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The Development of Polymeric Composite Membrane Containing Plant Waste Derived Activated Carbon and TiO_2 for Raw Petroleum Refinery Wastewater Treatment

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Abstract— **Petroleum industry results in a huge amount of harmful oily wastewater that must be properly treated. The fabricated composite membrane comprising polyvinylidene fluoride (PVDF), cassava peel-derived activated carbon (AC), and TiO_2 was then developed for treating raw petroleum refinery wastewater containing high loading of organic and inorganic pollutants. This study applied different ratios of AC/TiO_2 by 0, 1, 2, and 3% in the membrane fabrication solution to study the effect of composite ratio on the quality of permeate, fouling rate, and membrane tensile strength. The experimental work found that the ratio of AC/TiO_2 by 3% in the composite membrane outperformed the other ratios. The additives addition by 3% could achieve more than 40, 71, 40, and 67.5% of COD, oil, BOD_5, and phenol removal efficiency, respectively. It also reached a lower flux decline by 25% within 250 min of filtration time. Hermia's model was then applied to study the fouling mechanism occurring during membrane operation. Additionally, it was indicated that membrane tensile strength was also influenced by composite ratio, where higher PVDF amount resulted in higher membrane tensile strength. Overall, this study concluded that composite membrane could be a reliable alternative for treating raw industrial wastewater to existing wastewater treatment technologies. Further research related to the composite membrane integration system with other treatment techniques can then be conducted to lengthen membrane lifespan and improve the permeate quality.**

Keywords— **Cassava peel derived activated carbon; PVDF composite membrane; raw petroleum refinery wastewater; TiO_2; flux.**

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I. INTRODUCTION

Petroleum refinery industries need a huge amount of water to run several manufacturing processes. Consequently, they also produce a massive amount of wastewater containing high levels of impurity materials [1], [2]. To cope with this problem, petroleum industries should have the appropriate wastewater system to treat their refinery wastewater before its disposal can flow into the open environment. Untreated petroleum refinery wastewater (PRW) is harmful, affecting human health and other living creatures in the ecosystem due to its toxic organic and inorganic compounds [3], [4]. Therefore, to remove those hazardous pollutants and enhance the wastewater quality before discharging into the environment, some research attempted different treatment modes such as biological techniques, dissolved air flotation, adsorption, and skimming methods, or chemical approaches like coagulation, flocculation, and advanced oxidation processes [2], [5], [6].

However, there are still some challenges regarding those techniques, such as the need for a post-treatment stage, less efficiency, more chemicals needed, time-consuming reaction, and larger space for treatment device [7], [8]. Literature reported that adsorption studies using green resources have also been increasing for treating contaminated water and wastewater [9]. Those low-cost organic materials could be reliable alternatives to expensive commercial activated carbon [10].

Another prominent technique to treat wastewater is membrane filtration. Membrane technology offers advantageous features such as satisfactory output quality, simple apparatus design, and relatively simple operation[11], [12]. However, membrane technology still faces a problem called membrane fouling, and this fouling decreases permeate flux leading to high maintenance costs for membrane regeneration. There are some membrane types commonly used to treat industrial wastewaters, including ultrafiltration (UF) and microfiltration (MF) membranes fabricated from certain polymers, including cellulose acetate (CA), polyvinylidene fluoride (PVDF), polysulfone (PS), and polypropylene (PP) [13], [14].

In order to enhance membrane performance and mitigate membrane fouling rate, applying a pre-treatment or hybrid system and involving certain additives in membrane materials can be reliable solutions. A study compared sole nano-porous membrane and integrated adsorption/nano-porous membrane, for example, reported that single treatment was less effective than integrated system for reducing COD and TOC concentration [15]. Other works applied separate pre-treatment processes to treat wastewaters prior to entering the membranes system, and they also reported positive output of those double or triple treatment stages [7], [16], [17]. To date, combining pre-treatment and main treatment in one system is still challenging as they need to be compatible. Therefore, in terms of membrane application that has sensitive characteristics to oily wastewaters that specifically have high loading of organic and inorganic pollutants, some other studies have further tried to fabricate membrane enriched with several additives including graphite, ZnO, certain metals or minerals for better efficiency and lower fouling rate [12], [18].

Although increasing research has been devoted to better membrane performance, literature shows that research about the development of polymeric composite membranes containing specific additives such as plant waste-based carbon and photocatalytic particles to increase membrane capability in treating raw industrial wastewater is still limited. Other than that, very few studies still thoroughly focused on the influence of composite membrane ratio on more specific aspects.

Therefore, this work aimed to fill the gap in those existing research reports by; (1) assessing the effect of AC and TiO_2 ratio in the composite membrane on permeate quality, flux decline, membrane permeability, membrane surface morphology, tensile strength, and fouling mechanism, (2) examining plant waste as basic material for making AC, and (3) comparing the performance of commercial and composite PVDF based membrane developed in this work.

II. MATERIALS AND METHOD

A. Petroleum Refinery Wastewater

Refinery wastewater was collected from the wastewater pond of Pertamina Pty Ltd, a state-owned oil and natural gas company located in South Sumatera, Indonesia. The sample was previously filtered to eliminate any solid particles greater than a millimeter in size.

TABLE I
CHARACTERISTICS OF RAW PRW USED IN THIS STUDY

No.	Parameter	Value
1	BOD_5 (mg/L)	29500
2	COD (mg/L)	74102
3	Oil (mg/L)	368
4	Dissolved sulfide (mg/L)	0.72
5	Ammonia (mg/L	379
6	Phenol (mg/L)	0.8
7	Temperature (°C)	27.6
8	pH	6.03

Then characterization of the PRW was conducted immediately after arriving in the laboratory to avoid physical and chemical deterioration due to biological influence (Table 1). Then all the samples were put in a laboratory refrigerator for further usage.

B. Chemicals and Analytical Reagents.

This work used analytical reagents grade. The solution of either 0.1 M HCl or 0.1 M NaOH was used to adjust pH level. Titanium oxide (TiO_2), PVDF, N-methyl-2-pyrrolidone (NMP), and deionized water were obtained from Merck, Indonesia. Spectrophotometer UV-VIS Varian Cary 50 was applied to measure BOD_5, COD, and oil concentration based on the standard procedures (SNI 66-2503, APHA 5220, and SNI 06-6989). Laboratory oven Carbolite PN60, pH meter Eutech, furnace Carbolite AAF 11/7, scanning electron microscopy (SEM) FEI Inspect-S50 type, and hydraulic universal tester 50 Kn were also used in this work.

C. Activated Carbon Preparation

Different biomaterials, namely banana peel, orange peel, melon peel, watermelon peel, rambutan peel, and cassava peel, were previously investigated as potential activated carbon base materials. Those materials were washed several times with distilled water, followed by drying at 100 °C for 24 h in a laboratory oven. They were then crushed by a mechanical grinder to obtain powder form and passed through a sieve. The carbonization process was conducted by putting the prepared material in a laboratory furnace to 400 °C and maintained for 1 hour. It was then left to adapt to room temperature before storing properly. After evaluation, cassava peel was found to have a relatively higher removal efficiency of COD, oil, BOD_5, and phenol in raw petroleum refinery wastewater. Therefore, cassava peel was further chosen as an additive for composite PVDF membrane fabrication along with TiO_2 particles.

D. Membrane Composite Preparation

The fabrication of composite PVDF membrane were initially prepared by non-solvent induced phase separation (NIPS) technique [18], [19]. Different amounts of AC and TiO_2 nanoparticles were pre-dispersed in NMP solution followed by NH_4Cl in a beaker and then sonolyzed for 30 min to prevent agglomeration. The varying ratio of PVDF was then added to the mixture at temperature of 40 °C, and under constant stirring for at least 12 h until a homogeneous solution was obtained. Different ratios in the composite are revealed in Table 2. The casting solution was kept at 60 °C for 24 h, and the cast was put on a suitable glass plate by an auto casting knife followed by immersion in deionized water. Eventually, after coagulation, the cast membrane was stored in deionized water over night to remove any residuals before storing for further usage.

TABLE II
COMPOSITE MEMBRANE RATIO

Membrane	PVDF (wt%)	NH_4Cl (wt%)	NPM (wt%)	AC+TiO_2 (wt%)
I	15	0	85	0
II	13	1	85	1
III	11	2	85	2
IV	9	3	85	3

E. Membrane Flux and Fouling Analysis

A dead-end conducted flux measurement stirred cell (Amicon Corp) supported by a nitrogen cylinder for the needed pressure (Fig. 1). the membrane was conditioned with deionized water at a pressure of 2 bar. The volume of the permeate water was collected and measured every 10 min. The same experimental setup was applied for raw petroleum refinery wastewater. Two tests were performed for each membrane. To determine the permeation flux of the membrane, the following equation was applied [7], [20]:

$$J = \frac{V}{At} \quad (1)$$

where J, V, A, and t are the permeation flux (L/m^2 h), the volume of collected permeate (L), active membrane area (m^2), and time taken in permeate collection (h), respectively.

To evaluate the removal efficiency of targeted pollutant, the following equation can be used:

$$\text{Removal efficiency, } (\%) = (1- \frac{C_p}{C_f})\ x\ 100 \quad (2)$$

Where C_p (mg/L) and C_f (mg/L) are the concentration of a targeted pollutant in permeate and in feed, respectively.

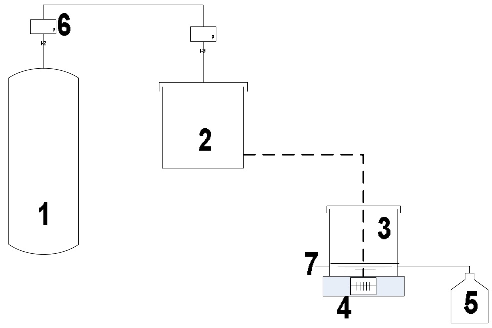

Fig. 1 PVDF Membrane system (1) N_2 gas cylinder, (2) Feed tank, (3) Dead-end stirred cell (4) Magnetic stirrer (5) Beaker glass (6) Pressure gauge (7) Membrane composite

The analysis of membrane fouling phenomena could be done by applying Hermia's model with its general equation written as follows [21]:

$$\frac{d^2t}{dV^2} = K\left(\frac{dt}{dV}\right)^n \quad (3)$$

Using the above equation involving the derivation of flux (J) with time (t), specific Hermia's formula then can be rewritten as follows:

$$\frac{dJ}{dt} = -K\,(J - J_{ss})J^{2-n} \quad (4)$$

where K is a constant, J_{ss} is permeated flux in steady condition, n = 0, 1, 1.5, and 2 refer to cake filtration, intermediate blocking, standard blocking, and complete blocking model, respectively. The cake formation model occurs when pollutant particles are bigger than the average pore size building up a cake layer thickening by the time forming multiple layers of pollutant particles. In intermediate blocking, particles block some pores creating intermediate fouling related to the equivalent size of pollutant particles and membrane pores. In the standard model, a decrease in pore diameter is caused by the non-uniformity of a particle trapped on the membrane pore, while the complete pore blocking model assumes that particles with a bigger size than membrane pores settle on the membrane surface, causing flux decline [4], [22].

III. RESULTS AND DISCUSSION

A. Activated Carbon and Characterization

Cassava peel-derived AC seemed to have relatively better adsorption capacity than other selected plant waste materials investigated in this study. It then was utilized and used in further experimental works. The characteristics of prepared AC are figured in Table 3. It can also be observed that AC has a relatively high proportion of carbon at 75.5%.

TABLE III
CASSAVA PEEL AC CHARACTERISTICS

No.	Parameters	Obtained percentage (%)	SII standard (%)
1	Volatile matter	18	25
2	Water content	7.5	15
3	Ash content	6.5	10
4	Activated carbon content	75.5	65

B. Effect of Additives Ratio on Permeate Quality

Fig. 2 illustrates the pollutants removal efficiency of PVDF composite membranes having different ratios. The additives addition led to relatively significant improvement in membrane performance to decrease COD, oil, BOD_5, and phenol concentration in raw PRW. This phenomenon can be related to the capability of both additives to reduce the contact angle of water on the membrane surface leading to an increase in membrane hydrophilicity [18]. This contributes to purer permeate output due to a better rejection of the targeted pollutants.

The experimental work showed that the more additives substances used in the membrane composite, the better the performance of the membrane to remove targeted pollutants. The addition of AC/TiO_2 by 3% could reach 40% and 71% of COD and oil removal efficiency, respectively. In contrast, a membrane that has no AC/TiO_2 depicted the lowest COD and oil removal efficiencies by 20% and 50%, respectively. Under BOD_5 and phenol, the fabricated PVDF membrane has also proven its potentiality. PVDF membrane could reduce the concentration of both BOD_5 and phenol by 50% and 20%, respectively.

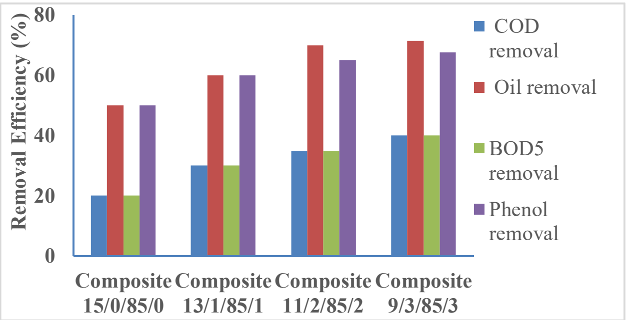

Fig. 2 Effect of additives substance and membrane composite composition (PVDF: NH_4Cl: NPM: AC/TiO_2) on COD, oil/grease, BOD_5 and phenol removal efficiency

However, it is worthy mentioning that the existence of additive substances has boosted membrane performance in terms of BOD_5 and phenol removal efficiency. In general, PVDF membrane has a better ability to simultaneously decrease COD, oil, BOD_5, and phenol contents when it has additional support from compatible materials such as AC/TiO_2 due to the prowess of those additives in reducing membrane fouling and increasing permeate quality. It is believed that photocatalytic substances, including TiO_2 can do a self-cleaning process related to the antifouling effect by preventing oil from contacting the membrane surface, while AC contributes to the membrane efficacy by firstly adsorb pollutant particles through its finite and active pores sites [12], [18], [23].

C. *Membrane Permeability*

Membrane permeability can be defined as the ability of membrane pores to pass in and out of specific substances. It is influenced by membrane type, material, and parameters, including pressure difference, feed velocity, and wastewater characteristics [7], [24]. Generally, deionized water is used to measure membrane permeability.

Fig. 3 shows the permeability measurement of each composite membrane fabricated in this work. Water flux of fresh membrane (J_{wi}), membrane water flux after wastewater treatment (J_{ww}), and membrane water flux after cleaning process (J_{wc}) are three parameters used for analyzing membrane permeability. In this work, the cleaning process was conducted using deionized water for around 60 min to clean membrane pores and surfaces. As can be observed, both additives (TiO_2 and AC) increased membrane permeability. This trend continued with the increase in additives ratio where membrane having 3% of additives could reach the highest values of $J_{wi,}$ J_{ww} and J_{wc} by 197, 167, and 187 L/m^2h, respectively.

These values were significantly higher than membrane having no TiO_2/AC addition, which reached $J_{wi,}$ J_{ww} and J_{wc} by 124, 87, and 112 L/m^2h of, respectively. This phenomenon can be attributed to those additives affecting hydrophilicity, water contact angle, and enlarging membrane porosity. After one cycle of treatment, the decrease in J_{wc} value was noticeable for all ratios, and this may be caused by a strong bonding between residual pollutants and membrane pores even after a certain cleaning process. Therefore, some chemicals and operating conditions could be considered for better membrane recovery.

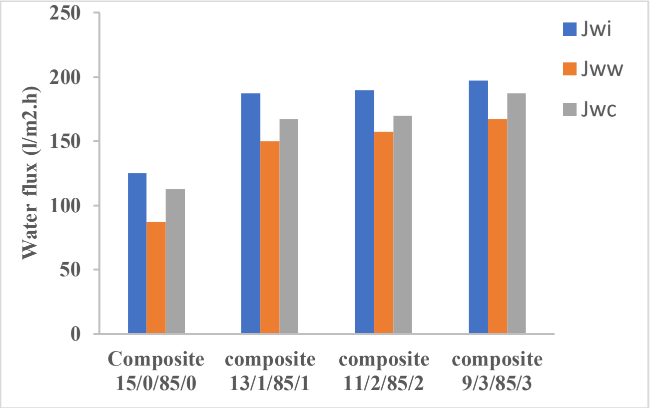

Fig. 3 The permeability flux of composite membrane used in this work

D. *Effect of Composite Ratio on Permeate Flux*

To assess the influence of composite ratio on permeate flux, different ratios of membrane casting solution were investigated, and the results were depicted in Fig. 4. Membranes were tested in terms of their flux changes during filtration time. Based on the experimental work displayed in the figure, it can be assumed that the effect of additive substances on permeate flux is noticeable where membrane containing higher ratio of AC/TiO_2 showed higher flux and, at the same time, lower membrane fouling rate [25].

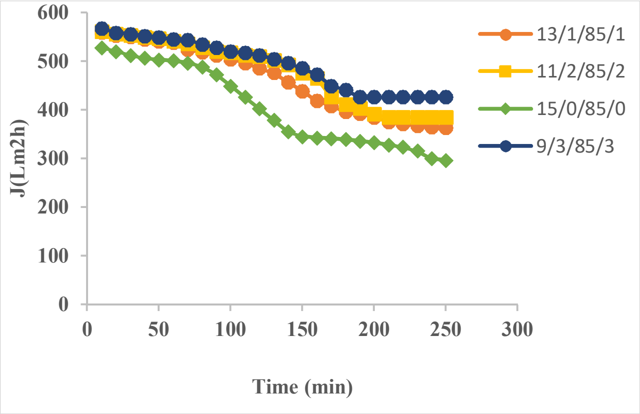

Fig. 4 The comparison of membrane flux profile regarding its composition ratio (PVDF/NH4Cl/NPAC + TiO_2)

According to Fig. 4, all membranes containing additives tended to have similar flux values within the first 100 min before experiencing visible different flux profiles. Reversely, PVDF membrane with no AC/TiO_2 addition showed lower starting flux and faster fouling rate by about 38% of flux decline within 250 min of filtration time. This might be related to the struggling PVDF-based membrane to filter polluted fluid causing earlier clogged pores solely. The composite membrane containing additives could perform better due to the antifouling effect of the photocatalytic and AC[18].

From another point of view, there are obvious differences in permeate flux decline at the end of filtration time among composite membranes containing AC/TiO_2 where a higher additives ratio resulted in a lower fouling rate. Specifically, composite membranes having 3% TiO_2/AC experienced just 25% flux decline while others with 2, 1, and 0% TiO_2/AC additives faced 31, 35, and 43.8% of flux decline, respectively. This indicated that those additives positively contribute to the performance of the membrane by protecting the membrane pore from severe clogging.

Furthermore, in order to assess the specific benefit of this research, the comparison between fabricated and commercially available equivalent PVDF membrane was investigated regarding flux decline percentage (Fig.5). The same operating condition and feed characteristics were applied in this evaluation.

Based on Fig. 5, it can be noticed that fabricated composite membrane containing TiO_2/AC could perform better than commercial membrane by having lower percentage of flux decline. Overall, in the last 10 min, flux decline percentages for fabricated composite membrane

($PVDF/TiO_2/AC$), commercial, and fabricated PVDF membranes were 25, 28.5, and 43.8 %, respectively.

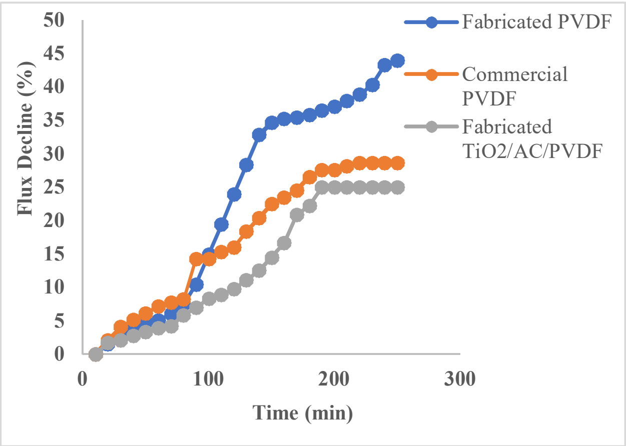

Fig. 5 The comparison of flux decline percentage among commercial PVDF, fabricated PVDF, and fabricated TiO2/AC/PVDF membranes

E. *Membrane Morphology and Tensile Strength*

SEM analysis was conducted to thoroughly study membrane morphology and verify the differences of surface and pore appearance, SEM analysis was then conducted, and the images are shown in Fig. 6. Based on the figures, it can be seen that each fabricated membrane has its unique micro pattern.

Fig. 6 indicates tangible differences in surface morphology. As portrayed in Fig. 6.1, the membrane without AC/TiO_2 seemed to have a smoother surface. The addition of blended additives then changed the membrane surface and pores images (Figs. 6.2-6.4).

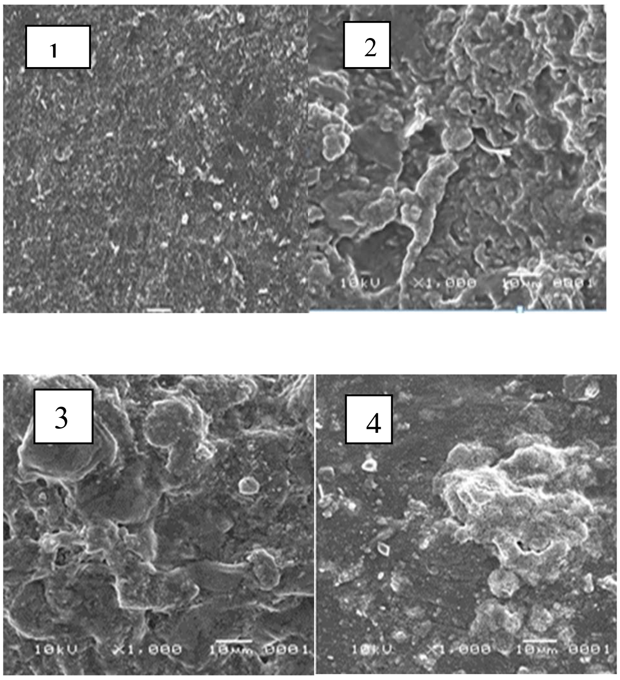

Fig. 6 SEM images of membrane composites with various compositions of PVDF/ NH_4Cl/ NPM/ AC+TiO_2 (1) 15/0/85/0, (2) 13/1/85/1, (3) 11/2/85/2, (4) 9/3/85/3

However, the pore size could be assumed different amongst those $PVDF/AC/TiO_2$ composite membranes. Overall, composite membrane blended with AC/TiO_2 seemed to have rougher surface structures. It may be caused by the higher viscosity level of the casting solution as well as the increase in membrane surface porosity [18], [26].

Furthermore, membrane tensile strength can be defined as the maximum stress that can be withstood by membrane material when it gets stretched, leading to a breaking point. The tensile strength of a membrane is measured by a tensile testing device dividing the maximum stress before the specimen fails or breaks with the initial value of the cross-section area of the membrane sample. Tensile strength values of membrane materials fabricated in this work were measured using a hydraulic Universal Tester 50 Kn, and the experimental results were depicted in Table 4.

TABLE IV
TENSIL STRENGTH MEASUREMENT

Membrane composite composition PVDF/ NH_4Cl/ NPM/ AC+TiO_2	**Tensile strength value (N/mm^2)**
15/0/85/0	5,33
13/1/85/1	4,43
11/2/85/2	4,27
9/3/85/3	4,19

Tensile strength influences the membrane's durability, which is experiencing continuous mechanical solicitations during filtration operations. As can be seen in Table 4, the composite membrane having 15/0/85/0 of PVDF/ NH_4Cl/ NPM/ AC+TiO_2 ratio showed higher tensile strength. This can be attributed to the higher amount of PVDF polymer, which has better flexibility and durability characteristics supporting the strength of the composite.

F. *Fouling Mechanism by Hermia's Models*

In accordance with the fouling rate on membrane pores and surface, Hermia's model can be a reliable model to understand the probable mechanism occurring during filtration. The modeling results summarised in Table 5 shows a correlation coefficient (R^2) indicating the fouling pattern of each composite membrane. Based on the table, the membrane with no TiO_2/AC addition appears to be in intermediate pore blocking mode, with R^2 of 0.943. This indicated that pollutant particles blocked the pores resulting in fouling occurrence where the pollutant particle could have the equivalent size to the membrane pores [27]. This outcome also indicates that Hermia's model is applicable in most of the membrane types giving probable explanation of fouling mechanism during filtration process [28], [29], [30]. In contrast, all composite membranes containing AC/TiO_2 additives followed the complete pore blocking model with R^2 values of 0.9679, 0.9258, and 0.9464 for 1, 2, and 3% of TiO_2/AC ratio, respectively.

TABLE V
FOULING MECHANISM USING HERMIA'S MODEL

Composite membrane	**Correlation coefficient (R^2)**			
	Complete blocking	**Standard blocking**	**Intermediate blocking**	**Cake**
15/0/85/0	0.95	0.952	0.954	0.952
13/1/85/1	0.967	0.963	0.958	0.946
11/2/85/2	0.925	0.918	0.910	0.894
9/3/85/3	0.946	0.942	0.937	0.926

The complete pore blocking model assumes that every pollutant particle is having a bigger size than membrane pores would settle on it. However, it is not superimposed upon others, creating a single layer of pollutant particles blocking all pores, not the area within them. In this case, the function of additives previously predicted to have the capability of antifouling and self-cleaning has been proven by decreasing the fouling rate.

IV. CONCLUSION

The PVDF composite membranes having 0, 1, 2, and 3% of AC/TiO_2 ratio were fabricated for treating raw PRW in order to know the effect of selected compositions on permeate quality, membrane permeability, flux decline, and tensile strength. This work then found that the sole PVDF membrane could reduce the concentration of COD, oil, BOD_5, and phenol to some extent. Then, adding AC/TiO_2 to PVDF membrane at any percentage has boosted the removal efficiency of those measured parameters. Other than that, the effect of those additives could positively increase composite membrane performance by lowering fouling rate and increasing permeate flux. The fouling mechanism analyzed by Hermia's models follows the complete pore blocking model. In addition, despite having a higher fouling rate, PVDF membrane with no AC/TiO_2 addition showed higher tensile strength. Eventually, in terms of fouling rate and permeate flux increase, fabricated PVDF membrane having 3% of AC/TiO_2 showed better performance compared to both commercially available equivalent PVDF membrane and fabricated PVDF without the additives.

NOMENCLATURE

J	permeation flux	$\mathrm{L\ m^{-2}\ h}$
V	volume of collected permeate	L
A	active membrane area	$\mathrm{m^2}$
t	time taken in permeate collection	h
C_p	permeate concentration	mg/L
C_f	feed concentration	mg/L
K	a constant	
J_{ss}	permeate flux in steady condition	$\mathrm{L\ m^{-2}\ h}$

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