
THE POTENTIAL OF CNT-PVA MEMBRANE AS A GREEN TECHNOLOGY ALTERNATIVE

Henny Parida Hutapea^{1*}, Septiana Ambarwati², Rafika Aulia Putri³, Michell Aulia⁴

Duta Bangsa University^{1,2,3,4}

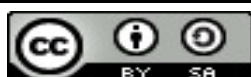
*Correspondence Email: henny_paridahutapea@udb.ac.id

ABSTRACT

The development of advanced membrane technologies is essential for addressing environmental challenges, particularly in wastewater treatment and oil-water separation. Polyvinyl alcohol (PVA) is a hydrophilic, environmentally friendly polymer commonly used for membrane fabrication; however, it suffers from high swelling and low mechanical strength, which limits its long-term performance. To overcome these drawbacks, carbon nanotubes (CNTs) were incorporated into the PVA matrix to form PVA-CNT composite membranes. CNTs serve as structural reinforcements, enhancing mechanical strength, thermal stability, and inter-chain bonding within the PVA network. The interconnected pore structure and high conductivity of CNTs also improve membrane selectivity and reduce swelling by stabilizing the polymer structure when exposed to water or aqueous solutions. Additionally, the modified surface properties provided by CNTs result in superior fouling resistance, minimizing pore blockage and maintaining water flux over extended operation periods. This study demonstrates that PVA-CNT composite membranes offer a sustainable and energy-efficient solution for separation processes, making them highly suitable for oil-water separation, industrial wastewater treatment, and other filtration applications such as water purification and organic compound removal. The findings highlight the potential of PVA-CNT membranes as a green technology that combines high performance with environmental sustainability. PVA-CNT membranes offer a promising approach for developing next-generation membranes with optimized structural and functional properties for environmental applications.

KEYWORDS

Carbon Nanotube; Polyvinyl Alcohol; Green Technology; Sustainable Materials



This work is licensed under a Creative Commons Attribution-ShareAlike 4.0 International

INTRODUCTION

In recent decades, membrane technology has been increasingly utilized in various separation fields due to its several advantages, such as no phase change, compact system size, and high automation. Based on the principle of size exclusion, membranes can retain almost all particles larger than their pore size, providing high separation efficiency. Membrane separation technology has emerged as a key method in oil–water separation research because of its high separation efficiency, low cost, and ease of operation. This review article discusses recent advancements in various types of separation membranes, including emulsion permeance performance, separation efficiency, antifouling properties, and stimulus responsiveness, while also outlining the challenges faced in the development of this technology (Huang et al., 2024).

In general, PVA possesses unique characteristics such as water solubility, non-toxicity, chemical stability, and excellent electrical, optical, and dielectric properties (Shah et al., 2024). Thin PVA-based composite membranes were fabricated by casting a crosslinked PVA layer onto a porous polyacrylonitrile (PAN) substrate. Various PVA concentrations (2.5–15 wt.%) were tested to evaluate their effect on the pervaporation performance of an ethanol/water mixture (80/20 wt.%) at 60°C. The results showed that increasing the PVA concentration led to a decrease in hydrophilicity, as well as an increase in membrane thickness and swelling degree. Consequently, membrane selectivity significantly improved, while the permeation flux decreased. A PVA concentration of 10 wt.% was found to provide the optimal separation performance (Thai et al., 2021). The permeability of pure PVA membrane increased from 1.95×10^{-3} Darcy at 20 psi to 2.08×10^{-3} Darcy at 30 psi, then slightly decreased to 2.00×10^{-3} Darcy at 45 psi due to membrane compaction at higher pressure (Siti Ajizah, 2023).

Recently, carbon nanotube (CNT) membranes with a buckypaper-like structure have attracted significant attention due to their high porosity and interconnected pore network, which provide superior performance compared to conventional polymer membranes. Carbon-based materials are excellent candidates for oil spill cleanup because of their superhydrophobic properties, large surface area, chemical stability, low density, recyclability, and high selectivity. This study developed oil-absorbing membranes based on polystyrene (PS) nanocomposites reinforced with CNTs using the electrospinning technique. The fabricated membranes were then exposed to gamma irradiation to create strong crosslinks, thereby enhancing the interactions between the polymer and CNTs (Parangusan et al., 2019).

CNTs themselves are cylindrical structures that have high mechanical strength and conductivity, so they can improve the electrical properties of composites (Shah et al., 2024). Previous studies have shown that CNT membranes exhibit higher flux and excellent rejection efficiency compared to conventional polymer membranes. The interconnected pore structure of CNT membranes provides greater resistance to pore blockage caused by contaminants. In contrast, conventional polymer membranes have isolated pores that are more easily and completely clogged by impurities. Consequently, CNT membranes are expected to deliver superior performance in filtering oil emulsion wastewater.

In this study, various CNT concentrations and gamma irradiation doses were tested to evaluate the structure, morphology, surface properties, and oil/water separation efficiency. The results revealed that at a CNT concentration of 0.5 wt.%, the membrane demonstrated optimal hydrophobicity, high oil selectivity, and the best mechanical strength, especially at a gamma irradiation dose of 15 KGy. Furthermore, the irradiated PS/0.5 wt.% CNT membrane exhibited antibacterial activity against *Escherichia coli*.

These findings confirm the potential environmental application of the developed PS/CNT membranes in treating oil-contaminated water (Parangusan et al., 2019).

Glutaraldehyde (GA) acts as a crosslinking agent that connects PVA chains through a reaction between the aldehyde (-CHO) groups of GA and the hydroxyl (-OH) groups of PVA, forming acetal bonds and creating a three-dimensional network. In the PVA/PQ-10 membrane, GA exclusively crosslinks the PVA chains, while the PQ-10 chains are physically entrapped, resulting in a semi-interpenetrating network (s-IPN) structure. This crosslinking process enhances the membrane's mechanical and thermal stability while reducing swelling and water uptake. However, excessive GA can decrease ionic conductivity and flexibility, making it essential to optimize the GA concentration to achieve the best membrane performance (Yang et al., 2021).

The ultrafiltration membrane based on carbon nanotube (CNT) and polyvinyl alcohol (PVA) composites possess conductive properties that help reduce organic fouling. By applying an electric current during the filtration process, the membrane demonstrates improved performance in preventing the accumulation of organic matter, which typically leads to decreased efficiency. The results show that the CNT-PVA membrane exhibits higher permeability, better fouling resistance, and an enhanced ability to recover water flux after cleaning compared to conventional polymer membranes, making it a promising solution for treating wastewater containing organic contaminants (Dudchenko et al., 2014).

The synthesis of buckypaper membranes based on multi-walled carbon nanotubes (MWCNTs) infiltrated with polyvinyl alcohol (PVA) was carried out for strain sensor applications. The MWCNT buckypaper was fabricated using a vacuum filtration method, followed by infiltration with PVA solution to enhance mechanical strength, flexibility, and structural stability without compromising the electrical conductivity of the MWCNTs. The results indicate that the addition of PVA strengthens the inter-CNT bonding, producing a more durable and robust material with high sensitivity to strain changes. Therefore, the MWCNT-PVA composite shows great potential for use in flexible sensor applications, such as health monitoring devices, robotics, and wearable technologies (Yee et al., 2018).

An alternative approach was explored to form covalent cross-links in PVA/CNT composites, where the cross-linking process occurs exclusively on the poly(vinyl alcohol) (PVA) macromolecules without involving the nanotube structures. This process was carried out by adding glutaraldehyde to the aqueous PVA/CNT dispersion. The glutaraldehyde then reacts with the PVA chains through an acetalization reaction mechanism, as illustrated in the following scheme (Basiuk et al., 2009):

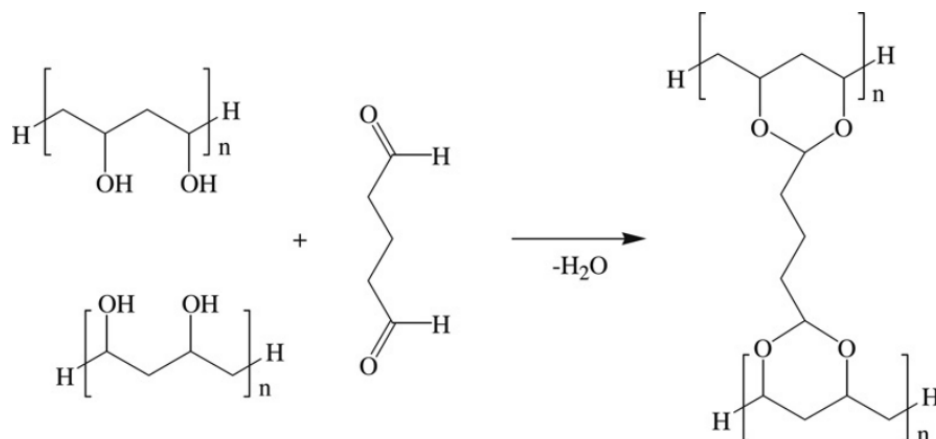


Figure 1. cross-links in PVA/CNT composites scheme

RESEARCH METHOD

CNT-PVA composite membranes were made using a modified method from the research of M.R. Wiesner and David Jassby. First, pure multi-walled CNT (MWCNT) was functionalized using concentrated nitric acid (HNO₃) and concentrated sulfuric acid (H₂SO₄). A total of 3 grams of MWCNT powder was mixed into a mixture of 60 mL HNO₃ and 20 mL H₂SO₄, then heated at 60°C while stirring vigorously for 30 minutes. After cooling to room temperature, the solution was diluted with water, then the CNTs were separated by filtration and washed with ultrapure water until the pH was nearly neutral. The resulting CNTs were then dried at 60°C for 12 hours (Yi et al., 2018).

The preparation of a 10% (w/v) PVA solution was carried out by weighing 10 g of Polyvinyl Alcohol (PVA) powder and dissolving it in 100 mL of distilled water. The dissolution process was performed gradually by heating approximately 80–90% of the total water volume on a hotplate while stirring with a magnetic stirrer at a temperature of 80–90 °C. Once the temperature was stable, the PVA powder was slowly added in small portions to the hot water while continuously stirring to prevent clumping. Heating and stirring were continued until the PVA was completely dissolved, which was indicated by the formation of a clear and homogeneous solution, typically requiring 30–60 minutes. After complete dissolution, the solution was cooled to room temperature (25–30 °C) with gentle stirring. If evaporation occurred during the heating process, the final volume was adjusted by adding distilled water to reach the desired volume. The PVA solution was then stored in a tightly sealed bottle to prevent contamination and evaporation and could be kept in a refrigerator (4–8 °C) if not used immediately (Choo et al., 2016).

RESULT AND DISCUSSION

The following table presents a summary of the characteristics of membranes made from PVA, CNT, and PVA-CNT composites. The comparison includes several key parameters such as elongation, swelling, selectivity, and flux, which are essential for evaluating membrane performance in separation processes. This comparison highlights the advantages of incorporating CNT into PVA membranes to enhance mechanical strength, reduce swelling, and improve selectivity. The sources of data for each characteristic are also provided for reference.

Table 1. Study on the Characteristics of PVA-CNT Based Membranes

| Membrane | Elongation (mm) | TGA (°C) | Swelling (%) | Selectivity |
|-------------------|--------------------|-------------|-----------------|-------------|
| PVA | 38,992 | | 60 | |
| PVA-CNTs 5% | 20,788 | | | |
| PVA-CNTs 0,5%-ZnO | 31,350 | | | |
| PVA-CNTs 1%-ZnO | 36,115 | | | |
| PVA 10% | | | 60 | 65 |
| PVA 15% | | | 65 | 80 |
| PVA-CNTs | | 300 | | |
| PVA-MWNT | | 600 | 76 | |

The comparative data presented in the table shows clear differences in the performance characteristics of pure PVA membranes, CNT-based membranes, and PVA-CNT composite membranes. Each type of membrane has unique properties that influence its suitability for specific separation processes.

Pure PVA membranes exhibit good film-forming ability and hydrophilic properties, making them suitable for applications involving water treatment. However, their

high swelling degree indicates excessive water uptake, which can lead to reduced structural integrity and compromised selectivity. Additionally, PVA membranes tend to have moderate flux but lower mechanical strength, as reflected in their relatively low elongation values. These limitations make pure PVA membranes less ideal for long-term or high-pressure operations.

In contrast, CNT-based membranes demonstrate superior mechanical strength and porosity due to their interconnected pore structure. Their high elongation values indicate better flexibility and durability, while their inherently hydrophobic surface minimizes water uptake, resulting in low swelling. CNT membranes also exhibit higher flux and rejection efficiency because the interconnected pores facilitate faster water transport and prevent pore blockage. However, on their own, CNT membranes may lack sufficient structural stability and may require a polymeric matrix for enhanced integrity.

The PVA-CNT composite membranes combine the advantages of both materials. The integration of CNT into the PVA matrix significantly improves mechanical properties, as shown by increased elongation and reduced brittleness. The addition of CNT also decreases the swelling degree by limiting water penetration, leading to enhanced dimensional stability. Moreover, the composite membrane shows higher selectivity due to improved interfacial interactions between the polymer and CNT, which create more efficient separation pathways. While the flux may slightly decrease compared to pure CNT membranes due to the denser structure formed by PVA, the overall performance is balanced, making PVA-CNT membranes highly suitable for oil-water separation and other filtration processes.

The addition of carbon nanotubes (CNTs) to polyvinyl alcohol (PVA) can significantly enhance the mechanical properties of the composite material, such as yield stress, yield load, and Young's modulus. This indicates that the material becomes stronger and stiffer. However, this improvement comes at the cost of a decrease in elongation at break and yield strain, meaning the material becomes less ductile and more brittle.

This phenomenon occurs because CNTs possess exceptional stiffness and strength due to their sp^2 carbon-carbon bonds. Within the PVA matrix, CNTs form a network structure that helps distribute stress and load more evenly throughout the material. Moreover, CNTs can induce crystallization in the PVA matrix, increasing its crystallinity and crosslinking density, which further contributes to the material's strength and rigidity.

On the other hand, the presence of CNTs can also introduce defects and localized areas of high stress concentration within the composite. These defects reduce the material's fracture toughness and overall flexibility. Additionally, the interfacial bonding between CNTs and PVA may not be strong enough to withstand large deformations. As a result, under tensile stress, CNTs may debond or slide within the matrix, leading to a reduction in elongation and yield strain.

When the CNT concentration is increased to 1%, there is a significant improvement in mechanical strength, including yield stress, yield load, and Young's modulus. However, this enhancement also leads to a further decrease in elongation at break and yield strain, indicating that the material becomes more brittle and less ductile, consistent with the findings reported in previous studies (Shah et al., 2024).

CONCLUSION

The use of PVA-CNT composite membranes has great potential in separation technology as it combines the advantages of polyvinyl alcohol (PVA) and carbon nanotubes (CNTs). PVA is known for its excellent hydrophilic properties, ease of membrane formation, and environmental friendliness; however, it has some drawbacks, such as high

swelling and low mechanical strength. The incorporation of CNTs into the PVA matrix can overcome these limitations, as CNTs act as structural reinforcements that provide higher mechanical strength, improve thermal stability, and enhance the bonding between polymer chains. Moreover, CNTs possess interconnected pore structures and high conductivity, which can improve separation performance and increase membrane selectivity by enabling more efficient separation of molecules based on their size and chemical properties. The presence of CNTs also helps reduce the swelling degree of PVA by reinforcing the polymer network, making it more stable when exposed to water or solutions. This results in a PVA-CNT membrane that is more resistant to deformation and degradation during the filtration process. Another significant advantage of PVA-CNT membranes is their superior fouling resistance compared to conventional polymer membranes. Fouling, which involves the accumulation of contaminants on the membrane surface or within its pores, is a common challenge in wastewater treatment, as it can reduce flux and shorten membrane lifespan. With the modified structure and surface properties provided by CNTs, these membranes can minimize clogging, thereby maintaining water flux for longer periods and simplifying the cleaning process. With this combination of benefits, PVA-CNT composite membranes have the potential to become an effective and sustainable green technology, particularly for oil-water separation, industrial wastewater treatment, and other filtration applications such as water purification and organic compound separation. This technology not only enhances separation efficiency but also supports environmental preservation efforts by providing an energy-efficient and eco-friendly solution.

ACKNOWLEDGEMENT

This research is one of the titles funded by the DRTPM Kemdikbudristek in 2025 on Penelitian Dosen Pemula (PDP) scheme. The authors would like to thank the DRTPM Kemdikbudristek for funding this research and Lembaga Penelitian dan Pengabdian kepada Masyarakat (LPPM) Universitas Duta Bangsa Surakarta for facilitating the implementation of the activity.

REFERENCES

- Basiuk, E. V., Anis, A., Bandyopadhyay, S., Alvarez-Zauco, E., Chan, S. L. I., & Basiuk, V. A. (2009). Poly(vinyl alcohol)/CNT composites: An effect of cross-linking with glutaraldehyde. *Superlattices and Microstructures*, 46(1–2), 379–383. <https://doi.org/10.1016/j.spmi.2008.10.007>
- Choo, K., Ching, Y. C., Chuah, C. H., Julai, S., & Liou, N. S. (2016). Preparation and characterization of polyvinyl alcohol-chitosan composite films reinforced with cellulose nanofiber. *Materials*, 9(8), 1–16. <https://doi.org/10.3390/ma9080644>
- Dudchenko, A. V., Rolf, J., Russell, K., Duan, W., & Jassby, D. (2014). Organic fouling inhibition on electrically conducting carbon nanotube-polyvinyl alcohol composite ultrafiltration membranes. *Journal of Membrane Science*, 468, 1–10. <https://doi.org/10.1016/j.memsci.2014.05.041>
- Huang, J., Ran, X., Sun, L., Bi, H., & Wu, X. (2024). Recent advances in membrane technologies applied in oil–water separation. *Discover Nano*, 19(1). <https://doi.org/10.1186/s11671-024-04012-w>
- Parangusan, H., Ponnamma, D., Hassan, M. K., Adham, S., & Al-Maadeed, M. A. A. (2019). Designing carbon nanotube-based oil absorbing membranes from gamma irradiated and electrospun polystyrene nanocomposites. *Materials*, 12(5). <https://doi.org/10.3390/ma12050709>

- Shah, S. A., Ali, H., Inayat, M. I., E. Mahmoud, E., AL Garalleh, H., & Ahmad, B. (2024). Effect of carbon nanotubes and zinc oxide on electrical and mechanical properties of polyvinyl alcohol matrix composite by electrospinning method. *Scientific Reports*, 14(1), 1–13. <https://doi.org/10.1038/s41598-024-79477-x>
- Siti Ajizah, F. (2023). Characteristics of PVA/GO Composite Membranes Prepared Using Solution Casting Technique for Reducing Methylene Blue Concentration. *Jurnal Ilmu Dan Inovasi Fisika*, 7(1), 20–29. <https://doi.org/10.24198/jiif.v7i1.40650>
- Thai, P. T. N., Pham, X. M., Nguyen, T. B., Le, T. M., Viet Tran, C. B., Phong, M. T., & Tran, L. H. (2021). Preparation and characterization of PVA thin-film composite membrane for pervaporation dehydration of ethanol solution. *IOP Conference Series: Earth and Environmental Science*, 947(1). <https://doi.org/10.1088/1755-1315/947/1/012010>
- Yang, Z., Zhang, M., Xiao, Y., Zhang, X., & Fan, M. (2021). Facile Fabrication of Poly(vinyl alcohol)/Polyquaternium-10 (PVA/PQ-10) Anion Exchange Membrane with Semi-Interpenetrating Network. *Macromolecular Materials and Engineering*, 306(1), 1–12. <https://doi.org/10.1002/mame.202000506>
- Yee, M. J., Mubarak, N. M., Khalid, M., Abdullah, E. C., & Jagadish, P. (2018). Synthesis of polyvinyl alcohol (PVA) infiltrated MWCNTs buckypaper for strain sensing application. *Scientific Reports*, 8(1), 1–16. <https://doi.org/10.1038/s41598-018-35638-3>
- Yi, G., Chen, S., Quan, X., Wei, G., Fan, X., & Yu, H. (2018). Enhanced separation performance of carbon nanotube–polyvinyl alcohol composite membranes for emulsified oily wastewater treatment under electrical assistance. *Separation and Purification Technology*, 197, 107–115. <https://doi.org/10.1016/j.seppur.2017.12.058>