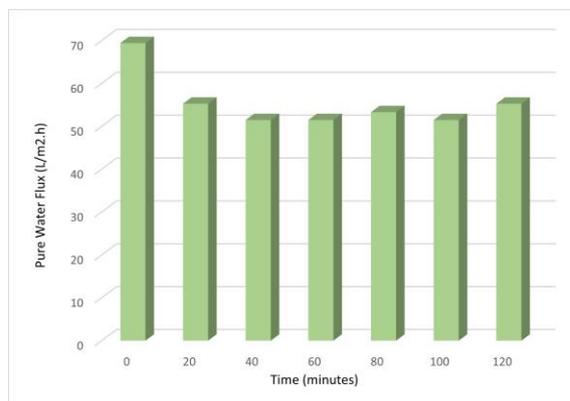


Figure 2. Pure Water Flux of PP Membrane

Although the graph shows testing using demineralized water (free from contaminants and dissolved particles), the pure water flux still experiences an initial decline. This phenomenon can be explained by internal physical phenomena of the membrane, not by fouling from external particles.

High initial flux is often caused by greater surface tension at the beginning of the test. However, water causes a reorganization of the membrane's polymer structure over time, especially in polymer-based membranes such as polysulfone, which can undergo mechanical compression or "membrane compaction" under constant operating pressure (Ou et al., 2017).

In addition, water adsorption on the membrane's pore walls can change the internal surface tension and reduce the membrane's effective porosity, even in the absence of solutes (Abdoli et al., 2018).

3.2 Performance of Polypropylene Membrane on Permeation

The permeate flux was evaluated by filtering a humic acid solution through a polypropylene (PP) membrane at room temperature under 1 bar pressure for 120 minutes. As shown in Figure 3, the flux declined from approximately $42 \text{ L/m}^2 \cdot \text{h}$ to $33 \text{ L/m}^2 \cdot \text{h}$ over the duration, a typical trend in ultrafiltration due to membrane fouling by organic matter.

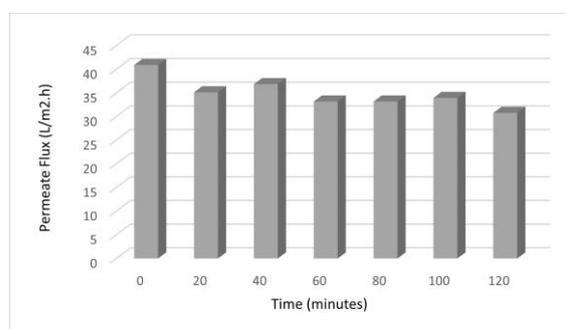


Figure 3 Permeate Flux of Humic Acid

The initial sharp decrease in flux is attributed to rapid surface fouling, where humic acid particles accumulate and clog membrane pores. This phase is followed by a semi-stable period, where a fouling layer establishes constant resistance, slowing further blockage (Abdoli et al., 2018; Tataru et al., 2018).

Minor fluctuations after 40 minutes may result from pressure variations or temporary fouling redistribution. Studies also note that increased membrane pore size and hydrophilicity enhance initial flux but do not prevent overall fouling (Ou et al., 2017).

Humic acid, a complex organic molecule with aromatic rings and functional groups like carboxyl (-COOH) and phenol (-OH), interacts physically and chemically with membranes (Figure 4). In PP membranes, which are hydrophobic and neutrally charged, non-polar segments of humic acid are attracted via Van der Waals forces and hydrophobic interactions, leading to adsorption and formation of a dense fouling layer (Yuan and Zydney, 2000; Koh,

Clark and Ishida, 2006; Shao, Hou and Song, 2011a).

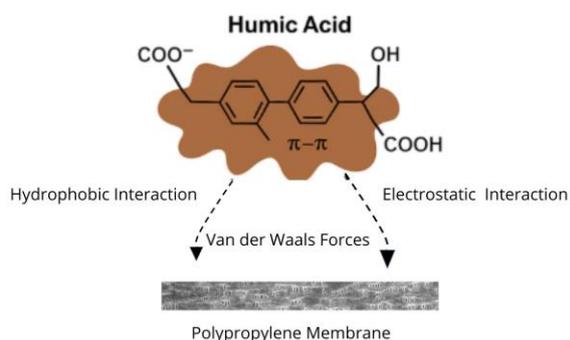


Figure 4. Interaction between PP Membrane and Humic Acid

Additionally, fouling occurs through pore blocking, where small humic molecules (0.5–5 nm) penetrate membrane pores, causing standard, complete, or intermediate blocking (Saha and an Das, 2015). Concentration polarization further exacerbates flux decline by forming a high-concentration boundary layer near the membrane, increasing osmotic pressure, and reducing the effective filtration gradient (Liu et al., 2022).

Chemically, humic acid's functional groups can engage in hydrogen bonding or weak electrostatic interactions, particularly under neutral to basic pH, where the molecules carry a negative charge. These interactions are more likely if the membrane surface contains reactive groups, either from manufacturing impurities or mild oxidation (Saha and an Das, 2015).

3.3 Performance of Polypropylene Membrane on Humic Acid Rejection

Although the PP membrane does not significantly affect permeate flux, it substantially rejects humic acid. At the beginning of filtration shown in Figure 5, humic acid rejection reached 90%, then gradually decreased to around 75% after 120 minutes of operation.

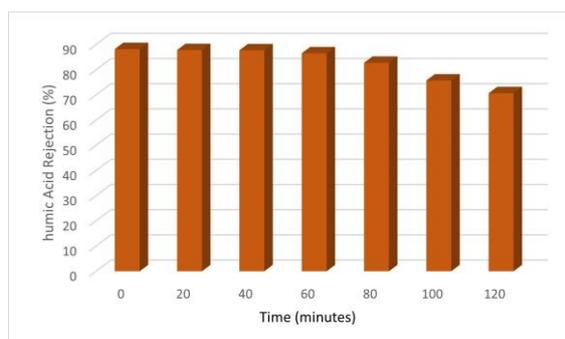


Figure 5. Humic Acid Rejection of PP Membrane

The rejection of humic acid by ultrafiltration membranes is determined by two main factors: the pore size and distribution of the membrane and the surface interaction between the membrane and humic acid molecules.

Humic acid is a complex organic molecule with varying molecular sizes between 0.5 and 5 nm and a negative charge. Rejection at the beginning of filtration is high due to the size exclusion effect, where larger particles cannot pass through the membrane pores. New or clean membranes also show optimal separation efficiency (Shao, Hou and Song, 2011b).

The decrease in humic acid rejection shown in the graph indicates fouling and changes in membrane surface dynamics. Organic fouling by humic acid can lead to several effects: (1) Pore deformation or channel expansion due to long-term pressure (Zhu et al., 2018), (2) Adsorption of humic acid that changes the surface charge and hydrophobicity of the membrane, (3) accumulation of a fouling layer (cake layer) that interferes with selective flow and creates leakage paths (Combe et al., 1999; Samreen et al., 2025).

PP membranes are neutral and hydrophobic, theoretically less than ideal for handling polar organic contaminants such as humic acid. Hydrophobic interactions tend to increase the adsorption of humic acid to the membrane surface, damaging its selective structure. In addition, PP membranes do not have a negative electrostatic charge that could promote rejection based on charge repulsion, as do membranes made of polyacrylate or oxidized cellulose (Prorok et al., 2023).

4. CONCLUSION

This study shows that hollow fiber polypropylene (PP) membranes have a relatively good initial performance in the

ultrafiltration process of humic acid solutions. Pure water flux initially reached 72 L/m²·h but decreased due to membrane compaction phenomena. Humic acid permeate flux also showed a significant decrease from 42 to 33 L/m²·h over 120 minutes, which was indicated as a result of fouling due to physical and chemical interactions between humic acid and the membrane surface. The initial rejection of humic acid at 90% decreased to 75%, indicating a degradation of separation performance due to pore clogging and concentration layer formation.

The hydrophobic and electrically neutral characteristics of polypropylene (PP) membranes are significant contributors to membrane fouling by polar organic compounds such as humic acid. Key mechanisms responsible for flux decline include surface adsorption, pore blocking, and concentration polarization. While PP membranes are advantageous due to their mechanical robustness, chemical resistance, and low production cost, their inherent hydrophobicity necessitates surface modification to enhance hydrophilicity and mitigate fouling. These findings contribute to developing more efficient and durable membrane-based water treatment systems for removing dissolved organic contaminants like humic substances.

REFERENCE

- Abdoli, S.M., Shafiei, S., Raoof, A., Ebadi, A., Jafarzadeh, Y. and Aslannejad, H., (2018). Water flux reduction in microfiltration membranes: a pore network study. *Chemical Engineering & Technology*, [online] 41(8), pp.1566–1576. <https://doi.org/10.1002/CEAT.201800130>.
- Ahmad, A.L., Pang, W.Y., Mohd Shafie, Z.M.H. and Zaulkiflee, N.D., (2019). PES/PVP/TiO₂ mixed matrix hollow fiber membrane with antifouling properties for humic acid removal. *Journal of Water Process Engineering*, [online] 31, p.100827. <https://doi.org/https://doi.org/10.1016/j.wpe.2019.100827>.
- Ambarita, A.C., Mulyati, S., Arahman, N., Bilad, M.R., Shamsuddin, N. and Ismail, N.M., (2021). Improvement of Properties and Performances of Polyethersulfone Ultrafiltration Membrane by Blending with Bio-Based Dragonbloodin Resin. *Polymers*, 13(24). <https://doi.org/10.3390/polym13244436>.
- Ariono, D. and Wardani, A.K., (2017). Review of hydrophilic PP membrane for organic waste removal. *AIP Conference Proceedings*, [online] 1840(1), p.90002. <https://doi.org/10.1063/1.4982310>.
- Combe, C., Molis, E., Lucas, P., Riley, R. and Clark, M.M., (1999). The effect of CA membrane properties on adsorptive fouling by humic acid. 154.
- Koh, M., Clark, M.M. and Ishida, K.P., (2006). Adsorptive fouling of a polypropylene microfiltration membrane with dissolved natural organic matter: do membranes possess an adsorption capacity? *Water Science & Technology: Water Supply*, [online] 6(2), pp.25–30. <https://doi.org/10.2166/WS.2006.044>.
- Li, X., Jiang, L. and Li, H., (2018). Application of Ultrafiltration Technology in Water Treatment. *IOP Conference Series: Earth and Environmental Science*, [online] 186(3), p.12009. <https://doi.org/10.1088/1755-1315/186/3/012009>.
- Liu, B., Yang, K., Li, G.Z. and Shi, Z., (2022). Alleviation of Ultrafiltration Membrane Fouling by ClO₂ Pre-Oxidation: Fouling Mechanism and Interface Characteristics. *Membranes*, [online] 12(1), p.78. <https://doi.org/10.3390/membranes1201078>.
- Mozia, S., Tomaszewska, M. and Morawski, A.W., (2005). Studies on the effect of humic acids and phenol on adsorption–ultrafiltration process performance. *Water Research*, [online] 39(2), pp.501–509. <https://doi.org/https://doi.org/10.1016/j.watres.2004.10.012>.
- Mundarti, S., (2019). Pengaruh Coating Polidopamin Pada Membran Polipropilena Untuk Filtrasi Senyawa Organik. Master's Thesis, Institut Teknologi Bandung.
- Ou, R., Wang, Y., Zhang, H., Wang, H. and Xu, T., (2017). Preparation of high-flux ultrafiltration membranes by blending strongly charged polymer. *Journal of Applied Polymer Science*, [online] 134(10), p.44570. <https://doi.org/10.1002/APP.44570>.
- Prorok, V., Movrin, D., Lukić, N.L. and

- Popovic, S., (2023). New Insights into the Fouling of a Membrane during the Ultrafiltration of Complex Organic–Inorganic Feed Water. *Membranes*, [online] 13(3), p.334. <https://doi.org/10.3390/membranes13030334>.
- Saha, S. and an Das, (2015). Analysis of Fouling Characteristics and Flux Decline during Humic Acids Batch Ultrafiltration. *Journal of Chemical Engineering & Process Technology*, [online] 6(5), pp.1–7. <https://doi.org/10.4172/2157-7048.1000252>.
- Samreen, S., Satyanarayana, C.V. V, Edukondalu, L., Beera, V. and Rao, V.S., (2025). Membrane Fouling Dynamics in the Clarification of Pomegranate Juice Using Egg Albumin Pretreatment: A Study on Hermia's Empirical Models. *Archives of current research international*, [online] 25(1), pp.134–150. <https://doi.org/10.9734/acri/2025/v25i11045>.
- Shao, J., Hou, J. and Song, H., (2011a). Comparison of humic acid rejection and flux decline during filtration with negatively charged and uncharged ultrafiltration membranes. *Water Research*, [online] 45(2), pp.473–482. <https://doi.org/10.1016/J.WATRES.2010.09.006>.
- Shao, J., Hou, J. and Song, H., (2011b). Comparison of humic acid rejection and flux decline during filtration with negatively charged and uncharged ultrafiltration membranes. *Water Research*, [online] 45(2), pp.473–482. <https://doi.org/10.1016/j.watres.2010.09.006>.
- Tataru, L., Nedeff, V., Barsan, N., Mosnegutu, E., Lehadus, M.P., Sandu, I. and Chitimus, D., (2018). Studies of Humic Acid Removal from Aqueous Systems by Using Polymeric Membrane Ultrafiltration Process. *Materiale Plastice*, [online] 55(4), pp.680–685. <https://doi.org/10.37358/MP.18.4.5100>.
- Wan Osman, W.N.A., Mat Nawati, N.I., Samsuri, S., Bilad, M.R., Khan, A.L., Hunaepi, H., Jaafar, J. and Lam, M.K., (2021). Ultra low-pressure filtration system for energy efficient microalgae filtration. *Heliyon*, [online] 7(6). <https://doi.org/10.1016/j.heliyon.2021.e07367>.
- Yang, L., Man, S.Y., Zhao, N., Li, J., Bo, S. and Zhang, M., (2024). Hot-Pressed Polypropylene (PP) Membranes Modified With SiO₂ Nanoparticles for Advanced Emulsified Oil/Water Separation. *Journal of Applied Polymer Science*. <https://doi.org/10.1002/app.56602>.
- Yuan, W. and Zydney, A.L., (2000). Humic acid fouling during ultrafiltration. *Environmental Science & Technology*, [online] 34(23), pp.5043–5050. <https://doi.org/10.1021/ES0012366>.
- Zhu, R., Diaz, A.J., Shen, Y., Qi, F., Chang, X., Durkin, D.P., Sun, Y., Solares, S.D. and Shuai, D., (2018). Mechanism of humic acid fouling in a photocatalytic membrane system. *Journal of Membrane Science*, [online] 563, pp.531–540. <https://doi.org/https://doi.org/10.1016/j.memsci.2018.06.017>.