



Article

The Influence of Activated Carbon Particle Size on the Properties and Performance of Polysulfone Composite Membrane for Protein Separation

Gunawan Setia Prihandana ^{1,*} , Aisyah Dewi Muthi'ah ¹, Tutik Sriani ² and Muslim Mahardika ³

¹ Department of Industrial Engineering, Faculty of Advanced Technology and Multidiscipline, Universitas Airlangga, Jl. Dr. Ir. H. Soekarno, Surabaya 60115, Indonesia; aisyahdm@ftmm.unair.ac.id

² Department of Research and Development, PT. Global Meditek Utama—IITOYA, Sardonoarjo, Ngaglik, Sleman, Yogyakarta 55581, Indonesia; tsriani@gmail.com

³ Department of Mechanical and Industrial Engineering, Faculty of Engineering, Universitas Gadjah Mada, Jalan Grafika No. 2, Yogyakarta 55281, Indonesia; muslim_mahardika@ugm.ac.id

* Correspondence: gunawan.prihandana@ftmm.unair.ac.id; Tel.: +62-881-0360-00830

Abstract: The superiorities provided by polymeric composite membranes in comparison to the original membrane have generated increased attention, particularly in the field of protein separation applications. This work involved the fabrication of polysulfone composite membranes using variable loadings of activated carbon particle sizes, namely, 37 μm , 74 μm , 149 μm , and 297 μm . The membranes were fabricated via the phase-inversion method, employing water as the coagulant. In this study, the impact of the AC powder particle sizes on membrane morphology, water contact angle, porosity, average pore size, molecular weight cutoff, pure water flux, and protein rejection was examined. Different membrane morphologies and properties were achieved by incorporating a variety of AC particle sizes. A porous membrane with the maximum pure water flux was generated by the loading of finer AC particles. Concurrently, protein rejection is increasing as a result of the use of AC particles as an infill in the composite membrane. In comparison to all fabricated membranes, the AC filler with a particle size of 149 μm exhibited the highest rejection of the lysozyme protein, reaching up to 73.9%, with a relatively high water permeability of 33 LMH/Bar. In conclusion, this investigation provides recommendations for the selection of AC particle sizes for protein separation in conjunction with PSF ultrafiltration membranes.

Keywords: polysulfone; ultrafiltration; membranes; activated carbon; good health; protein separation; clean water



Citation: Prihandana, G.S.; Muthi'ah, A.D.; Sriani, T.; Mahardika, M. The Influence of Activated Carbon Particle Size on the Properties and Performance of Polysulfone Composite Membrane for Protein Separation. *J. Compos. Sci.* **2024**, *8*, 483. <https://doi.org/10.3390/jcs8110483>

Academic Editors: Patricia Krawczak and Ludwig Cardon

Received: 13 September 2024

Revised: 11 November 2024

Accepted: 16 November 2024

Published: 19 November 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Ultrafiltration has emerged as a highly efficient technique in the field of separation processes. The cheap investment cost and energy needs, combined with the high separation efficiency, make it highly advantageous for a range of applications, such as food and pharmaceutical processing, as well as the chemical products industry. High-purity proteins have become indispensable raw materials in the food and medical treatment industries [1]. As a result, there has been a huge increase in the need for strong and effective techniques for separating and purifying proteins. Various methods, such as membrane technology, centrifugation, precipitation, sedimentation, electrophoresis, and chromatography, have been created to obtain elevated degrees of protein purification [2–5]. Membrane technology is considered an efficient method for protein treatment because of its high separation efficiency. It preserves the structure, integrity, and efficacy of proteins while minimizing the need for chemical usage [6]. The operational principle of separation membranes is impeding molecules that exceed the size of the membrane pore. As a result, membranes of varying pore sizes can separate molecules according to their molecular weight. Membrane filtration

can be used in a wide range of separation situations by choosing membranes with suitable pore diameters [7]. Ultrafiltration membranes are often effective for concentrating and separating proteins because their pore sizes are large enough to prevent protein molecules from passing through the membrane pores [8]. Membrane fouling, which involves the adsorption of solute on the membrane surface and the blocking of pores, greatly decreases the effectiveness of membrane separation [9]. Membranes are commonly produced using polymers such as polysulfone, polyethersulfone, polyamide, polyethylene, polypropylene, and polyvinyl fluoride. These polymers are chosen for their excellent mechanical, thermal, and chemical resistance properties [10].

Polysulfone (PSF) possesses exceptional thermal stability, elevated mechanical strength, and chemical resistance, rendering it an ideal substance for the production of filtration membranes [11,12]. Consequently, research endeavors on PSF membranes have experienced a substantial surge in the past decade. PSF membranes can be modified using techniques including physical mixing, chemical grafting, and coating. These processes are used to improve the performance of the membranes [13]. Although surface coating and chemical grafting onto premade membranes are convenient and flexible techniques, they encounter notable difficulties, such as pore structural deformation. This deformation results in a decrease in permeate flux, which ultimately restricts membrane performance [14]. In order to tackle these difficulties, the inclusion of inorganic elements in the solution is of great importance as it can improve the hydrophilicity, permeation, and antifouling properties [15,16]. The use of carbon-based materials in polymers to create ultrafiltration membranes has gained significant attention due to its potential to regulate pore shape, boost hydrophilicity, improve antifouling characteristics, and increase mechanical strength [17]. Nevertheless, the findings suggest that an excessive inclusion of inorganic particles has a negative impact on the selectivity, permeability, and shape of the membranes. The main goal is to employ appropriate inorganic materials with a comparatively lower proportion of inorganic fillers, improving the hydrophilicity of the membrane without compromising its permeability and selectivity [18].

Composite membranes composed of PSF and carbon nanotubes have been reported to demonstrate enhanced hydrophilicity, protein rejection, and water permeate flux [19]. Carbon-based nanoparticles are effective modifiers due to their hydrophilic surface functional groups, chemical stability, lack of toxicity, and excellent surface area [20]. Hosseini et al. [21] developed mixed matrix nanofiltration membranes based on polyethersulfone embedded with activated carbon nanoparticles (ACNPs) to remove sulfate and copper from water. The results indicate that the addition of super activated carbon nanoparticles to the casting solution significantly enhanced the rejection of sulfate and copper ions. Sharma et al. [22] fabricated polyethersulfone nanofiltration membranes by incorporating activated carbon nanoparticles at concentrations ranging from 0 to 1 wt.% to separate lignin from black liquor. The study's findings showed that AC-based membranes with a concentration of 0.1% had improved efficiency in separating lignin from black liquor, while also reducing membrane fouling. Nevertheless, while choosing additive materials, it is crucial to take into account not only their performance but also factors such as their local availability and cost-effectiveness [23]. Consequently, in light of the exorbitant expense associated with synthesizing nanoparticles, scientific investigation has prioritized the search for more economical alternatives in terms of powder size. Remy et al. [24] found that the addition of powdered activated carbon raised the critical flow by 10% and considerably prolonged the filtration time of the membrane.

Liu et al. [25] showed that the inclusion of powdered activated carbon resulted in longer membrane filtering time cycles, suggesting a decreased likelihood of membrane fouling. Torretta et al. [26] discovered that using a certain concentration of powdered activated carbon was the most efficient method for improving membrane flow, resulting in a notable 26% increase. To summarize, the use of powdered activated carbon as a filler has been found to improve the performance of composite membranes through multiple functions and mechanisms. Nevertheless, there is a dearth of research investigating the

selectivity and permeability of composite membranes with different diameters of powdered activated carbon. The aim of this work is to manufacture a PSF composite membrane with varying loadings of AC particle sizes and investigate how these parameters impact the membrane characteristics and performance in protein separation.

The work involved incorporating activated carbon particles of different sizes (37 μm , 74 μm , 149 μm , and 297 μm) into a solution of PSF membrane. This was carried out to create composite ultrafiltration membranes using the wet phase inversion procedure. An assessment was conducted to determine the influence of activated carbon particle size on hydrophilicity, porosity, average pore size, pore structure, and molecular weight cutoff. The surface morphology of the membranes was examined using scanning electron microscopy (SEM). The evaluation of water permeability and protein separation efficacy of the membranes was performed using standardized experimental methodologies.

2. Materials and Methods

2.1. Materials

Polysulfone pellets (PSF, 80,000 g/mol, P.T. Solvay Chemicals Indonesia, Jakarta, Indonesia), N-methyl-2-pyrrolidone (NMP; PT. Samiraschem Indonesia, Jakarta, Indonesia), Activated Carbon (AC) powder, Filtrasorb 100 (Calgon Carbon Corporation, Pittsburgh, PA, USA), and an adjustable film applicator (Elcometer 3540/4 Four-Sided Film Applicators, Elcometer Limited, Manchester, UK) were purchased to prepare the PSF-AC membranes. Lysozyme (12 kDa), pepsin (35 kDa), and bovine serum al-bumin (BSA) (69 kDa) proteins from HiMedia Ltd. in Mumbai, India were purchased to prepare protein solutions. Pure water was used as a gelatinization medium for the membrane fabrication, and droplet for the water contact angle test and membrane permeability test. The Scanning Electron Microscopy (SEM) instrument, Phenom ProX (Thermo Fisher Scientific, Waltham, MA, USA) was employed to analyze the cross section and morphological properties of the membranes. AC powder was utilized as received, and its specifications are detailed in Table 1.

Table 1. Specification of AC powder.

Iodine Number, mg/g	850
Moisture by Weight	2%
Effective Size	0.8–1.0
Uniformity Coefficient	2.1
Abrasion Number	75
Screen Size by Weight, US Sieve Series	
On 8 mesh	15%
Through 30 mesh	4%
Apparent Density (tamped)	0.58 g/cc
Water Extractables	<1%
Non-Wettable	<1%

2.2. Membrane Fabrication

Activated carbon-polysulfone (AC-PSF) membranes for protein separation were prepared by dissolving PSF in NMP, incorporating AC carbon as an additive, casting a thin film from the resulting solution (containing PSF, NMP, and AC) at varying concentrations (as indicated in Table 2) onto a non-woven support, and immersing the non-woven substrate in a pure water bath using the wet phase inversion method. The casting solutions consisted of PSF with specific particle size distributions: pristine membrane, 297 μm , 149 μm , 74 μm , and 34 μm , all at a concentration of 1 wt.% as membrane filler.

Table 2. Membrane prepared in this study.

No.	Membrane Code	AC Powder Size (μm)
1	PSF-CA0	-
2	PSF-CA1	297
3	PSF-CA2	149
4	PSF-CA3	74
5	PSF-CA4	37

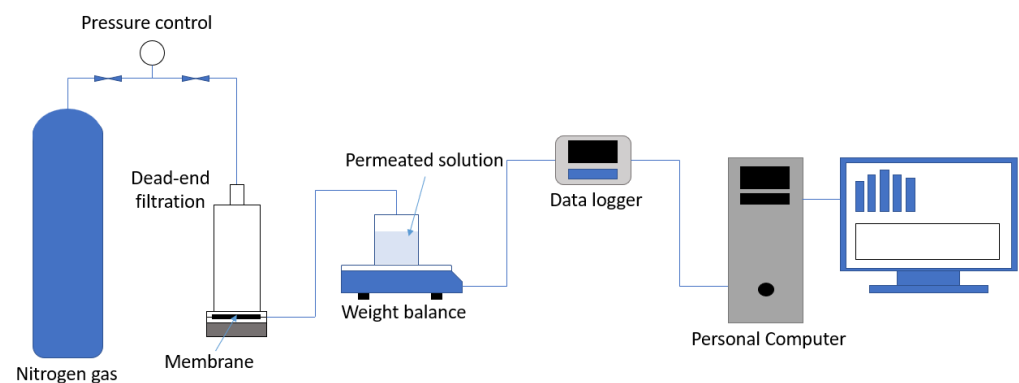
2.3. Water Contact Angle Test

A digital microscope (Dinolite Edge 3.0 AM73915MZTL, AnMo Electronics, New Taipei City, Taiwan) was employed to capture images of a 0.5 μL pure water droplet on the dried surface. Additionally, CAD software was utilized to measure the contact angle of the membranes. The contact angles for each membrane were measured three times and then averaged to ensure accuracy.

2.4. Ultrafiltration Experiments

2.4.1. Water Flux Test

The water flux of all membranes, including both pristine and PSF-AC membranes, was assessed using a dead-end cell setup, as depicted in Figure 1. This setup incorporates valves and pressure gauges to regulate the pressure within the feed chamber. Initially, the membrane sample was positioned at the bottom of the dead-end cell, with an active surface area of 14.6 cm^2 . Above the tested membrane, there was a feed chamber with a capacity of 300 mL. A pressure regulator was employed to control the applied pressure from the nitrogen cylinder to the feed chamber. A working pressure of 2 bar was applied to the fabricated membranes. The permeate weight was automatically recorded by a computer every 2 min over a duration of 30 min in this filtration system. Subsequently, at least three replicate samples were assessed for each membrane, and the average value, along with its standard deviation, was reported.

**Figure 1.** Experiment setup of the water flux test.

We employed the following equations [27] to calculate both the volumetric flux (J_v) and the permeability:

$$\text{Flux}(J_v) = \frac{Q}{A \times \Delta t} \quad (1)$$

$$\text{Permeability}(L_p) = \frac{J_v}{\Delta P} \quad (2)$$

In the context of membrane analysis, Q represents the quantity of permeate water (measured in liters) during the sampling time, Δt denotes the duration of the sampling period (in hours), A signifies the membrane area (measured in square meters), and ΔP represents the pressure difference (in bars).

2.4.2. Protein Separation

A solution containing 0.1 wt.% of BSA, pepsin, and lysozyme was carefully produced in a phosphate-buffered solution with a pH of 7.2. The ensuing protein separation experiment was carried out using a dead-end cell filtration test, with a fixed pressure of 2 bar. In order to measure the concentration of the protein solution that passed through, we used an UV–visible spectrophotometer set to a wavelength of 280 nm. The solute rejection (SR) was determined through meticulous analysis.

$$\%SR = \left[1 - \frac{C_p}{C_f} \right] \times 100 \quad (3)$$

In the context of protein concentration analysis, C_p represents the concentration of the permeated solution, while C_f denotes the concentration of the feed solution.

2.5. Porosity

The study focused on analyzing the membrane's porosity in order to assess the impact of including AC powder on the size of the membrane pores. This was carried out by the gravimetric method, using the following equation [28]:

$$\varepsilon(\%) = \frac{W_w - W_d}{\rho_{H_2O} \times A \times L} \times 100 \quad (4)$$

where W_w is the weight of the wet membrane, W_d is weight of the dry membrane, the effective area of the membrane is denoted by A , the thickness of the membrane is denoted by L , and the density of water (0.998 g/cm^3) is denoted by ρ_{H_2O} .

2.6. Average Pore Size

The average pore size of the surface membranes was estimated using the ultrafiltration experimental findings. In order to determine the average pore size of the membranes, we employed the molecular weight of the solute, which demonstrated a solute rejection (SR) greater than 80%, by utilizing the following equation:

$$\bar{R} = 100 \left(\frac{\alpha}{\%SR} \right) \quad (5)$$

In this context, $\bar{R}$ indicates the mean pore size (radius), while α refers to the solute radius, which is calculated by the Stoke radius computed from the solute's molecular weight using Sarbolouki's approach [29].

2.7. Molecular Weight Cutoff (MWCO)

The molecular weight cutoff (MWCO) has a direct correlation with the pore size of the membrane. The objective of evaluating the MWCO of a membrane is to determine the minimum size of an unreactive solute that exhibits protein rejection within the range of 80% to 100% in an ultrafiltration experiment [30]. The BSA, pepsin, and lysozyme were chosen as the proteins to be evaluated in order to assess any decrease in pore size that may occur as a result of introducing activated carbon powder.

3. Results

3.1. Contact Angle Analysis

In order to confirm the hydrophobic nature of the membranes, we conducted additional measurements of the water contact angle for various composite membranes using pure water. A membrane surface that is more hydrophilic is indicated by a lower water contact angle value.

Figure 2 displays the static water contact angles of several PSF-CA composite membranes. The pristine PSF membrane (PSF-AC0) exhibits an initial contact angle of around

67.6° due to its intrinsic hydrophilicity. The introduction of a 1% AC particle at any range of powder size resulted in a lower contact angle of 59.5°, 59°, 58.2°, and 57.5° for PSF-AC1, PSF-AC2, PSF-AC3, and PSF-AC4, respectively, in comparison to pristine PSF membranes (67.6°). This indicates that the contact angle was further reduced by further reducing the size of AC particles. This decline indicates a significant rise in the hydrophilicity of the membranes.

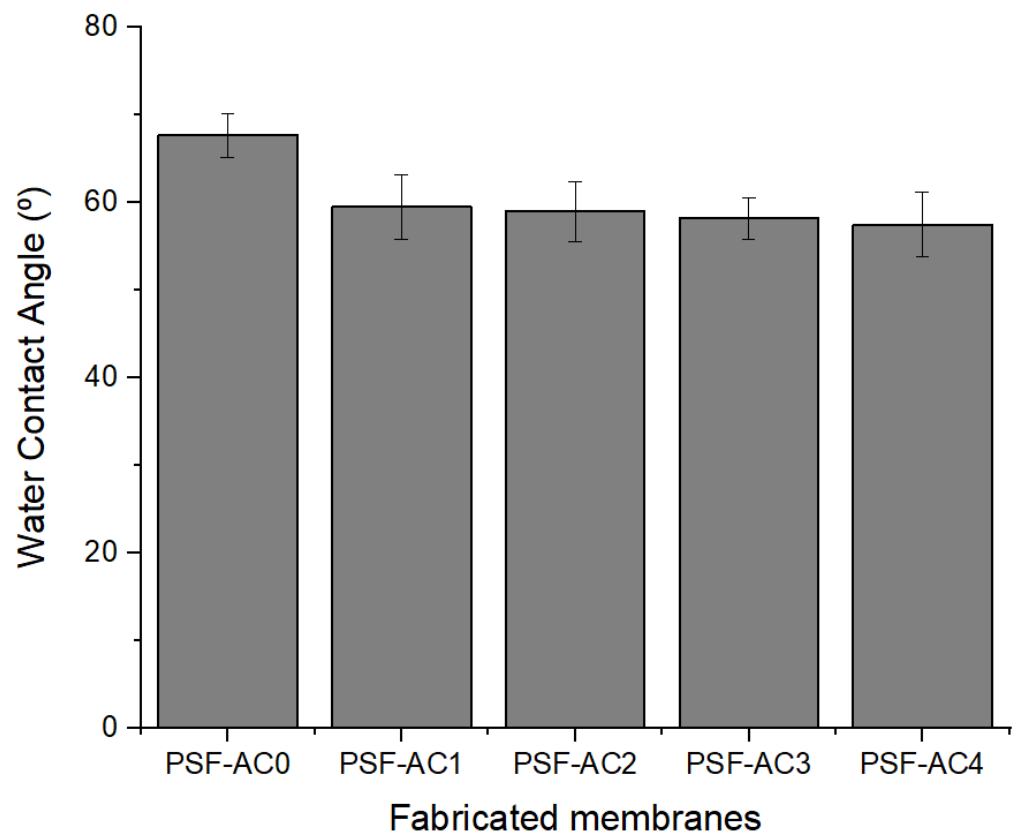


Figure 2. Water contact angle of the fabricated membranes.

The physical characteristics of powder, such as the size of particles, can affect how well it can be wetted. Powders with a greater particle size have superior wetting characteristics due to the enhanced ability of water to permeate the bigger interstitial gaps between the particles [31]. However, in this scenario, the interstitial gaps between larger particles are the surface of the membrane, which possesses a higher water contact angle. Consequently, this circumstance results in the PSF-AC membrane with larger particle sizes exhibiting more hydrophobicity.

3.2. Membrane Permeability Test

Figure 3 illustrates the water permeability of membranes with varying AC particle sizes incorporated. Upon comparing the water permeability of the pristine PSF membrane (14.75 LMH/Bar) with the fluxes of the composite membranes, it was observed that the fluxes increased as the AC particle size decreased, at the same AC concentration of 1 wt.%. The permeate flux with an activated carbon particle size of 297 microns is 32.5 LMH/bar, reaching a maximum value of 42.5 LMH/bar with activated carbon particles of 37 microns. According to Figure 2, it is evident that the improved water flux is a result of the heightened hydrophilicity, since the PSF-AC4 membranes exhibit the highest level of hydrophilicity. As stated in the contact angle analysis section, when the particle size is small, it aids in achieving a uniform distribution on the surface of the membrane, hence

facilitating water to permeate through its porous multichannel structure, resulting in a higher water permeability.

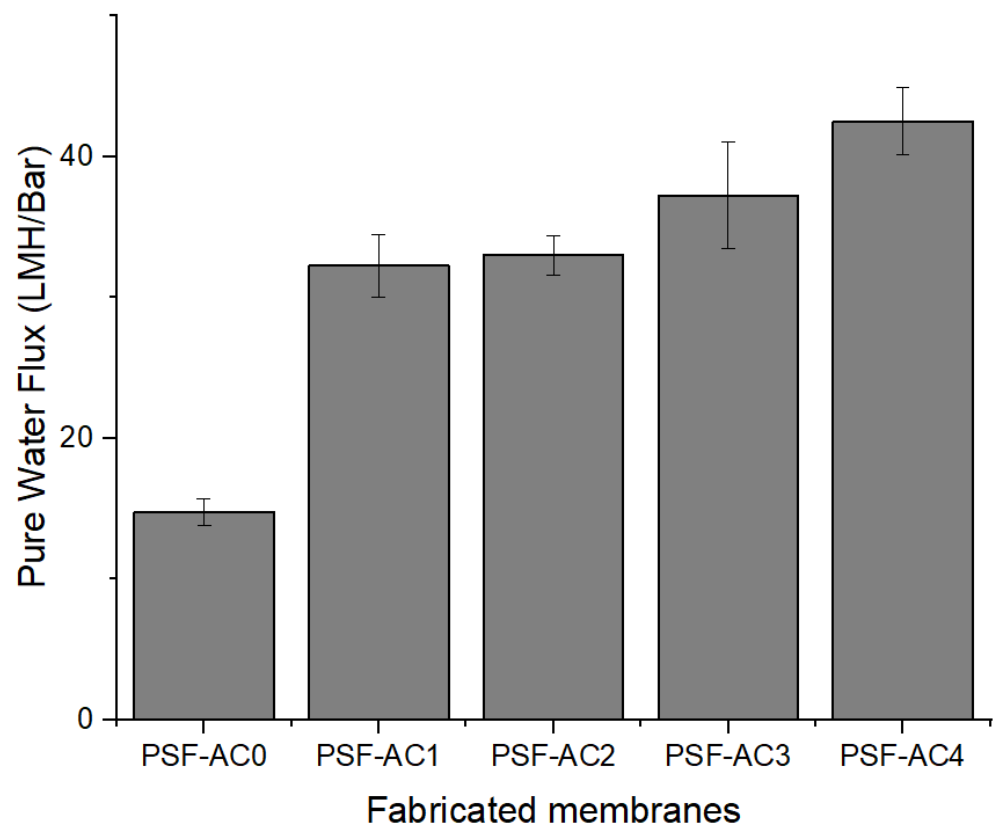


Figure 3. Water flux of the fabricated membranes.

Moreover, the addition of larger particle sizes increases the likelihood of aggregation, which, in turn, widens the space between particles and exposes the hydrophobic surface of the membrane. This leads to a decrease in effective porosity by blocking channels that allow vapor to pass through [32]. As a result, the water transfer facilitated by the porous multichannel structure of AC is compromised.

3.3. Effect of AC on Protein Rejection

The rejection and permeate capabilities of membranes depend on their morphological structure in conjunction with the parameters of the feed solution. In this study, three distinct proteins of varying sizes were employed as representative solutions for protein separation experiments. The solution containing BSA, pepsin, and lysozyme dissolved in water is used as a feed solution to study how the membranes reject these substances. Figure 4 displays the percentage of membrane protein rejection results, which were determined using Equation (3). The figure illustrates that the manufactured membranes exhibit a BSA rejection rate of 90%. All membranes have a pore size that is smaller than the molecular weight of BSA. The separation test for the membranes was extended using pepsin and lysozyme as solutes due to their smaller molecular sizes compared to BSA. Figure 4 demonstrates that the composite membrane exhibits a somewhat higher rejection of pepsin in comparison to the pristine membrane. A substantial disparity in protein rejection was seen when employing a lysozyme solution, with composite membranes exhibiting notable rejection of lysozyme. As shown in Figure 4, the PSF-AC2 membrane has the highest lysozyme rejection rate (73.9%), whereas the PSF-AC0 membrane has the lowest (49.3%). This can be elucidated by considering the size of the pores in the membrane. The membrane PSF-AC2 has the smallest pore size compared to all other produced membranes, resulting in the maximum

rejection of lysozyme. On the other hand, the membrane PSF-AC0 has the biggest pore size and exhibits the lowest rejection of lysozyme. Hence, the membrane demonstrated superior size-exclusion separation properties, allowing it to reject protein more effectively than other membranes that were fabricated [33].

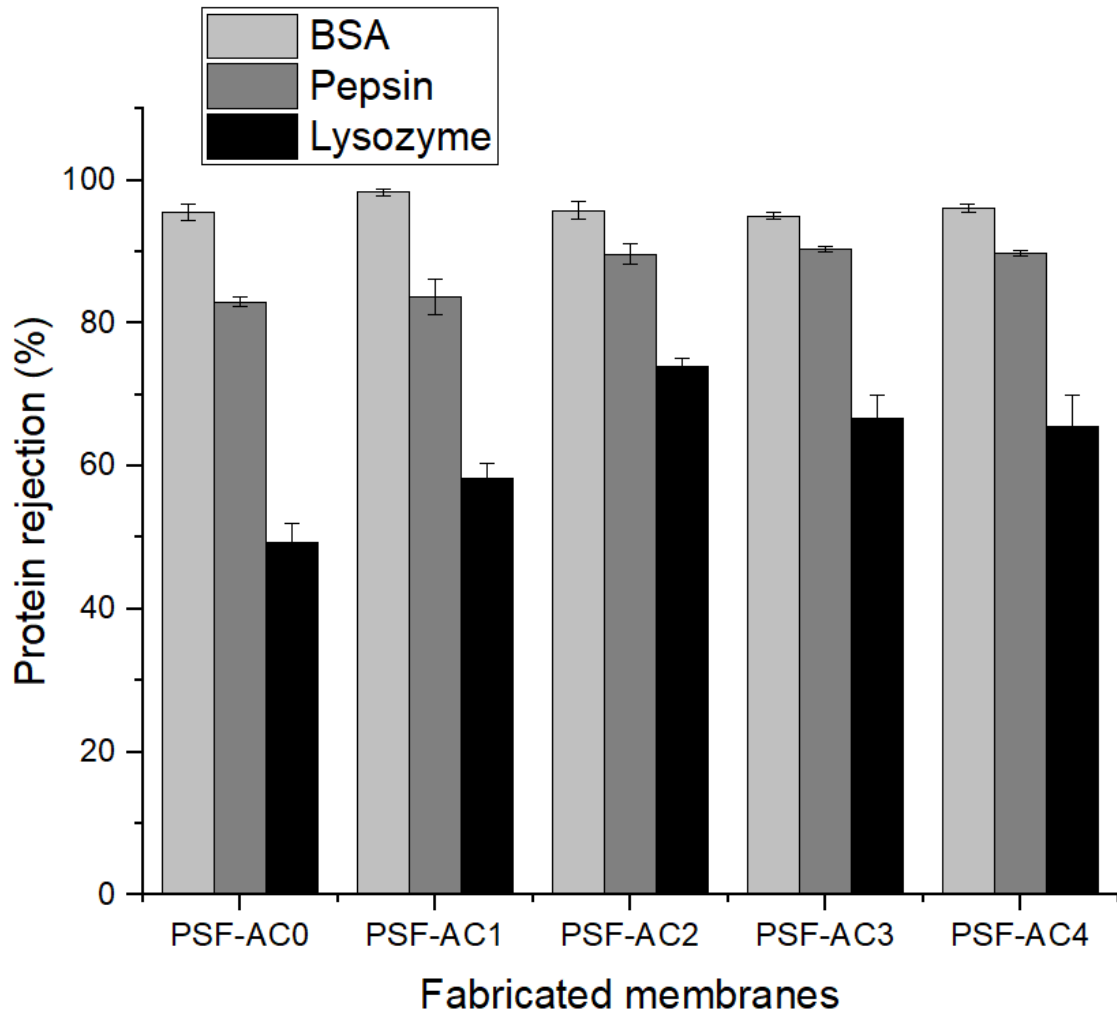


Figure 4. Protein rejection of the fabricated membranes.

3.4. Effect of AC on Membranes Porosity

An analysis of membrane porosity was used to evaluate the effect of nAC on the modified polymer membrane. The results of the apparent porosity of membranes that were incorporated with AC as an additive are presented in Table 3. The particulate size of the membranes was varied, with pristine membrane (PSF-AC0), 297 μm (PSF-AC1), 149 μm (PSF-AC2), 74 μm (PSF-AC3), and 37 μm (PSF-AC4). The pristine membrane had the lowest reported porosity values (63.41%) when AC was not present. The manufactured membrane reached its maximum porosity of 66.92% when the smallest AC powder of 37 μm was loaded. This phenomenon arises due to the fact that, when the concentration of AC is held constant at 1 wt.%, the decrease in particle size causes a larger quantity of particles, that, in turn, leads to a membrane with higher porosity and, as a result, an increased water flux [34].

Table 3. Porosity, pore radius, and MWCO of the fabricated membranes.

Membrane Code	Porosity (%)	Pore Radius, $R(A^\circ)$	MWCO (kDa)
PSF-AC0	63.41	30.12	35
PSF-AC1	63.46	29.88	35
PSF-AC2	66.02	27.88	35
PSF-AC3	66.61	27.66	35
PSF-AC4	66.92	27.83	35

3.5. Measurement of Average Pore Size

The average pore size of the fabricated membranes was calculated using Equation (5) and is depicted in Table 3. The results suggest that the introduction of activated carbon particles into the membrane matrix resulted in a decrease in the size of the pores in composite membranes. Specifically, a smaller average pore radius was observed as the activated carbon powder size decreased from 297 μm to 37 μm in the case of composite membranes.

The pristine membrane, PSF-AC0, had a pore size of 30.12 A° , whereas the composite membranes had pore sizes ranging from 27.83 A° to 29.88 A° . Nevertheless, the membrane made with the smallest AC powder particles (37 μm) had a larger pore size (27.83 A°) compared to PSF-AC3, 74 μm (27.66 A°). The membranes exhibited the following order of pore size: PSF-AC0 (pristine membrane) > PSF-AC1 (297 μm) > PSF-AC2 (149 μm) > PSF-AC4 (74 μm) > PSF-AC3 (37 μm). The smaller powder will yield a reduced pore size since it tends to spread uniformly across the membrane structure, resulting in smaller pores. The introduction of the 37 μm AC particle led to a little increase in pore size, possibly attributable to the non-uniformity of the powder size, as the powder was separated using a sieve shaker, resulting in smaller particles under 37 μm remaining during the separation process.

3.6. Molecular Weight Cutoff Measurement (MWCO)

Protein solutions with molecular weights of 14 kDa for lysozyme, 35 kDa for pepsin, and 69 kDa for BSA were employed to ascertain the molecular weight cutoff measurement for both pristine and PSF-AC composite membranes, with varying powder sizes of AC. The result of MWCO is presented in Table 3. As shown in Table 3, the molecular weight threshold of all membranes was initially 35 kDa, which is higher than the MWCO of lysozyme (14 kDa). Nonetheless, whereas all membranes exhibit an identical molecular weight cutoff (MWCO) of 35 kDa, there are substantial variations in the rejection % of lysozyme across each membrane. For instance, as illustrated in Figure 4, AC with a powder size of 149 μm exhibits a gap of 73.9% relative to the lysozyme molecular weight cutoff standard of 80% [31], indicating that, despite the membranes possessing identical MWCO levels, they demonstrate varying percentages of solute rejection.

3.7. Membrane Morphology

In this study, we provide the surface morphology SEM images of the PSF pristine membranes and the membranes filled with AC powder as presented in Figure 5. Undoubtedly, the impact of AC powder on the surface morphology of the PSF membranes is substantial. Initially, the PSF composite membranes have a uniformly dispersed surface with an increased number of pores. This phenomenon can be elucidated by the capacity of the fine particles to infiltrate and establish a more compact structure with reduced pore size [35].

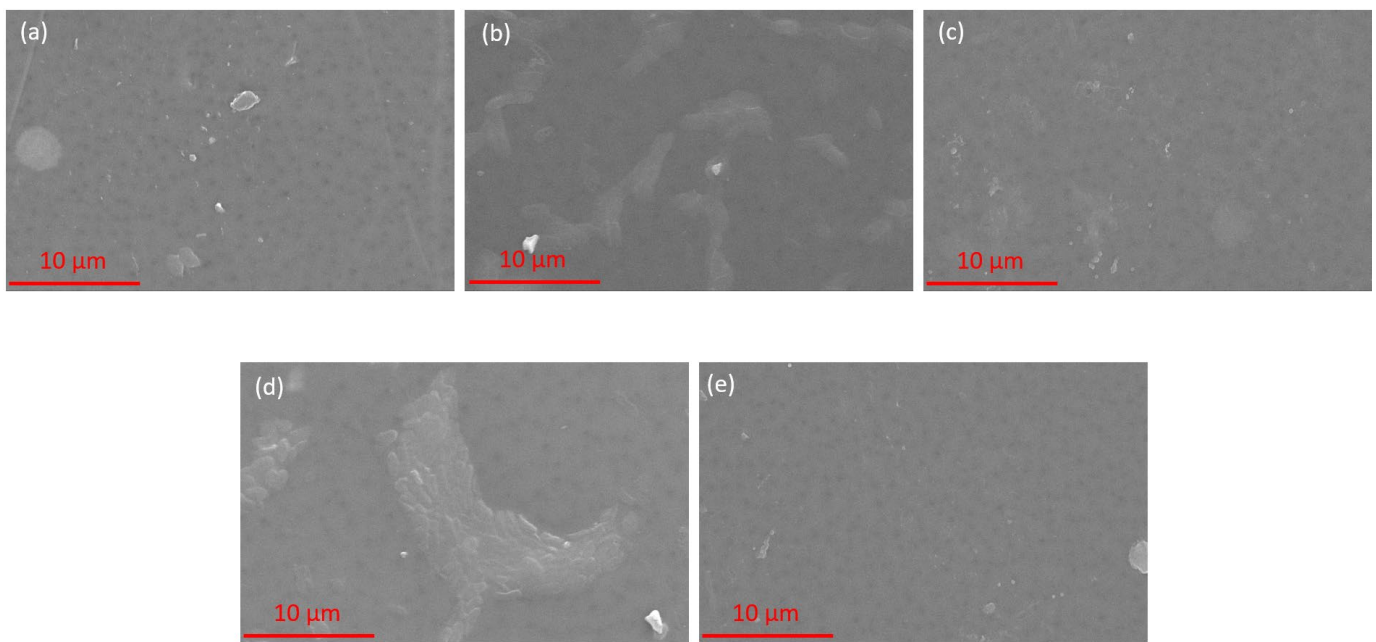


Figure 5. Surface morphology SEM images of the fabricated membranes: (a) PSF-AC0; (b) PSF-AC1; (c) PSF-AC2; (d) PSF-AC3; and (e) PSF-AC4.

The fabricated membranes are illustrated in cross-sectional SEM images in Figure 6. The PSF membranes are composed of a sponge-like layer and a finger-like layer. The selective layer is situated on the side of the membranes that contains the finger-like layer. The cross-sectional morphology of the modified membranes, which contain AC powder, is comparable to that of PSF pristine membranes. When coarse powder was utilized, the SEM images reveal that the membrane exhibited a greater tendency to form macro void structures rather than sponge-like structures [36]. The reduction of finger-like macro gaps by fine particles can be attributed to their capacity to efficiently fill the interstices, therefore enhancing the density and reducing the porosity of the structure [35].

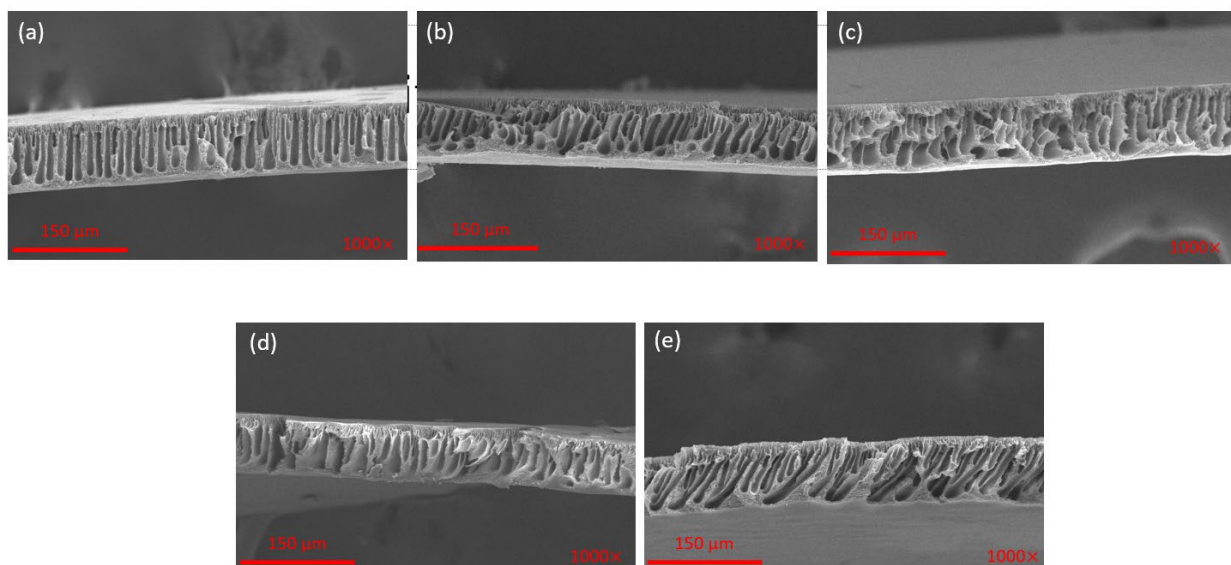


Figure 6. Cross section SEM images of the fabricated membranes: (a) PSF-AC0; (b) PSF-AC1; (c) PSF-AC2; (d) PSF-AC3; and (e) PSF-AC4.

Table 4 compares previous studies on PSF membrane performance that included AC and other additives as the principal modulator. In comparison to its modified counterpart, each column displays the value of the PSF membrane. AC, silver-nanoparticles, polyethylene glycol (PEG), polyvinylpyrrolidone (PVP), carbon nanotubes (CNT), CuCl₂, FeCl₃, and APS were among the additives that were incorporated. The majority of membrane modification investigations utilized PEG and PVP as pore formers. Additional additives at varying sizes and concentrations were employed to change the membrane. Each method improves the membranes' hydrophilicity, flux, porosity, average pore size, rejection capabilities, and morphology. Although the AC particles remained unmodified before application, the optimal membrane in this study exhibits a performance akin to comparable membranes from previous research regarding water permeability, protein rejection, and water contact angle. The membrane modification technique outlined in this work may serve as a pivotal factor for the expansion of membrane production in the protein separation sector.

Table 4. Overview of research on HA blending in polymeric membranes comparing the native membrane and modified membranes.

Polymer	Modifier	Permeability Flux, LMH/Bar	Pore Size, nm	Rejection (%)	ε (%)	WCA (°)	Ref.
PSF	AC	53.3/116.3	34.1/43.2	~93.2 (BSA)	-	-	[37]
PSF/PVP	Silver-nanoparticle	12/55	-	~48 (BSA)	-	81.2/60.9	[38]
PSF	PANI-CuCl ₂ , PANI-FeCl ₃ and PSF/PANI/APS	30/450	92.12/122.94	96 (BSA)	-	41.8-80	[39]
PSF/PEG	AC	5/9.8	6.4/7.2	81–93 (total phenolic compounds)	83–86	48.2–66.7	[40]
PSF	CNT	1/8	2.82/10.18	98 (lignin) 98 (BSA)	74.4/75.5	57.6/74.4	[28]
PSF	AC	15/42.5	27.66/30.12	90 (Pepsin) 73.9 (Lysozyme)	63.41/66.92	57.5/67.6	This work

4. Conclusions

A series of polysulfone ultrafiltration composite membranes were fabricated by altering the size of AC particles via phase inversion processing. The membranes exhibited a unique shape and performance dependent on the various sizes of AC particles used as a filler. The inclusion of particles of varying sizes has an impact on the morphology of the membrane, namely, in relation to the finger-like and sponge structure of the membrane. All the composite membranes demonstrated a superior water flux and rejection of the lysozyme in comparison to the pristine membrane. The findings derived from this investigation are indicative of the significance of the AC particle size in influencing the structure and functionality of the composite PSF membrane. Hence, the appropriate choice of particle size is crucial in assessing the effectiveness of membrane performance for different applications, especially in the context of protein separation and purification.

Author Contributions: Conceptualization, G.S.P., M.M. and T.S.; methodology, G.S.P.; software, A.D.M. and T.S.; validation, G.S.P., M.M. and T.S.; formal analysis, T.S. and G.S.P.; investigation, G.S.P. and T.S.; resources, G.S.P., A.D.M. and M.M.; data curation, T.S.; writing—original draft preparation, G.S.P.; writing—review and editing, T.S. and M.M.; visualization, T.S.; supervision, M.M.; project administration, A.D.M.; funding acquisition, G.S.P., A.D.M. and M.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Hibah Direktorat Riset, Teknologi dan Pengabdian kepada Masyarakat 2023, Scheme: Penelitian Fundamental Reguler (Grant No. 0536/E5/PG.02.00/2023; 114/E5/PG.02.00.PL/2023; 1338/UN3.LPPM/PT.01.03/2023).

Data Availability Statement: The data presented in this study are available upon request from the corresponding author.

Acknowledgments: The authors would like to thank the staff of Lembaga Penelitian Dan Pengabdian Masyarakat, Universitas Airlangga, Indonesia, for their administrative support.

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. Qiang, T.; Song, Y.; Zhu, R.; Yuan, W. Biomass material derived hierarchical porous TiO₂: Adjustable pore size for protein adsorption. *J. Alloys Compd.* **2020**, *829*, 154512. [\[CrossRef\]](#)
2. Kluszczynska, K.; Peczek, L.; Rozanski, A.; Czernek, L.; Duchler, M. U6/miR-211 expression ratio as a purity parameter for HEK293 cell-derived exosomes. *Acta Biochim. Pol.* **2022**, *69*, 409–415. [\[CrossRef\]](#) [\[PubMed\]](#)
3. Arbita, A.A.; Paul, N.A.; Cox, J.; Zhao, J. Amino acid sequence of two new milk-clotting proteases from the macroalga *Gracilaria edulis*. *Int. J. Biol. Macromol.* **2022**, *211*, 499–505. [\[CrossRef\]](#) [\[PubMed\]](#)
4. Ogata, M.; Sakamoto, M.; Yamauchi, N.; Nakazawa, M.; Koizumi, A.; Anazawa, R.; Kurumada, K.; Hidari, K.I.P.J.; Kono, H. Optimization of the conditions for the immobilization of glycopolypeptides on hydrophobic silica particulates and simple purification of lectin using glycopolypeptide-immobilized particulates. *Carbohydr. Res.* **2022**, *519*, 108624. [\[CrossRef\]](#)
5. Chai, M.; Ye, Y.; Chen, V. Separation and concentration of milk proteins with a submerged membrane vibrational system. *J. Membr. Sci.* **2017**, *524*, 305–314. [\[CrossRef\]](#)
6. Ding, L.; Zhang, W.; Ould-Driss, A.; Jaffrin, M.Y.; Tang, B. Concentration of milk proteins for producing cheese using a shear-enhanced ultrafiltration technique. *Ind. Eng. Chem. Res.* **2016**, *55*, 11130–11138. [\[CrossRef\]](#)
7. Xu, Z.; Ma, G.; Rana, D.; Matsuura, T.; Lan, C.Q. Engineering polyvinylidene fluoride membranes using charge tunable dendrimer polyamidoamine to enable microporous membranes for protein separation. *Sep. Purif. Technol.* **2025**, *354 Pt 4*, 128887. [\[CrossRef\]](#)
8. Kassa, S.T.; Hu, C.C.; Keshebo, D.L.; Ang, M.B.M.; Lai, J.Y.; Chu, J.P. Surface modification of high-rejection ultrafiltration membrane with antifouling capability using activated oxygen treatment and metallic glass deposition. *Appl. Surf. Sci.* **2020**, *529*, 147131. [\[CrossRef\]](#)
9. Jin, Z.; Shen, Y.; Chen, X.; Qiu, M.; Fan, Y. Construction of high-performance Ce-doped TiO₂ tight UF membranes for protein separation. *Appl. Surf. Sci.* **2023**, *610*, 155468. [\[CrossRef\]](#)
10. Simari, C.; Capri, A.; Rehman, M.H.U.; Enotiadis, A.; Gatto, I.; Baglio, V.; Nicotera, I. Composite anion exchange membranes based on polysulfone and silica nanoscale ionic materials for water electrolyzers. *Electrochim. Acta* **2023**, *462*, 142788. [\[CrossRef\]](#)
11. Chen, S.; Li, L.; Feng, M.; Huang, T.; Zhang, N.; Wang, Y. UV-activated superwetting ability of electrospun polysulfone/titanium dioxide membranes toward highly efficient methylene blue removal and oil/water separation. *J. Membr. Sci.* **2024**, *695*, 122450. [\[CrossRef\]](#)
12. Mamah, S.C.; Goh, P.S.; Ismail, A.F.; Suzaimi, N.D.; Yogarathinam, L.T.; Raji, Y.O.; El-badawy, T.H. Recent development in modification of polysulfone membrane for water treatment application. *J. Water Process. Eng.* **2021**, *40*, 101835. [\[CrossRef\]](#)
13. Pagidi, A.; Saranya, R.; Arthanareeswaran, G.; Ismail, A.F.; Matsuura, T. Enhanced oil–water separation using polysulfone membranes modified with polymeric additives. *Desalination* **2014**, *344*, 280–288. [\[CrossRef\]](#)
14. Khan, A.; Sherazi, T.A.; Khan, Y.; Li, S.; Naqvi, S.A.R.; Cui, Z. Fabrication and characterization of polysulfone/modified nanocarbon black composite antifouling ultrafiltration membranes. *J. Membr. Sci.* **2018**, *554*, 71–82. [\[CrossRef\]](#)
15. Zhang, Y.; Cui, P.; Du, T.; Shan, L.; Wang, Y. Development of a sulfated Y-doped nonstoichiometric zirconia/polysulfone composite membrane for treatment of wastewater containing oil. *Sep. Purif. Technol.* **2009**, *70*, 153–159. [\[CrossRef\]](#)
16. Arthanareeswaran, G.; Thakur, R.S. Effect of inorganic particle on the performance of polyethersulfone-cellulose acetate ultrafiltration membranes. In *Sustainable Membrane Technology for Energy, Water, and Environment*; Ismail, A.F., Matsuura, T., Eds.; John Wiley & Sons: Hoboken, NJ, USA, 2012; p. 11.
17. Abid, Z.; Abbas, A.; Mahmood, A.; Rana, N.F.; Khan, S.J.; Duclaux, L.; Deen, K.M.; Ahmad, N.M. Water Treatment Using High Performance Antifouling Ultrafiltration Polyether Sulfone Membranes Incorporated with Activated Carbon. *Polymers* **2022**, *14*, 2264. [\[CrossRef\]](#)
18. Manawi, Y.; Kochkodan, V.; Hussein, M.A.; Khaleel, M.A.; Khraisheh, M.; Hilal, N. Can carbon-based nanomaterials revolutionize membrane fabrication for water treatment and desalination. *Desalination* **2016**, *391*, 69–88. [\[CrossRef\]](#)
19. Khalid, A.; Al-Juhani, A.A.; Al-Hamouz, O.C.; Laoui, T.; Khan, Z.; Atieh, M.A. Preparation and properties of nanocomposite polysulfone/multi-walled carbon nanotubes membranes for desalination. *Desalination* **2015**, *367*, 134–144. [\[CrossRef\]](#)
20. Bagheripour, E.; Moghadassi, A.R.; Hosseini, S.M.; Ray, M.B.; Parvizian, F.; Van der Bruggen, B. Highly hydrophilic and antifouling nanofiltration membrane incorporated with water-dispersible composite activated carbon/chitosan nanoparticles. *Chem. Eng. Res. Des.* **2018**, *132*, 812–821. [\[CrossRef\]](#)
21. Hosseini, S.M.; Amiri, S.H.; Khodabakhshi, A.R.; Bagheripour, E.; Van der Bruggen, B. Activated carbon nanoparticles entrapped mixed matrix polyethersulfone based nanofiltration membrane for sulfate and copper removal from water. *J. Taiwan Inst. Chem. Eng.* **2018**, *82*, 169–178. [\[CrossRef\]](#)
22. Sharma, M.; Alves, P.; Gando-Ferreira, L.M. Effect of activated carbon nanoparticles on the performance of PES nanofiltration membranes to separate Kraft lignin from black liquor. *J. Water Process Eng.* **2023**, *52*, 103487. [\[CrossRef\]](#)

23. Noorani, N.; Barzegar, B.; Mehrdad, A.; Aghdasinia, H.; Peighambaroust, S.J.; Kazemian, H. CO₂ capture in activated pyrolytic coke/metal oxide nanoparticle composites. *Colloids Surf. A Physicochem. Eng. Asp.* **2023**, *679*, 132554. [[CrossRef](#)]
24. Remy, M.; Potier, V.; Temmink, H. Why low powdered activated carbon addition reduces membrane fouling in MBRs. *Water Res.* **2010**, *44*, 861–867. [[CrossRef](#)] [[PubMed](#)]
25. Wang, Y.; Xu, Y.; Zhang, S.; Li, Y.; Liu, W. Effects of powdered activated carbon dosage on the performance of membrane bioreactors treating biochemical tail water. *Sci. Total Environ.* **2023**, *874*, 162429. [[CrossRef](#)] [[PubMed](#)]
26. Torretta, V.; Urbini, G.; Raboni, M.; Copelli, S.; Viotti, P.; Luciano, A.; Mancini, G. Effect of powdered activated carbon to reduce fouling in membrane bioreactors: A sustainable solution. Case study. *Sustainability* **2013**, *5*, 1501–1509. [[CrossRef](#)]
27. Kanagaraj, P.; Nagendran, A.; Rana, D.; Matsuura, T.; Neelakandan, S.; Malarvizhi, K. Effects of Polyvinylpyrrolidone on the permeation and fouling-resistance properties of Polyetherimide ultrafiltration membranes. *Ind. Eng. Chem. Res.* **2015**, *54*, 4832–4838. [[CrossRef](#)]
28. Ferreira, I.; Alves, P.; Gil, M.H.; Gando-Ferreira, L.M. Lignin separation from black liquor by mixed matrix polysulfone nanofiltration membrane filled with multiwalled carbon nanotubes. *Sep. Purif. Technol.* **2021**, *260*, 118231.
29. Sarbolouki, M.N. A general diagram for estimating pore size of ultrafiltration and reverse osmosis membranes. *Sep. Sci. Technol.* **1982**, *17*, 381–386. [[CrossRef](#)]
30. Nagendran, A.; Mohan, D. Cellulose acetate and polyetherimide blend ultrafiltration membranes: II. Effect of additive. *Polym. Adv. Technol.* **2008**, *19*, 24–35. [[CrossRef](#)]
31. Fitzpatrick, J.J.; Salmon, J.; Ji, J.; Miao, S. Characterisation of the Wetting Behaviour of Poor Wetting Food Powders and the Influence of Temperature and Film Formation. *KONA Powder Part. J.* **2017**, *34*, 282–289. [[CrossRef](#)]
32. Wu, C.; Dai, X.; Sun, X.; Zhang, J. Preparation and characterization of fluoroalkyl activated carbons/PVDF composite membranes for water and resources recovery by membrane distillation. *Sep. Purif. Technol.* **2023**, *305*, 122519. [[CrossRef](#)]
33. Aziz, M.H.A.; Othman, M.H.D.; Hashim, N.A.; Rahman, M.A.; Jaafar, J.; Hubadillah, S.K.; Tai, Z.S. Pretreated aluminium dross waste as a source of inexpensive alumina-spinel composite ceramic hollow fibre membrane for pretreatment of oily saline produced water. *Ceram. Int.* **2019**, *45*, 2069–2078. [[CrossRef](#)]
34. Almanassra, I.W.; Jaber, L.; Chatla, A.; Abushawish, A.; Shanableh, A.; Ali Atieh, M. Unveiling the relationship between MOF porosity, particle size, and polyethersulfone membranes properties for efficient decontamination of dye and organic matter. *Chem. Eng. J.* **2023**, *471*, 144616–144634. [[CrossRef](#)]
35. Awang Chee, D.N.; Ismail, A.F.; Aziz, F.; Mohamed Amin, M.A.; Abdullah, N. The influence of alumina particle size on the properties and performance of alumina hollow fiber as support membrane for protein separation. *Sep. Purif. Technol.* **2020**, *250*, 117147. [[CrossRef](#)]
36. Tsung-Shou, Y.; Michael, D.S. Effect of particle size distribution on the sintering of alumina. *Commun. Am. Ceram. Soc.* **1988**, *487*, 484–487.
37. Ji, Y.; Ke, J.; Duan, F.; Chen, J. Preparation and characterization of modified activated carbon/polysulfone blended ultrafiltration membrane. *Desalination Water Treat.* **2017**, *98*, 78–84. [[CrossRef](#)]
38. Mollahosseini, A.; Rahimpour, A.; Jahamshahi, M.; Peyravi, M.; Khavarpour, M. The effect of silver nanoparticle size on performance and antibacteriability of polysulfone ultrafiltration membrane. *Desalination* **2012**, *306*, 41–50. [[CrossRef](#)]
39. Goel, V.; Tanwar, R.; Saikia, A.K.; Mandal, U.K. Separation characteristics of surface modified polysulfone ultrafiltration membrane using oxidative catalytic polymerization of aniline. *J. Polym. Mater.* **2022**, *39*, 283–305. [[CrossRef](#)]
40. Erragued, R.; Kujawski, W.; Kujawa, J.; Gando-Ferreira, L.M.; Bouaziz, M. Optimizing operating conditions for olive leaf valorization using activated carbon mixed matrix membrane. *J. Water Process Eng.* **2024**, *59*, 105036. [[CrossRef](#)]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.