EFFECT OF CARBON NANOMATERIALS CONCENTRATION IN NANOCOMPOSITE MEMBRANE FOR METHYL BLUE DYE REMOVAL

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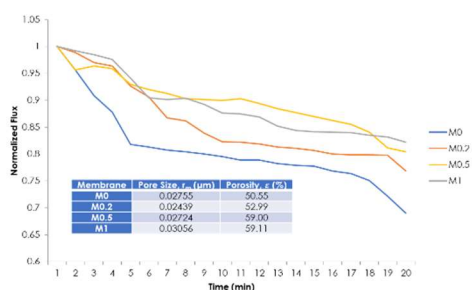[b]Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600, UKM Bangi, Selangor Darul Ehsan, Malaysia

Graphical abstract

Abstract

It is reported that the membrane properties can be enhanced by nanomaterials. However, agglomeration will occur due to the overdose of nanomaterials subsequently deteriorating membrane performance. The project aims to investigate the effect of concentration of cabon nanomaterials: multiwalled carbon nanotubes (MWCNTs) and graphene oxide (GO), on nanocomposite membrane for methyl blue (MB) dye removal. The GO/MWCNTs nanocomposite membranes were synthesized using direct blending method with three concentrations (0.2, 0.5, and 1 wt.%). The synthesized membrane was characterized by surface hydrophilicity, pore size and porosity, surface charge, functional group, and surface morphology. Besides, the performance of the synthesized membrane was evaluated by water permeability test, dye rejection test, and antifouling test. The result shows that the surface hydrophilicity was enhanced when the concentration of nanomaterials increased up to 0.2 wt%. However, higher concentration of nanomaterials reduces the membrane hydrophilicity due to the agglomeration of nanomaterials. The membrane with nanomaterials concentration of 0.2 wt.% (M0.2) has the best performance as it showed 6.85% and 32% improvement in water permeability and dye rejection when compared to the pristine membrane. Besides, M0.2 membrane has relatively good antifouling properties indicated by normalized flux (0.8043). This is due to the enhancement of hydrophilicity and zeta potential of M0.2 membrane by carbon nanomaterials. In short, optimum concentration of carbon nanomaterials are essential to enhance the membrane performance as agglomeration of nanomaterials occurs at high concentration.

Keywords: Carbon nanomaterials concentration, graphene oxide, multiwalled carbon nanotubes, nanocomposite membrane, membrane antifouling

Abstrak

Dilaporkan bahawa sifat membran boleh dipertingkatkan oleh bahan nano. Walau bagaimanapun, penggumpalan nano akan berlaku kerana kepekatan bahan nano yang tinggi seterusnya merosot prestasi membran. Projek ini bertujuan untuk mengkaji kesan kepekatan bahan nano kabon:

nanotiub karbon multi-dinding (MWCNTs) dan grafin oksida (GO), pada membran nanokomposit untuk penyingkiran pewarna metil biru (MB). Membran nanokomposit GO/MWCNTs telah dihasilkan menggunakan kaedah pengadunan langsung dengan tiga kepekatan (0.2, 0.5, dan 1 wt.%). Membran dicirikan oleh hidrofilik permukaan, saiz liang dan keliangan, cas permukaan, kumpulan berfungsi, dan morfologi permukaan. Selain itu, prestasi membran dinilai dengan ujian kebolehtelapan air, ujian penolakan pewarna, dan ujian anti-kotoran. Keputusan menunjukkan bahawa hidrofilik permukaan dipertingkatkan apabila kepekatan bahan nano meningkat sehingga 0.2 wt.%. Walau bagaimanapun, kepekatan bahan nano yang lebih tinggi mengurangkan hidrofilik membran disebabkan oleh penggumpalan bahan nano. Membran dengan kepekatan bahan nano 0.2 wt.% (M0.2) mempunyai prestasi terbaik kerana ia menunjukkan peningkatan 6.85% dan 32% dalam kebolehtelapan air dan penolakan pewarna jika dibandingkan dengan membran tulen. Selain itu, membran M0.2 mempunyai sifat anti-kotoran yang agak baik dibuktikan oleh fluks ternormal (0.8043). Ini disebabkan oleh peningkatan hidrofilik dan potensi zeta membran M0.2 oleh bahan nano karbon. Ringkasnya, kepekatan bahan nano karbon yang optimum adalah penting untuk meningkatkan prestasi membran kerana aglomerasi bahan nano berlaku pada kepekatan tinggi.

Kata kunci: Kepekatan bahan nano karbon, grafin oksida, *nanotiub karbon multi-dinding,* membran nanokomposit

1.0 INTRODUCTION

Nowadays, the textile industry has become a mainstream industry and contributor to many national economies. However, the textile industry produces wastewater that is harmful to the environment and ecosystem. Wastewaters produced by textile are mainly contaminated with dye, metals, and solvents. These contaminants will significantly damage the water bodies by increasing the biochemical and chemical oxygen demand and retard photosynthesis of the aquatic plant [1]. Thus, wastewater treatment played a vital role in protecting the water bodies by reducing the effects of contaminants [2].

Wastewater treatment is a process to remove impurities and contaminants from wastewater before it is discharged to the aquifer or natural bodies. The textile industry wastewaters must be treated by complying with the environmental protection laws before discharge into the water body. Generally, textile industry wastewater can be remedied by different treatment methods such as physical, chemical, and biological methods [3]. Recently, membrane technology has become a noble separation technology due to its advantages. The membrane could operate with low chemical consumption and relatively low energy, with no complex instrumentation and clean technology at ease of operation [4].

However, membrane fouling is a significant issue for membrane technology after the membrane has been used for a long period. Membrane fouling is the accumulation of particles on the membrane surface or within membrane pores, leading to worsening membrane performance. Thus, the membrane can be modified by adding carbon-based nanomaterials such as carbon nanotubes (CNTs) and graphene oxide (GO) to reduce membrane fouling. GO is a suitable nanoparticle that can be used to enhance the hydrophilicity of the membrane. CNTs have been used as nano-filler to improve membrane performance in terms of thermal, mechanical, electrical, and optical characteristics. Previous studies have shown the performance of nanocomposite membrane incorporated with GO/MWCNTs as nanofillers has improved attributed to the synergistic combination of both nanomaterials [5–8].

Additionally, it is reported that the concentration of nanomaterials significantly affects the nanocomposite membrane characteristics and performance. According to Abidin et al. [9], membrane hydrophilicity increases as the concentration of carbon nanomaterials increases. This is because the contact angle is decreased and the porosity of the membrane is increased, which results in enhanced pure water flux. Besides, Khoerunnisa et al. [10] have reported that the permeability of nanocomposite membrane increased when the carbon nanomaterials concentration increased. This is because the mean pore size is expanded and increased in hydrophilicity by adding more carbon nanomaterials. However, the surface roughness increased when the carbon nanomaterials concentration increased due to the swells at the membrane surface by nanomaterials aggregation. Therefore, it can be concluded that the membrane properties can be enhanced by increasing the concentration of carbon nanomaterials. However, agglomeration will occur due to the overdose of

carbon nanomaterials subsequently deteriorating membrane performance.

Hence, this research investigates the effect of carbon nanomaterials concentration in nanocomposite membranes for dye removal. This research aims to synthesize the GO/MWCTNs nanocomposite membrane by using direct blending method with different concentration of carbon-nanomaterials, characterize the synthesized GO/MWCNTs nanocomposite membrane and evaluate the performance of synthesized membrane. The performance of the nanocomposite membrane is evaluated by the water permeability, rejection and fouling propensity towards the synthetic dye using methyl blue (MB).

2.0 METHODOLOGY

2.1 Materials

GO sheet was obtained from previous research and used in membrane modification [8]. MWCNTs with diameters of 12-15 nm and length of <10 μm were purchased from NanoAmor, USA. Polyvinylidene fluoride (PVDF) powder was obtained from ThermoFisher Scientific, USA and was used to synthesize membrane. N-N-di-methylacetamide (DMAc) (assay GC area > 99%) was supplied by Merck Co., Germany was used as solvent to dissolve the PVDF. MB was the dye used to evaluate the membrane performance and was purchased from Alfa Aesar, USA.

2.2 Membrane Synthesis

The GO/MWCNTs nanocomposite membrane was synthesized using a direct blending method by incorporating the GO and MWCNTs carbon nanomaterials into the membrane matrix [8]. GO and MWCNTs were first dispersed in DMAC under 30 minutes of sonication using an ultrasonicator Symphony 97043-932 (Avantor Inc., USA). After that, PVDF was added into the carbon nanomaterials suspension and stirred mechanically at 65 ℃ and 250 rpm for 4 hours. The mixed solution was then stirred for another 4 hours at 40 ℃ and 250 rpm to dissolve the PVDF completely in the mixture. The dope solution was kept in a desiccator overnight to remove the air bubbles trapped inside the solution. The subsequent step was to cast the membrane polymer film on a glass plate by slowly pouring the prepared dope solution on a flat nonwoven polyester membrane support (CU414 Opti, Neenah US) mounted on a glass plate with a support of a motorized film applicator. In the casting process, the 200 μm thickness of the membrane was produced. The glass plate affixed by dope solution was then immersed into the RO water bath for 24 hours to ensure the phase inversion was completed. The weight ratio of PVDF: DMAc and carbon nanomaterial ratio were fixed at 15:85 and 1:1,

respectively [11]. Table 1 shows the membrane formulation in studying the effect of carbon nanomaterials concentration towards the membrane.

Table 1 Formulation of dope solution in studying the effect of carbon nanomaterial concentration

Membrane	Concentration of nanomaterial (wt%)	Carbon nanomaterial ratio (g/g)	
		GO	MWCNTs
M0	0	0	0
M0.2	0.2	1	1
M0.5	0.5	1	1
M1	1.0	1	1

2.3 Membrane Characteristics

Surface Hydrophilicity

Membrane hydrophilicity was determined by the contact angle obtained through captive bubble contact angle using a contact-angle meter [12]. Firstly, the membrane sample was attached tightly onto a glass slide with the support of double-sided tape. A water droplet with approximately 13 μL was dropped on the membrane surface. The image was captured immediately by a high-speed camera at a frequency of 100 pcs/s. The obtained image was analyzed using Drop Shape Analysis (DSA) software to compute the contact angle of the membrane surface. The contact angle was measured at three different spots to get more accurate experimental data to minimize the experimental error.

Pore size and Porosity

The gravimetric method was used to determine the porosity of the membrane [13]. The working principle of this method is to determine the weight of liquid occupied in membrane pores based on the water sorption. This method began with the membrane being immersed in the distilled water for 12 hours to ensure all the membrane pores are filled with water. Then, the wetted membrane was cut into small pieces with a dimension of 1 cm × 1 cm. The wetted membrane was wiped gently with filter paper to remove the water droplet on the membrane surface. This wetted was weighed using a weighing machine. Then, the wetted membrane was dried at 50 ℃ for 24 hours to remove the water inside the membrane and weighed again. The porosity was measured by using Equation 1 [8].

$$\varepsilon = \frac{\frac{w_1 - w_2}{\rho_w}}{\frac{w_1 - w_2}{\rho_w} + \frac{w_2}{\rho_p}} \tag{1}$$

where, ε = membrane porosity (%), W_1 and W_2 = wet and dry weight of the membrane (g), ρ_w = density of distilled water (0.998 g/mL), ρ_p = density of polymer (PVDF = 1.765 g/mL)

Guerout-Elford-Ferry formula was used to determine the membrane mean pore size as stipulated in Equation 2 [14].

$$r_m = \sqrt{\frac{(2.90 - 1.75\varepsilon)8V\mu\delta}{\varepsilon PA}} \qquad (2)$$

where, r_m = membrane mean pore radius, (m), ε = membrane porosity (%), μ = water viscosity (8.9×10^{-4} Pa.s), l = membrane thickness (m), Q = permeate volumetric flowrate (m³/s), A = membrane area (m²), and ΔP = operational pressure.

Surface Charge

Membrane surface charge or zeta potential is most often determined using the streaming potential method using zeta sizer. Firstly, the membrane sample was cut into the size of 5 mm × 3.5 mm. Then it was immersed in 0.1 nM NaCl together with 300-350 nm latex particles at neutral pH value. Membrane surface charge was determined based on the mobility of latex particles at multiple distances away from the membrane surface by applying a 25 V/cm electric field [8].

Functional Groups

FTIR spectrometer Spectrum 100 (PerkinElmer Inc., USA) was used to determine the functional group on the synthesized membrane by the chemical reaction properties of the compound [15]. The spectroscopy was operated at wavelengths from 500 cm^{-1} to 4000 cm^{-1}.

Surface Morphology

The membrane surface images were investigated by field emission scanning electron microscopy (FESEM) SUPRA 55VP (Zeiss, Germany). The membrane with the appropriate size was mounted tightly on the sample stud with the help of double-sided carbon tape. During FESEM analysis, the outer surface of the membrane sample was coated with a thin layer of iridium under vacuum to avoid surface charging on the membrane. Surface topology of the membrane sample was inspected at a potential of 10.00 kV under 5.00 k× magnification.

2.4 Membrane Performance

Membrane Permeability Test

Membrane performance was studied using a dead-end membrane filtration system, MA-01730 (MILLIPORE, U.S). During the membrane permeability test, constant pressure of 3 bar was applied for 20 minutes to minimize the impact of compaction [16]. Different transmembrane pressure (TMP) of 1.0, 1.5, 2.0 and 2.5 bar are applied in the filtration process for 10 minutes of each TMP to determine the membrane permeate flux using Equation 3. Membrane permeability is obtained from the gradient of permeation flux against TMP [17].

$$J = \frac{\Delta V}{A\Delta t} \qquad (3)$$

where, J= permeate flux (L/m².h), ΔV = permeate flux (L), A = membrane filtration area (m²), and Δt = permeation time (hr)

Dye Rejection Test

The ability of dye rejection of the synthesized membrane was tested using 30 ppm of MB solution at 1 bar. The percentage of dye removal is determined using Equation 4 [18].

$$R = (1 - \frac{C_p}{C_f}) \times 100 \qquad (4)$$

where, R= membrane rejection (%), C_p= concentration of permeate (ppm), and C_f= Concentration of feed (ppm)

Membrane Antifouling Test

The antifouling properties of the synthesized membrane were tested with 30 ppm of MB solution at 1 bar and room temperature. Then, the 30 ppm of dye solution is filtered through the membrane for 20 minutes and the average flux is recorded every 1 minute [19]. Normalized flux is determined by using Equation 5 below:

$$Normalized\ flux = \frac{J_f}{J} \qquad (5)$$

where, J_f = flux of prepared dye solution that filtered through the membrane and J = Pure water flux calculate in Equation 3

3.0 RESULTS AND DISCUSSION

3.1 Membrane Characteristics

Surface Hydrophilicity

As tabulated in Table 2, the result shows the improvement of membrane hydrophilicity as the carbon nanomaterials were incorporated into the membrane where the contact angle was decreased from 68.99° of (M0 membrane) to 65.97° (M0.2 membrane). This can be explained by a higher amount of hydrophilicity group (-COOH) in carbon nanomaterials being transferred to the outer surface of the membrane during the phase-inversion process in water bath [20]. The inclusion of hydrophilic polar functional groups onto the surface of membrane increases the electrostatic interaction and binding

energy between membrane and water molecules and facilitate water permeation hence lower contact angle [21]. However, the membrane contact angle increases when the concentration of carbon nanomaterials increases further to 0.5 wt% and 1.0 wt%, meaning membrane hydrophilicity of M0.5 membrane and M1 membrane was slightly decreased. This was probably due to the higher concentration of carbon nanomaterials partially agglomerated on the membrane affecting the membrane surface properties such as membrane morphology. This result is consistent with Shokouhian and Solouki [22], where the membrane contact angle was decreased initially until the amount of nanomaterials (p-phenylenediamine-grafted MWCNT) increased to a specific loading (0.02 wt%). The membrane contact angle increases gradually, which could be related to the aggregation of MWCNTs in the membrane structure, hence increased in the membrane surface roughness. As the surface roughness increases, the maximal angle of the liquid on the hydrophobic surface increases [23].

Table 2 Contact angle of synthesized membrane

Membrane	Contact Angle (°)
M0	68.99 ± 0.55
M0.2	65.97 ± 3.66
M0.5	69.27 ± 1.98
M1	69.49 ± 0.53

Pore Size and Porosity

Table 3 shows the porosity and mean pore radius of the synthesized membrane. It can be observed that the pore size of the synthesized membrane was in the range of 0.02679-0.03056 μm. Hence, these membranes are classified as ultrafiltration (UF) membranes. Studies have shown that UF membranes incorporated with 2% of GO demonstrated highly efficient removal of organic dyes including MB molecules [24]. Besides, it can be observed that the porosity of the nanocomposite membrane showed an increasing trend. This was because of the increases in diffusion rate between DMAc solvent and water during phase inversion. A higher amount of GO/MWCNTs nanomaterials has higher hydrophilicity properties enhancing the thermodynamic instability of the solution by speeding up the DMAc outflux and water influx [5].

Table 3 Pore size and porosity of synthesized membrane

Membrane	Pore Size, r_m (μm)	Porosity, ε (%)
M0	0.02755	50.55
M0.2	0.02439	52.99
M0.5	0.02724	59.00
M1	0.03056	59.11

However, the mean pore radius of the synthesized membrane decreased when the concentration of GO/MWCNTs increased to 0.2 wt%, but at concentration above 0.2 wt% an increasing trend was seen. This trend was similar to the contact angle result in Table 2. Although high loading of carbon nanomaterials is hypothesized to improve membrane performance due to increased hydrophilicity, the tendency of carbon nanomaterials agglomeration is also likely to increase accordingly. Agglomeration of nanoparticles will lead to the formation of non-selective interfacial voids inside the nanomaterial aggregates. This leads to defective membranes, which are detrimental to the permeation and rejection capabilities [25, 26].

Surface Charge

Figure 1 shows the result of zeta potential for the synthesized membranes. It can be seen that the highest zeta potential is the M0.2 membrane and the lowest is the M1 membrane. The zeta potential of membrane was increased from -19.9 mV to -0.446 mV when incorporating 0.2 wt% GO/MWCNTs to the pristine membrane. This is because at low concentration of 0.2 wt%, the membrane was manifested by the MWCNTs which are positive charges subsequently increase the zeta potential of synthesized membrane [27]. When the carbon nanomaterial concentration was increased from 0.2 to 1 wt%, the zeta potential of membrane was decreased. This can be explained by the presence of the negatively charged functional group in GO which is the carboxyl group (-COOH) that can promote negative surface charge of the membrane [28]. Therefore, a higher concentration of carbon nanomaterials that consists higher amount of carboxyl group will significantly reduce the zeta potential of the synthesized membrane.
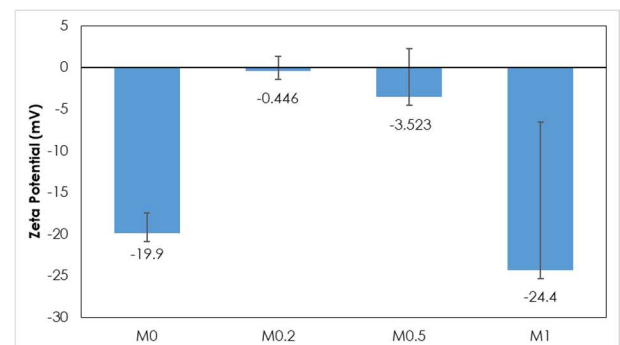

Figure 1 Zeta potential of synthesized membrane

This is consistent with studies by Rodrigues et al. [29], who studied the synthesis of nanohydroxyapatite nanocomposites by depositing MWCNTs-GO at 1–3 wt%. They reported that at lower concentrations of MWCNT-GO, the water loss of nanocomposite was significantly lower due to the typical hydrophobic character of MWCNTs-GO. Nevertheless, the increase in MWCNTs-GO concentration led to higher water pointing out the increase in the hydrophilic character of the nanocomposites.

Functional Groups

Figure 2 shows the result of FTIR for synthesized membrane. It can be seen that a peak with the wavelength of 1718 cm^{-1} is obtained for all synthesized membranes. This peak was attributed to the carboxyl group (C=O) in GO nanoparticles which were incorporated into the membrane. Besides that, the spectrum also showed the two peaks with the wavelengths of 1250 cm^{-1} and 1101 cm^{-1} which indicated the presence of the epoxy (C-O-C) and alkoxy (C-O) group, respectively [30]. As the concentration of GO/MWCNTs carbon nanomaterials increased, the peak became more intense and apparent. This was because the absorbance (intensity) of the peak is directly proportional to the concentration of the sample according to the Beer-Lambert Law [31].
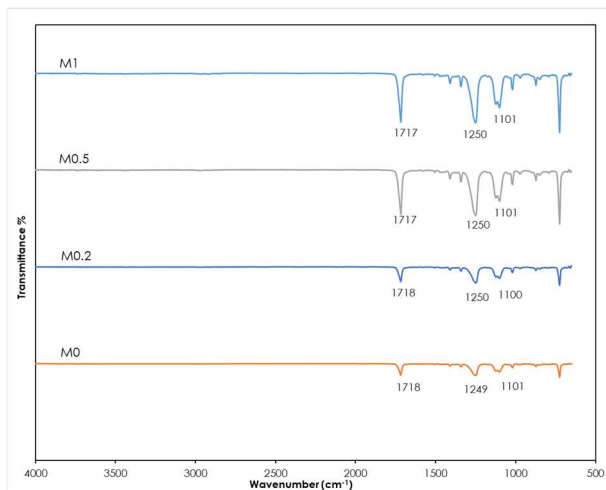
Figure 2 FTIR spectrum of the synthesized membrane

3.2 Membrane Performance

Membrane Permeability Test

Figure 3 shows the result of the water permeability test for the synthesized membrane. The M1 membrane has the highest permeability while the M0 membrane has the lowest water permeability. By incorporating the GO/MWCNTs into the membrane, water permeability was increased up to 36% when compared to the pristine membrane (M0). The value of water permeability for pristine M0 membrane was 68.77 L/m^2.h.bar and by increasing the concentration of carbon nanomaterials to 1 wt%, the value of water permeability increased significantly to 93.48 L/m^2.h.bar. This was because the mean pore radius of the M1 membrane (0.03056 nm) was significantly larger when compared to the pristine PVDF membrane (0.02755 nm).
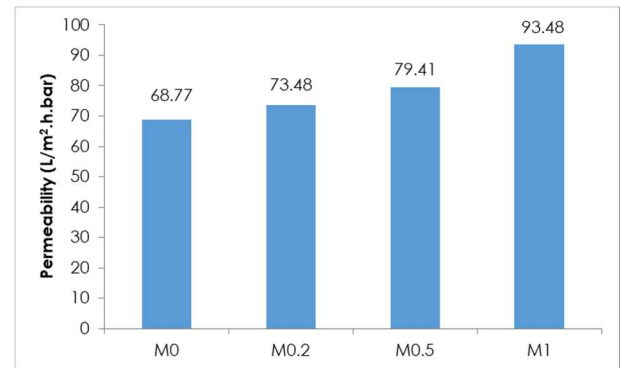
Figure 3 Water permeability result of synthesized membrane

As the concentration of GO/MWCNTs carbon nanomaterials increased to 0.2 wt% (M0.2), 0.5 wt% (M0.5) and 1.0 wt% (M1.0), the water permeability was increased by 6.8%, 15.4%, and 36% when compared to pristine PVDF membrane. The result of porosity in Table 3 could support this result. The porosity was increased when the concentration of carbon nanomaterials was increased. Hence, more water molecules were allowed to pass through the membrane. Besides, the membrane permeability result is in accordance with the membrane topology shown in Figure 4. It can be observed that membrane pore size and porosity increased drastically when 0.2 wt% and 1 wt% of carbon nanomaterials were added into the membrane matrix. The effect of the incorporation of carbon nanomaterials on membrane permeability was also studied and reported in previous studies. Majeed et al. [32] have found that the water flux of membrane increased by 63% by adding hydroxyl functionalized MWCNTs into the membrane which shows the enhancement in water permeability compared to the pristine membrane.
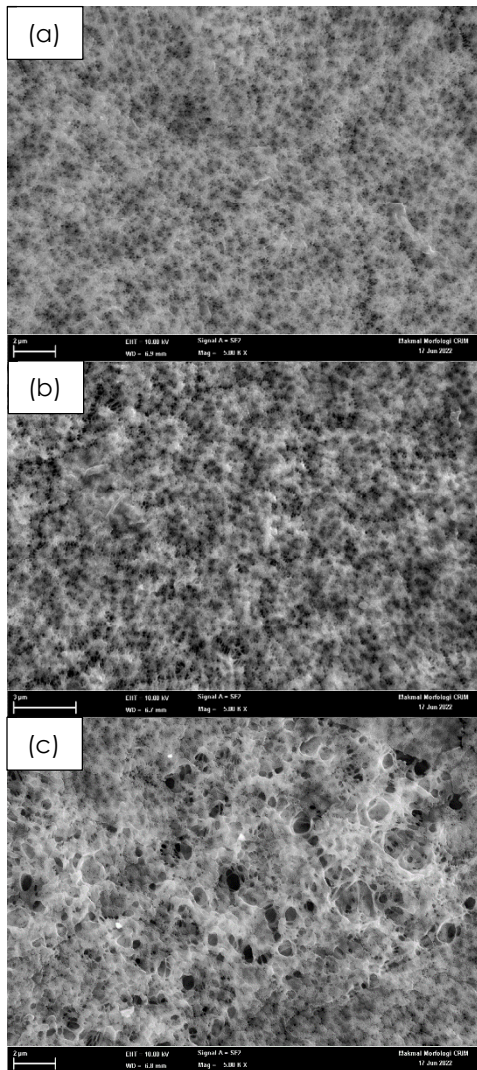
Figure 4 Surface topology of (a) M0, (b) M0.2, and (c) M1 membrane

Dye Rejection Test

Figure 5 shows the dye rejection result for synthesized membranes at 10 minutes and 20 minutes. It can be seen that the dye rejection is almost the same at 10 minutes for all synthesized membranes where the rejection value is around 97.84-99.84%. At 20 minutes, the membrane started to foul and the rejection value has dropped for all synthesized membranes. After 20 minutes of filtration, it can be observed that the pristine M0 membrane has the lowest dye rejection (65.35%) and the modified M0.2 membrane has the highest dye rejection value (97.00%). This result was supported by the surface charge of the membrane. Typically, the dye removal mechanism of the membrane was assigned to the repulsive electrostatic interaction between the negatively charged dye and negatively charged membrane surface [18]. However, MB was positively charged when dissolved in water [33]. Hence, the negative charge of

membrane will attract the dye particle on the membrane surface, leading to membrane fouling and reducing the dye rejection value. M0.2 membrane has higher zeta potential value (-0.466 mV) which is less negatively charged when compared to pristine M0 membrane (-19.9 mV). Hence, this results in more MB dye being rejected. Besides that, M0.2 membrane has smaller pore size (0.02439 nm) when compared to pristine membrane M0 (0.02755 nm) which also help to block the dye on the membrane surface through particle size sieve mechanism.

However, the membrane dye rejection was decreased by 27.62.9% when the concentration of carbon nanomaterials increased from 0.2 wt% to 0.5 wt%. This was probably because the zeta potential was decreased in M0.5 membrane (-3.523 mV) which the membrane surface was more negatively than the M0.2 membrane (-0.446 mV). Hence, positively charged MB tends to attract M0.5 membrane and lead to lower rejection. Besides that, the pore size of M0.5 membrane (0.027 nm) was higher than M0.2 membrane (0.024 nm), resulting in the dye molecules passing through the membrane at lower resistance. For the first 20 minutes, although M1 membrane with 1 wt% concentration of carbon nanomaterials has the lowest surface charge (-24.4 mV) and largest pore size (0.031 nm), but the dye rejection was increased to 91.45% which is significantly higher than the M0.5 membrane (69.38%). This was probably due to the cake layer formation on M1 membrane awing to the high permeation drag created by high water permeability. This has resulted in a sieving mechanism for solute rejection. Thus, the solutes can be removed by sieving/ adsorption onto the cake layer that has been formed over the fouled membrane surface [34].
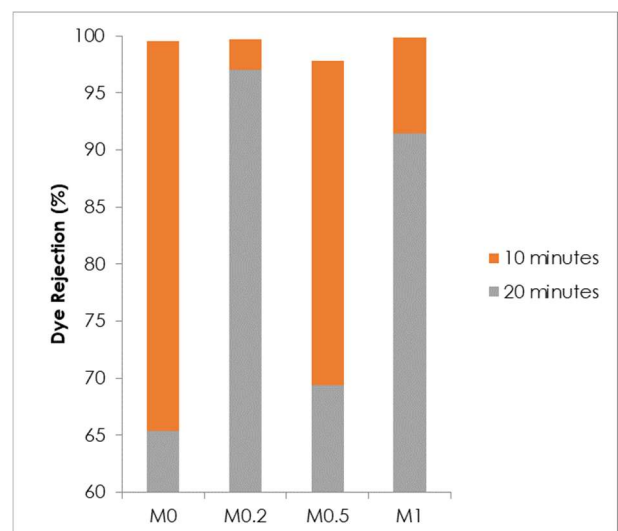

Figure 5 Dye rejection of synthesized membrane

Membrane Antifouling Test

Figure 6 shows the normalized flux of the synthesized membrane for the antifouling test. Normalized flux is

determined by dividing the permeate flux by pure water flux. Therefore, higher normalized flux has a higher permeate flux which indicates the membrane has better antifouling behaviour. It can be seen that M1.0 membrane has the highest normalized flux which indicates the best antifouling property for the modified membrane. At the same time, the lowest normalized flux was pristine PVDF membrane (M0) among the synthesized membranes.
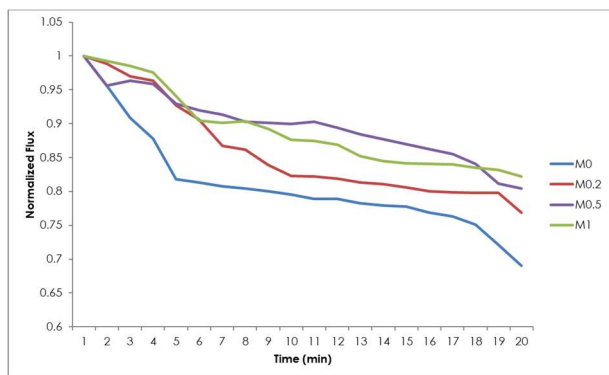

Figure 6 Result of antifouling test of synthesized membrane

In general, GO/MWCNTs nanocomposite membrane has better antifouling property than pristine membrane (M0) in which M0.2, M0.5, and M1.0 membranes have higher recorded normalized flux values of 0.7686, 0.8043, and 0.8225 after 20 minutes operation when compared to 0.6902 for pristine membrane. This was mainly contributed by many factors, particularly the porosity of GO/MWCNTs nanocomposite membrane. Rahimi et al. [18] have found that the antifouling performance was enhanced in composite membrane with higher porosity. They have reported that the antifouling property was enhanced with higher porosity synthesized membrane. Generally, higher porosity has a higher amount of empty space in a membrane reducing the membrane resistance and providing a larger space to block the dye particles. As the concentration of GO/MWCNTs increased for synthesized membrane, the porosity was also increased for the synthesized membranes M0 (50.55%) < M0.2 (52.99%) < M0.5 (59%) < M1 (59.11%). Therefore, the normalized flux value for the synthesized membranes also follows the same sequence, in which M0 < M0.2 < M0.5 < M1.

4.0 CONCLUSION

In conclusion, the research investigated the effect of carbon nanomaterials (GO/MWCNTs) as nanofillers on the performance of nanocomposite membrane. The result shows that the surface hydrophilicity was enhanced when the concentration of nanomaterials increased up to 0.2 wt%. However, higher concentration of nanomaterials reduces the

membrane hydrophilicity due to the agglomeration of nanomaterials. The M0.2 membrane with nanomaterials concentration of 0.2 wt.% has the best performance as it showed 6.85% and 32% improvement in water permeability and dye rejection compared to the pristine M0 membrane. Besides, M0.2 membrane has relatively good antifouling properties indicated by normalized flux (0.8043). This is due to the enhancement of hydrophilicity and zeta potential of the M0.2 membrane by carbon nanomaterials. In short, the optimum concentration of carbon nanomaterials is essential to enhance membrane performance as the agglomeration of nanomaterials occurs at high concentrations.

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