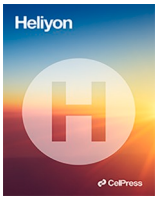
Heliyon 9 (2023) e21350

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon



Performance of modified hollow fiber membrane silver nanoparticles-zeolites Na–Y/PVDF composite used in membrane bioreactor for industrial wastewater treatment

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ARTICLE INFO

Keywords:
Membrane bioreactor
Hollow fiber membrane
Silver nanoparticles
Zeolites
Wastewater treatment

ABSTRACT

Membrane bioreactor (MBR) deteriorates due to fouling on the membrane pores, which can reduce the membrane performance. To reduce membrane fouling, the addition of inorganic filler can enhance the antifouling properties. This study investigates two different membrane preparation by thermally induced phase separation (TIPS) and dip coating methods to modify hollow fiber membrane with Silver Nanoparticles (AgNPs)-Zeolites used in MBR for industrial wastewater treatment. Performance was evaluated by analyzing the flux of water and wastewater, rejection, water content, and antifouling properties. Characterization result represented the synthesized silver nanoparticles had similar diffraction peak with commercial AgNPs, then the micrograph of AgNPs and zeolites addition membrane showed that the inorganic material had an octahedral shape representing zeolite crystal and irregular shape representing AgNPs. The addition of zeolites and AgNPs resulted in satisfying performance, increased flux, rejection, and antifouling properties.

1. Introduction

The effluent from wastewater-treatment plants (WTPs) is being considered a viable water resource for reclamation due to water scarcity, environmental degradation, and resource depletion [1,2]. Among other wastewater treatment technologies, membrane bioreactors (MBR) have received increased attention in recent years in both municipal and industrial wastewater treatment [3,4]. A

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https://doi.org/10.1016/j.heliyon.2023.e21350
Received 10 April 2023; Received in revised form 8 October 2023; Accepted 19 October 2023
Available online 22 October 2023

membrane bioreactor (MBR) is a wastewater treatment system that uses a membrane submerged inside or outside the bioreactor to separate activated sludge from water that has been treated in the bioreactor [5,6]. In addition, MBR technology has been widely applied in industry because of its advantages, which can decrease levels of chemical oxygen demand (COD), biological oxygen demand (BOD_5), and total suspended solids (TSS) up to 95 %, resulting in excellent effluent water quality, short water treatment times, minimal waste, and low space [6–8]. However, MBR also has a major challenge, including fouling, which can reduce the flux rate. Moreover, continuous fouling will cause failure in the wastewater treatment system [9,10].

Fouling occurs due to the accumulation of pollutants in wastewater and the formation of biofilms by activated sludge [11–13]. In recent years, many studies on the modification of membrane material by mixing zeolite into the membrane to increase the flux have been reported. The addition of Zeolite 4A to polysulfone membranes had better flux and antifouling values (flux 8.5 L m^{-2} h^{-1}, reversible fouling 83.5 %, and irreversible fouling 22.5 %) when compared to no addition (flux 2.5 L m^{-2} h^{-1}, reversible fouling 76.4 %, and irreversible fouling 19.8 %) [14]. In addition, the mixing of antimicrobial materials, such as silver nanoparticles, prevents the formation of biofilms on the membrane surface [15]. However, zeolite and silver nanoparticles decrease elasticity and increase stiffness, causing the membrane to be easily damaged when exposed to pressure [16]. The inorganic fillers reduced the elasticity of the membrane due to the difference in filler hydrophilicity with the membrane polymer's (Polysulfone) hydrophobicity, thus producing weak bonding among membrane polymers [17].

The preparation procedures significantly affect the properties and performance of the obtained membrane. Therefore, appropriate membrane preparation is an essential part of overcoming the issues. The thermally induced phase separation (TIPS) fabrication method produced a narrow pore size distribution of insoluble polymers in general diluents at room temperature [18]. In a previous study, the TIPS-fabrication based membrane showed good anti-fouling and high flux due to the higher hydrophilicity of the membrane, as indicated by the low contact angle value of 33.8° [19]. The membrane prepared using the TIPS method had a high water permeation flux, although the concentration of the polymer solution increased. This happens because the high temperature will decrease the viscosity of the solution so that it accelerates the mass exchange between the solvent and nonsolvent, resulting in a non-solid pore structure. Polyvinylidene membrane (PVDF) made by the TIPS method and the addition of perfluorosulfonic acid (PSFA) as an additive increased the porosity, surface hydrophilicity, and permeation flux of pure water (507.2 $\pm$ 1.2 L m^{-2} h^{-1} bar^{-1}) for membrane impressions at 90 °C [20].

The surface modification by dip coating method offers antifouling and stable mechanical properties [21,22]. The dip coating method coats both sides easier with excellent homogeneity than vacuum methods with similar filtration performance. Surface modification-based membranes are scratch-resistant and very durable in a harsh environment. This method is also applicable to a wide range of diameters or membrane pane sizes. It was reported that superhydrophilic silica NPs on PMAA-grafted membranes reduce foulant membrane interfacial forces, indicating their antifouling property [23]. On the other hand, introducing Cu NPs on the PVDF membrane surface exhibited high BSA rejection (80.5 %) with stable normalized flux. Moreover, it produced the highest FRR namely, 63.3 % [24].

Even though the performance of membranes is influenced by their fabrication methods, which have been previously studied, a thorough review of the literature reveals that no publication has directly compared various membrane preparation procedures for Ag-modified MBR. In this research, we examined two specific preparation methods: thermally induced phase separation (TIPS) and dip-coating. The orientation of the Ag position in the membrane contributes to the antifouling performance. Thus, investigating the different Ag modifications in membranes would provide the optimal process for Ag NPs modification. Therefore, this study develops a membrane with a zeolite and silver nanoparticle coating through the TIPS and dip-coating methods for industrial wastewater treatment applications.

2. Material and methods

2.1. Materials

The used materials, including sodium aluminate (NaOH, 99 %), sodium silicate (Na_2SiO_3, 99 %), silver nitrate ($AgNO_3$, 99.8 %), polyvinylidene fluoride (PVDF, Kynar®740, 99 %, Arkema Inc. Philadelphia), *N*-methyl-2-pyrrolidone (NMP, 99 %, Across Chemicals), polyvinyl alcohol (PVA, 99 %, Mw = 72.000 g/mol, Merck), glutaraldehyde (GA, 50 % (w/w), Merck), sulfuric acid (H_2SO_4, 98 %, Merck), and N-metil-2-pirrolidon (NMP, 99.5 %, Merck) were supplied by Merck. PVDF hollow fiber membranes were purchased from Hangzhou Kaihong Membrane Technology Co., Ltd., China, while distilled water was obtained from the local market.

2.2. Synthesis of Zeolite NaY

Zeolite NaY was synthesized using the hydrothermal method. The synthesis process followed some steps: (i) seed gel preparation; (ii) feedstock gel preparation; and (iii) overall gel preparation [25]. For the first step, 0.3469 g of NaOH and 1.924 g of sodium aluminate were mixed with 23.991 g of distilled water well mixed by a magnetic stirrer. Then 17.518 g of sodium silicate was added to the mixture and stirred for 10 min. Then, the mixture was aged for 24 h under room temperature. Then, for the second step, 0.1 g of NaOH and 13.85 g of sodium aluminate were mixed with 157.518 g of distilled water until well mixed by a magnetic stirrer. Then 107.212 g of sodium silicate were added to the mixture and stirred at 500 rpm. The feedstock gel was stored. For the last stage, 16.5 g of seed gel was added to the feedstock gel and stirred for 20 min at 600 rpm. Then, the overall gel was aged for 24 h at room temperature. After the aging process was completed, further hydrothermal processes were conducted in the autoclave [26–28] to form zeolite crystals at 100 °C for 7 h. The obtained product was filtered to separate the supernatant and washed with water until the pH was less

than 8.

2.3. Synthesis of AgNPs-Zeolite NaY

The 5 g of zeolite Na–Y was mixed into 10 ml of silver nitrate 0.1 M, then stirred to form a homogeneous solution. Then, 10 ml of 0.05 M trisodium citrate was added and stirred for 30 min at 6–10 °C, 3000 rpm. 10 ml of sodium borohydride 0.05 was dripped gently, followed by the addition of sodium hydroxide 1.25 M.

2.4. Membrane preparation

2.4.1. TIPS method

The preparation of dope solutions was modified from previous studies [22,29]. A few zeolite Na–Y or AgNPs/Zeolite Na–Y (0.1, 0.2, 0.3, and 0.4 g) were dissolved in NMP, followed by PVDF dissolution. The mixture was stirred at 60 °C for 18 h to obtain a homogenous mixture. The mixture was sonicated using the Ultrasonicator Faithful (Hebei China, Series FSF-020S, 40 kHz) then stored at room temperature to remove air bubbles. The mixture was heated to 90 °C followed by casting on a roller tube. The obtained cast sample was then immersed in water at room temperature.

2.4.2. Dip coating

The hollow membrane was coated by dip coating by modifying a previously reported procedure [21]. The hollow fiber membrane was coated with a polyvinyl alcohol solution. The coating solution was prepared by stirring 1.5 wt% PVA and 0.5 wt% GA in water for 4 h at 400 rpm. In addition, added Zeolite Na–Y or AgNPs/Zeolite Na–Y fillers into the solution and stirred until homogenized for 4 h at 400 rpm. Subsequently, the dope solution was sonicated for 30 min to remove the bubbles. The surface of the hollow fiber membrane was coated by soaking it in polyvinyl alcohol solution for 5 h and the membrane was dried at room temperature overnight.

2.5. Characterization

Rigaku Miniflex II instrument from Japan observed X-ray diffractograms (XRD) of prepared samples using CuK radiation at = 0.154, 40 kV, and 30 mA to confirm the crystal structure formation of the zeolite, AgNPs, and their composite. Employing scanning electron microscopy (SEM, Vega3 Tesc, Czech), the morphology of the membrane was analyzed. For the water quality assessment, the analysis methods of chemical oxygen demand (COD), biological oxygen demand (BOD), chemical oxygen demand (COD), and total suspended solids (TSS) corresponded to the Indonesian national standards, namely SNI 6989.73:2019, SNI 6989.72:2009, and SNI 6989.3:2019, respectively. The membrane thickness was measured using Vernier calipers.

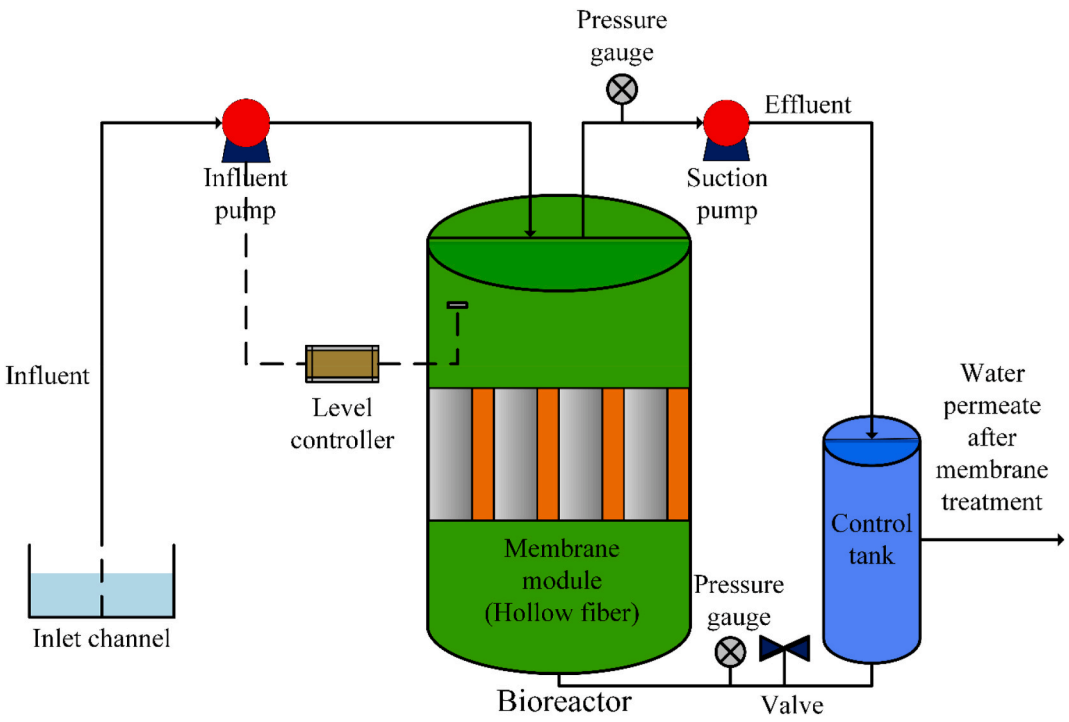

Fig. 1. Ultrafiltration reactor configuration.

2.6. Ultrafiltration process

The ultrafiltration test was carried out with a cross flow reactor system (Fig. 1) to evaluate the permeability, flux, rejection, water content, dan antifouling performance. The flux and rejection were calculated by following equations (1) and (2), respectively.

$$Flux = \frac{V}{A \times t} \tag{1}$$

$$Rejection = \frac{Initial\ water\ quality - Filtered\ water\ quality}{Initial\ water\ quality} \times 100\% \tag{2}$$

where V represents volume of permeate, A is membrane area that used, and t is operation time for filtration process. The water quality was indicated as COD, BOD, TSS, and TDS. Therefore, four rejection criteria will be obtained, corresponding to each water quality. The antifouling performance was measured by FRR (flux recovery ratio), reversible fouling resistance (R_{rev}), irreversible fouling resistance (R_{irev}), and total fouling resistance (R_{tot}) using Equations (3)–(6), respectively.

$$FRR = \frac{Permeation\ after\ washing}{Permeation\ before\ washing} \times 100\% \tag{3}$$

$$R_{rev} = \frac{Permeation\ after\ washing - Flux\ before\ washing}{Permeation\ before\ washing} \times 100\% \tag{4}$$

$$R_{irev} = \frac{Permeation\ after\ washing - Permeation\ before\ washing}{Permeation\ before\ washing} \times 100\% \tag{5}$$

$$R_{tot} = \left(1 - \frac{Flux\ before\ washing}{Permeation\ before\ washing}\right) \times 100\% \tag{6}$$

2.7. Membrane performance evaluation

The inoculated activated sludge was obtained from an industrial wastewater treatment plant in Gresik, Indonesia. The inoculated activated sludge has characteristics including MLSS 2750–2950 mg/L and DO 2–2.7 mg/L. The membrane performance evaluation was conducted at 0.5–0.7 bar. The wastewater from industrial production processes and corporate household activities, which are the main pollutants, was fed into the reactors. The wastewater quality was as follows: COD 2040 mg/L, TSS 415 mg/L, TDS 2050 mg/L, pH 7.11 $\pm$ 0.5, BOD 632 mg/L and temperature 28 °C.

The separation efficiency was evaluated by chemical oxygen demand (COD), biological oxygen demand (BOD_5), total dissolved solid (TDS), and total suspended solid (TSS) of the filtrate according to Indonesian National Standards such as COD, BOD_5, and TSS based on SNI 6989.73:2019, SNI 6989.72:2009, and SNI 6989.3:2019, respectively. The statistical technique employed to assess the significance of various membrane preparations was analysis of variance (ANOVA), conducted using Minitab 16 software (Minitab Inc., Institut Teknologi Sepuluh Nopember, Indonesia) [26,30].

3. Results and discussion

3.1. Materials characterization

3.1.1. X-ray diffraction analysis

The materials were characterized by XRD to determine their structure [31,32]. The synthesized zeolite Na–Y and silver nanoparticles have the same characteristic peak as JCPDS No. 39–1380 and JCPDS No. 04–0783, respectively. The synthesized zeolite Na–Y diffractogram shows a typical diffraction peak at 2θ of 6.2°, 10.1°, 11.8°, 20.2°, 23.5°, 26.9°, 30.5°, 31.2°, and 32.2° corresponding to (1 1 1), (2 2 0), (3 1 1), (4 4 0), (5 3 3), (6 4 2), (8 2 2), (5 5 5), and (8 4 0), respectively (Supplementary Fig. 1). The peak of zeolite-Y crystals with the highest intensity appears at 2θ of 6.2° which indicates high crystallinity and that no other phase is formed besides zeolite-Y. Furthermore, the XRD results of the synthesized silver nanoparticles (AgNPs) showed four XRD peaks of silver nanoparticles at 2θ of 38.3°, 44.3°, 64.6°, 77.5°, and 81.7° corresponding to (1 1 1), (2 0 0), (2 2 0), (3 1 1), and (2 2 2), respectively (JCPDS No. 04–0783) [33].

In the AgNPs-Zeolite NaY sample, a distinctive peak of silver nanoparticles appeared, indicating that the impregnation of silver nanoparticles on the surface of the zeolite Na–Y was successful. Moreover, the XRD peaks revealed that the composite consisted of PVDF and AgNPs/zeolites, with the structure of AgNPs being face-centered cubic (FCC) crystals. The average crystal size of silver nanoparticles calculated by the Debye-Scherrer equation was 27.69 nm.

3.1.2. Scanning electron microscope analysis

SEM analysis was performed to observe the morphology of the AgNPs/Zeolite Na–Y coated membrane (Supplementary Fig. 1). It can be seen that there are octahedral-shaped lumps, which indicate the presence of zeolite Y. SEM results show differences in dark and

light images that are influenced by its constituent elements. Compounding elements that have a higher atomic number will produce a brighter color image when compared to elements that have a lower atomic number [34–36]. In this study, the constituent elements of the AgNPs/Zeolite Na–Y coated PVDF membrane were Ag, Si, Al, Na, O, C, and F. In the SEM image, the brightest nodules, which are Ag elements, are spread evenly throughout the membrane surface. Therefore, the dip-coating process in this study was successfully carried out. The even distribution of these particles can increase the antifouling and antimicrobial properties of the membrane.

3.1.3. Tensile test

Stress, modulus young, and strain tests were carried out on PVDF membrane, Na–Y zeolite membrane, and AgNPs/Zeolite Na–Y membrane, as shown in Fig. 2a, b, and c, respectively. From the results of stress, strain, and Young's modulus, there is no significant difference between the three materials. it indicated that modifying the coating did not change the mechanical properties of the membrane [37]. Adding zeolite Na–Y to a PVDF membrane can alter its mechanical properties in several ways. Zeolite Y is a material with a high surface area that, when incorporated into a polymeric matrix, can modify the membrane's mechanical behavior. Zeolite Y is well-known for its exceptional mechanical strength and rigidity [38]. Incorporating zeolite particles into a polymeric membrane enables them to serve as reinforcing additives, thereby improving the membrane's overall mechanical properties. It can increase the membrane's tensile strength, flexural strength, and resistance to deformation [39]. In addition, zeolite Y has a greater modulus of elasticity than the vast majority of polymers [40]. It can enhance the composite material's rigidity when added to a polymeric membrane, making it less susceptible to warping or sagging under load. This can be especially beneficial in applications where membrane stability and dimensional integrity are essential. As a result, introducing Zeolite Y into the PVDF membrane improved the stress point while also decreasing the strain due to the membrane becoming stiffer. On the other hand, the addition of silver nanoparticles (AgNPs)/Zeolite Na–Y to PVDF membrane provided more impact on the improvement of the stress point and the reduction of membrane strain. This result was consistent with a prior study that demonstrated the AgNPs/SPS nanocomposite films became more resistant to break, stiffer, and less stretchable compared to the control SPS films [41]. The enhanced mechanical properties of the membrane were contributed by strong interactions between AgNPs/Zeolite Na–Y and the polymer matrix, facilitating adequate interfacial adhesion.

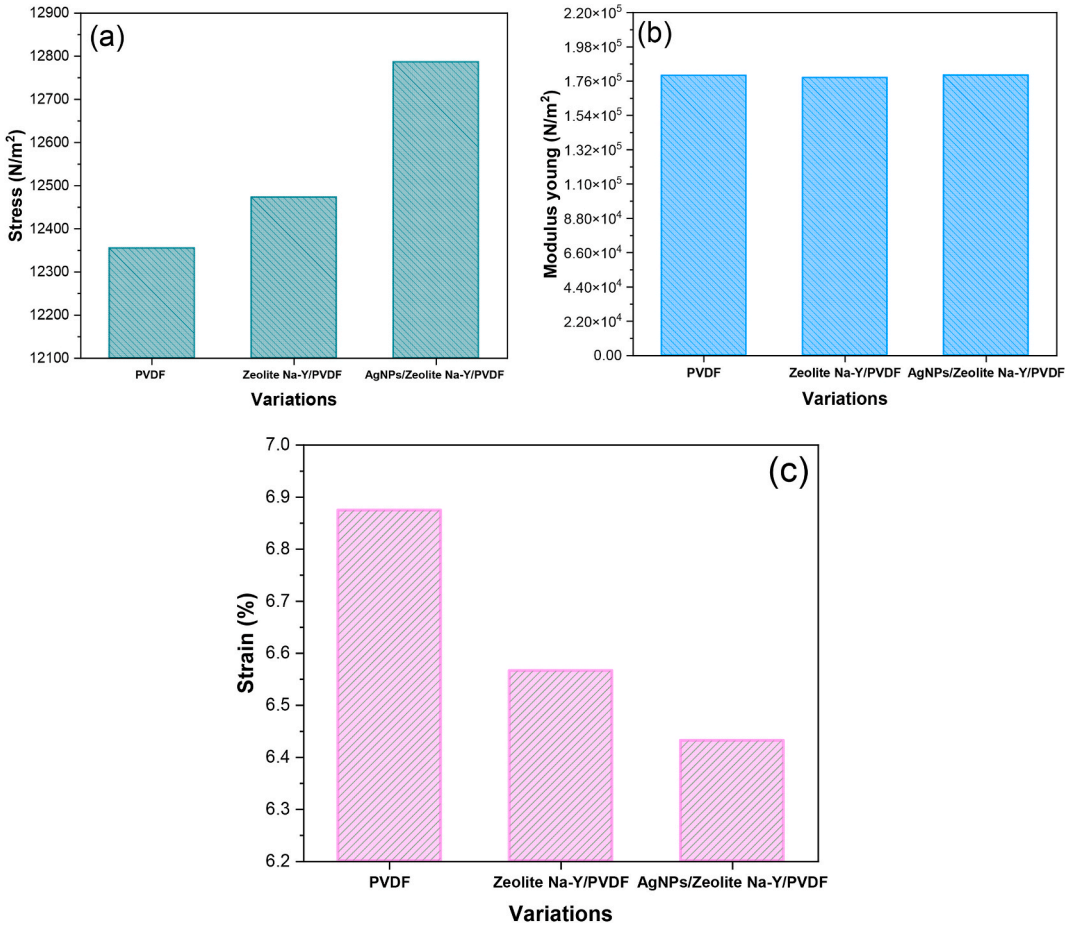

Fig. 2. (a) Stress, (b) modulus young, and (c) strain testing of the membrane.

3.2. Membrane performance

The membrane bioreactor's performance was evaluated by analyzing the flux, rejection, and water content of water and wastewater. Fig. 3a, b, c, and d depict flux performances of hollow fiber and flat sheet membrane using water and wastewater, respectively. The addition of zeolite and silver nanoparticles significantly increased the flux because of increased water absorption on the surface, since zeolites had high micropores and mesopores and thus easily absorbed water [42]. The structure of zeolites consists of alumina and silica linked with oxygen and hydroxyl groups. The hydroxyl group can strongly interact with water so that the zeolites can easily absorb water, as shown in Fig. 4 [42–44]. The highest water flux of membranes was obtained at 204.54 L/m^{2}h by the addition of 0.2 g AgNPs/Zeolite Na–Y into the PVDF matrix polymer membrane bioreactor. However, a higher loading concentration did not significantly increase the flux. It could also indicate that there is an optimal loading filler concentration beyond which further increases do not significantly enhance the system's performance. The addition of silver nanoparticles provided higher flux performance after washing when compared to zeolite addition alone [18]. This was consistent with the previous result result, which reported that introducing Ag nanoparticles aid to promote hydrophilicity of the membrane [45]. In addition, the presence of silver nanoparticles was also an inhibitor of the growth of microorganisms [25,46]. The antimicrobial activity of AgNPs was determined by two different processes, including the dissolution/release of AgNPs and Ag^+ and their reaction with cells [47]. On the other hand, hollow fiber membrane showed slightly higher flux than flat membrane. The hollow fiber provided better porosity, which decreased the resistance of the flow rate. In addition, hollow fiber modulation offers high specific surface area and packing density, and the associated membrane module is simple to build in the absence of spacers such as support mesh and grid [48].

Fig. 4 shows the water content of the membranes. The addition significantly increased the water content. A high water content indicates a higher hydrophilicity of the membrane. Previous reports have mentioned that the high hydrophilicity of the surface

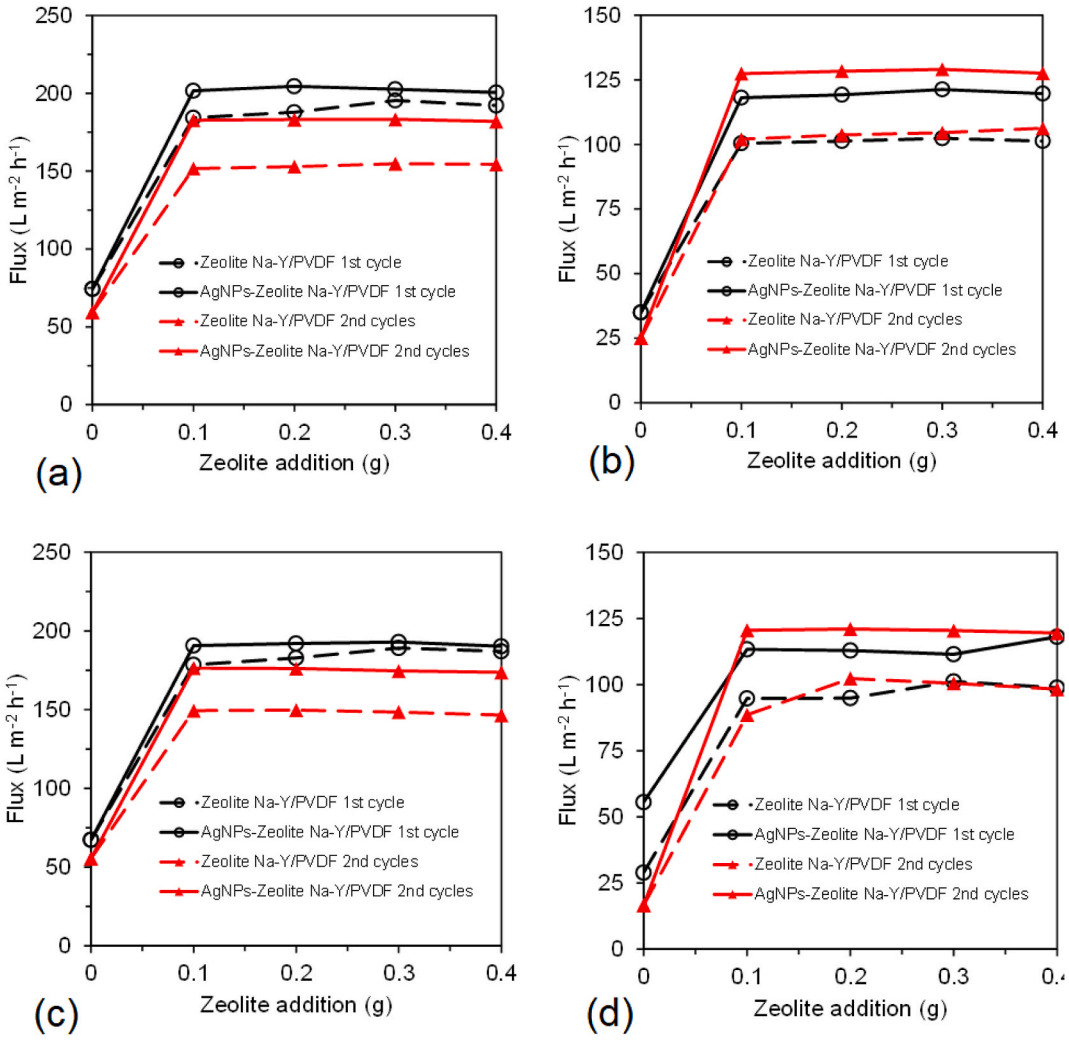

Fig. 3. Flux performance of hollow fiber membrane using water (a) and wastewater (b) as a feed, and flat sheet membrane using water (c) and wastewater (d) as a feed.

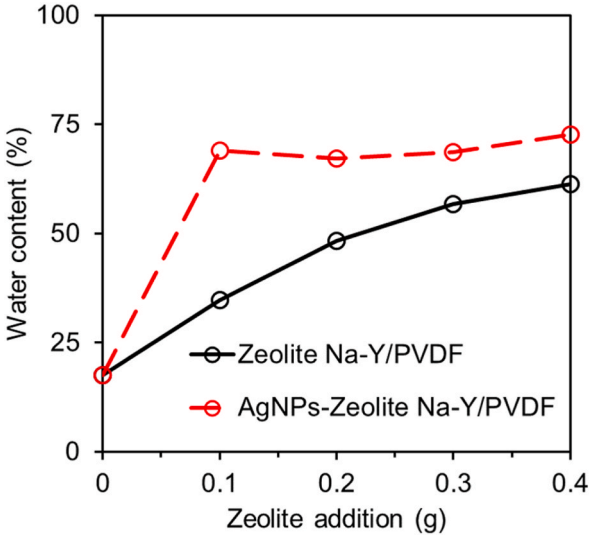

Fig. 4. The effect of zeolite addition on water content of Zeolite Na–Y/PVDF membrane and AgNPs-Zeolite Na–Y/PVDF membrane.

increases the water flux due to its positive capillary force [44,49,50]. As a result, AgNPs surface contains hydroxyl groups that attach water to form a water moiety.

Antifouling performances of Zeolite Na–Y hollow fiber membrane, AgNPs-Zeolite Na–Y/PVDF hollow fiber membrane, Na–Y Zeolite flat sheet membrane, and AgNPs- NaY/PVDF flat sheet membrane were shown in Fig. 5a, b, c, and d, respectively. The membrane with the zeolite's addition provided better antifouling properties compared to the neat membrane. Total fouling resistance (R_T), reversible fouling resistance (R_R) and irreversible fouling resistance (R_{IR}) decreased by zeolite addition. The reversible fouling resistance indicates that the clogging is removable. Low R_R and R_{IR} imply the fouling by removable and unremovable pollutants is minimal, respectively. The AgNPs showed low R_{IR} and contributed to the low R_T. AgNP zeolite Na–Y increases the hydrophilicity of the membrane significantly due to its high-water content, thus preventing irreversible fouling. A hydrophobic pollutant, organic matter, or bacteria will repel from the surface before it enters the pores of the membrane, preventing it from being an irreversible pollutant. In contrast, zeolite Na–Y alone had a lower water content, so the pollutant could penetrate the membrane pores, creating an irreversible pollutant. The prevented irreversible pollutant becomes a reversible pollutant due to its presence on the membrane surface. As shown in Fig. 5, the reversible pollutant was higher for AgNPs/zeolite Na–Y than zeolite Na–Y. In addition, silver nanoparticles acted as an inhibitor and degraded pollutants or microorganisms [20].

Fig. 6a, b, c, and d show rejection performances of hollow fiber membrane with addition zeolite, AgNPs-zeolite, and flat sheet membrane with addition zeolite, and AgNPs-zeolite, respectively. The membrane modified with zeolite and silver nanoparticles gave good COD, BOD_5, and TSS removal performance. However, the performance of TDS removal did not yield significant results. The permeation of TDS contaminants through the ultrafiltration membrane was facilitated by the presence of cations and anions [51]. The TDS removal performance with the addition of zeolite and silver nanoparticles was higher than without the addition of filler. Zeolite performs ion exchange [52], hence that it absorbs anions and cations contained in the TDS pollutants [37]. The highest rejection performance of the membrane was shown by filler loading 0.3 (g) zeolites addition and filler loading 0.3 (g) AgNPs/zeolite Y addition, namely water flux (195.45; 202.62 L/m^2.h), wastewater flux (104.53; 129.07 L/m^2.h), rejection (COD: 98.5; 99 %, BOD5: 98.5; 99 %, TSS: 99.8; 99.9 %, and TDS: 65.6; 66 %), FRR (82.5; 95 %), R_{rev} (26.84; 30.6 %), R_{irrev} (18.62; 9.28 %), and R_{tot} (4608; 4015 %), respectively.

Table 1 shows a comparison of the performance of membrane bioreactors with different fillers. The flux varied from 2.87 to 484 L m^{-2} h^{-1} [53–58]. The rejection rate also varies from 89.04 to 99.99 %. The membrane bioreactor with PVDF/PANI-TiO_2 filler has the highest flux value of 484 L m^{-2} h^{-1}, but the rejection rate is only 90 % [59]. When compared with other fillers, membranes with PVDF/Zeolite Na–Y and PVDF/Zeolite Na–Y AgNPs fillers have high flux values and high rejection rates. Hollow fiber membrane with PVDF/Zeolite Na–Y composite has a flux value of 106.26 L m^{-2} h^{-1} and a rejection rate of 98.382 %. While the hollow fiber membrane with PVDF/Zeolite Na–Y AgNPs composite has a flux value of 127.57 and a rejection rate of 99.019 %. Based on this, it can be concluded that the contribution level of AgNPs/zeolite composite membrane separation significantly improves membrane performance. The addition of AgNPs to the membrane also increases the hydrophilicity of the membrane, thereby reducing the hydrophobic interactions between the foulant molecules and the modified membrane surface. Thus, it can be concluded that the dip-coating method on PVDF membranes, Na–Y zeolite membranes, and AgNPs/Zeolite Na–Y membranes has advantages in flux values, COD rejection, and antifouling without affecting the mechanical properties of the membrane.

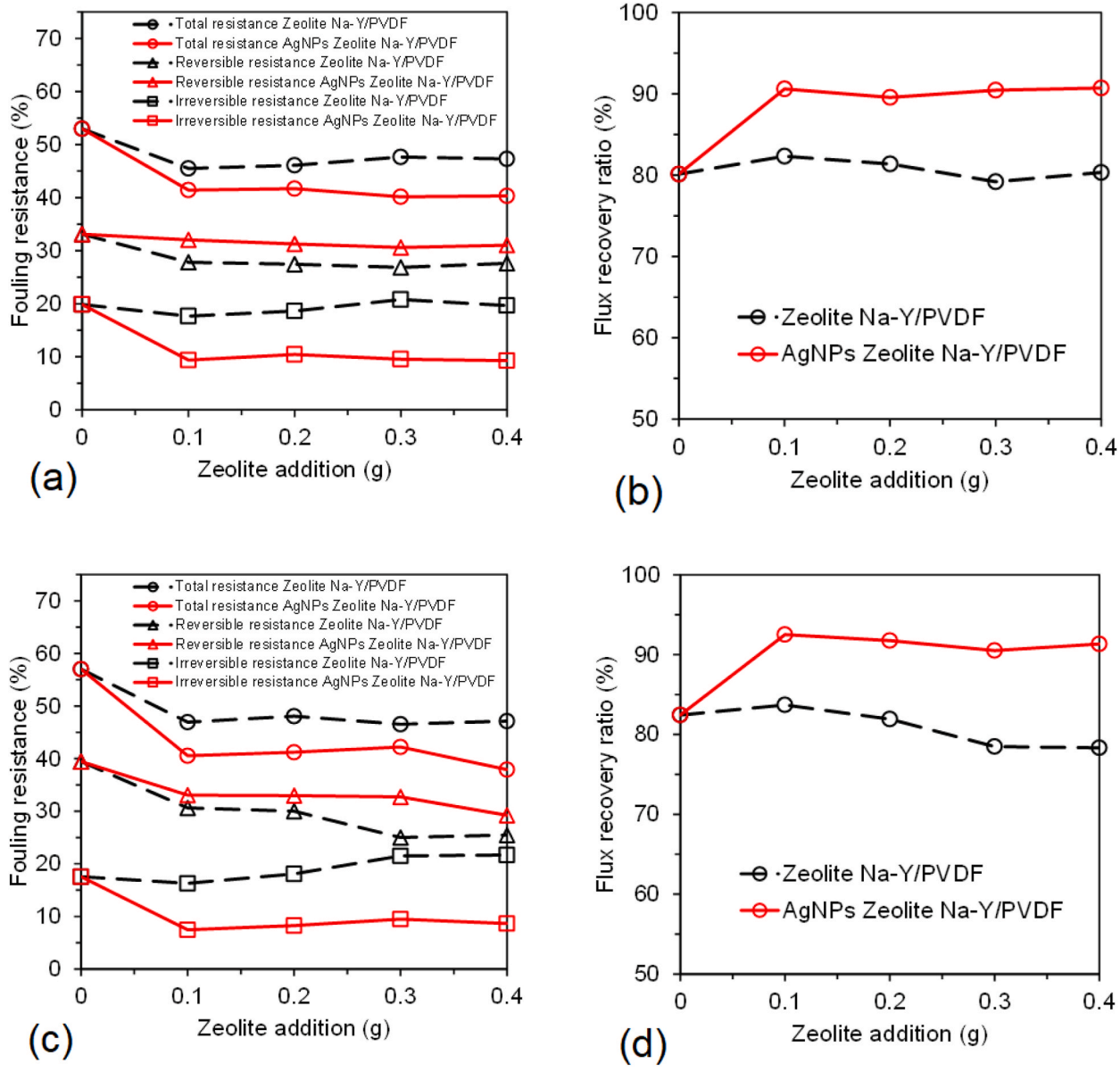

Fig. 5. Antifouling performance a) Zeolite Na–Y hollow fiber membrane, b) AgNPs-Zeolite Na–Y/PVDF hollow fiber membrane, c) Na–Y Zeolite flat sheet membrane, d) AgNPs- NaY/PVDF flat sheet membrane.

4. Conclusion

In this work, different types of membrane preparation, including TIPS and dip coating, were performed and analyzed. Hollow fiber modulation demonstrated more favorable with higher performance compared with flat sheet membranes due to the different surface areas of the membranes. In addition, zeolite Na–Y and AgNPs/Zeolite Na–Y at various loadings (0.1–0.4g) were introduced in the PVDF membrane bioreactor matrix and studied for their contribution to improving performances. The highest water flux of membranes was obtained at 204.54 L/m^2h by the addition of 0.2 g AgNPs/Zeolite Na–Y into the PVDF matrix polymer membrane bioreactor. The highest rejection performance of the membrane was shown by filler loading 0.3 (g) zeolites addition and filler loading 0.3 (g) AgNPs/Zeolite addition, namely COD: 98.5; 99 %, BOD_5: 98.5; 99 %, TSS: 99.8; 99.9 %, and TDS: 65.6; 66 %), FRR (82.5; 95 %), R_{rev} (26.84; 30.6 %), R_{irrev} (18.62; 9.28 %), and R_{tot} (4608; 4015 %), respectively. Moreover, the addition of zeolites and silver nanoparticles resulted in satisfying performance, increased flux, rejection, and antifouling properties. The best-performing membrane was obtained by the addition of zeolites and AgNPs/zeolite. The value of the performance was water flux (195.45; 202.62 L/m2.h) and Rejection (COD: 98.5; 99 %, BOD5: 98.5; 99 %). Furthermore, the addition of silver nanoparticles (AgNPs)/Zeolite Na–Y to PVDF membrane provided more impact on the improvement of the stress point and the reduction of membrane strain. This study suggested that adding

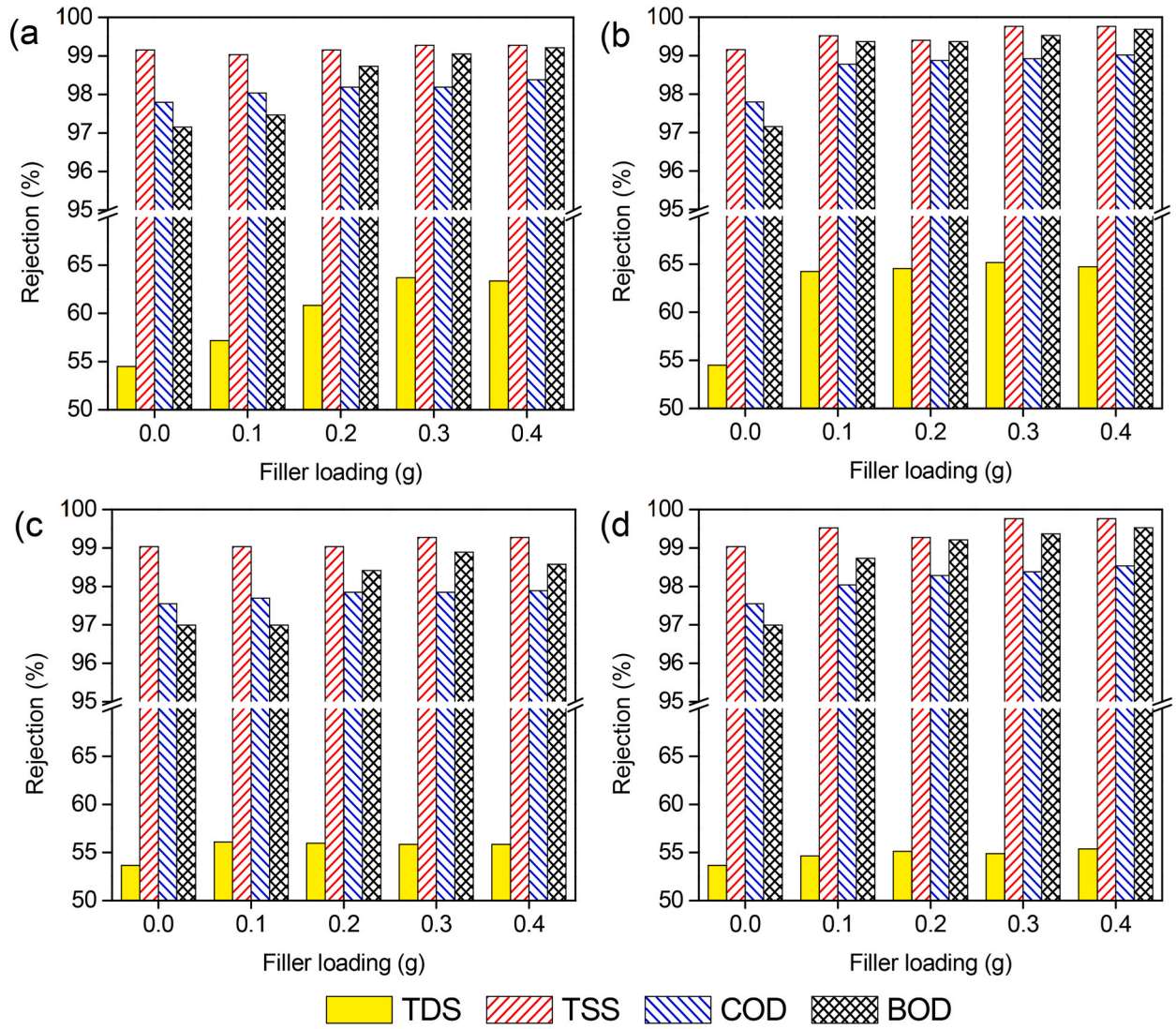

Fig. 6. Rejection performance of hollow fiber PVDF membrane with addition (a) zeolite Na–Y (b) AgNPs-zeolite Na–Y, and flat sheet PVDF membrane with addition (c) zeolite Na–Y (d) AgNPs-zeolite Na–Y.

AgNPs/Zeolite Na–Y filler into the PVDF membrane bioreactor matrix has advantages in flux values, COD rejection, and antifouling without adversely affecting the membrane's mechanical properties.

Funding declaration

The authors have no relevant financial or non-financial interests to disclose.

Data availability

Data will be made available on request.

CRediT authorship contribution statement

Rizki Fitria Darmayanti: Conceptualization, Data curation, Writing – original draft, Funding acquisition. **Maktum Muharja:** Data curation, Writing – review & editing, Investigation, Project administration. **Arief Widjaja:** Funding acquisition, Resources, Supervision. **Nurul Widiastuti:** Funding acquisition, Resources, Supervision, Validation. **Rahadian Abdul Rachman:** Data curation, Formal analysis, Investigation, Methodology, Writing – original draft. **Alvin Rahmad Widyanto:** Methodology, Validation, Writing – review & editing. **Abdul Halim:** Software, Visualization, Writing – review & editing. **Dendy Satrio:** Software, Validation, Visualization.

Table 1
Comparation of several membrane bioreactor performances.

Fillers	Method of Preparation	Remarks	Flux	Rejection	Rate of Rejection (%)	Ref.
PVDF/TiO_2SiO_2	Phase Inversion	Flat-sheet Membrane	11 kg/m^2h	Gallic Acid in Feed	99.9	[54]
PVDF/PANI-TiO_2	Phase Inversion	Nanocomposites Membranes	484 L/m^2h	Dye Rejection	90	[59]
Polypropylene/PVA	Dip-Coating	Hollow Fiber Microfiltration Membrane	25.7 L/m^2h	Salt Rejection	92.8	[55]
PVDF-HFP/TiO_2	Dip-Coating	Nanofiber	40 L/m^2h	Salt Rejection	99.99	[56]
PVDF/MOF-F300	Phase Inversion Process	Support Material/Nanofiber	2.87 kg/m^2h	Salt Rejection	99.99	[53]
PAN/Chitosan	Free radical graft copolymerization	Nanofiltration (NF) Membrane	229.82 L/m^2h	Dye Rejection	89.04	[57]
PVDF/H_3PO_4	Electrospinning	Flat-sheet composite membranes	12.50 kg/m^2h	Salt Rejection	91	[58]
PVDF/Zeolite Na–Y	Dip Coating	Flat-Sheet Membrane	98.3 L/m^2h	COD Rejection	97.84	This Work
PVDF/Zeolite Na–Y	Dip-Coating	Hollow Fiber membrane	106.27 L/m^2h	COD Rejection	98.38	This Work
PVDF/Zeolite Na–Y-AgNPs	Dip-Coating	Flat-Sheet Membrane	119.5 L/m^2h	COD Rejection	98.53	This Work
PVDF/Zeolite Na–Y-AgNPs	Dip-Coating	Hollow Fiber membrane	127.57 L/m^2h	COD Rejection	99.02	This Work

Bambang Piluharto: Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

The authors thank the Indonesia Endowment Fund for Education (LPDP) of the Ministry of Finance of the Republic of Indonesia through the Independent Research Grant with contract number 021/E4.1/AK.04.RA/2021.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2023.e21350.

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